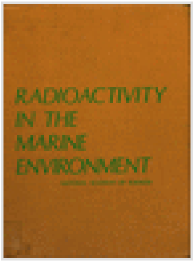

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FOREWORD

Fourteen years ago, the Academy published *Effects of Atomic Radiation on Oceanography and Fisheries*, one of a series of Academy studies on the biological effects of atomic radiation. That 137-page report summarized knowledge and problems known at that time and made suggestions for future programs for additional research. During the years that have followed, much new knowledge has been added to our understanding of the marine environment, the paths of movement that radioactive materials take within it, and the effects of such materials on other components of the environment.

Thus, it was appropriate that the Academy, in response to a request from the Atomic Energy Commission, undertake to prepare a second comprehensive review of our understanding of this area. The task was subsequently assigned to the Panel on Radioactivity in the Marine Environment of the Committee on Oceanography. This volume, the product of that review, brings together the results of the research of many scientists. A representative and highly competent group was assembled at Woods Hole, Massachusetts, in the fall of 1967 to prepare various sections of this report. Subsequently, the Panel has filled gaps and revised and updated this material to present a more comprehensive summary of the current state of understanding in this field.

It is particularly appropriate that this contribution to our understanding of the marine environment be available at a time when man is increasingly concerned with the ways in which his own actions may affect his environment. Though this work is specifically addressed to radioactivity in the marine environment, many of the concepts that pertain to our understanding of this problem can be applied effectively to studies of other wastes discharged into the marine environment, including industrial wastes, municipal sewage, pesticides, nutrients, heavy metals, and heat. It is perhaps ironic that, of the many substances that man has introduced into his environment over the centuries, he understands best and controls most rigorously the radioactive materials that have been produced only during the past quarter century. We are indeed fortunate that our intense concern for public safety and protection from radioactivity since 1950 has stimulated much basic research that can be applied to other serious environmental problems that we are just beginning to recognize.

Philip Handler
President
National Academy of Sciences

PREFACE

This report provides an account of what has been learned about radionuclides in the marine environment since the publication of a report about the same general subject by the National Academy of Sciences-National Research Council (NAS-NRC) in 1957. The present report has been written by the Panel on Radioactivity in the Marine Environment of the National Academy of Sciences' Committee on Oceanography.* Thirty-one persons from twenty organizations within the United States—university departments, national laboratories, and federal agencies—and from three laboratories outside the United States (Canada, England, and Monaco) have contributed to one or more of the ten chapters in the report.

Death came to one of the contributors, Joseph Rivera, and to one of the Panel members, Milner B. Schaefer, before the report was completed. Mr. Rivera, a scientist at the Atomic Energy Commission's Health and Safety Laboratory, New York City, devoted his research efforts to the measurement of man's exposure to radiation from fallout and contributed information from his research to the discussion, in Chapter 8, of the ecological aspects of radionuclides in the ocean. Dr. Schaefer was a contributor to the 1957 publication and also was a longtime member of both the National Academy of Sciences' Committee on Oceanography and the Panel on Radioactivity in the Marine Environment. He was educated as a fisheries biologist and was recognized internationally for his broad understanding of the ocean, including the impact of radionuclides on the marine environment. Dr. Schaefer had accepted the responsibility for writing the Introduction for this report but later asked to be relieved of this responsibility because of illness.

I wish to express our appreciation to Albert Bove, Editor for the Division of Earth Sciences, and Richard Vetter, Executive Secretary of the Ocean Affairs Board, for their invaluable editorial and administrative help.

Financial support by the United States Atomic Energy Commission, Division of Biology and Medicine, for expenses related to the writing of the report is gratefully acknowledged.

Allyn H. Seymour, *Chairman*
Panel on Radioactivity in
the Marine Environment

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*Died July 26, 1970.

CONTENTS

Chapter One

INTRODUCTION

A. H. Seymour

The first significant release of radionuclides to the marine environment began in late 1944 with the discharge of effluent from the reactors at the Hanford atomic plant to the northeast Pacific Ocean via the Columbia River. In July 1946, the detonation of two nuclear devices at Bikini Atoll introduced radionuclides into the north equatorial current system of the Pacific Ocean. Since that time, fallout from other nuclear detonations and radioactive wastes of various kinds have been added to the oceans of the world. Radionuclides are now found in all of the oceans; they present a potential health hazard and thus are of concern to the people of many nations. However, radionuclides as tags for chemical elements are a valuable tool for the study of biological, chemical, and physical processes of the ocean, and, in this report, both the beneficial uses and the potentially harmful effects of radionuclides are considered.

Studies of radionuclides in the marine environment and their impact upon populations of fish and invertebrates began at Bikini Atoll in 1946. During subsequent nuclear detonations at Bikini and later at Eniwetok Atoll, small-scale radioecological studies provided information about the biological and geographical distribution of fallout radionuclides near the test sites, but this information was mostly semiquantitative and semiqualitative. In the 1950's, the number of nations testing nuclear devices, as well as the number of nuclear detonations, increased; the technology of radiation detection and measurement improved greatly; and public concern about fallout in both the terrestrial and marine environments grew. In 1956–1957, the National Academy of Sciences–National Research Council (NAS–NRC) published a series of reports on the biological effects of atomic radiation. One of these reports, ***The Effects of Atomic Radiation on Oceanography and Fisheries*** (NAS–NRC Publication No. 551), is an appraisal of radioactivity in the marine environment at a time when fallout from the detonation of nuclear devices was the principal source of artificial (man-produced) radionuclides in the environment. Now, 25 years after the detonation of the first nuclear device, radionuclides in the ocean continue to be a subject of worldwide concern, although attention is now centered on the input of radionuclides from the peaceful uses of nuclear energy rather than on "fallout," which has become a nearly forgotten word.

The present report, prepared by the Panel on Radioactivity in the Marine Environment of the National Research Council's Committee on Oceanography, provides an account of what has been learned about radionuclides in the ocean since 1957. The subjects selected by the Panel for discussion are sources of radionuclides; the distribution of ^{137}Cs and other fallout radionuclides; physical processes of water movement; chemical systematics and elementary reactivities in seawater; sorption and scavenging properties of sediments; accumulation and distribution of radionuclides by marine organisms; radioecological interactions and considerations; and the effects of radionuclides on marine organisms and man.

Sources of natural and artificial radionuclides in the ocean are discussed by Joseph, Gustafson, Russell, Schuert, Volchok, and Tamplin in Chapter 2. Natural radionuclides include radionuclides and their radioactive daughters that have persisted since the earth's formation and radionuclides that are being produced constantly in the earth's atmosphere. The radioactivity of seawater from all natural radionuclides is about 750 dpm/liter, 97 percent of which derives from ^{40}K. Because of the long half-lives of many natural radionuclides, all organisms living in the ocean have been and will continue to be exposed to an essentially constant level of background radiation.

The principal sources of artificial radionuclides in the ocean are fallout from nuclear detonations, direct or indirect wastes from nuclear reactor operations, and wastes from medical, scientific, and industrial uses of radionuclides. Since the second nuclear test ban treaty in 1963, fallout has decreased markedly, and in recent years only small quantities of solid radioactive wastes have been disposed of in the sea by the United States. However, the number of nuclear reactors has increased steadily, and, at this time, wastes from nuclear reactor operations are considered to be the greatest source of radioactivity in the marine environment.

Only small amounts of radionuclides are released during the normal operation of nuclear reactors. The sources of these radionuclides, when they do occur, are excess primary coolant, produced by expansion of the coolant during reactor warm-up, and miscellaneous wastes. The eight original production reactors at Hanford are an exception in that the primary coolant is not recycled in a closed system but is returned directly to the Columbia River. Hence, greater amounts of radionuclides are released directly to the aquatic environment from a Hanford production reactor than from other types of reactors of similar capacity. Although only a few curies of radionuclides are released directly to the environment by nuclear reactors, thousands of curies are produced within the fuel elements of the reactor. These radionuclides, when chemically separated from the unused fuel element, create a major disposal problem. The present policy in the United States is to convert these highly concentrated liquid wastes into a chemically inert solid for underground storage. Previously, these liquid wastes were stored in underground tanks, and, at one time, their disposal in containers in the deep ocean was considered, but the policy was never adopted.

The oceanic distribution of fallout radionuclides is discussed in Chapter 3 by Volchok, Bowen, Folsom, Broecker, Schuert, and Bien. One of the important considerations is whether ^{137}Cs and ^{90}Sr are present at depths greater than 1,000 m. There are three expert opinions–one that they are present, another that they are not present, and a third that they are present in particulate but not in ionic form. Because of the significance of this issue to the understanding of ocean-water circulation and because no consensus was reached by the Panel, the unusual procedure was adopted of allowing proponents of each view to present synopses of their arguments.

The presence of ^{137}Cs and ^{90}Sr in deep water is also discussed in Chapter 4, where the conclusion is reached that these two radionuclides could be expected at depths between 500 and 2500 m at middle and low latitudes but not in quantities as great as some authors have reported.

The principal subjects of discussion in Chapter 4, by Pritchard, Reid, Okubo, and Carter, are those advective and nonadvective processes of water movement that influence the distribution of radionuclides in the ocean. The simplest situation, the instantaneous release of radionuclides in a physicochemical state in which complete and immediate solution occurs, is given first consideration; more complicated situations are then considered, in which the radioactive materials are released by several methods and in different physicochemical states.

The radioactive "cloud" formed in the ocean by the introduction of radionuclides is moved horizontally by local currents and vertically by small-scale turbulent-motion processes. The eventual dispersion of the cloud is affected by the size of the field-of-motion eddy. Eddies larger than the cloud produce advection of the cloud as a whole, while eddies smaller than the cloud produce internal shearing and stirring. Eddies the same size as the cloud produce shear in the velocity field and in this way significantly influence the shape of the cloud and contribute to its dispersion as well. As the cloud grows in size, the scales of motion that contribute to its movement and mixing change. Models designed to predict the shape of the cloud or the concentration of the radionuclides at a specific place or time must take into account the nature of these changes.

The subject of "concentration factors" is considered in several chapters, and tables of concentration factors for marine organisms are given in Chapters 5, 7, and 8. Concentration factors are useful for the prediction of the routes and rates of transfer of radionuclides from sea to man, and also for the identification of the nonconservative elements in seawater, that is, elements whose concentration may be significantly altered by biological processes.

Several conditions may limit the effective use of concentration factors. First, by definition, the concept loses meaning when applied to organisms that accumulate a chemical element or radionuclide in some way other than directly from seawater. Second, the value for the amount of the chemical element or radionuclide in the organism should be that value reached when equilibrium between the organism and seawater is reached. Third, the value for the amount of the chemical element or radionuclide in seawater should be the representative value for the entire period of accumulation of the chemical element or radionuclide by the organism and not the value for a single sample. Also, there may not be a single concentration factor for any common group of organisms, including those of the same species, because of the changes in concentration in the organism related to the physiological and environmental factors that influence metabolism. Further discussion of the use of concentration factors can be found in Chapter 8.

Each of the tables of concentration factors in the three chapters was used for a different purpose. The table in Chapter 5, "Marine Chemistry," was used to provide some insight into the accumulation and distribution of specific elements by plants and animals. For this purpose, large errors were acceptable, and the marine organisms were therefore classified only as either plants or animals and the concentration factors were expressed as logarithms. Also, as discussed in Chapter 5 by Goldberg, Broecker, Gross, and Turekian, the concentration factors and the ratio of elements in both deep water and surface water were used to identify the nonconservative elements.

The nonconservative elements are identified in greater detail in Chapter 7, "Accumulation and Redistribution of Radionuclides by Marine Organisms," by Lowman, Rice, and Richards. In addition, the elements that are concentrated by phytoplankton by a factor of 1,000 or more are listed, along with important radionuclides of these elements, that are of interest in the consideration of biological transport. The list includes structural, catalytic and easily hydrolyzed elements, heavy halogens, and heavy divalent ions—some of unknown biological function—all reported to be present either in fallout from nuclear detonations or in effluent discharged into the sea from nuclear reactors or reactor fuel-processing plants.

The most comprehensive table of concentration factors is given by Bowen, Olsen, Osterberg, and Rivera in Chapter 8, "Ecological Interactions of Marine Radioactivity," where the concentration factors for chemical elements are listed by trophic level and by plant and animal group. Although clear-cut conclusions could not be made, the authors of this chapter were of the opinion that concentration factors, in general, were inversely related to trophic level. In this chapter, the use of simulation models as the ultimate method of predicting what happens to artificial radionuclides introduced into the marine environment is also discussed. Like other models of ecological systems, the applicability of the model is inversely related to the amount of pertinent information left outside the model. The success of models in the management of certain fishery resources and the special promise that the use of simulation models has for marine ecology indicate the possible success of models for describing the dynamics of radionuclides in the marine environment. However, before present models can move beyond the "early model" stage, as defined in Chapter 8, much more information is needed.

Radionuclides can be removed from seawater by the biota and also by sediments and sedimentary particles, as described by Duursma and Gross in Chapter 6. The sorption capacity of a sediment for a radionuclide appears to be controlled by the physicochemical state of both the sediment and the radionuclide. After sorption, especially to a clay-mineral particle, the radionuclide may move within the structure of the particle while the particle is being moved by currents and wave action. Bottom grazers and burrowing organisms are important factors in the translocation of radionuclides in the surface layer of bottom sediments.

The use of sediments to scavenge radionuclides from seawater after an accidental release of radionuclides has been suggested. By this process, the radionuclides would merely be transferred from the water to the bottom sediments, which may or may not be advisable. The large quantity of sediments that would be needed under the most favorable circumstances raises the question of practicability and cost.

The effects of radionuclides in the ocean on marine organisms and man are discussed in the last two chapters. The discussion of the effects on marine organisms in Chapter 9 by Templeton, Nakatani, and Held points out that the results of laboratory experiments in various countries are not in agreement about the sensitivity of fish larvae to ionizing radiation. The results of some experiments indicate that there was no significant increase in abnormal larvae in fish exposed to ^{90}Sr + ^{90}Y concentrations a million times greater than the ^{90}Sr + ^{90}Y concentrations that in other experiments were reported to have produced significant increases in abnormal larvae. The concentration of ^{90}Sr + ^{90}Y in the experiments in which effects were observed was only one third the concentration of naturally occurring ^{40}K in seawater. These investigators also believe that embryo mortality greater than 10 percent will significantly reduce the population size of the adult stock. For fecund species, the accuracy of this statement is not obvious. The radiosensitivity of the embryos and larvae of fishes is pertinent to the question of radioactive-waste disposal in the sea; hence, the need for further study of the radiosensitivity of fish larvae is evident.

The radiation dose to plaice, a bottom fish, living in the vicinity of the discharge of radioactive effluent into the Irish Sea from the Windscale Chemical Reprocessing Plant is exceptionally well documented and can be extrapolated in a general way to other species and other areas. The dose was predicted from calculated values for the concentrations of radionuclides in seawater and later determined empirically by use of thermoluminescent dosimeters (TLD's). The contribution of natural radionuclides to the total dose of 7.4 rad for a plaice 2 miles from the discharge point was 1 percent; the major contribution was from the bottom sediments, with a small contribution from seawater and internal radionuclides. The dose to a fish living in surface water would be considerably less, because the contribution by bottom sediments would be essentially zero.

The predicted doses and the doses measured by the

TLD's attached to plastic tags on the fish were in good agreement. The small size of TLD's permits their use either internally or externally for fish, oysters, and other organisms of a similar or larger size. Future use of TLD's for *in situ* measurement of radiation doses to living organisms is expected to provide information not previously available.

In considering the effects of ionizing radiation on marine organisms, the primary concern is with populations, not individuals. Unless deaths by radiation reduced the stock below the level for maximum sustained yield, these deaths would not jeopardize the population and would merely be another type of mortality. In the appraisal of the genetic effects of ionizing radiation on marine organisms, it should be recognized that genetic damage at the population level is reparable by natural selection. A quotation in Chapter 9 from Purdom (1966) summarizes the effect of ionizing radiation on marine organisms as follows:

> It would seem likely that the genetic response of populations is relatively unimportant and that general mortality and infertility would be the limiting factors in the extent to which populations may overcome radiation exposure.

The primary concern about the presence of radionuclides in the ocean is their effect on man, as discussed by Foster, Ophel, and Preston in Chapter 10. Man may be exposed to ionizing radiation from seawater in various ways. Swimming, walking on beaches, and handling contaminated fishing gear are some of the ways, but none is as important as the ingestion of seafoods. The question, "How much seafood can be eaten safely?" then arises and leads to the concept of an "acceptable dose." The acceptable-dose concept implies that exposure to ionizing radiation from any source entails some risk of a biological effect; therefore, a dose is considered acceptable only if the benefits are greater than the risks and the risks are acceptable both to the individual and to society as a whole.

The acceptable dose recommended by the International Commission on Radiological Protection (ICRP) is 5.0 rem per 30 years or 0.17 rem per year. This dose is the acceptable average whole-body radiation dose for the general population, exclusive of radiation from natural background sources and from medical uses of ionizing radiation. The recommended dose limits, either for groups that can accept a risk greater than the acceptable risk for the general population or for single organs or tissues, are greater than 0.17 rem per year. The ICRP recommendations have been made on the assumption that dose and effect are linearly related, i.e., that there is no threshold effect.

The introduction of radionuclides into the ocean necessitates an assessment of the environment both before and after contamination occurs. The pre-event assessment is needed to provide information for the prediction of effects, and the postevent evaluations are needed to determine the actual effects. Predictions can be made by either the "critical pathway" or the "specific activity" approach.

The critical-pathway approach identifies the route and estimates the concentration of the radionuclide at each step in the route from seawater to man. The specific-activity approach establishes a maximum permissible value for radionuclides in seawater by equating the specific activity in seawater to the specific activity in the critical organ. If the specific activity of the critical organ is not exceeded in seawater, then the allowable body burden of the radionuclide in man cannot be exceeded, regardless of the amount of seafood eaten. The specific activity is simple to calculate and does not require the information needed by the critical-pathway approach in regard to food webs, concentration factors, transfer coefficients, and man's use of seafoods. The specific-activity approach, however, assumes that reliable information is available for the amounts of chemical elements present in critical organs and cannot be used for radionuclides that are poorly assimilated and thus have the gastrointestinal tract as the critical organ. Values derived by this approach are unduly conservative because full compensation cannot be made for radioactive decay that occurs as the radionuclide moves from the sea to man and because all of man's food is assumed to come from the contaminated area of the sea.

Both the specific-activity and the critical-pathway approaches can be used to predict the potential dose to man from the introduction of radionuclides into the ocean. The simplicity of the specific-activity approach suggests that it be tried first if the nuclides involved do not have the gastrointestinal tract as the critical organ. If the predicted value for the concentration of a radionuclide released in seawater is less than the maximum permissible concentration value in seawater as calculated by the specific activity approach, then the predicted release of radionuclides will not lead to a radiation exposure greater than the recommended limits. If the predicted radionuclide value is greater than the conservative value derived by the specific-activity approach, or if a more precise estimate of the probable dose is needed, then the critical-pathway approach should be used.

The specific-activity approach was suggested by an *ad hoc* committee of the National Research Council as a method for regulating radioactive waste disposal off the Pacific coast of the United States at a time when disposal of radioactive wastes to the ocean was being considered (National Academy of Sciences–National Research Council, 1962). The critical-pathway approach has been used frequently: The National Academy of Sciences–National Research Council (1959a, b; 1962) and the International Atomic Energy Agency Panel on Radioactive Waste Disposal into the Sea used this method to calculate maximum permissible concentrations of radionuclides in seafoods and in seawater, and the United States, the United Kingdom, France, Sweden, and India have employed it in the management of the disposal of radioactive wastes. The release of radionuclides from the Windscale Chemical Reprocessing Plant is an interesting example of the use of the critical-pathway approach. It was found that ^{106}Ru released from the plant was accumulated by an alga that is used in the preparation of a food eaten by a community of Welshmen. As a consequence, the release of ^{106}Ru controls the release of radioactive wastes from this plant.

Significant but still incomplete data are now available for an evaluation of the biological consequences of the introduction of radionuclides into the ocean. The ability to predict the distribution of radionuclides in the sea, as well as the ability to keep human exposure within the guidelines specified by the International Commission on Radiological Protection and the Federal Radiation Council, has been adequately demonstrated by the predictions and follow-up surveys that already have been made. These guidelines are based on many factors, not all perfectly known, and are subject to change when new and better information becomes available. Therefore, the present guidelines are subject to revision, although there is no evidence that the past and present policies and practices for radioactive waste disposal in the sea have jeopardized man or any marine species or ecosystems.

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Chapter Two

SOURCES OF RADIOACTIVITY AND THEIR CHARACTERISTICS

A. B. Joseph, P. F. Gustafson, I. R. Russell, E. A. Schuert, H. L. Volchok, A. Tamplin

Prior to 1942, man's exposure to ionizing radiation was limited, essentially, to natural radioactivity and medical x rays. Since then, the use of atomic energy has become an important part of the modern way of life. The first controlled, self-sustaining nuclear chain reaction occurred in December 1942, and the first atomic bomb was tested successfully in July 1945. Both kinds of events create man-made radioactivity. Over the past 25 years, the uses of atomic energy have grown more numerous, more diverse, and more widespread. In only a quarter of a century, man has created more radioactivity than existed naturally in the world in 1942, and the future will see quantum jumps in production. Table 1 indicates the diversity of types and applications of man-made atomic energy.

To increase our understanding of radioactivity in man's environment–more specifically, in the oceanic environment–it is helpful to examine the uses of atomic energy, the conditions of the creation of radioactivity, its control and management, and where and how radioactivity is used; these factors bear on the role of radionuclides in the physics, chemistry, and biology of the sea. Not all uses of atomic energy release radioactivity to the environment; some never do, some always do, and some do only under accidental conditions. The spectrum of radioisotopes released into the environment varies with the kinds of applications, the chemical and physical form of the radioactive material, and the environmental conditions at the place and time of use.

Two sources of atomic energy create fission-product radioactivity–nuclear reactors and nuclear explosives, both of which involve the fission of heavy elements. Nuclear explosives may also involve fusion of light elements. During the fission and fusion process, neutrons are released that may interact with surrounding materials to create new radionuclides. All of the other sources of atomic energy (see Table 1) involve the radioactivity derived from the fission or fusion processes–either fission products or activation products. The greatest future source of radioactivity will be nuclear reactors, assuming that the atmospheric nuclear testing moratorium will continue. In 1967, in the United States, there were 17 commercial nuclear-powered generating plants with a total capacity of 2,000 electrical megawatts (MW_e). In the early 1970's, the number of U.S. plants will be increased by 32 and power production by 23,000 MW_e–a tenfold increase in nuclear generating capacity (U.S. Atomic Energy Commission, 1967a). It has been estimated that 145,500 MW_e will be generated by nuclear plants by 1980 (U.S. Atomic Energy Commission, 1967b), not including reactors outside of the United States or reactors for ships, research, and special purposes.

It is evident that significant amounts of fission products will be generated in nuclear power plants. The fission of 1 g of ^{235}U produces approximately 1 g of fission products and 1,000 kilowatt-days of energy (at 100 percent efficiency). At the end of one MW-year, the "important" fission-product

TABLE 1 Sources and Applications of Atomic Energy

Energy Source	Output	Applications
Nuclear reactors (controlled nuclear fission)	Steam, electricity	Electric power–stationary or portable plants, desalination, propulsion of submarines and surface ships
	Heat, electricity, neutrons	Spacecraft and satellite power, spacecraft propulsion, research and special materials production
Nuclear explosives	Kinetic energy	Military and civilian applications: large-scale earth moving, subsurface excavation, mineral extraction from underground
Encapsulated radioisotopes (sealed sources)	Electricity	Marine navigation aids, unmanned weather stations, spacecraft project power, artificial human organs
Encapsulated radioisotopes	Gamma and beta radiation	Food preservation, polymerization, sterilization of medical supplies, thickness gauges
Radionuclides	Beta and gamma radiation	Medical uses, tracers in scientific research, measures of manufacturing processes

activity would amount to approximately 700,000 Ci (U.S. Public Health Service, 1960). However, under normal operating conditions, nuclear reactors release little radioactivity into the environment (see Table 11).

To date, the largest source of radionuclides in the ocean has been the atmospheric nuclear weapons testing programs of the United States, the Soviet Union, the United Kingdom, the People's Republic of China, and France. Meteorological processes distribute radioactive test debris throughout much of the stratosphere, which in turn results in worldwide deposition of fallout on the earth's surface. Only China and France continue atmospheric weapons tests and add to the worldwide fallout burden. On the whole, the inventory of nuclear debris in the stratosphere has declined since cessation of widespread atmospheric testing in 1963.

Future uses of nuclear explosives are contemplated for large-scale construction projects and for programs in conservation of natural resources; the former are to be underground explosions with less release to the atmosphere than past nuclear testing programs, and the latter are expected to be contained explosions.

Encapsulated radioisotopes release no radioactivity to the environment unless the container is ruptured. However, because of the substantial amounts of radioactivity involved, it is important to consider the environmental implications of accidents.

Scientific, medical, and industrial uses of controlled amounts of radioisotopes may release some activity to drainage systems that flow to the sea; generally, these quantities are small. In the United States, disposal of packaged wastes to the sea has been largely discontinued in favor of land burial. However, other countries still use the sea for disposal of low-level packaged radioactive waste.

NATURAL RADIOACTIVITY

More than 60 radionuclides occur naturally in the earth's environment. These derive primarily from two sources: the rocks and minerals of the solid earth and cosmic-ray activity.

The solid-earth radionuclides include 32 daughters of the long-lived uranium and thorium isotopes. There are 11 other radioisotopes spread across the periodic table whose common characteristic is a long half-life, i.e., between 10^7 and 10^{15} years. Some of these isotopes probably originate from the spontaneous fission of uranium *in situ* (Parker and Kuroda, 1958).

The radionuclides of the earth's crust are believed to have been created when the earth was formed. Wind and water erode and leach the radionuclides as part of the process of rock weathering; they are then carried to the oceans by runoff or by the winds as gases and particulates. In the oceans, the materials are found throughout the water column and in the sediments, according to their physical and chemical states, as described in Chapter 5.

Cosmic rays originating in outer space continuously bombard the earth's gaseous envelope. Some of these particles collide with atoms of nitrogen, oxygen, and argon in the atmosphere, producing radioactive elements. These radionuclides are brought down in solution to the earth's surface by precipitation, are sorbed on particles that settle out or are carried down by precipitation, or behave as a gas and enter into equilibrium reactions with water and other materials on the earth's surface. At least 14 radionuclides are created in this manner, and a number of these have been measured in seawater and in ocean sediments (see Table 2). Two of these radionuclides, ^{3}H and ^{14}C, also are produced by nuclear detonations.

TABLE 2 Concentrations of Natural Radionuclides in the Sea[a]

Radionuclide	Half-life	In Seawater		In Sediments (g/g)
		g/liter	dpm/liter	
		Terrigenous Origin		
Potassium-40	1.25×10^{9} yr	4.7×10^{-5}	670	$(0.8\text{–}4.5) \times 10^{-6}$
Rubidium-87	4.7×10^{10} yr	3.4×10^{-5}	64	–
Indium-115	6.0×10^{14} yr	–	–	–
Iodine-129	1.7×10^{7} yr	1.6×10^{-10}	0.06	–
Lanthanum-138	2.0×10^{11} yr	–	–	–
Neodymium-144	5.0×10^{15} yr	–	–	–
Samarium-147	1.3×10^{11} yr	–	–	–
Lutetium-176	2.4×10^{10} yr	–	–	–
Tungsten-180	10^{14} yr	–	–	–
Rhenium-187	5.0×10^{10} yr	–	–	–
Platinum-190	10^{12} yr	–	–	–
Thallium-207	4.79 min	$<1.2 \times 10^{-23}$	<0.005	2.1×10^{-21}
Thallium-208	3.10 min	4.1×10^{-24}	0.003	6.7×10^{-22}
Lead-210	19.4 yr	1.1×10^{-15}	0.2	4.5×10^{-14}
Lead-211	36.1 min	$<9.0 \times 10^{-23}$	<0.005	1.6×10^{-20}
Lead-212	10.6 hr	2.4×10^{-21}	0.007	3.9×10^{-19}
Lead-214	26.8 min	2.9×10^{-21}	0.2	1.2×10^{-19}
Bismuth-210	5.01 day	7.8×10^{-19}	0.2	3.1×10^{-17}
Bismuth-211	2.16 min	$<5.6 \times 10^{-24}$	<0.005	1.0×10^{-21}
Bismuth-212	60.5 min	2.2×10^{-22}	0.007	3.7×10^{-24}
Bismuth-214	19.7 min	2.1×10^{-21}	0.2	8.8×10^{-20}
Polonium-210	138.4 day	2.2×10^{-17}	0.2	8.8×10^{-16}
Polonium-211	0.52 sec	$<6.8 \times 10^{-29}$	$<1.5 \times 10^{-6}$	1.2×10^{-26}
Polonium-212	3.04×10^{-7} sec	1.2×10^{-32}	0.005	2.4×10^{-29}
Polonium-214	1.64×10^{-4} sec	3.0×10^{-28}	0.2	1.1×10^{-27}
Polonium-215	1.83×10^{-3} sec	$<8.1 \times 10^{-29}$	<0.005	1.4×10^{-26}
Polonium-216	0.158 sec	1.0×10^{-26}	0.007	1.7×10^{-24}
Polonium-218	3.05 min	3.4×10^{-22}	0.2	1.4×10^{-20}
Radon-219	3.92 sec	$<1.7 \times 10^{-25}$	<0.005	3.1×10^{-23}
Radon-220	51.5 sec	3.3×10^{-24}	0.007	5.4×10^{-22}
Radon-222	3.8 day	6.3×10^{-19}	0.2	2.5×10^{-17}
Francium-223	22 min	$<7.0 \times 10^{-24}$	$<6.0 \times 10^{-4}$	1.4×10^{-21}
Radium-223	11.68 day	$<4.4 \times 10^{-20}$	<0.005	8.5×10^{-18}
Radium-224	3.64 day	2.1×10^{-20}	0.007	3.4×10^{-18}
Radium-226	1,622 yr	1.0×10^{-13}	0.2	4.0×10^{-12}
Radium-228	6.7 yr	1.4×10^{-16}	0.05	2.3×10^{-15}
Actinium-227	21.6 yr	$<1.0 \times 10^{-15}$	<0.2	5.9×10^{-15}
Actinium-228	6.13 hr	1.5×10^{-20}	0.075	2.4×10^{-19}
Thorium-227	18.17 day	$<7.0 \times 10^{-20}$	<0.005	1.3×10^{-17}
Thorium-228	1.91 yr	$<4.0 \times 10^{-17}$	<0.07	7.0×10^{-16}
Thorium-230	7.52×10^{4} yr	$<3.0 \times 10^{-13}$	<0.014	2.0×10^{-10}
Thorium-231	25.6 hr	8.6×10^{-20}	0.1	2.9×10^{-20}
Thorium-232	1.42×10^{10} yr	1.0×10^{-10}	2.4×10^{-15}	5.0×10^{-6}
Thorium-234	24.1 day	4.3×10^{-17}	2.2	1.4×10^{-17}
Protoactinium-231	3.43×10^{4} yr	$<2.0 \times 10^{-12}$	<0.2	1.0×10^{-11}
Protoactinium-234	1.14 min	1.4×10^{-19}	220	4.7×10^{-20}
Uranium-234	2.48×10^{5} yr	1.9×10^{-10}	2.3–2.9	8.1×10^{-11}
Uranium-235	7.13×10^{8} yr	2.1×10^{-8}	0.09–0.17	7.1×10^{-9}
Uranium-238	4.5×10^{9} yr	3.0×10^{-6}	2.0–2.5	1.0×10^{-6}
		Cosmic Origin		
Hydrogen-3	12.26 yr	1.7×10^{-18}	0.036	–
Beryllium-7	53 day	$<4.9 \times 10^{-17}$	<38	–
Beryllium-10	2.5×10^{6} yr	2.2×10^{-17}	10^{-6}	$(1\text{-}3) \times 10^{-13}$
Carbon-14	5,570 yr	$(2\text{-}3) \times 10^{-14}$	0.2–0.3	$(0.1\text{-}1) \times 10^{-13}$
Sodium-24	2.6 yr	–	–	–
Aluminum-26	7.4×10^{5} yr	2.9×10^{-19}	1.2×10^{-8}	–

TABLE 2 (Continued)

Radionuclide	Half-life	In Seawater g/liter	In Seawater dpm/liter	In Sediments (g/g)
Silicon-32	710 yr	5.0×10^{-19}	2.4×10^{-5}	$(0\text{–}2) \times 10^{-16}$
Phosphorus-32	14.3 day	$<1.5 \times 10^{-18}$	–	–
Phosphorus-33	25 day	$<3.1 \times 10^{-18}$	–	–
Sulfur-35	87 day	$<1.8 \times 10^{-18}$	–	–
Chlorine-35	3.1×10^{5} yr	7.7×10^{-17}	5.5×10^{-14}	–
Chlorine-39	1 hr	–	–	–
Argon-37	35 day	–	–	–
Argon-39	270 yr	3.8×10^{-20}	2.9×10^{-6}	–

[a] Compiled from Koczy and Rosholt (1962) and Lal and Peters (1967).

TABLE 3 Number of Nuclear Detonations through 1968, by Local Environment and Elevation of Explosion

	Continental	Arctic Islands	Coral Islands	Open Ocean
Surface	58[a]	79[a]	16	–
Tower	45	–	11	–
Air and balloon[b]	47	–	48	1
High-altitude and rocket[c]	4	–	–	10
Barge and ship	–	–	35	–
Underwater	–	1	3	2
Underground	116	–	–	–

[a] Includes Soviet and Chinese shots indicated only as "atmospheric."
[b] Approximately 1,000–86,000-ft altitude.
[c] Approximately 141,000-ft to 300-mi altitude.

NUCLEAR EXPLOSIVES

Through December 1968, some 470 nuclear explosives had been detonated in many parts of the world in the testing programs of the United States, the Soviet Union, the United Kingdom, France, and China. All nuclear tests, with the possible exception of contained, underground explosions and those in outer space, result in radioactivity being introduced into the oceans. The kinds, amount, and characteristics of the radioactivity introduced varies with the number, size, and materials of the devices themselves. The environmental media that react with the heat and fission and activation products of the explosion determine the physical and chemical characteristics of the debris and its subsequent rate and pattern of distribution. Table 3 is a composite summary of known or detected tests conducted by the five nuclear powers through 1968 (Glasstone, 1964; various press announcements of Chinese and French tests). Through 1968, nuclear weapons tests have been conducted in both hemispheres under a wide variety of environmental situations—on, over, and under continental land masses, coral and arctic islands, and the open sea.

It has been estimated that there were some 194 megaton equivalents of fission produced by nuclear testing between 1945 and 1963: 139 megatons as air bursts, 54 as coral surface or barge bursts, and one megaton as tower bursts (Federal Radiation Council, 1963).

The total of 194 megatons corresponds to about 2.8×10^{28} fissioning atoms of uranium or plutonium. Most of the fission occurred in ^{238}U in thermonuclear explosions. Hence, to a good approximation the fission product mass yield curve reported by Hallden *et al.* (1961) will apply.

The half-lives of ^{137}Cs and ^{90}Sr are, respectively, 30 and 28 years. Using the fission yields reported by Hallden *et al.* (1961), for thermonuclear-induced fission, it is estimated that approximately 21 MCi of ^{90}Sr and 34 MCi of ^{137}Cs have been introduced into the earth's atmosphere and onto the surface of the globe.

The production of other fission products can be estimated similarly, using the fission yield and half-life data tabulated by Hallden *et al.* (1961). A few specific nonfission inputs are worthy of mention, such as ^{181,185}W during Operation Hardtack* in 1958, ^{102}Rh in the high-altitude ORANGE* explosion in 1959, and ^{109}Cd and ^{113m}Cd in the STARFISH* 400-km explosion in 1962. The high-yield Soviet explosions in 1961 and 1962 produced much ^{124}Sb,

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*Operational code names; a chronology is given in Glasstone (1964).

^{54}Mn, and ^{55,59}Fe. The MIKE* thermonuclear explosion in 1952 generated considerable transuranium isotopes, including higher mass isotopes of plutonium. A sensible fraction of the debris from MIKE was deposited in the Pacific, thus providing a number of unique tracers with fairly localized input, such as ^{241}Pu and ^{242}Pu.

It appears that approximately 10^{28} atoms of ^{239}Pu (and perhaps one tenth as much ^{240}Pu) have been generated by thermonuclear explosions in the course of testing. Neptunium-237 has also been produced in quantities comparable to ^{239}Pu.

One hundred and forty megatons, or 72 percent of the total yield, were produced as airbursts, i.e., under conditions in which the fireball did not intersect the ground. Documented cases of tower-burst inputs represent a small fraction of the total—in the vicinity of 0.5 percent—and may be taken to be of trivial significance in the context of the marine environment. However, the third category, surface bursts, while representing only about 28 percent of the total, deserves special attention because of highly localized input, the majority of which was in the Marshall Islands, in the 1952 Ivy, 1954 Castle, 1956 Redwing, and 1958 Hardtack series.

Characteristics of Radioactive Debris

The characteristics of the particles formed in a nuclear explosion are determined by a large number of factors. Among the more important are the yield of the device and the quantity and kind of nearby environmental materials interacting with the nuclear explosion. The biological availability of the radioactivity in the debris particles will depend on particle size, the chemical form of the radioelement, and the matrix in which the radioactivity is imbedded or to which it is attached.

PARTICLE SIZE DISTRIBUTION

Airbursts In general, more than 90 percent of the radioactivity in airburst debris is to be found in very finely divided particles, less than 1 μ in diameter. The literature on the particle size distribution of debris produced by airbursts is sparse. Nathans suggests a log-normal size distribution with a peak diameter in the differential size distribution below 1 μ in diameter (M. Nathans, Tracerlab Inc., personal communication). Sherer proposed an exponential form of distribution for particles larger than 1 μ, based on microscopic sizing of particles of verified radioactivity, e.g., the particle size distribution as a function of its diameter is proportional to $e^{-D/b}$, where b is a parameter having a value of 1-2 μ^{-1} for nominal-size (20-kt) explosions and D is the diameter in microns (J. Sherer, Lawrence Radiation Laboratory, Livermore, Calif., personal communication).

Whatever the true form of the size distribution function, it is clear that the particle population increases rapidly with decreasing particle size to below 1 μ. There is considerable indirect evidence that particle size is related to the yield of the device, or more precisely, to the yield to mass ratio. The size distribution curve is shifted to smaller sizes as the yield increases. Since the bulk of the radioactivity in airbursts resides in submicron particles, and these are slowly acted upon by gravitational forces, the worldwide stratospheric distribution of airburst debris is relatively insensitive to the exact form of the size distribution. Very few particles in excess of 10 μ in diameter are found at any yield.

Tower Bursts In tower bursts, a considerable portion of the supporting structure will be volatilized or melted. Few quantitative data have been published that reveal details of the size distribution of tower burst events. It is clear from environmental studies accompanying tests, however, that the particle population contains particles much larger than are found in airbursts of the same yield. For example, Larson (1966) reports that significant fractions of the radioactivity are to be found in particles in excess of 44 μ. Hence, a large fraction of the radioactivity is deposited locally, and proportionately less enters the long-range–long-term atmospheric distribution system.

Coral Island Surface Bursts A number of explosions of moderately high to high yield have been conducted on coral island surfaces or on barges, sometimes in relatively shallow water. In these cases substantial quantities of coral may be melted and vaporized, and, together with the vaporized constituents of seawater, may become major radioactive debris constituents.

Data on the size distribution of radioactive debris from multimegaton explosions on coral islands are limited. The sheer magnitude of the event precludes collection of adequate samples from the prompt fallout and the residual nuclear cloud. The samples that have been retrieved and analyzed inevitably suffer from lack of definition concerning the precise fraction of the total radioactive particle population the sample represents.

Figure 1 gives a composite size distribution of radioactive particles for BRAVO as obtained from prompt fallout recorded on nearby islands. Note that the maximum observed in the vicinity of 150 μ reflects only that particles smaller than 150 μ were incompletely deposited at the collection point because of their finite gravitational settling rates.

Nathans *et al.* (1970), in studying the size distribution of cloud samples of nuclear debris of BRAVO and other coral island surface explosions, found that the results for all particles (radioactive and inert) over the range 1-100 μ fit a power law of the form $f(d)$ *a* CD^{-n}, where n may have the value of about 4. Power law distributions are rather com-

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*Operational code names; a chronology is given in Glasstone (1964).

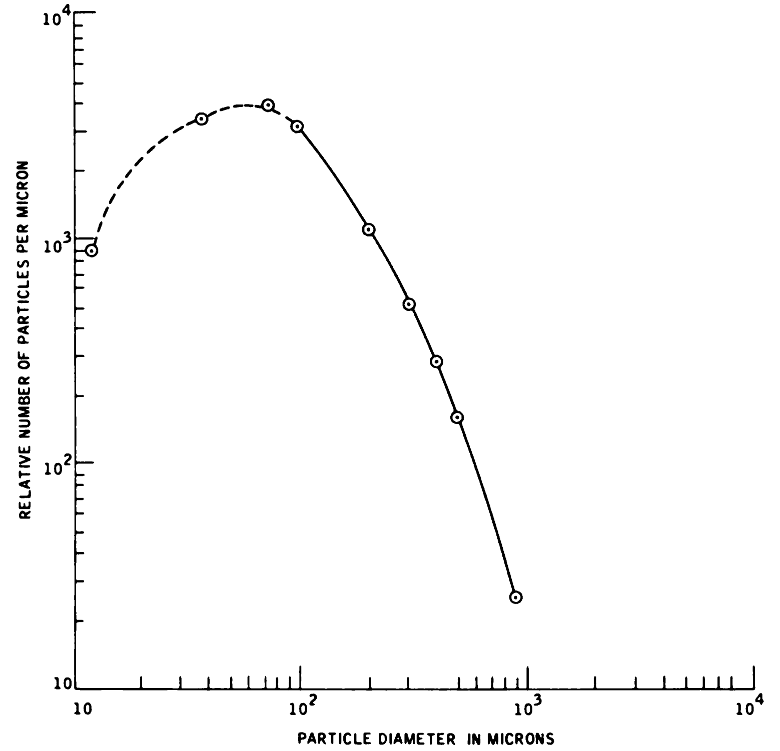

FIGURE 1 **Composite differential particle size spectrum—BRAVO.**

monly observed for natural aerosols, for zodiacal dust, and for stratospheric dust. Russell (1964) found a power law description for cloud samples of a low-yield surface explosion. Heft (1970) described a cloud from a coral surface explosion as representing the superposition of two log-normal distributions. However, because of the late time of sampling, Heft's samples, similar to Nathans', contained very little of the larger end of the particle size spectrum. In any event, there appears to be a physical basis for a multimode set of particle distribution functions describing the debris as a whole. At the larger end of the particle size spectrum, particles are formed by coalescence of smaller particles characteristic of the pre-shot soil. Melting and shock compaction are presumably partly responsible. A second phase, distributed about a smaller median diameter, is attributed to unmelted, still-crystalline debris swept into the nuclear clouds by the strong afterwinds.

ACTIVITY CONCENTRATIONS OF RADIOACTIVE DEBRIS

Radioactivity per unit mass of particle varies in both kind and amount with the size and shape of the particles of debris formed in nuclear tests. A term used to describe the radioactive content of particles is "activity concentration."* Activity of radioactive debris may be reported as atoms of radioactive species per gram of debris, as disintegrations per minute of radioisotope per gram, and so on. In this section, activity concentration is given in units of equivalent fissions per gram of debris. An equivalent fission is the number of fissile atoms (e.g., ^{235}U, ^{239}Pu, or ^{238}U) required to give the observed number of fission product atoms, or, alternatively, the number of fissioning atoms associated with a given number of product atoms of any kind.

The situation is complicated by a phenomenon called "fractionation." If the debris sample were unfractionated, i.e., representative of the true proportions in which the radioactivity was produced in the device, then equivalent fissions calculated for all isotopes in the sample will be identical. Because of differing chemical and physical properties, the elements may fractionate with respect to each other during the condensation and subsequent cooling phase in the fireball and nuclear cloud, or may fractionate by selective attachment onto pre-existing condensed materials. The elements in the chains that are gaseous at ordinary temperatures, or low-boiling, may experience sharp differentiations from elements whose metals or oxides are high-boiling. The more refractory elements or oxides (those condensing above 1500° K) will have condensed out well before 60 sec have elapsed, even for megaton explosions.

One can calculate the theoretical population of chain members in the first few minutes by using the charge division hypothesis of Glendenin *et al.* (1951), assigning probable nuclear charges based on the method of Wahl (1958), and using estimated half-lives after the method of Bolles and Ballou (1956).

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*In view of a different use and connotation of "specific activity" in subsequent chapters, in this chapter we will use "activity concentration."

The average activity concentration of airburst debris is calculable if the fission yield and mass of the device structure are known. Denoting activity concentration as S,

$$\bar{S} = 1.45 \times 10^{17} \; (W/M).$$

Here W is the yield in kilotons and M is the mass in tons. The units of $\bar{S}$ are equivalent fissions per gram.

However, because of fractionation of the radioelements during the cooling phase, no specimen of debris may correspond to this value, except by chance. Benson *et al.* (1965) have presented data that imply that the activity concentration of airburst particles over the range of a few to 15 microns in diameter is roughly constant. They found that the ^{95}Zr content of the particles is roughly related to the cube of the particle diameter. Barium-140 shows a radial dependence, roughly as D^2: 3-2.6 over this size range. These larger particles are strongly depleted in ^{140}Ba and all other volatile or semivolatile chains. Hence, the bulk of their activity is contributed by zirconium and rare-earth nuclides and a few additional refractory or semirefractory species, leading to a quasiconstant activity concentration behavior.

Typically, it is found that the fraction of the mass found in particles in excess of 2 μ will exceed by a factor of three or more the fraction of the most refractory fission product found in this size interval. Thus, in the above example, 30 percent of the mass might be found in particles larger than 2 μ associated with only 10 percent of the ^{95}Ar. This result may be attributable to poor mixing between the volatilized device materials and the fission products. It may also arise from concentration effects, insofar as they modify the condensation temperature of the higher-boiling elements or their oxides. The fission products may represent only 10^{-3} to 10^{-4} of the concentration of the structural components in the fireball vapors.

In particles smaller than 2-3 μ, however, the activity concentration increases sharply for all nuclides, as required by mass and activity balance constraints. There are few published details on this region, and the general behavior must be inferred from gross beta particle specific activities.

In the case of surface or near-surface bursts, the expanding fireball will be admixed with inert soil, some of which will have been vaporized, some melted, and some of which will remain essentially unaltered. The extent and time sequence of mixing between inert debris and fission products will be variable from event to event and would be expected to be dependent upon explosion conditions. Soils are variable in composition, and water vapor can represent a significant and undetermined component of the system, particularly in coral surface bursts.

Despite the complexity of the chemical and physical processes attending a land-surface nuclear explosion, certain consistent patterns of debris behavior are observed.

Empirical observations of land-surface nuclear debris suggest that a distinction be made among at least three classes of particles.

1. Completely volatilized material that recondenses to finely divided particles consisting of the device structural members, the fission products, and some environmental material.
2. Melted but not completely volatilized environmental material into which fission products may diffuse or onto which they attach. The melted entities may coalesce to form larger particles.
3. Unmelted environmental material, swept into the nuclear cloud by the afterwinds, to which fission products or small condensed radioactive agglomerates attach by surface impaction.

To these may be added a fourth class—spherical particles of very high activity concentration (up to 10^{16} equivalent fissions per gram) are found. These may represent coalescence of condensed vaporized soil and fission product droplets, with rapid agglomeration to larger particles. Some coalescence of this component with melted inert droplets may also occur, leading to a spherical end product.

Figure 2 depicts empirical activity concentration versus particle size data for a 15-megaton coral island explosion. This curve is drawn on the basis of radioactive particles only. It was determined by radioautography that the fraction of radioactive particles diminished rapidly with decreasing size below 44 μ. This effect is responsible for the steep slopes of both classes of chains, refractory and volatile, below 44 μ. Consequently, the absolute magnitude of the slopes of the curves below 44 μ is not to be taken too literally, since there is some uncertainty in the assessment of the number of radioactive particles. If correction is not made for radioactive particles, the activity concentration curve for chain 89 is flat over the entire range of particle sizes.

In Table 4 are assembled some activity concentration data, expressed in equivalent fissions per gram of fallout, collected near the fallout site. Data for a refractory-chain ^{99}Mo, a volatile-chain ^{89}Sr, and a chain of intermediate volatility ^{140}Ba are given. These data represent activity concentrations that would be observed if the total yield of the device were due to fission, i.e., they have been normalized to 100 percent fission yield for comparative purposes. No attempt has been made to distinguish between radioactive and nonradioactive particles in the samples, with the exception of the 15-Mt case.

An admixture of inert (nonradioactive) debris will in general represent material physically displaced by the energy of the explosion to the sampling position.

Some significant points become apparent from a study of Table 4:

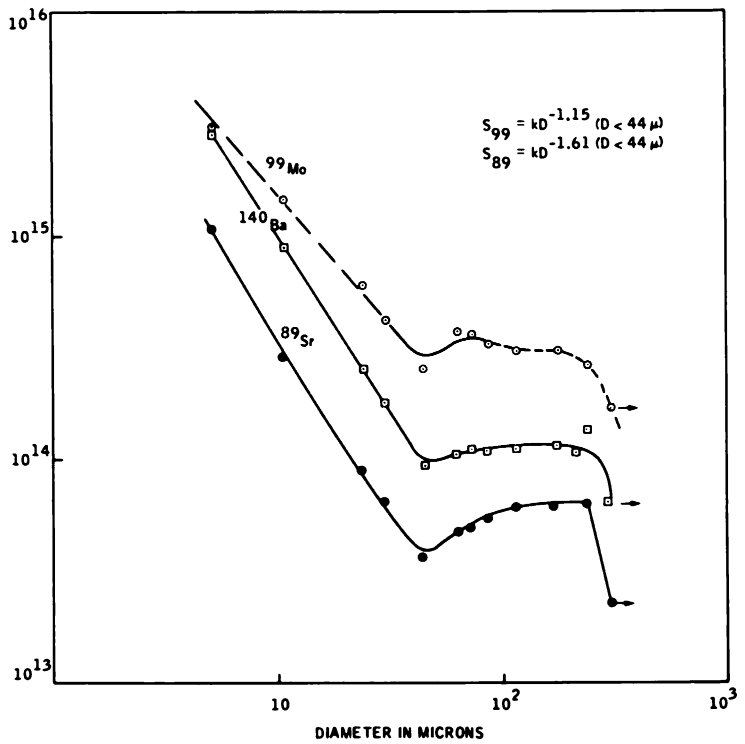

FIGURE 2 Empirical activity concentration *versus* particle size data for 15-megaton coral island explosion. S_{99}, activity concentration of chain 99; S_{89}, activity concentration of chain 89.

1. The normalized refractory-chain activity concentrations from coral surface explosions are relatively insensitive to yield, and, indeed, to particle size between about 50 μ and 250 μ in diameter. The average ^{99}F/g for the four events is 6.9×10^{14} over this range of sizes. The average deviation from the average is about 36 percent. The greatest variability is found at the large-particle end of the spectrum.

2. If columns 2 and 3 are compared, corresponding to fallout near zero and 80 km downwind, the activity concentration of the debris transported downwind is seen to be uniformly higher for all classes of isotopes. This suggests a longer opportunity for debris attachment processes to occur when the mean residence time in the cloud is lengthened. (However, absolute fallout intensities downwind will normally be lower because the number of grams of fallout deposited per unit area is smaller.)

3. Spherical particles (column 5) are uniformly higher in activity concentration for chain 99, a refractory chain, than are the irregular particles. [Tompkins and Krey (1956) report that about 25 percent of the total activity at 10 days after the explosion was attributable to spherical particles in the 5.1-Mt explosion, but only a few percent of the mass. Spherical particles were difficult to find in size fractions below 177 μ. One should not, however, infer their absence.]

4. Volatile chains show a somewhat greater variability in activity concentration from event to event than do refractory chains. The mean residence time in the cloud exerts a strong influence. (Compare 3.5 Mt near ground zero and 80 km downwind.) Also, the degree of mixing in the cloud appears to have been somewhat more effective in the 5.1-Mt case than in the other instances. Whereas spherical particles were strongly enriched in refractory chains relative to irregulars of the same size, this is not the case with volatile chains.

5. An activity concentration maximum in the vicinity of 300 μ is a recurring freature, which is most pronounced for the spherical population. Volatile chains appear to decrease in activity concentration roughly according to D^{-2} past 300 μ; the behavior is more in accord with D^{-1} for refractory chains, with the slope being slightly flatter for spheres than for irregulars. These features, if present in the 15-Mt case, were not revealed by the analysis that grouped all particles larger than 300 μ in a single fraction.

In summary, few unequivocal statements can be made concerning the activity concentration behavior of coral surface debris. However, certain of the observations have at least a partial explanation in the thermal and mixing history to which the debris was subjected.

We believe the large, high-specific-activity spheres to represent, in part, fireball condensate that coalesced to large droplets and, in part, melted soil particles that experienced no condensation history but that coalesced with high-activity-concentration droplets of condensate origin. Electron microprobe analysis of spheres from coral island explosions

TABLE 4 Activity Concentration and Particle Size, Coral Island Surface Explosions (Equivalent fissions × 10^{-14} per gram)[a]

$D_g(\mu)$	3.5 Mt (Prompt Fallout near Ground Zero)	3.5 Mt (Fallout Collected 80 km Downwind)	5.1 Mt	5.1 Mt (Spheres Only, near Ground Zero)	15 Mt (Radioactive Particles Only, Shot Atoll, Spheres plus Irregulars)	0.04 Mt (All Particles, Shot Atoll, Spheres plus Irregulars)
Chain 99 (^{99}Mo)						
57	4.8	16.0	10.2[b]	–	7.2	2.5
88	4.9	10.6	8.9[b]	–	6.6	4.0
125	5.8	9.8	8.4[b]	–	6.2	4.7
177	6.0	12.5	9.0[c]	35	6.0	5.7
297	12.4	13.2	15.2[c]	100	4.8	4.5
594	11.9	21.3	5.7[c]	68	3.4	1.6
840	3.1	24.3	4.7[c]	58	–	–
Chain 89 (^{89}Sr)						
57	0.075	0.24	0.36[b]	–	0.086	0.063
88	0.065	0.17	0.28[b]	–	0.11	0.074
125	0.046	0.19	0.24[b]	–	0.12	0.082
177	0.042	0.14	0.18[c]	0.24	0.12	0.062
297	0.043	0.12	0.22[c]	0.26	0.13	0.044
594	0.044	0.11	0.11[c]	0.063	0.046	0.063
840	0.075	0.070	0.042[c]	0.031	–	–
Chain 140 (^{140}Ba)						
57	0.32	1.28	0.67[b]	–	0.20	0.25
88	0.27	0.74	0.54[b]	–	0.22	0.28
125	0.20	0.99	0.45[b]	–	0.22	0.30
177	0.17	0.77	0.39[c]	0.48	0.23	0.23
297	0.15	0.75	0.49[c]	0.74	0.25	0.18
594	0.16	0.67	0.31[c]	0.18	0.13	0.24
840	0.031	0.41	0.12[c]	0.085	–	–

[a] Derived from Morgenthau *et al.* (1960).
[b] Irregulars plus spheres, near ground zero.
[c] Irregulars only, spheres removed, near ground zero.

show a number of instances in which spheres arose by coalescence of heterogeneous droplets (Norman and Winchell, 1967).

The $1/D^2$ behavior observed for volatile isotope chains in particles in excess of 300 μ is explicable on two grounds. Vapor phase controlled diffusion may lead to an activity proportional to the diameter of the particle for a given exposure time and hence to an activity concentration inversely as the square of the diameter. Also, larger particles are falling out of the volatile-rich cloud faster than smaller particles, leading to a decrease in activity concentration with increasing diameter. It is of some interest also that the uptake of refractory chains proceeds as $1/D$, indicating incorporation as the area for these large particles. The refractory-containing droplets are probably attached very early (in the first few minutes); vapor phase controlled diffusion probably plays no role here.

The activity concentration maximum of a volatile chain is understandable in terms of residence time in the cloud. A 100-μ particle found in the prompt fallout pattern could not have originated from so high a position in the cloud as a 300–500-μ particle and hence would have been exposed to volatile chain vapors for a shorter time.

Attempts to achieve a basic understanding of fractionation behavior as a function of explosion conditions and yield have been made by a number of workers. The thermodynamic model of Miller (1960) was a major step in placing fallout studies on a scientific basis. Norman *et al.* (1970) have carried the Miller model forward with a number of significant refinements, both conceptual and experimental. The radial distribution model of Freiling (1961, 1963) leads to considerable systemization in the treatment of debris behavior. Heft (1970) has achieved considerable success in describing fractionated systems with a nonphenomenological model that introduces a minimum number of basic assumptions.

Laboratory studies by Freiling (1970) and Adams *et al.* (1967a, 1967b) have recently focused on the kinetic aspects of the pickup of fission-product vapors by molten oxide droplets. Similar studies are under way in a number of other laboratories, using methods of high-temperature chemistry. A satisfactory physical model of debris formation processes must probably include due consideration of vaporization, condensation, and agglomerative processes. Equally important is a better understanding of the early thermal and mixing history within the ball of fire and developing nuclear cloud.

RELATIVE VOLATILITIES OF SOME FISSION PRODUCT CHAINS

It is appropriate to mention briefly fundamental chemical differences between subclasses of volatile fission product mass chains. Chain 89 is volatile mainly because of the 3.2-min ^{89}Kr; ^{89}Rb, another chain member, will behave as a volatile element during the initial stages of fireball cooling when only the most refractory elements have condensed. On the other hand, if the temperature is well below the boiling point of rubidium or its oxides, the latter will quickly attach to a debris particle surface. Similarly, the volatility of chain 131, usually measured as ^{131}I, is conferred by the relatively low boiling properties of Sn, Sb, and Te oxides. When the temperature has fallen to below 1000° C, these elements can be considered to be no longer volatile for the usual range of partial pressures.

It is apparent that radiochemical composition of the fallout must change proceeding downwind from ground zero, becoming more enriched in volatile relative to refractory chains.

The Special Case of Underwater Explosions

Only an insignificant fraction (less than 0.1 percent) of the total megaton equivalents of fission detonations (see Table 3) has resulted from underwater bursts. These events have the unique characteristic of being point source injections. The initial oceanic distribution is a function of the yield and depth of detonation. Based on limited test experience in oceanic situations, for shallow explosions, from one third to two thirds of the debris may be found in the mixed layer in a pool many hundreds of meters in radius and mixed to the existing thermocline (approximately 100 m). The history of such an injection as it was influenced by currents, turbulent diffusion, and decay, is depicted in Figure 3 as measured from an aircraft and in Figure 4 as measured by an *in situ* gamma probe (Riel, 1962).

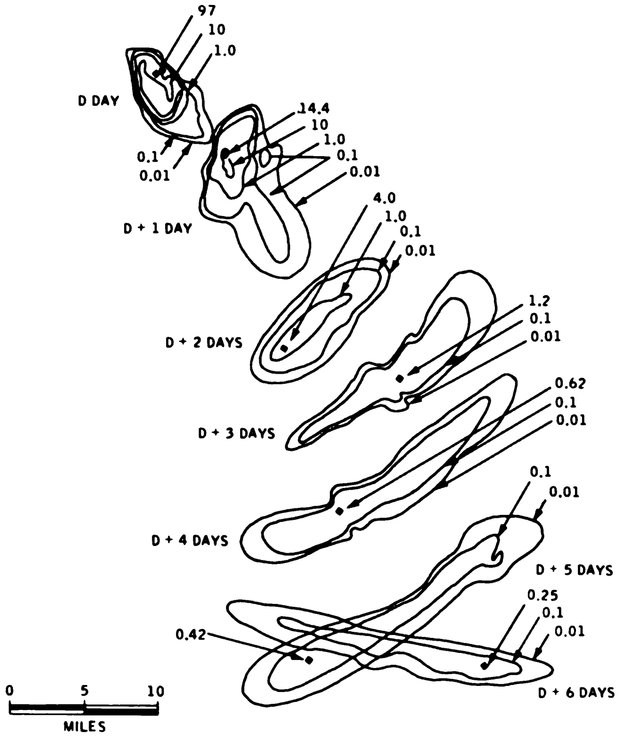

FIGURE 3 Surface pattern of distribution of radioactivity from a nominal-yield underwater explosion as measured from an aircraft at 500-ft (150-m) altitude. (Gamma contour values in mR/hr.)

Debris from deep explosions deposited in the thermocline layer will generally react to the same environmental influences; however, since the subsurface scale of turbulence is small, the predominant activity reduction mechanism is radioactive decay. Discrete lamina of radioactivity were observed in the thermocline layer after the WIGWAM test.* The fraction of the debris in these waters, as reported by Isaacs (1962), was estimated to be approximately two thirds of the total debris released by the explosion. Advection and diffusion of radioactivity in the ocean is discussed in detail in Chapter 4.

Fractionation of fission products created in underwater detonations is not severe (Freiling and Ballou, 1962). Individual fission product data from samples collected shortly after detonations indicated a variation of no more than 30 percent in predicted fission product ratios. An exception was ^{89}Sr from a deep burst, which varied by a factor of up to about 10 in relation to other fission product radionuclides.

*A 30-kt device detonated at a depth of 2,000 ft (610 m) in the Pacific at 29° N, 126° W, in 1955.

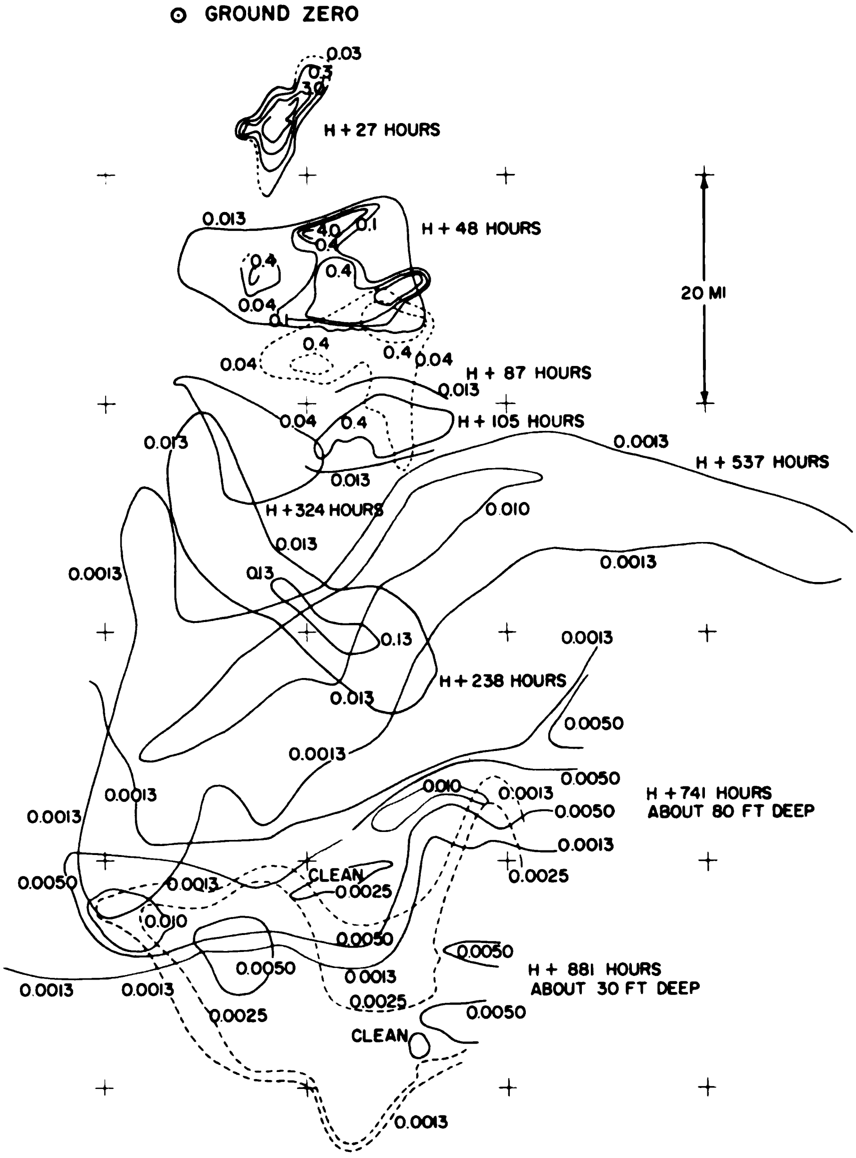

FIGURE 4 Subsurface pattern of distribution of radioactivity from a nominal-yield underwater explosion as measured by an *in situ* gamma probe (some lines are dashed for clarity of presentation). (Reprinted from Riel, 1962).

Nuclear Explosion Debris and Its Interaction with Seawater

The physical and chemical states of bomb debris in seawater are poorly understood. Some theoretical estimates have been made, a few determinations from actual debris have been reported, and some laboratory studies using stable element counterparts of detonation debris have been undertaken (Freiling and Ballou, 1962; Greendale and Ballou, 1954).

Radionuclide physicochemical states are largely determined by their state at the time of addition to the ocean (or by the initial state and the decay of their precursors) and by the effects of their chemical environment.

Little is known of the fundamental processes of formation of nuclear debris from an underwater explosion. In the absence of large quantities of bottom material, the initial processes can be visualized as forming debris consisting of vaporized and dissociated water, seawater salts, and device materials. For a 10-kt device detonated underwater, on the order of 7×10^6 kg of seawater will be vaporized. The bubble of debris will then contain, in addition to the vaporized device materials and the 7×10^6 kg of H_2O, 1.3×10^5 kg of chlorine, 7.7×10^4 kg of sodium and potassium, 1.1×10^4 kg of calcium and magnesium, 6.3×10^3 kg of sulfur, and less than 7×10^2 kg of any other seawater constituent. As this system cools, nucleation of the debris should be governed by iron, calcium, and magnesium oxides. The initial nuclei would therefore be expected to consist of particles of calcium, magnesium, and iron oxides of less than 20 μ diameter. These would contain the refractory-like radionuclides. Upon further cooling, these, in turn, would serve as nuclei for deposition of more volatile radionuclides.

The release of fission products to seawater from nuclear debris depends primarily upon particle solubility and leaching properties. It has been established that the ease of leaching gross beta activity from fallout particles increases in the following order: tower-shot debris, silicate-burst debris, airburst debris. Some authors report that the solubility increases as the size of the debris decreases. The debris from coral surface bursts is reported to be highly soluble in seawater (Adams *et al.*, 1960).

For underwater explosions, release to the seawater would include late-condensing soluble fission products and colloidal particles bearing most of the activity. Due to the large surface-to-volume ratio of small particles, leaching of soluble radionuclides would occur with high efficiency, leaving the insoluble radionuclides in the colloidal state. It has been shown that the halides, alkali metals, and alkaline earth metals will generally occur as soluble species in their normal oxidation states. Certain other elements, such as B, P, Ce, As, Se, Mo, Tc, and Te, are expected to occur as oxygenated anions that would be found partially in solution and partially colloidal or particulate. Nearly all of the remaining elements may be expected to exist as oxides or hydrated oxides associated with colloidal or gross particulate matter. However, our understanding of the interaction of radionuclides in such a complex mixture of inorganic compounds and organic and biological material is far from complete.

As a result of limited experiments, Freiling and Ballou (1962) summarized the nature of underwater nuclear explosion debris in seawater. They found that the total activity in the soluble phase increased from 35 percent at one day after the event to approximately 60 percent after two weeks. This increase was attributed to the slow conversion of some of the constituent elements into soluble material, rather than to the production of new elements through radioactive decay.

The percentages of soluble and insoluble fractions of a number of radionuclides were also determined by Ballou (1963), as shown in Table 5. These data generally indicate greater solubility than that earlier reported by Greendale and Ballou (1954).

Similar investigations were carried out with waterborne debris from an underwater burst in which the fireball contacted the bottom of a coral lagoon. These data, after Ballou (1963), are reported in Table 6. Particle sizes ranged from 0.001 μ to 10 μ in diameter. The distribution of radionuclides in surface water was collected 2 hr after detonation and analyzed 31 hr after detonation. "Solid phase" refers to material remaining in the ultrafiltrate. Standard deviations were calculated from duplicate analyses. Deviation of sums from unity indicates a lack of mass balance or is an indication of sample contamination.

For the data in Table 6, radiochemical analysis for selected radionuclides was performed on duplicate aliquots of the gross samples, ultrafiltrates, and dissolved membranes. Results from successively separated fractions did not reveal any change in the physical state distribution of any radionuclide with time, but mass balance deficiencies prohibit firm conclusions in some cases. The results from successive ultrafiltrations have therefore been averaged. The results for each radionuclide discussed below are compared with the results of the two previous investigations. Included in each discussion is the mass balance achieved and the effect of an

TABLE 5 Percentages of Activity of Radionuclides in Ultrafiltrates from Surface Water Samples, Underwater Burst

Sample	Centrifugation Time (hr from zero time)	Relative Activity in Ultrafiltrate at Centrifugation Time (%)						
		^{95}Zr	^{99}Mo	^{239}Np	^{237}U	RE[a]	^{137}Te	^{140}Ba
1	1	35	64	–	54	33	0	35
	7	–	–	42	–	–	–	–
	19	43	66	44	80	19	0	–
2	7	–	–	41	–	–	–	–
	19	–	–	53	–	–	–	–
3	10	48	67	67	77	42	0	–
	19	–	–	52	–	–	–	–
	116	43	63	–	71	39	0	–
Average		42	65	50	71	34		35

[a] Gross rare earths and yttrium.

TABLE 6 Radionuclide Distributions 31 hr after Detonation in Lagoon Burst; H + 2 Surface Water Sample

	Fraction Distribution		
Radionuclide	Solid Phase	Colloidal Phase	Soluble Phase
^{89}Sr	0.00 ± 0.00	0.11 ± 0.01	0.99 ± 0.00
^{95}Zr	0.38 ± 0.01	0.16	0.03 ± 0.00
^{95}Nb	0.45 ± 0.00	0.39 ± 0.01	0.00 ± 0.00
^{99}Mo	0.28 ± 0.00	0.06 ± 0.01	0.60 ± 0.00
^{103}Ru	0.60	0.25 ± 0.00	–
^{132}Te	0.60 ± 0.00	0.18 ± 0.00	–
^{140}Ba	0.00 ± 0.00	0.01 ± 0.01	0.99 ± 0.01
TRE[a]	0.83 ± 0.02	0.14 ± 0.06	0.03 ± 0.00
^{237}U	0.02 ± 0.00	0.11 ± 0.01	0.04 ± 0.06
^{239}Np	0.47 ± 0.00	0.46 ± 0.01	0.02 ± 0.00

[a] Total rare earths.

additional membrane on the physical state distribution and on the mass balance observed. Mass balance deficiency may be taken as a measure of contamination potential, since it presumably arises from adherence of the radioactive species to the walls of the polyethylene container or to the surfaces of the ultrafilter apparatus.

^{89}Sr An average value of 4 ± 1 percent of the ^{89}Sr was found to be in the insoluble phase, and an average mass balance of 116 ± 5 percent was obtained. The results show a somewhat greater solubility than that found in underwater arcing experiments, but not outside the precision of the methods. The results were unaffected by the presence of a second membrane.

^{95}Zr and ^{95}Nb Analyses were performed for both of these radionuclides, and no distinction between their behavior was evident. The presence of a second membrane increased both the mass balance and the percentage of activity appearing in the colloidal fraction. With either a single or double membrane, 0–8 percent of the activity was found in the soluble state, in fair agreement with underwater arcing results. With only a single membrane, an average of 48 percent was found to be insoluble, but the results were spread from 34 to 66 percent. The results from the deep underwater burst are within this range.

^{99}Mo Mass balance was consistently good, averaging 102 ± 5 percent. Using a single membrane, an average value of 68 ± 5 percent was found to be soluble, in good agreement with the deep underwater burst results. With a double membrane, 26 ± 5 percent was found to be soluble, in agreement with underwater arcing results.

^{103}Ru It was found that 82 ± 5 percent of the ruthenium was insoluble, the mass balance being 105 ± 6 percent. No double-membrane effect was noted. The solubility is therefore greater than indicated by the value of 1 percent from the underwater arcing experiments.

^{132}Te The insoluble fraction was found to contain 64 ± 9 percent of the activity and the mass balance was 81 ± 12 percent. No double-membrane effect was noted. These results are in better agreement with those from underwater arcing than with those from the deeper underwater burst.

^{140}Ba This radionuclide showed a much higher solubility than the single determination made for the deep underwater burst would indicate. Only 5 ± 1 percent was found in the insoluble fraction, the mass balance being 88 ± 4 percent. No double-membrane effect was noted. The results are similar to those obtained for ^{89}Sr.

Total rare earths The insoluble fraction contained 67 ± 3 percent of the activity, in accord with the deep underwater burst results, but the mass balance was only 71 ± 3 percent. No double-membrane effect was noted. Cerium-141, 144 was found to behave similarly. This behavior is not inconsistent with the results from the underwater arcing experiment, in which it was found to be soluble to the extent of 3 percent or less.

^{237}U The insoluble fraction contained 46 ± 5 percent of the activity, but the percentage of activity found in the soluble fraction was quite variable. The deep underwater burst results also indicate considerable variability. No double-membrane effect was noted.

^{239}Np With single membranes, 50 ± 2 percent of the activity was found to be insoluble, in agreement with the deep underwater burst results. With a double membrane, however, 93 percent was found to be insoluble. As in the case of ^{237}U, the percentage of activity found in the soluble fraction was quite variable.

Radioactive Fallout from Nuclear Weapons Tests

The specific character of the oceanic contamination from nuclear explosions depends very directly upon the nature, size, and physical location of the event. Thus, underwater bursts introduce radioactive contamination directly and immediately into the sea in a relatively small and well-defined area. Surface or air detonations, depending upon the size of the burst, distribute the debris in the atmosphere to varying altitudes and horizontal distances. The extent of these atmospheric dispersions directly influences the rate of fallout of the radioactive material and its ultimate distribution on the earth's surface.

The bulk of the nuclear debris from large surface or air bursts enters the stratosphere. Once in the stratosphere, this material is, for practical purposes, insulated from the earth's weather, and the rate and extent of its distribution are determined by meteorological processes in the stratosphere. In general, the stratospheric mixing rate is more rapid within the hemisphere of injection than either the fallout rate or the rate of intrahemispheric mixing. These factors result in a generally characteristic pattern of distribution of the radioactive aerosol within that hemisphere.

Removal of radionuclides from the stratosphere occurs by downward mixing and diffusion, with the major entry into the troposphere occurring in the middle and upper latitudes during late winter and early spring, in both hemispheres. The rate of stratospheric depletion of nuclear debris depends, to a degree, upon the latitude of the injection and the altitude of stabilization of the particles.

This rate, termed the "stratospheric half residence time" has been empirically observed to resemble a simple exponential decay. The minimum rate appears to be about 6 months, for nuclear tests in the lower stratosphere in polar regions. Debris injected into very high altitudes may take 2 years or longer before reaching the ground. It is interesting, however, that since the partial test ban treaty went into effect at the end of 1962, the stratospheric half residence time has remained quite constant at about 10 months.

The rate of intrahemispheric mixing in the stratosphere is slow, compared to the average observed stratospheric fallout rate; hence, the bulk of the nuclear debris remains within the hemisphere of introduction. Upon leaving the stratosphere, the debris is deposited on the earth's surface primarily as a direct result of precipitation and with a "tropospheric half residence time" of approximately 30 days or less.

The seasonal release of stratospheric debris into the troposphere, the probable latitudinal distribution of this release, and the indicated precipitation patterns on the ground combine to produce the observed spring maximum in fallout and global distribution with higher values in the mid-latitudes. Figure 5 illustrates a typical latitudinal distribution of stratospheric fallout.

Of the many radionuclides produced in the testing of nuclear weapons, ^{90}Sr has been examined the most intensively and extensively because its relatively high fission yield (~3.5 atoms per 100 fissions) and its bone-seeking character and long physical half-life make it potentially the greatest fallout hazard to man. Measurements of ^{90}Sr in a variety of environmental and biological media, including human specimens, have been made since the beginning of weapons testing. Atmospheric measurements coupled with the direct assay of ^{90}Sr in soil have made possible the assembly of a ^{90}Sr inventory. Interestingly enough, the inventory of ^{90}Sr in the atmosphere (mainly in the stratosphere) plus that present on the surface matches quite closely the total production of ^{90}Sr calculated from weapons testing to date. This does not imply detailed knowledge of the precise ^{90}Sr yield, or indeed the explosive yield, of each and every detonation, but it does apply to the time average of such events. Hence, we may conclude that we know the production and distribution (on a broad scale) of this particular radionuclide. The annual production and rate of deposition of ^{90}Sr are indicated in Table 7.

Table 8 lists the annual deposition of ^{90}Sr by 10° latitude bands for the period 1958-1966. These values were computed from the data of the worldwide sampling network and are based upon the assumption that there is no systematic difference in fallout efficiency between ocean and land surfaces. With that restriction, the information in Table 8 may be utilized as a direct measure of input to the oceans as a function of time.

Table 8 may also be used to derive stratospheric deposition values for other fission product nuclides on the further assumption that the ratio between such nuclides and ^{90}Sr is constant. There is substantial evidence that this can be assumed with some confidence for ^{137}Cs; hence, the ^{90}Sr values of Table 8 can be converted to ^{137}Cs values by multiplying by the constant 1.5. Calculation of amounts of other relatively long-lived fission products, such as ^{144}Ce and ^{95}Zr, from ^{90}Sr ratios should be used with caution and must be

TABLE 7 Worldwide Production and Deposition of ^{90}Sr (in MCi)

Year	^{90}Sr Produced	^{90}Sr Deposited
1945–1958	9.1	5.6
1959	0	1.1
1960	0	0.4
1961	2.5	0.4
1962	7.6	1.6
1963	0	2.6
1964	0	1.9
1965	0	1.0
1966	0	0.4

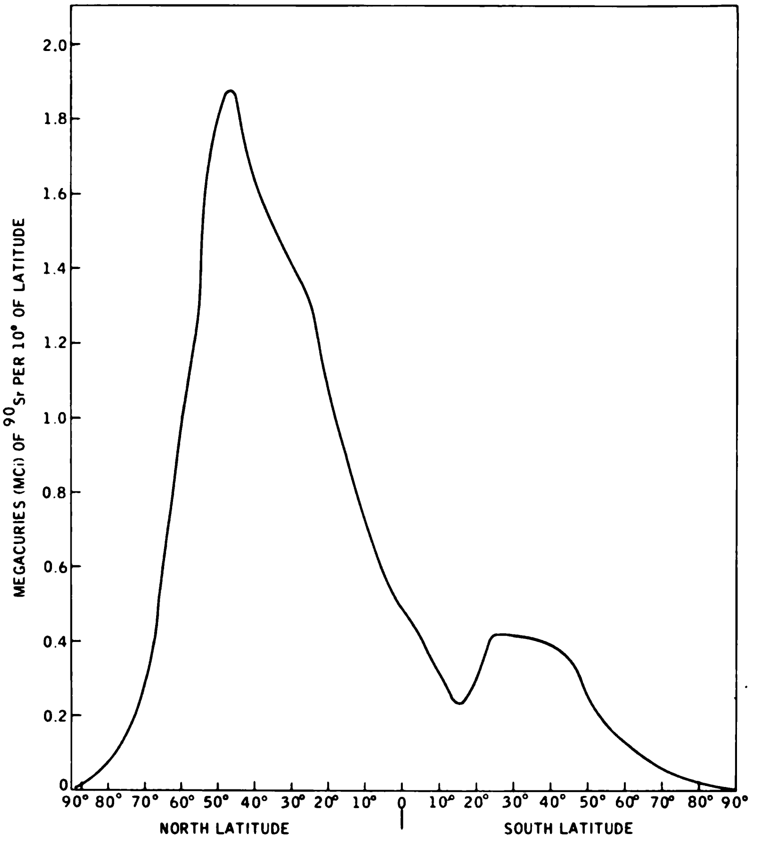

FIGURE 5 Latitudinal distribution of ^{90}Sr fallout (1958–1967).

corrected for radioactive decay, since their half-lives are relatively short.

A calculation of the distribution of ^{55}Fe produced as a product of activation by neutrons presumably on materials of the nuclear devices also was made based upon the ^{55}Fe/^{90}Sr ratio. A very substantial amount of this nuclide was made and injected into the stratosphere during the U.S. and Soviet test series in 1961 and 1962. The fallout data for ^{55}Fe, though sparse, suggest that it had apparently become rather well mixed with the ^{90}Sr in the stratosphere by 1963. On this basis, an average ratio of ^{55}Fe to ^{90}Sr of 9.4 was derived and used for the compilation of Table 9.

Future Applications of Nuclear Explosives

Some potential peaceful applications of nuclear explosives constitute a potential source for the introduction of radionuclides into the marine environment. The proposed applications that could contribute the most radioactivity to the oceans involve the use of nuclear explosives for the construction of harbors or canals.

The U.S. Atomic Energy Commission's Plowshare Program for developing potential uses for nuclear explosives is in its early developmental stages. While some experiments have been conducted in terrestrial situations, none has been attempted in the marine environment. Guidelines for the employment of nuclear explosives and for the design of the explosives themselves are still being developed and refined.

Environmental contamination resulting from nuclear excavations would differ significantly in at least three ways from that resulting from atmospheric weapons tests. First, most of the radioactivity produced would be retained in the broken rock that falls back into the crater and in the ejecta in the immediate vicinity of the detonation site, and any portion that moves from the detonation site will do so only by surface runoff or groundwater transport. Second, the radioactivity produced per megaton of explosive force will be substantially reduced by employing explosives with low fission yields, by using nonactivating neutron absorbers to reduce the neutron activation of components of the explosive and of the soil, and by using explosive components whose activation products are of low biological significance. Finally, because low-fission-yield devices will be employed, activation products will equal or surpass fission products in curies produced and in biological significance. In this respect, however, it is important to note that, of the radionuclides thus far introduced into marine or freshwater environments, the greatest potential hazard to man has involved

TABLE 8 Annual Deposition of ^{90}Sr Since 1958, by 10° Bands of Latitude (in MCi)

Latitude	1958	1959	1960	1961	1962	1963	1964	1965	1966	Total
80°-90° N	0.003	0.002	0.001	0.001	0.002	0.007	0.002	0.001	0	0.019
70°-80°	0.025	0.016	0.006	0.006	0.017	0.047	0.014	0.008	0.003	0.142
60°-70°	0.064	0.059	0.011	0.020	0.089	0.200	0.087	0.030	0.012	0.572
50°-60°	0.114	0.157	0.031	0.044	0.168	0.376	0.262	0.109	0.042	1.293
40°-50°	0.153	0.206	0.043	0.065	0.285	0.540	0.354	0.149	0.056	1.851
30°-40°	0.147	0.218	0.043	0.064	0.247	0.358	0.248	0.137	0.051	1.513
20°-30°	0.106	0.192	0.038	0.045	0.214	0.358	0.239	0.091	0.034	1.317
10°-20°	0.024	0.104	0.035	0.035	0.145	0.250	0.180	0.094	0.036	0.903
0°-10°	0.019	0.021	0.025	0.031	0.113	0.184	0.093	0.069	0.026	0.581
0°-10° S	0.044	0.034	0.024	0.021	0.096	0.063	0.073	0.039	0.015	0.409
10°-20°	0.041	0.023	0.018	0.017	0.028	0.033	0.036	0.028	0.011	0.235
20°-30°	0.055	0.031	0.027	0.044	0.048	0.056	0.068	0.062	0.024	0.415
30°-40°	0.067	0.032	0.022	0.033	0.032	0.050	0.075	0.068	0.026	0.405
40°-50°	0.030	0.022	0.028	0.024	0.036	0.049	0.076	0.071	0.027	0.363
50°-60°	0.018	0.015	0.017	0.015	0.017	0.023	0.032	0.026	0.010	0.173
60°-70°	0.010	0.007	0.008	0.008	0.009	0.012	0.016	0.012	0.005	0.087
70°-80°	0.004	0.002	0.003	0.003	0.003	0.004	0.005	0.005	0.002	0.031
80°-90°	0	0	0	0	0	0	0.001	0.001	0	0.002
Total	0.924	1.141	0.380	0.476	1.549	2.610	1.851	1.000	0.380	10.311

TABLE 9 Deposition of ^{55}Fe from Weapons Tests, by 10° Bands of Latitude (in MCi of ^{55}Fe, decay corrected to October 15, 1961)

Latitude	1962	1963	1964	1965	1966	Future[a]	Totals
80°-90° N	0.01	0.06	0.01	0.01	0	0	0.09
70°-80°	0.08	0.44	0.13	0.07	0.03	0.01	0.76
60°-70°	0.42	1.88	0.82	0.28	0.11	0.04	3.55
50°-60°	0.79	3.54	2.36	1.02	0.39	0.16	8.26
40°-50°	1.34	5.10	3.32	1.40	0.53	0.20	11.89
30°-40°	1.16	3.36	2.32	1.29	0.48	0.18	8.79
20°-30°	1.00	3.36	2.33	0.85	0.32	0.12	7.88
10°-20°	0.68	2.34	1.68	0.89	0.34	0.13	6.06
0°-10°	0.53	1.72	0.87	0.65	0.25	0.09	4.11
Total, northern hemisphere	6.01	21.80	13.74	6.46	2.45	0.93	51.39
0°-10° S	0.45	0.59	0.68	0.36	0.14	0.09	2.31
10°-20°	0.13	0.31	0.34	0.26	0.10	0.13	1.27
20°-30°	0.22	0.52	0.64	0.59	0.23	0.12	2.32
30°-40°	0.15	0.47	0.70	0.64	0.25	0.18	2.39
40°-50°	0.17	0.46	0.72	0.66	0.25	0.20	2.46
50°-60°	0.08	0.21	0.30	0.25	0.10	0.16	1.10
60°-70°	0.04	0.11	0.15	0.12	0.05	0.04	0.51
70°-80°	0.01	0.04	0.05	0.05	0.02	0.01	0.18
80°-90°	0	0	0.01	0.01	0	0	0.02
Total, southern hemisphere	1.25	2.71	3.59	2.94	1.14	0.93	12.56
Total, world	7.26	24.51	17.33	9.40	3.59	1.86	63.95

[a] Based on Stardust 0.2 MCi ^{90}Sr from pre-1963 tests. This material was evenly distributed over the northern and southern hemispheres; thus, fallout is assumed symmetrical. The ratio of ^{55}Fe to ^{90}Sr changes with time; using a constant ratio of 9.4 for the period 1962-1966 introduces an error.

activation products such as ^{32}P, ^{65}Zn, and ^{55}Fe. In underground explosions, ^{3}H contamination of groundwater may be a biological hazard, depending on the quantity produced.

There are three sources of the radionuclides produced in a thermonuclear cratering explosion: fission products, radionuclides induced in the device materials, and radionuclides induced in the environmental media surrounding the device. In addition, between 7×10^6 and 5×10^7 Ci of tritium, depending upon the nature of the thermonuclear reaction, may be produced.

In a thermonuclear explosion, roughly 10^{27} neutrons will be produced per megaton of yield. In addition, a large number of protons, deuterons, tritons, helium-e nucleii, and alpha-emitting radionuclides will be generated. These neutrons and charged particles will react with the materials of the device to produce a wide variety of radionuclides.

Estimation of the activation radionuclides produced in a nuclear event is difficult because many of the cross-section values necessary for the calculations are unknown. However, using the data that are known, such as Ng's calculations of the activation products in major rock types (1965), and using worst-case assumptions where needed, estimates of the potential biological hazard of a device can be made (Kaye *et al.*, 1969; Tamplin, 1967; Ng and Thompson, 1966; Burton and Pratt, 1967; Tamplin *et al.*, 1968; Martin, 1969).

When an explosive is detonated on a tower, all of the radioactivity produced is released to the atmosphere, but when the explosive is detonated underground, only a fraction of the activity is released to the atmosphere. When the explosive is buried deep enough, all of the activity remains underground. In cratering experiments, in which the expanding cavity ruptures the surface to form the crater, the material above the explosive acts like a filter bed and removes a large portion of the radioactive material that would otherwise be released to the atmosphere. The efficiency with which a radionuclide is removed depends on its chemical and physical properties or on those of its precursors during the venting process. Fission products such as ^{137}Cs and ^{90}Sr, which have rare-gas precursors during the venting process, are found in much higher relative concentrations in the cloud than are refractory radionuclides that do not have gaseous precursors, such as ^{95}Zr or ^{147}Nd. Therefore, to the extent that it is possible, the fraction released to the atmosphere must be determined for each specific radionuclide of interest.

The final physical and chemical forms of the radionuclides produced in a nuclear cratering explosion are poorly known, except for a few species. The final forms depend, as in the case of fractionation, upon the chemical nature of the species present during cavity expansion and venting. The chemical species present depend upon such factors as the water content of the medium and the overall oxidation-reduction potentials of the mixture. Large quantities of rock are melted and vaporized along with the materials of the device itself. As the cavity expands, the vaporized materials begin to condense, the order of condensation depending upon the volatilities of the materials. It is possible to alter this overall process by placing in the immediate vicinity of the device materials that could, for example, change the overall oxidation-reduction potential of the mixture and, therefore, the chemical species present. Whether such a procedure should be considered, of course, depends upon the associated biological hazard.

The U.S. Atomic Energy Commission (1967c) has released the following information concerning the radioactivity released to the atmosphere by cratering explosives:

> In order to plan for major excavation projects, the following factors relative to release of radioactive debris should be taken into account: The amount of radioactivity airborne in the cloud and in the fallout is minimized by scavenging during the venting process, by special emplacement techniques, by utilizing minimum fission explosives, and by employing extensive neutron shielding. Based on reasonable assumptions about these factors, the following information can be used in planning for cratering events of useful magnitude. For each individual nuclear explosive detonated, the sum of fission products airborne in the radioactive cloud and in the fallout can be expected to be as low as the equivalent of 20 tons. The tritium release may be less than 20 kilocuries per kiloton of total yield. The sum of activation products airborne in the radioactive cloud and in the fallout may be expected to be as low as the amounts shown in the following table:

Representative Set of Induced Radioactivities at Detonation Time (Total in Cloud and Fallout)

	Nuclide Production, Kilocurie for Yield of		
Nuclide	100 KT	1 MT	10 MT
^{24}Na	200	800	2,000
^{32}P	0.1	0.4	0.8
^{45}Ca	0.01	0.03	0.06
^{54}Mn	0.1	0.3	0.7
^{56}Mn	6,000	20,000	50,000
^{55}Fe	0.04	0.15	0.3
^{59}Fe	0.04	0.15	0.3
^{185}W	6	10	14
^{187}W	300	500	700
^{203}Pb	1,000	7,000	20,000
Other	15	20	40

NUCLEAR REACTORS

Nuclear reactors have been designed and built for a variety of purposes, as indicated by Table 1. Each purpose involves a design concept keyed to the desired form of energy output, which in turn depends on the intended application. Reactors are designed to produce power, neutrons, radioactive isotopes, and fissionable material. Each design presents different operating characteristics and could also influence the amount and type of wastes to be disposed of.

All present nuclear reactors are designed to induce fission and to sustain and control the chain reaction. Future nuclear power sources may include fusion when it is learned how to sustain and control temperatures of about a million degrees. The principle of operation of various fission reactors is the same in all reactors, but for different reactor concepts the major variants are the composition and construction of the fuel rods, neutron reflectors and moderators, control rod systems, and heat exchange materials and methods. Of special interest is the construction of fuel rods and the manner of their use, because radioactivity is created in and adjacent to the fuel rods.* Typically, fuel rods or elements are fabricated of fissionable uranium in the form of plates or pins that are completely encased (cladded) by a corrosion-resistant metal alloy such as stainless steel or zirconium alloy (zircaloy). The fuel elements are assembled into a cluster, forming the core of the reactor. The core is immersed in or bathed by a coolant fluid. In the United States, most reactors used for making electricity use isotopically light water that also acts as a neutron moderator. In the United Kingdom and other nations, nuclear electric-generating reactors having graphite moderation and gas cooling, generally air, are more common. Our discussion, however, will be concerned mainly with U.S. practices.

In water-cooled reactors, the core and coolant are contained in a vessel called the reactor vessel, usually of heavy steel construction. Heat is generated by fission of the fuel and is conducted through the walls of the cladding to the moderator-coolant. The size of a reactor is measured by its sustained thermal output, usually in kilowatts (kW_{th}).

Products created by the fission of uranium are retained within the cladding of the fuel elements unless there is a hole through which they can leak to the coolant. However, some neutrons do escape from the core to interact with impurities in the coolant and with the reactor vessel and other nearby materials. The neutron activation products in the coolant are the major source of radioactive wastes in reactor operations.

Nuclear reactors are generally housed in airtight steeel or concrete shells designed to contain airborne contamination from either normal or abnormal reactor operating conditions. The sites on which reactors are located are usually selected to restrict the exposure of nearby populations to radioactivity that may be released in a reactor accident.†

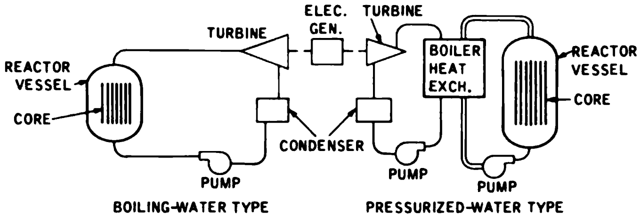

FIGURE 6 Schematic nuclear electric power reactors.

Nuclear Electric Power Reactors

Two systems have been devised for recovering the heat from nuclear reactors in order to generate steam and electricity. One, the boiling-water reactor, is direct; the other, the pressurized-water reactor, is indirect. These systems are illustrated diagramatically in Figure 6.

In the United States, all of the central-station electric power nuclear reactors are either of the boiling-water or the pressurized-water types. Power reactors exported by the United States are also of these types. These contrast with the natural-uranium-fueled, gas-cooled power reactors of the United Kingdom and France. As in the United States, foreign nations project increasing use of nuclear reactors for electric power generation (Table 10).

Thermal electric generating plants discharge some heat via their cooling systems because they are not 100 percent efficient in converting heat to electricity. Modern fossil-fueled generating plants have a thermal efficiency of about 38 percent, while nuclear plants are about 32 percent efficient. Future reactor plants, on the average, will have substantially larger generating capacities than those of today, and they will, therefore, require even more coolant flow. Some of this thermal capacity of large nuclear plants will be put to work in desalting seawater in dual-purpose plants.
It is expected that, as more generating capacity is installed, more plants will be constructed in coastal locations, where coolant water supply is essentially unlimited. This is already being done in the United Kingdom.

Nuclear power stations produce radioactive wastes in gaseous, liquid, and solid forms. Most of the fission products created within the fuel elements remain in the fuel elements until they are removed in chemical processing plants for recovery of the unused fuel.

A recent survey by Blomeke and Harrington (1968) of up to 7 years of experience in managing radioactive wastes at six operating nuclear power generating stations revealed that most of the radioisotopes present in the waste gases, liquids, and solids from power reactors originated in the primary reactor coolant. The isotopes appeared in the wastes as by-products of repurification of the coolant from the activation of corrosion products from chemical additives, from natural impurities in the water and even from the water it-

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*This discussion is concerned only with heterogeneous nuclear reactors; homogeneous reactors, in which the fuel is in a liquid or fluid state, are not yet beyond the conceptual research stage.

†There have been two known reactor accidents–"Windscale" and "SL-1"–and a preplanned reactor excursion–"Boxax." These are described by Dunster *et al.* (1958), U.S. Atomic Energy Commission (1961), and Kramer (1958).

TABLE 10 Nuclear Power Plants in Operation, under Construction, or Planned for the Near Future[a]

Country	Through 1967		1967-1973	
	No. of Plants	Capacity (MW_e)	No. of Plants	Capacity (MW_e)
Belgium	1	10	2	1456
Canada	2	220	5	2282
France	8	1177	5	2885
W. Germany	4	317	8	1891
India	–	–	5	980
Italy	3	607	2	685
Japan	2	165	6	2662
Netherlands	–	–	1	47
Pakistan	–	–	1	125
Spain	–	–	3	1073
Sweden	1	9	2	593
Switzerland	1	7	4	1756
United Kingdom	27	4125	11	6390
United States	14	2782[b]	61	46,941[c]

[a] After U.S. Atomic Energy Commission (1967b).
[b] U.S. Atomic Energy Commission (1967c).
[c] 41 having a combined generating capacity of 14,705 MW_e are under construction; projections are through 1974.

self, and from fission products that may leak into the coolant through defective cladding. Evidence indicated that some fission-product tritium may have diffused through stainless steel fuel cladding.

Gases are removed from the primary coolant stream in a variety of ways. In boiling-water reactors, over 99 percent of the radiolytic and fission gases are removed continuously by passing large volumes of air through the steam turbine air ejector. Short-lived gaseous isotopes are decayed by a 20- to 30-min delay before release. In pressurized-water reactors, gases are normally removed only when the coolant is withdrawn; some may leak out of the pressurized system. Pressurized-water reactors are provided with gas storage tanks, usually enough to allow several weeks' decay before release, if needed.

The primary coolant is treated by demineralization to keep the conductivity of the reactor water below 0.5 μmho/cm. This reduces corrosion rates within the system and holds down radiation levels in the piping system outside the core. In pressurized-water reactors, corrosion may be further inhibited by hydrogen overpressure and by addition of hydrazine, ammonia, or lithium hydroxide. Suspended solids in the coolant stream are removed in part by filters and by the demineralizer beds; depending on the age of the system, suspended solids are also deposited on surfaces throughout the primary system and occasionally may be "bumped" loose.

According to Blomeke and Harrington (1968), other sources and kinds of wastes associated with reactor operations are generally of routine importance, but under unusual circumstances they can be significant problems. Liquid wastes include the large volumes of water used for shielding during refueling operations and the wastes from decontamination operations, the laundry, the analytical laboratory, and the fuel storage pool; solid wastes—from replaced reactor components and instruments to contaminated tools, laboratory ware, laundry, removable floor coverings, and decontamination materials such as paper (wipes) and rags—have a large radioactivity range.

RADIOACTIVITY CHARACTERISTICS

Radionuclides in the gaseous wastes consist primarily of the activation products ^{13}N and ^{41}Ar and isotopes of the noble gas fission products, Kr and Xe, and halogens.

Radionuclides found in the coolant waters, and subsequently in the solid wastes, include most of the fission products and activated corrosion products such as ^{3}H, ^{32}P, ^{51}Cr, ^{59}Fe, ^{54}Mn, ^{58}Co, ^{60}Co, ^{65}Zn, ^{110m}Ag, ^{131}I, ^{134m}Cs, ^{137}Cs, ^{138}Cs, ^{139}Ba, and ^{140}Ba.

DISPOSAL PROCEDURES

If low in activity, liquid wastes are generally diluted with condenser cooling water to below 10^{-7} mCi/cc and discharged from the plant. Other liquid wastes, particularly from demineralizer regeneration, that contain relatively large amounts of activity and dissolved salts are collected in temporary storage tanks and then treated or decontaminated by evaporation. Evaporator overheads may be passed through mixed-bed demineralizers and returned to the plant system or discharged from the plant. Evaporator bottoms containing salt and radioactivity in the form of sludge are mixed with cement, put into drums, and shipped to regional land burial grounds.

Solid wastes contain about the same spectrum of radionuclides as the liquid wastes. These too are packaged and shipped to land burial sites.

Table 11 summarizes the radioactive-waste-management experience at four boiling-water and two pressurized-water nuclear reactor power stations.

Alan Preston, of the Ministry of Agriculture, Fisheries and Food of the United Kingdom, summarized in a private communication the annual liquid radioactive waste discharges of eight nuclear power stations operating in the United Kingdom in 1966, as shown in Table 12.

Materials-Production Reactors

At one time there were nine plutonium-producing reactors at the Hanford Plant, along the Columbia River approximately 360 miles upstream from its mouth on the Pacific Ocean; six of these were shut down between January 1965 and January 1969. One of the remaining reactors is dual-purpose, producing electric power (790-MW_e capacity) as well as plutonium; it was started up in 1963. These production reactors are designed for low-pressure, single-pass water cooling; the dual-purpose reactor has a closed-cycle coolant system similar to a pressurized-water reactor. In single-pass cooling systems, particulates in the river water are removed by coagulation, sedimentation, and filtration before the water enters the reactor. Sodium dichromate is added to reduce corrosion before the water passes through the reactor core and carries away the heat of fission. Impurities remaining in the water and corrosion products from the piping systems are neutron-radioactivated by the high neutron flux of the reactor core. In addition, fission products occur in the coolant because of natural uranium dissolved in the river water, uranium contamination on the surface of the fuel cladding, and occasional rupture of the fuel cladding.

Parker (1959) reports some 61 radioactive isotopes, as listed in Table 13, were detected in the Hanford reactor coolant discharge 4 hr after passage through the reactor. The reactor coolant stream is discharged to the Columbia River, where it mixes with the river water and is carried through four impoundment reservoirs enroute to the Pacific Ocean. The distance is approximately 360 miles, and travel time is 1 to 3 weeks, depending on stage of flow. The time of river flow permits radioactive decay, and interactions with the stream bed, sediments, and biota greatly reduce the spectrum of radioisotopes reaching the ocean. J. F. Honstead, of the Pacific Northwest Laboratories, Battelle Memorial Institute, Richland, Washington, in a private communication, estimates that annual amounts of radioactivity reaching the Pacific Ocean are as indicated in Table 14. In the early years of Hanford operations, methods for identifying and measuring extremely low concentrations of radionuclides were not yet developed. Consequently, samples were simply analyzed for "total beta" or "total alpha" content. The discrepancy between the "total beta" for 1960 and the sum of the isotopic analysis is explained by the fact that there is a low counting efficiency for ^{51}Cr and ^{65}Zn in a total beta analysis. The effect of decay time and river environment on the amount of radioactivity reaching the sea is seen in Table 15, which compares the concentrations at Richland, Washington, 34 miles downstream from the reactors, with concentrations at Bonneville Dam (the last impoundment before the sea), some 200 miles downstream.

Naval Propulsion Reactors

As of 1968, the United States had 76 nuclear-powered submarines in service, 21 under construction, and 8 more planned.* The United States also operated a two-reactor cruiser, an eight-reactor aircraft carrier, and two reactor-powered frigates. Three similar frigates, a deep-sea research vehicle, and a two-reactor aircraft carrier were planned or were being built (U.S. Atomic Energy Commission, 1967d). All nuclear-powered U.S. naval ships use pressurized-water reactors similar to the nuclear electric power stations described earlier. That is, they have closed primary and secondary cooling loops. The principal radionuclides considered as wastes are the neutron-activated corrosion and fission products that may leak through the fuel-element cladding. One major difference between the pressurized-water reactors on ships and on shore is the available physical space for tankage or other purposes. The reactor vessels in operational readiness must be kept full of water-moderator. At reactor start-up, the water expands when the reactor is brought up to operating temperature. In land-based plants, this expansion water is stored in tanks, whereas on submarines, it is dumped overboard. When the reactor is shut down and cooled, fresh water is added to keep the coolant system completely filled. On naval ships, reactors may be started up a few times per month on each ship, discharging on each heat-up an average of about 500 gallons (1,900 liters) of coolant (Iltis and Miles, 1959).

Other wastes that evolve from nuclear-ship operations include demineralizer resins used to decontaminate the coolant, reactor shield water, solid wastes from routine decontamination and maintenance operations, and laundry wastes.

**Janes Fighting Ships* (1967–1968) reports that the United Kingdom has 5 nuclear-powered submarines, France has 3 under construction, and the Soviet Union has 50 nuclear submarines and a nuclear-powered icebreaker in service and 2 nuclear-powered icebreakers under construction.

TABLE 11 Power Reactor Waste Management Experience[a]

	Boiling-Water Reactors				Pressurized-Water Reactors	
	Dresden-I	Big Rock Point	Humboldt Bay	Elk River	Indian Point-I	Yankee
Power rating	700 MW_{th}, 200 MW_e	157 MW_{th}, 50 MW_e	165 MW_{th}, 52 MW_e	58 MW_{th}, 24 MW_e	585 MW_{th}, 163 MW_e	600 MW_{th}, 185 MW_e
Operational period reported	Oct. 1959–Dec. 1966	Sept. 1962–Apr. 1967	Feb. 1963–Feb. 1967	Oct. 1961–Mar. 1967	Aug. 1962–Sept. 1966	Jan. 1961–Dec. 1966
Gross electricity generated, MWhr	6,600,000	1,053,000	1,055,000	393,000	3,489,000	6,362,000
Approximate capacity factor, %	65	45	80	70	50	70
Fuel cladding material	Stainless steel, zircaloy	Stainless steel	Stainless steel, zircaloy	Stainless steel	Stainless steel	Stainless steel
Max. assemblies with defective cladding, %	5	15	25	15	~0	~0
Gaseous wastes:						
Treatment	20-min delay, filtration	30-min delay, filtration	18-min delay, filtration	30-min delay, filtration	120-day delay, filtration	60-day delay, filtration
Stack exhaust rate, cfm	44,000	30,000	12,000	3,000	280,000	15,000
Height of stack, ft	300	240	250	97	400	150
Permissible annual average release[b]						
Activation and noble gases	700,000 μCi/sec	10^6 μCi/sec	50,000 μCi/sec	600 μCi/sec[c]	50,000 μCi/sec[c]	2,000 μCi/ml[d]
Halogens and particulates		3.6 μCi/sec[e]	0.18 μCi/sec[e]	0.003 μCi/sec[f]	0.24 μCi/cc[f]	
Range of annual average release						
Activation and noble gases	<100 to 25,000 μCi/sec	<20 to 35,000 μCi/sec	40 to 14,100 μCi/sec	0 to 109 μCi/sec	0.07 to 1.6 μCi/sec	0.002–22 Ci/yr
Percent of limit	<0.02–3.6	<0.002–3.5	0.08–28	0–18	0.00013–0.0026	0.001–0.03
Halogens and particulates	0.002–0.003 μCi/sec	<1.2 μCi/sec	10^{-5}–0.07 μCi/sec	<3 × 10^{-5} μCi/sec	~2 × 10^{-8} μCi/sec	
Percent of limit		<30	1–38	<1	$<10^{-5}$	

Liquid wastes:						
Principal type of treatment	Delay, filtration, demineralization	Delay, filtration, demineralization	Delay, filtration	Delay, filtration, demineralization	Delay, filtration, demineralization	Delay, filtration, evaporation
Volume collected, 10^3 gal/year	11,000	~1,000	~700	76	8,800	850
Percent returned to reactor use	84	~80	0	0	0	0
Activity range before dilution,[g] μCi/ml	10^{-5}-10^{-2} μCi/ml	10^{-4}-10^{-2}	10^{-4}-10^{-2}	6×10^{-8}-10^{-4}	3×10^{-6}-2×10^{-3}	10^{-6}-10^{-5}
Condenser water for dilution, gpm	167,000	50,000	100,000	28,000	260,000	140,000
Activity discharged						
Total curies/year[g]	4.1	5.8	1.3	0.01	11.1	0.01
Concentration range,[g] μCi/ml	10^{-8}-5×10^{-8}	2×10^{-8}-10^{-7}	2×10^{-9}-2×10^{-8}	10^{-10}-10^{-7}	3×10^{-10}-2×10^{-7}	3×10^{-10}-2×10^{-9}
Percent of limit	0.3-3.8[9]	1.9-9.9[i]	0.3-1.2[i]	0.0002-0.05[i]	0.03-4.7[i]	0.001-0.013[i]
Tritium, Ci/year (estimated)	5-10	20	20	10	500	1,300
Percent of limit (estimated)[j]	$<10^{-3}$	0.007	0.0035	0.006	0.03	0.16
Solid wastes:						
Average annual volume and activity[k]						
Resins	770 ft^3, ? Ci	250 ft^3, 120 Ci	50 ft^3, ? Ci	15 ft^3, 7 Ci	?	?
Other	3,200 ft^3, 100 Ci	250 ft^3, 100 Ci	1,000 ft^3, 250 Ci	400 ft^3, 10 Ci	900 ft^3, 1 Ci	3,600 ft^3, 500 Ci[l]

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[a] After Blomeke and Harrington, 1968.

[b] As stipulated in the respective AEC licenses.

[c] Based on an MPC for noble gas mixtures of 3×10^{-8} μCi/ml.

[d] Based on an MPC for noble gas mixtures of 3×10^{-7} μCi/ml, a concentration limit at point of discharge of 1,000 × MPC, and a stack exhaust rate of 15,000 scfm.

[e] Based on an MPC for halogens and particulates of 3×10^{-10} μCi/ml.

[f] Based on an MPC for halogens and particulates of 1×10^{-10} μCi/ml.

[g] Not including tritium.

[h] Limit is based on a continuous discharge (averaged over 12 consecutive months) of 10^{-6} μCi/ml of activity, including natural background in river water.

[i] Limit is based on a continuous discharge (averaged over 12 consecutive months) of 10^{-6} μci/ml of unidentified isotopes of plant origin.

[j] Limit is based on a continuous discharge of tritium (averaged over 12 consecutive months) of 3×10^{-3} μCi/ml.

[k] Solid waste activities are "best estimates" at the time of shipment. They vary on a "unit volume" basis over many orders of magnitude.

[l] This waste is mainly evaporator bottoms and incinerator ashes mixed with cement, but also includes resins.

TABLE 12 Radioactivity in Liquid Wastes Discharged by Nuclear Power Stations of the United Kingdom, 1964–1966

			Radioactivity Discharged (Ci)					
			1964		1965		1966	
Power Station	Location	Generating Capacity (MW_e)	^3H	Other	^3H	Other	^3H	Other
Berkeley	River Severn Estuary	2 x 138	1,008	1	306	5	225	23
Bradwell	Blackwater Estuary	2 x 150	37	4	471	19	583	29
Chapelcross	Open coast	4 x 45	–	4	–	5	–	28
Dungeness	Open coast	2 x 275	–	–	6	0.1	569	2
Hinkley Point	Parrett Estuary	2 x 250	–	0.01	174	2	32	18
Hunterston	Open coast	2 x 260	21	0.004	477	2	477	19
Sizewell	Open coast	2 x 290	–	–	–	–	29	1
Trawsfynydd	Lake Trawsfynydd	2 x 250	–	–	2	0.06	222	2
Oldbury	–	2 x 300	–	–	–	–	–	–
Calder Hall	Windscale	4 x 45	Discharges included in totals for fuel reprocessing operations (see Table 18, p. 32)					
Windscale AGR	Windscale	32						

TABLE 13 Hanford Reactor Effluent Isotopes in Order of Decreasing Concentration, Four Hours after Irradiation[a]

Major (90%)	Minor (8%)	Trace (2%)		
^{56}Mn[b]	^{69}Zn	^{152}Eu	^{131}I[b]	^{145}Pr
^{64}Cu[b]	^{72}Ga	^{153}Sm	^{141}Ce	^{151}Pm
^{24}Na[b]	^{92}Sr	^{187}W	^{142}Pr	^{60}Co
^{51}Cr[b]	^{239}U	^{141}La	^{14}C	^{143}Pr
^{239}Np[b]	^{133}I[b]	^{149}Nd	^{147}Nd	^{103}Ru
^{76}As[b]	^{92}Y[b]	^{140}La	^{45}Ca[b]	^{47}Sc
^{31}Si[b]	^{97}Nb[b]	^{132}I[b]	^{111}Ag	^{90}Sr[b]
	^{91}Sr[b]	^{157}Eu	^{91}Y	^{137}Cs
	^{65}Zn[b]	^{140}Ba[b]	^{59}Fe[b]	^{85}Sr
	^{32}P[b]	^{99}Mo	^{89}Sr[b]	^{238}U[b]
	^{90}Y[b]	^{156}Sm	^{54}Mn	^{239}Pu[b]
	^{135}I[b]	^{46}Sc[b]	^{95}Zr	^{227}Ac
	^{93}Y	^{115}Cd	^{149}Pm	^{210}Po[b]
		^{143}Ce	^{156}Eu	

[a] After Parker (1959).
[b] Routine measurements made on these isotopes.

TABLE 14 Annual Discharges of Radioactivity from the Hanford Reactor Operations to the Pacific Ocean (Ci)[a]

Year	Beta	^{32}P	^{51}Cr	^{65}Zn	^{239}Np
1950	2,500	–	–	–	–
1951	3,600	–	–	–	–
1952	7,100	–	–	–	–
1953	Insufficient data	–	–	–	–
1954	Insufficient data	–	–	–	–
1955	13,000	–	–	–	–
1956	17,000	–	–	–	–
1957	37,000	–	–	–	–
1958	44,000	–	–	–	–
1959	110,000	–	–	–	–
1960	93,000	6,200	310,000	14,000	26,000
1961	–	11,000	310,000	16,000	24,000
1962	–	4,700	240,000	11,000	11,000
1963	–	4,400	320,000	10,000	18,000[b]
1964	–	4,400	320,000	16,000	–
1965	–	4,000	290,000	18,000	–
1966	–	3,300	160,000	8,000	–
1967	–	4,400	224,000	15,000	–

[a] Since 1960 individual isotopes have been measured; previously, all isotopes were measured collectively.
[b] First 8 months only; analysis was subsequently discontinued.

The radionuclides commonly present in discharged reactor coolant are shown in Table 16, along with maximum and average concentrations obtained over 3 years of operations of the USS *Nautilus*. Similar results have been reported from other naval reactors (Iltis and Miles, 1959).

A summary of reported liquid-waste discharges and of amounts of activity discharged into U.S. harbors is given in Table 17. Harbor waters and sediments at shipyards, harbors, and submarine bases are monitored principally for ^{60}Co. In 1967, 55 sediment samples from three locations were reported to contain over 100 pCi of ^{60}Co per square centimeter, and 340 samples contained between 10 and 100 pCi/cm^2; the total bottom area with concentrations of ^{60}Co greater than 10 pCi/cm^2 was estimated to be approximately 0.6 km^2.

Waters in the harbors, on the other hand, showed no detectable ^{60}Co activity. In 1965, in Holy Loch, Scotland, ^{60}Co was detected in bottom sediment and on mud flats exposed at low tide, but none above background level was detected in the water. In 1966, levels were reported to have declined (Miles and Mangeno, 1967).

Demineralizer resins from shipboard reactors, containing as much as 12.5 Ci of activity, of which 10 Ci are attributable to ^{60}Co, are either transferred to shore facilities and subsequently handled as solid wastes or are dumped at sea more than 12 miles from shore while the ship is under way.

Solid radioactive wastes are transferred to shore facilities, packaged, and shipped to land burial sites.

TABLE 15 Annual Average Concentration of Several Radionuclides in Columbia River Water, 1966[a]

Radionuclides	Richland (pCi/liter)	Bonneville Dam[b]
R.E. + Y[c]	270	ID
^{24}Na	2,600	ID
^{32}P	140	23
^{51}Cr	3,600	1,300
^{64}Cu	1,400	ID
^{65}Zn	200	43
^{76}As	420	ID
^{90}Sr	1	ID
^{131}I	18	3
^{239}Np	770	ID

[a] After Honstead, 1967.

[b] ID, insufficient data.

[c] Rare earth + yttrium: ^{140}La, ^{152m}Eu, ^{153}Sm, ^{165}Dy, ^{90}Y, ^{91}Y, ^{93}Y, ^{141}Ce, ^{143}Ce, ^{144}Ce, ^{142}Pr, ^{143}Pr, ^{147}Nd, ^{147}Pm, ^{149}Pm, ^{151}Pm, ^{152}Eu, ^{156}Eu, ^{153}Gd, ^{159}Gd, ^{160}Tb, ^{161}Tb, ^{166}Ho, ^{169}Er, ^{171}Er.

TABLE 16 Concentrations of Radionuclides in Reactor Coolant Water, USS *Nautilus* (μCi/ml)

Nuclide	Maximum	Average
Corrosion Products		
^{18}F	2.7×10^{-2}	1.5×10^{-2}
^{24}Na	6.2×10^{-4}	2.2×10^{-4}
^{51}Cr	3.4×10^{-3}	3.7×10^{-4}
^{56}Mn	9.3×10^{-2}	2.1×10^{-2}
^{59}Fe	2.7×10^{-3}	5.1×10^{-4}
^{60}Co	2.5×10^{-2}	3.2×10^{-3}
^{64}Cu	4.7×10^{-3}	1.0×10^{-3}
^{65}Ni	1.5×10^{-3}	5.7×10^{-4}
^{182}Ta	2.9×10^{-2}	4.5×10^{-3}
^{187}W	9.0×10^{-3}	1.5×10^{-3}
Fission Products		
^{89}Sr		5.0×10^{-6}
^{90}Sr		5.0×10^{-8}
^{131}I		1.0×10^{-5}
^{140}Ba		1.0×10^{-6}
^{137}Cs		1.0×10^{-8}
^{144}Ce		1.0×10^{-7}

Civilian Propulsion Reactors

As of 1970, there were only two operational nuclear-powered civilian ships—the United States' NS *Savannah*, a cargo-passenger vessel, and West Germany's *Otto Hahn*, a 15,000-ton bulk carrier, which was undergoing sea trials. Japan has launched an 8,300-ton nuclear-powered freighter, the NS *Mutsu*, due for service in 1971. Italy has a nuclear-powered freighter under construction, and Japan and Germany have announced plans for two more freighters. The *Savannah* has provided experience to many maritime nations in how to deal with nuclear ships in harbors, coastal waters, and on the high seas.

The *Savannah*'s reactor is a pressurized-water type with a power rating of 80,000 kW_{th} and 22,000 shaft horsepower. The reactor vessel and its two primary coolant loops are contained within a vessel designed to withstand an internal pressure of 188 psi. The reactor has a dual shield: The primary shield is an annular water-filled tank around the reactor vessel with walls of lead 1 to 4 inches thick. The secondary shield, which surrounds the containment vessel, is made of concrete and steel in its lower half and lead and polyethylene in its upper half.

The reactor was first started up in December 1961, and in its first year of operation, the *Savannah* traveled some 30,000 miles, visiting ports on both U.S. coasts before returning to her maintenance base at Galveston, Texas. Since 1961, the *Savannah* has visited many of the major ports of the world and was in commercial service between U.S. and European ports. Through August 1968, she cruised 335,000 miles on her first fuel loading.

Although the *Savannah* was designed to contain all liquid radioactive wastes, with 1,350 ft^3 (38 m^3) of storage tank capacity, more wastes are generated than can be held. In its first year of operation, 23,700 ft^3 (670 m^3) of liquid wastes were evolved, primarily from leakage at the buffer-seal charge pumps, pressurized relief valve, and sampling-system relief valve. The first wastes contained 1.02 Ci of radioactivity and ranged in concentration between 3×10^{-8} and 5.4×10^{-4} mCi/ml. Of the first year's wastes, 8,300 ft^3 (235 m^3) were disposed of on land and the remainder dumped at sea. The wastes disposed of at sea were within the limits suggested by the National Research Council in its 1958 report (National Academy of Sciences–National Research Council, 1959).

It has been reported (Nuclear Safety Information Center, 1966) that in its first 6 months of commercial service (four round-trip voyages to European ports between August 1965 and March 1966), the *Savannah* discharged into the sea some 8,100 ft^3 (229 m^3) of liquid wastes containing 6×10^{-3} Ci of radioactivity, primarily ^{54}Mn and ^{60}Co. Solid wastes and demineralizer resins were disposed of on land. Off-gases from the containment vessel were passed through a series of filters prior to release to the atmosphere.

Aerospace Nuclear Reactors

Two classes of reactors are being developed for use in the aerospace program—one to provide electrical or other form

TABLE 17 Summary of Liquid Radioactive Wastes Discharged into Harbors by U.S. Navy Nuclear-Powered Ships[a]

Shipyard or Naval Harbor[b]	Liquid Radioactive Waste Discharges[c]				
		1966		1967	
	1961-1965 (Ci)	1,000 gal	Ci	1,000 gal	Ci
Portsmouth, N.H.	0.48	155	0.01	265	0.01
Quincy, Mass.	0.15	0	<0.005	–	<0.005
Groton–New London, Conn. (Electric Boat Div., state pier and submarine base)	7.18	1,274	0.03	606	0.01
Camden, N.J.	0.01	0	<0.005	–	–
Newport News, Va.	2.31	1,581	0.06	1,533	0.04
Norfolk, Va.	1.17	1,051	0.03	1,784	0.03
Charleston, S.C.	1.19	369	0.04	320	0.01
Pascagoula, Miss.	0	0	<0.005	–	<0.005
San Diego, Calif.	0.99	0	<0.005	–	<0.005
Vallejo, Calif.	2.33	270	0.19	–	<0.005
Bremerton, Wash.	0.03	0	<0.005	–	<0.005
Pearl Harbor, Hawaii	3.20	654	0.03	683	0.01
Apra Harbor, Guam	0.01	0	–	–	<0.005

[a] After Miles and Mangeno (1966, 1967, 1968).
[b] Other U.S. harbors have had less than 20,000 gal and less than 0.1 Ci discharged into them per year.
[c] Radioactivity measurements have been calibrated to ^{60}Co standard; volumes are prior to dilution.

of power in the kilowatt range for spacecraft missions, and the other, in the megawatt range, capable of propelling space vehicles into orbit. Both could introduce nuclear fission and activation products into the sea, either directly by launch accident or indirectly by fallout of burn-up materials from the atmosphere resulting from scheduled or unscheduled re-entry of space vehicles.

SNAP reactors (*S*ystems for *N*uclear *A*uxiliary *P*ower) are relatively small electrical generating plants designed to produce full power for more than a year without refueling. The aerospace SNAP reactors have fuel elements of uranium mixed with a neutron moderator–zirconium hydride (U-ZrHx). The fuel elements have thin-wall steel cladding and are arrayed within a stainless-steel reactor vessel. The neutron flux and chain reaction are controlled by beryllium reflectors located outside the reactor vessel. Heat is conducted from the fuel elements to the electrical generating system by a liquid metal, NaK, an alloy of sodium and potassium.

SNAP 10A* was launched in April 1965 and operated at full power for 43 days, at which time it was shut down by a spurious command from the satellite on which it was riding. This unit, designed to produce 500 electrical watts, used thermoelectric converters from a heat source of 60 kW_{th}. It is now in a 700-mile-high nearly circular polar orbit. The reactor is designed to break up, ablate, and disperse on re-entry into the earth's atmosphere. If SNAP 10A had operated for a year, it would have accumulated approximately 40,000 Ci of fission products. During its 43 days of operation, it accumulated about one tenth as much radioactivity. At the time of re-entry, in about 600 years, essentially only the longest-lived fission products–^{90}Sr and ^{137}Cs–would remain, and these would constitute only a millionth of the original amount.

SNAP 8, a 600-kW_{th} reactor system designed to produce 30 kW_{e} using liquid metal for coolant and a turboelectric generator, is being developed to supply power for manned oribital laboratories, lunar expeditions, communications satellites, or deep space missions. Methods for disposal of the reactor at the end of its operation are being evaluated. These methods include random re-entry and burn-up at high altitudes; intact re-entry into the deep ocean; intact earth impact and recovery; and boost into outer space (Nelson and Detterman, 1967).

Entry into the ocean is also possible in the event of a launch abort. If the reactor retains its components and configuration up to the time of impact with water, a nuclear excursion of 70 MW-sec (equivalent of 250-kW operation for 280 sec) would result. Although about half a million curies of total fission-product radioactivity would be created, it would consist largely of short-lived isotopes; in 2 hr, the radioactivity would have decayed to several thousand curies, in 4 days to about 40 Ci, and in 40 days to about 3 Ci (Kochendorfer, 1964).

In 1964, an experiment was performed to provide data and information for evaluation of the consequences of water immersion of an air-cooled aerospace reactor (Kessler *et al.*,

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*In SNAP terminology, even numbers denote nuclear reactor units, and odd numbers, isotopic-heat-generating units.

1964). A SNAP 10A reactor was injected into a large tank of water. A nuclear excursion of about 45 MW-sec occurred, which resulted in the rupture of all fuel elements in the reactor core and dispersal of the fuel (within the tank). The fuel lattice retained about 99 percent of the available fission products. Halogens that escaped from the fuel were retained in the water. Primarily noble gases and their daughters escaped, and these amounted to about 5 percent of the noble gas present in the core.

Nuclear rocket engines will be used to propel upper stages or large, advanced space vehicles after conventional chemical rockets have carried the nuclear stage well above the atmosphere. In all mission applications currently foreseen, the nuclear stage will be left in deep space after use and will never return to Earth. Hence, no radioactive materials are scheduled to enter the oceans from flight operations; ocean entry becomes a possibility only after a failure or accident.

There are two possibilities for failure. One would result from the booster failing during early ascent.* Since the reactor would not yet be started, it would not be radioactive when it falls back into coastal waters. However, rapid introduction of water into the core could override the control system and cause a nuclear excursion. The magnitude would depend on the water entry rate, which, in turn, would depend on the velocity and orientation of the vehicle on impact. In the worst case, up to 10^4 to 10^5 MW-sec of energy could be generated in a fraction of a second before the reactor destroyed itself. One would expect about two thirds of the fission products generated during the excursion to be contained in a low atmospheric cloud of gases and fine particulates and the rest to remain submerged in larger fuel fragments. The normal fission product inventory for thermal fission in ^{235}U (e.g., 3–30 Ci of ^{90}Sr) would be expected, plus a much lower number of activation products.

The other possibility for introducing radioactive material into the oceans would result from failures after the nuclear reactor is brought to power—about 4,000 MW. In some missions, the reactor would be started before Earth orbit is reached; in others, the reactor would be first started up in orbit. Depending on the start-up point, the reactor would operate from 10 to 20 minutes before reaching mission injection velocities—roughly Earth escape velocity. Complete failure at any time during operation would leave the highly radioactive reactor coasting in space. The period of time from failure to atmospheric re-entry and impact is extremely sensitive to the velocity attained at the instant of failure, and can range from less than an hour to hundreds of years. If the delay time is long enough for normal radioactive decay to reduce the inventory sufficiently, the failed machine will be allowed to remain in orbit. If the time before impact is too short, however, deliberate action will be taken, if necessary, to cause impact and disposal in deep ocean waters. In such cases, impact would occur within an hour. Again, atmospheric re-entry would break up the reactor so that the radioactive fuel elements and other debris would impact over an area of several square miles. No excursion would result, and the fission and activation products would remain within heavy pieces that would sink rapidly.

WASTES FROM THE PROCESSING OF REACTOR FUEL ELEMENTS

The vast amounts of fission products created in reactor fuel elements remain in the uranium matrix within the aluminum, stainless-steel, or zirconium-alloy cladding or jackets. Spent fuel elements are periodically removed from reactors and are shipped intact to chemical reprocessing plants, after a period of storage in water-shielded tanks to permit the short-lived radioisotopes to decay away. At the reprocessing plants, the cladding is removed, and the fission products are chemically separated from the unfissioned uranium in processes of two or more stages or cycles. First-stage waste normally contains more than 99 percent of the fission product radioactivity; this material, referred to as high-radioactivity or high-level waste, is kept in underground storage tanks. To our knowledge, no nation deliberately disposes of such high-level wastes to the environment.

Second and third stages of reprocessing unfissioned uranium result in larger volumes of wastes with significantly less radioactivity content. In the United States, these liquids may be still further decontaminated by evaporation or other process and discharged to underground formations. In the United Kingdom, such low-level wastes are discharged to coastal waters. Table 18 summarizes 11 years of radioactive-waste disposal by the Windscale plant. The Dounreay Experimental Reactor Establishment, in northern Scotland, also disposes of some chemical-processing plant wastes to the sea near Caithness, Scotland; in 1967, it was reported by the Fisheries Radiobiological Laboratory of the Ministry of Agriculture, Fisheries and Food that the Dounreay plant discharged 290 Ci of ^{90}Sr, 12,000 Ci of beta activity, and 14 Ci of alpha activity. Other plants around the coasts of the British Isles, where fuel elements are fabricated or radioisotopes are processed, dispose of smaller total amounts of activity than the fuel reprocessing plants.

In France, the chemical reprocessing plant at Marcoule disposes of about 1,800 Ci in liquid wastes per year to the

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*Not every ocean impact could result in an excursion. During launch and early portions of the ascent, auxiliary controls (neutron absorbers or poisons) may be used to override the effects of water entry and preclude an excursion. Beyond some point, however, failures could result in ocean impacts of such force as to dislodge the auxiliary poisons. Still later, atmospheric re-entry would cause reactor disassembly prior to impact, so that an excursion is impossible.

TABLE 18 Mean Activity Discharge Rates (Ci/mo) to the Irish Sea, Windscale Chemical Reprocessing Plant, United Kingdom[a]

Radionuclide	1957	1958	1959	1960	1961	1962	1963	1964	1965	1966	1967
^{106}Ru	2,218	3,522	2,956	3,302	2,095	1,916	2,781	1,924	1,752	2,077	1,436
^{103}Ru	300	492	746	964	265	153	800	100	150	205	186
^{90}Sr	137	210	129	43	41	85	46	81	97	76	116
^{89}Sr	248	72	170	82	114	42	14	16	14	7	12
^{144}Ce	215	497	583	74	180	200	116	267	288	571	1,142
^{91}Y + rare earths	300	567	506	83	201	125	90	90	73	75	203
^{137}Cs	310	516	165	76	91	92	31	111	97	100	132
^{95}Zr	59	210	415	196	140	78	47	1,797	1,479	1,172	1,566
^{95}Nb	535	510	845	523	658	356	272	1,735	2,803	1,947	2,143
Total beta	5,366	6,846	7,659	6,461	3,981	3,742	4,020	5,055	4,560	5,464	6,022
Total alpha	4.8	5.2	5.6	6.8	11.1	15.5	19.0	23.5	33.8	48.8	79.6

[a] After Howells (1966).

Rhone River; in 1965, the amount was 2,584 Ci (Rodier *et al.*, 1966). In addition, there is a new chemical reprocessing plant for irradiated fuel elements at La Hague on the Cotentin Peninsula with plans for disposal of treated wastes directly to the English Channel (Avargues and Jammet, 1966).

RADIOISOTOPIC POWER GENERATORS

SNAP isotopic devices are now employed over, on, and in the oceans. Tables 19 and 20 list the characteristics and applications of existing U.S. units. SNAP* units utilize the heat of radioisotopic decay. It has been proposed that in some aerospace units the heat be used directly for thrusters. In terrestrial applications, the heat is converted to electricity by thermoelectric converters.

The source of energy for the SNAP devices are the long-lived fission products or trans-uranics recovered from the tank-stored first-stage wastes from the processing of the irradiated fuel elements. These are first concentrated and then converted metallurgically to a dense mass most suitable for SNAP units. The isotopic power devices used in the United States are all of similar construction; only their size and output varies with the different fuels and thermoelectric converters. Figure 7 schematically illustrates the configuration of an isotopic power unit and the basic materials of construction. The materials of construction are chosen to provide a margin of safety in the event of a damaging accident.

Most of the SNAP devices in oceanic applications to date are fueled with ^{90}Sr in the chemical form of titanate, $SrTiO_3$. The titanate form is an inert ceramic that dissolves quite slowly on exposure to fresh or salt water. Future fuels may include the oxide forms of Sr, or Ce, or Co in metallic form, giving the most dense and efficient power-to-weight ratio. The amount of radioactivity in present SNAP units ranges from 30,000 Ci to 225,000 Ci of ^{90}Sr and is directly proportional to electrical output. Future units may contain up to 10^7 Ci. SNAP units for aerospace units use the most advantageous power-to-weight ratios and are fueled with metallic or oxide forms of somewhat shorter-lived isotopes, i.e., those having higher specific activity.

The isotopic fuel forms are jacketed or encapsulated in sealed (welded) corrosion-resistant metals and alloys such as Haynes, Hastelloy, or titanium. These canisters of fuel are completely surrounded by a heat-absorbing metal such as depleted uranium or tungsten. An insulating material around the fuel canister and heat-sink absorber keeps the isotopically generated heat at maximum temperature. The thermoelectric couples, which operate on the principle of maximum temperature drop, are linked at one end to the heat sink and at the other to the outside of the insulator. The insulation package is surrounded by a radiation-shielding material such as depleted uranium or lead, and this is encased in a container of heavy steel. For oceanic applications, the outer steel shell is designed to be a pressure vessel, as is the fuel capsule itself.

Corrosion Tests of SNAP Components

Hastelloy-C, the fuel-encapsulating metal of existing oceanic SNAP devices, has an average corrosion rate in seawater of 0.024 ± 0.008 mils per year (0.00061 ± 0.0002 mm per

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*SNAP is the U.S. designation; in Europe they are called RIPPLE (*R*adio*I*sotopic *P*ower *P*ackages for *E*lectricity).

TABLE 19 Characteristics and Applications of Oceanic SNAP Systems

Power System	Electrical Output	Fuel Form	Fuel Quantity (kCi)	Marine Application	Status as of November 1968
Past and Present Systems					
SNAP-7A	10 W	$SrTiO_3$	41	Coast Guard buoy	Post test analysis after 3-yr operation
SNAP-7B	60 W	$SrTiO_3$	225	CG lighthouse; then, offshore oil platform	Operating (relocated on oil platform in August 1966 after 2 yr on lighthouse)
SNAP-7D	60 W	$SrTiO_3$	225	Navy NOMAD buoy, Gulf of Mexico	Operating (implanted January 1964)
SNAP-7E	7.5 W	$SrTiO_3$	31	Sonar transducer at 15,600 ft depth	Operating (implanted July 1964)
SNAP-7F	60 W	$SrTiO_3$	225	Offshore oil platform	Post test examination of power decrease
Future Systems					
SNAP-21	10 W 20 W	$SrTiO_3$ or SrO	33 64	Sonar, cable boosters, navigation aid, research	Under development (environment test units 1969)
SNAP-23	25 W 60 W 100 W	$SrTiO_3$ or SrO	75 185 275	Weather buoy, navigation buoy, offshore oil platform	Under development (environmental test units scheduled 1971)
KW	1-10 kW	$SrTiO_3$, SrO, Co, or CeO	Up to 10,000	Man-in-the-sea research; offshore oil, mining, and exploration; communications	Application engineering and design study in progress

TABLE 20 Characteristics and Applications of Aerospace SNAP Systems

Power System	Power Output	Fuel Form	Fuel Quantity (kCi)	Aerospace Application	Design Mode of Re-entry	Status (1968)
NASA Goddard Microthruster	$60\ W_{th}$	^{147}Pm	170	Space vehicle thruster	Intact re-entry	Ground tests
Air Force DART	$157\ W_{th}$	^{238}Pu	4.7	Space vehicle thruster	Intact re-entry	Ground tests
SNAP-19	$60\ W_e$	^{238}Pu	34.5	NIMBUS B auxiliary power	960-km orbit; intact re-entry	Aborted launch in 1968, recovered intact from ocean
SNAP-27	$50\ W_e$	^{238}Pu	45	ALSEP mission	Superorbital; intact re-entry	
SNAP-3A	$3\ W_e$	^{238}Pu	1.6	Navy satellite	800-km orbit; burn-up on re-entry	2 launched 1961
SNAP-9A	$25\ W_e$	^{238}Pu	16	Navy satellite	960-km, 900-yr orbit; burn-up on re-entry	2 launched 1963 1 aborted 1964

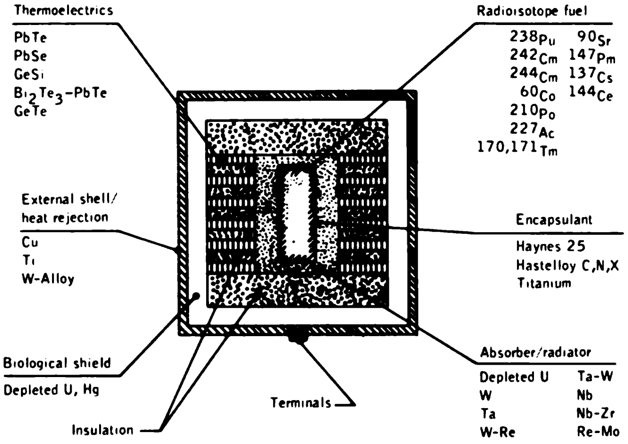

FIGURE 7 Schematic of SNAP isotopic power generators.

year) (Kubose and Cordova, 1966). It has been calculated that it would take some 300 years to corrode through the Hastelloy-C at that rate.

Laboratory and oceanic experiments were conducted to determine the dissolution rate of the SNAP fuels in seawater. Laboratory measurements of strontium titanate indicate a high initial release rate, 800 μg/cm^2 on the first day, followed by a slower rate of release (Zigman *et al.*, 1964). The release rate for extended periods was expressed as $R = 105\ (t + 6)^{-1}$ μg/cm^2/day (for $t - 1$), where t (time) is expressed in days.

Strontium titanate was also exposed to an oceanic environment at a depth of 30 m for 1,065 days (Goya *et al.*, 1966). Biological growth was evident on the fuel specimens. Measured dissolution rates varied from 4 to 14 μg/cm^2/day.

Plutonium metal reportedly reacts rapidly and completely with seawater, producing solid reaction products plus hydrogen (Lai and Goya, 1966). The solid reaction products have a solubility of about 1.5 to 2 μg/day/50–70 mg of solids. In another series of laboratory experiments, plutonium dioxide was immersed in seawater under various temperatures and pressures (Kubose *et al.*, 1967a). The long-term solubility rate was found to be lower at high temperatures, and this was attributed to the formation of a coating of calcium sulfate on the fuel-form specimens. The data were as follows:

Temperature (°C)	Pressure (psig)	Dissolution Rate (μg/mg/day)
20	15	4×10^{-3}
120	30	1.3×10^{-4}
190	180	0.6×10^{-4}

Later experiments indicated that the dissolved plutonium was absorbed from solution by ocean sediments; in one experiment, the amount in solution was reduced by a factor of 600 (Kubose *et al.*, 1967b).

SNAP isotopic units are also used in aerospace applications. Several are now in orbit, providing electrical energy for a satellite navigation system.

FALLOUT OF ^{238}Pu FROM THE SNAP-9A RE-ENTRY

In April of 1964, a SNAP-9A nuclear power generator aboard an aerospace vehicle re-entered the atmosphere following a malfunction during launch. The generator contained about 17 kCi of ^{238}Pu and was estimated to have entered the atmosphere of the southern hemisphere at an altitude of approximately 150,000 ft (45,000 m). (By contrast, approximately 400 kCi of ^{239}Pu have been dispersed from all nuclear explosions in the atmosphere.)

The plutonium of the device was assumed to have completely ablated during the intense heat of re-entry and to have been distributed in the atmosphere as particles of unknown size.

In the first 2 years since the re-entry and burn-up of the SNAP-9A, several programs were carried out to find, identify, and characterize the debris. The following conclusions briefly summarize the results of these studies:

1. More than 80 percent, or about 15 kCi, of the original 17 kCi in the device was still in the stratosphere in early 1966. Of this, some 80 percent was in the southern hemisphere and 20 percent in the northern hemisphere.
2. Particle size of the stratospheric ^{238}Pu from the SNAP-9A generator probably ranged from 5 to 58 mμ in diameter, with the arithmetic mean at 9.7 mμ.
3. By the end of 1966, approximately 1.5 kCi of the SNAP-9A plutonium had been deposited on the earth's surface. The southern hemisphere had received approximately 85 percent of this, while only 15 percent had been deposited in the northern hemisphere.
4. The distribution of the ^{238}Pu in the stratosphere over the southern hemisphere in mid-1966 approximated rather well the distribution of ^{90}Sr in early 1965 over the northern hemisphere. The altitude and latitudinal distribution of the highest core of activity in the southern hemisphere for the SNAP-9A materials generally mirror-imaged the older ^{90}Sr picture in the northern hemisphere.

Based on these observations, it is assumed that since the beginning of 1967, the fallout pattern of the ^{238}Pu particles from the SNAP-9A burn-up was and will continue to be similar to the pattern of stratospheric ^{90}Sr fallout observed over the past few years.

Table 21 is based on the above observations and assumptions, using a 10-month stratospheric half-residence-time. An additional assumption, based upon very qualitative observation of the ^{90}Sr stratospheric inventories, is that the stratospheric air of the two hemispheres mixes slowly and achieves

TABLE 21 Projected Deposition of ^{238}Pu from the SNAP-9A Re-entry (in kCi)

Latitude	1967	1968	1969	1970	1971	Total
80°-90° N	0	0	0	0	0	0
70°-80°	0.02	0.01	0	0	0	0.03
60°-70°	0.08	0.04	0.02	0.01	0.01	0.16
50°-60°	0.28	0.17	0.09	0.05	0.03	0.62
40°-50°	0.37	0.22	0.11	0.07	0.04	0.81
30°-40°	0.33	0.19	0.10	0.06	0.04	0.72
20°-30°	0.22	0.13	0.07	0.04	0.03	0.49
10°-20°	0.23	0.14	0.07	0.04	0.03	0.51
0°-10°	0.17	0.10	0.05	0.03	0.02	0.37
Total, northern hemisphere	1.7	1.0	0.5	0.3	0.2	3.7
0°-10° S	0.68	0.27	0.10	0.05	0.02	1.12
10°-20°	0.93	0.37	0.14	0.07	0.03	1.54
20°-30°	0.89	0.35	0.13	0.07	0.03	1.47
30°-40°	1.32	0.52	0.19	0.10	0.04	2.17
40°-50°	1.46	0.58	0.22	0.11	0.04	2.41
50°-60°	1.14	0.45	0.17	0.09	0.03	1.88
60°-70°	0.30	0.13	0.04	0.02	0.01	0.50
70°-80°	0.07	0.03	0.01	0	0	0.11
80°-90°	0.01	0	0	0	0	0.01
Total, southern hemisphere	6.8	2.7	1.0	0.5	0.2	11.2
Total, world	8.5	3.7	1.5	0.8	0.4	14.9

approximately equal concentrations of ^{238}Pu in about 4 years, which, in fact, was observed. In Table 21, the predicted annual fallout of ^{238}Pu from the SNAP-9A burn-up is given for each 10° band of latitude.

It is interesting to compare the probable sensitivity required for observing the ^{238}Pu deposited on land and in the sea. By the end of 1971, when virtually all of the SNAP-9A debris will have been deposited on the earth, the average concentration in the surface ocean water (assuming that the ^{238}Pu is homogeneously mixed to a depth of 100 m) will be approximately 0.0005 pCi per liter. Even within the latitude band of maximum deposition, 40°-50° S, and assuming no horizontal mixing, we would not find concentrations much higher than 0.001 pCi per liter. In contrast, the ^{238}Pu deposited on land in the 40°-50° S band will be about 1 mCi per km^2, or about 1,000 pCi per m^2. Even in the northern hemisphere, the deposits in mid-latitudes will be in excess of 100 pCi per m^2. These projected concentrations on land and in the sea seem to indicate clearly that the ^{238}Pu will be a very useful and practical tool for continental fallout studies. For marine studies, however, the concentrations will probably be beyond the range of practical measurement, using current equipment and technology. Therefore, if we are to make use of this radioactive source as an oceanic tracer, new systems for sampling of the sea—for determining chemical or physical concentrations of plutonium in seawater or for low-level alpha counting (or some combination)—must be developed.

PACKAGED RADIOACTIVE WASTE DISPOSAL

Any establishment working with radioactive materials probably evolves radioactive wastes, since anything the radioactive material comes into contact with is likely to become contaminated—i.e., some of the radioactive material is rubbed off or left behind. Such radioactive waste materials include laboratory ware and furniture; chemical hoods or their air filters; tubing and piping; protective plastics; experimental hardware; rags, wipes, and mops used for cleaning up; sludges from evaporators; and demineralizer resins. Uranium refineries, fuel fabrication plants, reactors, fuel reprocessing plants, radioisotope production plants, and laboratories working with uncontained radioisotopic materials all evolve radioactive wastes. In the United States, hundreds of establishments, both government and private, are licensed to use radioactive materials. The volume and level of activity of the wastes they may evolve vary in direct proportion to the amount of radioactivity being manipulated. Activity levels of wastes produced per year per establishment range from picocuries to kilocuries.

Since wastes occupy space, and since their accumulation may eventually build a radiation hazard, it is desirable to remove them to where they will be out of the way and create no further hazard. Remote, uninhabited places, such as abandoned mine shafts, government-owned and monitored sites, specially engineered burial vaults, or selected sites in the deep sea (>1,000 fathoms) have been suggested. Such places are limited in number, however, and as man extends his exploration and exploitation of the deep sea, possible sites for waste disposal there will diminish still further in number and remoteness. It will eventually be necessary to limit the number of sites used for this purpose and to monitor them in order to safeguard users of the sea floor.

Currently, in the United States, most of the solid wastes are buried on the sites of the federally owned atomic-energy plants or on state-owned lands. The latter are operated commercially and monitored by licensees, subject to government inspection.

In the early years of the atomic-energy industry in the United States, several establishments near the coasts used the sea for disposal of radioactive wastes. A few commercial companies were licensed by the AEC to practice sea disposal. However, since 1960, when the AEC adopted a policy leading to establishment of licensed regional land burial sites for private industry's radioactive wastes, sea disposal has waned. For sea disposal, containers of wastes were weighted, to assure sinking, and fabricated to accommodate deep-sea pressures. Land burial, on the other hand, requires only structural integrity for transporting; consequently, it is less expensive than sea disposal.

The United Kingdom has established two land burial sites in northern England for solid wastes containing small amounts of radioactive contamination. Such wastes constitute the largest fraction of all solid radioactive wastes (Morely, 1967). Many other nations do not have available as much remote land with required geohydrological characteristics as the United States. They look upon deep-sea disposal, therefore, as a safe and efficient means of getting rid of solid wastes. Their alternative is to use engineered burial vaults, which are more expensive to build and maintain than sea disposal.

Packaging for Sea Disposal

In the United States, the container most used for packaging radioactive wastes for sea disposal was the 55-gal (200-liter) oil drum. Paper or rag wastes were compressed into small bales, placed in the drum and completely surrounded by high-density material, usually concrete. Solids such as plastics and metals were mixed in the drum with concrete, the ratios of concrete to wastes depending directly on the levels of radioactivity. Sludges and demineralizer resins were usually contained within a carboy or smaller drum, then surrounded by concrete in the 55-gal drum. Several establishments packaged wastes in prefabricated steel-mesh-reinforced concrete boxes that varied in size from 20×20×44 in. (50×50×110 cm) to 5×7×8 ft (1.5×2.1×2.4 m). Wastes and concrete were mixed together in the box to achieve the desired specific gravity, 1.2. All packages, drums, and boxes were capped with 4 in. (10 cm) or more of clean concrete, and those with internal air spaces were fitted with vents to allow water to enter and equalize pressures on descent in the ocean (Joseph, 1957). Other nations package wastes for sea disposal similarly, especially using 55-gal drums.

In 1961, the AEC conducted two series of tests on the structural integrity of the various kinds of packages used for sea disposal. These tests provided information that was used to determine optimum combinations of steel and concrete needed to package various configurations of wastes. They also indicated that air spaces within the package of wastes must be pressure-equalized with vents in order to prevent container collapse (Pohlman and Pickett, 1962). Large concrete boxes with unvented internal air voids were found to be particularly vulnerable to rupture by pressure (Pneumo Dynamics, 1961b). In another test, smaller, unvented concrete boxes survived immersion to 6,000 ft (Brown *et al.*, 1962). Burns and Dunster (1967) report that the U.K. packaging practice also is to use reinforced concrete and pressure equalization devices.

Disposal at Sea

Tables 22 through 24 summarize the known packaged radioactive waste sea disposal operations conducted through 1967. Locations are approximate because of drifting of the ship during disposal and by the containers during descent through the mile-plus depths. Some of the U.S. disposals were conducted by transoceanic steamship companies that are licensed to dispose of wastes. Numbers of containers include all sizes, large concrete boxes as well as 55-gal drums. However, probably more than 95 percent of all containers were of the 55-gal size.

The radioactivity contained in the waste packages can only be estimated on the basis of labels describing the contents of waste receptacles coming into the packaging facility. Contaminated heterogeneous solids are the most difficult to assay; homogeneous sludges or slurries are the easiest. External radiation readings are marked on each container. The wastes generally include a wide spectrum of radionuclides, ranging from ^{3}H to the transplutonics. Alpha curies as a rule indicate thorium, uranium, or plutonium wastes, the isotopes having long half-lives.

In 1967, the European Nuclear Energy Agency coordinated a waste disposal demonstration "experiment" to establish common sea disposal operation principles and safety practices. Five nations participated, as indicated in Table 25 (European Nuclear Energy Agency, 1968).

In 1957 and 1960, the United States monitored the waste disposal sites off its Pacific Coast. Samples of water, biota, and sediments were collected and analyzed; none of the disposed radionuclides was detected in any of the samples (Faughn *et al.*, 1957; Pneumo Dynamics, 1961a).

MISCELLANEOUS SOURCES OF RADIOACTIVITY

There are several other possible sources of radionuclides that reach the oceans. These include low-level liquid wastes from medical and industrial users of isotopes and other research establishments (International Atomic Energy Agency, 1965). Such wastes, within established permissible concentrations of activity, are normally disposed of into municipal sewer systems. The widespread use of radioisotopes and the number of isotope users is becoming quite significant: in

TABLE 22 Summary of U.S. Sea Disposal Operations, 1951-1967 (Atlantic Ocean)

Year		Approximate Location	No. Containers, All Types	Estimated Activity at Time of Packaging (Ci)
1951-1958		42°25'N 70°35'W	4,008	2,440
		36°56'N 74°23'W	432	6.5
		Midocean	97	<0.1
		37°50'N 70°35'W	23,000	8,000
		38°30'N 72°06'W		
1959-1960		38°30'N 72°06'W	5,800	68,500[a]
	between	36°44'N 45°00'W	228	456
	and	36°50'N 74°23'W	204	24.5
		Midocean	22	<0.1
1961-1962		36°56'N 74°23'W	137	15.6
1963-1964		36°56'N 74°23'W	58	5.3
1965		36°56'N 74°23'W	6	4.3
1967		36°56'N 74°23'W	6	30.5
Totals			33,998	79,482.9

[a] Includes pressure vessel of Seawolf reactor—estimated 33,000 Ci of induced activity.

TABLE 23 Summary of U.S. Sea Disposal Operations, 1946-1966 (Pacific Ocean)

Year		Approximate Location	No. Containers, All Types	Estimated Activity at Time of Packaging (Ci)
1946-1958		37°39'N 123°26'W Wrs4 734	24,305	14,061
	between	40°07'N 135°24'W	38	0.5
	and	54°10'N 141°09'W		
1959-1960	between	40°07'N 135°24'W	53	0.4
	and	54°10'N 141°09'W		
		37°39'N 123°26'W	14,311	155.7
		32°00'N 121°30'W	3,467	3.6
		21°28'N 157°25'W	39	0.1
		34°58'N 174°52'W	7	14
1961-1962	between	40°07'N 135°24'W	41	0.5
	and	54°10'N 141°09'W		
		32°00'N 121°30'W	948	30
		37°39'N 123°26'W	8,913	235.1
		33°39'N 119°28'W	164	47.9
1963-1964	between	40°07'N 135°24'W	175	7.2
	and	54°10'N 141°09'W		
		37°39'N 123°26'W	8	16.1
1965	between	40°07'N 135°24'W	14	<0.1
	and	54°10'N 141°09'W		
		37°39'N 123°27'W	4	0.4
1966	between	40°07'N 135°24'W	40	88.7 (^{3}H)
	and	54°10'N 141°09'W		
		37°12'N 123°55'W	3	15.4 (^{3}H)
Totals			52,530	14,677.3

TABLE 24 Summary of Sea Disposal of Packaged Radioactive Waste by the United Kingdom, 1950-1967

Year	No. Containers	Estimated Activity (Ci)	
		Alpha	Beta
1951	100	0.5	5
1953	131	2.5	3
1955	2,519	12	33
1957	7,369	955	808
1958	4,330	695	1,085
1961	6,890	563	1,630
1962	648	17	163
1963	8,379	368	7,071
1964	10,634	444	15,090
1965	5,140	114	13,754
1966	3,362	78	2,742
1967[a]	1,268	91	1,682
	50,570[b]	3,331	44,096
1950–1963[c]	61,570	390	1,176

[a] Included in European Nuclear Energy Agency packaged radioactive waste disposal operation, 1967.

[b] Through 1961, the wastes were disposed of in an area near 34° N 20° W; after 1961, except for 1967, they were disposed in a rectangular area circumscribed by the coordinates: 40°20'N 13°53'W, 47°56'N 13°27'W, 48°43'N 13°05'W, and 48°19'N 12°39'W.

[c] Relatively low-activity wastes, including some from C.E.N.-Belgium and from Belchim, Belgium, disposed in the "Hurd Deep", an area near 50° N 01° W, by the United Kingdom.

TABLE 25 Summary of European Nuclear Energy Agency Packaged Radioactive Waste Disposal

Country of Origin	Number of Containers	Estimated Activity (Ci)	
		Alpha	Beta–Gamma
United Kingdom[a]	1,268	91	1,682
Germany	480	0.5	5
Netherlands	501	0.07	2
Belgium	1,945	–	190
France	896 (concrete)	1	10
	30,700	160	5,747
Totals	35,790[b]	253	7,636

[a] Also included in Table 24.

[b] Disposal area near 42° N 14° W.

1968, there were 8,321 licensees in the United States alone, many of them concentrated in urban population centers.

The fate of such wastes is indicated by two recent studies; one, of wastes discharged directly to a river system, and the other, of wastes reaching a sewage treatment plant. The Clinch River environment below the Oak Ridge National Laboratory was surveyed for radionuclides after nearly 20 years of low-level waste discharge that amounted to 1,110 Ci of ^{90}Sr, 660 Ci of ^{137}Cs, 6,600 Ci of ^{106}Ru, 1,240 Ci of rare earths, 270 Ci of ^{60}Co, and lesser amounts of ^{144}Ce, ^{95}Zr, ^{95}Nb, and ^{131}I, totaling almost 14,000 Ci. It was shown that about 200 Ci, principally cesium, were bound up with the sediments and that the remainder of the activity passed through this stretch of river in the aqueous phase (Parker *et al.*, 1966).

Folsom and Mohanrao (1960) studied the amount of gamma-emitting isotopes entering and leaving the sewage treatment plant of a large metropolitan city over a 2-year period. They found that in addition to a steady background level of fallout isotopes and natural radioactivity there were pulses or peaks of isotopes such as ^{137}Cs, ^{60}Co, ^{65}Zn, and ^{131}I entering the plant. They also found fractionation, caused by the sewage treatment process, i.e., about 55 percent of the ^{137}Cs was concentrated with the sludge, which undergoes aerobic bacterial digestion, and the remainder stayed with the liquid phase. In this particular plant, which was on the seacoast, the treated liquids and sludges were piped out to sea, where they were spread out over the sea floor and dispersed slowly.

Radioisotopes are also purposefully introduced to the marine environment in the course of scientific research projects, as tracers of physical, chemical, or biological processes. Several experiments have been carried out in the marine environment employing large quantities of short-half-life radioactive tracers to study the distribution of products from underwater chemical explosions (Gurney *et al.*, 1963). In 1961, for instance, four charges were detonated off the coast of California, at San Clemente Island. These shallow explosions of 10,000 pounds of HBX-1 were each seeded with 500 Ci of radionuclide. Three detonations employed ^{177}Lu (6.8-day half-life) in fine particulate oxide form; the fourth employed the noble gas ^{133}Xe (5.3-day half-life).

According to unpublished data of Schuert, samples collected on the day of the explosion and the day following it showed significant accumulation of the tracer by plankton (principally copepods). Activity ranging from 10^5 to 10^7 d/m/g of wet weight were measured in samples taken from water having no detectable radioactivity. Lower levels of radioactivity were measured in a number of species of fish, including jack mackerel, boccaccio, bonito shark, swell shark, and sheephead.

A number of experiments have been conducted along coastlines to study the littoral drift of sand by injecting the surf zone with batches of sand tagged with radioactivity. In these tests, the sand is either tagged directly with ^{133}Xe (similar to the kryptonating process) or coated with other nuclides and sealed with sodium silicate (Acree *et al.*, 1968). Such experiments have proven of great value in understanding the dynamics of shoreline coastal waters and harbor erosion or buildup.

Some years ago, consideration was given to the use of radioisotopes to study local diffusion and mixing processes, and several tests of this technique were conducted (Nuclear Science and Engineering Co., 1958). Since then, the technology of fluorometers has advanced considerably, and it is now possible to measure dye concentrations of the same low order as radioisotopes—0.5 parts per billion. Dyes largely have superseded isotopes in the measurement of water mixing processes. Selected radioisotopes such as ^{65}Zn and ^{32}P continue to be useful for studies of ecological systems (Pomeroy and Odum, 1966; Hooper and Ball, 1966).

SUMMARY

Radioactivity in the oceans can result from numerous sources. Natural atmospheric and geologic processes issue discrete radionuclides in certain forms and distributions; these are briefly described. Man's uses of nuclear energy result in another, wider spectrum of radionuclides of varying characteristics. The biological distribution in the ocean of man-made radionuclides depends greatly on the initial physicochemical form of the radioactivity, and this, in turn, depends on the particular use or application of nuclear energy. Nuclear energy applications are described, indicating the particular radionuclides involved in each application and the form of the radioactivity likely to be introduced into the ocean. The applications include nuclear explosions at various positions relative to the earth's surface; nuclear reactors for electric power, fuel production, ship propulsion, and space vehicle propulsion; nuclear fuel processing; waste disposal; auxiliary power generators; and radioisotopic tracers.

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Chapter Three

OCEANIC DISTRIBUTIONS OF RADIONUCLIDES FROM NUCLEAR EXPLOSIONS

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Bomb-produced radionuclides are of interest to ocean scientists for two primary reasons: (a) as potential contaminants of the ocean biosphere that could have a profound influence on the future development of some life forms and that might even present a hazard to man, and (b) as radioactive tracers for studies of water masses, sediment movement, and various biological parameters. In either case, the practical usefulness of fallout isotopes is limited by the precision of our knowledge of their temporal and spatial distribution in the oceans.

The magnitude of the difficulty of determining the distribution of bomb-produced radionuclides in the sea is in no way reflected by the amount of money or effort expended or the number of analyses performed to date. This is perhaps best illustrated by comparison with the scope of the studies of continental fallout. In 1966, for example, in the United States alone, approximately 2,000 land deposition samples were collected for ^{90}Sr analysis. The United Kingdom processed more than 800 precipitation samples for ^{90}Sr or ^{137}Cs in that year, and other European countries and Australia probably analyzed a similar number; thus, upwards of 3,500 samples of deposition on the land areas of the world were collected and analyzed in 1966. From the oceans, on the other hand, we find that since 1954, accounting for all samples from all countries a total of less than 3,000 analyses of ^{90}Sr or ^{137}Cs have been made. The rate has certainly increased in the last few years; in 1966, about 650 samples of ocean water were collected for fallout study, almost 20 percent of the number on land. Thus, we might reasonably conclude that, considering the area of the ocean surface (nearly three times that of the land) and the added dimension of depth, the data available for revealing the distribution of fallout radioisotopes in the oceans are indeed scant.

For this report, we have chosen to concentrate primarily on the distribution of ^{90}Sr in the sea and to make only brief mention of ^{14}C, ^{3}H, and other radionuclides. Based on a survey, reported in the section on the ratio of ^{137}Cs to ^{90}Sr in seawater (p. 71) ^{137}Cs is assumed to have remained well mixed with the ^{90}Sr after entering the oceans, in the more or less constant ratio generally observed in atmospheric samples. For convenience, we have approximated this ratio to be 1.5 and converted all ^{137}Cs data to ^{90}Sr accordingly. Thus, in this report, unless specifically referred to otherwise, all seawater ^{137}Cs values are expressed as the equivalent ^{90}Sr.

The motives for directing the major portion of this study to the distribution of the ^{90}Sr isotope are almost entirely dictated by very practical considerations: availability of seawater analyses and the preponderance of land data for comparison. Since the very earliest days of nuclear fallout studies, ^{90}Sr has been regarded as the fission product of greatest potential hazard to living things because of the unique combination of its relatively long 28-year half life, the very energetic beta particle of its ^{90}Y daughter, and its general resemblance to calcium in metabolic processes. For these reasons, great emphasis was put on ^{90}Sr analysis in biological as well as environmental samples, including seawater, in attempting to assay its current levels in the biosphere and to develop methods of predicting future levels. A rather large body of ^{90}Sr data has been accumulated over the last 15 years, resulting in good documentation of the distribution of that nuclide on the earth's land surfaces. Other geophysical parameters of nuclear fallout, such as its relationship to precipitation and season have also developed from these data. Consequently, for any type of synoptic analysis of deposited nuclear debris in the ocean and for comparison with land fallout, only ^{90}Sr offers enough information to be of use.

Strontium-90 also has the advantage of probably being useful as a water tracer through the apparent constancy of stable strontium in the sea at the relatively high concentration of about 8 mg per liter of water. If the fallout ^{90}Sr enters the sea in a soluble form, it must be a valid tracer of surface water.

Under the impetus of producing this chapter, many of the foremost investigators in the field were requested to contribute their data, often unpublished, for ^{90}Sr and ^{137}Cs in seawater. Practical considerations dictated that this chapter be limited to data summaries, and, therefore, in order to provide a viable reference to the original ^{90}Sr results and supporting data, they have virtually all been reported in a single volume of the Health and Safety (HASL) Quarterly Summary Report No. 197, dated July 1, 1968 (Bowen *et al.*, 1968b; Folsom *et al.*, 1968; Broecker and Simpson, 1968; Shirasawa and Schuert, 1968).

Early in the preparation of this report, it became apparent that certain areas of disagreement between some of the authors were not resolvable. The disputes concern interpretation of oceanic ^{90}Sr concentration data and, in some cases, the validity of analytical results; consensus on many of the interesting aspects and implications was impossible. Hence, in order to avoid embarrassment or withdrawal of any author, or a compromising of his views, we have remained neutral on a number of subjects. While this may appear less than satisfactory as a purely scientific endeavor, we consider the overall benefit derived from maintaining this group of authors to be of greater value.

^{90}Sr IN SURFACE OCEAN WATER

Strontium-90 has been measured in surface water samples since 1954 in the Atlantic Ocean and starting in 1957 in the Pacific Ocean. In total, almost 2,600 analyses of samples from the oceans and major seas have been performed and reported; the results are summarized in the following pages.

The concentrations of ^{90}Sr vary in the surface ocean as a function of latitude, longitude, and time in fairly complex ways, and thorough interpretation of the data is simply not possible within the scope of this volume. The two primary objectives, therefore, have been to summarize all of the available measurements in tables of practical and useful form and to provide as complete referencing as possible to the original published data and methodology. Secondarily, a few of the outstanding and noncontroversial features have been described briefly. Detailed interpretive reports on ^{90}Sr in the Atlantic surface waters (Bowen *et al.*, 1968b) and ^{137}Cs in the Pacific (Folsom *et al.*, 1968) have been included with the data compilations in the Health and Safety Laboratory Quarterly Report mentioned earlier. The variation of the ratio ^{137}Cs/^{90}Sr in surface ocean water is discussed in a later section of this chapter.

Tables 1–4 list the yearly averages of all known measurements of ^{90}Sr in surface ocean water, grouped by 10° bands of latitude, for the Atlantic, Pacific, and Indian oceans. In order to point up the striking concentration differences that have been observed, the Pacific Ocean was divided into eastern and western regions (east and west of 180°) in Tables 2 and 3. Table 5 summarizes the mean annual surface water ^{90}Sr concentrations in the major seas.

Intercomparison of Laboratory Results

Until very recently, no organized intercomparison program existed for laboratories engaged in radiostrontium analyses of seawater. Hence, for most of the period covered by Tables 1 through 5, and for most of the laboratories represented, we can compare the quality of the analyses only by comparing samples from similar areas and times. Recognizing the variability undoubtedly present over the course of a year, and within the oceanic area of a 10° latitude band, one is nevertheless virtually forced to group data in this manner, in order to observe a reasonable number of comparisons. In Tables 6, 7, and 8, examples were selected for the purpose of intercomparing both individual laboratories and countries of origin.

Tables 6, 7, and 8 are not easily interpreted without access to the original data that were averaged. The best example of a misleading case is the North Sea comparison, in Table 8. In the 22 values making up the German average of 146, several were about equivalent to the Finnish value of 392. This is, of course, partially reflected in the mean deviation error term. On balance, the agreement between laboratories seems to be quite acceptable in most cases, although a few, such as the North Atlantic Polish data and eastern Pacific equatorial data of 1961, may be exceptions.

Since 1966, the Health and Safety Laboratory (HASL) has sponsored a ^{90}Sr analysis laboratory intercomparison program. The program consists of providing any interested

TABLE 1 Mean Annual ^{90}Sr Concentrations[a] in Atlantic Ocean Surface Water by 10° Bands of Latitude

Latitude	1954		1955		1956		1957		1958		1959		1960	
Band	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
60°-70°N	–	–	–	–	–	–	–	–	–	–	–	–	3	18 (1)
50°-60°N	–	–	–	–	–	–	–	–	–	–	5	45±14 (2)	5	40±19 (6)
40°-50°N	–	–	1	111 (1)	2	19±10 (3)	2	13 (1)	2	19±1 (3)	–	–	6, 7, 12	82±61 (6)
30°-40°N	1	18 (1)	–	–	2	9±2 (3)	2, 3	11±2 (3)	2	16±5 (8)	3, 6, 12	16±4 (7)	6, 12	20±13 (5)
20°-30°N	–	–	–	–	2, 3	10±2 (6)	2	5 (1)	–	–	–	–	6, 12	19±9 (5)
10°-20°N	–	–	–	–	2	10 (1)	2, 3	8±2 (5)	2, 3	7±1 (6)	6, 12	14 (1)	8	18±4 (4)
0°-10°N	–	–	–	–	–	–	2	5±1 (2)	4	8±1 (3)	–	–	–	–
0°-10°S	–	–	–	–	–	–	4	4±1 (2)	4	4 (1)	–	–	–	–
10°-20°S	–	–	–	–	–	–	3	4±1 (2)	–	–	–	–	–	–
20°-30°S	–	–	–	–	–	–	–	–	4	7±1 (2)	–	–	–	–
30°-40°S	–	–	–	–	–	–	–	–	–	–	–	–	–	–
40°-50°S	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Total No. of Analyses		1		1		13		21		23		10		27

Latitude	1961		1962		1963		1964		1965		1966		1967	
Band	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
60°-70°N	9	23±4 (6)	15	21±2 (2)	9, 16, 17	65±15 (16)	3, 21, 22, 23	78±19 (24)	4, 57, 64	35±9 (19)	3, 27, 28	35±8 (16)	3	22±3 (2)
50°-60°N	3, 12	12±2 (5)	3	22±6 (34)	3	32±15 (27)	3	40±8 (24)	4	35±5 (37)	3	28±2 (24)	3	23±2 (11)
40°-50°N	3, 7, 10, 11, 12	27±15 (10)	3	30±10 (20)	3	42±12 (11)	3	42±10 (13)	4	46±8 (19)	3, 29, 30	45±12 (12)	3	29±4 (9)
30°-40°N	3, 11, 12, 13	22±7 (19)	3	26±7 (20)	3, 10, 18	41±10 (18)	3, 24	45±9 (19)	4	37±2 (11)	3, 29	39±4 (12)	3	36±3 (5)
20°-30°N	3, 10, 11, 12, 13	16±5 (17)	3, 10	28±6 (2)	18, 19	26±9 (6)	3, 24	39±3 (6)	4	46±2 (3)	3	35±2 (2)	–	–
10°-20°N	4, 10, 11, 12	13±3 (18)	–	–	3, 18	27±6 (10)	3, 24	25±6 (5)	4	16±1 (3)	3	21±2 (4)	3	18±1 (3)
0°-10°N	4, 12	8±3 (5)	–	–	3, 18	61±30 (12)	3, 24	28±3 (8)	4, 65	19±7 (4)	3	16±2 (4)	3	15±4 (2)
0°-10°S	4, 12, 13	8±3 (6)	–	–	3, 18	24±6 (23)	3, 24	22±4 (8)	4, 65	19±5 (4)	3	18±3 (2)	3	9 (1)
10°-20°S	12	8±3 (4)	–	–	18	16±5 (16)	24	21±4 (3)	4	28±2 (24)	3	19 (1)	3	15 (1)
20°-30°S	10, 12, 13	6±2 (7)	–	–	18, 20	21±4 (3)	24	15±4 (7)	4	45±12 (29)	–	–	–	–
30°-40°S	14	13±4 (7)	11	11±3 (5)	20	8±5 (3)	20, 24	10±5 (3)	–	–	–	–	–	–
40°-50°S	10	6 (1)	–	–	–	–	–	–	–	–	–	–	–	–
Total No. of Analyses		104		83		145		120		153		94		34

[a] ^{137}Cs data were converted to ^{90}Sr by the factor 0.67.

[b] (1) USAEC Health and Safety Laboratory (1958); (2) Bowen and Sugihara (1958); (3) Bowen *et al.* (1968b); (4) Bowen and Sugihara (1965); (5) Mauchline (1963); (6) Popov *et al.* (1962); (7) Dera *et al.* (1962); (8) Sugihara and Bowen (1962); (9) Kautsky (1968, private communication); (10) Rocco and Broecker (1963); (11) Patin *et al.* (1966); (12) Shvedov *et al.* (1963b); (13) Chulkov and Gorbunov (1963); (14) Schroeder and Cherry (1962); (15) Aarkrog *et al.* (1963b); (16) Aarkrog and Lippert (1964b); (17) Aarkrog and Lippert (1964c); (18) Ankudinov *et al.* (1967); (19) Broecker *et al.* (1966b); (20) Schroeder (1968, personal communication); (21) Umweltradioaktivitat und Strahlenbelastung (1965); (22) Aarkrog and Lippert (1965b); (23) Aarkrog and Lippert (1965c); (24) Gedeonov *et al.* (1967); (25) Aarkrog and Lippert (1966b); (26) Umweltradioaktivitat und Strahlenbelastung (1967b); (27) Aarkrog and Lippert (1967b); (28) Aarkrog and Lippert (1967c); (29) Aarkrog (1968, personal communication); (30) Umweltradioaktivitat und Strahlenbelastung (1966).

[c] dpm/100 liters ± mean deviation; numbers in parentheses are number of analyses.

TABLE 2 Mean Annual ^{90}Sr Concentrations[a] in Western Pacific Surface Water by 10° Bands of Latitude

Latitude Band	1957		1958		1959		1960		1961	
	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
60°-70° N	1	369±40 (7)	–	–	1	21±1 (2)	–	–	–	–
50°-60° N	–	–	–	–	–	–	–	–	–	–
40°-50° N	–	–	–	–	1	266 (1)	–	–	3	36±6 (1)
30°-40° N	–	–	–	–	1, 2	366±163 (3)	–	–	3, 5	37±7 (13)
20°-30° N	–	–	1	688 (1)	2	222 (1)	–	–	3, 5	62±26 (9)
10°-20° N	–	–	1, 2	121±57 (2)	2	102 (1)	–	–	3, 4, 5	35±18 (14)
0°-10° N	–	–	1	355 (1)	–	–	3	16 (1)	5	60±23 (5)
0°-10° S	–	–	–	–	–	–	3	19±7 (5)	5	44±18 (2)
10°-20° S	–	–	–	–	–	–	3	17±6 (3)	4, 5	16±1 (2)
20°-30° S	–	–	–	–	–	–	–	–	–	–
30°-40° S	–	–	–	–	–	–	–	–	–	–
40°-50° S	–	–	–	–	–	–	–	–	3	7±3 (4)
50°-60° S	–	–	–	–	–	–	–	–	3	3 (1)
60°-70° S	–	–	–	–	–	–	–	–	–	–
Total No. of Analyses		7		4		8		9		52

Latitude Band	1962		1963		1964		1965		1966	
	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
60°-70° N	–	–	–	–	–	–	–	–	3	61±9 (13)
50°-60° N	–	–	–	–	–	–	–	–	3	65±8 (40)
40°-50° N	6, 7	118 (1)	–	–	–	–	–	–	3	59±12 (42)
30°-40° N	3, 6, 7, 8	70±15 (7)	–	–	7	107 (1)	9	48±3 (3)	3	69±7 (32)
20°-30° N	3, 6, 7	64±15 (10)	–	–	7	83 (1)	–	–	3	44±6 (8)
10°-20° N	3, 6, 7	47±8 (10)	–	–	7	108 (1)	–	–	3	28±11 (14)
0°-10° N	3, 6, 7	27±4 (14)	–	–	7	45 (1)	–	–	3	21±3 (10)
0°-10° S	3	12±1 (11)	–	–	–	–	–	–	3	16±3 (15)
10°-20° S	3	12±2 (6)	–	–	–	–	–	–	3	16±2 (7)
20°-30° S	3	11±1 (2)	–	–	–	–	–	–	3	22±8 (11)
30°-40° S	–	–	–	–	–	–	–	–	3	43±14 (3)
40°-50° S	–	–	–	–	–	–	–	–	3	34±17 (4)
50°-60° S	–	–	–	–	–	–	–	–	3	9±0 (1)
60°-70° S	–	–	–	–	–	–	–	–	–	–
Total No. of Analyses		61				4		3		200

[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.

[b] (1) Miyake *et al.* (1961a); (2) Higano and Shiozaki (1960); (3) Folsom (1968, personal communication); (4) Higano *et al.* (1963a and b); (5) Popov *et al.* (1964b); (6) Popov *et al.* (1966a); (7) Tchumitchev (1966); (8) Sarahushi *et al.* (1962); (9) Shuert (1968, personal communication).

[c] dpm/100 liters ± mean deviation; numbers in parentheses are numbers of analyses.

TABLE 3 Mean Annual ^{90}Sr Concentrations[a] in Eastern Pacific Surface Water by 10° Bands of Latitude

	1959		1960		1961		1962		1963	
Latitude	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
70°-80°N	–	–	4	35 (1)	–	–	–	–	–	–
60°-70°N	1	21±1 (2)	–	–	4	41±1 (2)	4	54±4 (2)	–	–
50°-60°N	–	–	–	–	3	32±1 (3)	–	–	–	–
40°-50°N	–	–	–	–	3	34±6 (14)	–	–	–	–
30°-40°N	3	20±6 (7)	1, 2	18±6 (24)	3, 5	36±12 (24)	3, 9	54±6 (13)	–	–
20°-30°N	–	–	1, 3	26±6 (12)	3, 5, 6	36±9 (23)	3, 9	45±6 (6)	–	–
10°-20°N	–	–	3	21±4 (7)	3, 6	34±12 (13)	10, 12, 13	48±7 (7)	–	–
0°-10°N	–	–	3	12±3 (4)	3, 6, 7	32±10 (13)	10, 12, 13	49±16 (16)	–	–
0°-10°S	–	–	3	6±1 (2)	3, 6	19±10 (11)	11, 12, 13	68±15 (3)	14	10±0 (2)
10°-20°S	–	–	–	–	3, 5, 6	24±8 (8)	12, 13	73 (1)	14	7±3 (2)
20°-30°S	–	–	–	–	3, 7	11±5 (2)	–	–	–	–
30°-40°S	–	–	–	–	3	11±1 (2)	–	–	–	–
40°-50°S	–	–	–	–	3, 7	8±1 (3)	–	–	–	–
50°-60°S	–	–	–	–	3	12 (1)	–	–	–	–
60°-70°S	–	–	–	–	3	7 (1)	–	–	–	–
Total No. of Samples		9		50		120		48		4

	1964		1965		1966		1967	
Latitude	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
70°-80°N	–	–	–	–	–	–	–	–
60°-70°N	–	–	–	–	–	–	–	–
50°-60°N	15	99±1 (2)	15	95±11 (12)	3, 15	89±18 (8)	–	–
40°-50°N	3, 15	97±17 (5)	15	99±11 (11)	3, 15	113±21 (17)	3	117±7 (8)
30°-40°N	3, 15	78±30 (19)	3, 15	103±17 (25)	3, 16	128±20 (124)	3, 15	126±14 (6)
20°-30°N	3	60±11 (11)	–	–	3, 15	104±40 (110)	3	107±27 (24)
10°-20°N	–	–	–	–	3	47±9 (21)	3	45±13 (38)
0°-10°N	–	–	–	–	3	29±4 (9)	3	24±3 (29)
0°-10°S	–	–	–	–	3	22±8 (12)	3	15±2 (33)
10°-20°S	–	–	–	–	3	19±3 (14)	3	16±2 (30)
20°-30°S	–	–	–	–	3	29±7 (10)	–	–
30°-40°S	–	–	–	–	3	21±4 (30)	–	–
40°-50°S	–	–	–	–	3	24±7 (4)	–	–
50°-60°S	–	–	–	–	–	–	–	–
60°-70°S	–	–	–	–	–	–	–	–
Total No. of Samples		37		38		359		168

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[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.

[b] (1) Folsom *et al.* (1960); (2) Folsom and Mohanrao (1960); (3) Folsom (1968, personal communication); (4) Bowen and Sugihara (1964); (5) Higano *et al.* (1963a and b); (6) Popov *et al.* (1964b); (7) Rocco and Broecker (1963); (8) Mauchline (1963); (9) Saruhashi *et al.* (1962); (10) Higano *et al.* (1962); (11) Shiozaki *et al.* (1964); (12) Popov *et al.* (1966); (13) Tchumitchev (1966); (14) Broecker *et al.* (1966a); (15) Shirasawa and Schuert (1968); (16) Robertson and Perkins (1966b).

[c] dpm/100 liters ± mean deviation; numbers in parentheses are numbers of analyses.

TABLE 4 Mean Annual ^{90}Sr Concentrations[a] in Indian Ocean Surface Water by 10° Bands of Latitude

Latitude Band	1959		1960		1961		1962	
	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
20°-30°N	–	–	2	13±1 (2)	–	–	–	–
10°-20°N	–	–	1, 2	25±11 (11)	2	34±6 (2)	–	–
0°-10°N	–	–	1, 2	26±8 (8)	2	14 (1)	–	–
0°-10°S	1	24±6 (2)	1, 2, 3	23±4 (11)	2	34±9 (7)	4	13±4 (3)
10°-20°S	1	33±6 (4)	1, 2, 3	18±5 (19)	2	24±13 (4)	4	22 (1)
20°-30°S	1	28 (1)	2, 3	18±6 (6)	–	–	4	29±5 (3)
30°-40°S	1	15 (1)	2, 3	11±4 (6)	3	18±6 (2)	4	24 (1)
40°-50°S	–	–	3	4 (1)	–	–	–	–
Total No. of Analyses		8		64		16		8

[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.
[b] (1) Baranov *et al.* (1964); (2) Popov *et al.* (1964a); (3) Folsom (1968, personal communication); (4) Popov and Orlov (1967).
[c] dpm/100 liters ± mean deviation; numbers in parentheses are numbers of analyses.

analytical group with a series of "standard" seawater samples for ^{90}Sr assay. The samples are aliquots of a large volume of seawater obtained from a deep well at the Coney Island Aquarium, to some of which known amounts of ^{90}Sr were added. The basic Coney Island water contains about 40 dpm ^{90}Sr per 100 liters. The sample size is about 55 liters. By mid-1970, some 16 different laboratories from four countries had participated, and most had completed the analyses. In general, the participating laboratories represented by data in Tables 1 through 5, including the contractor organizations that carried out a portion of the Bowen *et al.* (1968; Bowen, personal communication) analyses, are proving to be in good calibration, almost all within 10 percent of the expected values. This program will be continued and hopefully expanded to include additional investigators.

Seasonal Variations

The spring peak of ^{90}Sr fallout deposition on land has been extensively documented and is generally attributed to seasonal meteorological phenomena that give rise to substantial interchanges of air between the stratosphere and troposphere. On the sea, neither direct measurements of ^{90}Sr in precipitation nor surface water samples seem to systematically reflect this annual fallout maximum at most sites.

In the North Atlantic, four weather observation ships, operated by the U.S. Coast Guard and the Environmental Sciences and Services Administration (ESSA), have been utilized as fallout collection platforms for the past several years (USAEC Health and Safety Laboratory, 1968). Similarly, in the North Pacific, the Naval Research Defense Laboratory (NRDL) has maintained a number of fallout observation posts on radar picket and weather ships. The data from these collections (Volchok, 1967; Volchok and Kleinman, 1968; Hammond *et al.*, 1966) do not uniformly indicate higher ^{90}Sr deposition during spring months, and, in several instances, suggest enhanced fallout during the winter.

At the North Atlantic weather stations, systematic monthly collections of surface seawater have been carried out for over 5 years (Bowen, personal communication; Bowen *et al.*, 1968). The ^{90}Sr concentrations from these collections, reported in Bowen *et al.* (1968), suggest the tendency for the maximum annual concentration to occur in the second half of each year, with a median peaking month of September. The authors conclude, based on detailed study of frequency and intensity of precipitation, as well as on oceanographic experience, that significant amounts of fallout are deposited in the ocean by means other than precipitation.

Schuert (1968, personal communication) reports the results of measurements of ^{90}Sr concentration in surface waters from the eastern North Pacific made periodically since late 1964 and states that no significant seasonal variation was observed. Bainbridge (1963b), reporting on surface water concentrations of tritium in the northern Pacific, presented a curve of average monthly specific activity for the years 1960 and 1961. Here the maximum tritium concentration occurred in the period December-January.

The evidence brought forth by the investigations briefly summarized above strongly indicates that the commonly observed spring peak in deposition of stratospheric ^{90}Sr on the land masses may be largely absent or substantially displaced in time on the sea. The authors cannot agree on whether or not this observation implies a difference in the basic fallout

TABLE 5 Mean Annual ^{90}Sr Concentration[a] in Surface Water of Various Seas

		North	Black	Baltic	Mediterranean	Adriatic	Tyrrhenian	Ligurian	Ionian
1958	Ref.[b]	1	–	–	–	–	–	–	–
	Conc.[c]	108 (1)	–	–	–	–	–	–	–
1959	Ref.[b]	2	5	7	–	–	–	–	–
	Conc.[c]	93±32 (2)	16±6 (16)	80 (1)	–	–	–	–	–
1960	Ref.[b]	3	5	3, 7	–	25	25	–	–
	Conc.[c]	135±25 (4)	63[d] (22)	134±77 (14)	–	40±4 (5)	14±1 (2)	–	–
1961	Ref.[b]	3, 4, 5	5	3, 7, 16, 11	5	25	19, 25	19	19
	Conc.[c]	78[d] (52)	58±12 (18)	139±72 (15)	19±13 (5)	27±6 (4)	69±44 (5)	101±49 (2)	46 (1)
1962	Ref.[b]	4, 6	13	7, 11	–	19, 25	19, 25	19	19
	Conc.[c]	87±11 (15)	115[d]	178±72 (27)	–	164±24 (10)	121±28 (5)	108±34 (5)	77 (1)
1963	Ref.[b]	7, 8, 9	13	7, 11	13, 19	19, 25, 29	13, 19, 25, 28	19	19
	Conc.[c]	153±49 (26)	140±8 (2)	151±36 (21)	85±18 (11)	108±37 (11)	111±36 (7)	62±11 (4)	59 (1)
1964	Ref.[b]	9	14	11, 17	20, 21, 22	21, 22, 26	20, 21, 22	21, 22	21, 22
	Conc.[c]	216 (1)	173±43 (13)	219±56 (18)	76±29 (7)	91±35 (11)	71±38 (8)	87±34 (8)	73±15 (2)
1965	Ref.[b]	30	15	11, 17	23, 24	23, 24, 27	23, 24	23, 24	23, 24
	Conc.[c]	73 (1)	166±9 (5)	206±31 (22)	24±13 (6)	58±29 (10)	15±5 (6)	8±4 (8)	25±21 (2)
1966	Ref.[b]	10, 11, 12, 30	–	11, 18	–	–	–	–	–
	Conc.[c]	108±20 (15)	–	176±33 (25)	–	–	–	–	–
1967	Ref.[b]	11, 30	–	11	–	–	–	–	–
	Conc.[c]	69±20 (38)	–	139±19 (24)	–	–	–	–	–

[a] ^{137}Cs data converted to ^{90}Sr by the factor 0.67.

[b] (1) Hollstein (1959); (2) Mauchline (1963); (3) Dera *et al.* (1962); (4) Umweltradioaktivitat und Strahlenbelastung (1962); Shvedov *et al.* (1963b); (6) Umweltradioaktivitat und Strahlenbelastung (1964); (7) Paakola and Voipio (1965a); (8) Aarkrog and Lippert (1964a); (9) Umweltradioaktivitat und Strahlenbelastung (1965); (10) Umweltradioaktivitat und Strahlenbelastung (1966); (11) Kautsky (1968, personal communication); (12) Umweltradioaktivitat und Strahlenbelastung (1967a); (13) Ankudinov *et al.* (1967); (14) Belyaev *et al.* (1966b); (15) Gedeonov *et al.* (1966); (16) Shvedov *et al.* (1963a); (17) Salo and Voipio (1966); (18) Salo and Voipio (1968, personal communication); (19) Argiero *et al.* (1965); (20) Gedeonov *et al.* (1967); (21) Comitato Nazionale de l'Energia Nucleare (1964); (22) Comitato Nazionale de l'Energia Nucleare (1965); (23) Comitato Nazionale de l'Energia Nucleare (1966a); (24) Comitato Nazionale de l'Energia Nucleare (1966b); (25) Cigna *et al.* (1963); (26) Rudjer Boskovic Institute (1965); (27) Rudjer Boskovic Institute (1966); (28) Schreiber (1966b); (29) Rudjer Boskovic Institute (1964).

[c] dpm/100 liters ± mean deviation; numbers in parentheses are numbers of samples.

[d] Original data not available.

TABLE 6 Atlantic Ocean ^{90}Sr^{a} Comparisons

Year	Latitude Band	Country	^{90}Sr Conc.^{b}	Reference
1959	30°-40°N	United States	16±3 (5)	Bowen *et al.*, 1968b
		Soviet Union	17±7 (2)	Popov *et al.*, 1962
1960	40°-50°N	Poland^{b}	137±53 (3)	Dera *et al.*, 1962
		Soviet Union	27±22 (3)	Popov *et al.*, 1962
1961	50°-60°N	United States	12±2 (5)	Bowen *et al.*, 1968b
		Soviet Union	12 (1)	Shvedov *et al.*, 1963b
1961	40°-50°N	United States	15±2 (2)	Bowen *et al.*, 1968b
		United States	18 (1)	Rocco and Broecker, 1963
		Soviet Union	19±4 (3)	Patin *et al.*, 1966
		Soviet Union	9 (1)	Shvedov *et al.*, 1963b
		Poland^{b}	65 (2)	Dera *et al.*, 1962
1965	60°-70°N	United States	29±5 (7)	Bowen *et al.*, 1968b
		West Germany	38±11 (9)	Kautsky, 1968, private communication
		Denmark	41 (2)	Aarkrog *et al.*, 1966b

^{a} ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.

^{b}dpm/100 liters ± mean deviation; numbers in parentheses are numbers of analyses.

^{c}These were sampled near shore, perhaps accounting for the high value.

TABLE 7 Pacific Ocean ^{90}Sr^{a} Comparisons

Year	Latitude Band	Country	^{90}Sr Conc.^{b}	Reference
Western Pacific				
1961	20°-30°N	United States	47±9 (4)	Folsom *et al.*, 1968
		Soviet Union	75±40 (5)	Popov *et al.*, 1964b
1961	10°-20°S	Japan	16 (1)	Higano *et al.*, 1963a and b
		Soviet Union	15 (1)	Popov *et al.*, 1964b
Eastern Pacific				
1961	0°-10°N	United States	17±1 (2)	Folsom *et al.*, 1968
		United States	6 (1)	Rocco and Broecker, 1963
		Soviet Union	45±12 (10)	Popov *et al.*, 1964
1962	20°-30°N	United States	43±6 (5)	Folsom *et al.*, 1968
		Japan	53 (1)	Saruhashi *et al.*, 1962
1967	30°-40°N	United States	124±14 (5)	Folsom *et al.*, 1968
		United States	134 (1)	Shirasawa and Schuert, 1968

^{a} ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.

^{b}dpm/100 liters ± mean deviation; numbers in parentheses are numbers of analyses.

TABLE 8 ^{90}Sr[a] Comparisons in the Indian Ocean and Other Seas

Year	Comparison	Country	^{90}Sr Conc.[b]	Reference
1960	Indian Ocean	United States	23±1 (4)	Folsom *et al.*, 1968
		Soviet Union	29±8 (3)	Baranov *et al.*, 1964
		Soviet Union	19±3 (4)	Popov *et al.*, 1964
1963	Tyrrhenian Sea	Soviet Union	101 (1)	Ankudinov *et al.*, 1967
		Italy	76±16 (2)	Cigna *et al.*, 1963
		Italy	108±48 (3)	Argiero *et al.*, 1965
		Italy	92 (1)	Schreiber, 1966b
1963	Mediterranean	Italy	88±22 (2)	Argiero *et al.*, 1965
		Soviet Union	84±17 (9)	Ankudinov *et al.*, 1967
1963	North Sea	Finland	392 (1)	Paakola and Voipio, 1965b
		Denmark	122±6 (3)	Aarkrog and Lippert, 1964a
		West Germany	146±70 (22)	Umweltradioaktivitat und Strahlenbelastung, 1964

[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.
[b] dpm/100 liters ± mean deviation; numbers in parentheses are numbers of analyses.

mechanism for deposition on land and sea. A hypothesis dealing with these data and explaining the relationships has been advanced by Bowen *et al.* (1968) and in summary suggests more frequent and less geographically confined penetrations of the over-ocean tropopause than believed to be the case over land.

^{90}Sr IN OCEAN WATER FROM SURFACE TO 700 METERS

Atlantic Ocean

Measurements of ^{90}Sr in water samples from depths of 700 m or less in the Atlantic Ocean have been collected; they are summarized in Tables 9 and 10. In the section of this chapter on the inventory of ^{90}Sr in the ocean (p. 62), these data were used to compute the total ^{90}Sr deposit in the oceans. Other features of these profiles also deserve emphasis–their average shape; their trend with time, when the data are expressed as fractions of the surface water concentration; and the correlation of shape with latitude.

The average shape of the curves of ^{90}Sr versus depth to 700 m is relatively smooth, with the maximum at the surface and rarely falling below 10 percent of the surface value at 700 m. Figure 1 shows the mean profiles for samples from north of 30°N, and between 30°N and 20°S. The apparent secondary maximum at 250 m is very probably an artifact of the averaging; very few samples are available from this depth. Compared to the shape, discussed below (p. 75), of depth profiles for ^{95}Zr-Nb, ^{144}Ce, or ^{147}Pm (each believed to move vertically largely as particulate material), the scarcity of subsurface concentration maxima is striking. In those cases that are well established (Rocco and Broecker, 1963; Bowen and Sugihara, 1960, 1965), the hydrographic situation has commonly indicated subsurface lateral advection along isopycnal surfaces outcropping to the surface further north or south. These curves appear to be clearly inconsistent with the idea that significant amounts of fallout ^{90}Sr sink by association with particles. In this context, especially convincing is the lack of any evidence for ^{90}Sr

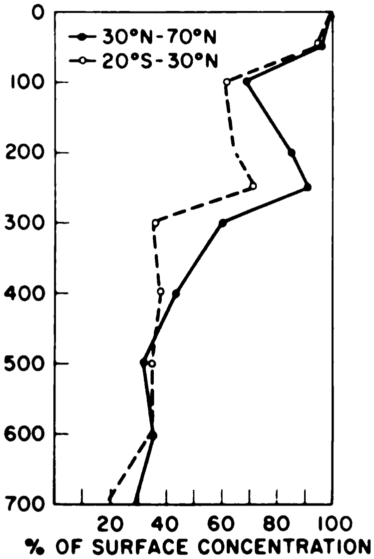

FIGURE 1 Mean profiles of relative ^{90}Sr in surface and intermediate Atlantic Ocean water.

TABLE 9 Mean ^{90}Sr^{a} Concentrations in Atlantic Ocean Surface to 700-m Profiles—Northern Hemisphere

	1957		1958		1959		1960		1961	
Depth (m)	Ref.^{b}	Conc.^{c}	Ref.^{b}	Conc.^{c}	Ref.^{b}	Conc.^{c}	Ref.^{b}	Conc.^{c}	Ref.^{b}	Conc.^{c}
60°-70°N										
Surface	–	–	–	–	–	–	1	18±2 (1)	–	–
30	–	–	–	–	–	–	–	–	–	–
100	–	–	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	1	67% (1)	–	–
700	–	–	–	–	–	–	1	45% (1)	–	–
50°-60°N										
Surface	–	–	–	–	–	–	–	–	–	–
100	–	–	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	–	–	–	–
700	–	–	–	–	–	–	–	–	–	–
40°-50°N										
Surface	–	–	–	–	–	–	9, 10, 11	82±61 (6)	–	–
30	–	–	–	–	–	–	10	184% (1)	–	–
50	–	–	–	–	–	–	10	184% (1)	–	–
60	–	–	–	–	–	–	–	–	–	–
100	–	–	–	–	–	–	–	–	–	–
200	–	–	–	–	–	–	–	–	–	–
250	–	–	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	10	132% (1)	–	–
500	–	–	–	–	–	–	–	–	–	–
600	–	–	–	–	–	–	–	–	–	–
700	–	–	–	–	–	–	–	–	–	–
30°-40°N										
Surface	3, 15	11±2 (8)	15	16±5 (8)	3, 9, 11	16±3 (7)	–	–	3, 11, 19, 20	22±7 (19)
50	–	–	3, 17	55% (2)	–	–	–	–	8	114% (1)
100	16	70% (1)	16, 17	57% (6)	8, 18	57% (3)	–	–	8	36% (2)
300	–	–	16, 17	50% (3)	8, 18	91% (2)	–	–	8	27% (2)
400	16	35% (1)	–	–	–	–	–	–	–	–
500	15	35% (1)	16, 17	28% (3)	8, 18	53% (2)	–	–	8	41% (1)
600	–	–	–	–	–	–	–	–	–	–
700	16	21% (1)	17	22% (1)	8	20% (1)	–	–	8	20% (1)
20°-30°N										
Surface	–	–	–	–	–	–	–	–	3, 11, 19, 20, 21	16±5 (17)
50	–	–	–	–	–	–	–	–	11	94% (1)
100	–	–	–	–	–	–	–	–	21	70% (3)
200	–	–	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	–	–	8, 21	30% (2)
400	–	–	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	–	–	8, 21	10% (3)
700	–	–	–	–	–	–	–	–	8	5% (1)
10°-20°N										
Surface	3, 15	8±2 (5)	–	–	–	–	26	18±4 (4)	11, 19, 21, 25	13±3 (18)
50	–	–	–	–	–	–	–	–	11	107% (2)
100	16	68% (1)	–	–	–	–	25	100% (3)	11, 25	42% (2)
250	25	55% (1)	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	–	–	25	35% (1)
400	16	27% (1)	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	25	50% (1)	25	15% (1)
700	–	–	–	–	–	–	25	20% (1)	25	15% (1)

1962		1963		1964		1965		1966		1967	
Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
–	–	–	–	2, 3, 4, 5	78±18 (24)	–	–	3, 6, 7	35±8 (16)	–	–
–	–	–	–	–	–	–	–	7	89% (2)	–	–
–	–	–	–	5	48% (4)	–	–	7	100% (2)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
3	22±6 (34)	–	–	3	40±8 (34)	–	–	3	28±2 (24)	–	–
8	46% (5)	–	–	8	48% (1)	–	–	–	–	–	–
8	39% (3)	–	–	8	50% (2)	–	–	–	–	–	–
8	28% (4)	–	–	9	14% (2)	–	–	–	–	–	–
8	31% (2)	–	–	8	30% (2)	–	–	8	65% (1)	–	–
–	–	–	–	3	42±10 (13)	–	–	3, 12, 13	43±12 (29)	3	29±4 (9)
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	14	115% (1)	–	–
–	–	–	–	–	–	–	–	12, 14	105% (3)	8	120% (2)
–	–	–	–	–	–	–	–	14	85% (1)	–	–
–	–	–	–	–	–	–	–	14	91% (2)	–	–
–	–	–	–	8	28% (2)	–	–	12, 14	71% (2)	8	105% (2)
–	–	–	–	8	13% (2)	–	–	14	60% (2)	–	–
–	–	–	–	–	–	–	–	12, 14	41% (4)	–	–
–	–	–	–	8	17% (1)	–	–	14	29% (1)	–	–
–	–	3, 21, 22	41±10 (19)	3, 23	46±9 (14)	–	–	3, 12	39±4 (12)	–	–
–	–	–	–	8	83% (2)	–	–	3	105% (1)	–	–
–	–	–	–	8	71% (1)	–	–	3, 12	116% (1)	–	–
–	–	–	–	8	36% (1)	–	–	3, 12	67% (1)	–	–
–	–	8	54% (1)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	3	14% (1)	–	–
–	–	–	–	–	–	–	–	12	26% (1)	–	–
–	–	–	–	8	18% (1)	–	–	–	–	–	–
3, 21	28±6 (3)	22, 24	26±9 (6)	3, 23	39±3 (6)	–	–	3	35±2 (2)	–	–
–	–	–	–	–	–	–	–	8	117% (1)	–	–
8	76% (1)	24	131% (1)	8	115% (1)	–	–	8	130% (1)	–	–
–	–	24	90% (1)	–	–	–	–	–	–	–	–
8	44% (1)	24	41% (1)	8	74% (2)	–	–	8	74% (1)	–	–
–	–	24	33% (1)	–	–	–	–	–	–	–	–
8	18% (1)	24	15% (1)	8	54% (1)	–	–	8	16% (1)	–	–
8	11% (1)	24	4% (1)	8	36% (2)	–	–	–	–	–	–
–	–	3, 22	27±6 (10)	–	–	–	–	3	21±2 (4)	–	–
–	–	8	42% (1)	–	–	–	–	–	–	–	–
–	–	8	57% (4)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	8	25% (4)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	8	14% (4)	–	–	–	–	8	5% (1)	–	–
–	–	8	13% (4)	–	–	–	–	–	–	–	–

continued overleaf

TABLE 9 (Continued)

	1957		1958		1959		1960		1961	
Depth (m)	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
1°-10°N										
Surface	–	–	–	–	–	–	–	–	11, 25	8±3 (5)
100	–	–	–	–	–	–	–	–	25	28% (1)
200	–	–	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	–	–	25	26% (2)
400	–	–	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	–	–	25	10% (1)
600	–	–	–	–	–	–	–	–	–	–
700	–	–	–	–	–	–	–	–	25	12% (2)

[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.
[b] See Table 10 for references.

"pile-up" in a layer just above the thermocline, as shown so clearly by Chesselet and co-workers (1964a, 1965a) for particulate radionuclides like ^{95}Zr-Nb and ^{144}Ce.

Expressed as fraction of the surface water concentration, profiles versus depth do not show any systematic trend with time. This seems to show a very remarkable mobility of the upper 700 m of the ocean. One would have expected rather to see, at times of high annual average fallout increment (Bowen *et al.*, 1968b), a decrease in the *percentage* concentrations present at depth, and at times of low annual fallout increment, the reverse. Examination of the original data (Dera *et al.*, 1962; Ankudinov *et al.*, 1967) shows that in several cases that appear as anomalously high percentages in Table 6, the same values, expressed as fraction of the *simultaneously measured* surface value, fall comfortably into line. The hypothesis that this uniformity represents simply a constant contamination of each deep sample we take to be contraindicated by the diversity of sampling gear used; in a few reports (Bodman *et al.*, 1961; Rocco and Broecker, 1963; Bowen and Sugihara, 1963, 1965), salinity determinations on the samples analyzed effectively rule out this possibility. Unfortunately, not enough stations are available to determine whether there may be a seasonal variation in shape of these curves, as would be suggested by an analysis such as that in Bowen *et al.* (1968b, Figure 6). Assuming that the data are correct, one might see in them evidence of unusual speed of "down-mixing" of fallout ^{90}Sr in the oceans.

As shown in Figure 1, the Atlantic Ocean data are fitted by two distinct curves, one representing stations north of 30°N, the other, those between 20°S and 30°N. It is evident from such discussions as those of Ryther and Menzel (1960) and Schroeder *et al.* (1959) that in the North Atlantic 30°N is about the southern limit of areas where water 18°C or colder usually crops out at the surface in winter. The data from the depths representing distinct differences between the two curves, 100 to 400 m, suggest that this may be caused by outcropping of 15° to 18°C water. It is, however, surprising that the effect is so large; as discussed by Koshlyakov (1967), for instance, a boundary closer to 50°N has mostly been predicted, depending on the combination of high average wind velocity and strong winter cooling of the surface ocean. Some data, still unpublished (Bowen, 1968, personal communication) but represented in the 1967 profiles 40°-50°N (Table 6) indicates the depth of mixing is much greater at these latitudes on the eastern side of the ocean; there may be evidence here for unsuspected organized shallow currents.

Pacific Ocean

^{90}Sr concentration profiles to depths of 700 m in the Pacific Ocean are displayed in Tables 11 and 12.

The three profiles of relative ^{90}Sr concentrations shown in Figure 2 are composites of a number of analyses for each point in the water mass grouped to illustrate the gross differences. The deepest penetration is seen in the Northwest Pacific, Station Victor, with almost constant concentrations from the surface down to about 250 m. At 700 m, the composite concentration for these profiles is seen to be about 35 percent of the surface value. Several reasons for this evidence of deep penetration have been put forth. Reid (1965) and others have presented hydrographic evidence indicating that relatively short paths may exist between surface areas a

1962		1963		1964		1965		1966		1967	
Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
–	–	3, 22	61±30 (12)	3, 23	28±3 (8)	–	–	–	–	–	–
–	–	3	18% (4)	23	89% (1)	–	–	–	–	–	–
–	–	22	31% (1)	23	61% (3)	–	–	–	–	–	–
–	–	3	9% (3)	–	–	–	–	–	–	–	–
–	–	–	–	23	37% (2)	–	–	–	–	–	–
–	–	3	10% (3)	–	–	–	–	–	–	–	–
–	–	22	40% (1)	–	–	–	–	–	–	–	–
–	–	3	12% (3)	–	–	–	–	–	–	–	–

[c] Surface values are dpm/100 liters ± mean deviation or reported error if only one analysis; subsurface values are percentages of surface values; numbers in parentheses are numbers of analyses.

few hundred miles further north and water masses passing 400–600 m below Station Victor. Also, a gyre center lies nearby, suggesting local recycling. Hence, ^{90}Sr fallout may not only penetrate quickly but also may stay longer at moderate depths in this western oceanic region.

In the Northeast Pacific Ocean, at Station November (30°N, 140°W), the composite profile is somewhat modified, indicating less penetration, not measurable at 700 m. Here, surface and excess of surface values persist down past 150 m, after which the concentrations decrease rather rapidly.

In the California Current and coastal waters, generally from about 10° to 33°N and 115° to 133°W, as Figure 2 clearly shows, the decrease with depth is even more rapid. In 1967, subsurface concentrations in individual profiles were found at as much as four times the surface values near 20°N. These maxima appeared in strata having low salinities, suggesting origin at the surface near 40°N. This further indicates a rate of thousands of miles of travel over 1-4 years with little destruction of water properties. Similar implications arise from inspection of certain salinity gradients in these strata and also from the relatively long persistence of certain radionuclide-tagged water masses that have been injected at intermediate depths (Folsom and Vine, 1957).

It is apparent then that fallout may persist so long at moderate depths that input patterns become distorted. Fallout budgets in the Pacific must take account of this. However, this persistence may afford means for following progress of intermediate waters more than half way round the Pacific.

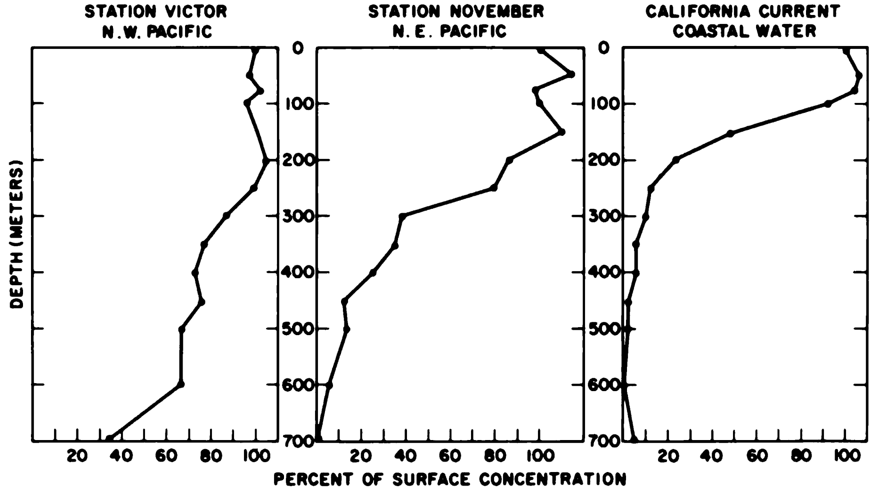

FIGURE 2 Mean profiles of relative ^{90}Sr activities in surface and intermediate Pacific Ocean water.

TABLE 10 Mean ^{90}Sr[a] Concentrations in Atlantic Ocean Surface to 700-m Profiles–Equatorial and Southern Hemisphere

	1957		1958		1959		1960		1961	
Depth (m)	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
1°N-1°S										
Surface	–	–	25	6±2 (1)	–	–	–	–	11, 20, 25	10±1 (1)
30	–	–	–	–	–	–	–	–	11	86% (1)
60	–	–	–	–	–	–	–	–	11	141% (3)
100	–	–	18, 25	75% (4)	–	–	–	–	11, 25	52% (2)
200	–	–	–	–	–	–	–	–	–	–
300	–	–	18, 25	79% (2)	–	–	–	–	25	40% (2)
500	–	–	18, 25	30% (1)	–	–	–	–	25	30% (1)
600	–	–	–	–	–	–	–	–	–	–
700	–	–	18, 25	45% (1)	–	–	–	–	25	20% (1)
1°-10°S										
Surface	25	4±1 (2)	–	–	–	–	–	–	–	–
30	–	–	–	–	–	–	–	–	–	–
50	–	–	–	–	–	–	–	–	–	–
100	–	–	–	–	–	–	–	–	–	–
200	–	–	–	–	–	–	–	–	–	–
300	15	25% (1)	–	–	–	–	–	–	–	–
400	–	–	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	–	–	–	–
600	–	–	–	–	–	–	–	–	–	–
700	–	–	–	–	–	–	–	–	–	–
	8	ND (2)								
10°-20°S										
Surface	–	–	–	–	–	–	–	–	–	–
250	–	–	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	–	–	–	–
30°-40°S										
Surface	–	–	–	–	–	–	–	–	27	13±4 (6)
100	–	–	–	–	–	–	–	–	21	92% (1)
200	–	–	–	–	–	–	–	–	21	46% (1)
300	–	–	–	–	–	–	–	–	21	29% (1)
500	–	–	–	–	–	–	–	–	21	18% (1)
40°-50°S										
Surface	–	–	–	–	–	–	–	–	21	6±1 (1)
200	–	–	–	–	–	–	–	–	21	270% (1)
400	–	–	–	–	–	–	–	–	21	81% (1)
600	–	–	–	–	–	–	–	–	21	68% (1)

[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.

[b] (1) Bowen (1968, personal communication); (2) Aarkrog and Lippert (1965b); (3) Bowen *et al.* (1968b); (4) Umweltradioaktivitat und Strahlenbelastung (1965); (5) Aarkrog and Lippert (1965c); (6) Aarkrog and Lippert (1965b); (7) Aarkrog and Lippert (1965c); (8) Bowen *et al.* (1968, personal communication); (9) Popov *et al.* (1962); (10) Dera *et al.* (1962); (11) Shvedov *et al.* (1963b); (12) Aarkrog (1968, personal communication); (13) Umweltradioaktivitat und Strahlenbelastung (1966); (14) Kautsky (1968); (15) Bowen and Sugihara (1958);

1962		1963		1964		1965		1966		1967	
Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]	Ref.[b]	Conc.[c]
–	–	3, 22	18±2 (1)	3, 23	28±2 (2)	3	14±3 (2)	3	17±3 (1)	–	–
–	–	8	72% (3)	–	–	8	100% (2)	8	94% (1)	–	–
–	–	8	43% (3)	–	–	8	100% (2)	8	100% (1)	–	–
–	–	8	15% (1)	–	–	8	50% (2)	8	35% (1)	–	–
–	–	22	230% (2)	23	51% (2)	–	–	–	–	–	–
–	–	8, 22	108% (2)	–	–	8	7% (2)	8	5% (1)	–	–
–	–	8, 22	72% (1)	–	–	8	5% (2)	8	6% (1)	–	–
–	–	–	–	23	30% (1)	–	–	–	–	–	–
–	–	–	–	–	–	–	40% (2)	–	–	–	–
–	–	3, 22	19±5 (1)	3, 23	22±3 (8)	3	12±1 (1)	–	–	3	9±1 (1)
–	–	–	–	–	–	–	–	–	–	8	100% (1)
–	–	–	–	–	–	–	–	–	–	8	100% (1)
–	–	8	21% (1)	23	68% (2)	–	–	–	–	8	66% (1)
–	–	–	–	23	68% (2)	–	–	–	–	–	–
–	–	8	26% (1)	23	73% (1)	–	–	–	–	8	16% (1)
–	–	22	41% (5)	–	–	–	–	–	–	–	–
–	–	8	16% (1)	–	–	–	–	–	–	–	–
–	–	22	34% (5)	23	40% (1)	–	–	–	–	–	–
–	–	–	–	–	–	8	15% (1)	–	–	–	–
–	–	22	16±5 (16)	–	–	–	–	–	–	–	–
–	–	22	81% (2)	–	–	–	–	–	–	–	–
–	–	22	75% (3)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–

(16) Bowen and Sugihara (1960); (17) Bowen and Sugihara (1963); (18) Bowen (1961); (19) Patin *et al.* (1966); (20) Chulkov and Gorbunov (1963); (21) Rocco and Broecker (1963); (22) Ankudinov *et al.* (1967); (23) Gedeonov *et al.* (1967); (24) Broecker *et al.* (1966); (25) Bowen and Sugihara (1965); (26) Sugihara and Bowen (1962); (27) Schroeder and Cherry (1962).

[c] Surface values are dpm/100 liters ± mean deviation or reported error if only one analysis; subsurface values are percentages of surface values; parentheses are numbers of analyses; ND = no detectable.

TABLE 11 $^{90}Sr^{a}$ Concentration Profiles in the Eastern Pacific Ocean, 0–700 m

	1960		1961		1962		1963		1964		1965		1966		1967	
Depth (m)[b]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]
60°–70°N																
0	–	–	2	41±1 (2)[e]	2	54±2 (2)[e]	–	–	–	–	–	–	–	–	–	–
30	–	–	2	107% (2)[e]	2	93% (2)[e]	–	–	–	–	–	–	–	–	–	–
50°–60°N																
0	–	–	–	–	–	–	–	–	11	99±1 (2)	11	95±11 (12)	4, 11	89±18 (8)	–	–
30	–	–	–	–	–	–	–	–	–	–	11	111% (1)	11	93% (1)	–	–
40	–	–	–	–	–	–	–	–	11	80% (1)	–	–	–	–	–	–
50	–	–	–	–	–	–	–	–	–	–	11	93% (3)	11	87% (1)	–	–
150	–	–	–	–	–	–	–	–	11	25% (1)	11	59% (4)	11	52% (2)	–	–
40°–50°N																
0	–	–	–	–	–	–	–	–	4, 11	97±17 (5)	–	–	–	–	–	–
50	–	–	–	–	–	–	–	–	11	111% (1)	–	–	–	–	–	–
150	–	–	–	–	–	–	–	–	11	71% (1)	–	–	–	–	–	–
300	–	–	–	–	–	–	–	–	11	19% (1)	–	–	–	–	–	–
450	–	–	–	–	–	–	–	–	11	3% (1)	–	–	–	–	–	–
700	–	–	–	–	–	–	–	–	11	0% (1)	–	–	–	–	–	–
30°–40°N																
0	1, 3	18±6 (24)	–	–	1, 16	54±6 (13)	–	–	4, 11	78±13 (19)	4, 11	103±17 (25)	4, 14	128±20 (124)	4, 11	126±14 (6)
10	–	–	–	–	–	–	–	–	–	–	–	–	14	110% (1)	–	–
40	1	161% (1)	–	–	–	–	–	–	–	–	11	113% (2)	14	121% (1)	–	–
50	–	–	–	–	–	–	–	–	–	–	13	94% (1)	13, 14	124% (3)	13	131% (2)
60	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	129% (2)
70	–	–	–	–	–	–	–	–	11	107% (1)	–	–	–	–	13	125% (2)
80	–	–	–	–	–	–	–	–	–	–	13	73% (1)	13, 14	111% (2)	13	141% (2)
90	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	115% (2)
100	–	–	–	–	–	–	–	–	–	–	11, 13	42% (3)	13, 14	125% (6)	13	114% (2)
150	–	–	–	–	16	133% (1)	–	–	11	60% (1)	13	52% (1)	13, 14	115% (6)	13	113% (6)
200	–	–	–	–	–	–	–	–	–	–	11, 13	26% (6)	13, 14	81% (9)	13, 14	72% (5)
250	–	–	–	–	–	–	–	–	–	–	11, 13	13% (2)	13, 14	62% (3)	13	62% (2)
300	1	61% (1)	–	–	–	–	–	–	11	40% (1)	13	7% (3)	13	48% (3)	13, 14	30% (2)
350	–	–	–	–	–	–	–	–	–	–	13	5% (2)	13, 14	21% (3)	13, 14	18% (3)
400	–	–	–	–	–	–	–	–	–	–	11, 13	5% (3)	13	24% (2)	13, 14	12% (3)
450	–	–	–	–	–	–	–	–	–	–	–	–	13	11% (1)	13	8% (2)
500	–	–	–	–	–	–	–	–	11	1% (1)	13	5% (1)	13	12% (3)	13, 14	5% (2)
550	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	0% (1)
600	1	<22% (1)	–	–	–	–	–	–	–	–	13	2% (1)	–	–	13, 14	2% (3)
650	–	–	–	–	–	–	–	–	–	–	13	2% (2)	–	–	–	–
700	–	–	–	–	–	–	–	–	–	–	11	1% (1)	–	–	13	0% (2)

TABLE 11 (Continued)

	1960		1961		1962		1963		1964		1965		1966		1967	
Depth (m)^b	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}	Ref.^{c}	Conc.^{d}
20°–30°N																
0	–	–	–	–	–	–	–	–	–	–	–	–	4, 11	104±4 (110)	–	–
10	–	–	–	–	–	–	–	–	–	–	–	–	11	41% (1)	–	–
20	–	–	–	–	–	–	–	–	–	–	–	–	11	40% (1)	–	–
50	–	–	–	–	–	–	–	–	–	–	–	–	13	101% (1)	–	–
80	–	–	–	–	–	–	–	–	–	–	–	–	11, 13	80% (2)	–	–
100	–	–	–	–	–	–	–	–	–	–	–	–	13	74% (1)	–	–
150	–	–	–	–	–	–	–	–	–	–	–	–	13	91% (1)	–	–
200	–	–	–	–	–	–	–	–	–	–	–	–	13	103% (1)	–	–
250	–	–	–	–	–	–	–	–	–	–	–	–	11, 13	52% (4)	–	–
300	–	–	–	–	–	–	–	–	–	–	–	–	13	34% (3)	–	–
350	–	–	–	–	–	–	–	–	–	–	–	–	13	30% (3)	–	–
400	–	–	–	–	–	–	–	–	–	–	–	–	13	13% (1)	–	–
450	–	–	–	–	–	–	–	–	–	–	–	–	13	11% (1)	–	–
500	–	–	–	–	–	–	–	–	–	–	–	–	13	11% (2)	–	–
650	–	–	–	–	–	–	–	–	–	–	–	–	11	0% (1)	–	–
10°–20°N																
0	–	–	4, 7	34±12 (13)	12, 17, 18	48±7 (7)	–	–	–	–	–	–	–	–	4	45±13 (38)
40	–	–	6, 7, 9	103% (1)	–	–	–	–	–	–	–	–	–	–	–	–
50	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	302% (1)
60	–	–	6, 7, 9	118% (1)	–	–	–	–	–	–	–	–	–	–	–	–
80	–	–	6, 7, 9	103% (1)	–	–	–	–	–	–	–	–	–	–	13	622% (1)
100	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	251% (1)
150	–	–	–	–	17	100% (2)	–	–	–	–	–	–	–	–	13	533% (1)
200	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	67% (1)
250	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	40% (1)
300	–	–	–	–	17	40% (3)	–	–	–	–	–	–	–	–	13	29% (1)
350	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	40% (1)
400	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	20% (1)
450	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	11% (1)
500	–	–	6, 7, 9	103% (1)	–	–	–	–	–	–	–	–	–	–	13	4% (1)
700	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	29% (1)

continued overleaf

TABLE 11 (Continued)

Depth (m)[b]	1960 Ref.[c]	1960 Conc.[d]	1961 Ref.[c]	1961 Conc.[d]	1962 Ref.[c]	1962 Conc.[d]	1963 Ref.[c]	1963 Conc.[d]	1964 Ref.[c]	1964 Conc.[d]	1965 Ref.[c]	1965 Conc.[d]	1966 Ref.[c]	1966 Conc.[d]	1967 Ref.[c]	1967 Conc.[d]
0°-10°N																
0	–	–	4, 7, 8	32±10 (13)	12, 17, 18	49±16 (16)	–	–	–	–	–	–	–	–	4	24±3 (29)
50	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	200% (1)
100	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	313% (1)
150	–	–	6, 7, 9	150% (2)	17	39% (5)	–	–	–	–	–	–	–	–	13	25% (1)
200	–	–	8	9% (1)	–	–	–	–	–	–	–	–	–	–	13	8% (1)
250	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	8% (1)
300	–	–	–	–	17	9% (5)	–	–	–	–	–	–	–	–	13	0% (1)
350	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	0% (1)
400	–	–	8	1% (1)	–	–	–	–	–	–	–	–	–	–	13	8% (1)
450	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	0% (1)
500	–	–	6, 7, 9	116% (1)	–	–	–	–	–	–	–	–	–	–	13	0% (1)
600	–	–	8	0% (1)	–	–	–	–	–	–	–	–	–	–	–	–
700	–	–	–	–	–	–	–	–	–	–	–	–	–	–	13	0% (1)
0°-10°S																
0	–	–	4, 7	19±10 (11)	12, 17, 18	68±15 (2)	10	10±0 (2)	–	–	–	–	–	–	–	–
30	–	–	–	–	–	–	10	166% (1)	–	–	–	–	–	–	–	–
100	–	–	–	–	–	–	10	101% (2)	–	–	–	–	–	–	–	–
200	–	–	6, 7, 9	153% (3)	17	35% (1)	–	–	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	10	14% (1)	–	–	–	–	–	–	–	–
400	–	–	6, 7, 9	121% (1)	–	–	10	14% (2)	–	–	–	–	–	–	–	–
10°-20°S																
0	–	–	4, 5, 7	24±8 (8)	–	–	10	7±3 (2)	–	–	–	–	–	–	–	–
100	–	–	–	–	–	–	10	109% (2)	–	–	–	–	–	–	–	–
200	–	–	6, 7, 9	146% (1)	–	–	10	59% (2)	–	–	–	–	–	–	–	–
300	–	–	–	–	–	–	10	22% (2)	–	–	–	–	–	–	–	–
400	–	–	–	–	–	–	10	11% (2)	–	–	–	–	–	–	–	–
500	–	–	–	–	–	–	10	19% (2)	–	–	–	–	–	–	–	–
600	–	–	–	–	–	–	10	10% (1)	–	–	–	–	–	–	–	–
700	–	–	–	–	–	–	10	3% (2)	–	–	–	–	–	–	–	–
20°-30°S																
0	–	–	4, 8	11±5 (2)	–	–	–	–	–	–	–	–	–	–	–	–
400	–	–	8	0% (1)	–	–	–	–	–	–	–	–	–	–	–	–

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[a] ^{137}Cs values converted to ^{90}Sr by the factor 1.5.
[b] Depths are rounded off to nearest 10 m for 0–100 m and nearest 50 m for deeper samples.
[c] See Table 12 for references.
[d] Surface values are dpm/100 liters ± mean deviation; subsurface values are percent of surface value; numbers in parentheses are number of analyses.
[e] These samples are actually from the Chukchi Sea.

TABLE 12 ^{90}Sr[a] Concentration Profiles in the Western Pacific Ocean, 0–700 m

	1961		1963		1964		1965		1966	
Depth (m)[b]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]
30°–40°N										
0	3, 7	37±7 (13)	–	–	–	–	11	48±5 (3)	4	69±7 (32)
30	–	–	–	–	–	–	11	106% (1)	–	–
50	–	–	–	–	–	–	–	–	13	92% (1)
70	–	–	–	–	–	–	11	94% (1)	–	–
80	–	–	–	–	–	–	–	–	13	103% (1)
100	–	–	–	–	–	–	–	–	13	129% (1)
150	–	–	–	–	–	–	11	85% (1)	13	107% (2)
200	–	–	–	–	–	–	–	–	13	135% (6)
250	–	–	–	–	–	–	–	–	13	102% (5)
300	–	–	–	–	–	–	11	80% (1)	13	107% (5)
350	–	–	–	–	–	–	–	–	13	82% (4)
400	–	–	–	–	–	–	–	–	13	79% (4)
450	–	–	–	–	–	–	–	–	13	76% (1)
500	15	95% (1)[e]	–	–	–	–	11	48% (1)	13	77% (2)
600	–	–	–	–	–	–	–	–	13	68% (2)
700	–	–	–	–	–	–	–	–	13	41% (2)
20°–30°N										
0	–	–	–	–	12	83 (1)	–	–	–	–
100	–	–	–	–	12	117% (1)	–	–	–	–
10°–20°N										
0	4, 5, 7	35±18 (14)	–	–	12	108 (1)	–	–	–	–
100	–	–	–	–	12	58% (1)	–	–	–	–
200	6, 7, 9	49% (1)	–	–	–	–	–	–	–	–
350	6, 7, 9	57% (1)	–	–	–	–	–	–	–	–
500	6, 7, 9	29% (1)	–	–	–	–	–	–	–	–
0°–10°N										
0	7	60±23 (5)	12	127±0 (1)[f]	12	47±5 (6)[f]	–	–	–	–
100	–	–	12	57% (1)[f]	12	87% (1)	–	–	–	–
150	–	–	–	–	12	121% (2)	–	–	–	–
200	6, 7, 9	32% (1)	–	–	–	–	–	–	–	–
250	–	–	–	–	12	36% (2)[f]	–	–	–	–
500	6, 7, 9	45% (1)	–	–	–	–	–	–	–	–
0°–10°S										
0	7	44±18 (2)	–	–	–	–	–	–	–	–
200	6, 7, 9	75% (1)	–	–	–	–	–	–	–	–

[a] ^{137}Cs values converted to ^{90}Sr by the factor 1.5.

[b] Depths are rounded off to nearest 10 m for 0–100 m and nearest 50 m for deeper samples.

[c] (1) Folsom and Mohanrao (1960); (2) Bowen and Sugihara (1964); (3) Folsom *et al.* (1968); (5) Higano *et al.* (1963a); (6) Popov *et al.* (1963a); (7) Popov *et al.* (1964b); (8) Rocco and Broecker (1963); (9) Popov *et al.* (1966a); (10) Broecker *et al.* (1966a); (11) Shirasawa and Schuert (1968); (12) Tchumitchev (1966); (13) Folsom (1968); (14) Robertson and Perkins (1966); (15) Miyake *et al.* (1964); (16) Saruhashi *et al.* (1962); (17) Higano *et al.* (1962); (18) Popov *et al.* (1966a).

[d] Surface values are dpm/100 liters ± mean deviation; subsurface values are percent of surface values; parentheses indicate number of analyses.

[e] Coastal sample.

[f] Samples obtained from 180°00′ long.

^{90}Sr IN DEEP OCEAN WATER

As briefly outlined and extensively referenced in earlier sections, the observations of ^{90}Sr in the surface and intermediate depths of the oceans have been usefully applied to oceanography, meteorology, and related fields. The ^{90}Sr data from deep ocean water, however, while less abundant because of the expense and difficulty in obtaining good samples, have become a source of considerable debate in the scientific literature. The core of the dispute lies in the findings (Bowen and Sugihara, 1958, 1960, 1965; Bowen *et al.*, 1966; Miyake *et al.*, 1961b, 1962a) of low but measurable concentrations of ^{90}Sr at depths of greater than 1,000 m in virtually all parts sampled of both major oceans. These data

TABLE 13 Mean ^{90}Sr Concentrations in Atlantic Ocean and Caribbean Sea Deep Water

	1957		1958		1959		1960		1961	
Depth (m)	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]
60°-70°N										
1,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	1	10±1 (1)	–	–
2,000	–	–	–	–	–	–	–	–	–	–
50°-60°N										
1,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
2,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
3,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
40°-50°N										
1,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
2,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
3,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
4,000	–	–	–	–	–	–	–	–	–	–
5,000	–	–	–	–	–	–	–	–	–	–
30°-40°N										
1,000	–	–	1, 3	2 ±0.9 (4)	1	3.5±0.3 (1)	–	–	1	0.9±0.1 (3)
	1	1 ±1 (1)	–	–	1	6.6±0.6 (1)	–	–	1	1.5±0.5 (2)
2,000	–	–	–	–	–	–	–	–	1	1 ±0.5 (2)
	–	–	–	–	–	–	–	–	–	–
3,000	–	–	–	–	–	–	–	–	–	–
	–	–	1	3.8±0.4 (1)	–	–	–	–	–	–
4,000	–	–	1	4.1±0.6 (1)	–	–	–	–	–	–
	–	–	1	2.6±0.3 (1)	–	–	–	–	–	–
20°-30°N										
1,000	–	–	–	–	–	–	–	–	1	3 ±1 (1)
	–	–	–	–	–	–	–	–	1	2 ±1 (1)
2,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	5	1.4±0.3 (1)
3,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
4,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
5,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–

are in direct conflict with conclusions concerning the rates of vertical mixing based on ^{14}C measurements (Broecker *et al.*, 1960; Bien *et al.*, 1963) and also with other investigators' deep water ^{90}Sr and ^{137}Cs analyses (Rocco and Broecker, 1963; Broecker *et al.*, 1966a). All the available deep water analyses of ^{90}Sr are summarized in Tables 13 (Atlantic Ocean and Caribbean Sea) and 14 (Pacific Ocean).

A consensus was impossible among the authors of this section, regarding the subject of ^{90}Sr or ^{137}Cs in the deep ocean. Hence, to obtain the most current views of the concerned scientist, each was invited to submit an essay of approximately 500 words to be published unabridged in this volume. The three submitted papers, plus tabulations of deep water profiles in both the Atlantic and Pacific oceans follow.

1962		1963		1964		1965		1966		1967	
Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
1	8 ±3 (4)	–	–	–	–	–	–	–	–	–	–
1	3 ±1 (3)	–	–	–	–	–	–	–	–	–	–
1	1.5±0.5 (3)	–	–	1	1.5±0.5 (1)	–	–	–	–	–	–
1	2 ±1 (1)	–	–	–	–	–	–	–	–	–	–
–	–	–	–	1	1.5±0.6 (1)	–	–	–	–	–	–
1	4 ±1 (1)	–	–	1	<1 (1)	–	–	–	–	–	–
–	–	–	–	1	4.5±2.5 (2)	–	–	2	3.1±0.3	–	–
–	–	–	–	1	3 ±3 (3)	–	–	2	4.5±1.1 (4)	–	–
–	–	–	–	1	3 ±2 (2)	–	–	2	2.7±0.8 (2)	–	–
–	–	–	–	–	–	–	–	2	1.8±0.3 (2)	–	–
–	–	–	–	1	1 ±0.4 (4)	–	–	2	2.9±0.1 (2)	–	–
–	–	–	–	1	2.3±0.4 (1)	–	–	–	–	–	–
–	–	–	–	–	–	–	–	2	3.4±1.1 (4)	–	–
–	–	–	–	1	4 ±1 (1)	–	–	2	3.8±2.0 (4)	–	–
–	–	–	–	–	–	–	–	4	4.2±1.7 (2)	–	–
–	–	–	–	1	1 ±0.3 (1)	–	–	–	–	–	–
–	–	–	–	1	6 ±2 (1)	–	–	4	2.8±1.7 (2)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	1	1 ±1 (1)	–	–	–	–	–	–
–	–	–	–	–	–	–	–	4	2.8±1 (2)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
1	1 ±2 (1)	1	2.5±3.6 (1)	1	14 ±0.5 (1)	–	–	1	1.3±0.0 (1)	–	–
–	–	1, 6	0.5±1 (2)	1	0.5±0.3 (2)	–	–	–	–	–	–
1	1.5±1.4 (1)	1, 6	0.5±1 (2)	–	–	–	–	1	4.2±0.0 (1)	–	–
–	–	6	0.7±1.5 (1)	1	10 ±0.5 (1)	–	–	–	–	–	–
–	–	1, 6	1.1±1.3 (3)	1	<0.5 (1)	–	–	1	1.5±0.0 (1)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	1.5±2 (1)	1	0.5±0.2 (1)	–	–	–	–	–	–
–	–	6	4 ±2 (1)	–	–	–	–	1	2.5±0.4 (1)	–	–
–	–	6	1.2±0.5 (1)	–	–	–	–	–	–	–	–
–	–	6	1.4±0.6 (1)	–	–	–	–	1	1.1±0.0 (1)	–	–

continued overleaf

TABLE 13 (Continued)

	1957		1958		1959		1960		1961	
Depth (m)	Ref.^{a}	Conc.^{b}	Ref.^{a}	Conc.^{b}	Ref.^{a}	Conc.^{b}	Ref.^{a}	Conc.^{b}	Ref.^{a}	Conc.^{b}
10°-20°N										
1,000	–	–	–	–	–	–	–	–	5	0 ±0.4 (1)
	–	–	–	–	–	–	–	–	7	3 ±0.3 (1)
	7	1.6±0.8 (1)	–	–	–	–	–	–	5	0.5±0.4 (1)
	–	–	–	–	–	–	–	–	7	4.5±0.4 (1)
2,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	7	2.8±0.3 (1)
3,000	–	–	–	–	–	–	–	–	5	5 ±1 (2)
	–	–	–	–	–	–	–	–	–	–
4,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
5,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
0°-10°N										
1,000	7	<0.3 (2)	–	–	–	–	–	–	7	3.5±0.3 (1)
	–	–	–	–	–	–	–	–	–	–
2,000	–	–	–	–	–	–	–	–	–	–
	7	0.3±0.6 (2)	–	–	–	–	–	–	–	–
3,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
4,000	–	–	–	–	–	–	–	–	–	–
	7	<0.5 (2)	–	–	–	–	–	–	–	–
5,000	–	–	–	–	–	–	–	–	–	–
0°-10°S										
1,000	7	<0.6 (1)	7	2.5±0.3 (2)	–	–	–	–	7	4.2±0.2 (1)
	–	–	–	–	–	–	–	–	7	3.2±0.2 (1)
2,000	–	–	7	2.4±0.3 (2)	–	–	–	–	–	–
	7	<0.4 (2)	–	–	–	–	–	–	7	3.2±0.5 (1)
3,000	–	–	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
4,000	7	<0.9 (1)	–	–	–	–	–	–	–	–
	7	<0.6 (1)	–	–	–	–	–	–	–	–
5,000	–	–	–	–	–	–	–	–	–	–
20°-30°S										
1,000	7	1.5±0.6 (1)	–	–	–	–	–	–	–	–
	–	–	–	–	–	–	–	–	–	–
2,000	–	–	–	–	–	–	–	–	–	–
	7	<1 (1)	–	–	–	–	–	–	–	–
3,000	7	<1.6 (1)	–	–	–	–	–	–	–	–
30°-40°S										
1,000	–	–	–	–	–	–	–	–	5	2.6±1 (1)
	–	–	–	–	–	–	–	–	–	–
2,000	–	–	–	–	–	–	–	–	–	–
40°-50°S										
1,000	–	–	–	–	–	–	–	–	5	15 ±1 (1)
	–	–	–	–	–	–	–	–	5	2.2±0.5 (1)
2,000	–	–	–	–	–	–	–	–	–	–

[a] (1) Bowen *et al.* (1968); (2) Kautsky (1968); (3) Bowen and Sugihara (1963); (4) Aarkrog (1968); (5) Rocco and Broecker (1963); (6) Broecker *et al.* (1966a); (7) Bowen and Sugihara (1965); (8) Gedeonov *et al.* (1967); (9) Ankudinov *et al.* (1967).

1962		1963		1964		1965		1966		1967	
Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]	Ref.[a]	Conc.[b]
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	3.5±0.5 (2)	–	–	–	–	1	0.8± 0.4 (1)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	2 ±0.6 (2)	–	–	–	–	1	1.5±0.2 (1)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	5 ±1 (2)	1	5 ±0.3 (1)	–	–	1	0.5±0.2 (1)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	1	<0.7 (1)	–	–	–	–	–	–
–	–	1	1.5±0.5 (1)	–	–	–	–	1	0.4±0.3 (1)	–	–
–	–	1	2 ±2 (1)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	1	0.6±0.5 (1)	–	–
–	–	1	3.5±3[c] (6)	8	10 ±7 (2)	1	0.3±0.4 (2)	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	5 ±3 (1)	–	–	–	–	–	–	–	–
–	–	1	4 ±2 (2)	–	–	–	–	–	–	–	–
–	–	1	1 ±1 (1)	–	–	1	0 ±0.4 (1)	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	3 ±4 (1)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	1	1.1±0.4 (1)	–	–	–	–
–	–	1	7 ±1 (1)	–	–	–	–	–	–	–	–
–	–	1	1 ±4 (3)[c]	8	5.5±0.5 (4)	–	–	1	0.6±0.4 (1)	–	–
–	–	1	0.5±1 (1)	–	–	–	–	–	–	–	–
–	–	1	1 ±0.5 (1)	8	4 ±1 (3)	–	–	1	0.9±0.7 (1)	–	–
–	–	1	2 ±3 (1)	–	–	–	–	–	–	–	–
–	–	1	5 ±2 (1)	–	–	–	–	1	0 ±0.6 (1)	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	1	0 ±1 (1)	–	–	–	–	1	0 ±0.6 (1)	–	–
–	–	9	5 (1)	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–
–	–	–	–	–	–	–	–	–	–	–	–

[b] Values are dpm/100 liters ± mean deviation or reported error if only one analysis; numbers in parentheses are number of analyses.

[c] Data omitted from average, 2 values in 0°–10°N, 1 value in 0°–10°S.

TABLE 14 Mean $^{90}Sr^{a}$ Concentrations in Pacific Ocean Deep Water

	1958		1959		1960		1961		1963		1964		1965		1966		1967	
Depth (m)[b]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]	Ref.[c]	Conc.[d]
50°-60°N																		
800	–	–	–	–	–	–	–	–	–	–	5	3.6±0.2 (1)	5	1.4±1.4 (4)	5	<0.5 (2)	–	–
1,000	–	–	–	–	–	–	–	–	–	–	–	–	–	–	3	1 ±1 (1)	–	–
1,300	–	–	–	–	–	–	–	–	–	–	–	–	5	<0.5 (1)	–	–	–	–
1,500	–	–	–	–	–	–	–	–	–	–	5	2.6±0.1 (1)	5	68.6±5 (1)	3, 5	1 ±1.3 (3)	–	–
2,400	–	–	–	–	–	–	–	–	–	–	–	–	5	1.8±0.2 (1)	–	–	–	–
2,500	–	–	–	–	–	–	–	–	–	–	5	6.7±0.2 (1)	5	2.8±1 (2)	5	<0.5 (2)	–	–
40°-50°N																		
800	–	–	–	–	–	–	–	–	–	–	–	–	–	–	3	6 ±1 (1)	–	–
1,300	–	–	–	–	–	–	–	–	–	–	5	<0.5 (1)	–	–	–	–	–	–
2,100	–	–	1	46.6±4 (1)	–	–	–	–	–	–	5	<0.5 (1)	–	–	–	–	–	–
3,000	–	–	1	26.6±4 (1)	–	–	–	–	–	–	–	–	–	–	–	–	–	–
5,000	–	–	1	28.9±4 (1)	–	–	–	–	–	–	–	–	–	–	–	–	–	–
6,000	–	–	1	0 (1)	–	–	–	–	–	–	–	–	–	–	–	–	–	–
30°-40°N																		
800	–	–	–	–	–	–	–	–	–	–	–	–	3, 5	3.0±1 (3)	–	–	–	–
900	–	–	–	–	–	–	–	–	–	–	–	–	5, 8	1.4±1 (3)	3	3 ±1 (1)	–	–
1,000	–	–	–	–	–	–	–	–	–	–	–	–	8	1.3±1 (2)	3	1 ±1 (1)	5	3.2±1 (1)
1,100	–	–	–	–	–	–	–	–	–	–	–	–	8	0.3 (1)	–	–	–	–
1,200	–	–	–	–	–	–	–	–	–	–	5	<0.5 (1)	–	–	–	–	5	0 (1)
1,300	–	–	–	–	–	–	–	–	–	–	–	–	5	<0.5 (1)	–	–	–	–
1,500	–	–	–	–	–	–	–	–	–	–	–	–	–	–	3	3 ±1 (1)	–	–
2,000	–	–	–	–	–	–	–	–	–	–	–	–	3, 8	4.3±3 (3)	3	0 ±1 (1)	–	–
2,100	–	–	–	–	–	–	–	–	–	–	–	–	8	2 (1)	–	–	–	–
2,400	–	–	–	–	–	–	–	–	–	–	–	–	5	1.3±1 (2)	–	–	–	–
2,700	–	–	–	–	–	–	–	–	–	–	5	<0.5 (1)	–	–	–	–	–	–
3,000	–	–	–	–	–	–	–	–	–	–	–	–	5	0.8±0.5 (1)	–	–	–	–
4,000	–	–	–	–	–	–	–	–	–	–	–	–	3	0 ±2 (1)	3	0 ±1 (1)	–	–
4,400	–	–	–	–	–	–	–	–	–	–	–	–	5	1.9±5 (1)	–	–	–	–
6,000	–	–	–	–	–	–	–	–	–	–	–	–	3	3 ±2 (1)	–	–	–	–
8,000	–	–	–	–	1	0 (1)	–	–	–	–	–	–	–	–	–	–	–	–
20°-30°N																		
800	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
900	–	–	–	–	–	–	6, 7	9 ±4 (1)	–	–	–	–	3	5 ±3 (2)	–	–	–	–
1,000	1	133±11 (1)	–	–	–	–	–	–	–	–	–	–	3	1 ±1 (2)	3	5 ±1 (1)	–	–
1,300	–	–	–	–	–	–	–	–	–	–	–	–	3	0 ±2 (2)	–	–	–	–
1,500	–	–	–	–	–	–	–	–	–	–	–	–	3	1 ±1 (2)	–	–	–	–
2,000	–	–	–	–	–	–	–	–	–	–	–	–	3	2 ±1 (2)	–	–	–	–
3,000	1	133±11 (1)	–	–	–	–	–	–	–	–	–	–	3	2 ±1 (2)	–	–	–	–
4,000	–	–	–	–	1	15.5±2 (1)	–	–	–	–	–	–	3	1 ±1 (2)	–	–	–	–
5,000	–	–	–	–	–	–	–	–	–	–	–	–	3	2 ±2 (2)	–	–	–	–
5,500	–	–	–	–	–	–	6, 7	4 ±1 (1)	–	–	–	–	–	–	–	–	–	–

10°-20°N																		
900	–	–	–	–	–	–	6, 7	1.4±1 (1)	–	–	–	–	–	–	–	–	–	–
1,000	–	–	–	–	–	–	6, 7	20 ±5 (4)	3, 4	1 ±2 (1)	–	–	–	–	–	–	–	–
2,000	–	–	–	–	–	–	6, 7	16 ±4 (2)	–	–	–	–	–	–	–	–	–	–
2,600	–	–	–	–	–	–	–	–	–	–	–	–	3	0 ±1 (1)	–	–	–	–
3,000	1	26.6±4 (1)	1	44.4±4 (1)	–	–	–	–	–	–	–	–	–	–	–	–	–	–
3,500	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
4,000	–	–	–	–	–	–	6, 7	17 ±3 (2)	3, 4	0.7±2 (1)	–	–	–	–	–	–	–	–
4,800	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
0°-10°N																		
800	–	–	–	–	–	–	2	0.1±1 (1)	–	–	–	–	–	–	–	–	–	–
900	–	–	–	–	–	–	6, 7	11 ±2 (1)	–	–	–	–	–	–	–	–	–	–
1,000	–	–	–	–	–	–	6, 7	17 ±6 (4)	3, 4	1.7±2 (1)	–	–	3	0 ±1 (1)	–	–	–	–
1,500	–	–	–	–	–	–	–	–	–	–	–	–	3	2 ±1 (1)	–	–	–	–
2,000	–	–	–	–	–	–	6, 7	9 ±2 (4)	3, 4	0 ±2 (1)	–	–	3	6 ±1 (1)	–	–	–	–
3,000	1	31.1±4 (1)	–	–	–	–	6, 7	7 ±3 (1)	–	–	–	–	–	–	–	–	–	–
3,500	–	–	–	–	–	–	–	–	–	–	–	–	3	3 ±1 (1)	–	–	–	–
4,000	–	–	–	–	–	–	6, 7	14 ±6 (1)	–	–	–	–	–	–	–	–	–	–
4,800	–	–	–	–	–	–	–	–	–	–	–	–	3	3 ±1 (1)	–	–	–	–
0°-10°S																		
1,000	6, 7	9±3 (2)	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
2,000	6, 7	10±7 (3)	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
3,000	6, 7	19±6 (1)	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
4,000	6, 7	1±1 (1)	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
5,600	–	–	–	–	–	–	–	–	–	–	–	–	3	0 ±1 (1)	–	–	–	–
10°-20°S																		
1,000	–	–	–	–	–	–	2, 6, 7	3 ±3 (3)	3, 4	1 ±2 (2)	–	–	3	1.5±1 (2)	–	–	–	–
1,300	–	–	–	–	–	–	2	0.7±0.5 (1)	–	–	–	–	–	–	–	–	–	–
1,500	–	–	–	–	–	–	–	–	3, 4	0.2±2 (1)	–	–	3	0 ±1 (1)	–	–	–	–
2,000	–	–	–	–	–	–	6, 7	<4 (2)	3, 4	0 ±2 (1)	–	–	–	–	–	–	–	–
2,500	–	–	–	–	–	–	–	–	3, 4	5 ±2 (1)	–	–	–	–	–	–	–	–
2,600	–	–	–	–	–	–	–	–	–	–	–	–	3	0.7±1 (1)	–	–	–	–
3,000	–	–	–	–	–	–	6, 7	<4 (2)	3, 4	0.5±2 (1)	–	–	3	2 ±1 (1)	–	–	–	–
3,600	–	–	–	–	–	–	2	0 ±2 (1)	–	–	–	–	–	–	–	–	–	–
4,000	–	–	–	–	–	–	–	–	3, 4	1 ±2 (2)	–	–	–	–	–	–	–	–
4,500	–	–	–	–	–	–	–	–	–	–	–	–	3	0 ±1 (2)	–	–	–	–
6,300	–	–	–	–	–	–	–	–	–	–	–	–	3	0 ±1 (1)	–	–	–	–

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[a] ^{137}Cs measurements were converted to ^{90}Sr by the factor 0.67.

[b] Rounded off to the nearest 100 m.

[c] (1) Miyake *et al.* (1962a); (2) Rocco and Broecker (1963); (3) Broecker and Simpson (1968); (4) Broecker *et al.* (1966a); (5) Shirasawa and Schuert (1968); (6) Popov *et al.* (1964b); (7) Popov *et al.* (1966a); (8) Folsom *et al.* (1968).

[d] Values are dpm/100 liters ± mean deviation or reported error if only one value; numbers in parentheses are number of analyses.

Evaluation of Deep Water Data

Wallace S. Broecker

If, as reported by Bowen and Sugihara (1960) and Miyake *et al.* (1962a), a significant fraction of the ^{90}Sr and ^{137}Cs added to the oceans by fallout of nuclear test debris have reached depths greater than 1,000 m in the ocean, then both the generally accepted rates of vertical mixing and the inventories of fallout production must be seriously in error. As I consider both of these estimates to rest on firm self-consistent evidence, I cannot accept the validity of any deep-water results that require this conclusion. The following paragraphs are a defense of this position.

The most difficult aspect of any measurements involving trace amounts of any radioactivity is the estimation of the blank. Errors due to other sources, such as chemical yield and counter efficiency, are relatively easy to pin down. The fact that the surface-water data from various laboratories is reasonably consistent rules out the possibility that disagreement on the analytical results for deep waters stems from any analytical error other than the blank. I feel that the investigations that yielded the most anomalous results (Bowen and Sugihara, 1960; Miyake *et al.*, 1962) did not adequately demonstrate that the deep-water results were not solely the result of contamination. Such obvious checks as running ^{90}Sr-free water, analyzing duplicates, and reanalyzing processed seawater were not reported. Indeed, in the case of the Woods Hole group who initially claimed to have no measurable blank, it was subsequently found that one of the reagents used in their analyses indeed contained enough ^{90}Sr to give a few dpm/100 liters.

Another reason for doubting the deep-water data is its lack of internal consistency. If ^{90}Sr is being rapidly carried to the deep sea, then the deep-water values should rise roughly in accord with the total amount of fallout added to the oceans. In 1960, Bowen and Sugihara reported finding several dpm/100 liters in samples collected in 1958 over a wide range of depth in the deep equatorial Atlantic. Their values reported here for samples collected in 1965 and 1966 from the same latitude bands average less than 1 dpm/100 liters. Other investigators have obtained similar low deep-water results. It is easier for me to believe that this marks a reduction in blank rather than a severalfold decrease in the deep-water ^{90}Sr inventory. Although the significance of the fraction of a dpm/100 liter values found in 1965 and 1966 can still be disputed, they are not nearly as difficult to live with as the earlier data. The roughly threefold increase in the surface values reduce by an order of magnitude the inequity between the mixing rates required to explain the fallout results and those based on natural radiocarbon. The inventory problem is also reduced to more manageable proportions.

In June 1965, the Lamont group took a series of samples in the area studied by Miyake and his co-workers. Instead of finding tens of dpm of ^{90}Sr and ^{137}Cs, we found almost no measurable activities for these isotopes. Thus, even if his analyses were valid, they reflect no more than a local anomaly and therefore cannot be used to calculate either oceanic mixing rates or fallout inventories.

I think it is safe to say that any finite amounts of ^{90}Sr and ^{137}Cs found prior to 1965 on samples from below 1,500 m are almost certainly due to undetected blanks. In fact, for this period, I feel that the best estimate of an investigator's blank is his results on deep-water samples. If they are reasonably consistent, they serve as a fair correction for the near-surface-water results. If they show a large spread, the validity of even the surface values for that investigator are open to serious question.

Penetration of Fallout Strontium-90 into Deep Water of the Atlantic Ocean

Vaughan T. Bowen

In Table 13 are collected and averaged 188 measurements of the concentration of strontium-90 in samples from 1,000 m or deeper in the Atlantic Ocean and Caribbean Sea; of the data available, four numbers only are omitted, all from Ankudinov *et al.* (1967), whose 1963 1,000-m samples are so high as to indicate probable sample contamination or, possibly, pretripping of the sampler. In two other instances, conflicting data points have been presented without averaging: in 1961, Rocco and Broecker (1963) found negligible ^{90}Sr at 1,000 and 1,500 m in the Caribbean Sea, whereas Bowen and Sugihara (1965) found measurable amounts at these depths in the eastern Sargasso.

Of the laboratories reporting these analyses, most have shown that they can extract from some seawater samples strontium containing less strontium-90 than they can detect; this is true for Ankudinov *et al.* and Gedeonov *et al.*, as it is for Bowen and his co-workers or for Broecker and his. In the excepted cases—Aarkrog (Denmark) and Kautsky (Germany)—the considerable bodies of data include enough analyses, 50 percent or less of their means for deep samples, to convince me they are not suffering seriously from "blank" problems. In spite of this, we have only one report (Broecker *et al.*, 1966a) in which a majority (4 of 7) of the samples were below detection limits; even in this case, no ^{137}Cs value was found to be zero! Although much has been made of them, Rocco and Broecker (1963) found only two below-detection ^{90}Sr values, of eight 1,000-m or deeper samples. Reference to Table 1 shows that other bodies of data show smaller proportions still. Clearly, the data available are consistent only with the view that most samples withdrawn from the deep Atlantic Ocean contain measurable amounts of strontium-90.

Since the depth of mixing during severe winter storms at high latitudes in both the North and the South Atlantic often exceeds 1,000 m, and since isotherms found much deeper than this at low latitudes reach depths from surface to only a few hundred meters, the mechanism for the observed penetration of ^{90}Sr is reasonably clear. Wüst and Defant, Stommel and Arons have described, either from geostrophic analysis of oceanographic density profiles or from theoretical considerations, patterns of narrow, rapidly moving deep currents capable of transporting down-mixed ^{90}Sr from high latitudes of either ocean to the equator in periods measurable in years or even months. It is of interest in this context that the data from 1957 show measurable ^{90}Sr at 1,000–1,500 m in both oceans at latitudes above 10°, but none about the equator; this was the last time so considerable a mass of deep Atlantic water was shown to be unlabeled.

The apparent contradiction between this data and that for the carbon-14 concentration of deep-ocean water has caused much concern, enough to induce others to suggest that ^{90}Sr, contrary to the extensive published predictions of its behavior as a solute in seawater, may be sinking with some unknown population of particles, as Schuert has suggested may be the case for ^{137}Cs. In the absence of positive evidence for this, the arguments already published appear conclusive, especially reinforced with the facts that rainfall ^{90}Sr is collectible with high efficiency on ion-exchange columns, that high ^{90}Sr has never coincided with the intermediate depth peaks of cerium-144 or promethium-147, which appear to represent real populations of labeled sinking particles, and that in the Baltic Sea, Salo and Voipio (1966, personal communication) have seen no evidence for deep penetration of ^{90}Sr except as a consequence of obvious injections of new labeled seawater. This seems especially convincing since the depths involved are so small (about 200 m), the halocline limiting down-mixing is so excessively steep, and the only-brackish surface water provides the smallest concentrations of stable Sr as "hold-back carrier" available in any well-studied marine situation.

It seems to me unavoidable to conclude that strontium-90 has penetrated deep Atlantic and Caribbean waters and that in doing so, it must have acted as a tracer of real water movements. That this same conclusion also follows from analysis of our extensive body of surface water ^{90}Sr values, I have demonstrated elsewhere (Bowen *et al.*, 1968b).

^{90}Sr and ^{137}Cs in Deep Ocean Water

Edward A. Schuert

The existence of trace quantities of the global fallout radionuclides ^{90}Sr and ^{137}Cs in deep ocean water has been reported by a number of investigators. In most cases, the quantities observed have been at such low levels that they have challenged the analytical techniques available. Nevertheless, enough positive evidence has been reported to form a consensus in favor of the existence of ^{90}Sr and ^{137}Cs at depths as great as 5,000 m.

Recent research by the author and T. Shirasawa of the NRDL may shed light on this problem. An in situ pump-adsorber system was developed and was used off the coast of California (32°N, 130°W) to extract ^{137}Cs from large volumes of seawater using the inorganic ion-exchange material KCFC. Profiles taken to 2,000 m, well into the deep water, were analyzed for both stable cesium and ^{137}Cs. This technique permitted the acquisition of uncontaminated samples from effective volumes of over 1,000 liters and further permitted an absolute error estimate to be placed on the results. Two profiles taken in January and June of 1967 showed positive evidence of ^{137}Cs to 340 m. Samples taken at 380 m, 450 m, 700 m, 970 m, 1,200 m, and 1,800 m contained no ^{137}Cs within the limits of detection (0 ± 0.2 dpm/100 liters). Water samples taken at the same time and processed in the laboratory showed trace amounts of ^{137}Cs existing as deep as 1,000 m.

The above unpublished results suggest that ionic ^{137}Cs is not mixing across the intermediate waters into the deep domain. The question of translocation by particulate or by the biomass remains to be evaluated. However, it should be pointed out that if ^{137}Cs or ^{90}Sr are to be exploited as tracers of ocean water masses, only the ionic component should be measured.

I feel that ionic ^{137}Cs and ^{90}Sr are excellent tracers for studying ocean circulation and mixing processes and that in the absence of advective processes, as in the case studied above, there has been no exchange between surface and deep waters in the time frame represented by the history of global fallout.

Data reporting the existence of global fallout at depth should be reviewed from the point of view of advective and other translocation mechanisms such as particulate and biomass activity.

THE INVENTORY OF ^{90}Sr IN THE OCEAN

Because of the limited number of ^{90}Sr and ^{137}Cs measurements and the disagreement regarding the validity of the deep water values, any overall oceanic inventory is bound to be subject to rather large uncertainties. For this reason, we have chosen an approach that lends itself to a variety of assumptions regarding the distribution of fallout.

The ^{90}Sr concentration C_x, at any depth x, in the ocean can be approximated by the following equation:

$$C_x = (C_S - C_D)\, e^{-\frac{0.693x}{x_{1/2}}} + C_D, \qquad (1)$$

where C_S and C_D are, respectively, the concentrations of ^{90}Sr at the surface and in the deep water mass. The parameter $x_{1/2}$ (called the half mixing depth) is the depth at which the surface water excess (i.e., $C_S - C_D$) decreases by a factor of two. Figure 3 diagrammatically represents the parameters of Eq. (1). Note that, for flexibility in computation, this method defines any parcel of water as consisting of both a "surface" and "deep" component. The surface concentration, $C_S - C_D$, decreases as described above; the deep concentration C_D remains constant throughout the depth of the sea.

Signifying the mean depth of the ocean by $\bar{x}$, and recognizing that $\bar{x}$ is large compared to $x_{1/2}$, Eq. (1) can be integrated with the following result:

$$C = C_D\bar{x} + (C_S - C_D)\frac{x_{1/2}}{0.693}. \qquad (2)$$

Since C_D and C_S are generally in units of dpm/100 liters and $\bar{x}$ and $x_{1/2}$ are in meters, the integrated activity would come out in the unit, dpm meter/100 liters. This is readily convertible to the commonly used fallout unit mCi/km^2 by the factor 4.5×10^{-3}. Taking $\bar{x}$ to be 3.8×10^3 m, we then have (in units of mCi/km^2)

$$C = 17C_D + 6.5 \times 10^{-3}\,(C_S - C_D)\,x_{1/2}. \qquad (3)$$

Before doing any detailed calculation, it is worthwhile to get some feeling for the relative magnitude of the two terms in the integration. To do this, we will consider the ocean as a whole. Regardless of how the deep water data are interpreted, a reasonable maximum for the oceanwide average for C_D would be 3 dpm/100 liters (see Table 12). Hence, the upper limit on the inventory of the deep component is given by

$$C_D\bar{x}A_O \leqq 17 \times 3 \times 3.6 \times 10^8 \leqq 18 \times 10^9 \text{ mCi}$$

or

$$C_D\bar{x}A_O \leqq 18 \text{ MCi},$$

where A_O equals the area of the entire world ocean, 3.6×10^8 km^2.

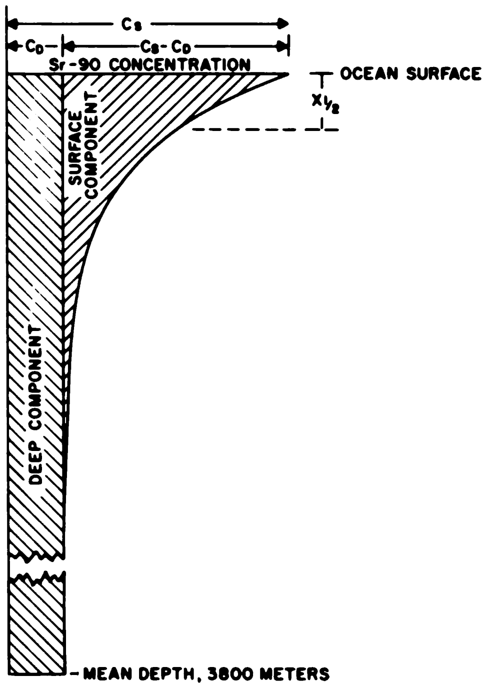

FIGURE 3 Diagrammatic representation of the parameters for computing oceanic ^{90}Sr inventories.

In estimating the surface component of the total inventory of ^{90}Sr (from Table 1 and Figures 1 and 2), if we take $x_{1/2}$ to be 200 m (it probably ranges from 50 m to at least 500 m) and $C_S - C_D$ to be 30 dpm/100 liters (it ranges from 7 to at least 100 dpm/100 liters) we obtain

$$(C_S - C_D)\frac{x_{1/2}}{0.693}A_O = 6.5 \times 10^{-3} \times 30 \times 200 \times 3.6 \times 10^8$$

$$= 14 \times 10^9 \text{ mCi}$$

$$= 14 \text{ MCi}.$$

Clearly, both terms must be considered!

Complete inventories of ^{90}Sr in the oceans have been carried out for 1961 and 1966. These years were picked because substantial data are available at almost all regions and depths. It seems quite clear, based upon the information developed in the earlier sections, that the surface ^{90}Sr values, C_S, are high enough in concentration and well enough cross-checked as to be open to very little question. The half mix-

TABLE 15 Oceanic Areas and Half Mixing Depths

Latitude	Area (10^8 km^2) Atlantic	Pacific	Total	$x_{1/2}$ (m)
>30°N	0.23	0.41	0.64	200
0–30°N	0.30	0.61	0.91	100
0–30°S	0.26	0.72	0.98	100
>30°S	0.26	0.80	1.06	200

TABLE 16 Surface Concentrations of ^{90}Sr (dpm/100 liters)

Latitude	Atlantic 1961	Atlantic 1966	Pacific 1961	Pacific 1966	Weighted Average 1961	Weighted Average 1966
>30°N	22	37	40	95	34	74
0–30°N	14	22	39	91	31	68
0–30°S	7	18	22	23	18	22
>30°S	12	18[a]	8	21	9	20

[a]Estimated value.

ing depth, $x_{1/2}$, and the average deep water concentration, C_D, are somewhat in doubt; hence, to establish limits on the inventories, "reasonable" limits for these two parameters were used. For the deep water, values of 0 and 3 dpm per 100 liters were chosen. While the intermediate depth profiles of Tables 9 and 10 and Figures 1 and 2 do not really permit definition of a systematic geographic variation, we have divided the main ocean bodies of each hemisphere into regimes north and south of 30° and assigned values for the half mixing depths of 100 m for the low-latitude regions and 200 m for the high latitudes. The areas used in the computation and the assigned half mixing depths are listed in Table 15.

The values of the surface water concentrations of ^{90}Sr were obtained from the data in Tables 1, 2, and 3. These are listed in Table 16 along with the averages, weighted by the appropriate areas.

The computed ^{90}Sr inventories for 1961 and 1966 are shown in Table 17 for the two cases of deep water concentrations mentioned. Also listed are the values calculated by extrapolating the land fallout observations, assuming no difference between land and sea. It is very striking that even under the most conservative assumptions used in this calculation, the results are double the land fallout per unit area. The overwhelming evidence, it must be pointed out, suggests that the deep ocean samples do contain small but measurable concentrations of ^{90}Sr and further that the half mixing depth chosen for the computation is lower than most of the observational data would suggest. Hence, these considerations lead one to conclude that the oceanic fallout appears to have been at least three, and perhaps more, times that on land, in agreement with the conclusion reached by Bowen *et al.* (1968b, 1969).

We cannot reconcile this conclusion with the best estimates of the total ^{90}Sr inventory (Volchok and Krey, 1967), which at most could accommodate about 50 percent more fallout on sea than on land. The additional ^{90}Sr suggested in this computation, more than 7 MCi in 1966, seems to be much more than can be attributed to either measurement errors or underestimates in the total ^{90}Sr production, and we are consequently left with no reasonable explanation for this inconsistency.

THE RATIO OF ^{137}Cs TO ^{90}Sr IN SEAWATER

Radiochemical analysis of most environmental samples studied in the investigations of nuclear fallout (stratospheric air, tropospheric air, and precipitation) yielded ^{137}Cs/^{90}Sr ratios generally ranging between values of about 1 and 3 (Friend *et al.*, 1961; United Nations Scientific Committee on the Effects of Atomic Radiation, 1964, 1966; Hardy and Chu, 1967). Since both of these nuclides have inert gaseous precursors in their fission chains and generally similar nonrefractory chemical characteristics, substantial fractionation from the time of their creation in the nuclear burst is considered unlikely. Hence, the major part of the variation in observations of the ratio of ^{137}Cs to ^{90}Sr in environmental samples has been thought to reflect errors in analysis.

The expected value of this ratio in global fallout, computed by Harley *et al.* (1965), based upon measured fission product yields of debris from megaton weapons and current data on half-lives and decay schemes, is 1.45. Thus, all of the fallout entering the sea is assumed to carry this ratio of ^{137}Cs to ^{90}Sr.

Tables 18 and 19 summarize data on the ratio of ^{137}Cs to ^{90}Sr for both the Atlantic and Pacific oceans, subdivided by depth regions. The overall weighted average for all of these data is 1.6 ± 0.3. It seems reasonably clear that no significant trends in the ratio have been manifested, either with time or depth in the sea. All of the apparent anomalous values (such as in the 1965 deep water Pacific) are readily explainable, generally due to the extremely low activities and consequent greater uncertainties encountered in these samples.

A limited amount of data is available for both ^{137}Cs and ^{90}Sr from the Black Sea. Gedeonov *et al.* (1966) reported a number of analyses on samples taken in mid-1965 from the surface to 500 m deep. The ^{137}Cs/^{90}Sr was extremely constant, averaging 1.7 ± 0.1.

A substantial number of samples from the Baltic Sea were analyzed for these isotopes covering the period 1960 through 1967 (Kautsky, 1968 personal communication; Paakkola and Voipio, 1965a; Salo and Voipio, 1966 and

TABLE 17 Calculated Oceanic Inventories of ^{90}Sr in 1961 and 1966 (in MCi)

Measured Deep Water Concentration (dpm/100 liters)	Calculated Surface Component	Calculated Deep Component	Total	Land Extrapolation
1961				
0	7.0	0	7.0	3.2
3	6.1	18.4	24.5	
1966				
0	14.3	0	14.3	7.6
3	13.1	18.4	31.5	

TABLE 18 Ratio of ^{137}Cs to ^{90}Sr in the Atlantic Ocean

Year	^{137}Cs/^{90}Sr[a]	Reference
Surface Water		
1961	2.5±1.0 (6)	Kautsky, 1968, personal communication
	1.7±0.2 (2)	Rocco and Broecker, 1963
1962	1.5±0 (2)	Rocco and Broecker, 1963
1963	1.8±0.1 (2)	Kautsky, 1968, personal communication
	1.8±0.5 (54)	Ankudinov *et al.*, 1967
	1.9±0.3 (2)	Broecker *et al.*, 1966a
1964	1.5±0.2 (8)	Umweltradioaktivitat und Strahlenbelastung, 1965
	1.7±0.3 (30)	Gedeonov *et al.*, 1967
1966	1.5±0.1 (6)	Umweltradioaktivitat und Strahlenbelastung, 1966; Kautsky, 1968
Intermediate Water		
1961	1.6±0.2 (5)	Rocco and Broecker, 1963
1963	1.7±0.5 (5)	Broecker *et al.*, 1966a
	1.7±0.5 (21)	Ankudinov *et al.*, 1967
1964	1.5±0.2 (7)	Gedeonov *et al.*, 1967
1966	1.6±0.1 (14)	Umweltradioaktivitat und Strahlenbelastung, 1966; Kautsky, 1968
Deep Water		
1961	0.5±0 (1)	Rocco and Broecker, 1963
1963	1.7±0.6 (5)	Ankudinov *et al.*, 1967
1964	1.7±0.3 (6)	Gedeonov *et al.*, 1967
1966	1.9±0.6 (20)	Umweltradioaktivitat und Strahlenbelastung, 1966; Kautsky, 1968

[a] Average value ± mean deviation; numbers in parentheses are numbers of analyses.

personal communication, 1968). The ratio in the Baltic is found to be not nearly as constant as was seen for the oceans and generally is significantly lower, averaging somewhat less than 1.0. Evidently, conditions in the Baltic Sea encourage the fractionation of these nuclides after deposition, probably by changes in the ^{137}Cs concentration rather than the ^{90}Sr (Salo and Voipio, 1966).

This brief discussion and the summaries of ratio data presented in the tables strongly indicate that barring the most unusual coincidence of some geochemical or biological process moving both nuclides in the sea in the same ratio of their supply, ^{90}Sr and ^{137}Cs (and hence their stable naturally occurring isotopes) must move by ocean circulation processes and are therefore valid tracers of the water movements.

TRITIUM IN THE OCEANS

The global inventory of bomb-produced tritium, primarily from the megaton hydrogen weapons tests of the late 1950's and early 1960's (Martell, 1963), far outweighs the amount present from natural production by cosmic interaction with air. Further, substantially all of the tritium, both natural and bomb-produced, combines to form tritiated water in

TABLE 19 Ratio of ^{137}Cs to ^{90}Sr in the Pacific Ocean

Year	$^{137}Cs/^{90}Sr^a$	Reference
Surface Water		
1957	1.2±0.4 (7)	Miyake *et al.*, 1962a
1958	1.2±0.3 (3)	Miyake *et al.*, 1962a
1959	1.1±0.3 (4)	Miyake *et al.*, 1962a
1961	1.6±0.2 (3)	Rocco and Broecker, 1963
1963	1.5±0.2 (4)	Broecker *et al.*, 1966a
1964	1.5±0.3 (8)	Shirasawa and Schuert, 1968
1965	1.5±0.2 (15)	Shirasawa and Schuert, 1968
1966	1.4±0.1 (2)	Shirasawa and Schuert, 1968
Intermediate Water		
1963	1.9±0.5 (5)	Broecker *et al.*, 1966a
1964	1.7±0.2 (10)	Shirasawa and Schuert, 1968
1965	1.5±0.2 (26)	Shirasawa and Schuert, 1968
1966	1.4±0.1 (8)	Shirasawa and Schuert, 1968
Deep Water		
1958	1.2±0.2 (4)	Saruhashi *et al.*, 1962
1959	1.4±0.5 (4)	Saruhashi *et al.*, 1962
1960	1.4±0 (1)	Saruhashi *et al.*, 1962
1964	1.1±0.1 (2)	Shipman, 1966
1965	3.5±1.9 (6)	Shirasawa and Schuert, 1968

[a] Average value ± mean deviation; numbers in parentheses are numbers of analyses.

the stratosphere, and is subsequently brought down into the troposphere, where both precipitation and molecular exchange serve to transfer the tritium into the surface ocean water. Hence, tritium could probably be called the ideal tracer for ocean water movements, and in light of the time elements involved since the major injections into the atmosphere and the probable concentrations in the sea from natural sources, it should be extremely useful in studies of rates and mechanisms of surface water movements.

In all, the data and application of the data on tritium in ocean water is sparse. Some 350 samples of surface water from the North Pacific collected between 1959 and 1966 were analyzed for tritium (Bainbridge, 1963a, b; Dockins *et al.*, 1967). They showed that the response of the ocean water to a pulsed input of tritium, such as the spring fallout high, is an instantaneous rise that decreases rather quickly, presumably due to distribution downward into the mixed layer. In addition, it was pointed out that since the peak surface concentration in 1963, the levels remained about constant through 1964 and 1965 and have subsequently decreased, although the decrease has not been as abrupt as that of the tritium concentration in precipitation in the northern hemisphere.

In the Atlantic Ocean, Ostlund and Rinkel (1967) studied mixing and movement of water in the equatorial current systems by use of a series of profiles of tritium concentration, temperature, and salinity as a function of depth. The samples were obtained in March of 1964 from an area between about 2°N and 2°S of the equator, and about 7°W to 15°W longitude. Their data strongly indicate that no measurable tritium penetrated below the thermocline (at approximately 100–200 m), and they conclude that the thermocline is an effective barrier for water. A typical tritium profile from this paper is shown in Figure 4.

Additional Atlantic Ocean depth profiles for tritium have been reported on samples taken over a year later from the Atlantic, over a range of about 8°S to 52°N latitude (Munnich and Roether, 1967; Roether and Munnich, 1967). The latitude of these was from about 20°W to 30°W. Some examples of the profiles from these studies are reproduced in Figure 4. Generally, the equatorial samples, to 15°N, show a rapid decrease of tritium with depth, although measurable values seem to persist somewhat deeper than was seen in the Ostlund and Rinkel profiles. Whether this deeper penetration of tritium can be attributed to the additional year between the samplings, or perhaps to the particular longitudes of the profiles, is not apparent.

Further north, as Figure 4 clearly illustrates, the penetration of tritium occurs to even greater depths; it is easily measurable at 100 m at all latitudes north of 38°N.

In Table 20 are presented parallel data for tritium and ^{90}Sr analyzed separately on samples collected simultaneously in July of 1967 from four stations in the North Atlantic. The tritium was analyzed by Roether and Munnich (1968, personal communication) and the ^{90}Sr by Bowen *et al.* (1968, unpublished data). The two stations marked

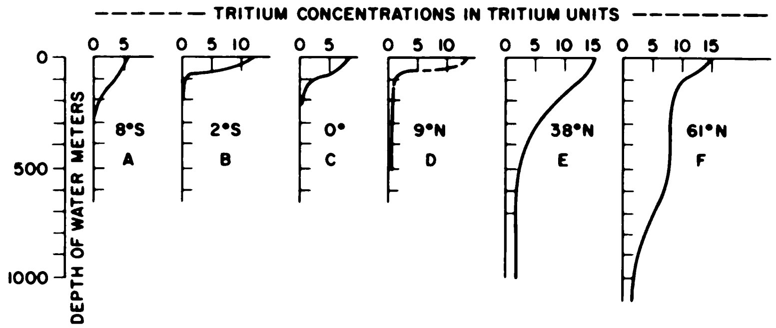

FIGURE 4 Tritium profiles in the Atlantic Ocean. (Profiles A, C, D, E, and F after Roether and Munnich, 1967; profile B after Ostlund and Rinkel, 1967.)

"Eastern" lay between 15°W and 30°W longitude, and the two marked "Western" lay between 35°W and 40°W. Although the actual ^{90}Sr and tritium results in some cases show significant differences with respect to both surface and "mixed layer" concentrations, remarkable agreement is seen when the data are expressed as percent of surface value.

The eastern stations show essentially uniform ^{3}H to 200 m and ^{90}Sr to 300 m, and only about 80 percent of the surface ^{90}Sr at 300 m. Even at 500 m, the eastern stations show about 60 percent of the surface ^{90}Sr or ^{3}H, while the western fraction is only about 40 percent, both nuclides agreeing well. At 1,000 m, on the other hand, the eastern stations show less than 5 percent of the surface ^{90}Sr and less than 10 percent of ^{3}H, whereas the western pair show 20-25 percent of the surface value, both nuclides again agreeing. The few samples in the 2,000-2,500 m range show no E-W dichotomy, the two nuclides again agreeing at 7-8 percent of the surface values.

Clearly, the data show no systematic vertical discrepancy between ^{90}Sr and ^{3}H, as was suggested by Ostlund and Rinkel (1967) for equatorial samples; Rooth (personal communication) has found their data consistent with the hypothesis that in 1963 no ^{3}H had penetrated the 10° isotherm about the equator. At the stations reported in Table 20, the 10° isotherm lay between 700 and 1,000 m on the eastern side, and between 500 and 600 m on the western; clearly, the data show penetration of this "barrier" in every case, but more extensively when the isotherm lies closer to the surface.

The noncorrespondence of absolute concentrations of ^{3}H and ^{90}Sr in both surface and deep water samples is taken as evidence that these nuclides are introduced to the ocean with differing efficiencies by rain or other processes. A detailed examination of this relationship might be expected to be quite illuminating with respect to mechanisms of over-ocean fallout.

TABLE 20 Tritium and ^{90}Sr in Atlantic Ocean Profiles, July 1967, 40°-50°N (percent of surface concentration)

	Western		Eastern	
Depth (m)	Tritium[a]	^{90}Sr[b]	Tritium[a]	^{90}Sr[b]
1	100	100	100	100
100	–	85.5	–	100
200	80.7	–	96	–
300	–	79	–	100
400	75.8	–	–	–
500	45	39	60	64
600	23.8	–	50	–
700	11	15.7	–	–
800	17.3	–	18.2	–
1,000	26.6	21	7.4	3.4
1,500	9.5	–	–	–
2,000	–	7.6	–	8.8
2,500	–	–	7.8	–

[a] Tritium analyses from Roether and Munnich (personal communication).
[b] ^{90}Sr analyses from Bowen *et al.* (unpublished data).

^{14}C IN THE PACIFIC OCEAN

The distribution of radiocarbon in the Pacific Ocean is best discussed under three headings: in the deep water, in the surface water, and in vertical profiles.

The investigations of radiocarbon in deep Pacific Ocean water confirmed the theory that the only source of deep water is from the south, flowing northward, and the strongest movement in deep water is along the western boundary (Bien *et al.*, 1965).

Unfortunately, no ^{14}C measurements of Pacific Ocean surface water were made prior to substantial contamination by radiocarbon from nuclear weapons tests. Broecker and Walton (1959) reported that approximately 10 percent of the bomb-produced radiocarbon had entered the ocean. This material served as a tracer of downward mixing, and Bien and Suess (1967) concluded that no penetration could be observed beyond a depth of 200 m in the Pacific Ocean.

The horizontal distribution of ^{14}C in the surface water of the Pacific Ocean is apparently very dependent upon latitude. In areas of known upwelling, where the water temperature is very obviously affected, the radiocarbon content follows the simple qualitative rule postulated by Burling and

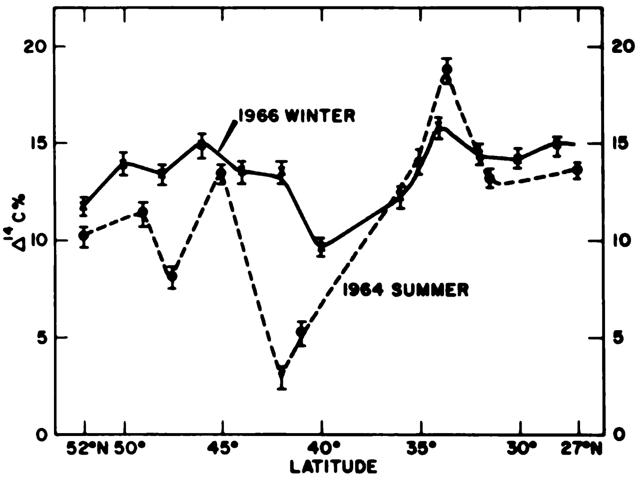

FIGURE 5 Seasonal characteristics of ^{14}C content in the surface water of the North Pacific Ocean, 155° W.

Garner (1959), i.e., that the warmer the surface water, the higher its radiocarbon content. The colder, ^{14}C-poor subsurface water acts to cool and dilute the warmer ^{14}C-rich surface water.

The seasonal characteristics of the ^{14}C content in the North Pacific is very well illustrated by the samples collected in a pair of traverses made in 1964 and 1966. During the summer of 1964, when the sea was calm and surface activity was at a minimum, samples were obtained along the 155° west meridian, from 27°N to 52°N. The ^{14}C concentration reached a peak at about 35°N, the center of the so-called subtropical gyre in the North Pacific Ocean. At about 42°N, the ^{14}C showed a minimum, indicating dilution by subsurface water, also confirmed by temperature measurements. In midwinter of 1966, in rough seas and maximum surface activity, the same traverse was repeated. The resultant curve followed the same general trends but was measurably damped, possibly because of the effects of increased advection and/or horizontal mixing in winter. In Figure 5 these results are illustrated in units of relative ^{14}C concentration.

OTHER FALLOUT RADIONUCLIDES IN OCEAN WATER

In a later section of this chapter, Table 21 lists all artificial radionuclides known to have been measured in seawater, with a key to the literature references in which these have been reported. The data fall into two classes: reports of occurrence, without enough detail of sampling either vertically or horizontally to support oceanographic or geochemical interpretation; and careful examinations of the vertical or horizontal distributions of a few nuclides, undertaken either by analysis of series of water samples or by the use of *in situ* gamma-ray spectrometers.

Reports of Occurrence

Some of these observations refer to radionuclides definitely not originating in fallout. There are a number of reports dealing with the Columbia River outflow, which carries cooling water radionuclides from the Hanford works into Northeast Pacific coastal water, and another series dealing with the outflow into the Irish Sea from the British fuel-reprocessing plant at Windscale. It should be noted that there are no reports of ^{32}P, ^{46}Sc, ^{51}Cr, ^{60}Co, ^{65}An, ^{110m}Ag, or ^{140}Ba in seawater, except from the Columbia River outflow. Of these, ^{60}Co, ^{65}Zn, and ^{110m}Ag are known to be disseminated in worldwide fallout, and others must be introduced to the oceans in course of the operations of nuclear-powered ships as well as in marine disposal of radioactive wastes (see Chapter 1), but their concentrations in seawater have not reached levels detectable by the methods so far used.

There is a much smaller number of reports in which the results are surprising: Nelepo (1960b), on the basis of *in situ* gamma spectrometry, reported both ^{85}Kr and ^{155}Eu. Consideration of their gamma spectra in relation to the relative insolubility in seawater of the former and to the quite low fission yield of the latter leads to the conclusion that each represents a misidentification of a gamma peak of ^{144}Ce that Nelepo did not report, in spite of its known considerable abundance, even though ^{155}Eu is shown by its demonstration, unequivocally, in marine sediments (Cerrai *et al.*, 1967) to have been present in seawater. A similar case is the report (Nulman and Vasquez Barete, 1967) of ^{131}I in coastal seawater from the eastern Pacific, Gulf of California, and Gulf of Mexico. The amounts reported are very high, a mean about 65 percent of the ^{90}Sr in five samples for 1966, and show no systematic change with time in the months from March to October. There is no question that for brief periods following any atmospheric test explosion, ^{131}I is a measurable constituent of fallout, and its identification in marine organisms has been made frequently; however, its presence in seawater had not been clearly demonstrated before.

Another small group (^{54}Mn and ^{125}Sb) of radionuclides, well known from reports of their presence in aerosols and in marine sediments to have been introduced as fallout to the oceans, have only rarely been reported from the open ocean. Antimony-125 was found in 1963-1964 samples from the Gulf of Mexico (Hood *et al.*, 1964; Slowey *et al.*, 1965) by gamma spectrometry on material concentrated from 1,000-liter samples; Bowen and Sugihara (1958) had been unable to detect this nuclide in 55-liter samples from the open Atlantic. Slowey *et al.* (1965), from the fact that ^{125}Sb concentrations were highest in near-shore samples, suggest the seawater reflected land run-off as well as direct fallout to the ocean; very little ^{125}Sb was found in other than "soluble" forms. Manganese-54 was found both in the Gulf of Mexico (Hood *et al.*, 1964; Slowey *et al.*, 1965) and in eastern Pacific coastal waters (Folsom *et al.*, 1963); Slowey *et al.* found much less than the expected ^{54}Mn in particulate forms, but they nevertheless found that this nuclide was moving downward in the water column at a rapid rate comparable to that of ^{144}Ce.

Other radionuclides about which we have very little information include ^{89}Sr (Cigna *et al.*, 1963), ^{239}Pu and ^{240}Pu (Pillai *et al.*, 1964; Bowen *et al.*, 1971), and ^{91}Y (Miyake, 1963a). In the case of ^{91}Y, Bowen and co-workers have a large number of still unpublished analyses of North Atlantic surface water samples taken early in 1962; by about June of that year, they found ^{91}Y no longer determinable in 55-liter samples. In the case of plutonium-239, Pillai *et al.* (1964) found, surprisingly, no evidence for separation of this nuclide from either ^{90}Sr or ^{137}Cs after reaching the sea surface. It seems likely that this was the result of their observing at a time when arrival of new fallout was so rapid as to obscure the effects of marine fractionation processes, since unpublished data of Pillai (Noshkin, Bowen, and Pillai, personal communication) showed that in shallow-water marine sediments the relative ^{239}Pu content was comparable to ^{144}Ce, indicating, as discussed below, considerable separation from ^{90}Sr or ^{137}Cs.

Nuclides Whose Vertical and Horizontal Distributions Have Been Extensively Studied

This group is seen by elimination to comprise ^{95}Zr-^{95}Nb, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce, and ^{147}Pm. Of these, ^{147}Pm has been reported by only one group (Bowen, 1961; Bowen and Sugihara, 1958, 1963, 1964, 1965; Sugihara and Bowen, 1962; Bowen *et al.*, personal communication), although its fallout onto the sea surface in determinable amounts has been confirmed by analyses of marine biota (Cerrai *et al.*, 1964a; Schreiber, 1966) and sediments (Cerrai *et al.*, 1964b, 1967; Schreiber, 1966b). As seen in Table 21, the others have each been studied by a number of investigators, both in open-ocean samples and, in the cases of ^{95}Zr-^{95}Nb and ^{106}Ru, in the effluent from the Windscale works. It had been predicted by Bowen and Sugihara (1958) that lanthanides in fallout should be largely associated with particulate matter in the oceans; the same conclusion was indicated by the studies of Greendale and Ballou (1954) and Freiling and Ballou (1962), although their data may not be applicable to "world-wide fallout" geochemistry. In the cases of ^{95}Zr-^{95}Nb, ^{106}Ru and ^{141}Ce and ^{144}Ce measurements by Chesselet and co-workers (Chesselet and Lalou, 1964a, b, 1965a, b; Chesselet *et al.*, 1965) and by Hood *et al.* (1964) and Slowey *et al.* (1965) have shown this prediction to be largely correct, although the last-named authors reported unexpectedly large percentages of each to be "soluble" at some depths sampled in the Gulf of Mexico. Both ^{95}Zr-^{95}Nb and ^{106}Ru appear to be largely particulate also in Irish Sea waters contaminated by Windscale effluent (Mauchline, 1963).

As might have been expected from this agreement of behavior, the vertical profiles of concentration of these four radionuclides agree in principal but not in detail. Curves are "wavy," with secondary maxima at depth and often even the maximum concentration at appreciable depth. In cases where sampling was close enough to show this detail, especially profiles by Chesselet, there is a tendency for the curve to peak at a depth just shallower than the top of the thermocline. With the exception of this regularity, the actual depths of subsurface concentration maxima vary widely (Bowen and Sugihara, 1965; Sugihara and Bowen, 1962; Higano *et al.*, 1963b; Riel, 1966); although no systematic examination of this question has yet been made, enough data are available to show that it is very unlikely that these subsurface maxima coincide with depths characterized by stable high concentrations of suspended particles as described by Jerlov (1955) or by Lisitsyn (1961). It appears rather that those fallout-labeled particles that penetrate the thermocline have quite rapid vertical velocities.

An important question about vertical transport referred to by Bowen and Sugihara (1965) as "populations of sinking particles" is whether these have significance for the geochemistry of the elements whose radionuclides are analyzed or whether they represent fallout nuclides locked into the debris of atmospheric tests (as "glassy spherules" condensed from the explosion cloud, for instance) and not equilibrating with elements in other phases of the ocean water column. Briefly, it is at least the majority opinion that little, if any, of the fallout analyzed in the oceans can be geochemically inert. We have discussed above the evidence that ^{90}Sr and ^{137}Cs are largely present in seawater in soluble form. Bowen and Sugihara (1963, 1965) and Sugihara and Bowen (1962) have discussed the evidence that ^{144}Ce and ^{147}Pm are physically separated after they enter the water column. Similar separations are seen for ^{95}Zr-^{95}Nb versus ^{106}Ru in the vertical profiles published by Chesselet and co-workers (1965), by Slowey *et al.* (1965), and by Riel (1966). That each of these radionuclides has been separated also from ^{90}Sr or ^{137}Cs is clear; this was explicitly discussed by Sugihara and Bowen (1962) and by Higano *et al.* (1964). Separations such as these appear possible only by solution of the fallout debris [the low pH conditions often operative in evaporating rain drops (Ericsson, 1957) must not be lost sight of] and entry of the individual radionuclides into appropriate radionuclides into appropriate geochemical cycles in the oceans.

This is not to say that one can be confident that the fallout radionuclides come into isotopic equilibrium with their stable elements in seawater and so act as "good" tracers geochemically. In most cases, we have no evidence appropriate to considering this question—no stable element concentration data for zirconium, ruthenium, or antimony, and really too little in such a case as manganese. The fact that fallout radionuclides enter the ocean largely by penetration of the sea surface, with its attendant organic chemical complexities still being unraveled, should surely raise suspicions in our minds. No evidence has yet appeared either in the Irish Sea or along the United States Pacific coast that uptake by organisms is different when particular nuclides are introduced either as fallout or as "Windscale effluent" or "Columbia River outfall." This is encouraging, but experiments have not been planned specifically to explore this question, and often the fallout nuclides have had to be studied at great dilution, producing large experimental uncertainties. Should later study bear out the hypothesis (Slowey *et al.*, 1965) that a large fraction of near-shore antimony-125 represents land runoff (and, by inference, is isotopically equilibrated with land-supplied stable antimony), this situation would appear very favorable for confirming the usefulness of ^{125}Sb as a geochemical tracer in seawater.

In the case of the lanthanides (^{144}Ce, ^{147}Pm, and, when measured, ^{155}Eu), recent evidence has further complicated the picture: Høgdahl *et al.* (1968) report that the relative abundance pattern of lanthanide elements appears to be a conservative property and to distinguish in the Atlantic such water masses as North Atlantic Deep Water, Antarctic Intermediate Water, and Antarctic Bottom Water from each other. Constituents of this degree of conservatism, bearing in mind that the Antarctic Intermediate Water mass is only about 700 m thick about the equator (Defant, 1961), could not have vertical velocities of the orders suggested for fallout ^{144}Ce or ^{147}Pm, and they probably could not have oceanic residence times as short as those estimated by Goldberg *et al.* (1963). In discussion of this latter discrepancy, Goldberg has suggested, as had others, that the short-residence-time elements undoubtedly represent two constituents: one that is removed very rapidly after its introduction from river outflow by geochemical processes peculiar to the continental shelves; the other that, after reaching the pelagic environment, has a very long residence time. It is clear that this latter portion has the geochemical properties to be a conservative constituent of major water masses. It does not, however, appear that fallout lanthanides are tracing, or, by inference, equilibrating with, either postulated constituent.

Further examination, in the case of each traceable fallout radionuclide, of the relationship between their behavior in the marine environment and that of the various forms of their related elements, should be a major task of students of fallout geochemistry in the oceans.

One final point should be emphasized: Studies of shallow vertical profiles of such relatively short-lived nuclides as ^{95}Zr-^{95}Nb, ^{103}Ru, ^{106}Ru, ^{141}Ce, and ^{144}Ce (Chesselet *et al.*, 1965) yielded very convincing evidence that for these fallout nuclides the deposit per unit ocean surface has considerably exceeded that per unit land surface at comparable latitude. Chesselet and his co-workers found a significant increase over land in fallout per unit area. The range of "over-ocean excess," on the Bay of Biscay or Mediterranean, was from twofold to sevenfold. Since in each case these integrations represent only the upper 150–300 m, they must be viewed as underestimating by whatever amounts have penetrated to deeper levels. Considerable interest attaches to this demonstration of the same order of magnitude of over-ocean fallout excess as has been described from measurement of vertical profiles of ^{90}Sr discussed earlier in this chapter (Bowen and Sugihara, 1960, 1963, 1965) or from study of the changes with time of surface ocean ^{90}Sr concentrations (Bowen *et al.*, 1968a, b).

NUCLIDE REFERENCE TABLE

Table 21 lists all of the known publications that either report results or describe methods of analysis for radioisotopes in seawater of particular interest to this chapter. In all, 26 radionuclides are represented, with more than 170 references; ^{90}Sr and ^{137}Cs are, naturally, the most prominent.

TABLE 21 References to Radionuclides Analyzed in Seawater

Nuclide	Reference
^{3}H	United Nations Scientific Committee on the Effects of Atomic Radiation (1966) Bainbridge (1963a, b) Begemann and Libby (1957) Dockins *et al.* (1957) Giletti (1957) Martell (1963) Munnich and Roether (1967) Ostlund and Rinkel (1967) Roether and Munnich (1967)
^{14}C	United Nations Scientific Committee on the Effects of Atomic Radiation (1966) Bien and Suess (1967) Bien *et al.* (1963, 1965) Broecker (1963) Broecker *et al.* (1960) Broecker and Rocco (1963) Broecker and Walton (1959) Burling and Garner (1959) Munnich and Roether (1967) Rafter and Ferguson (1957) Roether and Munnich (1967, personal communication) Thommert *et al.* (1965)
^{32}P	Chakravarti *et al.* (1964)

TABLE 21 (Continued)

Nuclide	Reference
^{46}Sc	Perkins *et al.* (1966)
^{51}Cr	Chakravarti *et al.* (1964)
	Osterberg *et al.* (1965)
	Perkins *et al.* (1966)
^{54}Mn	Folsom *et al.* (1963)
	Hood and Slowey (1964)
	Slowey *et al.* (1965)
^{60}Co	Chakravarti *et al.* (1964)
	Perkins *et al.* (1966)
^{65}Zn	Chakravarti *et al.* (1964)
	Perkins *et al.* (1966)
^{85}Kr	Nelepo (1960b)
^{89}Sr	United Nations Scientific Committee on the Effects of Atomic Radiation (1966)
	Cigna *et al.* (1964)
	Miyake and Sugiura (1955)
^{90}Sr	Aarkrog (1963; 1968, personal communication)
	Aarkrog and Lippert (1964a, b, c; 1965a, b, c; 1966a, b, c; 1967a, b, c)
	Aarkrog *et al.* (1963a, b, c)
	Agnedahl *et al.* (1962)
	Ankudinov *et al.* (1967)
	USAEC Health and Safety Laboratory (1958, 1968)
	Umweltradioaktivitat und Strahlenbelastung (1962, 1964, 1965, 1966, 1967a, b)
	Comitato Nazionale de l'Energia Nucleare (1963, 1964, 1965, 1966a, b)
	Rudjer Boskovic Institute (1964, 1965, 1966)
	United Nations Scientific Committee on the Effects of Atomic Radiation (1964, 1966)
	Ministry of Agriculture, Fisheries and Food (1967)
	Argiero *et al.* (1963, 1965)
	Azhazha and Chulkov (1964)
	Baranov and Khitrov (1964)
	Baranov *et al.* (1964)
	Blinov *et al.* (1965)
	Bowen (1961)
	Bowen and Sugihara (1957, 1958, 1960, 1963, 1964, 1965)
	Bowen *et al.* (1968a, b, c)
	Broecker (1966)
	Broecker and Simpson (1968)
	Broecker *et al.* (1968a, b)
	Cerrai *et al.* (1963a)
	Chakravarti *et al.* (1964)
	Cigna *et al.* (1963, 1964)
	Dera *et al.* (1962)
	Gedeonov *et al.* (1966, 1967)
	Hammond *et al.* (1966)
	Higano (1959)
	Higano *et al.* (1962, 1963a)
	Higano and Shiozaki (1960)
	Hishida and Yamamoto (1958)
	Hiyama and Ichikawa (1957)
	Hollstein (1959)
	Kautsky (1962; 1968, personal communication)
	Mahmond *et al.* (1961)
	Mauchline (1961, 1962, 1963)
	Mauchline and Templeton (1963, 1964)
	Miyake (1958a, 1963a)
	Miyake *et al.* (1960; 1961a, b, c; 1962a, b; 1964)
	Miyake and Sugiura (1955)
	Nulman and Vasquez Barete (1967)
	Paakola and Voipio (1965a, b)
	Park *et al.* (1965)
	Patin (1965)
	Patin *et al.* (1966)
	Petersen (1962)
	Popov (1966)
	Popov *et al.* (1962; 1963b; 1964a, b; 1966a, b, c)
	Popov and Orlov (1967)
	Popov and Patin (1966a, b)
	Rocco and Broecker (1963)
	Salo and Voipio (1966; 1968, personal communication)
	Saruhashi (1963)
	Saruhashi *et al.* (1962)
	Schreiber (1967)
	Schreiber *et al.* (1962, 1964, 1965)
	Schreiber and Tassi-Pelati (1965)
	Shiozaki *et al.* (1965)
	Shvedov *et al.* (1963a, 1964)
	Shvedov and Shirokov (1962)
^{91}Y	Miyake (1963b)
	Miyake and Sugiura (1955)
^{125}Sb	Bowen and Sugihara (1958)
	Hood and Slowey (1964)
	Slowey *et al.* (1965)
^{95}Zr	Ministry of Agriculture, Fisheries and Food (1967)
	Chakravarti *et al.* (1964)
	Chesselet *et al.* (1964a, b; 1965)
	Chesselet and Nordemanne (1962c)
	Cigna *et al.* (1963, 1964)
	Mauchline (1963)
	Mauchline and Templeton (1963)
	Miyake (1963a)
	Miyake and Sugiura (1955)
	Reil (1966)
	Reil *et al.* (1965)
	Slowey *et al.* (1965)
	Yamagata and Iwashima (1965)
^{103}Ru	Chakravarti *et al.* (1964)
	Chesselet *et al.* (1964a, b)
	Cigna *et al.* (1963, 1964)
	Mauchline (1963)
	Miyake and Sugiura (1955)
	Slowey *et al.* (1965)
^{106}Ru	Ministry of Agriculture, Fisheries and Food (1967)
	Chesselet *et al.* (1964a, b; 1965)

TABLE 21 (Continued)

Nuclide	Reference
^{106}Ru	Cigna *et al.* (1964)
	Hood and Slowey (1964)
	Mauchline and Templeton (1963)
	Miyake and Sugiura (1955)
	Perkins *et al.* (1966)
	Reil *et al.* (1965)
	Slowey *et al.* (1965)
^{110m}Ag	Perkins *et al.* (1966)
^{131}I	Nulman and Vasquez Barete (1967)
^{137}Cs	Ankudinov *et al.* (1967)
	Umweltradioaktivitat und Strahlenbelastung (1962; 1964; 1965; 1966; 1967a, b)
	Comitato Nazionale de l'Energia Nucleare (1964; 1965; 1966a, b)
	United Nations Scientific Committee on the Effects of Atomic Radiation (1964, 1966)
	Ministry of Agriculture, Fisheries and Food (1967)
	Argiero *et al.* (1963, 1965)
	Bainbridge (1963b)
	Baranov and Khitrov (1964)
	Broecker (1966)
	Broecker and Simpson (1968)
	Broecker *et al.* (1966a)
	Broecker and Rocco (1963)
	Chulkov and Gorbunov (1963)
	Cigna *et al.* (1963)
	Folsom (1968, personal communication)
	Folsom and Mohanrao (1960; 1962a, b)
	Folsom *et al.* (1960, 1961, 1965, 1968)
	Folsom and Saruhashi (1963)
	Folsom and Sreekumaran (1966a, b)
	Gedeonov *et al.* (1967)
	Kupferman (1971)
	Hammond *et al.* (1966)
	Higano *et al.* (1962, 1963)
	Higano and Shiozaki (1960)
	Kameda *et al.* (1962)
	Kautsky (1962; 1968, personal communication)
	Miyake (1958a, 1963a)
	Miyake *et al.* (1961a, b, c; 1962a, b; 1964)
	Mohanrao and Folsom (1963a, b)
	Nelepo (1960b)
	Nulman and Vasquez Barete (1967)
	Park *et al.* (1965)
	Perkins and Robertson (1970, personal communication)
	Robertson and Perkins (1966b)
	Rocco and Broecker (1963)
^{140}Ba	Perkins *et al.* (1966)
^{141}Ce	Chesselet *et al.* (1964a, b)
	Miyake and Sugiura (1955)
	Slowey *et al.* (1965)
^{144}Ce	Bowen 1961
	Bowen and Sugihara (1958, 1963, 1964, 1965)
	Chakravarti *et al.* (1964)
^{144}Ce	Chesselet *et al.* (1964a, b; 1965)
	Cigna *et al.* (1963)
	Higano *et al.* (1962, 1963a)
	Hood and Slowey (1964)
	Kameda *et al.* (1962)
	Mauchline (1963)
	Mauchline and Templeton (1963)
	Miyake and Sugiura (1955)
	Perkins *et al.* (1966)
	Shiozaki *et al.* (1964)
	Slowey *et al.* (1965)
	Sugihara and Bowen (1962)
^{147}Pm	Bowen (1961)
	Bowen and Sugihara (1958, 1963)
^{155}Eu	Nelepo (1960b)
^{239}Pu and ^{240}Pu	Pillai *et al.* (1964)
	Bowen *et al.* (1971)

SUMMARY

In this chapter we have endeavored to bring together all known measurements of fallout radionuclides in seawater, with special emphasis on ^{90}Sr and ^{137}Cs, and to discuss a number of important implications of these data. Additionally, an extensive bibliography has been compiled, covering measurements, analytical methodology, data reports, and applicable interpretive studies.

By far the major emphasis in seawater analysis has been on ^{90}Sr and ^{137}Cs. The ratio of ^{137}Cs to ^{90}Sr was relatively constant regardless of depth, time, or location of sample collection, averaging about 1.5. Hence for simplicity in comparing data, all ^{137}Cs were converted to ^{90}Sr by the factor 0.67.

This report summarizes the results of surface water analyses of 779 samples from the Atlantic Ocean, 1,181 from the Pacific Ocean, 96 from the Indian Ocean, and 543 from various seas. These results were reported by more than 20 different investigators from 8 different countries. The interlaboratory agreement on ^{90}Sr analyses, based on samples taken in reasonably close proximity, was found to be satisfactory. The nonuniformity of observed seasonal variations of ^{90}Sr in fallout over the oceans and also in surface water concentration, compared to the common spring peak on land, is not understood, and may reflect differences in the fallout mechanism.

Depth profiles of ^{90}Sr concentration, down to 700 m in the Atlantic, generally show regular decreases, with values in this stratum rarely falling below 10 percent of the surface value, and no significant trend with time. No correlation of these profiles with inferred fallout rates to the surface can be discerned. In the Pacific, much greater geographic variability is shown in the depth profiles; in the northwestern regions, the 700-m samples averaged about 35 percent of the surface concentration, while northeastern and California coastal profiles indicate much lower concentrations at depth.

A consensus of opinion by the authors of this chapter could not be reached concerning interpretation of the deep water ^{90}Sr concentration data and the total oceanic inventory. The conclusions drawn from these subjects have direct implications on the rate of mixing in the ocean, the age of deep water, and the global ^{90}Sr budget.

Tritium concentrations in the surface Pacific Ocean peaked in 1963 and have been decreasing since. In the eastern equatorial Atlantic, the thermocline appears to completely insulate the deeper layers from tritium penetration; however, north of about 15°, tritium was found at depths in excess of 2,000 m, with no systematic vertical discrepancy from ^{90}Sr. Radiocarbon, studied in the northeastern Pacific Ocean, indicates no penetration below 200 m and a very strong latitude dependence in surface water. Interesting seasonal patterns of ^{14}C in surface water suggest a relationship between weather and mixing.

Studies of other artificial radionuclides in the sea have shown that the lanthanides, as well as zirconium and niobium, are largely associated with particles and exhibit generally similar profiles with depth, in spite of the probability that the fallout debris is initially in solution on entering the ocean. The relationship between these nuclides and their stable isotopes in the sea is complex and, in the main not well understood.

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APPENDIX

Following is a listing of references to literature pertinent to this chapter which have appeared too late for inclusion in the bibliography, or which have only very recently come to our attention. These papers are not included in the Nuclide Reference Table (Table 21) in this chapter.

We acknowledge the wonderful cooperation of many colleagues who helped in supplying unpublished and unavailable data, and in providing literature listings to enhance the bibliography. In particular: Dr. I. L. Karol (USSR), Dr. H. Kautsky (Germany), Dr. A. Voipio (Finland), Mr. A. Aarkrog (Denmark), and Mr. B. W. Schroeder (WHOI).

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Chapter Four

PHYSICAL PROCESSES OF WATER MOVEMENT AND MIXING*

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INTRODUCTION

Radioactive isotopes introduced into the marine environment are subjected to the same physical, chemical, and biological processes that affect nonradioactive isotopes in the same physical state. The additional factor influencing the distribution of an introduced radionuclide is radioactive decay. For very short-life isotopes, radioactive decay influences distribution at all scales of space and time. For intermediate-life isotopes, decay is unimportant for space and time scales below some critical values and becomes increasingly important for space and time scales larger than these critical values. For long-life isotopes, decay influences the distribution in the ocean only for relatively large time and space scales.

The fate of a radioactive substance introduced into the ocean depends on the physical and chemical state of the substance, the manner of introduction, and the location of introduction.

This chapter considers the physical processes that advect the introduced material away from the source and those that disperse the material by nonadvective mechanisms. Motion in the ocean can be regarded as a continuous spectrum encompassing scales ranging from that of the molecular free path up to that of the ocean-wide circulation. For that part of the motion assigned to advective processes, the spatial and temporal distribution of the velocity field must be known; for that part of the motion assigned to the nonadvective, or diffusive processes, only certain statistical properties are required. The division between advective and nonadvective motion depends upon how much detail, in time and space, we require in our effort to determine the distribution of the concentration of a constituent of seawater or of an introduced material such as radioactive isotopes.

The general concepts are most conveniently explained by describing the fate of radioactive materials introduced into the sea by several different methods and under several different conditions of physicochemical state. The situation most amenable to description is that in which the radioactive substance is introduced as a local source over a short period of time and in a chemical and physical state such that complete and immediate solution in the receiving waters takes place. Further, the mass of introduced material is taken to be sufficiently small that no significant difference in density exists between the initial contaminated volume and the surrounding receiving waters. From a practical standpoint, the introduction of the contaminant will result in some initial mechanical dilution. The initial contaminated

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volume resulting from this mechanical mixing is termed the initial cloud.

The local current pattern will advect the contaminated cloud away from the point of introduction, and the small-scale turbulent motion will produce diffusion of the cloud, causing it to grow in volume and the concentrations within it to decrease. As the cloud increases in size, the scale of motion producing an advection of the cloud as a whole increases, as does the upper limit of the scales of motion that produce diffusion of the cloud. If our interest centers only on certain statistical properties of the spreading cloud, we may consider them with respect to scales of motion: eddies larger than the cloud produce an advection of the cloud as a whole, while eddies smaller than the cloud produce internal shearing and stirring, which together with processes producing uniformity are described mathematically under the general term "turbulent diffusion." In such an approach the energy available for diffusion is considered to increase in proportion to some measure of the size of the cloud, though the relationship may not be linear. This manner of treating the problem permits us to describe the time variation in the average root mean square spread of the cloud and the time-rate of decrease in peak concentration. With an adequate mathematical model, the area contained within the various isolines of concentration is also determined. However, the shapes of the cloud and of individual isolines of concentration are not described by this approach.

The above discussion deals primarily with the horizontal spread of the cloud. It is tacitly assumed that the cloud is bounded vertically either by the surface and bottom, by the surface and a boundary of large vertical stability (such as the thermocline), or by an upper and lower boundary of high stability. If the vertical density structure permits significant vertical fluxes to take place, then the spread and the concentration distribution within the cloud will be influenced by processes of vertical advection and diffusion. Scale will be less important in regard to the vertical processes than to the horizontal processes.

If we desire greater information about the moving and dispersing cloud, such as the general shape of the cloud and of the individual isolines of concentration and the probable positions of the points of maximum concentration and of the center of mass of the cloud (these may not be at the same position), then we need to look at the field of motion in greater detail. Eddies much larger than the cloud are then considered to be responsible for the advection of the cloud as a whole, and eddies much smaller than the cloud produce dispersion by turbulent diffusion. However, eddies approximately the same size as the cloud significantly influence the shape of the cloud and contribute to the dispersion as well. Essentially, eddies about the same size as the cloud produce shear, or spatial variation in the velocity field. The shear in the velocity field will tend to advect one part of the cloud faster than another, producing an elongated, somewhat elliptical shape. Shears in both the horizontal and vertical planes are effective in producing this elongation, but the process is most easily envisioned by considering vertical shear. Consider the cloud as initially shaped like a vertical cylinder. The vertical shear in the horizontal current pattern will produce a tilting of the cylinder, as the upper portion is moving at a different speed than the lower portion. Large vertical-concentration gradients are therefore produced by this shearing advective motion, which, when acted on by vertical diffusion, produces an elongated cloud in the horizontal plane with the axis of elongation in the direction of the shear.

As the patch grows in size, changes occur in the portion of the spectrum of motion that contributes to the diffusion process, the portion that produces the shear effects, and the portion that advects the cloud as a whole, with the boundaries between these various portions of the spectrum always moving toward larger scale. Consequently, any model intended to describe the changing shape and concentration distribution for any considerable length of time must take into account the changes in the scales of the motion contributing to the various aspects of the movement and mixing of the contaminated cloud.

If the material is introduced as a suspension of small particles rather than in solution, the same processes of movement and mixing will still act to advect and disperse the initial cloud. However, there will also be a motion of the individual particles due to their settling velocity. A mathematical model of the process would therefore include an added term to account for this added vertical flux.

If the material is introduced into the marine environment in a solution having significantly different density than the receiving waters, then the early fate of the radioactive material may be influenced to a considerable degree by the manner of introduction. If the initial solution is less dense than the receiving waters and the introduction is made at some depth below the surface, then the cloud will ascend as a result of the buoyant forces. As the cloud ascends, it will mechanically entrain diluting water from the environment, thus providing for an initial mechanical decrease in concentration and also for a reduction in the difference in density between the cloud and the receiving waters. If the point of introduction is deep enough, and if a vertical gradient in density occurs in the receiving waters, then the cloud will ultimately entrain sufficient water to bring it to the density of the surrounding water and will cease to rise. At this point the physical processes discussed above begin to act on the cloud. If the cloud reaches the surface with a significant density difference remaining, the processes of further dilution by turbulent mixing are somewhat inhibited.

If the cloud is initially denser than the receiving waters, it will sink, entraining diluting water enroute. A sequence of events similar to that described above for the ascending cloud of lower density will then occur.

If the radioactive material is introduced as a large planar source, such as fallout on the surface of the ocean, then our concern with scale depends only on how detailed a description we wish of the subsequent fate due to physical processes. The physical processes that are most important in determining the time history of the spatial distribution of the radioactive material introduced into the sea as a planar source at the sea surface are vertical diffusion; vertical advection; and sinking at source regions for intermediate and deep water and subsequent horizontal movement of the radioactive materials within the intermediate and deep water masses.

Hence, the large-scale ocean-wide circulation patterns, particularly those involving vertical motion, as distinguished from small-scale turbulent-diffusion processes, must play a major role in the long-term distribution of the introduced radioactive materials. In the early period after initiation of introduction, the vertical distribution at a given location will most likely depend primarily upon the vertical processes of turbulent diffusion and vertical advection. The length of time covered by this early period, during which the local vertical processes are of prime importance, is likely to be several decades.

In the remainder of this chapter, the physical processes briefly mentioned above are described in greater detail. In addition to discussions of the general ocean-wide circulation, turbulent diffusion processes, and the special features of inshore and estuarine environments, one part of the chapter is devoted to the oceanographic implications of the observed distributions of natural tracers and of fallout-derived radioactive isotopes.

OCEAN CHARACTERISTICS AND CIRCULATION

This discussion summarizes the essential features of the circulation of the oceans in relation to the associated distribution of properties. The density of seawater depends upon its temperature and salinity at a given pressure. In turn, through the geostrophic balance relation, the density governs in large measure the rate of change of current velocity with depth, excluding Ekman boundary layers (*cf.* Sverdrup *et al.*, 1942). The distribution of properties, on the other hand, is governed by currents and turbulent mixing processes. Accordingly, one must have a knowledge of both in order to achieve a proper understanding of either.

Our knowledge of the distribution of properties in the sea is at present much more complete than our knowledge of its circulation, with the possible exception of the surface currents. However, there have been particularly significant advances during the past 16 years in the development and application of methods for measuring the elusive intermediate and deep currents (Swallow, 1955; Volkmann *et al.*, 1956; Knauss, 1960; Metcalf *et al.*, 1962; Richardson *et al.*, 1963; Webster, 1963; Richardson and Schmitz, 1965; Pochapsky, 1966; Kolesnikov *et al.*, 1966; Maloney, 1967). Direct measurements in deep water are still sparse, but those available seem to support qualitative deductions based upon theory (Stommel, 1957; Stommel and Arons, 1960a, 1960b). The latter studies, and those of Lineykin (1955) and Robinson and Stommel (1959), represent major advances in our understanding of the dynamics and thermal convection of the ocean.

A reasonably up-to-date and comprehensive treatment of the subject is given in general texts on physical oceanography, such as those of Defant (1961) and Neumann and Pierson (1966), as well as in more specialized texts such as that of Stommel (1965).

Property Statistics

A very thorough statistical treatment of the temperature and salinity of the oceans has been presented in a series of papers by Cochrane (1958), Pollak (1958), and Montgomery (1958). These workers have examined the bivariate distributions of potential temperature and salinity in the Pacific, Indian, and Atlantic oceans, respectively. The potential temperature is the temperature that a seawater sample would have if it were brought adiabatically to the surface (atmospheric pressure) without change of its salinity. It differs from the *in situ* temperature of the sample by not more than 1.5°C (cf. Sverdrup *et al.*, 1942, p. 64). Salinity is essentially the total dissolved salts (in grams) in 1 kg of seawater (i.e., mass fraction in parts per mille).

A summary of some of these property statistics is given in Table 1. The statistics for the potential specific volume of seawater are given in the table, along with those of potential temperature and salinity. The potential specific volume is the specific volume a sample would have if brought adiabatically to atmospheric pressure. These values are presented in the form of an anomaly (expressed in centiliters per metric ton) from the specific volume of seawater at the standard conditions of 0°C, 35 per mille, and 1 atm. This standard specific volume has the value 97,264 cl/ton (Sverdrup *et al.*, 1942). Thus, from Table 1 it is seen that the mean specific volume of the world ocean is 97,320 cl/ton (or 0.097320 cl/g). This specific volume, in turn, corresponds to a density of 1.027538 g/ml (or a density anomaly from 1 g/ml of 27.538 mg/ml).

Table 1 indicates that 90 percent (by volume) of the water of the world ocean has potential temperatures lying in the range 0.0°C to 12.6°C, with a median value of 2.1°C and a mean value of 3.52°C (indicating a highly skewed distribution). Moreover, 50 percent of the world ocean has

TABLE 1 Statistics of Potential Temperature, Salinity, and Potential Specific Volume Anomaly for the Pacific, Indian and Atlantic Oceans and for the World Ocean as a Whole[a]

Ocean	Mean	5%	25% (Lower Quartile)	50% (Median)	75% (Upper Quartile)	95%
Potential temperature, °C						
Pacific	3.36	0.8	1.3	1.9	3.4	11.1
Indian	3.72	−0.2	1.0	1.9	4.4	12.7
Atlantic	3.73	−0.6	1.7	2.6	3.9	13.7
World	3.52	0.0	1.3	2.1	3.8	12.6
Salinity, per mille						
Pacific	34.62	34.27	34.57	34.65	34.70	34.79
Indian	34.76	34.44	34.66	34.73	34.79	35.19
Atlantic	34.90	34.41	34.71	34.90	34.97	35.73
World	34.72	34.33	34.61	34.69	34.79	35.10
Potential specific-volume anomaly, cl/ton						
Pacific	62	22	31	39	66	162
Indian	56	21	25	31	63	145
Atlantic	45	8	22	28	46	137
World	56	20	26	36	62	149

[a] Reprinted with permission from Montgomery, 1958.

potential temperatures in the narrow range of 1.3°C to 3.8°C (primarily associated with the deep water of the oceans). The total range of oceanic temperature is from about −2°C to 30°C (not indicated in the table). The 90 percent range for salinity in the world ocean is 34.33 to 35.10 per mille. Its extreme range is about 30.0 to 40.0 per mille, excluding estuaries and lagoons but including the arctic seas and the Red Sea.

The statistical and physical differences between the Atlantic, Pacific, and Indian oceans are small but significant. Montgomery (1958) points out that the joint statistical distribution of potential temperature and salinity in the world ocean shows a dominant mode at 1.5°C and 34.7 per mille, which he calls the "Common Water." This mode corresponds closely to the mode for the joint statistical distribution for the Pacific and Indian oceans individually. The Atlantic Ocean, on the other hand, has a dominant mode centered at a potential temperature of 2.25°C and salinity of 34.95 per mille. This is primarily associated with the North Atlantic Deep Water. A secondary mode occurs at 0°C and 34.65 per mille for the South Atlantic and is associated with the Antarctic Bottom Water. The relative volumes of the Pacific, Indian, and Atlantic oceans are, respectively, 53.0, 21.2, and 25.8 percent of the world ocean.

Ninety percent of the water in the world ocean has a range in potential specific volume of only about 130 parts out of 10^5. Nevertheless, such variations of specific volume are indeed important in computing currents related to the distribution of mass.

The ranges of temperature (*in situ*) and salinity for various water classes of the oceans are given in Table 2, according to Defant (1961). The water classes referred to are delineated geographically in Figure 1, and the temperature-salinity relations for these classes are shown graphically in Figure 2. The Common Water would be represented by a point at the base of the curves in Figure 2.

Spatial Distribution of Properties

Typical vertical profiles of temperature and salinity are shown in Figures 3 and 4, respectively, for ten different latitudes in the Atlantic Ocean. In the Arctic and Antarctic, the water column is nearly isothermal and isohaline below about 200 m. The minimum surface temperature in these regions is controlled by the freezing point of seawater, which is about −1.8°C at normal salinities (Sverdrup *et al.*, 1942). Surface salinities in the polar regions, particularly near the continents, vary considerably from summer to winter (being dependent upon the amount of meltwater).

At mid-latitudes and in the tropical belt, the temperature distributions are characterized by a nearly isothermal, surface mixed layer (ranging in thickness from 10 to 200 m), a nearly isothermal abyssal region below the 1,000-m depth, with a transitional intermediate region of great stability (the thermocline). Below about 500 m, the field of temperature shows little seasonal variation.

TABLE 2 Ranges of Temperature and Salinity for Different Subregions of the Three Oceans[a]

Water masses of the Atlantic Ocean

North Atlantic	Temp. (°C)	Salinity (‰)	South Atlantic	Temp. (°C)	Salinity (‰)
1. North Polar Water	−1 to +2	34.9	1. South Atlantic Central Water	+5 to +16	34.3–35.6
2. Subarctic Water	+3 to +5	34.7–34.9	2. Antarctic Intermediate Water	+3 to +5	34.1–34.6
3. North Atlantic Central Water	+4 to +17	35.1–36.2	3. Subantarctic Water	+3 to +9	33.8–34.5
4. North Atlantic Deep Water	+3 to +4	34.9–35.0	4. Antarctic Circumpolar Water	+0.5 to +2.5	34.7–34.8
5. North Atlantic Bottom Water	+1 to +3	34.8–34.9	5. South Atlantic Deep and Bottom Water	0 to +2	34.5–34.9
6. Mediterranean Water	+6 to +10	35.3–36.4	6. Antarctic Bottom Water	−0.4	34.66

Water masses of the Indian Ocean

	Temp. (°C)	Salinity (‰)
1. Equatorial Water	4–16	34.8–35.2
2. Indian Central Water	6–15	34.5–35.4
3. Antarctic Intermediate Water	2–6	34.4–34.7
4. Subantarctic Water	2–8	34.1–34.6
5. Indian Ocean Deep and Antarctic Circumpolar Water	0.5–2	34.7–34.75
6. Red Sea Water	9	35.5

Water masses of the Pacific Ocean

North Pacific	Temp. (°C)	Salinity (‰)	South Pacific	Temp. (°C)	Salinity (‰)
1. Subarctic Water	2–10	33.5–34.4	1. Eastern South Pacific Water	9–16	34.3–35.1
2. Pacific Equatorial Water	6–16	34.5–35.2	2. Western South Pacific Water	7–16	34.5–35.5
3. Eastern North Pacific Water	10–16	34.0–34.6	3. Antarctic Intermediate Water	4–7	34.3–34.5
4. Western North Pacific Water	7–16	34.1–34.6	4. Subantarctic Water	3–7	34.1–34.6
5. Arctic Intermediate Water	6–10	34.0–34.1	5. Pacific Deep Water and Antarctic Circumpolar Water	−1 to 3	34.6–34.7
6. Pacific Deep Water and Arctic Circumpolar Water	−1 to 3	34.6–34.7			

[a] Reprinted with permission from Defant, 1961.

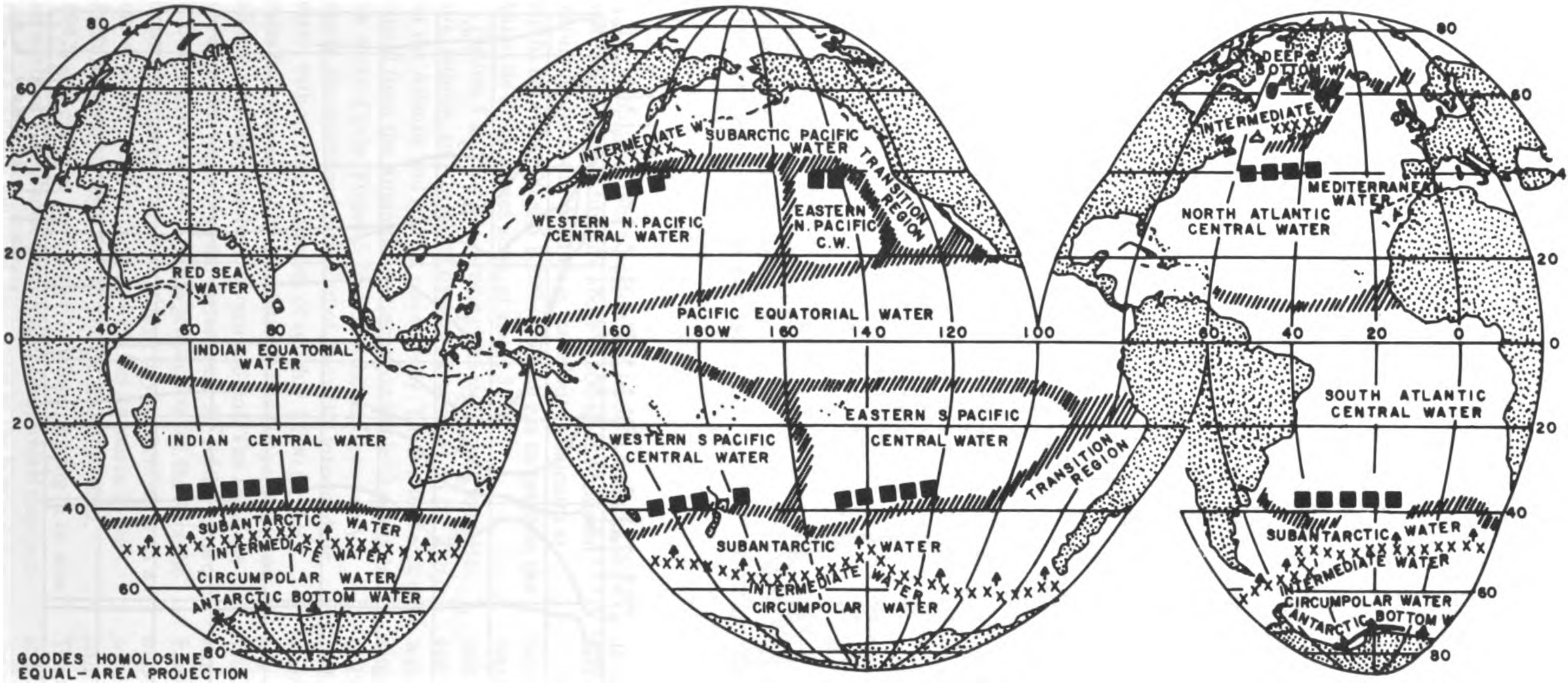


FIGURE 1 Approximate boundaries of the upper water masses of the ocean. Squares indicate the regions in which the central water masses are formed; crosses indicate the lines along which the Antarctic and Arctic Intermediate Waters sink. (Reprinted with permission from Sverdrup *et al.*, 1942.)

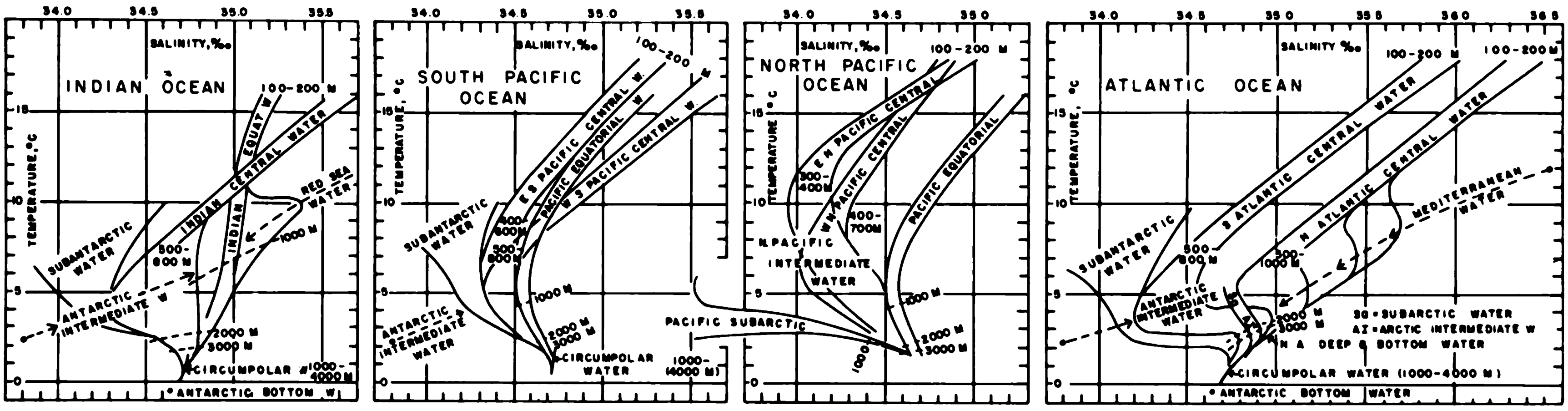
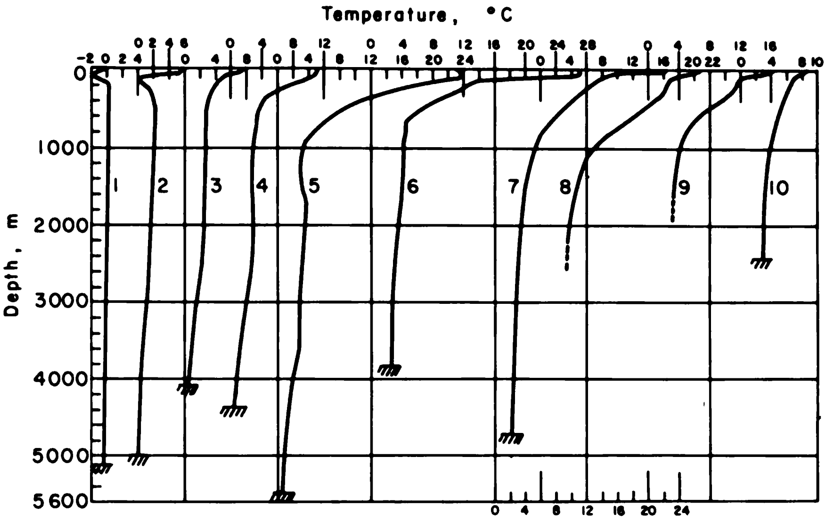


FIGURE 2 Temperature-salinity relations of the principal water masses of the oceans. (Reprinted with permission from Sverdrup *et al.*, 1942.)



Ship	Position		Date
1. *Will Scoreby* 554	63° 20′ S	17° 23′ W	5.ii. 1931
2. *Meteor* 58	48° 30′ S	30° 0′ W	7/8. x. 1925
3. *Meteor* 83	32° 9′ S	25° 4′ W	29. xi. 1925
4. *Meteor* 170	22° 39′ S	27° 55′ W	9. vii. 1926
5. *Meteor* 191	9° 7′ S	2° 2′ W	9/10. ix. 26
6. *Meteor* 212	0° 36′ N	29° 12′ W	19. x. 1926
7. *Meteor* 283	17° 53′ N	39° 19′ W	22/23. iii. 1927
8. *Dana* 1376	33° 42′ N	36° 16′ W	10. vi. 1922
9. *Armauer Hansen* 17	58° 0′ N	11° 0′ W	29. vii. 1913
10. *Fram* 20	78° 1′ N	9° 10′ E	22. vii. 1910

FIGURE 3 Vertical temperature distribution at a series of stations along a meridian in the Atlantic Ocean. (Reprinted with permission from Defant, 1961.)

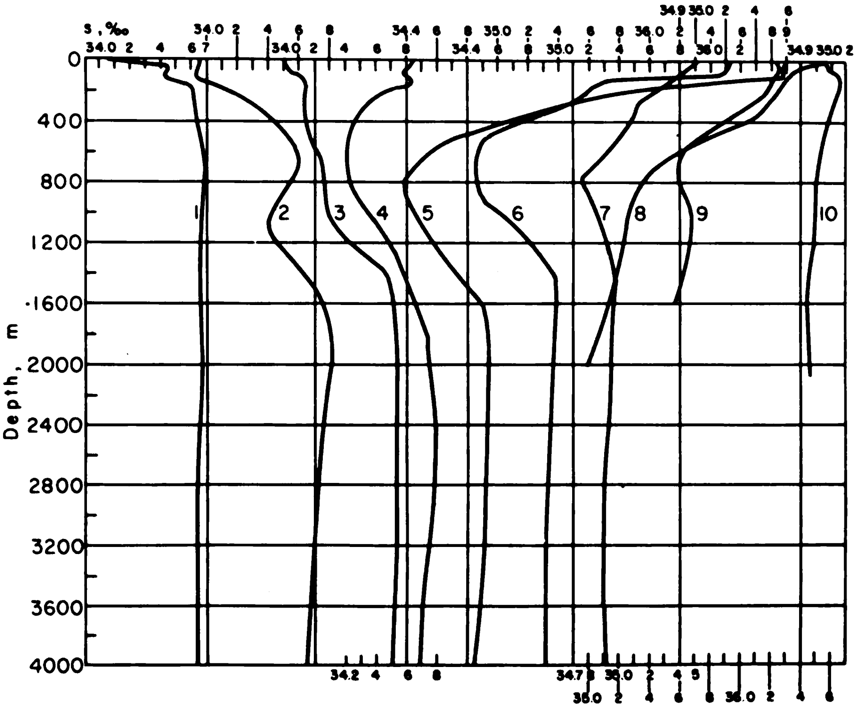


FIGURE 4 Vertical salinity curves for a series of oceanographic stations along a meridional section through the Atlantic (see legend of Figure 3); corresponding vertical temperature curves are shown in Figure 3. (Reprinted with permission from Defant, 1961.)

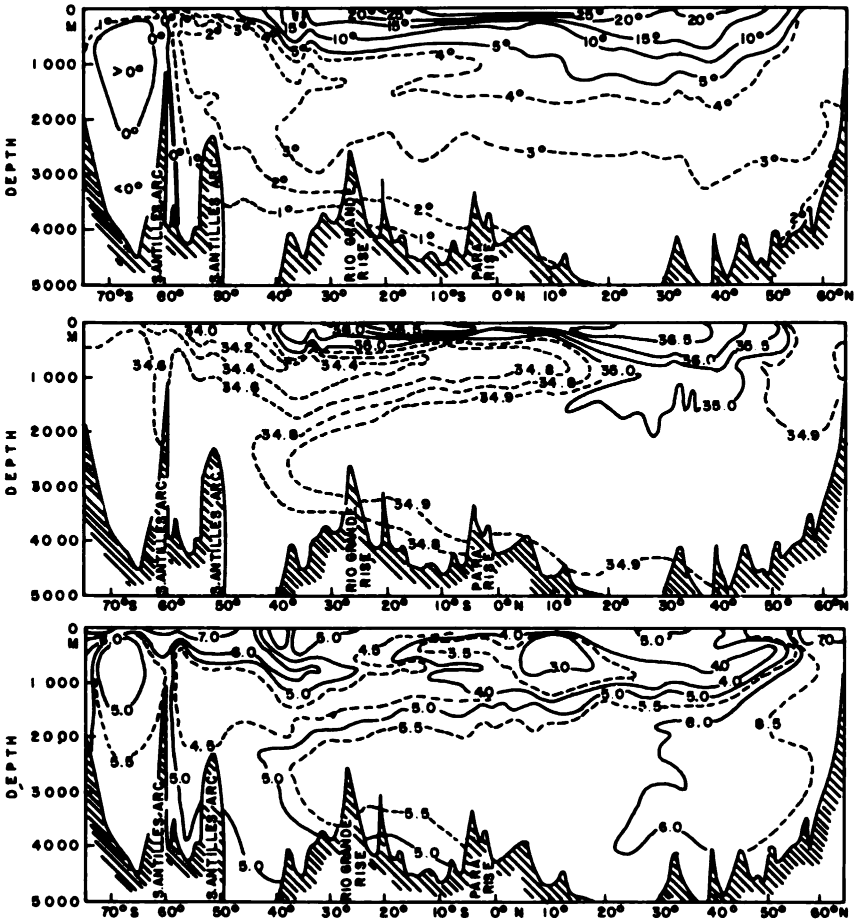


FIGURE 5 Vertical sections showing distributions of temperature, salinity, and oxygen in the western Atlantic Ocean. (Reprinted with permission from Wüst, 1935.)

The salinity profiles at middle and tropical latitudes show more complicated structure above 1,500 m. A dominant feature in the South Atlantic is the salinity minimum at depths between 800 and 1,200 m, which has its origin in the Antarctic Convergence zone, at about 50°S.

Figures 5 and 6 show vertical cross-sections of temperature, salinity, and dissolved oxygen (in units of ml per liter) for the Atlantic and Pacific, respectively. These cross-sections extend from the Antarctic continent (on the left) nearly to the Arctic Circle. Figure 5, for the Atlantic, shows very clearly the northward intrusion of Antarctic intermediate water, with its core at a depth of roughly 1,000 m; this water is characterized by its relatively low temperature, low salinity, and high dissolved oxygen content. The apparent source of this water is at the surface near 50°S (the Antarctic Convergence zone). The oxygen section for the Atlantic shows a southward intrusion of high oxygen content with a core at about 3,000-m depth, which has its source in the North Atlantic between 50° and 60°N. Sverdrup *et al.* (1942) give evidence for a region of winter sinking in an area just south of Greenland (the Irminger Sea), which they proposed as a primary source of deep water in the North Atlantic. Figure 6, for the Pacific, does not indicate any similar source region.

The temperature sections for both the Atlantic and Pacific suggest a northward intrusion, at the bottom, of cold water that apparently originates at the surface near the Antarctic continent with the onset of freezing during the southern winter. The northward intrusion of the Antarctic bottom water into the Atlantic is shown very clearly in a chart of temperature at 4,000 m, presented in a paper by Stommel (1955).

Figure 7 is a cross-section of density for the Atlantic Ocean. The values indicated on the contours represent density anomaly (in mg/ml), neglecting the influence of pressure. The value 27.0 corresponds to 1.0270 g/ml for a sample at 1 atm but at the *in situ* temperature and salinity. Except at great depth, the values of the density anomaly must increase with depth for gravitational stability of the water column. Hence, there is a noticeable absence of core structure, such as that indicated in the diagrams of Figure 5. The most stable water is located near the equator; the least stable water is in the northern latitudes, particularly during the winter season for the hemisphere concerned. Clearly, it

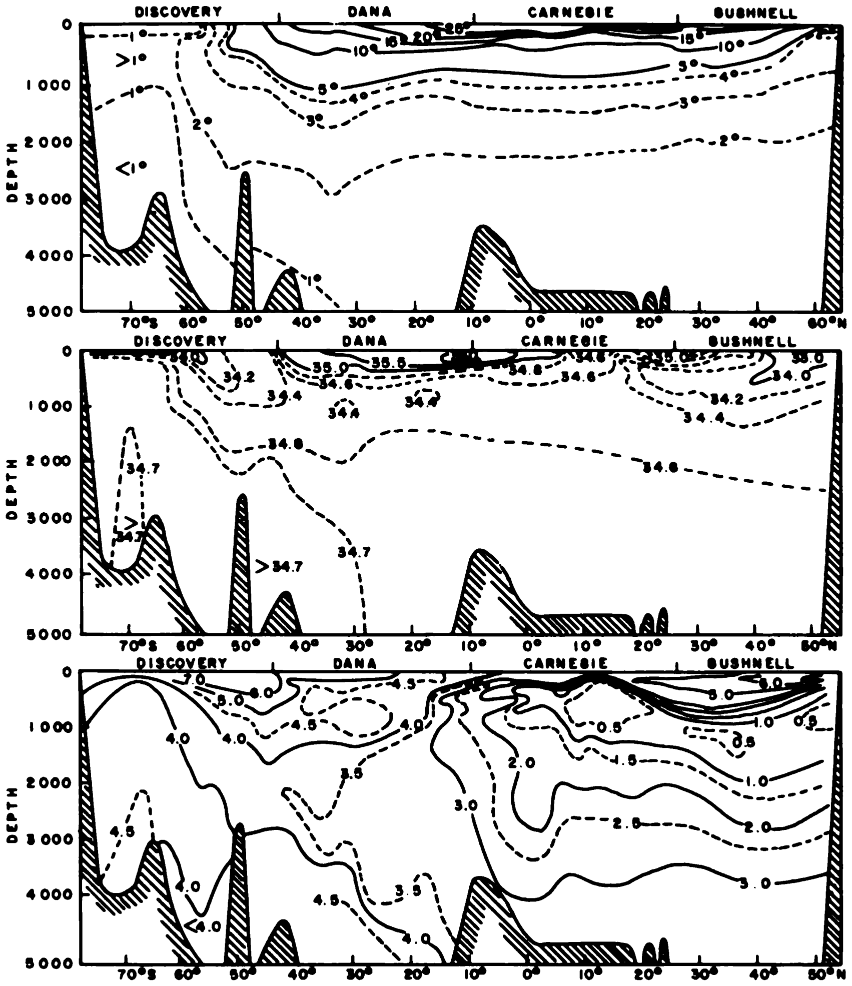


FIGURE 6 Vertical sections showing distribution of temperature, salinity, and oxygen in the Pacific Ocean, approximately along 170° W. (Reprinted with permission from Sverdrup *et al.*, 1942.)

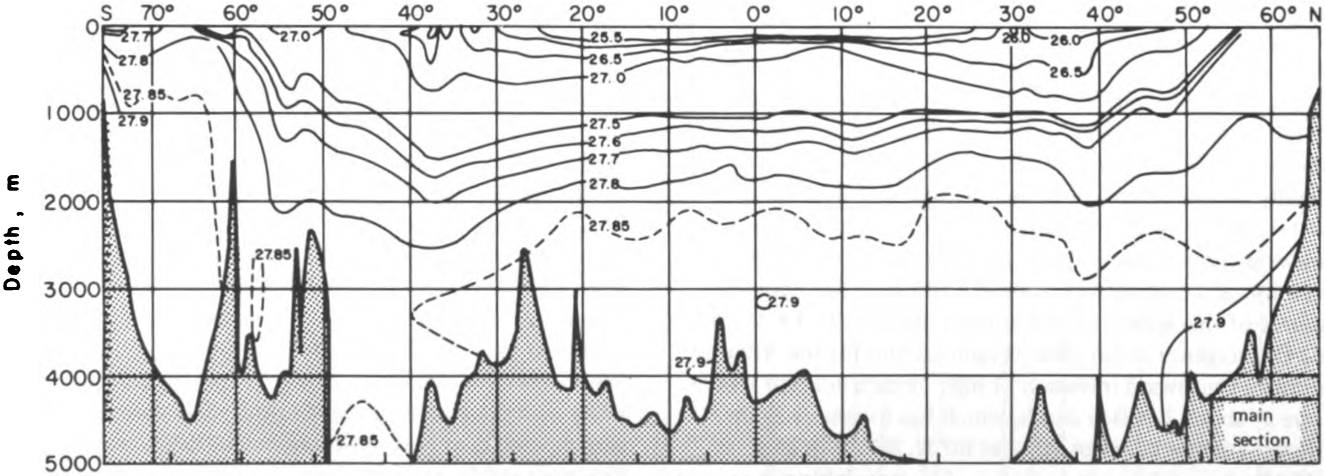


FIGURE 7 Vertical density section along the western trough of the Atlantic, corresponding to temperature-salinity sections of Figure 5. (Reprinted with permission from Defant, 1961.)

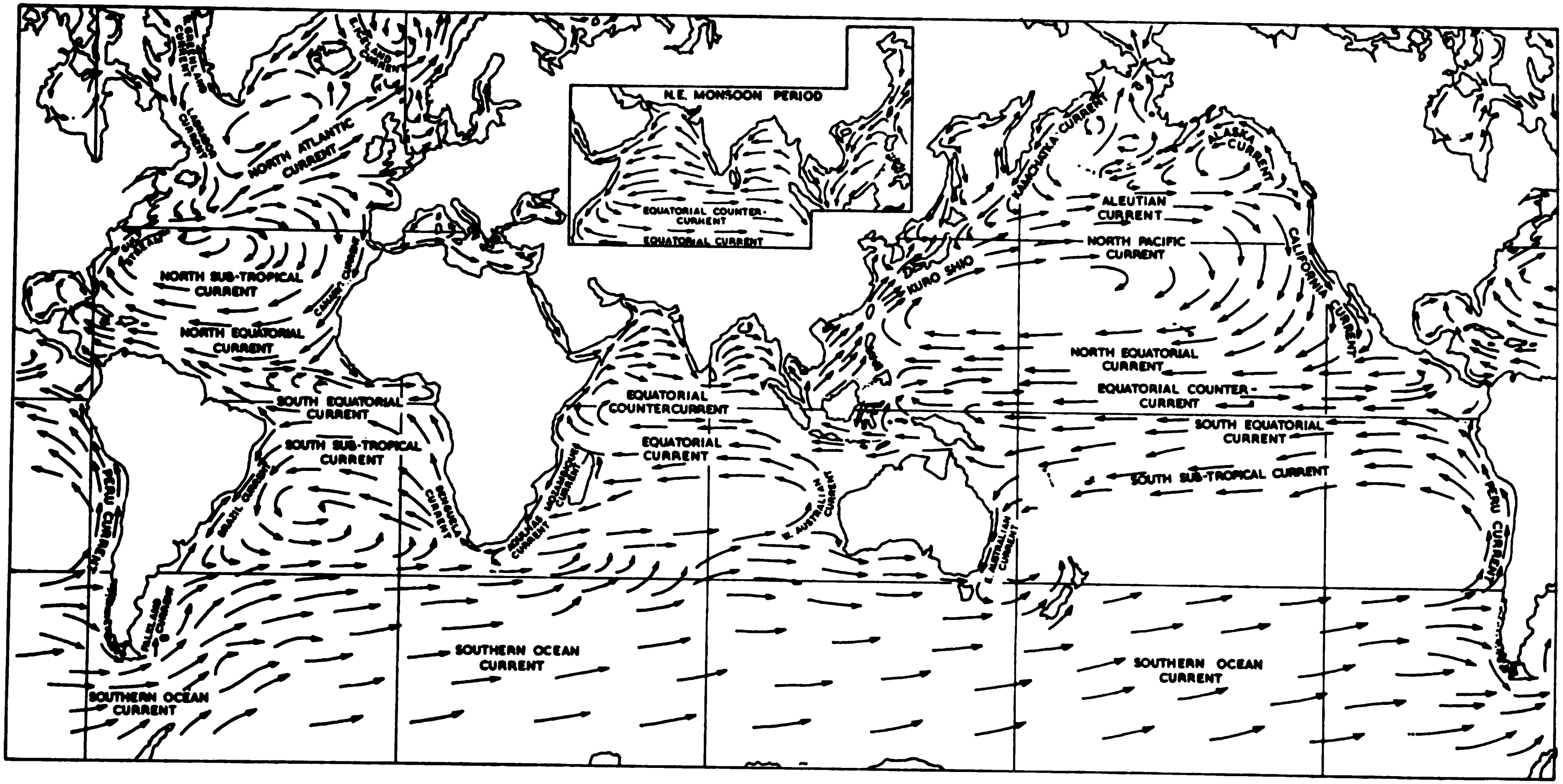


FIGURE 8 Surface currents of the ocean. (After Somerville and Woodhouse, 1950.)

is in the latter regions that excess cooling can result in vertical convection and provide the source for the deep and bottom water renewal. Complete renewal of the Common Water has been estimated by Bolin and Stommel (1961) to require on the average about 1,200 years. This can be compared to Montgomery's (1959) estimate of 10 years for the residence time of the surface mixed layers of the world ocean and to Bolin and Stommel's estimate of from 100 to 400 years residence time for the Atlantic Intermediate Water.

For more detailed information about the distribution of properties, the reader may consult the works of Wüst (1935, 1949), Defant (1961), Fuglister (1960, 1963), Reid (1965), and others. The paper by Reid presents a very complete and up-to-date analysis of the properties of the Pacific Ocean.

Surface and Near-Surface Currents

Figure 8 shows the general features of the mean surface current pattern of the world ocean. More detailed charts showing magnitudes are published by the U.S. Navy Oceanographic Office. Although such currents as the Gulf Stream and Kuroshio are semipermanent, they change in strength and configuration from month to month (cf. Stommel, 1957; Warren, 1963; Fuglister, 1963). The most striking, but perhaps least studied, variable current regime is that of the northern and equatorial part of the Indian Ocean, the variations being strongly coupled with the pronounced seasonal changes of the wind pattern (monsoons).

Magnitudes of the quasipermanent currents (i.e., nontidal currents) generally do not exceed about 250 cm/sec (5 knots). Values of this magnitude have been reported for the core of the Gulf Stream and Florida Current. More frequently, the stronger of the major current systems at the surface flow at less than 200 cm/sec. However, the lateral width of strong currents like the Gulf Stream is generally less than 100 km (the distance over which speeds exceed 10 percent of the core strength). Consequently, in the major area of the ocean surface, away from these "rivers of the sea," one finds typical speeds of 0 to 30 cm/sec, values that are comparable to the magnitude of tidal currents in the open sea.

The depth of the northward-flowing Gulf Stream is about 1,000 to 1,500 m, depending upon location, and its speed decreases with increasing depth (Stommel, 1957). This is in marked contrast to the depth of the major equatorial current systems (less than about 500 m). Indeed, the surface Equatorial Current, which flows toward the west in all three oceans, is now known to be a superficial feature, limited to perhaps a few decameters in depth. Hidden beneath this surface current, in all three oceans, lies the major, jet-like Equatorial Undercurrent, with speeds up to 150 cm/sec in its core and extending to depths of 400 to 500 m (cf. Knauss, 1960; Knauss and Taft, 1964; Metcalf *et al.*, 1962; Knauss, 1966; Cochrane, 1963; Stalcup and Metcalf, 1966).

The Equatorial Undercurrent of the Pacific has its core at the equator and is confined to the narrow band between about 1°N and 1°S. It has been traced as a well-defined subsurface stream from the Gilbert Islands, at 174°E, to the Galapagos Islands, at 92°W. The total transport has been estimated to be about 40 million m^3/sec (greater than that of the Florida Current, which is 26 million m^3/sec).*

The volume transports in the upper 1,000 m of the North Atlantic as estimated by Sverdrup *et al.* (1942) are indicated in Figure 9. The values for the Gulf Stream are now known to be somewhat low, the maximum transport off Cape Hatteras being closer to 80 sv than to 55 sv. Moreover, the amount of water sinking in the Subarctic Convergence region near Greenland has been estimated by Stommel and Arons (1960b) to be more nearly 20 sv instead of 4 sv. The latter value is only a rough estimate. Observational evidence for deep currents certainly supports the idea that Sverdrup's initial estimate of 4 sv for the North Atlantic rate of supply to the deep water is too small. If this is true, then the compensational flow northward across the equator from the South Atlantic must be proportionately greater than indicated in Figure 9. Nevertheless, the figure does display the major qualitative features of the flow in the upper 1,000 m.

The Antarctic Circumpolar Current is unique in that it represents the only current system on earth that is nearly a continuous zonal current circling the entire globe. This system is much deeper than 1,000 m, except in the region of the Drake Passage, between South America and the Antarctic continent. Its average total transport has been estimated to be in excess of 100 sv (Sverdrup *et al.*, 1942). However, this figure is quite uncertain in the light of arguments advanced by Stommel and Arons (1960b). In any event, the qualitative features of the flow as presented by Sverdrup (Figure 10) are probably valid.

Intermediate and Deep Circulation

The currents at a depth of 800 m as estimated from the field of density, using the geostrophic balance relation, are shown for the Atlantic Ocean in Figure 11. These estimates are based on a level of no motion, which is taken to be consistent with the spreading of the Antarctic Intermediate Water. Figure 11 indicates current directions substantially similar to those of the surface flow, except off the coast of Argentina. The speeds are generally less than 30 cm/sec.

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*For convenience in dealing with volume transports by currents, we will define a transport of 1 million m^3/sec as a sverdrup (abbreviated sv). For example, the average volumetric rate of exchange of water between the Mediterranean Sea and the Atlantic is about 1.7 sv.

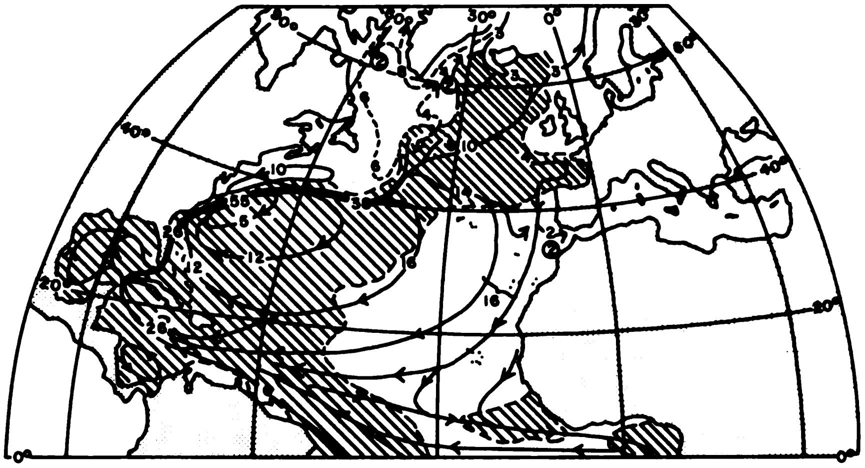

FIGURE 9 Transport of Central Water and Subarctic Water in the Atlantic Ocean. The lines with arrows indicate the direction of the transport, and the inserted numbers indicate the transported volumes in millions of cubic meters per second. Full-drawn lines show warm currents; dashed lines show cold currents. Areas of positive temperature anomaly are shaded. (Reprinted with permission from Sverdrup *et al.*, 1942.)

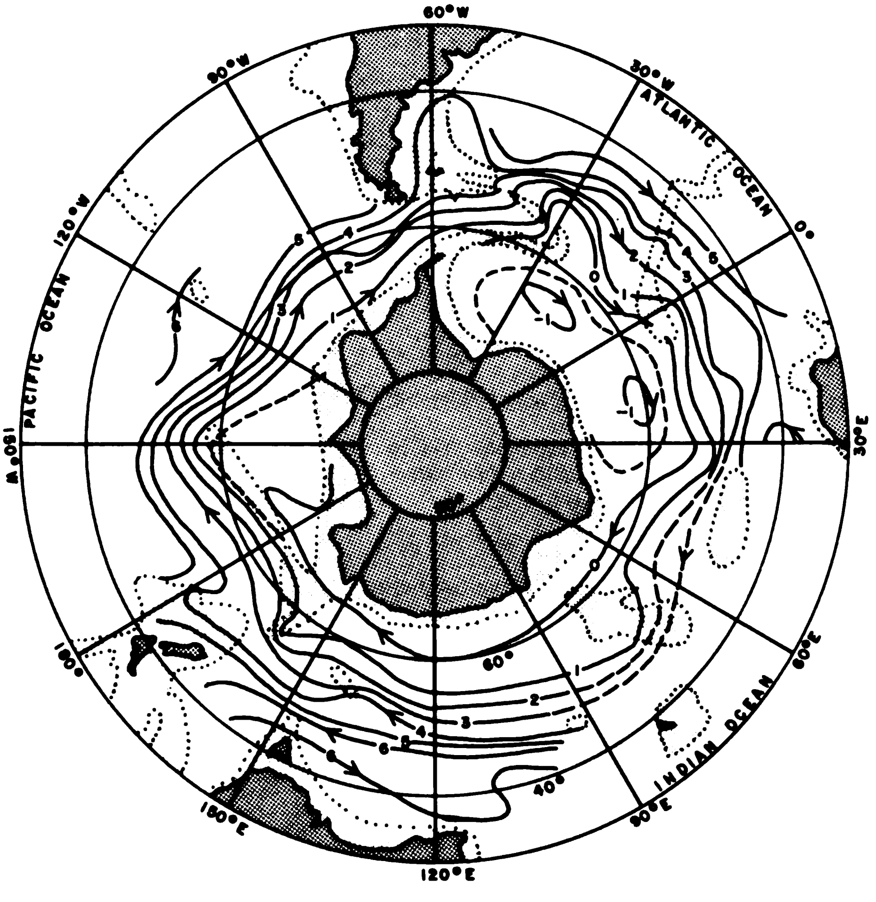

FIGURE 10 Transport lines around the Antarctic Continent. Between two lines, transport relative to the 3,000-decibar surface is about 20 million m^3/sec. (Reprinted with permission from Sverdrup *et al.*, 1942.)

Defant (1961) also estimated the currents at 2,000-m depth in the Atlantic based on the same reference level. The resulting picture of flow at this depth, as shown in Figure 12, is in striking contrast to those of flow at the surface (Figure 8) and at a depth of 800 m (Figure 11), particularly in the North Atlantic. At 2,000 m, a southward flow exists beneath the Gulf Stream and also along the coast of South America to 25°S, and current speeds are generally less than 20 cm/sec.

Although some question exists concerning Defant's selection of a spatially variable level of no motion, the fact remains that his picture of flow is strikingly similar to that predicted by Stommel and Arons (1960a, b), based on theoretical grounds. (We will return to this point later.) Further

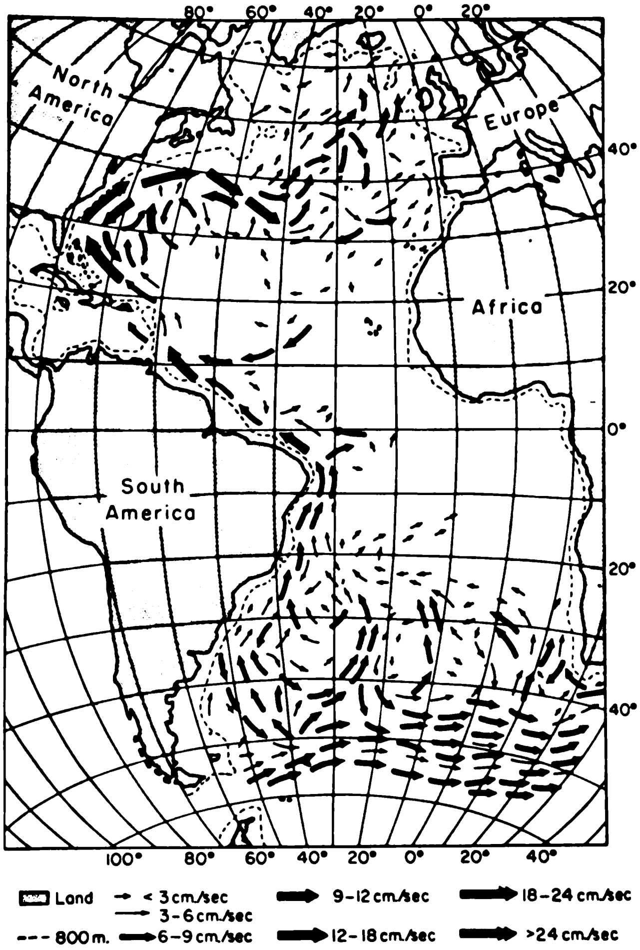

FIGURE 11 Current field at a depth of 800 m, computed from the absolute topography of the 800-decibar surface. (Reprinted with permission from Defant, 1961.)

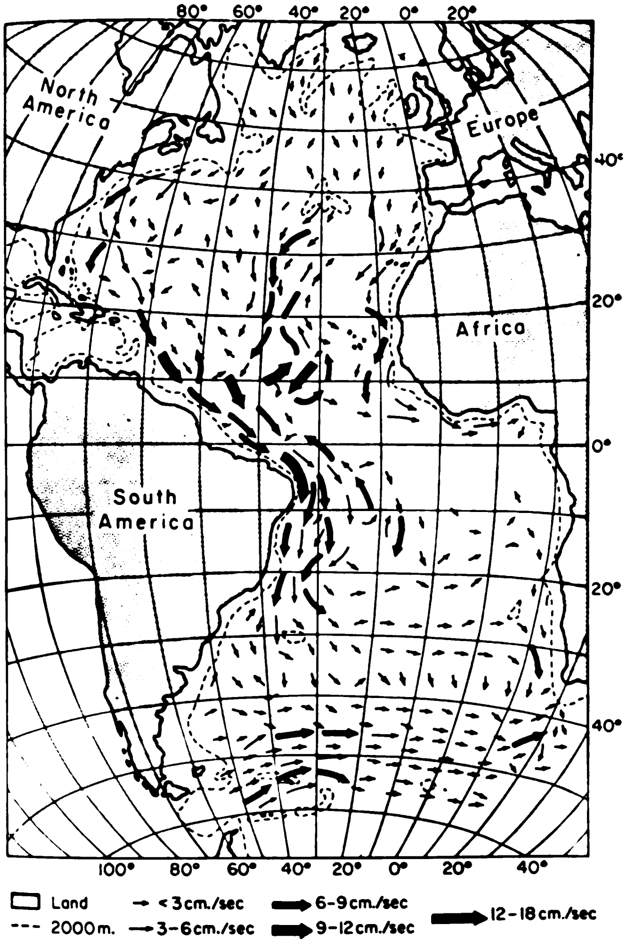

FIGURE 12 Current field at a depth of 2,000 m, computed from the absolute topography of the 2,000-decibar surface. (Reprinted with permission from Defant, 1961.)

support for the southward set of the deep currents along the western boundary of the North Atlantic is provided by the direct observations of Swallow and Worthington (1961), Volkmann (1962), and others. Table 3 is a reproduction of the results of deep current measurements in the region at or near 33°N, 75°W made by Swallow and Worthington with neutrally buoyant floats. The mean current indicated by these measurements, for the layer between 1,500 and 3,000 m, is about 8 cm/sec in a compass direction of 210°. Since the combined duration of these measurements is about 27 days, the tidal "noise" is essentially eliminated. However, the variability from one measurement to another is still considerable (a range of 18 cm/sec). Stommel (1963) and Longuet-Higgins (1965) have suggested that such variability might be attributed to planetary wave noise associated with the inherent variability of the atmospheric driving processes (primarily the stress of the winds).

Some notion of the three-dimensional structure of circulation in the Atlantic Ocean is provided in a schematic diagram constructed by Wüst (1949), which is reproduced in Figure 13. (Note that this figure shows north latitudes on the left and south on the right, in contrast to previous figures.)

Theoretical Aspects

In the central oceanic regime at mid-latitudes, away from the equator and the intense western boundary currents, the flow is characterized by very small Rossby number and small Reynolds number (based on the horizontal eddy viscosity).*

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*The Rossby number is fluid speed divided by a characteristic velocity associated with the earth's rotation, which may be taken as rf where r is the mean radius of the earth and f is the local vertical component of the earth's vorticity (Coriolis parameter). Specifically, $f = 2\Omega \sin \varphi$, where Ω is the earth's angular speed and φ is the latitude.

The Reynolds number is Vr/K_h, where V is the fluid speed and K_h is the horizontal eddy viscosity associated with lateral turbulent mixing processes.

TABLE 3 Summary of Deep Current Measurements with Neutrally Buoyant Floats, March-April 1957, near 33°N, 75°W[a]

Float	Launched		Last Fix		Mean Observed Depth ±SD (m)	Mean Velocity ±SD (cm/sec)	°T ±SD
	Time	Date	Time	Date			
B	1000	6	0743	11	2,040 ± 70	0.33 ± 0.11	108 ± 18
D	1715	17	0718	22	2,550[b] ± 40	4.27 ± 0.21	201 ± 1.4
						1.88 ± 0.11	235 ± 4
E	1527	20	1717	22	1,480 ± 50	6.42 ± 0.47	308 ± 4.7
						6.50 ± 0.29	231 ± 2.3
F	0954	23	0733	25	2,620[b] ± 80	8.99 ± 0.53	190 ± 2.3
G	2050	23	1032	26	2,600[b] ± 50	4.41 ± 0.26	218 ± 3.3
						7.08 ± 0.44	203 ± 2.3
H	0815	26	0605	29	2,910[c] ± 70	18.36 ± 0.28	182 ± 1.0
I	0854	26	1040	29	2,760[c] ± 190	6.02 ± 0.27	216 ± 2.6
						12.62 ± 0.39	196 ± 1.3
J	1110	30	1103	2	2,900[c] ± 120	10.24 ± 0.18	204 ± 1.7
						9.42 ± 0.22	185 ± 11
K	1156	30	0935	31	2,770[c] ± 200	12.95 ± 0.59	207 ± 5.2

[a] Reprinted with permission from Swallow and Worthington, 1961.
[b] Mean for D, F, and G = 2,580 m.
[c] Mean for H, I, J, and K = 2,840 m.

It can be shown (Stommel, 1957) that, under these conditions, the balance of torques on a fluid column extending from level z to the surface, under steady state conditions, is given approximately by the relation

$$\beta \int_z^0 u dz = fw + \left(\frac{\partial \tau_y}{\partial x} - \frac{\partial \tau_x}{\partial y}\right). \tag{1}$$

Here, x, y, and z represent local cartesian coordinates in the eastward, northward, and upward directions, respectively. The term f is the Coriolis parameter defined in the footnote, and β is the derivative of f in the y direction. The eastward and northward velocity components are denoted by u and v, and the vertical by w, at level z in equation (1). Finally, τ_x and τ_y represent the eastward and northward components of wind stress at the surface (nominally at $z = 0$). It is assumed that the level z is taken below the depth of frictional influence (Ekman layer), which may extend to depths of about 100 or 200 m.

The physical interpretation of relation (1) is as follows: The term in parentheses is a wind stress torque per unit area; the term fw is the vorticity tendency term associated with stretching or shrinking of the water column; and the term on the left is the planetary vorticity tendency term, which is associated with displacement of the water column to a different latitude. If z is taken at the variable elevation of the seabed $[z = -D(x,y)]$, then fw leads to a topographical influence that can be pronounced near the continental slopes (Warren, 1963).

In the central abyssal region, in which the seabed is regarded as horizontal, w must vanish at the bottom and Eq. (1) reduces to

$$\beta \int_{-D}^0 v dz = \left(\frac{\partial \tau_y}{\partial x} - \frac{\partial \tau_x}{\partial y}\right) \tag{2}$$

for a column extending to the seabed. This is Sverdrup's approximation (1947); from it, one can calculate the meridional volume transport resulting from the wind-stress field as derived from climatological information. Using the continuity equation for the column,

$$\frac{\partial}{\partial x} \int_{-D}^0 u dz + \frac{\partial}{\partial y} \int_{-D}^0 v dz = 0, \tag{3}$$

one can also evaluate the zonal component of volume transport, taking the latter as zero on the eastern boundary. In order to satisfy continuity on the western boundary of the ocean, we require a narrow frictional or inertial boundary current (Munk, 1950; Charney, 1955; Stommel, 1957). In the western boundary current regime, either lateral friction or inertial terms must be introduced into Eq. (2); indeed,

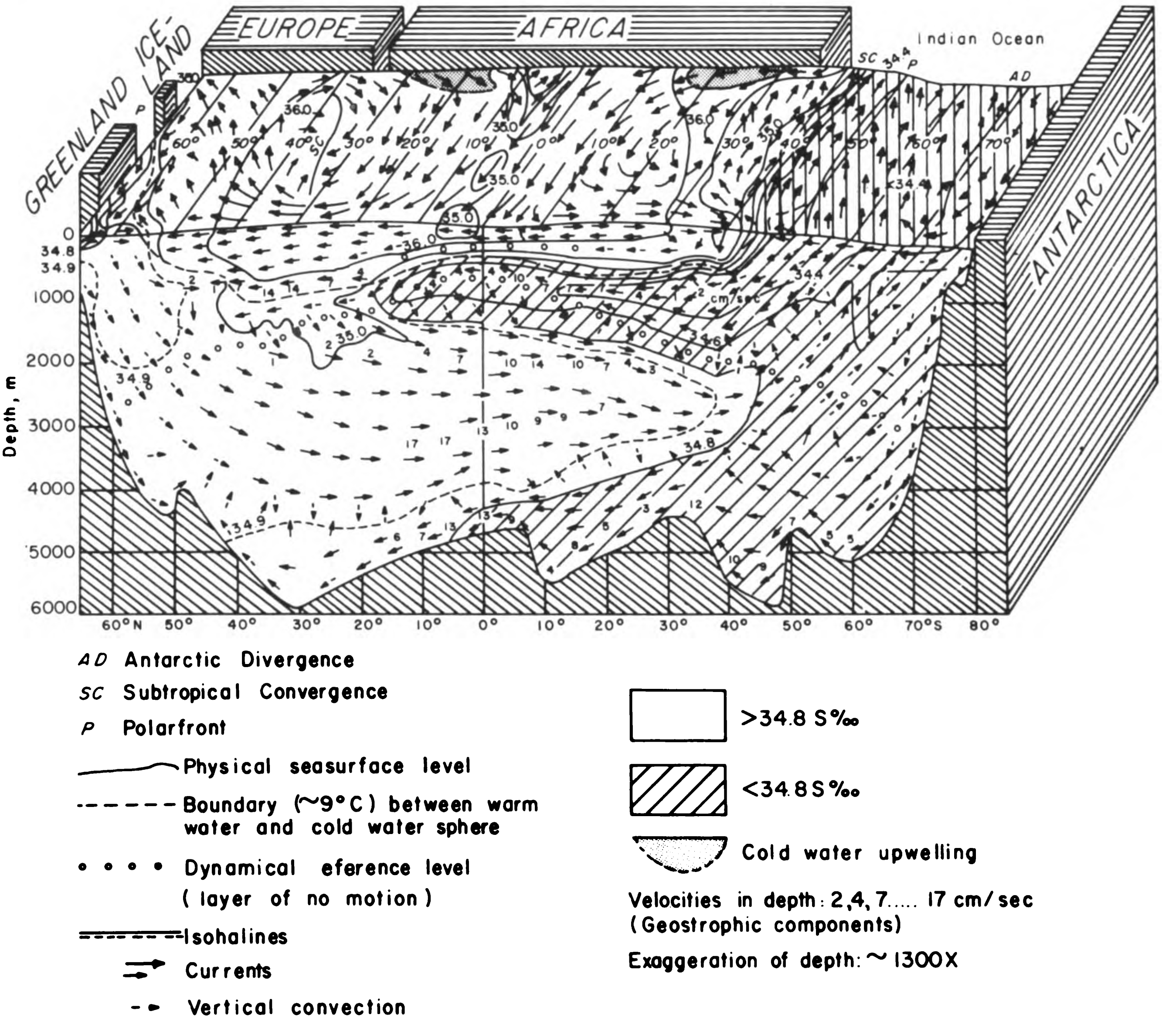

FIGURE 13 Schematic block-diagram of the surface currents, of salinity distribution, and of the deep sea circulation of the Atlantic Ocean. (After Wüst, 1949, corrected.)

these effects dominate over the wind torque term in the boundary current regime.

Relations (2) and (3) are valid in the central oceanic regime only for the total column of fluid (and even then only if the seabed is horizontal or if there is negligible motion at the bottom). These relations tell nothing about the internal vertical circulation or about differences of circulation in the upper and lower layers of the sea individually.

In order to examine the transports in a layer from the surface to, say, 1,000-m depth ($z = -1{,}000$ m), one must use Eq. (1) with w evaluated at that depth. On the other hand, for the layer from 1,000 m to the mean sea floor (–4,000 m), the relation

$$\beta \int_{-4{,}000}^{-1{,}000} v\,dz = -fw \tag{4}$$

is employed such that the sum of Eqs. (1) and (4) is consistent with Eq. (2). Moreover, the continuity relation for the upper 1,000 m is

$$\frac{\partial}{\partial x}\int_{-1{,}000}^{0} u\,dz + \frac{\partial}{\partial y}\int_{-1{,}000}^{0} v\,dz = w. \tag{5}$$

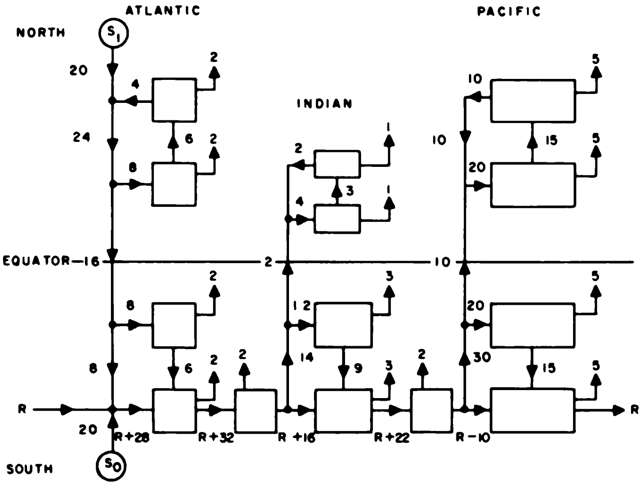

FIGURE 14 Schematic budget of transports below the 1,000-m depth in various portions of the world oceans. Numbers represent transports in units of 10^6 m^3/sec (sv). (Reprinted with permission from Stommel and Arons, 1960b.)

The relations (1), (4), and (5) can be useful only if the vertical velocity, *w*, can be determined independently of these relations. The estimation of *w* for the steady state oceanic convection regime has been examined theoretically by Robinson and Stommel (1959) and later by Robinson and Welander (1963) and others. Robinson and Stommel found that a vertical (turbulent) exchange coefficient of about 0.7 cm^2/sec leads to a realistic temperature profile for mid-latitude conditions. Moreover, they found an associated upward vertical velocity, typical of mid-latitudes, of about 10^{-5} cm/sec (about 1 cm/day). This upward motion is required in order to balance the downward diffusion of heat.

In a later paper, by Stommel and Arons (1960b), the above thermohaline theory is employed to estimate the total upward transport from the deep water of the world ocean to the upper 1,000 m of the ocean. A value of about 40 sv is obtained. This upward transport must be balanced by sinking in the deep and intermediate water source regions (in the North Atlantic and the Antarctic region). They estimate that half of this sinking (20 sv) occurs in the North Atlantic near Greenland.

Using Eqs. (1), (4), and (5) and an assumed uniform upwelling rate through the permanent thermocline of the world ocean, Stommel and Arons were able to obtain crude estimates of the deep-water transports within the westward boundary currents of the Atlantic, Indian, and Pacific oceans. With the exception of the Antarctic Circumpolar Current, they were also able to estimate the circulation within the deep water of the North Atlantic, the South Atlantic, and other areas. The results are shown schematically in Figure 14. The directions and amounts of the transport between various segments of the schematic deep ocean are indicated (R being an unknown deep-water transport around the Antarctic continent).

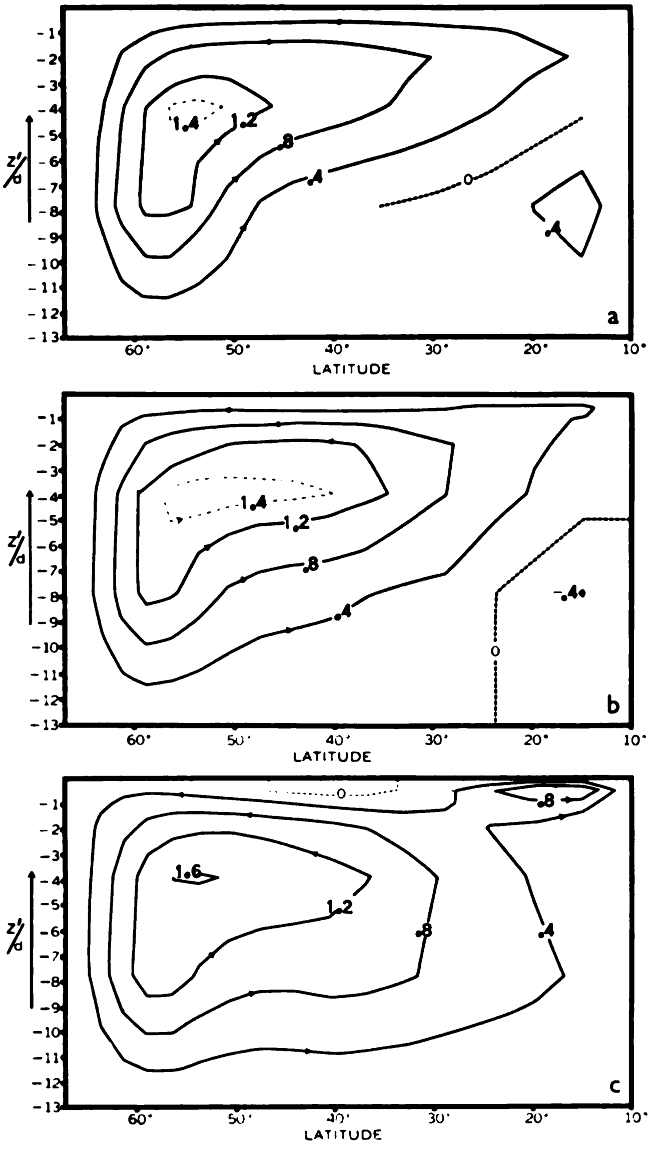

FIGURE 15 Streamlines for the total mass transport in a northern hemisphere ocean, in the vertical meridional plane, based on the numerical model of Bryan and Cox (1967): (a) no wind; (b) moderate zonal wind; and (c) strong zonal wind. (Reprinted with permission from Bryan and Cox, 1967.)

In the theory presented by Stommel and Arons, it is implied that the major current in deep water occurs on the western boundary. However, it must be borne in mind that their theory deals with an ocean of uniform depth. In the Atlantic, the Mid-Atlantic Ridge represents a formidable barrier to the deep currents, and indeed the deep-water flow pattern obtained by Defant (Figure 12) shows a strong deep current with southward set just east of the Mid-Atlantic Ridge. Defant's pattern is supported by a more recent analysis by Lappo (1963) that makes use of direct observations together with the observed density field.

Serious efforts have recently been directed toward the problem of numerical modeling of the ocean circulation (Bryan, 1963; Gormatyuk and Sarkisyan, 1965; Holland, 1966; Bryan and Cox, 1967). The most comprehensive of these models is that of Bryan and Cox, which employs the complete set of hydrodynamic equations and the thermal diffusion-advection equation. They carried out a parametric study of the numerical model for different Rossby numbers, Reynolds numbers, and relative wind strengths. The primary deficiency of their present model (presumably for the North Atlantic) is that there is no allowance for the important interchange with the southern oceans across the equator. Figure 15 shows a plot of the steady-state, zonally averaged streamlines in a vertical section from 10°N to about 65°N as calculated by Bryan and Cox. The strength of the vertical circulation is changed only slightly by the wind; the governing factor in controlling overturning is the impressed meridional temperature distribution at the sea surface.

The total upward transport for the average of these results is about 38 sv, based on realistic conditions for the parameters as stipulated by Bryan and Cox. This is about twice the estimate of Stommel and Arons (1960b) for the North Atlantic and nearly the same as the estimated total upwelling for the world ocean as a whole. Apparently, the reason for this excess overturn rate is that although the model is confined to a basin whose size and surface temperature distribution are similar to those of the North Atlantic, no allowance has been made for interchange with the remaining 8/9 of the world ocean. Accordingly, the estimated residence time in deep water of only about 100 years, implied by the Bryan and Cox model, is probably low by a factor of nine.

Aside from this limitation of the present application of the model, the numerical technique does offer considerable potential with respect to the analysis of the distribution of contaminant introduced into the sea.

MIXING PROCESSES IN THE MARINE ENVIRONMENT

In addition to local, continuous, extremely low-level sources, such as the processed liquid effluent from a shore-based nuclear installation, artificially produced radioactivity might also be introduced into the sea through local instantaneous release, such as might result from an accident to a seaborne or airborne nuclear power source, for example. In such releases, fairly local problems will be important, such as the maximum concentration in a patch of radioactivity after a known interval of time, or the concentration along the centerline of a plume of radioactivity released from a continuous source. To understand these problems, we must acquire fairly precise knowledge of the physical processes of movement and dispersion of material due to oceanic currents and turbulence rather than general knowledge of the overall rates of exchange, as in a "box model."

Movement and diffusion of a radioactive material released into the sea may be classified, according to the conditions under which it is released, as follows: instantaneous source, continuous fixed source, or continuous moving source. The intensity of the continuous fixed and continuous moving sources may vary with time, and each may be regarded as either infinitesimal or finite in size, depending upon the scale of diffusion.

In the theory of diffusion or conduction in solids, the solution for an instantaneous point-source is regarded as fundamental (Carslaw and Jaeger, 1959). Thus, by integrating the fundamental solution with respect to time with an appropriate source intensity, we obtain the solution for the continuous fixed point-source. By integrating with respect to appropriate space variables, we obtain solutions for instantaneous finite-sized sources. Solutions of a large number of other important problems can be obtained immediately from the fundamental solution. Such a fundamental solution, in a rigorous sense, does not exist for the diffusion problem for a turbulent field: knowledge of the concentration distribution for an instantaneous point-source is not sufficient to obtain the distribution of substance from a continuous point-source fixed in a turbulent field. In diffusion of the continuous point-source type, we must not only take into consideration diffusion relative to the centers of mass of individual elementary patches,* but we must also consider variabilities in the location of these centers of mass.

In practical problems, however, we may approximate, in our calculations, the concentration from a continuous fixed point-source simply by superimposing an infinite number of diffusing patches from an instantaneous source, all of which are assumed to move at a single mean velocity. More will be said about this approach later.

Instantaneous Releases

The horizontal scale of turbulence in the sea is usually so much greater than the vertical scale that, for many purposes, their effects on diffusion can be considered separately. Revelle *et al.* (1956) cited an example of diffusion of radioactive material released below the thermocline; the material spread over an area of 100 km^2 while its vertical extent did not exceed 1 m. Similarly, Folsom and Vine (1957) described the horizontal spread of radioactive tracer over an

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*Which may be considered as being of the instantaneous point-source type.

area of 40,000 km^2 in 40 days while it remained in a surface layer less than 60 m deep.

These experimental facts seem to indicate that the horizontal components of oceanic turbulence play an essential role in the horizontal spread of material introduced into the marine environment. A method of approach to the oceanic diffusion problem suggested by this finding has been used by various investigators. In this approach we first assume that the substance is subject to horizontal diffusion within a sufficiently thin homogeneous layer that all vertical variations in concentration may be ignored. We further assume that it is possible to describe the distribution; after some time has elapsed, with lines of equal concentration. In other words, we treat the problem as if the diffusion took place on a purely two-dimensional horizontal plane and was due to horizontal isotropic turbulence. The study of horizontal diffusion using this approach has permitted prediction, with some success, of the spread of introduced substance (Joseph and Sendner, 1958, 1962; Ozmidov, 1958; Okubo, 1962a).

The importance of vertical diffusion, however, when combined with vertical shear in a mean flow, should not be underestimated even though the diffusing material is practically confined within a very thin layer. As a matter of fact, another approach based on the interaction of a shearing current with transverse mixing does give rise to horizontal diffusion at a scale comparable to that of the purely horizontal spread due to horizontal eddies (Bowles *et al.*, 1958; Bowden, 1965; Okubo, 1966; Pritchard *et al.*, 1966). Presumably, both approaches are partially correct; each supplements the other. We shall discuss the two approaches separately.

RADIALLY SYMMETRICAL SOLUTIONS OF HORIZONTAL DIFFUSION DUE TO HORIZONTAL TURBULENCE

The following discussion considers primarily the two-dimensional horizontal diffusion of a radioactive substance introduced instantaneously at a point in the sea. The motion of a patch of diffusing radioactive material is a sort of irregular spreading superimposed on an overall wandering of its center of mass. The wandering motion is attributed to the combined motion of large-scale currents (see previous section). On the other hand, the spreading of the patch is controlled chiefly by eddies whose size is comparable to or smaller than that of the patch. We are interested mainly in discussing the distribution of radioactive material about the moving center of mass.

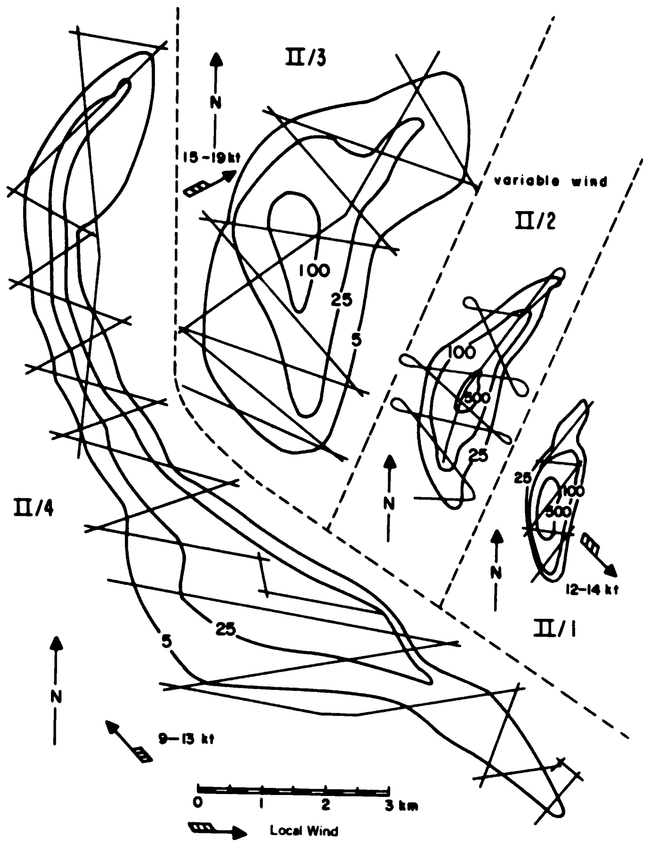

FIGURE 16 Patterns of dye patch on a horizontal plane. (Reprinted with permission from Joseph *et al.*, 1964.)

At any time after release, the shape of the isolines of any concentration will be very irregular (Figure 16); the patch of radioactivity will usually be elongated in one direction or another. In the discussion that follows, we shall visualize an infinite number of releases under identical oceanographic conditions, using for each the same amount of radioactive material. The centers of mass of the different patches will have different histories owing to the nonstationary and nonhomogeneous character of the large eddies. However, as far as the relative distribution of substance with respect to the center of mass is concerned, certain statistical properties of individual patches may be almost equivalent from one patch to another since the smaller eddies responsible for the relative diffusion are supposed to be stationary and homogeneous (and probably even isotropic) in a horizontal plane. Thus, if we take an infinite number of distributions, each observed after the same time interval following release, and if we superpose them in such a way that the centers of mass coincide with each other, and if we average all of the superposed distributions, we would then expect a radially symmetrical distribution of substance about the center of mass, which at the same time would be the point of maximum concentration, provided that the mean flow pattern is isotropic. In other words, the mean concentration is a function only of the diffusion time, *t*, and the distance, *r*, from the center of mass. The very irregular distribution of concentration in the individual release can be regarded, so far as the rate of dispersion is concerned, as a particular realization from the ensemble from which the averages are formed; thus, the area enclosed by a line of constant concentration at a time, t, after release, would be nearly the same for all the identical releases.

We shall now discuss a possible form of the radially symmetrical distribution. Assume that, with the passage of time, the pattern of concentration relative to the maximum concentration at the center of mass remains relatively unchanged as the characteristic length of dispersion grows and the peak concentration decreases. We may express this formally as

$$S(t,r) = S_0(t,o)\, F\left(\frac{r}{b(t)}\right) e^{-\lambda t}, \tag{6}$$

where $S_0(t,o)$ represents the peak concentration at the center of mass of a radioactive patch, $b(t)$ is the characteristic length of dispersion, F is a decreasing function of the argument r/b, and the factor $e^{-\lambda t}$ represents the decay of radioactivity.

Since the total amount of radioactive material released over a period of time is conserved if radioactive decay is taken into account, we have

$$\int_0^\infty S(t,r)\, e^{\lambda t}\, 2\pi r dr = M/D, \tag{7}$$

where M is the total amount of material released, say, so many curies, and D is the depth of water within which the radioactivity is assumed to be distributed uniformly. The substitution of Eq. (6) into Eq. (7) yields

$$S_0(t,o) = \frac{M/D}{2\pi\Phi b^2(t)},$$

where Φ is a form-constant, its value dependent upon the pattern of concentration.

Hence, we write

$$S(t,r) = \frac{M/D}{2\pi\Phi b^2(t)}\, F\left(\frac{r}{b(t)}\right) e^{-\lambda t}. \tag{8}$$

Other physical concepts must be introduced to determine the forms of F and b. One possibility, a Markov-process hypothesis, leads to an exponential pattern for F.

Thus far, proposed solutions that can properly describe oceanic horizontal diffusion are mostly of an exponential type. They are, apart from the decay factor:

$$S_I \equiv \frac{Se^{\lambda t}}{M/D} = \frac{1}{2\pi P^2 t^2}\, e^{-(r/pt)} \qquad \text{(Joseph and Sendner, 1958)} \tag{9}$$

$$S_I = \frac{1}{\pi\omega^2 t^2}\, e^{-(r^2/\omega^2 t^2)} \qquad \text{(Okubo and Pritchard, 1960)} \tag{10}$$

$$S_I = \frac{1}{6\pi\gamma^3 t^3}\, e^{-(r^{2/3}/\gamma t)} \qquad \text{(Ozmidov, 1958)} \tag{11}$$

$$S_I = \frac{1}{\pi\beta^3 t^3}\, e^{-(r^2/\beta^3 t^3)} \qquad \text{(Obukhov, 1959)} \tag{12}$$

$$S_I = \frac{1}{\frac{3}{4}\pi^{3/2} a^3 t^3}\, e^{-(r^{4/3}/a^2 t^2)} \qquad \text{(Okubo, 1962a)} \tag{13}$$

In particular, a Fickian solution is obtained when $b = 4kt$, where k is a diffusion constant; that is,

$$S_I = \frac{1}{4\pi kt}\, e^{-(r^2/4kt)}. \tag{14}$$

As is well known, Fickian diffusion fails in describing horizontal diffusion in the sea (Stommel, 1949). The Schönfeld solution (1959) does not belong to an exponential type; it is a special case of the similarity solution, Eq. (8), when $b = \omega t$ and

$$F\left(\frac{r}{b(t)}\right) = \left(1 + \frac{r^2}{\omega^2 t^2}\right)^{-3/2} \tag{15}$$

Table 4 presents some theoretical results derived from proposed solutions. They predict that the maximum concentration decreases as either t^{-2} or t^{-3} after the effect of radioactive decay is eliminated. Dye-release experiments reveal that the exponent of t lies, in general, between -2 and -3 (Okubo, 1962a) and sometimes varies even in the course of diffusion (Pritchard *et al.*, 1966). A similar situation is found in the spatial distribution of concentration; any one of the proposed solutions is able to fit, to some extent, the observed distribution. All of the proposed solutions, except Schönfeld's, show that the variance, i.e., the mean square distances from $r = 0$, increase with time as either t^2 or t^3. The Schönfeld solution is the type of Cauchy's distribution for which no moment of positive order is finite (Cramer, 1945). An ever-increasing power of dispersion is characteristic of the horizontal diffusion due to oceanic turbulence. For a Fickian diffusion, the variance is proportional to time so that a constant coefficient of diffusion may be defined.

TABLE 4 Some Theoretical Predictions Derived from Proposed Solutions

Proposed Solution by[a]	Peak Concentration, $S_I(t,o)$	Spatial Distribution, $\ln \dfrac{S_I(t,r)}{S_I(t,r)}$	Horizontal Variance,[b] σ^2
Joseph and Sendner	$\dfrac{1}{2\pi P^2} t^{-2}$	$-\dfrac{1}{Pt} r$	$6P^2t^2$
Okubo and Pritchard	$\dfrac{1}{\pi\omega^2} t^{-2}$	$-\dfrac{1}{\omega^2t^2} r^2$	ω^2t^2
Ozmidov	$\dfrac{1}{6\pi\gamma^3} t^{-3}$	$-\dfrac{1}{\gamma t} r^{2/3}$	$60\gamma^3t^3$
Obukhov	$\dfrac{1}{\pi\beta^3} t^{-3}$	$-\dfrac{1}{\beta^3t^3} r^2$	β^3t^3
Okubo	$\dfrac{1}{3/4\ \pi^{3/2}a^3} t^{-3}$	$-\dfrac{1}{a^2t^2} r^{4/3}$	$\dfrac{4}{\sqrt{\pi}} a^3t^3$
Schönfeld	$\dfrac{1}{2\pi w^2} t^{-2}$	$\ln \dfrac{w^3t^3}{(w^2t^2+r^2)^{3/2}}$	(none existent)

[a] Refer to Okubo (1962b) for further references and details regarding these solutions.

[b] Horizontal variance is defined as the mean squared value of the horizontal distance of substance particles from the center of mass. Mathematically, it is expressed by

$$\sigma^2 = \iint_{-\infty}^{\infty} (x^2 + y^2) S_I \, dx\, dy = \int_0^{\infty} r^2 S_I(r,t)\ 2\pi r dr.$$

For oceanic diffusion, on the contrary, the variance increases faster than t^1, so that an apparent coefficient of diffusion increases with time or with the scale of diffusion, l, e.g., the size of a diffusing patch of radioactivity. Roughly speaking, as the size of the patch increases over the course of time, its diffusion will be more and more subject to the large-scale eddies, which contain more energy than the small eddies, and consequently, the rate of dispersion will be accelerated. It can be shown that the t^3 law of the variance is equivalent to the $l^{4/3}$ law of an apparent coefficient of diffusion, where l represents the scale of diffusion, say, the standard deviation associated with the distribution. The 4/3 law of diffusion was first proposed by Richardson (1926) for atmospheric diffusion, and later by Stommel (1949), Inoue (1950), Ichiye and Olson (1961), and others, for oceanic diffusion. On the other hand, the t^2 law of variance corresponds to the $l^{1.0}$ law of an apparent coefficient of diffusion. It goes without saying that the radioactive decay does not affect the variance of the concentration distribution in a patch.

The proposed solutions can also be classified into categories on the basis of a characteristic parameter involved in the solution. One group contains a "diffusion velocity" as a parameter [see solutions (9), (10), and (15)], and the others are characterized by the rate of turbulent-energy transfer (cm^2/sec^3).

The basic concept involved in the latter group of solutions is that the eddies responsible for the horizontal spread of substance lie in the "inertial subrange" (Kolmogorov, 1941). These eddies receive their energy from the larger eddies and pass it to smaller ones. No direct energy is supplied to those eddies from external sources, and the energy dissipation due to viscosity is not significant. Their properties thus depend *only* on the rate of energy transfer, which

TABLE 5 Values of Characteristic Parameters

Proposed Solution by[a]	Characteristic Parameter	Region						
		Open Sea	Off Cape Kennedy	Off Cape Kennedy	Irish Sea	Santa Monica Bay	Santa Monica Bay	Off Tokaimura
Joseph and Sendner	P (cm/sec)	1.5	0.8	–	1.1	1.0	1.0	2.0
Okubo and Pritchard	ω (cm/sec)	2.4	1.2	–	1.9	0.7	0.8	1.5
Ozmidov	γ (cm$^{2/3}$/sec)	0.7×10^{-2}	0.9×10^{-2}	1.3×10^{-2}	2.4×10^{-2}	5.9×10^{-2}	7.8×10^{-2}	8.9×10^{-2}
Obukhov	β (cm$^{2/3}$/sec)	1.2×10^{-2}	2.0×10^{-2}	2.3×10^{-2}	4.4×10^{-2}	4.1×10^{-2}	5.5×10^{-2}	6.2×10^{-2}
Okubo	α (cm$^{2/3}$/sec)	1×10^{-2}	1.7×10^{-2}	2.1×10^{-2}	3.7×10^{-2}	5.1×10^{-2}	6.8×10^{-2}	7.8×10^{-2}
Schönfeld	ω (cm/sec)	1.9	1.1	–	–	0.6	0.6	1.2
Diffusion time, t (sec) (time after release)		3.5×10^{6}	1.9×10^{5}	10^{5}	4.5×10^{4}	$1.8 \times 10^{3} \sim 1.2 \times 10^{4}$	$1.2 \times 10^{3} \sim 4.8 \times 10^{4}$	$2.4 \times 10^{3} \sim 1.1 \times 10^{4}$
Scale of diffusion l (cm) = 4σ		3.2×10^{7}	8.7×10^{5}	5×10^{5}	3.6×10^{5}	$3.5 \times 10^{3} \sim 3 \times 10^{4}$	$2.7 \times 10^{3} \sim 1.5 \times 10^{4}$	$1.3 \times 10^{4} \sim 3.1 \times 10^{4}$

[a] Refer to Okubo (1960b) for further references and details regarding these solutions.

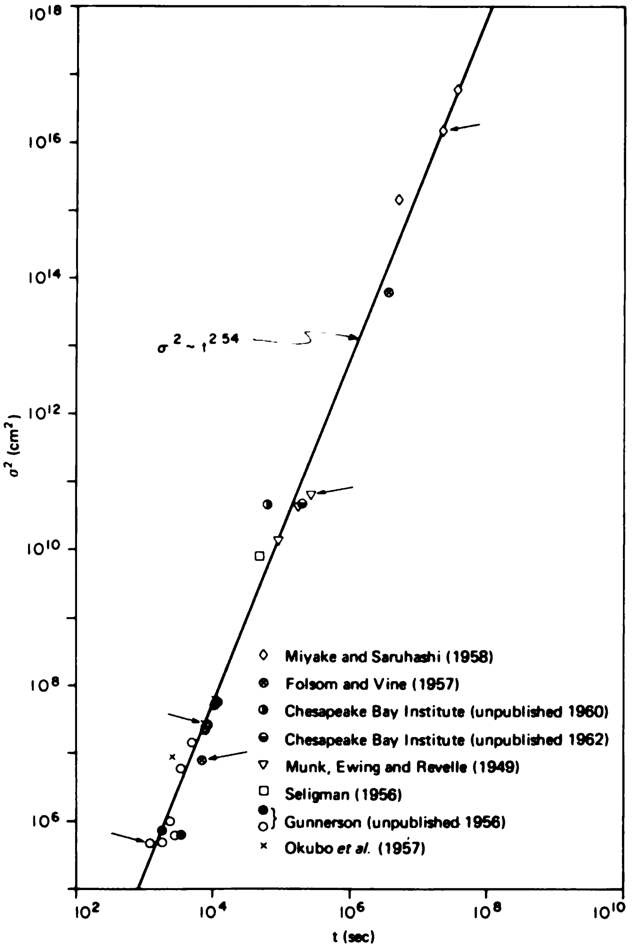

FIGURE 17 Relationship between the horizontal σ^2 and time of diffusion, t. (For details, see Okubo, 1962b.)

must be equal to the rate of energy dissipation if the energy of the eddies remains stationary. Under natural conditions, however, there is often the possibility of energy being added directly to such eddies by local storms, tidal currents, or, on a still smaller scale, by waves (Stommel, 1949; Ozmidov, 1965).* These excited eddies could not then be strictly in the "inertial subrange," so results derived from that concept would not necessarily be applicable. Ozmidov (1965), nevertheless, suggests the existence of the *local* inertial subranges separated by two scales of eddies where influx of external energy takes place. This, in turn, suggests that solutions of the Ozmidov, Obukhov, and Okubo types may be used locally with a different value of the rate of turbulent energy transfer. As the scale of diffusion increases, the expected value of this parameter decreases, since the local supplies of energy tend to be transferred from the larger eddies to the smaller eddies (for some exceptional cases see Webster, 1965). Table 5 suggests, as a matter of fact, that the estimated value of the rate of energy-transfer parameter decreases as the scale of diffusion increases. As a consequence, the results derived from these solutions would approach, to some extent, those parameterized by a "diffusion velocity."

The "diffusion velocity" model, on the other hand, is based on an intuitive concept that the rate of dispersion depends *only* on a "characteristic" velocity. Since the oceanic diffusion processes are a result of oceanic turbulence, a close connection between a "diffusion velocity" and the intensity of turbulence, i.e., the root-mean-square value of the fluctuation in velocity of the ambient water, must exist. The estimated value of the diffusion velocity is of the order of 1 cm/sec. Joseph and Sendner (1962), using a few available observations, presented a "diffusion-velocity spectrum" that indicates a weak dependence of the diffusion velocity on the scale of diffusion.

Constancy of each characteristic parameter would be desirable for the purpose of practical application of the proposed solutions to the horizontal diffusion of radioactivity from a source; actually, however, they vary with the intensity and structure of turbulence in the sea under consideration. A review paper by Bowden (1964) provides a general idea of oceanic turbulence.

In Figure 17 we plot the observed variance of the horizontal distribution from an instantaneous small source against the time since release. A linear fit to all points gives

$$\sigma^2 = 0.006 \times t^{2.5} \quad (\sigma\text{: cm; } t\text{: sec}). \qquad (16)$$

An apparent coefficient of horizontal diffusion may be defined by

$$K_a \equiv \sigma^2/4t. \qquad (17)$$

Eliminating t from Eqs. (16) and (17), we obtain an empirical relationship between the apparent coefficient of diffusion, K_a, and the scale of diffusion, l, which is arbitrarily defined as three times the standard deviation of the horizontal distribution σ.

$$K_a = 0.0087 \times l^{1.2} \quad (K_a\text{: cm}^2\text{/sec; } l\text{: cm}). \qquad (18)$$

Representative values of σ^2 and K_a for various times and scales of diffusion are tabulated in Table 6.

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*Long-term current measurements at depths of 50 and 100 m south of Bermuda were made by Day and Webster (1965). Using the techniques of spectral analysis, they obtained kinetic energy density spectra for currents. The spectra show dominant spectral peaks corresponding to the local inertial period on the 100-m depth spectrum and nearly to the local inertial period on the 50-m depth spectrum. Both spectra also show a peak at zero frequency or very large-scale eddies, and a broad low peak centered on the semidiurnal tidal period.

TABLE 6 Representative Values of the Horizontal Variance, σ^2, and an Apparent Coefficient of Diffusion, K_a

Diffusion Time, t	Horizontal Variance, σ^2 (cm^2)	Standard Deviation, σ	Scale of Diffusion, $l = 3\sigma$ (cm)	Apparent Coefficient of Diffusion, K_a (cm^2/sec)
1 hr	4.7×10^6	22 m	6.6×10^3	3.3×10^2
5 hr	2.6×10^8	160 m	4.8×10^4	3.7×10^3
10 hr	1.5×10^9	380 m	1.1×10^5	1.1×10^4
1 day	1.3×10^{10}	1.2 km	3.6×10^5	3.9×10^4
10 days	4.2×10^{12}	20 km	6.0×10^6	1.2×10^6
30 days	6.5×10^{13}	80 km	2.4×10^7	6.0×10^6
50 days	2.3×10^{14}	150 km	4.5×10^7	1.4×10^7
100 days	1.3×10^{15}	360 km	1.1×10^8	3.9×10^7
365 days	3.3×10^{16}	1,800 km	5.4×10^8	2.7×10^8

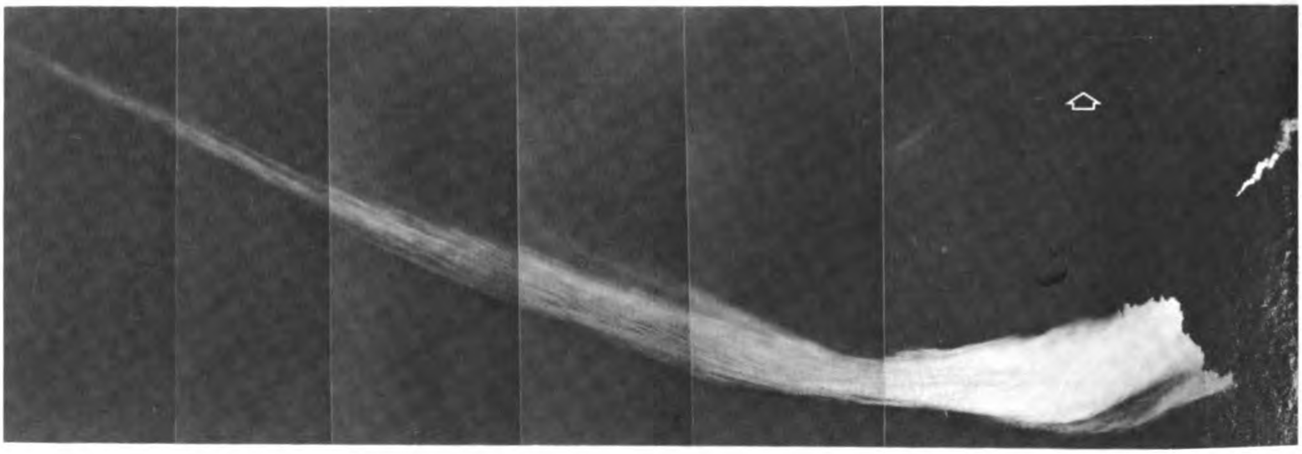

FIGURE 18 Dye patch 1 hr, 27 min after release, August 15, 1962. Wind direction is indicated by smoke bomb on sea surface, far right. Arrow points north.

SHEAR-DIFFUSION SOLUTIONS

Many aerial photographs of dye patches from an instantaneous source reveal that they are more or less elongated. Figure 18 is a photograph of one of these elongated dye patches. The dye, introduced as a vertical line source, began to elongate after about an hour and a half roughly in the direction of the local wind. The research vessel is approximately 30 m long. The leading portion ("head") of the dye patch contained a higher dye concentration than the trailing portion ("tail"). Observations revealed that the head is at or near the surface while the tail is at a lower level, probably several meters down. From the leading edge toward the tail, the dye was located at successively deeper levels. It is evident that the apparent elongation of the dye patch is a result of the vertical shear in the horizontal mean flow.

A remarkable feature of the elongation is the clockwise curvature (when looking from the head toward the tail) of the tail. In almost all observations of elongation, the curvature, when present, was clockwise in the northern hemisphere; a few observations revealed anticlockwise curvature of dye patches in the southern hemisphere (Katz *et al.*, 1965; Ichiye, 1967). This suggests that the curved tail may be due to the effect of the vertically differential advection by the wind-driven Ekman current, which is represented as a spiral on a horizontal projection. Katz *et al.* (1965) described schematically the development of the curvature by the Ekman flow (Figure 19).

The elongation of a substance patch due to the differential velocity in the mean flow gives rise to an effective diffusion when combined with transverse mixing due to small-scale random motions. Bowles *et al.* (1958), noticing the importance of this process in horizontal mixing, called it the "shear effect,"* by which they meant the distortion of a vertical column of substance due to the variation of mean

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*This effect has been recognized by fluid dynamicists since the beginning of this century. An analytical study of the shear effect on dispersion was first made by Taylor (1953, 1954) for the dispersion of solution through a tube and pipe, and also by Corrsin (1953) for an unbounded shear flow.

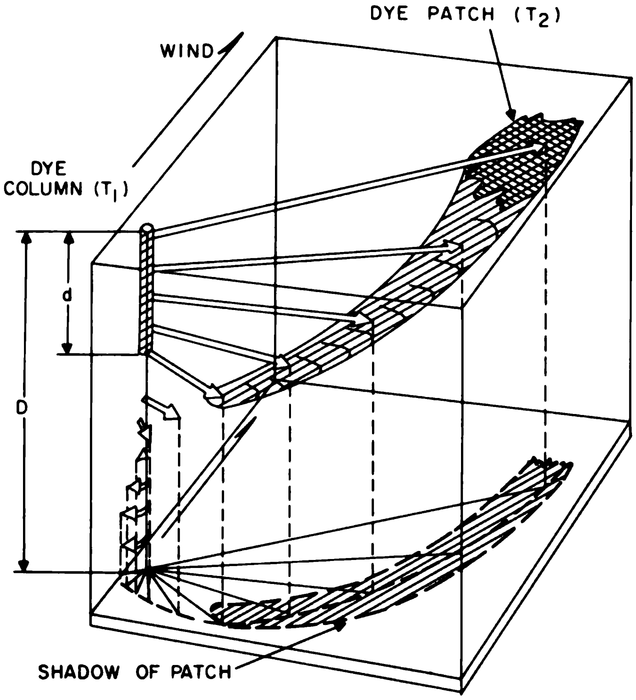

FIGURE 19 An idealized model showing the way a classical Ekman-type drift current in the northern hemisphere would distort a column of dye that initially extended vertically downward from the surface. d is the depth to which dye can be seen. D is the depth of penetration of the wind-induced current.

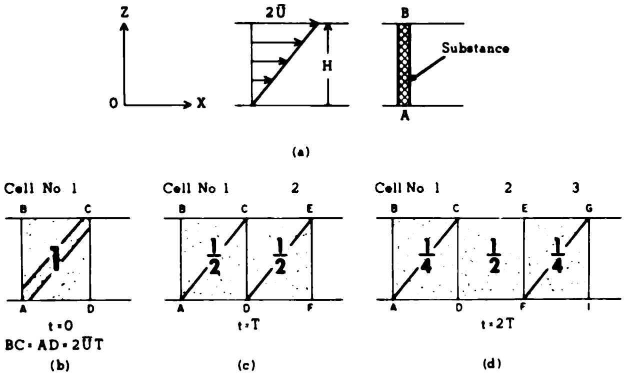

FIGURE 20 Longitudinal dispersion of a substance by the "shear effect." (a) Initial Condition; (b) t = 0; (c) t = T; (d) t = 2T.

velocity with depth combined with vertical mixing. However, any gradient of mean velocity combined with turbulent mixing leads to an effective diffusion. Thus, the shear effect (in a broad sense) may be associated with tidal currents, inertial currents, density currents, wind-driven currents, and so on.

A very simple model provides a clue to understanding the shear effect (Figure 20). Consider a shallow basin of constant depth H. The mean horizontal velocity is assumed to vary linearly with depth only; its value is $2U$ at the surface and zero at the bottom. The shear effect is simulated in such a way that for an interval of time, T, shear alone acts to distort a patch of substance; then, vertical mixing occurs and instantaneously produces a uniform concentration throughout the depth, H. The process is repeated at another interval, T, dispersing the substance in the direction of $\bar{U}$. Thus, after each interval T, a new cell of the length $2\bar{U}T$ is added to the substance patch.

It can be shown (Okubo and Carter, 1966) that after a time, $nT = t$, the distribution of substance among the cells is represented by a binomial distribution with the variance $n\,(\bar{U}T)^2 = \bar{U}^2 T\, t$. Since T is a measure of the time required for a given bounded system to attain substantial vertical homogeneity of material, T must be of the order of $H^2/\pi K_z$, where K_z is the coefficient of vertical eddy diffusion. Hence, the downstream variance is of the order of $\bar{U}^2 H^2 t/\pi^2 K_z$; a more elegant and exact derivation gives $\bar{U}^2 H^2 t/30 K_z$ (Saffman, 1962).

This simple model implies that horizontal diffusion can occur even without horizontal components of random movements of water. Furthermore, the value of the variance due to the shear-diffusion may well account for the scale of diffusion observed in shallow waters (Bowden, 1965). For example, if we take $\bar{U} = 10$ cm/sec, $H = 3$ m, and $K_z = 10$ cm^2/sec, then after $t = 10^4$ sec, the variance due to the shear diffusion amounts to 3×10^8 cm^2, or the size of patch is a few hundred meters.

The shear effect, however, appears in a different manner when the region is effectively unbounded, because the substance is allowed to diffuse indefinitely in the direction of shear, i.e., perpendicular to the mean velocity. The effect of shear on dispersion will be far more marked in an unbounded region than in a bounded one. For a uniform shear field, in fact, the longitudinal variance increases as t^3 in an unbounded region (Corrsin, 1953), whereas the variance is a linear function of time for the bounded case. For this reason, we shall first present a shear-diffusion model in an infinite sea and later consider the possible effect of boundaries.

In the radially symmetrical model, no allowance is made explicitly for factors such as shear in the mean flow. The shear-diffusion model, on the other hand, focuses attention on the actual pattern of flow that can contribute, together with the random motions of small-scale eddies, to the mixing. For simplicity, assume the mean velocity field to be sheared both laterally, Ω_y, and vertically, Ω_z. They are considered to be steady and homogeneous, that is,

$$U = U_0(t) - \Omega_y y - \Omega_z z,$$

$$V = W = 0,$$

where $U_0(t)$ is the time-dependent mean velocity in a plane $y = z = 0$. Since the time-dependent homogeneous flow has nothing to do with the spread of radioactive material with respect to the center of mass of the patch, we simply take a coordinate system whose origin moves with the center of mass of the patch with its x-axis directed with the mean flow, the y-axis lateral, and the z-axis vertical. We thus eliminate the $U_0(t)$ term completely from our discussion.

We further assume that the small-scale eddies responsible for the internal mixing can be described by eddy diffusivities A_x, A_y, and A_z in the x, y, and z directions, respectively. These are taken as constant in order to make the problem tractable.

The basic equation for the shear diffusion is then expressed by

$$\frac{\partial S}{\partial t} + (-\Omega_y y - \Omega_z z)\frac{\partial S}{\partial x} = A_x\frac{\partial^2 S}{\partial x^2} + A_y\frac{\partial^2 S}{\partial y^2} + A_z\frac{\partial^2 S}{\partial z^2} - \lambda S. \qquad (19)$$

The solution for an instantaneous point source at $x = y = z = 0$ is

$$S_I(t,x,y,z) \equiv \frac{S(t,x,y,z)}{M}e^{\lambda t}$$

$$= \frac{1}{8\pi^{3/2}(A_xA_yA_z)^{1/2}t^{3/2}\sqrt{1+\Phi_3^2t^2}}$$

$$\exp -\left\{\frac{\left[x+\frac{1}{2}t(\Omega_y y+\Omega_z z)\right]^2}{4A_xt(1+\Phi_3^2t^2)} + \frac{y^2}{4A_yt} + \frac{z^2}{4A_zt}\right\}, \qquad (20)$$

where S_I represents the concentration from a unit amount release, say, a 1-Ci source, with no decay, and Φ_3^2 is defined by $(1/12)\,[\Omega_y^2\,(A_y/A_x) + \Omega_z^2\,(A_z/A_x)]$. It is interpreted that Φ_3^{-1} represents a time at which the shears come into effect on mixing of substance.

Equation (20), having a quadratic form in x, y, and z, states that the contours of the concentration are a set of ellipsoids with common principal axes, the orientation of which varies with time. Thus, the patch of radioactive material is elongated in general. The degree of elongation depends, apart from t, on the shears and eddy diffusivities.

A long time after release, a very much elongated patch will line up in the direction of mean flow, and, at the same time, the combined effect of the shears and of the diffusion due to random eddies accelerates, to a great extent, the rate of dispersion of radioactive material around the moving center of mass. For an initial period of diffusion, however, the shear effect is hardly felt. A critical time, t_c, may be defined by Φ_3^{-1}. For the main period of shear diffusion, i.e., $t > \Phi_3^{-1}$, the following features are derived from equation (20):

maximum concentration,

$$C_pe^{\lambda t} = \frac{\sqrt{3}M}{4\pi^{3/2}(A_xA_yA_z)^{1/2}\sqrt{\Omega_y^2\dfrac{A_y}{A_x}+\Omega_z^2\dfrac{A_z}{A_x}}}\; t^{-2.5}$$

x-variance, $\sigma_x^2 = \left(\frac{2}{3}\Omega_y^2A_y + \frac{1}{6}\Omega_z^2A_z\right)t^3$

y-variance, $\sigma_y^2 = 2A_y t$

z-variance, $\sigma_z^2 = 2A_z t$

mean horizontal variance,

$$\sigma^2 \equiv \sigma_x\sigma_y = 2A_y\sqrt{\frac{1}{3}\Omega_y^2 + \frac{1}{12}\Omega_z^2\frac{A_z}{A_y}}\;t^2$$

degree of elongation on a horizontal plane,

$$\rho \equiv \frac{\sigma_y}{\sigma_x} = \sqrt{\frac{1}{\frac{1}{3}\Omega_y^2 + \frac{1}{12}\Omega_z^2\frac{A_z}{A_y}}}\;t^{-1}$$

apparent coefficient of horizontal diffusion,

$$K_e \equiv \frac{\sigma^2}{2t} = A_y\sqrt{\frac{1}{3}\Omega_y^2 + \frac{1}{12}\Omega_z^2\frac{A_z}{A_y}}\;t$$

Equation (20) describes three-dimensional diffusion under shears in an infinite ocean. For a release in the surface layer or near the bottom of the sea, we must take into consideration the (reflective) boundary effect at least at the sea surface ("naviface") or at the bottom; a lateral boundary such as a coastline may be treated in a similar way. Without the shears, or for the radially symmetrical model, it would be easy to treat the boundary effect simply by adding the image of the fundamental solution for infinite space with respect to the boundary (Carslaw and Jaeger, 1959). With shear we cannot use the image method in such a simple way, as might be expected.

However, the essential features of the shear diffusion would remain much the same for infinite and quasi-infinite seas, except for the fact that the distribution in the vicinity of the boundary becomes asymmetrical in that the position of the maximum concentration shifts toward the downstream direction. Saffman (1962) shows, as a matter of fact, that the longitudinal variance for the semibounded case differs only by a certain numerical factor from that for the infinite case. Obviously, the presence of the boundary does not affect the lateral and vertical variances. These results also indicate that the time behavior of the peak concentration would be unchanged.

In shallow waters the presence of both the sea surface and the bottom must be considered after an appreciable amount of the substance has diffused from the surface to the bottom. Then the shear diffusion due to vertical shear becomes decelerated and will be overwhelmed by the shear diffusion due to lateral shear. In other words, the depth-mean concentration can be discussed by a two-dimensional shear-diffusion model with the effective value of the longitudinal coefficient due to vertical shear (Bowden, 1965) added linearly to the eddy diffusivity, A_x.

The solution for the two-dimensional diffusion in a horizontal shear can be easily obtained by putting $\Omega_z = 0$ in Equation (20) and taking an average over the depth, D. We then have

$$S_I(t,x,y) = \frac{\bar{S}(t,x,y)}{M e^{-\lambda t}}$$

$$= \frac{1}{4\pi D (A'_x A_y)^{1/2}\, t\sqrt{1 + \Phi_2^2 t^2}}$$

$$\exp - \left[\frac{x^2 + t\Omega_y x y + \frac{A'_x}{A_y}\left(1 + \frac{1}{3}\Omega_y^2 \frac{A_y}{A_x} t^2\right) y^2}{4A'_x\, t\,(1 + \Phi_2^2)} \right], \qquad (21)$$

where $\Phi_2 \equiv \sqrt{1/12}\ \Omega_y \sqrt{A_y/A_x}$, D denotes the mixing depth over which the substance is distributed uniformly, and A'_x is the sum of A_x and the effective longitudinal coefficient due to vertical shear. Φ_2^{-1} is interpreted as a time at which the horizontal shear begins to come into effect on the mixing of substance.

The behavior of the characteristics of two-dimensional shear diffusion may be summarized as follows for $t > \Phi_2^{-1}$:

$$C_p e^{\lambda t} = \frac{\sqrt{3} M}{2\pi D A_y \Omega_y} t^{-2} \qquad \sigma^2 = \tfrac{2}{3} A_y \Omega_y t^2$$

$$\sigma_x^2 = \tfrac{2}{3} A_y \Omega_y^2 t^3 \qquad \rho = \frac{\sqrt{3}}{\Omega_y} t^{-1}$$

$$\sigma_y^2 = 2 A_y t \qquad K_e = \frac{1}{\sqrt{3}} A_y \Omega_y t$$

Essential features of the shear diffusion are the same for the three-dimensional and two-dimensional cases. The faster rate of decrease of the peak concentration in the three-dimensional case is simply a consequence of the fact that the substance has one more dimension to diffuse.

Carter and Okubo (1965) analyzed dye-release experiments in the Cape Kennedy area purely on the basis of shear-diffusion models. Offshore releases, being regarded as three-dimensional cases, exhibited the two time regimes, $t^{-1.5}$ and $t^{-2.5}$, in the decrease of the maximum concentration (Figure 21). On the other hand, near-shore and inshore releases in shallow waters showed the two time regimes, $t^{-1.0}$ and $t^{-2.0}$, in the decrease of the peak concentration. For both the deep- and shallow-water cases, the horizontal variance increased in direct proportion to the second power of time. Representative values for the shears and eddy diffusivities in the offshore region of the Cape Kennedy area are estimated as follows:

	Winter	Summer
$A_x \approx A_y$	4×10^3 cm²/sec	4×10^3 cm²/sec
A_z	19 cm²/sec	1.3 cm²/sec
Ω_z	1.8×10^{-3} sec^{-1}	6.6×10^{-3} sec^{-1}
Ω_y	$<10^{-4}$ sec^{-1}	$<10^{-4}$ sec^{-1}
t_c	4 hr	8 hr

As may be seen, there is an interesting resemblance between this simple shear-diffusion model and a "diffusion velocity" type of solution of the radially symmetrical model. Both models give the same dispersion rate: the horizontal variance increases as the second power of the time. As a matter of fact, a combined parameter, say, $\sqrt{\Omega_y A_y}$, which appears in the expression for σ^2 in the shear-diffusion model, has the dimensions of velocity. Using the values of shear and eddy diffusivities shown above, we obtain a value for the combined parameter on the order of 1 cm/sec, which is comparable to the value of a diffusion velocity estimated in the radially symmetrical model. This implies that the shear-diffusion model describes more explicitly the physical processes of horizontal diffusion from a source than the radially symmetrical model does.

CONTINUOUS RELEASE FROM A FIXED POINT

As mentioned earlier, the solution for an instantaneous source is not necessarily fundamental to the construction of the solution for a continuous source. However, there are no

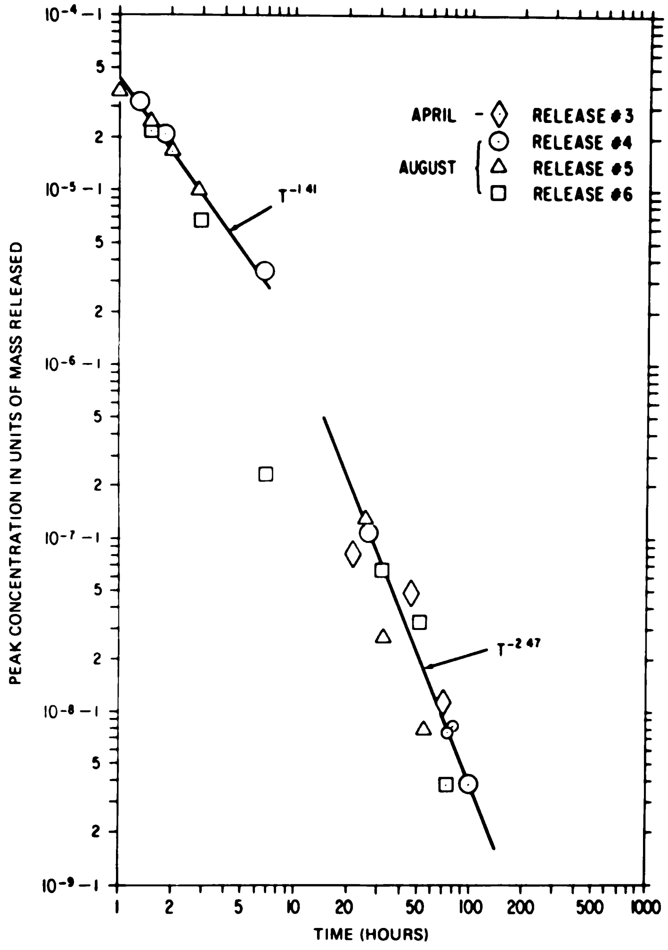

FIGURE 21 Variation of peak concentration with time for dye-release experiments in the sea off Cape Kennedy. (See Carter and Okubo, 1965.)

appropriate differential equations to be used as a basis for oceanic diffusion from a continuous source. It still seems appropriate, however, to consider plume-dispersion models, such as that of Frenkiel (1953) with an eye toward practical applications.

Plume-dispersion models in widespread use employ instantaneous patches as elementary components. The ideal plume model is assembled by superposition of an infinity of overlapping instantaneous patches, each released from a fixed origin and each translated by the mean velocity, U. Such a superposed-patch model is illustrated in Figure 22a. Thus, the concentration of radioactivity from a continuous source may be formulated as follows:

$$C(t,x,y,z) = \int_0^t q(t-t')\,S_I(t',x-Ut',y,z)\,e^{-\lambda t'}\,dt', \tag{22}$$

where $C(t,x,y,z)$ denotes the concentration of radioactivity at a point (x,y,z) and at a time t, elapsed since the start of releases; $q(t)$ is the rate of release at the source in curies per second; and S_I represents the concentration distribution for an instantaneous release of a unit amount without decay.

Any one of the solutions for an instantaneous release may be used for S_I. Only a few limited cases, however, permit one to carry out the integral in Eq. (22) analytically. For mathematical convenience, therefore, diffusion in the direction of the mean flow is neglected, leading to the "spreading-disk" model for plumes, as illustrated in Figure 22b. Thus, the formula for C in the disk model becomes

$$C(t,x,y,z)$$

$$= \int_0^t q(t-t') \int_{-\infty}^{\infty} S_I(t',x'-Ut',y,z)\,dx'\,e^{-\lambda t'}\delta\,(x-Ut')\,dt'$$

$$= q\left(t-\frac{x}{U}\right)\frac{1}{U}\int_{-\infty}^{\infty} S_I\left(\frac{x}{U},x'-x,y,z\right)dx'\,e^{\frac{-\lambda x}{U}} \text{ for } x \leqq Ut$$

and 0 for $x > Ut$. (23)

To illustrate, let us take the instantaneous solution (Eq. 10). The depth mean concentration from a continuous source is given by

$$\bar{C}(t,x,y) = \frac{q\left(t-\dfrac{x}{U}\right)}{\sqrt{\pi}\,\omega D x} \exp -\left(\frac{U^2y^2}{\omega^2x^2}\right) \exp -\left(\frac{\lambda x}{U}\right) \text{ for } x \leqq Ut$$

$$\text{and 0 for } x > Ut. \qquad (24)$$

That is, the centerline concentration in the plume from a constant rate of release without decay decreases inversely with the distance from the source.

Similarly, the instantaneous solution (Eq. 12) results in the following expression for the depth-mean concentration:

$$\bar{C}(t,x,y) = \frac{q\left(t-\dfrac{x}{U}\right)}{\sqrt{\pi}\,\beta^{3/2} D\, U \left(\dfrac{x}{U}\right)^{3/2}} \exp -\left(\frac{U^3y^2}{\beta^3x^3}\right) \exp -\left(\frac{\lambda x}{U}\right) \text{ for } x \leqq Ut$$

$$\text{and 0 for } x > Ut. \qquad (25)$$

Here, the plume concentration along the central line decreases in proportion to $x^{-3/2}$ when the rate of release is constant and the decay is not included.

The three-dimensional shear-diffusion solution (Eq. 20), when applied to the centerline concentration of a constant release plume, also gives rise to an x^{-1} law.

Real plumes present a far more complicated appearance (Figure 22c) than does the plume of the theoretical model. The motion of the individual elementary patches consists of two parts: spreading within individual patches and meandering of the center of mass of the element. The meandering is caused by the motion of the large-scale eddies. However, if we focus our attention on a meandering plume at a given instant and position a coordinate system with respect to the meandering plume, i.e., the x-axis along the centerline of the plume, the concentration distribution in the plume can be represented by the ideal plume discussed above.

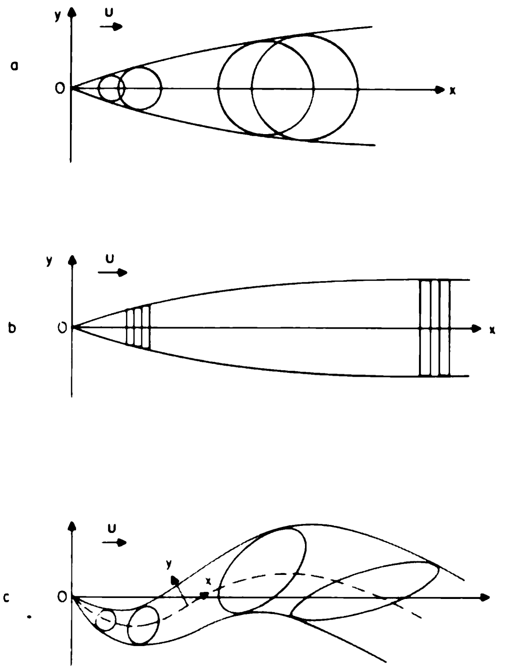

FIGURE 22 Schematic plan views of plumes: (a) ideal (theoretical) plume; (b) spreading-disk plume; (c) fluctuating single plume. (See Gifford, 1959.)

On the other hand, the mean concentration at a fixed point taken over a certain period longer than the time scale of the meandering will be smaller than the concentration in the ideal plume. According to Gifford (1959), a long-term mean concentration at a fixed point may be expressed as follows:

$$[C(t,x,y,z)] = \int_{-\infty}^{\infty} C(t,x,y-y_o,z)\, g(x,y_o)\, dy_o, \qquad (26)$$

where $g(x,y_o)$ represents the frequency function associated with the meandering of the central line of a plume about the x-axis, i.e., the direction of the overall mean velocity, U_m.

Thus far, very few experimental and theoretical results concerning the frequency function appear to have been reported. If it is assumed that the frequency function is Gaussian with the variance $\sigma_g^2(x)$, Eq. (26) can be integrated, at least in principle, for appropriate solutions for the ideal plume, C. Thus, the depth-mean solution (24) for C combined with a Gaussian form of g gives (Gifford, 1959):

$$[\bar{C}(x,y)] = \frac{q_o}{\sqrt{\pi}\,D\,(\omega^2x^2 + 2U_m^2\,\sigma_g^2)^{1/2}}$$

$$\exp -\left[\frac{y^2}{2\left(\dfrac{\omega^2}{2U_m}\,x^2 + \sigma_g^2\right)}\right] \cdot \exp -\left[\frac{\lambda x}{U_m}\right] \qquad (27)$$

where the rate of release is assumed to be a constant value, q_o. Csanady (1963) showed experimentally that the standard deviation, σ_g, increases linearly with the distance from the source. This implies that the overall mean concentration along the x-axis, without radioactive decay, still decreases inversely with the distance, with an effective diffusion velocity larger than that of the ideal plume.

Another important aspect of continuous releases must be mentioned. That is, from the standpoint of a safety estimate of radioactivity, not only the concentration in a steady-state portion of the plume but also the distribution of radioactivity in the transient-state portion, e.g., the advancing front of the plume, must be considered. The "disk" model is too crude to be used for this purpose, because the longitudinal diffusion of individual elementary patches primarily determines the concentration distribution in the frontal part of the plume. We must return to the general solution (23), substitute an appropriate instantaneous solution into S_I, and perform the integration.

Let us consider a tractable case, in which the duration time of release is very short in comparison with the half-life of the radioactive isotope in question. Then, using solution (10) for S_I, we obtain the concentration from a constant rate of supply as follows:

$$C(t,x,y) \simeq \frac{q_o}{2\sqrt{\pi}\,\omega D\,(x^2+y^2)^{1/2}}\, erfc\left[\frac{U}{\omega}\left(\frac{\sqrt{x^2+y^2}}{x_r}-1\right)\right], \quad (28)$$

where

$$erfc\,\eta \equiv \frac{2}{\sqrt{\pi}}\int_{\eta}^{\infty} e^{-a^2}\,da, \text{ and } x_r \equiv Ut.$$

In particular, the centerline concentration becomes

$$C(t,x,o) \simeq \frac{q_o}{2\sqrt{\pi}\,\omega D x}\, erfc\left[\frac{U}{\omega}\left(\frac{x}{x_r}-1\right)\right],$$

$$\text{for } x \geqq 0. \quad (29)$$

At the point $x = x_r$, the concentration is half what it would be if the source had been emitting for an infinite time. Beyond this point, the decrease in concentration is very sharp, depending upon the ratio of the values of the mean velocity and of a diffusion velocity.

Figure 23 illustrates schematically how the centerline concentration varies with the distance for a transient plume from a continuous source.

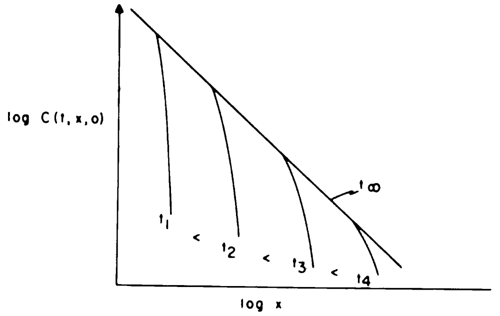

FIGURE 23 Schematic variation of the centerline concentration with distance, for a transient plume from a continuous source.

Equation (29) appears to satisfy some of the observed features of the oscillating plume in a tidal estuary (see Carter, 1965).

FINITE SOURCES

Thus far, we have regarded the size of the source as infinitesimal. This means that the discussed solutions cannot be applied to a certain initial period of patch diffusion or in the vicinity of a continuous source, since the source is actually finite. A measure of the critical time or distance beyond which a point-source solution is practical is discussed below.

First, consider an instantaneous release. Let σ_o^2 be the variance of the initial distribution from a finite-sized source. Then, the critical time, t_f, is defined as the time at which $\sigma^2(t_f) = \sigma_o^2$, where $\sigma^2(t_f)$ denotes the variance from an instantaneous point-source.

For practical purposes, the finite-source problem may be treated by the method of superposition of infinitesimal sources, even though this method is not correct in principle. Thus, the depth-mean concentration from a uniform source of rectangular shape, extending from $-a/2$ to $a/2$ in the x direction and from $-b/2$ to $b/2$ in the y direction, can be expressed as follows:

$$S_f(t,x,y) = \frac{m}{abD}\int_{-a/2}^{a/2} dx_o \int_{-b/2}^{b/2} dy_o\, S_I(t, x-x_o, y-y_o)\, e^{-\lambda t}, \quad (30)$$

where m denotes the amount of radioactivity released per unit area.

Similarly, continuous releases of radioactivity from a finite source can be treated by superposition. Thus, beyond the distance $x_f \equiv Ut_f$ from the source, we may, for practical purposes, disregard the size of the source; there the distribution of radioactivity would be nearly identical to that at a point source with the same total intensity.

OCEANOGRAPHIC IMPLICATIONS OF THE DISTRIBUTION OF NATURAL TRACERS AND FALLOUT ISOTOPES

The development of radioactive tracing and dating techniques in the last decade has provided oceanographers with a new tool for studying large-scale (oceanic) physical processes in the sea, such as diffusion and advection. We may hope that as tracer measurement techniques improve, our understanding of the worldwide distributions of these radioactive tracers will also improve, thus permitting us to focus our attention on smaller scale processes. As our understanding of the processes of advection and diffusion at all scales improves, so too will our ability to predict the capacity of our environment to receive radioactive wastes, from fallout and other sources. Accordingly, this section reviews the radiochemistry literature that applies to the oceans. Since most of the papers reviewed analyze the data by means of "box models," it seems appropriate to start with a general description and discussion of these models.

Box Models

Radiochemists are generally credited with developing the box model concept, although similar models were used in the early stages of the study of physical oceanography. Radiochemistry data is analyzed by this means probably because such data are sparse, and an oversimplified picture of the ocean is required in order to draw any conclusions at all. In this approach, the ocean is divided into a number of "boxes," or reservoirs, each containing water that is assumed to be well mixed. The entire system is generally assumed to be in a steady state, and the exchange of fluid-attached properties, i.e., salt, heat, stable carbon, ^{14}C, etc., is assumed to take place in accordance with first-order kinetics—that is, the transfer of matter follows an exponential exchange law (the flux of the property per unit area per unit time is proportional to the concentration of the property) as long as the internal mixing rate in each reservoir is large in comparison to the transfer rate out of the reservoir. The transfer processes are unspecified, although one physical interpretation may be arrived at as shown in Figure 24.

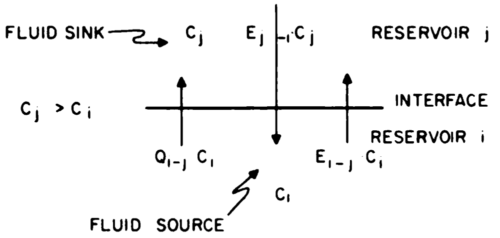

FIGURE 24 Schematic presentation of the transfer processes between two reservoirs.

Consider the interface between two liquid reservoirs such as those shown in Figure 24. Here, there is an advective flux of property $Q_{i-j} \cdot C_i$ from reservoir *i* to reservoir *j*, and a net diffusive flux $E_{i-j} \cdot C_i - E_{j-i} \cdot C_j$, in the opposite direction. It is assumed that $C_j > C_i$ and that continuity of mass is maintained by a fluid source in reservoir *i* and a fluid sink in reservoir *j* at unspecified points in these reservoirs. At steady state, we have at the interface

$$Q_{i-j} \cdot C_i + E_{i-j} \cdot C_i - E_{j-i} \cdot C_j = 0. \tag{31}$$

Equation (31) may be rewritten as follows:

$$\frac{Q_{i-j}}{V_i} \cdot N_i + \frac{E_{i-j}}{V_i} \cdot N_i - \frac{E_{j-i}}{V_j} \cdot N_j = 0, \tag{32}$$

where $\frac{N_i}{V_i} = C_i$.

Finally, from Eq. (32), we obtain

$$\left\{k_{i-j}^{ad} + k_{i-j}^{diff}\right\} N_i - k_{j-i} \cdot N_j = 0, \tag{33}$$

where

$$\frac{Q_{i-j}}{V_i} \equiv k_{i-j}^{ad} \equiv \frac{1}{\tau_{i-j}^{ad}} \qquad \text{(advection)} \tag{34a}$$

$$\frac{E_{i-j}}{V_i} \equiv k_{i-j}^{diff} \equiv \frac{1}{\tau_{i-j}^{diff}} \qquad \text{(diffusion)} \tag{34b}$$

$$\frac{E_{j-i}}{V_j} \equiv k_{j-i} \equiv \frac{1}{\tau_{j-i}} \qquad \text{(diffusion)} \tag{34c}$$

and where τ_{i-j} is the *residence time*, or "average" life, of a molecule of the species being considered in the reservoir *i* relative to the reservoir *j*. τ_{i-j} is identical with the τ defined in Craig (1957) as the quotient of the steady state contents of a reservoir and the flux.

An expression that is completely equivalent for the same exchange at the interface and for the form that is usually used in box models is

$$k_{i-j} \cdot N_i - k_{j-i} \cdot N_j = 0. \qquad (35)$$

Thus, one possible interpretation of residence times arrived at by use of equations similar to Eq. (35) is that the transfer coefficient, k_{j-i}, is some measure of the diffusion and k_{i-j} is some measure of the combined effects of diffusion and advection, since k_{i-j} corresponds to $k_{i-j}^{ad} + k_{i-j}^{diff}$ in Eq. (33).

Another important conclusion concerning box models of the chain* type may be drawn from Eqs. (32) through (35). A chain model does not permit a net flux of water molecules across the interface except for the evaporation-precipitation difference at the air-sea interface. Such a model is shown in Figure 25. The water balance requires that $E_{D-M} = E_{M-D}$ at steady state, and from Eqs. (34b) and (34c) we have,

$$\frac{k_{D-M}}{k_{M-D}} = \frac{V_M}{V_D} \qquad (36)$$

or

$$\frac{\tau_{D-M}}{\tau_{M-D}} = \frac{V_D}{V_M}. \qquad (37)$$

These relationships indicate that the residence times determined by this model will depend quite critically upon the mixed-layer depth that is selected. Models of this type were used by Craig (1957, 1963), Broecker (1966), and Plesset and Dugas (1967).

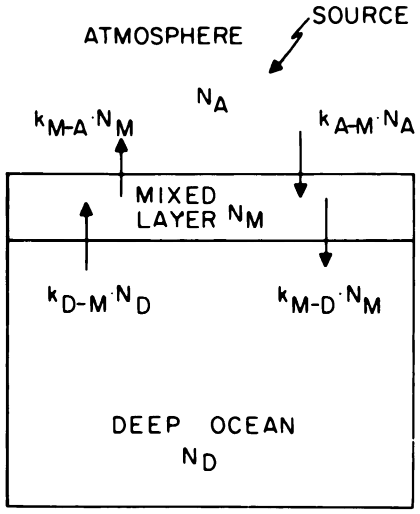

FIGURE 25 A box model of the ocean–atmosphere system of the chain type.

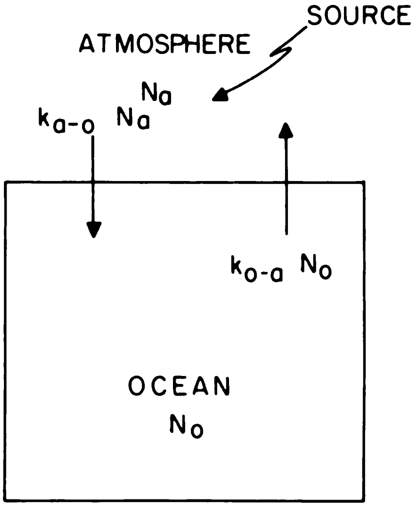

FIGURE 26 A simple box model of the ocean–atmosphere system.

Still another consideration is the strong interdependence between the model selected and the relationship between residence time and radiocarbon age. Attention has already been called to this by Craig (1963). In Figure 26, the residence time of the ocean reservoir is very nearly infinity, since water can only be exchanged between the atmosphere and the oceans by evaporation and precipitation. That is, if the ocean were completely mixed by some means, the resulting "age" would refer only to the residence time of the radiocarbon molecules in the ocean relative to those in the atmosphere, and not to the residence times of the water molecules. In the situation represented in Figure 25, the radiocarbon and water molecules in the deep ocean can be exchanged only with molecules in the mixed layer; therefore, if such an ocean could somehow be created, the residence times of the deep ocean water and of the radiocarbon molecules would be identical. If one now ventilates a portion of the deep water by exposure to the atmosphere, such as occurs at high latitudes, the radiocarbon age of the deep water will be reduced, since the radiocarbon molecules in the deep sea can now be exchanged with those of both the mixed water just above and the atmosphere. The residence time of the water molecules is, however, unchanged. It is

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*In a chain model, the reservoirs are arranged like beads on a string. If the deep ocean reservoir depicted in Figure 25 is ventilated by exposure to the atmosphere, the model would become a "cyclic" model with respect to ^{14}C, but would remain a "chain" model with respect to water molecules.

apparent from the foregoing discussion that the residence times of interest to the oceanographer are the residence times of the various water reservoirs relative to each other, such as a deep layer relative to a mixed layer or an intermediate layer relative to an overlying surface layer and an underlying deep layer. Any model that does not provide accurate estimates of the residence times of the water molecules in the various subdivisions of the real ocean is of limited value in interpreting circulation patterns and mixing processes.

At least three box models and one analytical model based on radioactive isotope data seem to meet the foregoing criteria. They are, in chronological order, the model of Broecker *et al.* (1961), the model of Bolin and Stommel (1961) for the Common Water* and Atlantic Intermediate Water, Broecker's (1966) transient-state model for predicting ^{90}Sr and ^{137}Cr vertical distributions resulting from bomb fallout and his steady-state model for the vertical distribution of ^{14}C, and Munk's (1966) analytical model for the deep Pacific water. Both the Bolin and Stommel model and the Munk model utilize, and are therefore consistent with, temperature and salinity distributions. These models are presented in Table 7. For comparison, even though they do not utilize radioactive isotope data, Montgomery's (1959) results for the residence times of subtropical surface water, based on his salinity model, are included.

Table 7 summarizes what seem to us to be the important inferences to be drawn from the various models. It is quite clear that the average residence time of a water molecule in the deep Pacific and Indian oceans is about 1,000–1,300 years. Surface water in all oceans is 10 years old on the average, and Atlantic Antarctic Intermediate Water is 100–400 years old. As used in this paragraph, "old" is synonomous with residence time relative to all routes of removal. From continuity considerations, it is easy to deduce estimates of vertical velocities at the interface between the deep water and the water above it (interface depth approximately 1,000 m). These vertical velocity estimates should be regarded as oceanic averages. Their order of magnitude is $0.3\text{-}1.4 \times 10^{-5}$ cm/sec. It is not possible to separate in an unambiguous manner the effects of deep horizontal advection and vertical mixing. The range of horizontal velocities for the northward velocity of the deep Pacific waters were arrived at by ignoring vertical diffusion for the lower limit and including its effect for the upper limit. Broecker's model (1966) is not consistent with what we believe to be the important processes in the formation of the thermocline, as he points out. It does, however, permit a comparison between mixing rates inferred from ^{14}C data, on one hand, and ^{90}Sr and ^{137}Cs data, on the other.

Comments about Some Conflicting Oceanographic Implications from Observations of the Vertical Distribution of ^{90}Sr and ^{137}Cs

A number of investigators, notably Miyake *et al.* (1962), Bowen and Sugihara (1958, 1960, 1965), and Rocco and Broecker (1963), have measured ^{90}Sr and ^{137}Cs concentrations at the surface and at various depths in the ocean and have interpreted the concentration distributions they observed. Some of these oceanographic interpretations conflict with each other, and some apparently conflict with the descriptions of physical processes in the oceans presented in earlier parts of this chapter.

Miyake *et al.* described measurements of ^{90}Sr and ^{137}Cs made in the western North Pacific Ocean in 1958, 1959, and 1960. They found significant amounts of radioactivity at depths as great as 5,000 m. They ascribed this deep penetration of radionuclides to vertical eddy diffusion from a source in the surface layers, and they computed a vertical diffusivity from their observed depth distribution for the depth interval 1,000 to 4,000 m. Their value of 200 cm^2/sec for vertical diffusivity far exceeds the values for the deep waters of the Pacific obtained by Munk (1966) as well as the values that can be deduced from the analysis of Bolin and Stommel (1961). Munk obtains estimates of K_z, the vertical diffusivity, using distributions of temperature, salinity, and ^{14}C, and shows that the value he computes of about 1.3 cm^2/sec is consistent with the distributions of radium and dissolved oxygen. Munk also showed that the associated value of the vertical velocity (1.2 cm/day), which is intimately tied to the value of K_z in his model, is consistent with the probable rate of production of the Pacific Deep Water. As Munk points out, the values he obtains for the vertical diffusivity and for the vertical velocity are consistent with the results obtained by Stommel and his collaborators in their manifold attack on the abyssal circulation (Stommel, 1957; Robinson and Stommel, 1959; Stommel and Arons, 1960a and b; Bolin and Stommel, 1961), and by Wooster and Volkman (1960), and Knauss (1962).

We have no reason to question the values for the concentrations of ^{90}Sr and ^{137}Cs determined by Miyake *et al.*; however, we find that we must reject their hypothesis regarding the source of the deep penetration of these radionuclides. Miyake's measurements were made following several series of weapons tests in the Pacific, involving detonations that could have produced close-in contamination to considerable depths. It is also significant that the very extensive but unpublished measurements of ^{137}Cs made throughout the North Pacific by Folsom (personal communication) during 1965, 1966, and 1967 do not show significant amounts of this isotope below a depth of about 400 m, even though these measurements were made after the accumulated fallout had more than doubled over that

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*This is the deep water of the Indian and Pacific oceans, the most voluminous water mass of the world. It was named by Montgomery (1958).

TABLE 7 Summary of the Residence Times, τ, the Vertical Coefficients of Eddy Diffusivity, K_z, the Vertical Velocities, w, and the Horizontal Velocities, v, Inferred from the Various Models

Model	Water Mass	Exchanges with	τ (yr)^{a}	K_z (cm^2/sec)	$w \times 10^5$ (cm/sec)	v (cm/sec)
Broeker *et al.* (1961)	Arctic	Atlantic Deep Water	45	–	–	–
	North Atlantic Surface Water	Arctic	10	–	–	–
	South Atlantic Surface Water	North Atlantic Surface Water	10	–	–	–
	North Atlantic Deep Water	Arctic, Antarctic	600	–	–	–
	Antarctic	North Atlantic Deep Water, Pacific and Indian Deep Water	100	–	–	–
	Pacific and Indian Surface Water	Antarctic	25	–	–	–
	Pacific and Indian Deep Water	Antarctic	1,300	–	–	–
Bolin and Stommel (1961)	Pacific and Indian Common Water	North Atlantic Deep Water, Antarctic Bottom Water, Pacific and Indian Intermediate Water	1,200	–	0.6	–
Bolin and Stommel (1961)	Atlantic Antarctic Intermediate Water	Antarctic Surface Water, Atlantic Deep Water, Atlantic Surface Water	100–400	–	0.3–1.0	–
Broecker (1966)	Caribbean (upper 750 m)	Layer below	Sr^{90} 4.8^{b}	1.5	9.5^{c}	–
	North Atlantic (upper 750 m)	Layer below	Cs^{137} 1.6^{b}	4.5	28.5^{c}	–
	South Pacific (upper 750 m)	Layer below	16^{b}	0.45	3^{c}	–
	Pacific–Indian Ocean main thermocline	Layer below	C^{14} 19	0.4	2.5	–
	North Atlantic	Layer below	5	1.5	9.5	–
Munk (1966)	Pacific (all > 1 km)	Vertical advection and diffusion with upper 1 km	1,000^{d}	1.3	1.4	0.07–1.0 (north)
Montgomery (1959)	North Pacific Surface (<100 m)	High-latitude Surface Water, Low-latitude Surface Water, and Deep Water	2	1.6^{e}	–	–
	South Pacific (<230 m)	High-latitude Surface Water, Low-latitude Surface Water, and Deep Water	12	1.4^{e}	–	–
	North Atlantic (<375 m)	High-latitude Surface Water, Low-latitude Surface Water, and Deep Water	23	1.9^{e}	–	–

TABLE 7 (Continued)

Model	Water Mass	Exchanges with	τ, (yr)[a]	K_z, (cm^2/sec)	$w \times 10^5$, (cm/sec)	ν, (cm/sec)
	South Atlantic (<190 m)	High-latitude Surface Water, Low-latitude Surface Water, and Deep Water	9	1.3[e]	–	–
	South Indian (<170 m)	High-latitude Surface Water, Low-latitude Surface Water, and Deep Water	6	1.5[e]	–	–
	Average (<213 m)	High-latitude Surface Water, Low-latutide Surface Water, and Deep Water	10	1.4[e]	–	–

[a] Residence time relates to exchange by all routes permitted by the model.
[b] Residence time in each 150-m layer computed from d/R where d is the layer depth, 150 m, and R is the inter-reservoir mixing rate $[L/T]$.
[c] R, the inter-reservoir mixing rate.
[d] Computed from w (mass of Common Water/surface area of Pacific); $M = 6 \times 10^{23}$g; $s = 1.37 \times 10^{18}$cm^2.
[e] Estimated from d^2/τ.

which existed at the time at which Miyake *et al.* obtained their measurements.

Of more concern are differences in the observed vertical distributions of ^{90}Sr and ^{137}Cs obtained by Broecker and his associates, on the one hand, and by Bowen and Sugihara, on the other. Broecker argues that his observations in the North Atlantic do not show ^{137}Cs radioactivity exceeding 2 dpm/100 liters and ^{90}Sr radioactivity exceeding 1 dpm/100 liters below a depth of about 500 m. Broecker's measurements in the equatorial Pacific are lower than these levels for ^{137}Cs and ^{90}Sr for all but a few samples below a depth of about 300 m. The latest published measurements by Broecker and his associates are for 1963.

Published measurements of ^{90}Sr radioactivity by Bowen and Sugihara were for the most part obtained in the Atlantic Ocean between 16°N and 16°S during the period 1957 through 1961. These measurements show significant ^{90}Sr radioactivity at depths as great as 2,500 m and also show secondary peaks at intermediate depths.

Measurements made by both groups are reasonably consistent for the near-surface layers. Both sets of measurements show a maximum of radioactivity at the surface and a fairly rapid decrease to a depth of about 500 m. Below this depth Broecker and his associates show ^{90}Sr radioactivity, or ^{137}Cs radioactivity converted to ^{90}Sr equivalents, of less than 1 dpm/100 liters (with a few exceptions, discussed below), while Bowen and Sugihara show ^{90}Sr radioactivity at depths between 500 and 2,500 m ranging between 2.8 and 4.5 dpm/100 liters. The vertical profile is irregular, showing intermediate secondary maxima.

Bowen and Sugihara have argued that their observations are consistent with what is known about the large-scale circulation. They do not consider that the ^{90}Sr radioactivity they find below about 500-m depth penetrates downward through the thermocline at middle and low latitudes. They argue instead that the source regions for deep and intermediate water masses in the North Atlantic occur in regions of relatively high fallout. The sinking of water at these sources and the subsequent southward movement of the intermediate and deep water masses along surfaces of constant potential density could bring radioactive materials to the intermediate and deep layers at middle and low latitudes.

The general concept proposed by Bowen and Sugihara is certainly a valid one. Some North Atlantic Intermediate Water appears to be formed in a region centered at about 50°N, and the source region for North Atlantic Deep Water is centered at about 60°N. Based on land measurements, fallout reaches peak concentrations just south of 50°N, and is still about 55 percent of the maximum at 60°N. The speeds in the core of the intensified western deep boundary current, which carries the North Atlantic Deep Water southward, are thought to be sufficient to carry water to the equator from the source region in about two years. Enroute, however, this flow feeds the deep water volume of the entire North Atlantic. Evidence from analysis of natural tracers indicates that the mean residence time for the Deep Water in the North Atlantic is at least 300 years, and for the entire North Atlantic Deep Water, including its penetration through the South Atlantic, the residence time has been shown to be 600 to 800 years. Any tracer added to the surface source waters for the North Atlantic Deep Water in the last decade should be considerably diluted at middle and low latitudes at this time, even though the minimum transit time to the equator would be great enough to permit some radionuclides to reach the equatorial Atlantic at depths of 2,000 to 3,000 m within 2 years after injection at the surface source.

The velocities associated with the intermediate waters formed in the North Atlantic are not well known, and existing evidence indicates that this water mass is not formed in very great amounts. Certainly, the North Atlantic Intermediate Water is much less important as a supplier of intermediate-depth water than the Antarctic Intermediate Water. However, fallout at the southernmost latitude of the source of Antarctic Intermediate Water is very small compared to fallout in the northern hemisphere. Thus, radionuclides injected into the source waters of the North Atlantic Intermediate Water within the last decade should be highly diluted at intermediate depths in the middle and low latitudes of the Atlantic.

The problem we face in explaining the relatively high ^{90}Sr radioactivity observed by Bowen, therefore, is not that there are no mechanisms that would ultimately bring radionuclides originally injected into the surface layers as fallout to depths between 500 and 2,500 m at middle and low latitudes, but rather that the radioactivity observed appears to be much higher than would be expected from known residence times for the intermediate and deep water. According to existing concepts, the older resident water simply represents too large a diluting volume for the recently initiated source of radioactivity.

A recent compilation of all ^{90}Sr and ^{137}Cs measurements made to date, now being prepared for publication by the New York Operations Office of the Atomic Energy Commission, indicates that observations of the deep penetration of these isotopes are not restricted to Bowen and Sugihara, but occur in the data of Broecker and his associates as well as of others. Even in their published data, Broecker *et al.* (1966) show one analysis of a sample collected at a depth of 4,330 m in the North Atlantic (34°49'N, 70°49'W) that has a ^{90}Sr radioactivity of 4.3 ± 1.8 dpm/100 liters and a ^{137}Cs radioactivity of 7.0 ± 1.2 dpm/100 liters. Thus, it appears that physical oceanographers must examine the evidence from fallout studies in greater detail than they have to date. It is quite possible that existing concepts must be modified to account for the relatively rapid deep penetration of these isotopes in the Atlantic.

Using a technique quite different from that of Broecker and his associates, Folsom (personal communication) has made extensive measurements of ^{137}Cs in the North Pacific. This technique involves the *in situ* extraction of the ^{137}Cs onto KCFC resin, and relative large volumes of water can be processed rapidly. Most of Folsom's measurements were made during 1965, 1966, and 1967, and they constitute a larger number of ^{137}Cs measurements than has been made by any other single source. These measurements agree with those reported by Broecker *et al.* (1966) for the equatorial Pacific in that Folsom's observations also show statistically significant radioactivity confined above 300 m.

Folsom's measurements show one feature not revealed by the observations of other investigators. Folsom finds very large peaks of ^{137}Cs radioactivity occurring in thin, subsurface lenses within the seasonal thermocline. *In situ* recording salinometers also show thin lenses of anomolous salinity maxima and minima in this same depth range.

Fission explosions produce ^{137}Cs and ^{90}Sr at a relatively fixed ratio of between 1.6 and 1.8. These isotopes are thought to be transported together and to exist in seawater in completely dissolved form without being subjected to any significant biological or geochemical transport. Consequently, it is possible to convert, within reasonable limits, observations of one isotope to equivalent concentrations of the other. Folsom's rather extensive sampling of the surface waters of the North Pacific for ^{137}Cs can therefore be compared to a collection of all available surface analyses of ^{90}Sr in the Atlantic Ocean that have been compiled, but not yet published, by Vaughn Bowen (personal communication). This comparison shows that for the same latitudinal bands, the amounts of ^{90}Sr and ^{137}Cs radioactivity in the North Pacific are at least twice the corresponding amounts at the surface of the North Atlantic. This difference cannot be explained as resulting from the greater amount of close-in fallout in the Pacific. Of approximately 200 megatons of fission materials produced in weapons tests to date, some 167 megatons, or approximately 84 percent, was injected into the stratosphere and would therefore return to the earth's surface as worldwide fallout. We are forced to conclude that the fallout nuclides are mixed in the surface layers of the North Atlantic to a depth about twice that to which such nuclides are mixed in the North Pacific. This finding is consistent with our existing knowledge of the vertical density structure in the upper layers of the two oceans.

The surface data supplied by Vaughan Bowen clearly show the surface mixing to be seasonal. We have averaged the surface ^{90}Sr radioactivity in the North Atlantic by latitudinal bands and by 6-month periods for the years 1961 through 1966. The two 6-month periods chosen for averaging were December through May (which we here designate as winter) and June through November (which we here designate as summer). Figure 27 shows the winter and summer averages for the three latitudinal bands of 30–40°N, 40–50°N, and 50–60°N. To illustrate our point, individual curves have been drawn through the average of the winter points for the three latitudinal bands, and through the average of the summer points. Both sets of curves reach a maximum in 1963–1964. The winter ^{90}Sr radioactivity observations are, however, about 30 percent lower than those in the summer, indicating that mixing occurs to greater depths in the winter.

Studies of residence times of natural tracers in the vari-

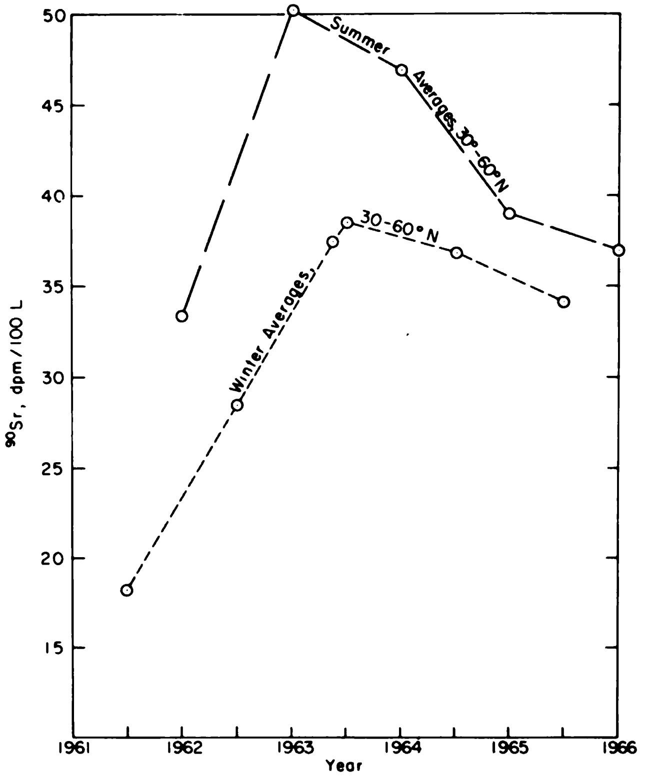

FIGURE 27 Average of surface ^{90}Sr activity in the North Atlantic by 6-month intervals. Summer and winter averages shown separately.

ous intermediate and deep water masses may help resolve the problem of interpreting the Bowen and Sugihara observations of the vertical distribution of ^{90}Sr. E. Schuert, of the U.S. Naval Radiological Defense Laboratory (personal communication), has made careful comparisons of two sampling and analysis procedures for the determination of ^{137}Cs in the ocean. Schuert took samples at three stations in the eastern North Pacific, two stations in January 1967 at 32°N, 130°W and 32°N, 123°W, and the third station in June 1967 at 34°N, 134°W. Two series of measurements were made at each station. In one series, 1,000 liters of water were pumped *in situ* through a device containing KCFC resin to extract the ^{137}Cs. In the second series, water samples were collected using the large-volume water sampler designed by Bodman *et al.* (1961), which is also the sampler used by Bowen and Sugihara since mid-1958. The KCFC resin used in the first series at each station was counted for ^{137}Cs activity before use, and this reading was subtracted from the activity determined after passing the seawater through the resin. In the second series, the seawater samples were treated chemically to concentrate and isolate the ^{137}Cs.

For each type of sampling, the three stations showed insignificant differences. Both techniques gave surface ^{137}Cs activities at all three stations of 200 to 210 dpm/100 liters. The radioactivity decreased sharply with depth. At 340 m, the KCFC technique showed ^{137}Cs activity of 0.8 ± 0.5 dpm/100 liter, and below 400 m, no statistically significant ^{137}Cs activity was found on the KCFC resin. The water samples collected using the Bowen and Sugihara technique, with subsequent chemical treatment before analysis, showed that the ^{137}Cs activity also reached essentially zero at about 400 m. However, a secondary maximum of 12 dpm/100 liters was observed at about 500 m, and another of about 5 dpm/100 liters at about 800 m.

It has generally been assumed that both ^{90}Sr and ^{137}Cs exist in seawater in completely dissolved form, and that no particulate transport is involved in the distribution of these isotopes. Thus, any method that satisfactorily extracts dissolved ^{90}Sr and ^{137}Cs should be suitable for the determination of the distribution of these isotopes. Presumably, the KCFC resin removes only the dissolved ^{137}Cs. Both the water and its contained suspended materials are treated chemically. One possible explanation for the differences in the vertical distributions observed by Schuert using the two methods of sampling and analysis is that cesium exists in seawater in two forms. Though the amount in true solution dominates, some small fraction may exist in particulate form. The KCFC method did not show any ^{137}Cs in solution below a depth of 400 m. The water sampling method, on the other hand, showed low but significant radioactivity at several depths below 500 m, which may have represented ^{137}Cs associated with suspended particles that had reached these depths by gravitational sinking.

One further piece of evidence may be cited. In a paper presented at the meeting of the American Chemical Society on October 3-7, 1966, D. E. Robertson and R. W. Perkins, of Battelle Memorial Institute, Pacific Northwest Laboratory, described a series of ^{137}Cs analyses made in the North Pacific. Their data represented the average of 10 vertical profiles taken along 39°N between 134°15′ and 134°59′W. This average profile is shown in Figure 28; the ^{137}Cs activity is expressed in pCi/100 liters. Note that at 270 m the ^{137}Cs activity decreased from a near-surface maximum to levels below the detection limit. This profile is relatively smooth compared to those observed by Folsom (personal communication). The data is consistent with the measurements made using the KCFC method in that no detectable concentrations were found below approximately 300 m. We have integrated this curve vertically and found that the accumulated ^{137}Cs corresponds to a total fallout of 85 ± 18 mCi/km^2. This may be compared to a value of 115 mCi/km^2 estimated for this latitude from land measurements. The difference (30 mCi/km^2) may result from any of the following: (a) the fallout over this area of the Pacific may not be the same as the average over land for this latitude; (b) the counting uncertainty was quite high (this could account for a significant portion of the difference; note that the upper limit of the range of standard error would give an integrated value of 103 mCi/km^2); (c) ^{137}Cs may exist below the 270-m depth at concentrations just below the limit of detection and through a sufficient depth range to contribute the 30 mCi/km^2 difference to the depth integral; (d) ^{137}Cs may exist both in the upper layers and below the 270-m level in the form not measured by the technique employed.

The difference of 30 mCi/km^2 is regarded as relatively small, and of opposite sign, compared to that reported by Bowen and Sugihara (1965). These investigators found that the depth-integrated profiles of ^{90}Sr for the equatorial Atlantic showed amounts eight times higher than those expected from fallout, determined from land measurements, in this latitudinal belt.

In the next section, some recent measurements of the vertical distribution of bomb-produced ^{14}C in the northeast Pacific Ocean are discussed. These measurements also support the contention that in the Pacific, bomb-produced radionuclides that are primarily waterbound (i.e., in true solution) have not penetrated in measurable amounts below a depth of 300 m.

Estimates of Vertical Velocity and Vertical Diffusivity through the Thermocline Based on the Temporal Change in the Vertical Distribution of Carbon-14 Produced by Weapons Tests

The use of the distributions of ^{14}C and stable carbon in box models to estimate residence times of the various subdivisions of the oceans, as described earlier in this chapter, involved the use of measurements made prior to large weapons tests and subsequent correction to post-weapons-test values in order to remove the effect of the added flux of bomb-produced ^{14}C from the data. The large input of artificially produced ^{14}C began to make a significant imprint on the vertical profile of ^{14}C concentrations sometime after 1959 and was particularly evident in data from the northwest Pacific Ocean by 1965. The observed temporal change in the vertical distribution of ^{14}C, particularly the progressive downward penetration of the new input, can be used to provide additional estimates of some of the physical advective and diffusive parameters.

Figure 29 shows observed vertical profiles of ^{14}C concentration taken in the northeast Pacific Ocean in October 1959, in June 1965, and in October 1966. Some additional observations made in August 1964 below a depth of 200 m are also shown in the figure. The curve for October 10, 1959, is drawn through observations made by G. S. Bien, Scripps Institution of Oceanography, at 30°04′N, 118°02′W. (Note that the range of uncertainty in the measurements is indicated for each observed point.) Observations also made by G. S. Bien at 49°00′N, 154°58′W, in August 1964 at depths of 210 m, 500 m, and 1,000 m indicate remarkable uniformity in time and space. Evidently, the additional flux of bomb-produced ^{14}C had not penetrated to 210 m by August 1964.

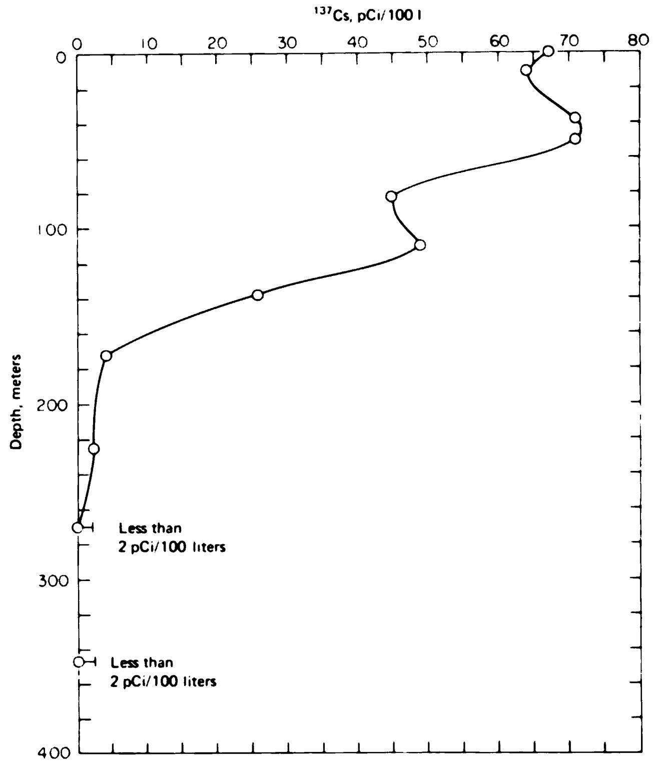

FIGURE 28 Cesium-137 in pCi/100 liters; average of 10 vertical profiles taken along 39°N, between 134° 15′W and 134°59′W, by Robertson and Perkins, February 12, 1966.

The curve for June 30, 1965, is drawn through observations made by A. W. Fairhall, University of Washington, at 48°29′N, 133°09′W. This curve shows that between October 1959 and June 1965 the influx through the surface of bomb-produced ^{14}C had significantly increased the ^{14}C concentrations above about 200 m. This curve joins the curve drawn through the observations made in October 1959 at a depth of about 240 m. The curve for October 18, 1966, was drawn through observations also made by A. W. Fairhall, at 47°00′N, 132°03′W. This curve shows the further increase in the near-surface ^{14}C concentration resulting from the increase of flux through the surface and also shows penetration to a greater depth. This curve joins that of October 1959 at a depth of 300 m.

For the region below 300 m, a single curve has been drawn, since for the most part the observations made in the several locations and at the several times do not show statistically significant differences at any depths.

The original data were given as δ^{14}C values, expressed as per-mille differences from a reference standard, and showed both positive and negative differences from the standard. For convenience in the numerical treatment of the data described below, we have here introduced an arbitrary zero

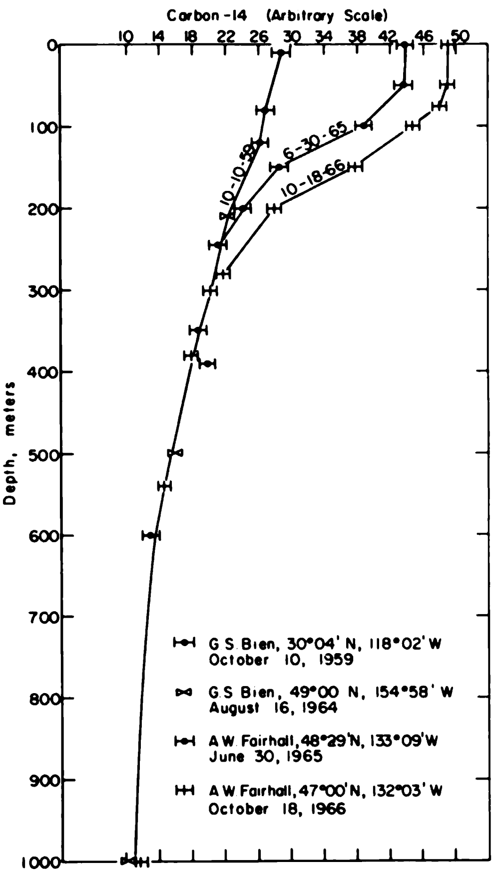

FIGURE 29 Carbon-14 concentration (arbitrary units) with depth, northeast Pacific Ocean.

shift so that all ^{14}C "concentrations" appear as positive numbers. As stated above, $\delta\,^{14}C$ is expressed as a per-mille difference from a standard; however, in the following treatment of these data, the absolute values of the concentration ultimately cancel out of the problem. Hence, for convenience in keeping track of units, we have here considered that the zero-shifted per-mille values for $\delta\,^{14}C$ represent a concentration of ^{14}C in arbitrary units per cubic meter, expressed as F/m^3 (F being an undefined unit of mass).

Assume that in the region of the ocean in which these ^{14}C data were obtained, and for the time scale being considered, the vertical variations in concentration, and particularly the time changes in the vertical distribution, are controlled by the physical processes of vertical flux. Specifically, for the time period represented by the 1965 and 1966 data, it is assumed that data relating to the horizontal advective and diffusive processes are not important in determining the vertical variation in concentration.

Under these assumptions, we have, for a given time and depth,

$$\frac{\partial \zeta}{\partial t} = -\frac{\partial (w\zeta)}{\partial z} + \frac{\partial}{\partial z}\left(K_z \frac{\partial \zeta}{\partial z}\right), \qquad (38)$$

where

ζ = concentration of ^{14}C
z = vertical coordinate, positive downwards
w = vertical velocity
K_z = vertical diffusivity

If this equation is integrated from the surface to the depth z, we have

$$\int_0^z \frac{\partial \zeta}{\partial t}\,dz' = -(w\zeta)\Big]_0^z + \left(K_z \frac{\partial \zeta}{\partial z}\right)\Big]_0^z . \qquad (39)$$

The surface limits on the right-hand side of this equation represent simply the flux of ^{14}C through the surface, here designated by q_o. Thus, we have

$$\int_0^z \frac{\partial \zeta}{\partial t}\,dz' = q_o + \left(K_z \frac{\partial \zeta}{\partial z}\right)_z - (w\zeta)_z \qquad (40)$$

The left-hand side of this equation can be approximated by

$$\int_0^z \frac{\partial \zeta}{\partial t}\,dz' = \frac{1}{\Delta t}\int_0^z (\zeta_2 - \zeta_1)\,dz', \qquad (41)$$

where ζ_1 represents the ^{14}C concentration from the June 30, 1965, profile, and ζ_2 represents the ^{14}C concentration from the October 18, 1966, profile. Then, $\Delta t = t_2 - t_1 = 475$ days, or 4.11×10^{11} sec. Equation (40) then becomes, on rearranging the order of terms,

$$(w\zeta)_z \cdot \Delta t - \left(K_z \frac{\partial \zeta}{\partial z}\right)_z \cdot \Delta t = q_o\,\Delta t - \int_0^z (\zeta_2 - \zeta_1)\,dz'. \qquad (42)$$

The term $q_o\,\Delta t$ represents the total flux through the surface during the time interval Δt. It is evident from the vertical profiles of ^{14}C concentration for 1959, 1965, and 1966 that the added bomb-derived flux across the surface considerably exceeded the flux of natural radiocarbon. It is also evident that the bomb-produced ^{14}C added to the surface layers of the ocean had not penetrated below 300 m in this area of the northeast Pacific as of October 1966. Therefore,

TABLE 8 Values of the Vertical Velocity, w, and the Vertical Coefficient of Eddy Diffusivity, K_z, Computed Using the Vertical Profiles of ^{14}C Concentration Shown in Figure 29 (Values of the pertinent parameters entering the equations also given)[a]

Depth (m)	$\bar{\zeta}$ (F/m^3)	$-\Delta \frac{\partial \bar{\zeta}}{\partial z}$, ($10^6 F/m^4/sec$)	$\Delta t \bar{\zeta}$, ($10^9 F/m^3/sec$)	$q_o \Delta t - \sum_o^z (\zeta_2-\zeta_1)\Delta z$ (F/m^2)	$-w$ (10^{-7}m/sec)	$-\omega\Delta\bar{\zeta}$, (F/m^2)	K_z (cm^2/sec)
0	46.4	–	1.91	1,542	0	0	–
20	46.35	0.10	1.91	1,437	0.3	53	149
40	46.3	0.77	1.90	1,330	0.6	107	18.6
60	45.6	2.37	1.87	1,217	0.8	157	5.8
80	44.0	4.02	1.81	1,094	1.1	203	3.2
100	41.7	5.87	1.72	971	1.4	240	3.1
150	32.7	7.06	1.34	606	2.1	272	1.2
200	25.9	4.12	1.06	262	2.1	224	1.2
250	22.4	2.11	0.92	73	2.1	193	1.2
300	20.3	1.44	0.84	0	2.1	175	1.2
400	17.7	0.97	0.73	0	2.1	153	1.6
500	15.5	0.90	0.64	0	2.1	134	1.5
600	13.5	0.62	0.56	0	2.1	117	1.9
700	12.5	0.29	0.51	0	2.1	107	3.7
800	11.9	0.25	0.49	0	2.1	103	4.1
1,000	11.0	0.16	0.45	0	2.1	95	6.0

[a] For symbols, assumptions, and method of computation, see text.

$q_o \Delta t$ is given approximately by the net change in accumulated ^{14}C above a depth of 300 m—that is,

$$q_o \Delta t = \int_0^{300} (\zeta_2 - \zeta_1)\, dz. \tag{43}$$

Note that, on the time scale considered here, the radioactive decay of ^{14}C, which would contribute a term to the equation having opposite sign to the flux of naturally produced ^{14}C, has also been neglected.

The vertical velocity, w, will be zero at the surface and would be expected to increase in magnitude with depth. Below 300 m, the right-hand side of Eq. (42) will be zero. Since the concentration decreases with depth, the balance required by Eq. (42) gives an upward-directed vertical velocity, at least to a depth of 1,000 m. Since the horizontal motion that must carry the adcending water back to regions of sinking is concentrated most strongly in the upper 200 m, we assume for our immediate purpose that the vertical velocity is constant from a depth of 1,000 m up to a depth of 150 m, and then decreases in magnitude to zero at the surface.

The vertical density gradient in this region is approximately constant through the depth range of 160 to 300 m, and the vertical diffusivity might also be expected to be constant through the same depth range. Making this assumption, data derived from the profiles of ^{14}C concentration given in Figure 29 can be used in Eq. (42) to find the vertical velocity, w, and the diffusivity, K_z. The procedure involved inserting into Eq. (43) the values of $(\zeta \Delta t)$, $\left(\frac{\partial \zeta}{\partial z}\Delta t\right)$, and $q_o \Delta t - \int_0^z (\zeta_2 - \zeta_1)\, dz'$, corresponding to 160-m depth and to 300-m depth. The resulting two simultaneous equations are then solved for w and K_z. Values of $(\zeta_2 - \zeta_1)$ were obtained directly from the profiles for June 30, 1965, and October 18, 1966, while values of ζ and $\partial\zeta/\partial z$, which enter the advective and diffusive flux terms, were determined from a slightly smoothed profile representing the average ζ_1 and ζ_2.

The values of vertical velocity and diffusivity thus obtained were $w = -2.1 \times 10^{-5}$ cm/sec (i.e., directed upward) and $K_z = 1.22$ cm^2/sec. Taking w to be constant at this value throughout depth interval of 150 m to 1,000 m and then to decrease linearly in magnitude to zero at the surface, Eq. (42) can be used to obtain values for K_z at all depths from just below the surface to 1,000 m. Table 8 shows the values of the pertinent parameters obtained from the profiles of ^{14}C concentrations given in Figure 29, and the computed values of K_z. This table shows that the computed values of K_z are essentially constant over the depth interval 160 to 300 m, supporting the assumptions that allowed the equation to be solved for both w and K_z.

The high value, of 149 cm^2/sec, for the vertical diffusivith at a depth of 20 m, shown in Table 8, indicates the rapid mixing above 50 m within the surface layer in this area of the oceans. The computed magnitudes of the vertical velocity, of approximately 2×10^{-5} cm/sec, and of K_z in the thermocline, of approximately 1 cm^2/sec, are consistent with a relatively long residence time for the waters below 1,000 m in the Pacific Ocean. Though K_z increases slowly with depth below 400 m, reaching a value of 6 cm^2/sec at 1,000 m, the computations here do not support the work of Miyake *et al.* (1962); they are more in line with those of Munk (1966), as discussed in the previous section of this chapter.

NEAR-SHORE AND ESTUARINE ENVIRONMENTS

Man's most intimate contact with the marine environment occurs near the coast and in estuaries and other embayments, and he makes the most intensive use of this segment of the environment for the harvest of seafood and nonliving resources and for waste disposal. Consequently, it is to be expected that some radioactive materials from man's peaceful uses of nuclear energy will enter the near-shore and estuarine waters; it is necessary to ensure that man's use of these waters and their products will not be limited by the introduction of these radioactive materials.

Knowledge of the physical processes of movement and turbulent diffusion in estuaries has grown significantly in the last 15 or 20 years. The effort expended in study of the near-shore waters along the open ocean has, however, been relatively small. The region from just outside the surf zone to 20 or so miles offshore remains poorly understood, although the use of this near-shore environment as a receiver of man's unwanted waste materials is likely to increase greatly in the near future.

Two reports have been published by the National Academy of Sciences-National Research Council dealing with the disposal of low-level radioactive waste in coastal waters. These reports were written by working groups of the Committee on Oceanography and for the most part treat the subject of the disposal of packaged waste into designated disposal areas. The first of these, ***Radioactive Waste Disposal into Atlantic and Gulf Coastal Waters*** (National Academy of Sciences-National Research Council, 1959), was prepared by a working group under the chairmanship of Dayton E. Carritt. The second, ***Disposal of Low-Level Radioactive Wastes into Pacific Coastal Waters*** (National Academy of Sciences-National Research Council, 1962), was prepared by a working group under the chairmanship of John D. Isaacs. Despite the emphasis on packaged-waste disposal, a problem of little current concern, these reports still contain considerable information about the physical and biological character of coastal waters that can be used in considering the probable environmental consequences of other possible sources of radioactive materials in these waters.

The term *near-shore zone* is used here to designate the strip of water adjacent to the shoreline in which the adjacent land boundary, freshwater runoff from the land, and local meteorological conditions contribute significantly to the physical processes of movement and mixing. The width of this zone is highly variable, and the outer boundary is not well defined. The near-shore zone may be quite narrow along coasts where the continental shelf is narrow and where oceanic conditions in which the motion of the water is dominated by the "permanent" currents, discussed earlier in this chapter, penetrate close to shore. Conversely, off a coast where the continental shelf is quite wide, the near-shore zone may be several tens of miles wide. The near-shore zone is not, however, coincident with the continental shelf. Where the shelf is very narrow, the near-shore zone may extend beyond the shelf, and where the shelf is very wide, the near-shore zone may extend out from shore for only a fraction of the shelf width. The critical features defining this zone are the dominance of boundary effects and of transient local effects on the water movement.

There is considerable temporal and spatial variation in the movement of the waters in the near-shore zone. Except near inlets, the tidal component of the motion is generally parallel to the coast and oscillatory in character, with the onshore–offshore component increasing with distance from the coast. Inshore from the point where surface waves are breaking—that is, in the surf zone—a longshore current usually develops in a direction dependent upon the angle at which the waves approach the shoreline. Thus, along a coastline oriented in a north–south direction, with the ocean to the east, waves approaching the shoreline from the northeast will produce a longshore current that flows southward. Outside the surf zone, the local wind and the salinity gradient, which is related to runoff from the land, combine to dominate the nontidal current in the near-shore zone. Since the waves breaking on a shoreline at any given time may have been for the most part generated by storm systems some distance away, the direction from which the waves approach the coast and the local wind direction are generally not correlated. Consequently, the direction of flow inside the surf zone may be opposite to the direction of the currents seaward of the surf zone.

Along coasts having relatively large freshwater inflows to the ocean, such as the Atlantic coast of the United States, the salinity, and hence the density, of the water increases with distance offshore. The average net nontidal flow in the near-shore zone will, along such coasts in the northern hemisphere, be directed such that the shoreline is to the right of an observer looking downstream. Such currents are particularly well developed along coasts to the right (looking seaward) of the mouths of major estuaries–southward from the mouth of the Chesapeake Bay, for example.

At any particular time, however, the currents in the near-shore zone may be dominated by the local wind or by density-induced effects resulting from the wind. In the shallow waters characteristic of the near-shore zone, the direct, wind-induced transport of the near-surface waters is directed slightly to the right of the wind. Hence, an offshore wind will transport the surface waters offshore, particularly a wind blowing at an angle to the shoreline such that an observer with his back to the wind has the shoreline on his left hand. If this occurs at a time of the year when there is a vertical density gradient with warmer waters at the surface, these warmer waters are transported offshore and the cooler waters from offshore are transported along the bottom and well up along the shoreline. The resulting density distribution will produce, in the northern hemisphere, a current flowing along the coastline with the shore to the left of the current (looking downstream).

When such an offshore or longshore wind ceases, the warmer surface layers slosh back toward the shore. Dye-tracer studies along open coastlines have shown that in this way pollutants introduced from an offshore outfall are most likely to be carried onto the beach following the cessation of a wind that has produced an offshore surface-layer transport. In the presence of an onshore wind, on the other hand, a slope zone develops near the shore and the wind-induced transport turns and runs parallel to the shoreline at a point just outside the breaker zone.

Radioactive materials introduced into the near-shore zone will be transported away from the point of discharge by the current present in the vicinity of the discharge at the time of discharge, and will be mixed into the receiving waters by turbulent diffusion. Where there is continuous discharge of radioactive materials into the near-shore zone, a diverging plume extending downcurrent from the source will develop, with concentration levels in the plume decreasing with distance from the source. Because of the transient character of the current pattern in the near-shore zone, the plume will show temporal variations in direction, sometimes folding back on itself, but more often, when the current direction approximately reverses, the remnants of the older, lower concentration plume will be advected along a path somewhat offset from the path of the new plume extending from the source in the new direction of the current. Over a long period of time, a general low-level concentration field will be built up in the vicinity of the source, upon which the transient, higher concentration plume is superimposed. The steady-state level of this general background concentration will depend upon the rate at which the waters of the near-shore zone are exchanged with waters of the adjacent outer shelf or oceanic region.

We have but meager knowledge of these exchange processes by which the waters of the near-shore are renewed. Where the inflow of fresh water from the land into the near-shore zone is sufficient to produce measurable salinity gradients, and where the rates of freshwater inflow can be determined, the mean residence time for the water of the near-shore zone can be estimated. Such an estimate was made by Ketchum and Keen (1955) for the waters of the entire continental shelf along the Atlantic coast of the United States between Cape Hatteras and Cape Cod.

Assuming that the source of the water that exchanges with the waters over this segment of the continental shelf is the "slope water," which is formed between the Gulf Stream and the continental shelf, these investigators used the observed salinity distribution to compute the total volume of river water resident over the shelf. Dividing this accumulation of water derived from land drainage by the annual volume rate of flow from all rivers discharging from the adjacent coast gave a mean residence time for this segment of the continental shelf of about 1½ years. Conceivably, this approach could be modified to apply to any segment of the near-shore zone along this coastline or any other coastline where similar conditions of freshwater inflow prevail.

Along coastlines where freshwater drainage from land is too small to develop measurable salinity variations, the use of some other tracer, either natural or artificial, will be required in order to determine the exchange rates between the waters of the near-shore zone and waters of the adjacent outer continental shelf or open ocean.

As stated earlier, estuary studies during about the last 15 years have been rather extensive, and a considerable volume of literature has been published concerning the distribution of properties and the circulation patterns in the various types of estuaries. Cameron and Pritchard (1963) contributed a review of this literature, which also contains a representative list of references concerned with estuaries through about 1960. The book entitled *Estuaries* (Lauff, 1967), published by the American Association for the Advancement of Science in 1967, refers to several more recent papers dealing with the kinematics and dynamics of estuaries. Other references that treat studies of this subject include those of Rattray and Hansen (1962), Hansen and Rattray (1965, 1966), and Pritchard (1955).

An estuary has been defined (Pritchard, 1967a) as a semi-enclosed coastal body of water that has a free connection with the open sea and within which seawater is measurably diluted with fresh water derived from land drainage. From a geomorphological standpoint, four primary subdivisions of estuaries are recognized: drowned river valleys, fjord-type estuaries, bar-built estuaries, and estuaries produced by tectonic processes.

Drowned river valleys are the classical estuaries of the physical geographer. Because they are generally confined to coastlines with relatively wide coastal plains, these waterways have also been called coastal-plain estuaries. These estuaries are widespread throughout the world, and are common along the eastern seaboard of the United States. This class of estuaries has also been studied the most, and most of the existing literature about estuaries deals with them.

Coastal-plain estuaries may be further subdivided into four principal types (Pritchard, 1955, 1967b), differing from one another in the character of the nontidal circulation pattern, in the intensity of vertical stratification, and in the extent of lateral homogeneity. These types are the salt-wedge estuary, typified by the mouth of the Mississippi River; the partially mixed estuary, typified by the Chesapeake Bay and its tributary estuaries; the vertically homogeneous estuary; and the sectionally homogeneous estuary.

Most estuaries fall into the category of partially mixed estuaries, and further discussion here will therefore be limited to this type. This discussion of the movement and mixing in partially mixed coastal-plain estuaries covers the processes of importance in estuaries in general and makes adequate reference to the literature dealing with other categories of estuaries.

In a partially mixed estuary, the salinity increases with depth as well as in the seaward direction. There is usually a surface layer in which the vertical salinity gradient is small, an intermediate layer in which the salinity increases relatively rapidly with depth, and a deep layer in which the rate of increase of salinity with depth is small, as in the surface layer. While the intermediate layer is one of relatively high stability, the density gradient is not sufficiently steep to completely inhibit vertical mixing between the surface layer and the bottom layer.

The oscillatory ebb and flood of the tide are the predominant motions in the estuary. Superimposed on the tidal currents is a net circulation pattern in which there is a net seaward flow in the surface layers and a net flow from the mouth toward the head of the estuary in the deeper layers. There is also a small net vertical motion from the deeper layers to the surface layers. The volume of water flowing toward the head of the estuary per unit time decreases from the mouth to the head of the estuary, since water is simultaneously being transferred through vertical motion from the deeper layers to the surface layers. Hence, the volume rate of seaward flow in the surface layers increases from the head toward the mouth of the estuary.

A pollutant initially introduced into the bottom layers in a partially mixed coastal-plain estuary, in addition to participating in the oscillatory movement of the tidal currents, is carried in the net motion toward the head of the estuary. At the same time, turbulent mixing leads to horizontal dispersion in the longitudinal and lateral directions and to vertical dispersion into the surface layers. Pollutant that becomes mixed with the surface layers is carried in the net flow toward the mouth. Seaward from the point of introduction, pollutant being carried toward the ocean in the surface layers is partially mixed downward into the deeper layer and reintroduced into layers moving toward the head of the estuary.

A pollutant introduced into the surface layers is initially carried in the net flow toward the mouth of the estuary. Mixing leads to horizontal and vertical spreading, and the wastes are thus added to the deeper layers, which have a net flow toward the head of the estuary.

In the region of the estuary headward from the point of introduction, the concentrations of pollutant will always be greater in the deeper layers than in the surface layers, while seaward from the point of introduction the converse will be true. These conditions prevail regardless of whether the wastes are initially introduced into the surface layers or into the deeper layers.

The pollutant is ultimately flushed from the estuary in the seaward flow of the surface layers.

In order to understand these various processes in greater detail, consider a waste effluent introduced via an outfall into the estuary. The subsequent fate of the introduced pollutant will be influenced by the physical properties of the effluent, the method of introduction, and the depth of introduction. If the density of the effluent stream is lower than that of the receiving waters, and the stream is introduced at the surface of the estuary, then vertical mixing of the waste materials with the receiving waters will be inhibited. However, if such a waste stream is introduced into the estuary near the bottom, the effluent will initially rise as a buoyant plume, entraining diluting water from the environment en route to the surface. Since the estuary is seldom deep enough that the ascending plume reaches the density of the receiving waters at an intermediate depth, the plume, still somewhat lower in density than the surrounding estuarine waters, will spread out on the surface.

If the waste effluent is denser than the estuarine waters, the waste stream will tend to spread out on the bottom. Initial mechanical dilution can be enhanced in this case by introduction of the waste stream near the surface, thus providing for a descending plume that will entrain diluting water en route to the bottom.

The major sources of pollution to an estuary are usually introduced by way of effluent streams that are less dense than the receiving waters, and as noted above, the most effective way to introduce such a waste stream would be as a bottom discharge. The effectiveness of the dilution of the ascending plume is enhanced by discharging the waste stream through a multiport diffuser, designed so that the ascending plumes overlap slightly when they reach the surface. This method produces an elongated volume source in the surface waters, which is then further subjected to the physical processes of movement and dispersion in the estuary.

In some situations, the volume rate of discharge of the effluent stream is so great that little mechanical dilution can be obtained from a rising plume in the depth range available in the estuary without a complex and costly diffuser system. An alternative method of obtaining effective initial mechanical dilution is to introduce the waste stream as a high-velocity jet directed across the waterway.

If the method of introduction produces sufficient initial mechanical dilution to significantly reduce the density difference between the waste stream and the estuarine waters, further dilution by mixing is enhanced.

In the surface layers, the diluted effluent will be extended into an elongated horizontal plume by the prevailing tidal currents. During ebb tide, the plume will extend down the estuary, and during flood tide the plume will extend up the estuary. Horizontal and vertical turbulent diffusion will act along the length of the plume to spread the waste materials and to continually reduce the contaminant concentration with distance from the source. Each reversal of the tidal current will result in a folding back of the spreading plume. However, because of the large-scale turbulent eddies, the plume will seldom fold exactly back on itself and will also seldom follow the same path on successive tides. Thus, there will develop a widespread contaminant field of relatively low concentration, on which is superimposed, with each tide, a relatively narrow plume of higher concentration.

Tidal oscillations past irregularities in the shoreline are an important mechanism in the longitudinal dispersion of an introduced pollutant. This is made most evident by considering a contaminated volume, produced by an instantaneous release, as it is carried up and down the waterway by the tidal currents. Frequently, eddies associated with slight embayments or with points of land that project into the waterway will temporarily trap water containing high concentrations of pollutant as the contaminated volume moves past these shore features on one or the other phase of the tide. The main bulk of the contaminant is carried on past the shore feature by the tidal current, while the material trapped by the shore feature slowly spreads out into the main stream, leading to an effective dispersal behind the bulk of the contaminated volume. When the tide reverses, the process is repeated, with a resulting dispersion on the opposite side of the contaminated volume.

The theoretical treatment of mixing discussed earlier in this chapter can be modified to treat the distribution of concentration of a pollutant discharged into an estuary either as a local nearly instantaneous source or as a local continuous source. The modification includes the effects of the side boundaries and of the boundary conditions assumed at the river and seaward ends of the estuary. Determination of the probable time-dependent distribution in a plume originating from a continuous source requires further consideration of the oscillatory nature of the tidal current. Carter (1965) has described a theoretical approach that appears to satisfy many of the observed features of the oscillating plume in a tidal estuary.

The use of theoretical relationships for mixing appear to be most useful in describing the detailed distribution of pollutant concentration within, for example, a tidal segment of an estuary (i.e., the segment defined by the excursion from its source of a particle of water in an estuary, headward on the flood tide and seaward on the ebb tide). A number of investigators have, on the other hand, treated the broader aspects of the flushing of an estuary. In such studies, concern is directed toward the broad mechanisms of the movement of the pollutant through the estuary and of its ultimate discharge to the open ocean. Ketchum (1950), Stommel (1953), Kent (1958), and Dorrestein (1960) all treat the flushing of estuaries as a one-dimensional (longitudinal) problem. Stommel and Kent deal with solutions to the one-dimensional advection–diffusion differential equation, while Ketchum and Dorrestein use a finite, segmented approach, sometimes referred to as a box model.

More recently, Pritchard (1969) described a two-dimensional box model that is particularly suited to partially mixed estuaries. In this approach, the estuary is divided into segments along the length of the estuary, and each such longitudinal segment is further subdivided vertically at the depth of the maximum vertical salinity gradient, which approximately defines the boundary between the net non-tidal seaward-flowing upper layer and the deeper layer in which the flow is directed headward in the estuary. Using volume and salt continuity requirements for each segment, a series of simultaneous equations is developed from which the larger scale advective flows and vertical-exchange coefficients can be determined.

It is theoretically possible to obtain numerical solutions to the basic hydrodynamic and kinematic equations using high-speed computers. These equations are so complex that to date it has not been possible to treat the complete transient-state equations with even the largest of high-speed computers. However, numerical modeling of estuarine circulation, and of the distribution of salinity and introduced wastes, using models of considerably greater complexity than have been employed where analytical solutions were sought, is developing rapidly.

In numerical modeling of estuaries, the equations of motion are used to determine both the mean circulation and the tidal flow. To date, the Navier-Stokes form of the equations of motion, in which the local and field acceleration terms are neglected, has been used for the mean flow, and the classical wave equations for the computations of tidal flow, using the rise and fall of the tide at the entrance as input. The continuity equations for mass and salt must be solved simultaneously with the equations of motion to predict the temporal and spatial distribution of both the circulation and the salinity. Prediction of the distribution of concentration of a waste requires the simultaneous solution of the convective-diffusion equations and the equations of motion.

In all cases to date, the equations employed in the numerical model still represent considerable simplification of the complete hydrodynamic and kinematic equations. In particular, transient-state solutions for the nontidal circulation will ultimately be required, although such has not been done to date. Also, to treat the majority of estuarine problems, the vertical dimension must be included, since the characteristic two-layered estuarine circulation dominates so many of the estuaries in this country.

The greatest problem with numerical modeling of the estuary is that the relationships between the diffusion parameters that enter the equations and predictor variables such as tide, weather, and circulation are not known. There are two schools of thought on how to overcome this problem. One contends that if the field of motion is determined in sufficient detail in space and time, the diffusion terms become relatively unimportant, and only rough estimates of the diffusion parameters are required. The second school advocates determining the pertinent relationships by which the diffusion parameters applicable to any desired averaging scale can be computed. The advantage of the first approach is that at least two-dimensional modeling is possible now, though at considerable cost in computer time, since very short intervals in time and space are involved. The advantage of the second approach is that, for situations requiring much less detail in time and space, less computer memory and computer time is required, and the possibility of extending the model to three spatial dimensions appears more favorable.

Hydraulic models of estuaries have been and are being used to study the physical dispersion of proposed waste discharges. A hydraulic model is inherently three-dimensional, and time variations in tidal input and freshwater inflow are readily included in the model. Properly verified hydraulic models of estuaries are capable of producing valid information on the physical dispersion of waste materials introduced into the estuary at intermediate and large scales of averaging. There is some indication that the details of concentration distribution near the point of discharge may not be properly scaled in the model as a result of overmixing at small scales, which is associated with the roughness elements required for distorted hydraulic models. Simmons and Lindner (1965) discuss the uses of hydraulic models of tidal waterways, and Ippen (1966) shows that while dispersion phenomena in such models are properly scaled in the saltwater section, there are questions regarding the scaling of such phenomena in the freshwater tidal-river section. The latter report also contains a chapter on the general scaling requirements for hydraulic models of estuaries.

It is doubtful that the costs of construction of a hydraulic model of an estuary can be justified on the basis of waste management alone; however, the use of existing hydraulic models, built for other purposes, does appear feasible. This is particularly true for problems where all three spatial dimensions are important, since proven alternative means of solution are not yet available.

SUMMARY

In this chapter the physical processes that result in movement and mixing of radioactive materials within the ocean are described. The current state of our knowledge of the essential features of the circulation of the oceans in relation to the associated distribution of properties is summarized, and recent theoretical studies of ocean circulation are reviewed.

Considerable progress has been made over the last ten years in understanding the processes of mixing in the ocean and in adjacent coastal and estuarine waters. Consequently, a major portion of this chapter is devoted to a description of our current knowledge of turbulent diffusion in the ocean. Comparisons between experimental and theoretical results are also given.

Some oceanographic implications of the observed distribution of natural tracers, such as carbon-14, and of fallout isotopes, are discussed. Particular attention is given to the conflicting evidence concerning the rate of vertical penetration of the dynamically passive isotopes strontium-90 and cesium-137, discussed elsewhere in this publication. Variations of carbon-14 with depth in the interval 1959 through 1966 at a location in the northeast Pacific Ocean are used to compute the vertical components of the mean velocity and of the eddy diffusivity. The results of this analysis are in line with those of Munk (1960), and contrary to the conclusions reached by Miyake *et al.* (1962).

Because man's most intimate contact with the marine environment occurs in coastal areas and in estuaries and other embayments, a section of this chapter is devoted to a brief description of the processes of movement and mixing in these waters. References are given to several recent papers containing more detailed descriptions of the state of our knowledge of the physical processes in estuaries and other coastal waterways.

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Chapter Five

MARINE CHEMISTRY

E. D. Goldberg, W. S. Broecker, M. G. Gross, K. K. Turekian

INTRODUCTION

Chemical species introduced into the oceans will initially be partitioned among three phases: the living biosphere, seawater, and inorganic and organic particles. On different time scales and in various sites, the ultimate fate of all such elements is removal to the sea floor or discharge into the atmosphere. Although it is not possible to treat in detail the paths of all chemical species in the various types of marine environments, we can attempt to systematize available chemical data for radionuclides in seawater and in sediments and to direct attention to those areas where information is lacking.

Marine environments can be conveniently categorized into two major domains: the coastal ocean and the open ocean. The coastal ocean includes estuaries, lagoons, the water over the continental shelves, and many marginal seas. The open ocean is that part not significantly affected by its boundaries with the continents or by the shallow-ocean bottom.

Obviously, there is no clear-cut boundary between the coastal and open ocean; therefore, for this report, we will consider water deeper than 1,000 m to be part of the open ocean.

The different physical and biological conditions of these two oceanic domains, relevant to problems of elemental distribution, are summarized in Table 1. The remainder of the chapter elaborates on these concepts.

CHEMICAL SYSTEMATICS AND ELEMENTAL REACTIVITIES IN SEAWATER

The formulation of models to study the dispersion of radioactive species introduced into the marine environment can be approached within the framework of chemical characteristics and the behaviors of their stable counterparts in seawater. The assumption is made that the dispersion paths of radionuclides introduced in soluble forms will be the same as those of stable nuclides introduced or existing in the marine environment. This implies that the chemical speciation is the same for both stable and radioactive nuclides in seawater.* Hence, it is possible to utilize existing knowledge of oceanic chemistry in formulating models. The complexities of seawater as an electrolyte solution and the inadequacies in our quantitative description of the oceanic

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*There are evidences in the literature of exceptions to this assumption. For example, stable zinc appears to exist in a number of complex species, some of which exchange slowly with the uncomplexed forms. Zinc-65, introduced into the marine environment from nuclear installations, may not attain the same chemical speciation in short time intervals as its stable isotopic counterparts. Some of the reported differences of specific activities between organisms and the water may result from the preferential uptake of a specific dissolved species of zinc. Another explanation may be that organisms may have picked up these elements from water masses where the specific activities were different but the speciation was similar. At present, resolution of this difficulty is not evident.

TABLE 1 Primary Factors That Can Alter the Chemical Composition of Seawater

Factor	Effect upon Elemental Behavior	Coastal Ocean	Open Ocean
Primary productivity	Fixation of elements in biomass, with subsequent transfer to deeper waters or to sediment	High, with local variability	Generally low, except in areas of divergence and high latitude
Particle input by river runoff, or resuspension from bottom	Provide surfaces for reactions of dissolved species and sites for bacterial activity	High	Low
Reservoirs for element accumulation	Storage of elements for various periods of time	Exist in sediments of ocean bottom	Exist in deep water and sediments of ocean bottom
Water circulation	Dispersion or retention of introduced species	Partial retention near coast	Dispersion dominant

system are well known. Substantial advances have been made in the chemical description of seawater over the past years, especially in regard to the speciation of the elements and to their relative reactivities. This new information permits meaningful statements about the expected behavior of at least some of the nuclides.

Table 2 lists average values for the concentration of elements in the ocean, together with what appears to be, from thermodynamic considerations, the most important chemical form, or forms, in solution. Column 4 divides the variability in the abundance of the elements into the following three categories.

A. Concentration is directly proportional to the salinity (such elements are referred to as "conservative").

B. There is a well-developed and readily described variability in concentration, as a function of depth, ocean basin, or both.

C. Reported variations in abundance are independent of salinity and not clearly dependent on depth or oceanic basin.

The elements in Class A are, in general, unreactive and display a remarkable stability in solution, while elements in Class B are usually involved in biological cycles, perhaps in inorganic processes that may result in an inhomogeneous oceanic distribution. The behavior of Class C elements in the ocean is not well understood.

There are several approaches to the description of the reactivity of an element in the oceanic chemical system, the most useful of which are residence times and the degree of undersaturation or supersaturation.

Residence time is defined as the average time an element spends in ocean water between introduction and incorporation into the sediments. Reactive elements generally have relatively short residence times in the oceans, while chemically inert species generally have much longer residence times, assuming a steady-state system in which the amount of an element entering the marine environment is compensated by the transfer of an equivalent amount from seawater to the sediments. The residence time, T, is defined by the relationship

$$T = A/(dA/dt),$$

where A is the total amount of the element in solution in the oceans and dA/dt is the amount introduced, and therefore precipitating, per unit time. Table 2 gives values of the residence times based upon values of dA/dt calculated from stream-input data.

Because of the oversimplified nature of the model, the absolute values of these residence times should be taken as a measure of reactivity rather than as a meaningful chronological number. The alkali metals and the alkaline-earth metals, for example, with long residence times, are characterized by the lack of reactivity of their ions in solution, while those elements intimately involved in biological cycles—phosphorus and silicon—have short residence times (10^3 to 10^4 years). The shortest residence times are calculated for elements primarily associated with lithogenous particles, such as aluminum, titanium, and thorium.

A second measure of reactivity derives from the degrees of undersaturation of ions with respect to their least soluble compound and their most stable dissolved species. It has been noted that, for reactive elements whose expected concentrations are calculated on the basis of their least soluble salts (Table 3) or on the basis of stable complexes (Table 4), and that are classified according to residence times (except for the rare earths and thorium), the observed concentrations are much lower than the limiting ones for oxygenated seawater. This indicates that phenomena other than solu-

TABLE 2 Geochemical Characteristics of the Elements[a]

Element	Seawater Concentration (μg/liter)	Principal Dissolved Species	Category[b]	Concentration Dissolved in Stream Waters (μg/liter)	Residence Time in Ocean (yr)
H	1.1×10^8	H_2O	A	–	–
He	7×10^{-3}	He (gas)	A	–	–
Li	1.7×10^2	Li^+	A	3	2.3×10^6
Be	6×10^{-4}	–	–	–	–
B	4.5×10^3	$B(OH)_3$, $B(OH)_4^-$	A	10	1.8×10^7
C	2.8×10^4	HCO_3^-, CO_3^{-2}	A	–	–
C (org.)	1×10^2	–	A	–	–
N	1.5×10^4	N_2 (gas)	A	–	–
N	6.7×10^2	NO_3^-	B	–	–
O	8.8×10^8	H_2O	A	–	–
O	6×10^3	O_2	B	–	–
O	1.8×10^6	SO_4^{-2}	A	–	–
F	1.3×10^3	F^-	A	100	5.2×10^5
Ne	0.12	Ne (gas)	A	–	–
Na	1.1×10^7	Na^+	A	6,300	6.8×10^7
Mg	1.3×10^6	Mg^{+2}	A	4,100	1.2×10^7
Al	1	–	–	400	1.0×10^2
Si	3×10^3	$Si(OH)_4$, $SiO(OH)_3^-$	B	6,500	1.8×10^4
P	90	HPO_4^{-2}, $H_2PO_4^-$, PO_4^{-3}	B	20	1.8×10^5
S	9.0×10^5	SO_4^{-2}	A	–	–
Cl	1.9×10^7	Cl^-	A	1,800	1×10^8
Ar	4.5×10^2	Ar (gas)	A	–	–
K	3.9×10^5	K^+	A	2,300	7×10^6
Ca	4.1×10^5	Ca^{+2}	A	15,000	1.0×10^6
Sc	$<4 \times 10^{-3}$	$Sc(OH)_3^0$	–	0.004	$<4 \times 10^4$
Ti	1	$Ti(OH)_4^0$	–	3	1.3×10^4
V	2	$VO_2(OH)_3^{-2}$	–	0.9	8.0×10^4
Cr	0.5	CrO_4^{-2}, Cr^{+3}	–	1	2.0×10^4
Mn	2	Mn^{+2}	C	7	1.0×10^4
Fe	3	–	–	670	2.0×10^2
Co	0.4	Co^{+2}	C	0.1	1.6×10^5
Ni	7	Ni^{+2}	C	0.3	9.0×10^4
Cu	3	Cu^{+2}	C	7	2×10^4
Zn	10	Zn^{+2}	C	20	2×10^4
Ga	3×10^{-2}	–	–	0.09	1×10^4
Ge	7×10^{-2}	$Ge(OH)_4$	–	–	–
As	2.6	$HAsO_4^{-2}$, $H_2AsO_4^-$	–	2	5×10^4
Se	9×10^{-2}	SeO_4^{-2}	C	0.2	2×10^4
Br	6.7×10^4	Br^-	A	20	1×10^8
Kr	0.2	Kr (gas)	A	–	–
Rb	1.2×10^2	Rb^+	A	1	5×10^6
Sr	8×10^3	Sr^{+2}	A	70	4×10^6
Y	1×10^{-3}	$Y(OH)_3^0$	C	–	–
Zr	3×10^{-2}	–	–	–	–
Nb	0.01	–	–	–	–
Mo	10	MoO_4^{-2}	A	0.6	7×10^5
Ru	–	–	–	–	–
Rh	–	–	–	–	–
Pd	–	–	–	–	–
Ag	0.3	$AgCl_2^-$	C	0.3	4×10^4
Cd	0.1	Cd^{+2}	–	–	–
In	<20	–	–	–	–
Sn	0.8	–	–	–	–
Sb	0.3	–	C	2	7,000
Te	–	–	–	–	–
I	60	IO_3^-, I	A	7	4×10^5
Xe	5×10^{-2}	Xe (gas)	A	–	–

TABLE 2 (Continued)

Element	Seawater Concentration (μg/liter)	Principal Dissolved Species	Category[b]	Concentration Dissolved in Stream Waters (μg/liter)	Residence Time in Ocean (yr)
Cs	0.3	Cs^+	A	0.02	6×10^5
Ba	20	Ba^{+2}	C	20	4×10^4
La	3×10^{-3}	$La\ (OH)_3^0$	C	0.2	6×10^2
Ce	1×10^{-3}	$Ce\ (OH)_3^0$	C	–	–
Pr	0.6×10^{-3}	$Pr\ (OH)_3^0$	C	–	–
Nd	3×10^{-3}	$Nd\ (OH)_3^0$	C	–	–
Sm	0.5×10^{-3}	$Sm\ (OH)_3^0$	C	–	–
Eu	0.1×10^{-3}	$Eu\ (OH)_3^0$	C	–	–
Gd	0.7×10^{-3}	$Gd\ (OH)_3^0$	C	–	–
Tb	1.4×10^{-3}	$Tb\ (OH)_3^0$	C	–	–
Dy	0.9×10^{-3}	$Dy\ (OH)_3^0$	C	–	–
Ho	0.2×10^{-3}	$Ho\ (OH)_3^0$	C	–	–
Er	0.9×10^{-3}	$Er\ (OH)_3^0$	C	–	–
Tm	0.2×10^{-3}	$Tm(OH)_3^0$	C	–	–
Yb	0.8×10^{-3}	$Yb\ (OH)_3^0$	C	–	–
Lu	0.1×10^{-3}	$Lu\ (OH)_3^0$	C	–	–
Hf	$<8 \times 10^{-3}$	–	–	–	–
Ta	$<3 \times 10^{-3}$	–	–	–	–
W	0.1	WO_4^{-2}	–	0.03	1.2×10^5
Re	0.008	–	–	–	–
Os	–	–	–	–	–
Ir	–	–	–	–	–
Pt	–	–	–	–	–
Au	1×10^{-2}	$AuCl_2^-$	C	0.002	2×10^5
Hg	0.2	$HgCl_4^{-2}$, $HgCl_2^0$	C	0.07	8×10^4
Tl	<0.1	Tl^+	–	–	–
Pb	0.03	$PbCl_3^-$, $PbCl^+$, Pb^{+2}	C	3	4×10^2
Bi	0.02	–	–	–	–
Po	–	–	–	–	–
At	–	–	–	–	–
Rn	6×10^{-13}	Rn (gas)	–	–	–
Ra	1×10^{-7}	Ra^{+2}	C	–	–
Ac	–	–	–	–	–
Th	$<5 \times 10^{-4}$	$Th(OH)_4^0$	–	0.1	<200
Pa	2.0×10^{-6}	–	–	–	–
U	3	$UO_2(CO_3)_3^{-4}$	A	0.04	3×10^6

[a] Compiled from Goldberg (1965) and Turekian (1969).
[b] See text for explanation of letters.

TABLE 3 Expected Equilibrium Concentrations for Some Elements, Based on Insoluble Salts of Phosphate, Carbonate, Hydroxide, and Sulfide (Concentrations in log moles/liter)[a]

Element	PO_4^{-3} (log $a = -9.3$)	CO_3^{-2} (log $a = -5.3$)	OH^- (log $a = -6$)	S^{-2} (log $a = -9$)	Observed in Seawater
La^{+3}	−11.1	–	0	–	−10.7
Ce^{+3}	−10.0	–	–	–	−10.2
Th^{+4}	−11.8	–	–	–	−11.7
Cr^{+3}	−11.3	–	–	–	−8.0[b]
UO_2^{+2}	9.2	–	–	–	−7.8[c]
Fe^{+3}	−10.6	–	–	–	−7.3[d]
Fe^{+2}	–	–	–	−6.4	−7.3[d]
Mn^{+2}	–	−3.1	+0.2	−2.6	−7.4
Co^{+2}	−4.4	−6.5	−2.2	−12.1	−8.2
Ni^{+2}	−2.9	−0.6	−3.2	−10.7	−6.9
Cu^{+2}	−5.1	−3.5	−5.8	−26.0	−7.3
Ag^{+1}	−2.0	−2.7	−1.5	−19.8	−8.5[e]
Zn^{+2}	−3.5	−3.7	−3.5	−14.1	−6.8
Cd^{+2}	−3.7	−5.0	−0.5	−16.2	−9.0
Hg^{+2}	–	–	−12.5	−43.7	−9.1[e]
Al^{+3}	–	–	−12.0	–	−8.3
Ga^{+3}	–	–	−16.0	–	−9.3
Sn^{+2}	–	–	−15.0	−16.0	−8.2
Pb^{+2}	−6.8	−6.8	−2.0	−16.6	−9.8[e]

[a] Calculations made with the following activity coefficients: monovalent ions, 0.7; divalent ions, 0.1; trivalent ions, 0.01.

[b] Occurs primarily as CrO_4^{-2}.

[c] Occurs primarily as $UO_2(CO_3)_3^{-4}$.

[d] May occur as particulate phases.

[e] Occurs primarily as chloride complexes.

TABLE 4 Complexes Formed in Seawater and Their Expected and Observed Equilibrium Concentrations (in moles/liter)

Element	Complex	Expected[a]	Observed
Silver	$AgCl_2^-$	−4.2	−8.5
Mercury	$HgCl_4^-$	+1.9	−9.1
	$HgCl_2^0$	−0.3	
Lead	$PbCl_3^-$	−5.6	−9
	$PbCl^+$	−5.8	

[a] Using least soluble salt, from Table 3.

bility equilibria are determining elemental concentrations.

For lanthanum, cerium, and thorium, the solubility of the phosphate appears to govern their concentrations in seawater, keeping them at a remarkably low level (Table 3). Their removal from seawater, or their transfer from surface to deeper waters or to the sediments, may be enhanced by the biological cycles in which phosphate is regenerated by the decomposition of organic detritus descending through the water column.

COASTAL OCEAN

Several characteristics of the coastal ocean appear to be especially significant in determining the behavior of elements. They are (a) rapid mixing of substances injected into the ocean; (b) circulation patterns that tend to favor retention near the coast of substances introduced into the coastal ocean; (c) relatively intense biological activity; and (d) the abundance of particles (both biogenous and lithogenous) suspended in the water.

The discharge of a river quickly mixes with a volume of seawater several times as large to form a low-salinity surface layer that flows into the coastal ocean, mixing continuously. Such discharges can be identified on the basis of salinity and other parameters. The distribution of dissolved substances discharged by rivers usually replicates the patterns shown by these identifying parameters. Most soluble substances introduced along the shore will be mixed fairly rapidly, even when not clearly associated with a major river discharge.

The circulation of the coastal ocean tends to favor the retention of dissolved substances near the coast. The thin plume of low-salinity water formed by the discharge of a river moves along the coastline for many kilometers, carrying with it many of the substances injected into it. The sub-

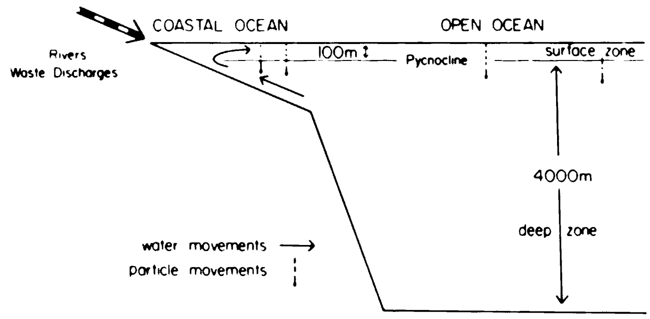

FIGURE 1 Schematization of the coastal and open ocean.

surface circulation also tends to favor, near the coast, the retention of most substances injected into the coastal ocean. Where precipitation and runoff exceed evaporation, an estuarine-like circulation results (see Chapter 2) in which fresh water added to the ocean surface mixes with salt water from below and moves generally seaward (Figure 1). A shoreward subsurface flow replaces the salt water that has moved upward into the surface layers. In areas where evaporation predominates, the coastal ocean does not exhibit estuarine-like circulation, but such areas are relatively uncommon. The increased nutrient supply due to this vertical circulation and the nutrients supplied by rivers are major causes of the relatively large primary production in the coastal ocean, because subsurface waters brought into the surface layers supply nutrients to the photic zone.

Certain substances in the surface waters become associated with the descending organic debris. Chemical species released by the decomposition of particles sinking out of the surface layer will tend to be moved landward in the subsurface flow, to return eventually to the surface layer. Although materials are lost to the sediments or to the surface layers of the open ocean, the circulation of the surface and subsurface waters tends to retain some chemical species in the coastal ocean.

Dissolved oxygen in the near-bottom waters or in the sediment may be depleted or even completely exhausted, depending on the supply rate of dissolved oxygen relative to the rate of consumption in the decomposition of organic matter formed in the photic zone. Where exhaustion of dissolved oxygen occurs, sulfate-reducing bacteria are involved in the production of H_2S. Complete exhaustion of the dissolved oxygen in the water column is not common in the coastal ocean except where water circulation is greatly restricted, as in certain fjords, or where primary productivity is extremely high. In areas of large primary productivity, where the supply rate of dissolved oxygen is large enough to prevent oxygen depletion in the near-bottom waters, H_2S may nevertheless form in the sediments because the rate of oxygen diffusion into the sediment is slow.

The presence of hydrogen sulfide may act as a control on the concentration of many metallic ions in seawater, as can be seen in Table 3. A sulfide ion activity of 10^{-9} moles/liter, corresponding to that of a "stinking mud," results in an environment favorable to the removal of metals that form highly insoluble sulfides. Certain metals will be depleted even in moderately sulfide-rich water, i.e., $a_{S^{-2}} = 10^{-13}$ moles/liter.

Manganese and iron appear to be enriched in sulfide-rich waters inasmuch as the sulfides of the reduced forms of these elements are relatively soluble (Table 3). Higher concentrations of manganous ions in the anoxic waters of the Black Sea, relative to the oxygenated surface waters, have been reported, apparently confirming the existence of this process of enrichment.

Reactive elements brought into the ocean may become associated with land-derived solid materials. Such materials derived from organisms incorporate certain elements into their tissues or skeletons, often concentrating them manyfold relative to their concentrations in seawater. After an organism's death, some of these elements are incorporated in the bottom sediment, along with undecomposed organic remains. Nutrient elements, such as phosphorus and nitrogen, may be released as soluble species during decomposition of the organic remains in the water.

Particle-associated chemical species and reactive elements that were taken up by the organisms and not quickly released by decomposition are eventually incorporated in the sediment, which is a major reservoir for such elements.

OPEN OCEAN

The open ocean differs significantly from the coastal ocean; thus, there appear to be major differences in elemental behavior (Table 1). Among the significant differences between the coastal and open ocean are the general nutrient deficiencies and relative scarcities of organisms and particles in surface layers of the latter. The well-developed density stratification in the open ocean inhibits large-scale vertical mixing and the upward movement of nutrients from deep waters, except at divergences or in high latitudes. Unreactive elements generally pass through the coastal ocean into the open ocean. Reactive elements tend to become associated with particles or to be utilized by organisms and thus are removed from seawater in the coastal ocean. Elements with short residence times tend to accumulate in the sediment deposited in the coastal ocean; elements with long residence times tend to accumulate in the open ocean. Uranium, which can be reduced to a tetravalent state in coastal deposits, is an exception—it has a long residence time in the ocean but tends to accumulate in inshore deposits.

In the open ocean, some chemical species removed from the surface layers by organisms are released in adjacent deeper waters as the particles decompose before reaching the bottom. Among these are elements such as phosphorus and nitrogen, which are retained by the partially decom-

TABLE 5 Ratios of Elemental Concentrations in Deep Waters C_{deep} and in Surface Waters C_{surf} of the Pacific Ocean

Element	C_{deep}/C_{surf}
Li	1.0
Na	1.0
K	1.0
Rb	1.0
Cs	1.0
Mg	1.0
Ca	1.02
Sr	<1.1
Ba	4
Ra	4
CO_2	1.2
S as SO_4^{-2}	1.0
P as phosphate ions	>10.0
Si as $Si(OH)_4^0$	>10
N as NO_3^-	>10.0
Mn	~1
Mo	~1
U	~1
Br	~1

posed organic matter and deposited with sediments in the coastal ocean. Consequently, open ocean sediments are relatively less important as a reservoir for reactive elements than are the coastal bottom deposits.

Downward transport and subsequent decomposition of the particulate remains of marine organisms are the major cause of chemical inhomogeneities within the ocean. Various elements fixed by marine organisms in the surface waters of the ocean are transported to, and released in, depths where solution or oxidation of the debris takes place. The net result is a depletion of these elements in surface waters and an enrichment in deep waters.

The ratio of the concentration of an element (normalized to a standard salinity) in the deep waters to that in the surface waters of the Pacific Ocean provides a convenient index of the effectiveness of this process. Values are given in Table 5 for several elements.

Nitrogen, phosphorus, and silicon are removed very efficiently, the last element primarily as the opaline tests of diatoms and radiolaria, and the first two as basic constituents of the organic material itself. The fact that the nitrogen/phosphorus ratio (16/1) is nearly constant in all the major open ocean water masses and in all major types of organisms attests to the dominance of organic activity in the generation of inhomogeneities in the concentrations of these two elements. Carbon is depleted not only by its incorporation in organic molecules but also by its fixation and removal as $CaCO_3$ (in foraminifera, coccoliths, pteropods). Calcium owes its depletion to $CaCO_3$ and possibly to calcium phosphate precipitation. The absence of significant concentration variations for relatively unreactive elements such as cesium strongly suggests that transport by organisms is unimportant compared to transport by physical mixing in the water column.

Nonsystematic variations in the strontium/chlorinity and cesium/chlorinity ratios have been reported, but the results are difficult to interpret. Possibly, the strontium concentration of seawater is altered by the dissolution of large numbers of celestite (strontium sulfate) radiolaria. For cesium no mechanism is evident.

The depletion factors given in Table 1 can be easily converted to residence times for the element in surface water, T_s, relative to transfer to the deep sea. Assuming that such elements as Na, K, Mg, Cl, and S are transported only by physical mixing, we can use their residence times as references. From simple material-balance considerations, it can be shown that

$$T_s \text{ element} = \frac{C_{surf}}{C_{deep}} \qquad (T_s \text{ chlorine})$$

where C_{surf} and C_{deep} are the concentrations of the element in surface and in deep water, respectively. Radium, for example, because of its fourfold enrichment in deep water, apparently resides in surface water only one quarter the time that chlorine resides there. The silicon residence time is on the order of one tenth the chlorine residence time.

The residence time of a relatively nonreactive element is about 20 years in the mixed layer for the world ocean—this estimate is based upon physical mixing processes solely. Thus, barium or radium would have passage times of about 5 years in the mixed layer, while silicon would be removed in about 2 years.

The variations with depth of many trace elements in seawater are poorly known, as indicated in Table 2. For such elements, another approach is needed to delimit their reactivities in biological cycles. One possibility is to use the chemical composition of marine plankton. The degree to which a given element is enriched by marine organisms should be related to the importance of its transport by particles. Three types of solid phases must be considered: organic detritus, calcareous exoskeletal materials, and opaline exoskeletal materials. The total ocean-wide productions can be estimated from the deep-water excesses (concentration in deep water minus concentration in surface water) of silicate, of total dissolved inorganic carbon or carbonate alkalinity, or of nitrate and phosphate.

The deep Pacific excesses are as follows:

P	3×10^{-6} moles/liter
N	5×10^{-6} moles/liter
C	4×10^{-4} moles/liter
Si	2×10^{-4} moles/liter
Carbonate alkalinity	2×10^{-4} equivalence/liter

TABLE 6 Elemental Concentration Factors for Marine Organisms (Expressed as the Logarithm)[a]

Element	Plants	Animals
Al	4	4.5
As	3	2
Ba	–	1
Be	3	3
B	0.1	0.5
Cd	2	5
Ca	0.5	1
Ce	2.5	2
Cs	0.5	1
Cl	–	-1
Cr	4.5	5
Co	3	2
Cu	2	3
F	0.1	–
Ga	1.5	1.5
Au	3.5	1.5
I	3.5	–
Fe	4.5	3.5
Pb	4.0	4.0
Li	–	-0.3
Mg	-0.2	-0.2
Mn	3.5	3.5
Mo	1.5	1.5
Ni	2.0	3.0
Nb	3.0	5.5
P	3.7	4
Pu	3	3
K	-0.1	0.5
Ra	–	–
Ru	2.0	1.0
Sc	4.5	3.5
Si	2.3	2.0
Ag	3.0	3.0
Na	-1.5	-0.8
Sr	1.0	0.5
S	-0.5	-0.5
Sn	2.5	2.0
Ti	3.5	3.5
W	3.0	2.0
U	–	–
V	2.5	2.0
Zn	3.0	4.0
Zr	3.0	3.2

[a] Concentration factors for organisms from the open and coastal areas are included. Data is insufficient to ascertain differences that might result from systemic variations in elemental concentrations in these two marine domains.

TABLE 7 Concentration Factors between Seawater and Plankton Ash for Some Trace Elements[a]

Element	Seawater (μg/liter)	Plankton Ash (μg/g) Plants (Sargassum)	Plankton Ash (μg/g) Animals	Concentration Factors (liters seawater per grams plankton ash) Plants (Sargassum)	Concentration Factors (liters seawater per grams plankton ash) Animals
P	88	20,000	20,000	230	230
Ag	0.3	0.3	0.3	1	1
Al	1	65	300	65	300
B	4,450	1,200	140	0.27	0.031
Ba	20	120	52	6.0	2.5
Cd	0.11	8	13	72	120
Co	0.4	3	3	7.5	7.5
Cr	0.5	9	7	18	14
Cu	2	270	270	135	135
Li	170	6	40	0.04	0.2
Ni	6.6	27	12	4	2
Sr	8,100	8,500	930	1	0.1
Ti	1	26	120	26	120

[a] Based on unpublished data for marine plankton species from G. Thompson and V. T. Bowen (Woods Hole), H. Curl (Oregon State), G. Nicholls (Manchester), and K. K. Turekian (Yale).

For the total dissolved inorganic carbon, 3×10^{-4} moles/liter result from the combustion of organic carbon, and 1×10^{-4} moles/liter from the fallout of $CaCO_3$. The carbonate alkalinity, nitrogen, phosphorus, and carbon data are internally consistent, and show that for each mole of $CaCO_3$ dissolving in the deep sea, organic material containing 3 moles of organic carbon must be oxidized. The silicon data suggest that for each mole of $CaCO_3$, there must be 2 moles of opaline silica precipitated. The corresponding weight ratios would be–dry organic material: $CaCO_3 : SiO_2 = 0.7 : 1.0 : 1.3$.

The elemental concentration factors for marine organisms, with respect to seawater, are given in Table 6. Although there is an uncertainty of at least an order of magnitude in these values for many of the elements, they provide an entry to the problem of the dissemination of elements through biological activity. Where reliable concentrations are known, we can estimate the deep-water excesses of trace elements. If we assume, for example, that any element incorporated into plant tissue is released in a manner similar to that of phosphorus without fractionation, then the enrichment values in Table 6 assume an importance. Those elements that are reported to be enriched to the same level as phosphorus, or higher (Al, Sc, Pb, Fe, and Cr in Table 6), would be expected to have significant deep-water enrichment. Such elements are removed from surface water by sinking particles. Since phosphorus can attain enrichments of an order of magnitude or so in the deep ocean relative to the surface ocean, elements that are enriched in marine plants by even one order of magnitude less than phosphorus would be expected to have detectable nonhomogeneous vertical distributions, whereas, if the enrichment of an element relative to phosphorus is down by more than one order of magnitude, no significant vertical-concentration changes are to be expected.

Concentration factors based upon direct measurements of elemental contents in ashed organisms are given in

Table 7 in the arbitrary units of liters of seawater per gram of ash. On this scale, the concentration factor for phosphorus is about 200 (or more, if we use the lower phosphorus concentration of surface waters). These more recently determined values provide an additional frame of reference for considerations of vertical distributions of elements in the marine environment.

CONCLUSION

The chemical factors governing the dispersion of species introduced into the marine environment are in need of better definition, especially with regard to coastal environments. The involvement of many elements in primary plant productivity is not known. The comparative chemistries of oxic and anoxic waters warrant much additional work. The natures and roles of suspended inorganic and organic phases are poorly understood. An elaboration of such problems would provide firmer bases from which to systematize the general dispersion problem. In particular, we see need for chemical investigations along the following lines:

1. Elemental concentrations should be sought for the marine plants that are most important in the fixation of carbon in both coastal and open ocean areas. The relative importance in the coastal oceans of attached plants to planktonic species with regard to the uptake of specific elements warrants attention.
2. For the coastal ocean, the relative effects of interactions of introduced species with nonliving particulate organic and inorganic phases and with viable organic phases should be ascertained.
3. The distribution of heavy metals in the open ocean and their distribution among dissolved, colloidal, and particulate states should be determined.

SUMMARY

Although initially incorporated in the biosphere, or dissolved in seawater or associated with particles, materials introduced into the ocean are eventually removed from seawater and deposited on the ocean bottom. The available data are systematized for certain important radionuclides in seawater and sediments.

Marine environments may be divided into coastal ocean (above the continental shelf) and open ocean, where the water is deeper than 1,000 m. Physical and biological conditions are different in these ocean areas, and these in turn influence the behavior of the elements in the ocean. Elements are classified as (A) conservative elements, (B) those exhibiting well-developed and regular variability in concentration with depth or ocean basin or both, and (C) those whose concentration is independent of depth or ocean basin. Elements in class A are generally unreactive in seawater. Elements in class B are usually involved in biological cycles. Elements in class C are not well understood. Elements have also been classified according to their residence time and reactivity in seawater.

The coastal ocean is characterized by rapid mixing of substances, partial retention close to coast of solids and certain reactive elements, relatively intense biological activity, and abundance of particles. Many reactive elements become associated with particles and are deposited near the continents.

Open ocean waters are characterized by relative deficiency of nutrients in surface waters and relative scarcity of organisms and particles. Elements with long residence times tend to accumulate in open ocean waters. Sediments depositing in deep ocean areas are less important as a reservoir for reactive elements than are sediment deposits in coastal areas. Downward transport and subsequent decomposition of particles is the dominant cause of depletion of the element in surface waters and enrichment in subsurface waters.

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Chapter Six

MARINE SEDIMENTS AND RADIOACTIVITY

E. K. Duursma, M. G. Gross

Sedimentary particles have a substantial capacity to remove radionuclides from the ocean, either depositing them on the bottom out of contact with man or concentrating them on particles that may be picked up by filter-feeding organisms, some of which are used by man for food (Templeton and Preston, 1966; Chapters 7 and 8, this report). The role of marine sediments in these processes has been studied less than most other aspects of marine radioactivity, as was already pointed out by Koczy and Rosholt (1962). This is perhaps surprising but understandable. Marine sediments deposited at great depths and far from waste-disposal localities contain little fallout and waste activity. The explanation is that a soluble radionuclide introduced into the sea will have a fate similar to that of the natural elements (see Chapter 5) and that the yearly input of radionuclides into marine sediments is related to the residence time of stable elements in seawater. For the deep ocean this input to the sediments has remained quite small since the mid-1950's, when man-made radionuclides first appeared in large amounts in the oceans.

Because of the diverse composition of marine sediments, it is still difficult to predict a complete quantitative picture for areas with highly contaminated seawater. It is possible, however, to approximate satisfactorily the general distribution and deposition behavior to be expected and the relative magnitude of the effects, based on laboratory experiments and field studies.

To understand the role of sediments in the uptake of radionuclides, it is necessary to know the capacity of sediments and sedimentary particles, in relation to their physical and chemical properties, to sorb (or desorb) radionuclides. It is also necessary to know in which areas of the ocean sediments are being deposited or in which large quantities of suspended sediment might sorb and remove radionuclides from seawater. These problems will be discussed in relation to the known mechanisms of sediment–radionuclide interactions. In addition, a preliminary evaluation of the possible role of sedimentary materials in removing radionuclides from contaminated seawater will be discussed.

SORPTION OF RADIONUCLIDES BY MARINE SEDIMENTS

Sediment Composition

Sediments can sorb radionuclides of a variety of elements. Our limited knowledge prevents accurate predictions, but important relationships and general principles can be demonstrated. For instance, several investigators have reported that radionuclides are more concentrated in fine-grained than in coarse-grained sediments (Hamaguchi, 1962; Noshkin and Bowen, 1965; Kautsky, 1966; Templeton and Preston, 1966). Experiments by Duursma and Eisma (unpublished) with various radionuclides and mostly fine-

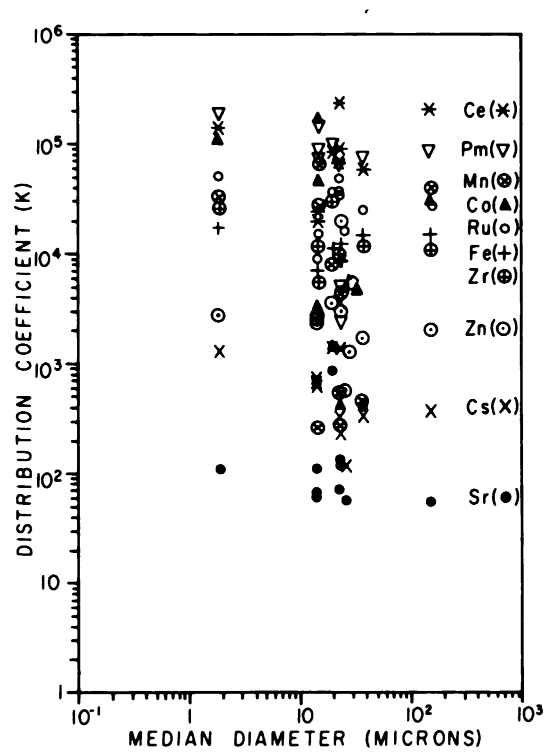

FIGURE 1 Distribution coefficients (K) for ten radionuclides as a function of the median grain size (D_{50}), in microns, in various marine sediments. Determined with the thin-layer technique (Duursma and Bosch, 1969); filters with 10-mg sediment are exposed to 150 ml seawater enriched with radionuclides.

The marine sediments used were sampled by French, German, U.S., Soviet, Italian, and Finnish research vessels at the following locations (Duursma and Eisma, unpublished):

Area	Depth (m)	Location
Mediterranean	2,710	43°N, 7½°E
Black Sea (used oxygenated)	?	44°N, 35½°E
Caribbean	?	15°N, 69°W
Romanche Trench	7,200	0°N, 18°W
Pacific	5,210	18°N, 132°W
Atlantic	4,150	27°N, 39°W
Antarctic	3,970	50°S, 127½°W
Gulf of Trieste	30	46°N, 14°E
Red Sea (Discovery Deep)	2,080	20°N, 38°E
Baltic	220	57°N, 20°E

grained marine sediments demonstrated a lack of correlation between radionuclide sorption and grain size. Uptake in these sediments (Figure 1) may be the result of poor sorting, as there were relatively large amounts of fine particles in all the samples. On the other hand, the selective sorption of fine particles is not always directly proportional to the specific surface area. It depends on the specific minerals and radionuclide involved (Table 1). Nevertheless, in Irish Sea sediments near the pipeline outlet, at Sellafield, the amount of radioactivity in different grain-size fractions was nearly proportional to the specific surface area of the grains (Jones, 1960; Jefferies, 1968).

In sediments with large cation exchange capacities, as calculated from the mineral composition (Duursma and Eisma, unpublished), the radionuclides were somewhat more strongly sorbed (Figure 2). The sorption capacity for a specific nuclide, calculated as the distribution coefficient,* did not on the average differ by more than a factor of four between the sediments with extremely high and extremely low cation exchange capacities. Increased carbonate content is slightly correlated with decreased radionuclide uptake (Figure 3); in part, this can be explained as resulting from lower cation exchange capacities of the carbonate minerals.

Sediments with higher iron content have larger distribution coefficients (Figure 4). Iron in sediments may occur as concretions or particle coatings (Carroll, 1958), enhancing the formation of sorption complexes for ^{106}Ru (Jones, 1960). Sorption of ^{59}Fe, however, shows no obvious correlation with the total iron content of the sediment used in the experiments.

Radionuclide Sequence

Some radionuclides are more strongly sorbed to sediment particles than others; the specific sequence varies with different sediments. In some Atlantic, Pacific, and Indian Ocean and Mediterranean Sea sediments, the sequence of sorption of radionuclides from seawater (from those weakly sorbed to those strongly sorbed) was (Duursma, 1969):

$$^{45}\mathrm{Ca} < {}^{90}\mathrm{Sr} < \mathrm{U, Pu}, {}^{137}\mathrm{Cs} < {}^{86}\mathrm{Rb} < {}^{65}\mathrm{Zn} < {}^{59}\mathrm{Fe}, \\ {}^{95}\mathrm{Zr/Nb}, {}^{54}\mathrm{Mn} < {}^{106}\mathrm{Ru} < {}^{147}\mathrm{Pm}.$$

The radionuclides ^{144}Ce and ^{60}Co fall somewhere between ^{59}Fe and ^{147}Pm in the ease with which they are sorbed. However, in the Irish Sea, ^{106}Ru is less sorbed than, for

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*For the purpose of this discussion, some relevant terms are defined as follows:

Distribution coefficient–A dimensionless ratio of the amount of bound radionuclide per unit volume of dry sediment to the amount of radionuclide per unit volume of seawater. The values presented are apparent-equilibrium values and are about 0.4 to 0.5 the value of similar coefficients defined on a weight basis. The larger the distribution coefficient, the stronger the binding or sorption of the radionuclide to the sediment.

Diffusion coefficient–A diffusion coefficient for a medium not in motion and in which the transport of radionuclides is caused by concentration gradients in the interstitial water; side reactions such as sorption are included.

TABLE 1 Distribution of Sorbed Radionuclides and Total Radioactivity in Different Size Fractions in Three Types of Sediment[a]

Sediments	Size Fractions		Distribution Coefficients for Each Size Fraction (value × 10^2)									
Origin	μ	% Weight	^{90}Sr	^{137}Cs	^{106}Ru	^{59}Fe	^{65}Zn	^{60}Co	^{147}Pm	^{54}Mn	^{95}Zr/Nb	^{144}Ce
Dutch Wadden Sea[a]	>64	51.1 ± 4.9	0.0	0.0	0.0	0.0	0.0	5.8	0.0	0.0	0.3	0.4
	32–64	21.7 ± 4.5	0.0	1.6	3.1	76.0	0.0	3.7	0.0	1.4	42.0	3.3
	16–32	9.5 ± 3.2	0.0	5.4	0.0	53.0	260.0	65.0	0.0	35.0	66.0	120.0
	8–16	5.6 ± 2.0	0.0	12.0	7.3	370.0	490.0	59.0	0.0	8.0	1,040.0	950.0
	4–8	8.4 ± 1.9	0.0	16.0	5.2	510.0	380.0	430.0	320.0	480.0	1,220.0	540.0
	<4	3.7 ± 0.8	26.0	6.2	4.7	540.0	112.0	220.0	280.0	97.0	670.0	124.0
Mediterranean off Monaco[a]	>64	0	–	–	–	–	5.2[b]	–	–	–	–	–
	32–64	3.0 ± 1.3	15.0	2.8	41.0	15.0	68.0	380.0	24.0	19.0	117.0	32.0
	16–32	15.2 ± 4.2	2.5	1.3	11.0	101.0	140.0	540.0	0.8	41.0	290.0	73.0
	8–16	36.5 ± 8.6	7.6	1.4	22.0	118.0	150.0	730.0	4.5	61.0	150.0	82.0
	4–8	39.1 ± 7.5	9.3	2.3	34.0	183.0	140.0	820.0	101.0	76.0	310.0	147.0
	<4	6.2 ± 1.9	5.9	0.5	7.8	63.0	97.0	140.0	130.0	23.0	160.0	41.0

Sediments	Size Fractions			
Origin	μ	% Weight	Radioactivity μCi/g dry weight	
Irish Sea Pipeline outlet, Sellafield[c]	100–200	26.0	2.0×10^{-4}	Specific Radioactivity = $1.3 \pm 0.4 \times 10^{-6}$ μCi/cm^2
	50–100	27.4	3.0×10^{-4}	
	20–50	22.6	9.0×10^{-4}	
	10–20	14.0	2.5×10^{-3}	
	4–10	6.1	4.6×10^{-3}	
	<4	4.0	6.2×10^{-3}	

[a] The Dutch Wadden Sea and Mediterranean sediments were suspended in radionuclide-enriched seawater for one month (30 g sediment in 20 liters of seawater); size fractions were separated by sedimentation techniques (Duursma and Eisma, unpublished).

[b] For ^{65}Zn, another Mediterranean sediment was used with 3.3%, >64 μ; 7.6%, 32–64 μ; 49.2%, 16–32 μ; 24.9%, 8–16 μ; 12.7%, 4–8 μ; and 2.3%, <4 μ.

[c] The Irish Sea sediments were exposed in the field to radionuclides discharged near Sellafield (Jones, 1960).

example, ^{95}Zr/Nb (Preston, personal communication), a result that agrees with the figures given for the sediments mentioned in Table 1. Similar results have been obtained for freshwater sediments (Garder and Skulberg, 1964), except that ^{137}Cs was more strongly sorbed in fresh water.

Details of the mechanisms controlling such sorption sequences are still unknown; there is, however, an extensive literature on ion-exchange processes in soils and minerals (see Robinson, 1962, for a discussion and for references). The weak sorption of ^{90}Sr and ^{45}Ca on marine sediments suggests that these radionuclides are involved in exchange reactions with their stable isotopes, which are relatively abundant in seawater. For other nuclides, however, the concentrations of their stable isotopes in seawater are so low that it is unlikely that the stable forms of the elements involved play any significant role in ion exchange processes. At low concentrations, the concentration of the stable carrier typically does not greatly influence the amount of sorption of ^{60}Co and ^{65}Zn (Duursma and Bosch, 1970) indicating that exchange reactions with other ions dominate over ion exchange with stable isotopes of the same element.

Processes causing sorption of radionuclides on sediment particles seem to be affected by the chemical properties of the radionuclides involved and by the physicochemical and biological conditions of the sediment (pH, reduction-oxidation state, zeta-potential, bacterial activity) rather than by the bulk sediment composition. Jenne and Wahlberg (1968) studied the processes by which ^{90}Sr, ^{60}Co, and ^{137}Cs are bound to Clinch River sediments downstream from the discharge from Oak Ridge National Laboratory. They found that ^{90}Sr was associated with carbonates precipitated *in situ*. The radionuclide ^{60}Co was associated with Mn and Fe oxides, and ^{137}Cs was taken up by incorporation in the lattice of certain clay minerals. Similar mineral-related reactions, each one highly specific, probably control radionuclide uptake by marine sediments.

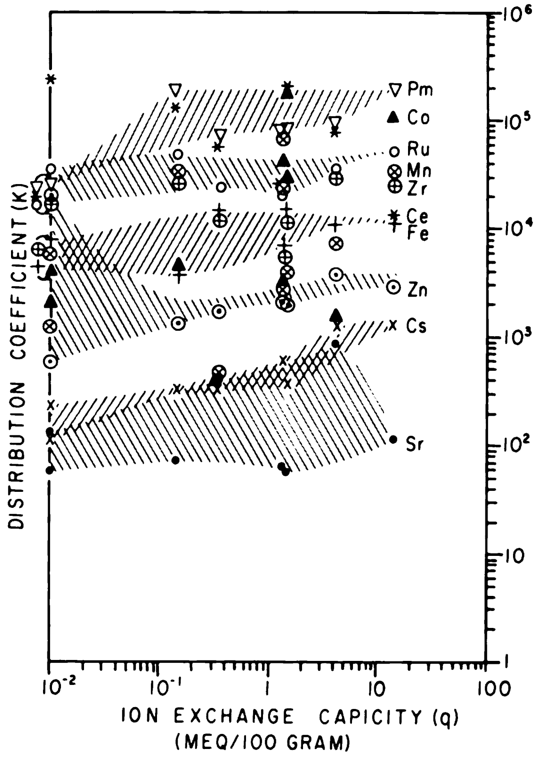

FIGURE 2 Distribution coefficients (K) for ten radionuclides and the sediments mentioned in Figure 1, as a function of calculated cation exchange capacity (q), in milliequivalents per 100 grams (meq/100 g). The cation exchange capacity is calculated as follows (Duursma and Eisma, unpublished):

$$q = \frac{n}{100}\sum k - \frac{P}{100},$$

where n = % fraction <2 μ, P = % clay mineral in the fraction of <2 μ, and k = 100 meq/100 g for montmorillonite
25 meq/100 g for chlorite and illite
10 meq/100 g for kaolinite
0 meq/100 g for other minerals.

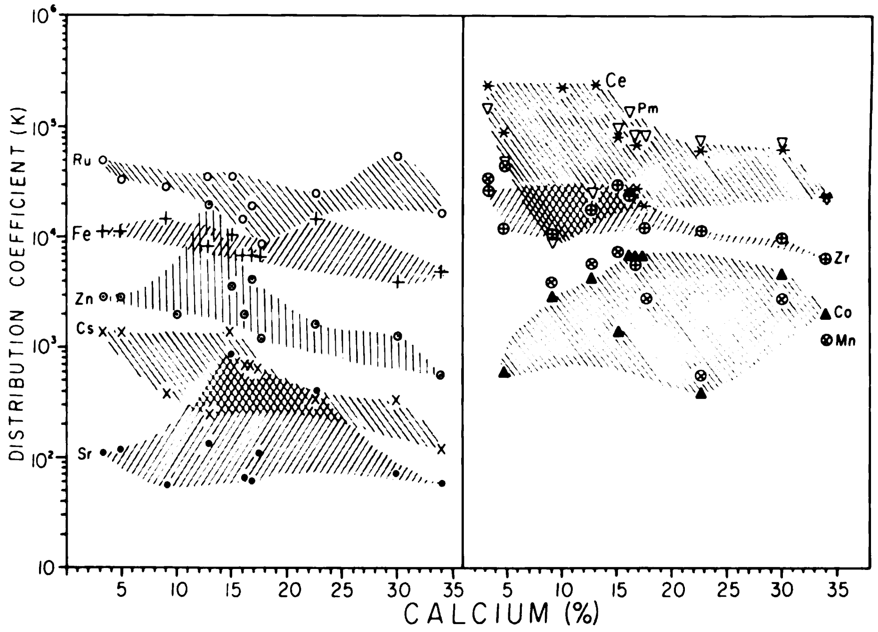

FIGURE 3 Distribution coefficients (K) for ten radionuclides and the sediments mentioned in Figure 1, as a function of the total Ca + Mg content of the sediments (Duursma and Eisma, unpublished).

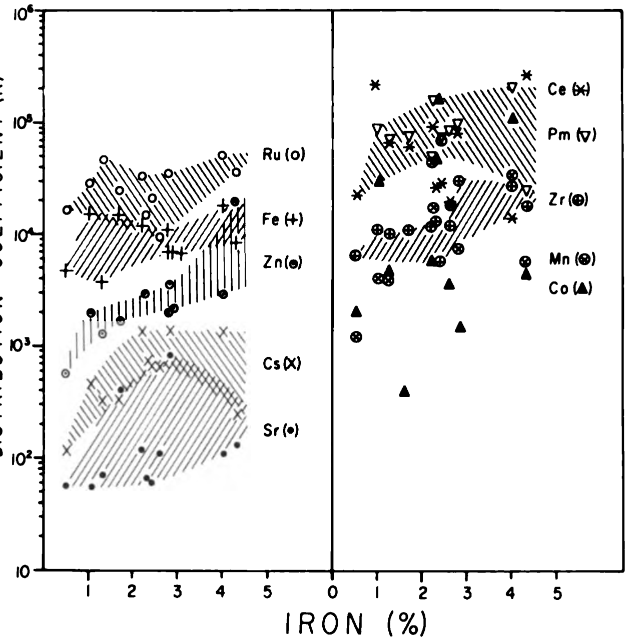

FIGURE 4 Distribution coefficients (K) for ten radioisotopes and the sediments mentioned in Figure 1, as a function of the total Fe content of the sediments (Duursma and Eisma, unpublished).

Equilibria

In sorption experiments, the apparent equilibrium distribution between seawater and sediment for different radionuclides is approached at different rates, depending on the nuclides involved. The nuclides ^{137}Cs, ^{90}Sr, and ^{65}Zn approach the apparent equilibrium much faster than do ^{59}Fe and ^{95}Zr/Nb. Cobalt-60, ^{106}Ru, ^{144}Ce, ^{54}Mn, and ^{147}Pm exhibit apparent reaction rates intermediate between these extremes. Adsorption resulting from slow precipitation may cause the slow reaction (several days) for ^{59}Fe and ^{95}Zr/Nb. Sorption by chemical binding or ion exchange may cause the relatively high reaction rates observed (several hours) for ^{137}Cs, ^{90}Sr, and ^{65}Zn (Duursma and Eisma, unpublished).

The term equilibrium implies that the reactions are reversible, but this is often difficult to demonstrate for radionuclides strongly held by sediments. The activity of radionuclides released to seawater may be so small that it is difficult to determine accurately, as in the experiments involving ^{144}Ce- and ^{147}Pm-labeled sediment and nonradioactive seawater. For ^{60}Co, it was possible to demonstrate loss of the nuclide from the sediment (Figure 5). In this case, the sorption-desorption process is due to several quasi-reversible reactions in which the rate of nuclide loss is demonstrably slower than the rate of uptake. This is explainable, in part, as resulting from a set of several processes, each with different rates, so that the overall reaction does not proceed at the same speed when it is reversed.

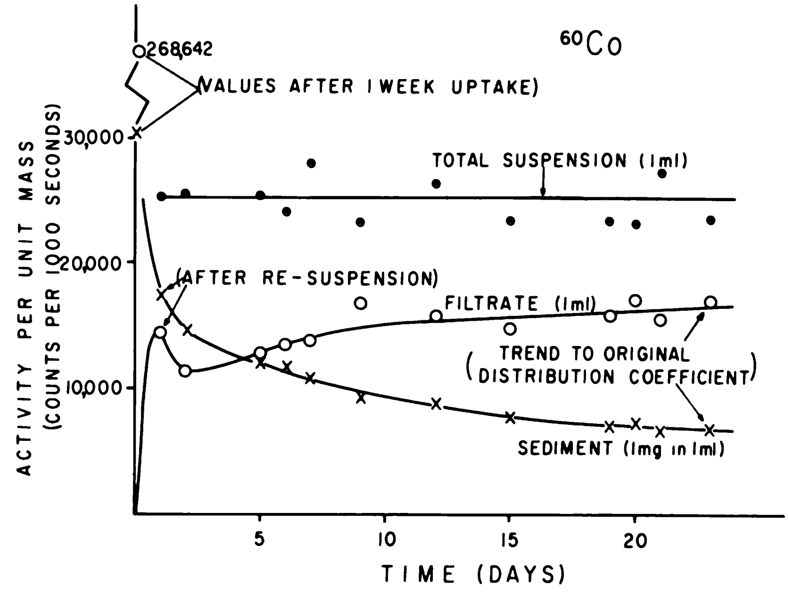

FIGURE 5 Desorption of ^{60}Co as a function of time from Mediterranean sediment, previously suspended for one week in seawater (stirring) containing ^{60}Co, and then resuspended in seawater containing no ^{60}Co (Duursma and Bosch, 1970).

Experiments on the uptake of ^{45}Ca by sediments show a stepwise set of reactions (Duursma and Bosch, 1970). The first step is relatively rapid, occurring in the first few seconds or minutes of contact. The final step is much slower—measurable only after days or weeks—and apparently involves diffusion into the sediment particles. Furthermore, there is a suggestion of an intermediate step that requires from a few minutes to a day for completion. This intermediate reaction may result from migration of the radionuclide in small capillaries between broken ends of crystal lattices or from diffusion through thin layers of organic matter coating the particles (Chave, 1965), or both.

Anomalies

Under different experimental conditions, Duursma and Bosch (1969), using the same radionuclide and the same sediment, found that it was often easier to obtain reproducible results with one radionuclide than with another. Specifically, ^{60}Co sometimes gives different results when different methods are used, a situation not encountered with other radionuclides. Even using the same sediment, the amount of ^{60}Co uptake depends on whether the sediment was in contact with quiet (unstirred, thin layer, or settling) seawater or suspended in rapidly stirred water. Each experiment came to apparent equilibrium states that were distinctly different (Figure 6). Changing the experimental conditions afterwards from quiet to stirred caused the desorption of some ^{60}Co and the establishment of another apparent equilibrium. However, a change from stirring to quiet conditions did not result in increased sorption of the nuclide. Hence, in using experimental data to predict sorption processes in nature, it is necessary to consider whether the experimental conditions approximate the expected field conditions and whether the history of the water-sediment systems was similar prior to the introduction of the radionuclide.

Anoxic Systems

Thus far, the discussion has been concerned only with sediments under oxygenated conditions. Anoxic conditions (complete oxygen depletion) are rare in open-ocean water (Richards, 1957) but moderately common in interstitial water of sediments, especially those deposited in highly productive nearshore areas. Consequently, we must consider radionuclide behavior under anoxic conditions. Experiments using Black Sea sediment under oxidizing and reducing conditions show that there is little change in the distribution coefficients of many radionuclides (Table 2). Typical

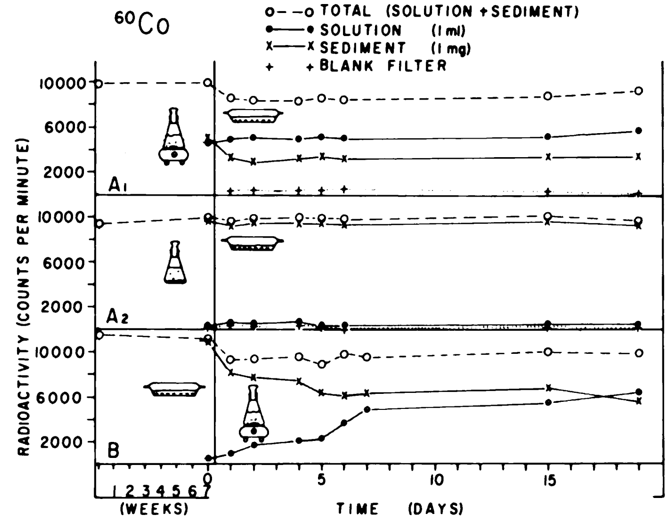

FIGURE 6 Results of a seven-week experiment in which Mediterranean sediment was exposed under various conditions to seawater containing ^{60}Co, followed by a 19-day experiment in which the conditions were changed. Note the relatively rapid adjustment to the new conditions in experiment B, but not in experiment A1 (Duursma and Bosch, 1970).

TABLE 2 Distribution Coefficients of Sorption for 10 Radioisotopes as Determined under Oxygenated and Anoxic Conditions in Black Sea Sediment[a]

Radionuclide (As Chloride)	Distribution Coefficients	
	Oxygenated[b]	Anoxic[c]
^{90}Sr	1.2×10^2	4.7×10^3
^{137}Cs	1.4×10^3	3.1×10^3
^{106}Ru (nitrate)	3.4×10^4	9.0×10^3
^{59}Fe	1.2×10^4	4.0×10^3
^{65}Zn	3.0×10^3	3.2×10^4
^{60}Co	6.0×10^3	1.4×10^4
^{147}Pm	4.8×10^4	5.2×10^3
^{54}Mn	4.6×10^4	4.3×10^3
$^{95}Zr/^{95}Nb$	1.2×10^4	9.0×10^3
^{144}Ce	9.0×10^4	1.2×10^4

[a] Duursma and Eisma, unpublished data.
[b] Dissolved oxygen concentration: 5 ml O_2/liter.
[c] Hydrogen sulfide concentration: 2-3 mg H_2S/liter.

changes are reduction of Fe^{+3} to Fe^{+2}, which is more soluble even in the presence of H_2S, and the possible precipitation of various metals, such as Zn, as sulfides. (See Chapter 5, for further discussion of this point.)

Marine sediments in many areas have an oxygenated surface layer a few millimeters to a few centimeters thick, which probably is formed by O_2 diffusion, burrowing of marine organisms, and mixing due to currents and waves. Below this surface zone, dissolved oxygen in interstitial water is depleted. Sediment in the lower zone is thought to represent the sediment that is below the level of reworking by currents and by physical and biological phenomena, while the upper zone is essentially in transit (Hayes, 1964; Rhoads, 1967).

Under reducing conditions, the distribution coefficients are lower (Table 2) for ^{106}Ru, ^{59}Fe, ^{147}Pm, ^{54}Mn, $^{95}Zr/Nb$, and ^{144}Ce. Consequently, these radionuclides dissolve in the reduced zone and then move into the overlying oxygenated zone; other radionuclides (^{90}Sr, ^{65}Zn, and ^{60}Co) are less soluble under reducing conditions, and a reverse transport may be expected.

Results similar to these for stable Fe, Mn, Co, and Zn have been reported by Sevastyanov and Volkov (1967), Brooks *et al.* (1968), and Presley and Kaplan (1968), based on the distribution of trace elements in interstitial waters from marine sediments. In many reactions, the different chemical behaviors under reducing conditions result from formation of different complexes rather than from direct reduction of the element.

VERTICAL MOVEMENT OF RADIONUCLIDES IN MARINE SEDIMENTS

Problems

Sediment deposits contact the overlying seawater through the sediment-water interface, a two-dimensional surface that does not facilitate efficient uptake by sediments of radionuclides from seawater. Consequently, radionuclides are added to sediment by the deposition of particles with associated radionuclides or, in some circumstances, by movement of radionuclides through the interface into (or out of) the sediments. Thus, it is necessary to consider movements of radionuclides within the sediments.

Large distribution coefficients for common radionuclides associated with sediments and sedimentary particles indicate that migration of radionuclides in these deposits cannot be very rapid. Rates of nuclide movement are at least partially determined by whether nuclides sorbed by sedimentary particles can be released to the interstitial seawater or whether the nuclide remains associated with the particles. Diffusion of radionuclides into particles themselves further decreases the probability of subsequent radionuclide migration. However, although the rate of radionuclide loss from particles is slower than the rate of uptake (Figure 5; Pomeroy *et al.*, 1966), radionuclides associated with particles on or very near the water-sediment interface may redissolve in the overlying water as well as diffuse deeper in the sediment.

Diffusion

Two types of movement of radionuclides on the ocean bottom may be distinguished. First, and perhaps simplest, is diffusion of radionuclides into (or out of) the sediment owing to concentration gradients in the interstitial waters or overlying seawater, in combination with sorption and desorption on the sediment particles. Second, and more complicated, are the diffusive processes where the medium itself may be moving. Laboratory and field experiments were made of the diffusion processes, using ^{36}Cl ions that were not sorbed by the sediment particles (Duursma and Bosch, 1970). These results, combined with observed distribution coefficients of sorption, were used to calculate diffusion coefficients for the same sediments and radionuclides discussed earlier. The results obtained for ^{36}Cl agree with those (about 10^{-6} cm^2/sec) obtained by Shiskina (1966) using a different approach and *in situ* measurements. The ^{90}Sr values are in satisfactory agreement with those (about 10^{-8} cm^2/sec) obtained for ^{226}Ra (Koczy and Bourret, 1958). Chemical composition of interstitial waters (Horne, 1969, p. 395–400) also plays an important role in controlling either radionuclide bonding to sediment particles or formation of soluble ligands.

The results of these experiments and calculations indicate that diffusion in sediments is extremely slow for most radionuclides. This can be seen from Figure 7, which shows the time required for the movement of a 10 percent concentration front from a constant source. Note also that the sequence of diffusion coefficients is the reverse of the sequence of the distribution coefficients (page 148). In other words, the stronger the binding of a particular radionuclide to a sediment, the smaller the diffusion coefficient and the slower the diffusion process.

Current and Wave Action

Currents and wave action cause substantial mixing of sediment, both vertically within a given sediment mass and horizontally, so that sediment-associated radionuclides may go into suspension from an initial depositional site and be moved substantial distances before final deposition. The upper few centimeters of cores often contain radionuclide

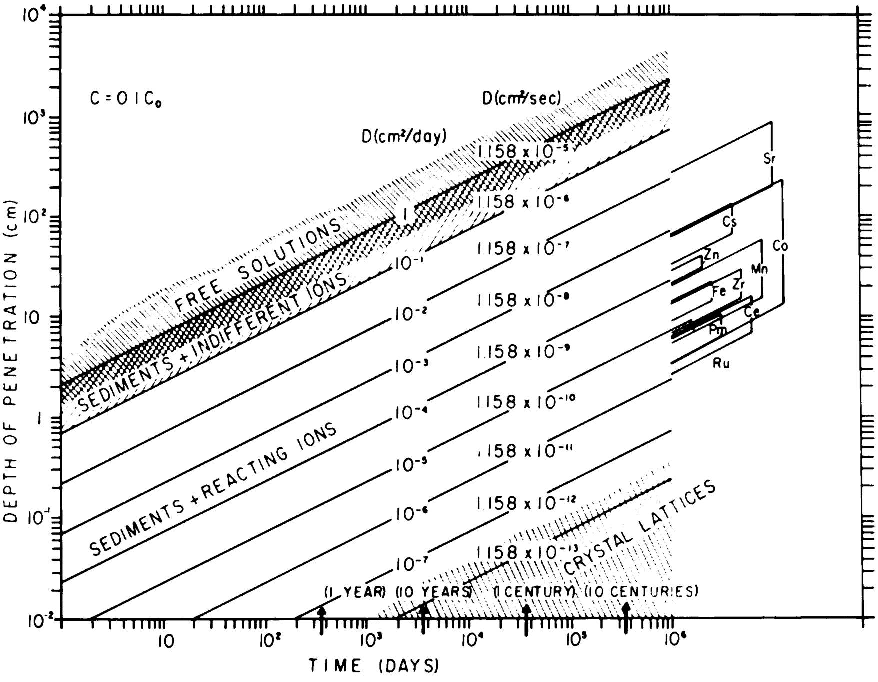

FIGURE 7 Depth penetration of a 10 percent concentration front into a sediment from an overlying water mass with a constant radionuclide concentration, as a function of time, for various diffusion coefficients. The range of diffusion coefficients found for various elements and marine sediments is shown at the right.

concentrations that are aberrant with respect to those at greater depths in the cores (Barnes and Gross, 1966; Jefferies, 1968). This may be caused by physical processes, by biological activity, or possibly by disturbance of the water-sediment interface during the sampling operation, if special care has not been taken to avoid such disturbance.

Biological Activity

Activity of burrowing organisms probably causes much vertical movement of radionuclides in shallow-water sediments. Burrowing by benthic organisms in subtidal muds causes the reworking of the upper 2 or 3 cm several times annually; the upper few millimeters is probably cycled daily. In subtidal sands, burrowing occurs to a depth of 30 cm (Rhoads, 1967). Barnes and Gross (1966) suggested that biological activity could account for the apparent rapid mixing of radionuclides in the upper 2 cm of several cores taken near the Columbia River mouth.

Mixing due to biological activity can be treated as a diffusive process, and a biological diffusion coefficient can be calculated. Using data from Haven and Morales-Alamo (1966), who studied the depth distribution in sediment of fluorescent particles, a diffusion coefficient of about 10^{-6} cm²/sec can be calculated, a result of diffusion plus motion of the medium.

In the Irish Sea near the Windscale discharge outlet (Templeton and Preston, 1966), mixing by benthic organisms may be the cause of ^{106}Ru penetration in the sediments to depths of 25 cm below the interface (Figure 8). Assuming that the concentration in the sediment is the integral of continuous diffusion processes from instantaneous sources (deposits on top of the sediment) and taking into regard the decay of ^{106}Ru, the depth of penetration indicates an apparent diffusion coefficient of about 10^{-7} cm²/sec, which is much closer to an effect predicted from biological activity than to diffusion, where the coefficient for ^{106}Ru should be about 10^{-11} to 10^{-12} cm²/sec (Duursma and Bosch, 1969).

Interstitial Water Movement

Upward movement of water through the water-sediment interface is still little known. However, sediment accumulation does cause compaction with a resulting upward movement of interstitial water. In the case of diffusion, this effect will be less than that of burial by sediment accumulation (see below). Probably a more important mechanism is the movement of water through the sediment owing to groundwater discharge from submarine aquifers on the continental margins. Evidence for submarine discharge from both fresh and brackish waters has been found as much as 120 km off the Florida coast, as well as elsewhere on the continental shelf and slope (Manheim, 1967). Such submarine discharges are especially likely off land areas with high rainfall that are underlain by suitably permeable aquifers (Kohout, 1967). Considering the slow rates of diffusion, it seems probable that such upward movement of water through the sediment-

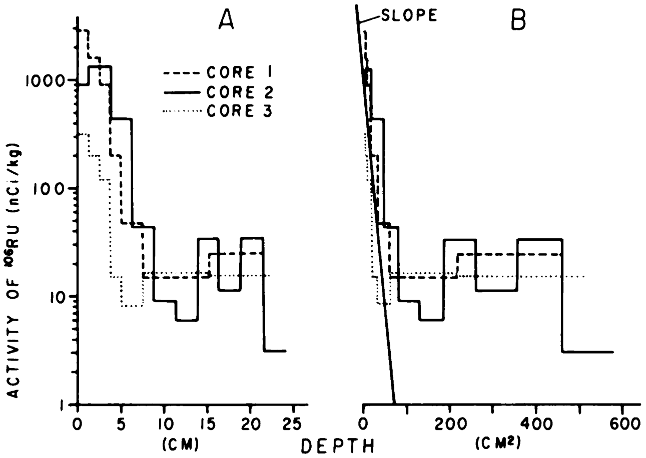

FIGURE 8 Concentration of ^{106}Ru in sediment cores from the Irish Sea (Templeton and Preston, 1966) (A) as a function of depth below the sediment-water interface and (B) replotted as a function of the square of the distance. The slope of the line in B indicates an apparent diffusion coefficient of 10^{-7} cm^2/sec. (Calculated according to Duursma and Hoede, 1967.)

water interface could be an important process moving radionuclides from the sediment back into the overlying water.

Another possibility in areas with high organic debris in their sediments is gas production (Reeburg, unpublished Ph.D. thesis, The Johns Hopkins University), which may cause partial mixing of interstitial water, resulting in redistribution of radionuclides.

Burial by Sediment Accumulation

If sediment accumulation is fairly rapid, a single layer of contaminated sediment will be mixed with or buried by later sediment deposits, thus inhibiting exchange of radionuclides across the sediment-water interface. Under such circumstances and where current, waves, and biological activities are almost absent, sediments can assimilate larger amounts of radionuclides than in areas where the rate of sediment accumulation is much lower. Figure 9 indicates the relative importance of diffusion of a 10 percent concentration front from a thin contaminated sediment layer and the competing process of burial by later sediment deposits. Since diffusion does not proceed uniformly, the answer to whether the sedimentation or the diffusion is faster depends on the time scale of the process. The diagram (Figure 9) gives a rough indication for periods of 1 to 10^7 years.

For most of the deep-ocean floor where sediment accumulates at rates of about 1 mm per thousand years (Sverdrup *et al.*, 1942) and for the 70 percent of the world's continental shelf not covered by sediment deposited in the past 3,000-5,000 years (Emery, 1968), diffusion is probably the dominant process. In these areas, loss and exchange of radionuclides from the sediments to the overlying seawater will be primarily dependent on the reactions of the radionuclides.

On the shallow continental shelf near major rivers, where sediment is accumulating relatively rapidly, the radioactive sediment may be deposited rapidly enough that it is not substantially disturbed by physical processes (currents and wave action) and before it can be extensively burrowed by benthic organisms. Under such conditions, thin contaminated sediment layers may be covered. In parts of the northern Adriatic and Ligurian seas, such sediment layers containing fallout nuclides (^{90}Sr, ^{144}Ce, ^{147}Pm, ^{155}Eu) deposited during the period 1958-1965 were detected (Schreiber, 1966; Schreiber *et al.*, 1968; Cerrai *et al.*,1967; Albini *et al.*, 1968), where depth below the interface is proportional to the time since deposition (Figure 10). Considering that the diffusion coefficient of ^{90}Sr (see Figure 7) is about 10^{-8} to 10^{-7} cm^2/sec and that the diffusion coefficient for the three rare-earth metals is 10^{-11} to 10^{-10} cm^2/sec, and further that the deposition rate is 2 cm/year in the North Adriatic, it can be concluded from Figure 9 that for ^{90}Sr the diffusion should have been "faster" during the eight years of deposition of radioactive sediment but that the rare-earth radionuclides ^{144}Ce, ^{147}Pm, and ^{155}Eu have been covered up.

Where low dissolved-oxygen concentrations occur seasonally in near-bottom waters, layers formed by individual phytoplankton blooms and varves formed by seasonal changes in sediment sources can be distinguished (see Gucluer and Gross, 1964, for an example of sediment accumulation under such conditions). Consequently, one might expect to find sediment layers containing distinctive radionuclides from a single event, such as a period of unusually large amounts of atmospheric fallout. In oxygenated waters such distinctive layers would be quickly mixed with other nonradioactive sediment by burrowing organisms.

RADIONUCLIDE TRANSPORT BY SEDIMENTS

Our consideration of radionuclide transport by sediments, though restricted to the continental shelf and near-shore waters, may have applications to deeper waters. Sediment-associated radionuclides are more likely to prove troublesome in near-shore waters, either through direct contact with humans or through uptake by food organisms, especially filter-feeding organisms. Among potential depositional sites are beaches, estuaries and their tidal flat areas, and open continental shelves.

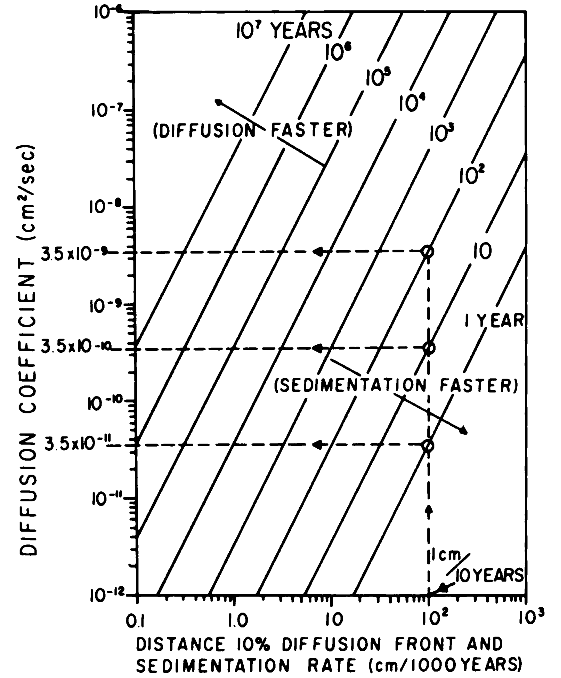

FIGURE 9 Relation between the distance at which 10 percent of the concentration of a thin contaminated sediment layer may be found and the diffusion coefficient, for different times after the instantaneous contamination of this layer. Examples follow:

1. If the sediment is covered by a layer of newly deposited sediment in a certain time (say 10 years at sedimentation rate 1 cm/10 years), then the diffusion is regarded as "faster" (based on 10 percent front distance) than the sedimentation for radionuclides having a diffusion coefficient higher than 3.5×10^{-10} cm²/sec.
2. For the same sediment accumulation rates, the diffusion coefficients for 1 and 100 years are 3.5×10^{-11} cm²/sec and 3.5×10^{-9} cm²/sec, respectively.

Note the agreement for the Po river mouth (see Figure 10), where the sedimentation rate is 2 cm/year; for periods of 1 to 10 years sedimentation dominated because the diffusion coefficients of the radionuclides involved were smaller than 3×10^{-9} cm²/sec (except for ^{90}Sr).

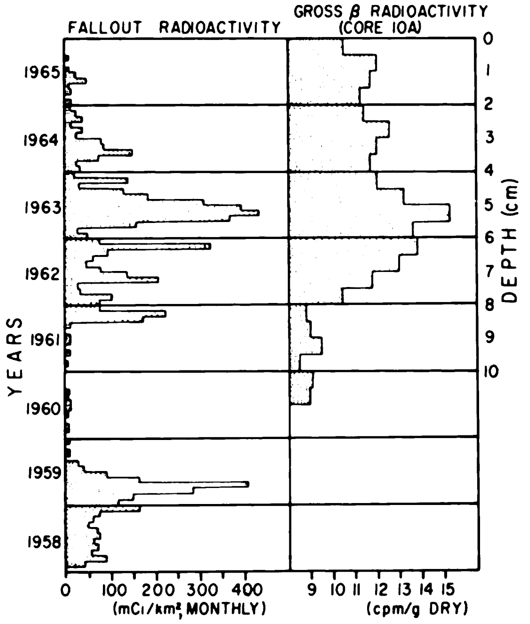

FIGURE 10 Variation of gross beta activity from atmospheric fallout observed at Ispra, Euratom Centre, Italy, and gross beta activity in sediment from a core collected in the Adriatic Sea near the Po river mouth in 1966 (Schreiber *et al.*, 1968). The apparent rate of sediment accumulation is approximately 2 cm/year.

Beaches

Sediment deposited on beaches is more or less continually stirred by wave action. As a result, the sediments typically are well-sorted sands with grain sizes of 100 to 200 μ (Postma, 1967a and b; Jefferies, 1968). Although there is often a pronounced landward movement of water along the bottom (Morse *et al.*, 1968), there is commonly relatively little artificial radioactivity associated with sands, even near a major discharge of radioactivity (Templeton and Preston, 1966). The low level of activity of the radionuclides is probably the result of the lack of fine-grained particles (see Table 1) and the continual abrasion of the grain surfaces. Sediment may be moved fairly rapidly in the area between the breakers and the beach when incoming waves strike the beach at an angle (Galvin, 1967). Longshore currents transport substantial quantities of sediment, and near discharges of radioactive effluent, this may be an important route of nuclide movement (Templeton and Preston, 1966).

Estuaries

Estuaries act as sediment traps (Postma, 1967a and b). A relatively small fraction of riverborne sediment or sediment moving along the coast escapes in a short time over most of the world's continental shelves (Emery, 1968). Instead, large quantities of fine-grained sediment (<100 μ) accumulate, at least temporarily, in the estuaries or on tidal-flat areas (Postma, 1967a and b), where the fine particles are passed in large quantities through filter-feeders (Verwey, 1952), some of them important as food resources. In an estuarine system, sediments are susceptible to resuspension and movement by tidal or river currents (Conomos, 1968). During major floods or periods of strong wave action, sediments are washed out from the estuaries to be deposited on the continental shelf. Where there are river deltas, the fine-grained sediment ($<50\ \mu$) can remain suspended near the point where the near-bottom flow changes from a net-downstream direction in the river to a net-upstream flow in the estuary (Postma, 1967a and b).

Klingeman and Kaufman (1965) demonstrated deposition of riverborne sediment-associated radionuclides in San Francisco Bay. Radionuclides bound to river sediments may be desorbed when they enter the sea. De Groot (1966) and Eisma (1968) showed that Fe in suspended matter was desorbed at low salinities and later was precipitated in seawater.

Continental Shelves

Information about sediment movement on continental shelves comes from interpretation of the distribution of various sedimentary characteristics. Studies of radioactive sediment dispersal on the continental shelf provide some of the best information about actual sediment movement in this area. Large amounts of radioactive materials discharged from the Windscale plant on the Irish Sea (Templeton and Preston, 1966) and the Columbia River discharge from the Hanford plutonium-producing reactors (Gross, 1966) provide enough tracer materials to permit sediment movements to be followed over substantial distances. In both areas, the radioactive sediment moved generally parallel to the coastline.

Near-bottom currents on the continental shelf are little known, but they probably play a major role in transporting sediment-associated radionuclides. From the few studies of near-bottom currents available, it appears that near-bottom currents tend to parallel the adjacent coast except near major estuarine systems, where near-bottom currents flow toward the estuary (Bumpus, 1965; Templeton and Preston, 1966; Rehrer *et al.*, 1967; Morse *et al.*, 1968). On the Atlantic coast of the United States, near-bottom currents are rather similar to the surface currents, setting essentially southward at about 0.5 km/day (Bumpus, 1965). On the Pacific coast of the northwestern United States, the near-bottom currents generally set toward the north, although the variable surface currents set southward for about six months a year (Morse *et al.*, 1968). Reported speeds of seabed drifters, devices used to study near-bottom currents, are about 1-2 km/day. Although such studies indicate probable directions of sediment movements, they do not indicate the speed. Sediment-associated radionuclides, however, may be used to determine apparent speeds of movement of radioactive particles (Gross and Nelson, 1967) under certain conditions.

Submarine Canyons

Submarine canyons appear to be the major routes of sediment movement across the continental shelf (Shepard, 1965). Where rivers bring large amounts of sediment to the ocean, frequent turbidity currents seem to empty the nearby canyons and carry large amounts of sediment onto the deep-ocean floor. Where continental shelves are wide, submarine canyons appear to be inactive at present. Where shelves are narrow and the canyons head close to shore, they appear to have sediment continually moving through them by creep, sand flows, and slumps (Shepard, 1965).

Except for a report of ^{65}Zn and ^{51}Cr in sediment in a submarine canyon near the Columbia River (Osterberg *et al.*, 1963), there have been no reports of deep-ocean accumulation of sediment-associated nuclides from the various coastal discharges of radioactivity.

Arctic Areas

Movement of sediment-associated radionuclides in high latitudes, where transport by ice may be involved, has not been included in our discussion largely because of the lack of available information about the behavior of sediments under those conditions. However, it is known that glacially derived clays in seawater strongly sorb many common radionuclides (Duursma and Eisma, unpublished).

SCAVENGING ABILITY OF SEDIMENTS

In areas where sediments are resuspended by tidal currents, wave activity, and river currents, there will be high concentrations of particles in the water. The sorption capacity of these particles for dissolved radionuclides may cause scavenging and deposition of the radionuclides on the bottom. Removal of radionuclides from the water will depend on the distribution coefficients of the radionuclides and particles involved, the rates of sorption, and the settling velocities of the sediment particles.

Because of their settling velocities, sediment particles will have a relatively short time of contact with the water for sorption of soluble nuclides to occur. Thus, complete sorption equilibrium between particles and water may not be achieved. This, however, is probably not too significant, because the first step in the sorption process is relatively rapid, and for most of the radionuclides at least 80 percent of the equilibrium is achieved in several hours.

Using data on distribution coefficients presented in this chapter, it is possible to calculate the effect of scavenging for a given concentration of sediment settling through a

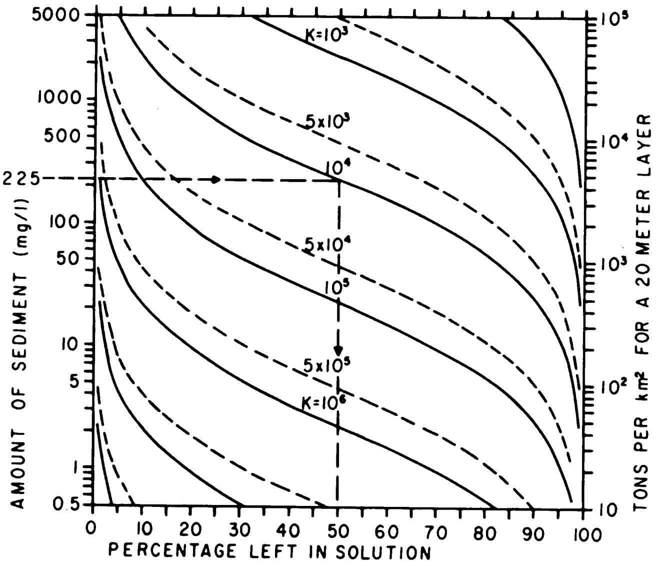

FIGURE 11 Predicted relationship between the fraction of the original radionuclide concentration remaining in solution as a function of the amount of sediment that has settled out from suspension, for different distribution coefficients. The assumed dry sediment density is 2.27 g/ml.

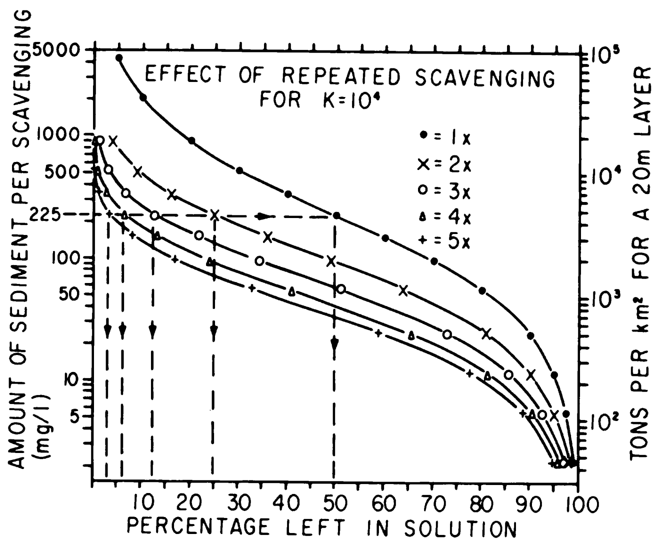

FIGURE 12 The effect of five successive scavengings by equal amounts of sediment with a density (dry) of 2.27 g/ml for a radionuclide with a distribution coefficient of 10^4.

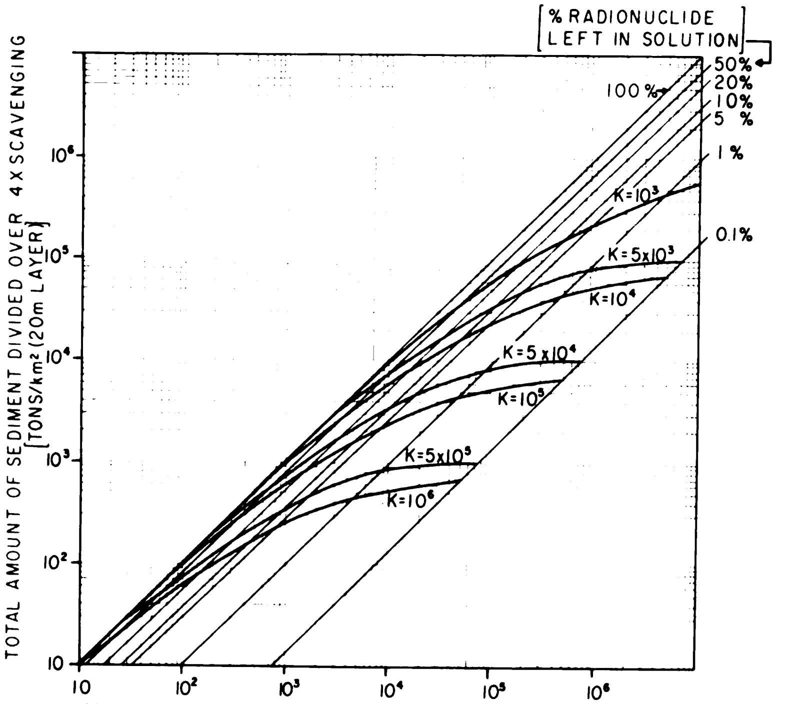

FIGURE 13 The relation between the amount of sediment necessary to obtain the same effect of scavenging, if divided into four parts or if used at once, for different distribution coefficients (density of dry sediment 2.27 g/ml). Abscissa indicates the amount of sediment if used only once; ordinate, the total amount of four successive treatments. Note that scavenging dividing up into subsequently smaller amounts has less effect when the quantities are too small (where the full lines approach the 45° line).

contaminated layer of seawater. Figure 11 is a model in which the amount of radionuclide remaining in seawater after settling of the particles is plotted as a function of the concentration of suspended sediment for different distribution coefficients. For example, 225 mg (dry weight) of sediment per liter of seawater with a distribution coefficient of 10^4 for the radionuclide involved will reduce the concentration of the radionuclide by 50 percent. Repeated suspension and settling of smaller amounts of sediment will remove more of such radionuclides than a single release of an equivalent total amount of sediment (shown by the dotted lines in Figure 12; see also Figure 13).

It must be remembered that the model is based on preliminary results using a relatively small number of laboratory experiments; the postulated removals have not been tested experimentally under natural conditions. Scavenging is known to occur, however, under natural conditions—rough weather in the Irish Sea near the Windscale pipeline outlet causes more rapid removal of radioactivity from the water and deposition in the bottom sediments (Templeton, personal communication).

This model is presented as an example of possible uses of sediments or other solids to remove waterborne pollutants such as radionuclides, causing them to be deposited on the bottom. Whether it is desirable to do this rather than depending on natural dilution and burial processes depends on local conditions and other factors. The results of these calculations suggest that such applications of sediments would be restricted to dealing with accidents or other releases in coastal areas such as bays, lagoons, harbors, rivers, or lakes (Duursma, 1969).

SUMMARY

Sediments and sedimentary particles have a substantial capacity to remove radionuclides from seawater. The particles may be deposited on the ocean bottom or picked up by filter-feeding organisms that may concentrate the radionuclides and pass them into man's food supply.

Certain radionuclides are more strongly sorbed on sediment particles than others. Mineral-related reactions, each with a degree of specificity, appear to be controlled by chemical species of the radionuclides and the physicochemical conditions of the sediments and sedimentary particles. In some laboratory experiments, the amount of radionuclides taken up by sediments and sedimentary particles depends on the history of the sediment–water system prior to the introduction of the radionuclide; this makes it difficult to predict radionuclide behavior in sediment-laden water.

Radionuclides may move through the sediment after deposition. Among the processes by which radionuclides move are diffusion through the interstitial water, movement with the interstitial water expelled from the sediment by compaction, and groundwater discharge through the sea bottom. Where sediment accumulation is sufficiently rapid, the radioactive particles may be covered by later sedimentary deposits before these relatively slow movements can move the nuclides through the sediment–water interface. On a large part of the continental shelf and on much of the deep-ocean bottom, rates of sediment accumulation are slow enough that diffusion processes may be dominant.

Sediment particles and their associated radionuclides may be mixed or moved by burrowing activity of benthic organisms, near-bottom currents, wave action, turbidity currents, longshore currents, or by ice.

Sediments or sedimentary particles can remove radionuclides from seawater and deposit them on the sea bottom. Calculations have been given to indicate the effect of such applications of sediments, and criteria have been presented to guide the selection of materials to be used in such applications.

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Chapter Seven

ACCUMULATION AND REDISTRIBUTION OF RADIONUCLIDES BY MARINE ORGANISMS

F. G. Lowman, T. R. Rice, F. A. Richards

The transport and distribution of radionuclides introduced into the sea are dependent upon the interactions of the water, organisms, and sediments with the added material and with each other. The organisms, detritus, and sediments accumulate several elements to levels many times greater than those in seawater and may influence the distribution of the corresponding radionuclides in areas of high biological activity or rapid sedimentation.

In the open sea, the bottom sediments are far removed from most of the vertical water column, and the interaction of these sediments with surface-introduced material may be limited almost entirely to their constituting a floor for insoluble particles carried down by gravity. In the surface waters of these areas, the biota, too, may provide insignificant mass and exert only minor influences upon the transport of most trace elements.

In regions of upwelling and in shallow near-shore areas, however, biomass is often relatively great. The influence of the organisms upon the distribution patterns of some trace elements or radionuclides added from land runoff or from direct fallout may be significant. In the shallow near-shore areas, sediments and suspended particulate matter may alter biological influences. The high degree of adsorption by these sediments may, however, be partly due to biological activity of the attached periphyton.

In marine ecosystems, the plants and animals are in direct contact with trace elements in their environment, thus allowing direct transfer of elements as ions, colloids, or particulates at the phase boundaries between the hydrosphere and biosphere. This close relationship may result in rapid and strong mutual interactions between the organisms and the environment when fluctuations of biomass or abundance of trace elements or radionuclides occur. Thus, when other factors are not limiting, the abundance of available nutrients controls the biomass. Large increases in biomass may, in turn, deplete some nutrient elements from localized areas. Not only may marine organisms alter the distribution of stable elements normally present in marine waters, but they may also influence the seasonal, horizontal, and vertical distributions of radionuclides and stable elements introduced into the sea by man. These interactions should be considered in studies of the effects of biological activity upon the distribution patterns of radionuclides and trace elements in the sea.

The sources of radioactivity and the inorganic interactions of the radionuclides and associated stable elements with the sea have been reviewed in detail in Chapter 2 and will be discussed here only with respect to their influences upon the availability of radionuclides to marine organisms. Biological processes that result in the passage of radionuclides through food webs to man will be considered, as well as the relative importance of physical and biological transport on the distribution patterns of radionuclides introduced into the marine environment.

INFLUENCE OF NONBIOLOGICAL FACTORS ON THE AVAILABILITY OF RADIONUCLIDES TO MARINE ORGANISMS

Source of Radionuclides

The availability of radionuclides to the marine biota depends largely upon their chemical and physical forms and the rates and modes of addition to the marine environment. Most of the nuclear detonations thus far have occurred under oxidizing conditions. Devices exploded on or near the surface of land incorporated large amounts of coral, water, device material, support structures, and air into the fireball, delivering large amounts of oxides. In marine areas receiving large amounts of fallout, precipitation of calcium and ferric hydroxides effectively scavenged some radionuclides to deeper water, out of reach of the plankton and pelagic animals but in direct contact with the benthic animals. Tower or air bursts, in contrast to surface detonations, introduced only small amounts of contaminants into localized marine areas but contributed a majority of the radionuclides in oxidized form in worldwide fallout. The total weight of the stable material in this fallout was only 1,000 to 10,000 times the weight of the radionuclides, and this extremely small amount of material would not be significant in scavenging radionuclides from the surface waters. Thus, these radionuclides would be available to the organisms in the upper waters and would be subject only to limited transport to the bottom.

Over long periods of time, the insoluble radionuclides associated with oxides of iron and manganese would sink, however, as a result of their being incorporated into fecal pellets by filter-feeding zooplankton. Manganese-54, whose stable counterpart occurs as the Mn^{+2} ion in seawater, has been reported by several investigators to exist in seawater mainly as particulate manganese dioxide (Slowey *et al.*, 1965; Schelske *et al.*, 1966). Filter-feeding sponges have been observed to accumulate ^{54}Mn in preference to stable Mn in the sea, whereas benthic algae from the same area accumulate their manganese mainly from solution (Lowman *et al.*, 1967a). Ruthenium-103 and ruthenium-106 in worldwide fallout also is present in particulate forms at ratios 4 to 13 times those for stable ruthenium in seawater (Dixon *et al.*, 1966).

Nuclear explosions under the surface of the sea also occur under oxidizing conditions, partly because of dissolved oxygen in the water. Although the metal in the device and the support sources would provide less mass than the sodium chloride in the vaporized seawater, the centers of nucleation would be provided by the oxides of iron, calcium, and magnesium.

Underwater detonations in shallow bodies of water may produce sufficient heat to convert interstitial water of the bottom sediments into steam, dislodging sediments that may be incorporated into the fireball. A detonation of this type on a calcium carbonate bottom of a lagoon resulted in little or no precipitation of ^{89}Sr by the suspended calcium oxide and hydroxide. Although this radionuclide would be expected to be coprecipitated, it remained in the soluble fraction. During the time of calcium precipitation, the precursors of ^{89}Sr (4.4 sec ^{89}Br, 3.2 min ^{89}Kr) constituted the major isotopes of fission chain 89, and only a small fraction of the ^{89}Sr was available for coprecipitation. Thus, the chemical characteristics of the precursors of a long-lived fission product at the time of detonation influence the availability of the radionuclides to the biota.

Underground nuclear explosions, such as those tested for excavating, differ in several ways from detonations in the air, on the surface of the ground, or in the water. Underground detonations (a) introduce large amounts of stable elements from the vaporized and pulverized rocks into the fallout, which could cause significant precipitation and coprecipitation of radionuclides as well as isotope dilution in seawater; (b) reduce the amounts of radionuclides reaching the sea, since a major fraction of the total radioactivity would be adsorbed onto the surfaces of the material that falls back into or near the lip of the excavation (the fallout and cloud would contain only 5 to 10 percent of the radioactivity); (c) produce a reducing environment as a result of the presence of vaporized metal from the device, support structure, and the casing of the drill hole, electrons from fission, hydrogen from fusion, and elementary carbon, hydrogen, and native metals in the vaporized geological material.

Several elements, ^{54}Mn and ^{56}Mn, for example, would descend in fallout from underground detonations in a partly or completely reduced state and would exhibit increased solubility when added to seawater, compared to the oxidized forms. The behavior of radioisotopes of tungsten also appears to be influenced by this type of detonation. Radiotungsten produced under oxidizing conditions was complexed tightly to the waxy leaves of land plants; however, no foliar or root uptake into the plant could be detected (Lowman, 1960). In an underground detonation at the Nevada test site, Romney *et al.* (1967) observed a high degree of uptake of radiotungsten produced under reducing conditions through the roots but reported no chelation onto the leaves. The distribution in the sea of radiotungsten produced under oxidizing conditions suggested that a physical or chemical change occurred after its introduction into the water. The radiotungsten constituted about 50 percent of the total radioactivity in the water and plankton of an open-sea area about 180 miles in diameter. About half of the tungsten in the plankton (25 percent of the total radioactivity in the area) was associated with silica, probably the skeletons of marine diatoms. During a second survey of the same body of water three weeks later, all of the radiotungsten had dissociated from the plankton but was still present in the water (Lowman *et al.*, 1959).

Environmental Factors

Environmental factors may vary geographically and seasonally. Biological productivity is usually higher near the land than in the open sea, and a greater fraction of the total radionuclides might be expected to be associated with the biota in the near-shore regions. However, other mechanisms may operate near the shore to reduce the availability of individual radionuclides to the plants and animals. These include fractionation of radionuclides during runoff from the watersheds, isotope dilution, chemical precipitation and coprecipitation, adsorption by suspended and bottom sediments, and vertical and horizontal diffusion.

Estuarine and other near-shore areas receive variable amounts of river runoff from the adjacent land mass. Fractionation of radionuclides of different elements in water draining from watersheds occurs because of differences in solubility, particle size, and adsorptive characteristics of the individual radionuclides and stable elements. Radionuclides that are easily dissolved or are not strongly adsorbed to the larger solid particles occur in the runoff in large amounts compared with the insoluble or strongly adsorbed elements. Radionuclides present in enhanced amounts in the runoff will, however, usually be accompanied by the corresponding stable element. In these cases, isotope dilution may reduce the specific activity* of the radionuclides in the marine organisms near the outflows of rivers to levels below those occurring in the offshore animals and plants contaminated by direct fallout. Although the specific activity may be lower in the near-shore organisms, they may contain greater total amounts of element and thus more radionuclide (Duke, 1967). In these cases, the total ash content of plants and animals living in areas of heavy runoff from the land may be two to three times greater than that in the same species from other areas (Lowman *et al.*, 1967a).

Radionuclides added to the sea from river outflows are diluted by the corresponding stable elements normally present in seawater. In areas of upwelling, relatively large amounts of trace elements are supplied to the surface from the deeper layers. Phytoplankton, zooplankton, and omnivorous fish collected in these areas often contain smaller amounts of fallout radionuclides than organisms of the same groups collected in other marine areas where upwelling does not occur (Avila, 1969).

In areas of mixing of fresh water and seawater, radionuclides in ionic form, adsorbed onto suspended river sediments, may be released into the estuaries as a result of the presence of electrolytes in seawater (Schutz and Turekian, 1965a). Colloids of iron and aluminum may simultaneously be precipitated and act as scavengers for radionuclides, including ^{55}Fe, ^{59}Fe, ^{65}Zn, ^{54}Mn, ^{95}Zr, ^{144}Ce, ^{147}Pm, ^{106}Ru, and ^{110}Ag. In areas of extensive scouring and resuspension of bottom sediments by tidal currents, several radionuclides introduced by river waters may be adsorbed onto sediment surfaces. Some of the adsorption may be due to inorganic processes, but significant amounts of added radionuclides and trace elements may be bound to sediment surfaces by the periphyton (Pomeroy *et al.*, 1965).

Radionuclides introduced into the surface waters of the open sea become distributed in the upper mixed layer. Between 50°N and 45°S, the oceans usually consist of an upper "warm-water sphere" separated by a vertical transition zone of rapid temperature and density change from a deeper "cold-water sphere" that reaches to the bottom of the sea (Dietrich, 1963). The warm upper waters vary in depth but are often about 100 m thick and are rapidly mixed by winds. In this layer the temperature, salinity, and density are nearly uniform. A shear zone often exists just below the mixed water in the layer of rapid change in density. This layer, the pycnocline, constitutes a barrier for the downward movements of soluble material from the upper mixed layer or for the upward movement of dissolved elements in the deeper waters (Revelle, 1956; Lowman, 1960).

Radionuclides introduced into the sea exist as solutes, colloids, or particulates, depending upon their chemical characteristics. Soluble and colloidal forms tend to follow the water masses into which they are placed unless they become associated with particles through physicochemical or biological activity. On the average, the bottom is at a depth of about 3,800 m, and the accumulation of radionuclides by the benthic biota from surface introduction occurs mainly through activity of the plankton and the sinking of organic debris and inorganic particulates from the surface waters.

BIOLOGICAL FACTORS THAT INFLUENCE THE AVAILABILITY OF RADIONUCLIDES TO MARINE ORGANISMS

Concentration Factors

Biological concentration factors for nonconservative elements are generally greater than those for the major, or conservative, elements, which are present in amounts directly related to the salinity or chlorinity of the water and are effectively transported and distributed in the sea by currents. Nonconservative elements do not follow these patterns, however, and marine biologists have often postulated that marine plants and animals participate in the transport of these elements. This concept is based mainly upon the observations that marine organisms concentrate several

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*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 in microcuries of radioisotope per gram of total element.

elements to levels many times those in seawater and that the organisms often are capable of moving relatively quickly in directions different from those of the water masses in which they live. This was noted originally for phosphorus and silicon.

Several elements occur in marine organisms in amounts exceeding those in the water. The term "concentration factor" has been used in this relationship and may be defined as the ratio of the concentration of an element or radionuclide in an organism or its tissues to that concentration directly available from the organism's environment under equilibrium or steady-state conditions. This definition of a concentration factor is complicated by the fact that aquatic organisms do not normally derive all of their nutrients or radionuclides from one source but instead accumulate them from a variety of sources, including food, water, and suspended or deposited sediment. Concentration factors are indicators only and are not absolute. They may be altered by biological and environmental factors.

Polikarpov (1966), in his often-used definition of "concentration factor," stated that this factor is "the ratio of the concentration of a radionuclide (or corresponding stable element) in the organism and in the aqueous solution." The restrictive character of this definition is illustrated by his added statement that "the capacity of an organism to accumulate radioactive substances is expressed by the ratio of its radioactivity to that of the aqueous medium *or the preceding food link** in which the radionuclide was concentrated." In many marine organisms, the concentration factor is based on the preceding food link. The limited practical utility of a concentration factor concept based entirely on water becomes even more apparent when one considers some infaunal marine organisms that do not live in water and do not eat "normal" food but rather live in bottom sediments and feed on detritus. Because many of these organisms ingest large amounts of sediment along with the organic detritus, one is faced with the problem of selecting the environmental basis for comparing the amounts of elements or radionuclides in the organisms. Comparisons of the amounts of trace elements in an infaunal organism with the amounts in seawater are not representative, since, for many elements, even the concentrations of dissolved elements in the interstitial water of sediments differ significantly from those in "normal" seawater (Brooks *et al.*, 1968). Comparisons of elemental concentration in organisms with the total amount of element per unit weight of sediment also introduce large errors due to the variable amounts of certain elements that cannot be metabolized by the organisms because they are incorporated in mineral matrices that are not broken down by biological activity. Concentration factors based on comparisons of elemental content of marine organisms with amounts of the element associated with organic detritus in the near-shore areas of the sea may be subject to large errors. Here active precipitation and adsorption may occur for several elements introduced by rivers in soluble or colloidal form. Not only do these elements coprecipitate and adsorb onto the surfaces of organic detritus, but they often adsorb onto the surfaces of clay and mineral particles that may present a greater total surface area than organic detritus. Sediment is ingested along with the food, and only a small fraction of the total elements accumulated by the organisms may have come from the food. The removal of adsorbed stable or radioactive nuclides from the surfaces of organic detritus and sediment during digestive processes of marine infaunal invertebrates is strongly influenced by pH, total electrolyte content, enzymic digestion of biological substrates, and other factors that determine the degree of association between adsorbed ions or colloids and the exposed surfaces of ingested material. Not all trace elements associated with organic detritus in estuarine areas are adsorbed onto the surface but may have been incorporated into the living material. Some elements do flocculate rapidly from solution or colloidal suspension when river water is mixed with seawater in estuaries. For these elements, the amounts adsorbed onto the surfaces of organic detritus may exceed by factors of several hundred the amounts biologically incorporated into the detritus (Phelps, personal communication).

The problem of defining "concentration factor" in benthic organisms is related to the complex relationships of these organisms to the total available trace elements or radionuclides that surround them. Many benthic organisms are of importance from the viewpoint of human food sources and may comprise an important fraction of the total biomass in marine areas of economic importance (Sanders, 1956, 1958, 1960; Sanders *et al.*, 1962; Phelps, 1967).

In contrast to infaunal and benthic organisms, concentration factors for plankton and nekton may be based directly upon concentrations of the elements in seawater. On the basis of the total oceans, plankton and nekton provide most of the biomass, and, of these, the phytoplankton constitute the largest group in terms of both volume and exposed surface area. Phytoplankton accumulate nutrient elements directly from water, and their concentrations of stable or radioactive nuclides may be compared directly with the amounts in water for calculating concentration factors without the complication of intervening trophic levels. Water is also the ultimate source of nuclides for the zooplankton and nekton, although the radionuclides may be preconcentrated or discriminated against as they pass through one or more trophic levels before being incorporated into the individuals of any given step in a food web or chain. Even for those animals constituting the higher trophic levels, an equilibrium may be established between the animals, their food, and the amount of the element or radionuclide in the water. Because the intervening food orga-

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*Emphasis added.

nisms approach a given equilibrium with the water and are in turn eaten by the animals of the higher trophic level, so too will the organisms higher in the food web approach an equilibrium with the amount of the material in the water.

Turnover Rates

In the natural environment, the capacity of an organism to achieve equilibrium with a radionuclide will be directly related to the biological turnover rate of the corresponding stable element in the organism and inversely related to the rate of environmental dilution. The length of time required for an organism to exchange half of its total content of a given element is referred to as the biological half-life of the element. The shorter the biological half-life, the faster the organism may achieve equilibrium with the water. Whether equilibrium has become established may be determined by measuring the specific activity of the radionuclide in the organism and in the water (or food, if equilibrium between organism and food is of interest). When the specific activity of the organism is equal to that in the source, empirical equilibrium has been achieved.* In larger animals, the specific activities within various organs often differ from each other and from that in the water for periods of time measured in weeks or months following the introduction of the radionuclides, even in restricted environments. In many organisms, the total amount of a given element or radionuclide may consist of several pools, each with a different biological half-life varying from hours to weeks (Figure 1). Since the amount of a given radionuclide in the water from a single introduction usually decreases with time because of turbulent diffusion or other causes, the different body pools of the radionuclide in an organism living in the area will follow the decrease in environmental activity at different rates, each dependent upon a particular turnover rate. Under these conditions, the organism is unable to arrive at equilibrium with the environment. The smaller marine organisms, including the phytoplankton and some zooplanktons, frequently have turnover rates for trace elements with half-times measured in hours. In these organisms, "equilibration" with the changing levels of a radionuclide in the water may be achieved.

Although all organisms may approach equilibrium with the environment with respect to a given element, most marine organisms in higher trophic levels probably are not capable of achieving true equilibrium with introduced radionuclides of medium or short physical half-life. Failure to attain equilibrium is often caused by relatively rapid changes in environmental levels of radionuclides in marine areas surrounding restricted sites of introduction. It may also be caused by changes in vertical and horizontal distribution of radionuclides. The changes in radionuclide content in a given area of the open sea above the thermocline or in estuarine areas often occur rapidly in comparison with biological turnover rates,* especially for the higher trophic levels. However, if a chronic disposal situation exists for many years, organisms that grow up in that environment may be at equilibrium even with radionuclides of medium physical half-lives. Mauchline and Taylor (1964) have discussed equilibrium conditions for the long half-lived ^{137}Cs and ^{90}Sr under these conditions.

Surface Adsorption and Incorporation of Nuclides by Organisms

The degree to which an element and its radionuclides are concentrated by an organism depends upon the physicochemical interactions of the element with the environment and with the organism as well as upon the requirements of the organism. Goldberg (1957) showed that, in general, the ability of marine organisms to concentrate metals from the sea paralleled the order of stability of metal ligand complexes, and Szabo (1967) reported that the levels of accumulation of alkaline earths by mixed plankton occurred in the same sequence as that for cation exchangers such as Dowex-50 in which the order of association is radium > barium > strontium > calcium > magnesium. Grim (1953) proposed that the control of the alkali metals sodium, po-

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*The use of specific activity to determine if equilibrium has been achieved is valid only if the radionuclide and the corresponding stable element are present in the same physicochemical form.

*The reciprocal exchange of atoms of an element between an organism and the environment is referred to as turnover. Turnover rate refers to the fraction of atoms exchanged per unit weight of an organism in a given time period. The net movement of atoms in or out of an organism may change as a result of growth or other changes in the physiological condition of the organism or as a consequence of environmental changes. Not all atoms of an element that move in or out of an organism are necessarily involved in turnover. The net movement of atoms in or out of an organism may be explained as follows:

U = Uptake of element by organism
L = Loss of element by organism
When $U = L$ (net movement = zero, all movement is the result of turnover)
When $U > L$ (turnover occurring but net movement = the increase in amount of element in the organism)
When $U < L$ (turnover occurring but net movement = the decrease in amount of element in the organism)

Turnover rates of an element in an organism have been obtained by measuring the amount of a radioisotope of the element retained by the organism when it is placed in seawater not containing the radionuclide. Many investigators have used this method to follow the loss of a radionuclide from an organism with time.

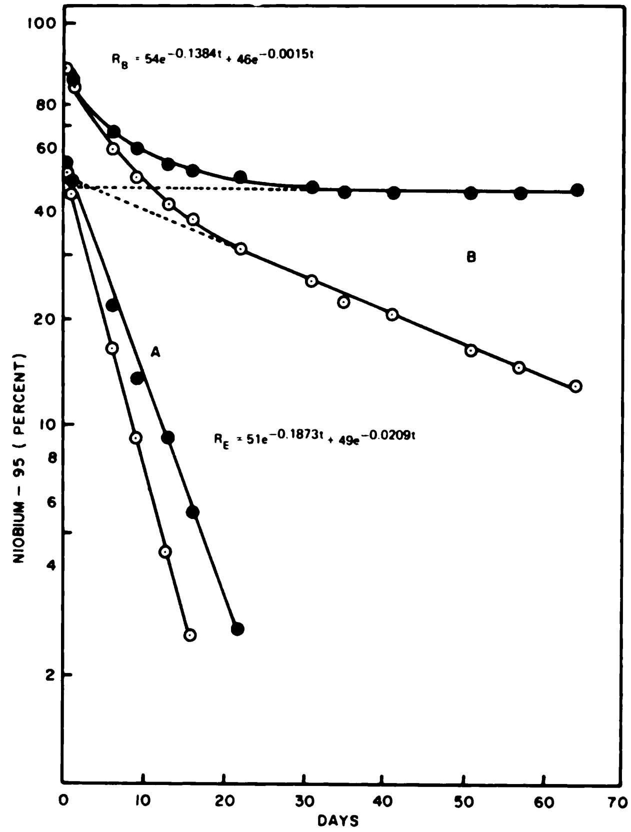

FIGURE 1 Retention of ^{95}Ni by the Atlantic croaker, *Micropogon undulatus*, showing separation of the curve into two components (A and B). Solid circles represent biological retention (R_B, corrected for physical decay); open circles represent effective retention (R_E, not corrected for decay). (Baptist *et al.*, 1970; reproduced from *Health Physics* 18:141-148, 1970, by permission of the Health Physics Society.)

tassium, rubidium, and cesium in the sea involved ion-exchange reactions with the clay minerals of the bottom sediments, with retention on the clay increasing with the ionic radius of the element. Similar ion-exchange reactions for the alkali metals may occur at the surfaces of marine organisms.

Several investigators have postulated that surface sorption, including ion exchange, may play an important role in the incorporation of trace elements into food webs by phytoplankton. For the heavier alkaline earths and alkali metals, at least, the major uptake mechanism probably is provided by ion-exchange mechanisms, although some other elements may be adsorbed mainly by chelating processes. Fukai (1966), in a study of the binding of radionuclides on ion-exchange resins, observed that radionuclides of chromium, manganese, iron, cobalt, zinc, and cerium rapidly dissociated from Dowex-50 when the loaded resins were placed in seawater, but that more than 90 percent of all of these elements remained chelated to Chelex-100 after elution with seawater. The resin showed an especially high affinity for transition elements.

Although ion-exchange resins will not remove most metals from seawater, the resins are capable of extracting several elements from seawater if they are first converted to chelating resins. Carritt (1965) reported that by absorbing the chelating agent dithizone onto Dowex-1-12x, 100 g of the resulting chelating resin could concentrate cobalt, copper, nickel, zinc, cadmium, and lead from 2,000 liters of seawater before saturation occurred (see also Boni, 1966, and Callahan *et al.*, 1966). Goya and Lai (1967) studied the adsorption of 37 elements from water by Chelex-100 and observed that more than 95 percent of manganese, iron, cobalt, chromium, nickel, zinc, cadmium, lead, bismuth, and several of the lanthanides and actinides were chelated onto the resin from seawater at the natural pH. More than 50 percent of the mercury, tin, and ruthenium and more than 20 percent of the barium, silver, gallium, strontium, calcium, scandium, titanium, and technetium were chelated by the resin. Less than 0.6 percent of the sodium and molybdenum were taken up, although 1.9 percent of the cesium was adsorbed.

If the average concentration factors of the marine phytoplankton, zooplankton, and attached algae for elements in seawater are plotted against the percentage of the corresponding elements sorbed from the water by Chelex-100, uptake by organisms and resin in general are positively related (Figure 2). The dashed lines in the figure represent the subjective relationship between the two variables. Five of the elements—titanium, scandium, silver, gallium, and zirconium—appear to be concentrated by the organisms to values higher than would be expected from their uptake by Chelex-100. Titanium, scandium, gallium, and zirconium share a common characteristic of being easily hydrolyzed and forming more or less insoluble hydroxides and ion complexes at the pH of seawater. Also included in those that form insoluble hydroxides are aluminum, yttrium, iron, the lanthanides, and plutonium. All of these elements are concentrated by plankton and seaweeds with average concentration factors as follows: aluminum, 70,000; iron, 25,000; zirconium, 29,000; titanium, 10,000; gallium, 5,500; plutonium, 2,200; scandium, 1,700; cerium, 51,100; and yttrium, 500.

RESULTS OF BIOLOGICAL ACTIVITY

Accumulation and Concentration of Elements by Benthic Algae, Phytoplankton, Zooplankton, Molluscs, Crustacea, and Fish

The average concentration factors for benthic algae, phytoplankton, zooplankton, and the muscle tissue of molluscs, crustacea, and fish are shown in Table 1. The concentration

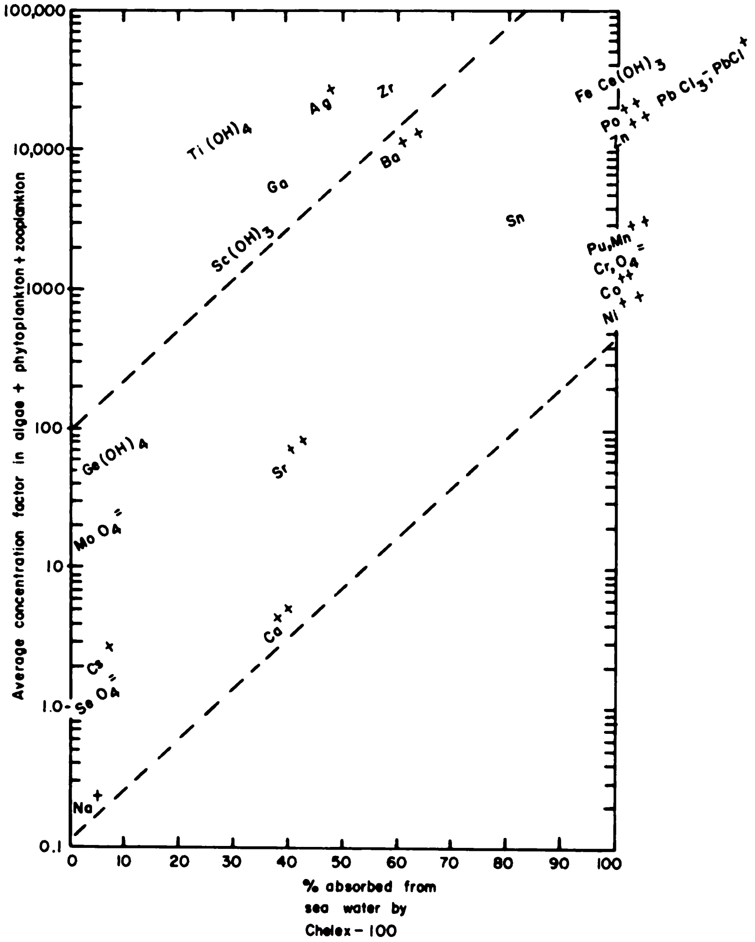

FIGURE 2 Relation of concentration factor of several elements in algae, phytoplankton, and zooplankton to their absorption from seawater by Chelex-100.

factors are taken from a large number of references, and comparisons of factors between individual elements cannot be made in most cases; however, general patterns of uptake may be selected. Here, as described before, the alkali metals, sodium through cesium, and the alkaline earths, magnesium through radium, show a general increase in uptake with increased ionic radius. Beryllium, however, is concentrated more strongly than magnesium and calcium, although its ionic radius is much smaller. Beryllium has several properties relating it to zinc and/or cadmium, including a tendency for covalent bonding, formation of complexes, and formation of amphoteric hydroxides. Beryllium thus might be expected to resemble, in part at least, the IIB elements in enhanced uptake by marine organisms.

In general, the elements that are concentrated significantly in marine organisms (Table 1) may be grouped into at least one of five categories. These are (a) structural elements: carbon, nitrogen, phosphorus (silicon, calcium, and strontium, in some cases); (b) catalyst elements: iron, copper, zinc, manganese, and cobalt (nickel, chromium, cadmium, and silver may follow these elements); (c) elements easily hydrolyzed at seawater pH: aluminum, gallium, scandium, yttrium, cerium, plutonium, titanium, and zirconium; (d) heavy halogens: bromine and iodine; and (e) heavy divalent ions: barium, radium, and lead.

Marine plants and animals concentrate the different isotopes of a given element to the same degree, provided the isotopes are in the same chemical and physical form.* The degree to which radionuclides are incorporated into food webs also depends upon the availability of the corresponding stable element and metabolically similar elements and the physiological and behavioral characteristics of the organisms in food webs and chains. Because several radionuclides are accumulated by marine organisms to concentrations several thousand times the amounts in the water, several investigators have suggested that biological transport may be a factor in altering their distribution in the sea.

Investigations of the presence of radionuclides in the marine environment have been concerned mainly with the release of contaminants produced by nuclear technology, including testing of nuclear weapons, as well as reactor cooling and fuel processing. Radionuclides that are accumulated

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*Except for lighter elements, including H, C, O, and S (Vogel, 1969; Degens, 1967; Münnich and Vogel, 1959; Ault, 1959a and b; Silverman and Epstein, 1958; Craig, 1954).

TABLE 1 Average Concentration Factors for Benthic Algae, Plankton, and Mollusc, Crustacean, and Fish Muscle[a]

Chemical Group		Benthic algae	Phytoplankton	Zooplankton	Mollusc Muscle or Soft Parts	Crustacean Muscle	Fish Muscle
IA	Li	–	–	–	0.28	1.2	0.47
	Na	–	–	–	0.2	0.3	0.13
	K	–	–	–	8	13	13
	Rb	–	–	–	16	13	17
	Cs	–	–	–	8	23	15
	Fr	–	–	–	–	–	–
IB	Cu	100	30,000	6,000	5,000	–	1,000
	Ag	–	23,000	9,000	7,100	7	–
	Au	470	–	–	400	400	60
IIA	Be	110	1,000	15	–	–	–
	Mg	2	2	4	1	–	0.2
	Ca	2	2.5	5	0.4	120	1.5
	Sr	96	–	–	1	3	0.1
	Ba	–	17,000	900	–	–	8
	Ra	1,400	12,000	190	1,300	140	130
IIB	✓Zn	410	15,000	8,000	11,000	2,000	500
	Cd	200	–	–	–	–	1,000
	Hg	–	–	–	–	–	–
IIIA	B	<1	–	1.6	–	–	2
	Al	15,000	100,000	100,000	9,000	12,000	10,000
	Ga	1,300	8,000	7,000	2,000	2,000	–
	In	–	–	–	–	–	–
	Tl	–	–	–	–	–	–
IIIB	Sc	2,000	2,000	1,000	–	300	750
	Y	480	1,000	105	12	–	250
	La	–	–	–	–	–	–
	Ce	670	90,000[b]	1,000	360	2	0.3
	Pu	1,300	2,600	2,600	260	3	3
IVA	C	~4,000	3,600	2,800	4,700	3,600	5,400
	Si	100	2,000	300	50	–	20
	Ge	50	–	–	–	–	–
	Sn	–	6,000	450	–	–	–
	Pb	700	40,000	3,000	40	–	–
IVB	Ti	4,100	25,000	17,000	–	–	–
	Zr	2,200	60,000	25,000	2	2	<1
	Hf	–	–	–	–	–	–
VA	N	–	36,000	24,000	47,500	44,000	65,000
	P	10,000	34,000	13,000	6,000	24,000	33,000
	As	2,000	–	–	650	400	700
	Sb	–	–	–	–	–	–
	Bi	–	–	–	–	–	–
VB	V	600	600	700	1,700	330	110
	Nb	1,000	1,000	–	7	3	100
	✓Ta	–	–	–	–	–	–
VIA	S	1	–	–	0.3	–	1
	Se	1	–	–	–	–	–
	Te	–	–	–	–	–	–
	Po	1,000	–	–	–	–	–

TABLE 1 (Continued)

Chemical Group		Benthic algae	Phytoplankton	Zooplankton	Mollusc Muscle or Soft Parts	Crustacean Muscle	Fish Muscle
VIB	Cr	1,600	2,400	1,900	440	100	70
	Mo	8	–	26	60	10	10
	W	5	–	–	20	2	3
VIIA	F	1	–	–	–	–	–
	Cl	1	1	1	1	1	1
	Br	–	–	–	–	–	–
	I	5,000	–	3,000	50	30	12
	At	–	–	–	–	–	–
VIIIB	Mn	2,300	4,000	1,500	12,000	1,900	80
	Tc	–	–	–	–	–	–
	Re	–	–	–	–	–	–
VIII	Fe	4,800	45,000	25,000	9,600	2,400	1,600
	Co	800	1,500	700	600	500	10
	Ni	1,000	5,000	3,000	–	–	–
	Ru	390	200,000[b]	34,000[b]	3	100	0.05
	Rh	–	–	–	–	–	–
	Pd	–	–	–	–	–	–
	Os	–	–	–	–	–	–
	Ir	–	–	–	–	–	–
	Pt	–	–	–	–	–	–

[a] Data derived from Polikarpov (1966), Templeton (1965), Goldberg (1962), Vinogradova and Koval'skiy (1962), Fukai and Meinke (1962), Nicholls *et al.*, (1959), and from unpublished work of Lowman.

[b] Slowey *et al.* (1965).

in easily detected amounts by marine organisms include the fission products ^{95}Zr, ^{99}Mo, ^{103}Ru, ^{106}Ru, ^{131}I, ^{140}Ba, ^{141}Ce, ^{144}Ce, and the neutron-induced radionuclides ^{32}P, ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{51}Cr, ^{57}Co, ^{60}Co, ^{65}Zn, ^{110}Ag, ^{181}W, ^{185}W, and ^{187}W, and natural ^{40}K. All of these radionuclides, except for ^{40}K, represent stable elements that occur in seawater at much less than one part per million. If marine organisms are capable of altering the distribution patterns of the radionuclides listed above from those expected to occur from physical and chemical factors in the sea, then they should also be capable of influencing the distribution of the corresponding stable elements. The present discussion will concern both stable and radioactive isotopes.

Accumulation and Biological Transport

In seawater the major or conservative stable elements are transported by currents as solutes, soluble chelates, or dispersed colloids and may undergo limited movement from one water mass to another by eddy diffusion. Elements associated with biological material also are distributed by these processes but, in addition, are transported by vertical and horizontal migrations of animals, are anchored in place on the bottom through incorporation by benthic organisms, or are carried to the bottom from the surface by the effect of gravity on dead organisms, fecal pellets, moulted exoskeletons, and other organic detritus.

Any influence of marine organisms (or of physical or chemical processes) that changes the distribution pattern of an element from that of the conservative elements depends upon the conversion of the element from soluble or colloidal to particulate form. For marine organisms to change the distribution of a stable element (or radionuclide), they must alter a significant fraction of the total element present in the water in a period of time equal to or less than the time required for transport by water. This is most likely to occur if the radionuclide is introduced into an area of intense biological activity in which there is a shortage of trace elements resulting from a high rate of utilization.

The rate at which nutrients may be incorporated into phytoplankton is directly related to the biomass. However, the incorporation of nutrients is also limited by the amount of the organic material produced. Only those nutrients present in the euphotic zone at any given time may be incorporated into the biomass. As the phytoplankton population increases, so does the absorption of light in the water.

Photosynthesis, thus, becomes more and more restricted to a thin layer near the surface in which nutrients are rapidly depleted. If vertical mixing occurs, bringing nutrients into the euphotic layer, the same mixing may carry phytoplankton to deeper waters where lower levels of illumination will reduce photosynthesis and eventually cause their death.

In contrast to photosynthesis, which is restricted to a narrow layer at the surface, catabolism, excretion, death, and decomposition occur at all depths, including the euphotic zone. Over most of the oceanic area, there is a shallow upper zone of incorporation of elements, a downward movement of dead organisms and particulate excretion products, and a broader zone of regeneration; a net downward movement of nutrients occurs with time. The vertical distribution of the elements will depend upon the total amount of each element accumulated in the biomass, the sinking rate, and the rate at which the individual elements are regenerated into solution. Those elements regenerated more rapidly tend to remain in the upper layers, while those with slower regeneration rates become distributed into the deeper waters.

Distribution of the Structural Elements Nitrogen, Phosphorus, Carbon, and Silicon

Four structural elements—nitrogen, phosphorus, carbon, and silicon—are present in phytoplankton at levels from 1,000 to 40,000 times the amounts found in seawater. In some diatoms, the concentration factor for silica may be as high as 55,000. These concentration factors are of the same order of magnitude as those observed for the uptake of several radionuclides by these and other plankton. Thus, an examination of the distribution patterns of nitrogen, phosphorus, carbon, and silicon in the waters of areas of high biological productivity may provide an insight into possible effects of biological activity upon radionuclide transport and distribution, provided turnover rates for the nutrient elements and the radiocontaminants are comparable.

The elements carbon, hydrogen, nitrogen, and phosphorus are involved in the marine food webs as essential components of protoplasm and—particularly carbon and phosphorus—as constituents of skeletal parts and shells. In plankton organisms these elements are found in atom ratios that approximate H:C:N:P = 268:106:16:1 (Sverdrup *et al.*, 1942), which provided the basis for the C:N:P ratios and the hypothetical planktonic composition $(CH_2O)_{100}(NH_3)_{16}H_3PO_4$ suggested by Redfield *et al.*, 1963. The C:N:P ratios exclude carbon in calcareous structures and phosphorus of skeletal structures of higher forms. The ratios of shell and skeletal carbon and phosphorus are highly variable, but reasonable limits of variability in the structures may be established.

The amount of carbon, hydrogen, nitrogen, and phosphorus that can be photsynthetically incorporated into plant material, and thus ultimately into animal material, is limited by the available supply and the stoichiometric composition of the resulting biomass. Although it has been suggested that several elements may act as limiting parameters for the production of the total biomass, it is increasingly evident that the potential productivity of the sea is primarily a function of the availability of phosphorus or nitrogen or both.

Potential fertility may be defined as the quantity of organic matter that photosynthesis can produce in a volume of seawater if allowed to proceed until the limiting nutrient is exhausted (Redfield *et al.*, 1963). Thus, the potential productivity is chemically limited and tends to represent upper limits of primary productivity. However, the limiting is not subject to a sharp cutoff, although phytoplankton normally assimilates nitrogen and phosphorus in the atom ratio of approximately 15:1 until very low concentrations of the nitrogen are present in the water. Even after the nitrogen is almost completely depleted in the water, the phytoplankton cells apparently continue to incorporate phosphorus, which is still present in relatively large amounts in solution (Ketchum *et al.*, 1958). According to Ketchum *et al.* (1958), eddy diffusion may bring nitrogen and phosphorus to the euphotic layer from the deeper layers in higher amounts than normally occurs in the euphotic layer; thus, the phytoplankton could use the element in short supply as rapidly as it became available. In this way, the phytoplankton would continue to form cells of normal composition and incorporate nitrogen, which apparently was present in extremely small amounts in the water. Even here, the rate at which nitrogen was supplied from the deeper water to the euphotic layer would exert a limiting effect.

The maximum potential productivity of the open ocean is probably represented by the deep waters of the North Pacific where the phosphate content is about 3.5×10^{-6} *M*. The photosynthetic utilization of all this phosphorus would take place as shown in Table 2.

It is evident that, in the example chosen, the total realization of the potential productivity of this water would bind all of the nitrate, most of the phosphate, about 10 percent of the carbon, and a negligible fraction of the hydrogen into the bodies of the plants. Following the same reasoning used by Redfield *et al.* (1963), this would result in the fixation of 371 mg-atoms of carbon per m^3* or 4.452 g of carbon per m^3. If the carbon constitutes 50 percent of the dry weight of the plankton and the dry/wet ratio is 0.2, the wet weight of the plankton would be 44.5 g/m^3. This is approximately the biomass that has been observed in redtide blooms (41 cc/m^3, Ketchum and Keen, 1948). If the plankton has a density the same as that of seawater, this amount of productivity would represent the following concentra-

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*3.5 mg-atom $C/m^3 \times 106$ (atom ratio for carbon) = 371 mg-atom C.

TABLE 2 Amount of Nutrient Elements in "Richest" Seawater and the Rates of Availability and Utilization by Phytoplankton

(1) Element	(2) mg-Atoms/m^3 in Richest Seawater	(3) Relative Availability in Richest Seawater	(4) Atom Ratio Utilized in Phytoplankton	(5) Relative Availability (3)/(4)
P (PO_4)	3.5	1	1	1
N (NO_3-)	52.5	15	16	0.94
C (HCO_3-)	3,559.5	1,017	111	9.6
H (H_2O)	108,000,000	30,850,000	260	1.2×10^5

tion factors: phosphorus: 2.2×10^4; nitrogen: 2.4×10^4; carbon: 2.3×10^3; hydrogen: 0.195.

Nowhere has the estimated annual production of plankton populations been observed to exceed the potential productivity of the richest oceanic water cited above, assuming a photic depth of 50 m. If the mg-atom ratios for the four elements in "average" seawater were used (P = 2.3; N = 34.5; C = 2,340; H = 1.08×10^8) instead of those for "richest" seawater, 244 mg-atoms or 2.93 g of carbon per m^3 would be fixed. This would amount to 29.3 g of wet plankton per m^3. The concentration factors for the four elements under these conditions would be, for phosphorus, 3.4×10^4; for nitrogen, 3.6×10^4; for carbon, 3.6×10^3; and for hydrogen, 0.195.

In each case, the concentration factors for phosphorus and nitrogen are approximately 3×10^4; carbon, 3×10^3; and hydrogen, 0.2. These concentration factors are probably representative for all of the more productive areas of the world. The potential productivity in the vicinity of Woods Hole, Massachusetts, might be assumed to be related to a concentration of phosphate of 2×10^{-6} *M* (62 μg P/liter). This could yield 2.54 g C/m^3 or, in a photic zone 50 m deep, 127 g C/m^2. A weighted mean average for the region is 120 g C/m^2/year (Ryther, 1963). This agreement is probably fortuitous, but it might be interpreted as meaning that, at the average rate of production it would take a year for all of the phosphate and nitrogen in the photic zone to be incorporated into the bodies of plants and animals and that replacement water, if any, had the same productivity potential when introduced into the photic zone.

The turnover rates for nitrogen and phosphorus may equal or exceed, in a short-time interval, the annual average, and the fractions of both elements that are not passed to higher trophic levels or to the bottom through the sinking of fecal pellets, moults, and dead organisms are probably recycled between the primary producers and the water several times during an annual cycle.

Even the degree of association suggested by the annual average for the accumulation of other elements is sufficient that local populations with movements independent of the motion of the water would be expected to contain and transport significant amounts of total phosphorus and nitrogen accumulated in the euphotic layer. If the marine organisms do indeed transport nitrogen and phosphorus in patterns different from those resulting from physical mechanisms, they might also be expected to exert a similar influence upon the distribution patterns of other elements, and their radionuclides, that are accumulated to the same level.

The nutrient elements do exhibit vertical distribution patterns different from those of the conservative elements (Figure 3). Phosphate, nitrate and silica are found in the intermediate and deep water in greater amounts than in the euphotic layer. These three elements may be removed from the surface layers by the settling of phytoplankton; by incorporation of the phytoplankton into the zooplankton, which undergo diurnal migration; or by excretion or loss as fecal pellets, moults, dead zooplankton, and organic debris, all of which will sink.

Most of the phosphorus in the animals undergoes rapid turnover. Varying amounts of it are incorporated into fecal pellets, other excretory products, dead organisms, and into the skeletal structure of fishes, which are deposited upon the bottom after the death of the animal. Skeletal apatite slowly dissolves in the deeper parts of the ocean, and a large part of the phosphorus is eventually returned to solution in these areas.

Upon the death of phytoplankton and zooplankton, phosphorus appears to be regenerated back into the water slightly more rapidly than nitrogen, and nitrogen more rapidly than carbon, since the C : N : P ratios increase with depth (Richards *et al.*, 1965; Holm-Hansen *et al.*, 1966; Vaccaro, 1963). However, the relative regeneration rates of carbon and nitrogen may vary according to local conditions, since Menzel and Ryther (1964) observed low ratios of carbon to nitrogen below the euphotic zone to depths of 4,000 m and reported that similar conclusions could be derived from the data of Parsons and Strickland (1962). In any event, the turnover rate of phosphorus is sufficiently rapid that large variations of phosphorus concentration with time of day have been noted in Chesapeake Bay (Newcombe

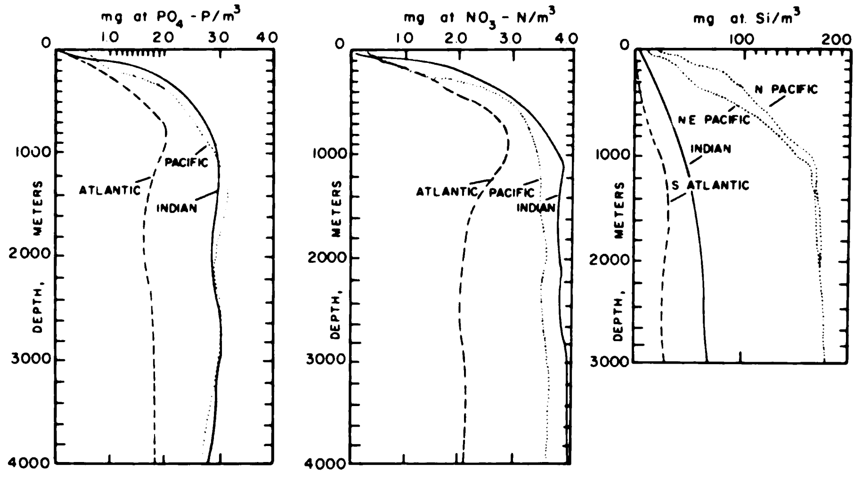

FIGURE 3 The vertical distribution of phosphate, nitrate and silica in the Atlantic, Pacific, and Indian oceans. (Reprinted with permission from Sverdrup *et al.*, 1942.)

and Lang, 1939), with the phosphate in the surface waters decreasing slowly after sunrise from 13 μg P/liter to a minimum value of 3 μg P/liter at 3:00 p.m. After sunset, the phosphorus increased rapidly to maximum values about 2 hours after midnight. Newcombe and Brust (1940) reported similar observations. Phosphorus is also turned over rapidly in dead or dying phytoplankton. Hoffman (1956) reported that, upon the death of diatoms, autolysis rapidly released up to 25 percent of the total phosphorus as inorganic phosphate. Raymont (1963) observed that immediately after death of phytoplankton about one third of the total phosphate was released as dissolved organics and that within 2 days, 70 percent of the remaining phosphorus was remineralized. Menzel and Ryther (1964) stated that, in comparison to carbon and nitrogen, phosphorus was regenerated very rapidly, since no phosphorus was found associated with organic particulates at depths below 200 m in the western North Atlantic.

In some cases, the regeneration of phosphorus in zooplankton may be even more rapid than in phytoplankton. Pomeroy *et al.* (1963) reported that zooplankton were capable of excreting 100 percent of the contained phosphorus daily, a large part of which was apparently tied up in soluble organic compounds and was not excreted as phosphate.

The regeneration of silica from diatom skeletons might be expected to differ from phosphorus in that the regeneration of phosphorus and nitrogen involves the removal of both elements as constituents of organic matter. In addition, zooplankton incorporate phosphorus and nitrogen, but not silica. The silica tests are ejected and would sink to dissolve at greater depth while the two other elements would be retained in the upper layers (Redfield *et al.*, 1963).

In summary, the vertical distribution of phosphorus, nitrogen, and silica in the sea is affected by biological transport, and the concentrations of the three elements are not related to salinity or the distribution of the conservative elements. The vertical distributions of the three elements differ in the Atlantic, Pacific, and Indian oceans (Figure 3) but follow a generally similar pattern. Phosphorus, nitrogen, and silica are usually present in the surface waters in low amounts or are depleted and rise gradually in value to a maximum for phosphorus between 300 and 1,000 m, but for nitrogen and silica the maximum is at 1,000 m or deeper.

Carbon is concentrated by phytoplankton at factors about an order of magnitude lower than phosphorus or nitrogen, and only about 10 percent of the available carbon dioxide is utilized under conditions of maximum uptake. The total amounts of carbon dioxide increase with depth because of its incorporation into plants in the euphotic layer and the production of carbon dioxide when dead material is oxidized at deeper levels. Low oxygen concentrations and pH values in areas with relatively large amounts of carbon dioxide tend to characterize water masses that are poorly ventilated and that receive large amounts of decaying organic matter (Skirrow, 1965). Carbon dioxide content of seawater often increases measurably with depth in the first few hundred meters. Keeling *et al.* (1965) reported observations from the equatorial Pacific in which the carbon dioxide increased from 310 ppm in the top 100 m to a value of 620 ppm at 450 m.

The oxidation of carbon compounds appears to proceed more slowly in those containing nitrogen (i.e., the proteins). Thus, Menzel and Ryther (1964) observed that the C:N ratios in nonliving detritus collected in the euphotic zone were the same as those in living phytoplankton (C:N 11.5:1 in January and 5.3:1 in April). In the deeper waters, to depths of 4,000 m, the C:N ratio decreased to 2.5:1. The average C:N ratio for protein is about 2.9:1 (Sverdrup *et al.*, 1942). Menzel (1967) reported, however, that the surface standing crop at any given site had no influence on the amount of organic particles directly below—that the distribution of organic particles in all areas of the sea was homogeneous in the deeper waters and that no consistent decrease in dissolved carbon occurred with depth. Redfield (1942) considered that, in the North Atlantic, the high nutrient levels at depth were derived from material sinking at the Antartic Convergence. Sheldon *et al.* (1967) also found that between depths of 200 and 4,000 m the amount of particulate carbon was constant in different areas of the sea. Hobson (1967), however, observed seasonal and vertical variations in particle concentrations in the northeast Pacific Ocean. He tentatively attributed these variations to differences in the production of particles in the euphotic zone and their subsequent sinking and horizontal transport by currents.

Uptake of Nonstructural Elements and Their Radionuclides by Phytoplankton

Twenty-six elements have been reported to be concentrated in marine organisms by factors approximately equal to or greater than that for carbon. In addition, plutonium, which occurs naturally in only insignificant traces but is produced by man, has been observed to be accumulated by mixed plankton to levels 2,600 times those in the surrounding water (Pillai *et al.*, 1964).

Those elements that are concentrated at least 1,000 times in marine phytoplankton are shown in Table 3. Also shown are the radioisotopes most likely to be formed by fission and by activation by neutrons, protons, deuterons, and alpha particles. Except for a few radionuclides, only those resulting from fission and neutron activation would be produced in relatively large amounts. Of the 25 radionuclides listed, ^{28}Al, ^{76}As, ^{64}Cu, ^{131}I, and ^{47}Sc have short physical half-lives and would undergo physical decay before biological transport could significantly alter their distribution in the oceans. Of the remaining nuclides, two, ^{115m}Cd and ^{63}Ni, would not be produced in large amounts by known sources (Beasley and Held, 1969). Thus, 7 of the 25 radionuclides listed in Table 3 would not have radioisotopes subject to significant biological accumulation and may be eliminated from further consideration. The remaining 18 radionuclides include several of known biological importance. These include ^{32}P, ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{57}Co, ^{58}Co, ^{60}Co, and ^{65}Zn. The radionuclides of elements that do not have known biological functions include ^{46}Sc, ^{95}Zr, ^{95}Nb, ^{110m}Ag, ^{144}Ce, ^{210}Pb and ^{239}Pu.

Of special interest is the observation that, of the above-

TABLE 3 Elements Accumulated by Phytoplankton to Levels at Least 1,000 Times Those in Seawater and Principal Radionuclides of Interest in Considerations of Biological Transport

Element	Radionuclide	Half-life	Principle Reaction for Production	Reported Concentration in Phytoplankton
Al	^{28}Al	2.31 min^{a}	^{27}Al (n, γ)	1×10^5
As	^{76}As	26.4 hr^{a}	^{75}As (n, γ)	–
Be	^{7}Be	53.6 day	^{6}Li (d, n)^{b}	1×10^3
C	^{14}C	5,730 yr	^{14}N (n, p)	4×10^3
Cd	^{115m}Cd	43 day	^{114}Cd (n, γ)^{b}	–
Ce	^{144}Ce	275 day	Fission	9×10^4
Cr	^{51}Cr	17.8 day	^{50}Cr (n, γ)	2×10^3
Co	^{57}Co	270 day	^{56}Fe (d, n)	1×10^3
			^{56}Fe (p, γ)	–
Co	^{58}Co	71.3 day	^{55}Mn (a, n)	1×10^3
Co	^{60}Co	5.26 yr	^{59}Co (n, γ)	1×10^3
Cu	^{64}Cu	12.8 hr^{a}	^{63}Cu (n, γ)	3×10^4
I	^{131}I	8.05 day^{a}	Fission	–
Fe	^{55}Fe	2.6 yr	^{54}Fe (n, γ)	4×10^4
Fe	^{59}Fe	45.6 day	^{58}Fe (n, γ)	4×10^4
Pb	^{210}Pb	20.4 yr	Daughter ^{226}Ra^{c}	4×10^4
Mn	^{54}Mn	303 day	^{56}Fe (d, a); ^{54}Fe (n, p)	4×10^3
Ni	^{63}Ni	92 yr	^{62}Ni (n, γ)^{b}	5×10^3
Nb	^{95}Nb	35 day	Fission	1×10^3
P	^{32}P	14.3 day	^{31}P (n, γ)	3×10^4
Pu	^{239}Pu	24.4 yr	^{238}U (n, γ)	2.6×10^3
Sc	^{46}Sc	83.9 day	^{45}Sc (n, γ)	2×10^3
Sc	^{47}Sc	3.43 day^{a}	Daughter ^{47}Ca	–
			^{46}Ca (n, γ)	2×10^3
Ag	^{110m}Ag	255 day	^{109}Ag (n, γ)	2×10^4
Zn	^{65}Zn	245 day	^{64}Zn (n, γ)	2×10^4
Zr	^{95}Zr	65.5 day	Fission	6×10^4

[a] Radionuclide has too short a half-life for effective biological transport.

[b] No known source of large-scale production. Small-scale production of ^{63}Ni reported by Beasely and Held, 1969.

[c] ^{210}Pb reported to be formed in explosives used for peaceful uses of nuclear explosives.

listed radionuclides, all except ^{210}Pb have been reported to be concentrated by marine organisms from radioactive contamination introduced into the sea from nuclear weapons test sites and from outflows from production reactors and reactor fuel reprocessing plants. Lead-210 has not been produced in sufficient amounts by nuclear explosives to have been studied in detail. Naturally occurring ^{210}Pb has been reported in marine organisms (Beasley *et al.*, 1969).

Biological Transport by Zooplankton

Phytoplankton and zooplankton are the marine organisms most likely to influence the distribution patterns of radionuclides introduced into the sea. These organisms constitute the largest biomass, have the greatest total exposed biological surface areas, exhibit the highest concentration factors and turnover rates for many trace elements, and, in the case of zooplankton, migrate vertically. The magnitude and extent of diurnal vertical migration of zooplankton living in the upper layers, however, is often overestimated, and the plankton populations are sometimes considered to correspond to a large biological "blotter," which daily moves up and down to great depths, scavenging and redistributing radionuclides and trace elements in its path.

Plankton, to significantly alter the distribution of radionuclides from that caused by physical and chemical processes, must effectively transport the material at rates comparable with movement due to currents, turbulent diffusion, density gradients, and chemical precipitation or must alter the availability of the radionuclides for physical transport processes. Biological transport may be accomplished by direct uptake and loss of the radionuclide accompanied by horizontal or vertical movement, by the excretion of fecal pellets, and by the sinking of moulted exoskeletons or dead or moribund plants and animals.

Direct transport of radionuclides by vertical migration of zooplankton is dependent upon the ability of the organisms to travel relatively great distances in comparison with their size. In many areas of the world, the upper mixed layer is 80 to 100 m deep, and effective transport by the plankton may thus occur only if they migrate vertically beyond this depth.

Diurnal Migration of Deep-Water Plankton and Biological Transport Up from Depths

Vertical migrations of zooplankton through great distances have been reported for some species, and the importance of these migrations has sometimes been given undue emphasis, since these species constitute only a small fraction of the total zooplankton populations. Zooplankton exist at great depths in the sea; however, their diurnal vertical migrations are usually limited to a few hundred meters, and the ratio of their volume to that of the water is usually of the order of 10^{-8}. Amphipods, copepods, and ostracods have been collected from depths of 6,000 to 7,000 m by Russian investigators (Wolff, 1960), and concentrations of zooplankton have been observed at 1,500 to 2,000 m in the Atlantic (Leavitt, 1938). A significant fraction of these deep-living zooplankton undergo migrations although the depths at which they live preclude regular excursions into the upper mixed layer. Waterman *et al.* (1939) showed that amphipods, decapods, euphausids, and mysids migrated from 1,000 m upward to 200 to 600 m, and Bainbridge (1961) reported that the upper limit of migration for *Thysanopoda acutifrons* and *Acanthephyra purpurea* was about 200 m below the surface. The zooplankton from the deeper waters would, therefore, exert much less effect upon vertical transport of surface-introduced radionuclides than the animals in the upper layers, because those in deeper waters constitute a relatively small portion of the total population, and most of them do not pass into the upper mixed layer.

Ketchum and Bowen (1958) calculated the relative effectiveness of vertical transport of elements upward from the deeper waters by biological and physical processes using the mean residence time of 300 years (Craig, 1957) in the deep water (assuming an average depth of 3,800 m and a 100-m upper mixed layer). They reported that a concentration factor of 340 in zooplankton would be required for upward biological transport to equal that provided by physical transport. The numerical values used in these calculations, however, represented conditions that would result in near-maximum biological transport. They probably are not typical of the usual conditions. The ratio of zooplankton volume to water was assumed to be 10^{-6} and corresponds to the biomass of zooplankton found in areas of upwelling, which are sites with high rates of biological productivity. In many areas of the sea, smaller amounts of zooplankton are found. In addition, plankton in deeper waters are usually present in smaller quantities than in the surface layers. Bogorov (1946) found that 95 percent of the zooplankton in the Artic seas lived in the upper 200 m and only 5 percent of the total were below this depth. Even if the zooplankton in the deeper waters constituted four times this amount, the results cited above would be high by a factor of four.

The calculations were based upon the premise that the animals reached equilibrium with the radionuclides during the time spent in each water layer. This would require rapid turnover rates. In a study of zooplankton contaminated with radionuclides, Kuenzler (1965) found hourly turnover rates of 1 to 6 percent of the body pool for ^{131}I, ^{54}Mn, ^{58}Co, ^{60}Co, ^{55}Fe, and ^{65}Zn. In the copepods, which constituted 62 percent of the total zooplankton, the rates were 2.4 percent/hr for ^{131}I, 4.5 percent/hr for ^{58}Co and ^{60}Co, and

1.2 percent/hr for ^{65}Zn. Turnover rates of this magnitude could result in organisms approaching equilibrium at least for cobalt, if the observed rates of turnover were maintained continuously. These values, however, as well as most others that have been reported, were measured at surface temperatures. At these temperatures the turnover rates are 1.5 to 3.5 times those that occur in the deeper, cooler waters considered in the calculations. If those factors are taken into account, as well as the effect of vertical plankton distributions, concentration factors of about 10^4 would be required for biological transport upward to equal physical transport.

The average ratios of zooplankton volume to water in different marine areas have been tabulated by Ketchum (1957). Shown in Table 4 are the locations, the zooplankton biomass, and the concentration factors required for a rate of biological transport upward that would equal physical transport. The concentration factors are corrected for temperature and vertical distribution of plankton as described previously.

Mean concentration factors (CF) in zooplankton rarely exceed 10^4, as shown in Table 1. Elements that are concentrated to the highest level by zooplankton include aluminum (CF 100,000), zirconium (CF 25,000), nitrogen (CF 24,000), titanium (CF 17,000), phosphorus (CF 13,000), and iron (CF 25,000). Only zirconium, phosphorus, and iron have radionuclides that are normally produced in nuclear technology. For these radionuclides, biological transport from the bottom waters upward would be significantly lower than physical transport in all open ocean areas cited in Table 4 because of the relatively low concentration factors that exist for these elements. In the nearshore areas, including the Gulf of Maine, coastal waters, and the North African Upwelling, physical transport from deeper waters to the surface is at least one order of magnitude greater than the Atlantic average, and here also, physical transport would be more effective than biological transport in moving radioactive contaminants from the deeper waters to the surface.

Diurnal Migration of Zooplankton from the Upper Mixed Layer and Biological Transport Down from the Surface

Although zooplankton in the deeper waters may not contribute significantly to the transport of radioactivity from the bottom to the surface, surface-living species may transport relatively large amounts of radionuclides downward. Several authors have reported that the greatest amounts of zooplankton are found in the top 800 m of the sea and that most of the animals appear to live in the top 400 m. Johnson (1957) reported that during the daytime a major part of the diurnally migrating plankton was concentrated in one or more horizontal layers down to 400 m. Bogorov (1946), as mentioned before, observed that 83 percent of the zooplankton lived in the top 10 m of Arctic waters, while 12 percent lived between 10 and 200 m and 5 percent lived between 200 and 750 m. This general pattern of vertical distribution does not appear to be limited to the polar regions. Fraser (1936) showed that the larval stages of *Euphausia superba* lived at about 200 m and migrated upward diurnally. The young stages of the same species remained near the surface and did not migrate vertically at all. *Calanus finmarchicus*, the major copepod in the North Atlantic, occurs in surface waters at night and migrates downward to a midday depth of about 70 m (Nicholls, 1933). According to Farran (1947, 1949) marine ostracods appear at two levels, one at 100 to 300 m and the other at 250 to 400 m. Michael (1911) showed that *Sagitta bipunctata* migrated between the surface and a depth of 50 m off California, and Hardy and Gunther (1936) reported that the amphipod *Parathemisto* migrated from the surface to a depth of 80 to 100 m. According to Bainbridge (1961), the available data showed that most marine copepods migrated vertically 50 to 150 m but that vertical movements could not be demonstrated for *Anomalocera patersoni*, *Rhincalanus gigas*, *Calanoides acutus*, *Microcalanus pygaeus*, *Oithona*

TABLE 4 Biomass of Zooplankton in Different Marine Waters[a] and the Concentration Factors Required for Zooplankton to Transport Radionuclides Upward at a Rate Equal to the Physical Transport

Location and Character	Volumes (cc) of Zooplankton/m^3	Concentration Factors Required for Equal Biological and Physical Transport Upward
Sargasso Sea	0.006–0.09	75,000–1,000,000
Eastern North Pacific	0.042	55,000
Eastern Tropical Pacific	0.055	40,000
Peru Current	0.124	55,000
Gulf of Maine	0.08–1.0	6,800–85,000
Coastal waters	0.08–0.8	8,500–85,000[b]
North African Upwelling	1.0	6,800

[a] Data from Ketchum (1957).

[b] Not directly applicable because the rates of vertical transport from physical processes in these areas greatly exceed the rates used for the open sea. The concentration factors shown are correspondingly low.

frigida, and *Centropages typicus*. In some cases, zooplankton will not move into the upper mixed layer from deeper waters if a thermocline is present (Clarke, 1934).

The observations cited above, and others, suggest that the majority of zooplankton inhabit a relatively thin upper layer of the sea. This should be expected since the upper few meters constitute the site of major food production. Except for the settling of moribund and dead phytoplankton, the herbivorous zooplankton must feed in the upper layers of the sea, since net production occurs only above the level at which the oxygen produced by photosynthesis equals that consumed by metabolic processes in the autotrophs. In the temperate regions, this depth is usually 5 to 50 m (Marshall and Orr, 1928; Clarke, 1946) although it may extend to 150 m in tropical waters (Riley *et al.*, 1949; Wimpenny, 1966).

Soluble-colloidal radionuclides introduced at the surface of the sea distribute through the mixed layer by physical processes but do not diffuse rapidly down through the pycnocline. Those plankton that normally live below the mixed layer must move into the upper mixed layer to be able to accumulate radionuclides directly from the water or to feed upon contaminated phytoplankton. If they derive their food from sinking organisms, detritus, or particulate excretory products or if they prey upon zooplankton that have moved down from the mixed layer, then the food must have been in the upper mixed layer in order for these deep-living zooplankters to become significantly contaminated.

Large sources of detritus would be required in the upper zones of the sea to support significant populations of detritus-feeding zooplankton. The food for these animals, derived from plants and animals living in contaminated surface waters, would contain varying amounts of radionuclides. Incorporation of radionuclides would occur through metabolic uptake by the food organisms or through accumulation directly onto the nonliving debris by adsorptive processes. Detritus often accounts for from 40 percent to more than 90 percent of the suspended organic particulates in the upper mixed layer where phytoplankton are most abundant (Krey, 1961), and this nonliving material may provide significant amounts of food and radionuclides for omnivores if its origin is in the upper mixed layer. Because the primary source of food is in the surface layers of the sea and usually decreases in amount with depth, it is not surprising that the herbivorous and omnivorous zooplankton live mainly in the upper waters. Carnivorous zooplankton must feed where their prey are available, and although they may have a greater vertical range than the omnivores and herbivores, they, too, often tend to become concentrated in the upper layers.

Zooplankton living in the upper mixed layer metabolize radionuclides whether or not they migrate into deeper waters during the daylight hours. For those zooplankton that do not undergo vertical migration in and out of the upper mixed layer, all of the biological transport of radionuclides into the deeper water would result from the influence of gravity upon fecal pellets, moulted exoskeletons, and dead animals. At equilibrium, the only fraction of the ingested radionuclides that would not be carried down by this mechanism would be that incorporated by growth and the amounts excreted by the animals back into the water of the mixed layer in soluble or colloidal form plus the fraction of the radionuclides dissociated into solution or colloidal suspension from the fecal pellets and moults as they settled. If migrating and nonmigrating individuals feed at equal rates in the surface waters, individuals that migrate downward diurnally from the mixed layer would not incorporate as much radionuclide into their body structure or into fecal pellets per 24 hours as would those animals not migrating, because of the shorter time spent in the contaminated zone. In the migrating forms, only a fraction of the total ingested radionuclide would be transported by vertical migration. The remainder would be transported by fecal pellets and moults. Since filtration rates of zooplankton generally decrease with decreased temperature (Raymont, 1963), the zooplankton remaining in the warmer surface waters would ingest more food per day than those migrating to the colder deeper waters where their metabolic rates would also decrease.

One of the most definitive investigations of the role of zooplankton in the incorporation, turnover, and transport of a radionuclide by vertical migrations in and out of a contaminated marine environment was carried out in the Pacific Ocean by Kuenzler in 1962 (Kuenzler, 1965, 1969).

In the area sampled by Kuenzler, the copepods constituted 62 percent of the total plankton to a depth of 500 m, and 34 percent to 40 percent of this group stayed in the upper mixed layer at all times of the day. This distribution is in general agreement with those observed by Nicholls (1933) and other investigators. The total volume of plankton, down to a depth of 500 m, was only 6.8 g/m^2. An average 4 g/m^2, or 58 percent of the total zooplankton, spent at least part of each day in the upper mixed zone, at depths less than 100 m. About 41 percent of the plankton in the surface layers underwent diurnal migration down into the thermocline. Thus, only about 24 percent of the total zooplankton population, living to a depth of 500 m, migrated in and out of the upper mixed layer into which the radioactivity was introduced. This observation is in contrast with the assumption often made that a major fraction of the zooplankton participate in vertical biological transport of radionuclides out of the upper mixed layer.

If the amounts of migrating and nonmigrating zooplankton in the upper mixed layer are known, one may estimate the relative efficiency of vertical transport by the different biological mechanisms, including diurnal vertical migration and the sinking of fecal pellets and dead or moribund animals. The relative effect of these different transport mechanisms is modified by several biological processes, including (a) feeding rates, (b) food utilization, and (c) the relative utilization of trace elements and radionuclides by zooplankton compared with that for nutrient elements.

Effects of Feeding Rates and Utilization Efficiencies upon Vertical Transport

Feeding rates in zooplankton are highly variable and subject to the influence of many physical and biological limits. An average value for daily ingestion of food equal to about 30 percent of their own body weight appears to be a good estimate for zooplankton. Riley (1947) observed that *Acartia* required a daily amount of food equal to almost 30 percent of its own weight, and Smayda (1966) is of the opinion that the value may sometimes equal 100 percent. At the higher feeding rates, however, the rate of utilization of the ingested food is reduced, primarily as a result of rapid passage of the material through the gut, with limited digestion.

The utilization of ingested food, like the feeding rate, is highly variable both within and between species of zooplankton. Part of this variability is due to differences in the fraction of ingested food excreted or voided as feces (Strickland, 1965). A low utilization efficiency would result in enhanced transport of food material to deeper water layers by descending fecal pellets. A high utilization efficiency would favor transport downward by metabolic turnover in zooplankton undergoing diurnal vertical migration.

A utilization efficiency of 10 percent between trophic levels has sometimes been assumed; however, direct measurements of food conversion in several species of marine zooplankton suggest that efficiencies may be significantly greater than this. Reeve (1963) wrote of his investigation on growth efficiency in *Artemia salina*, "The work reported . . . indicates that the conversion of plant to animal tissue could, by special choice of conditions, be maintained at an efficiency of well over 50%." Conover (1964) fed *Calanus hyperboreus* three species of phytoplankton and reported "if the average caloric content of the algae . . . are used to recompute the gross growth efficiencies . . . greater than 60% efficiency can be attained when food concentrations are relatively low."

Monakov and Sorokin (1961) observed 40 percent efficiency in food utilization by the freshwater zooplankter *Daphnia pulex*, and efficiencies for marine zooplankton have been reported as follows:

Organism	Efficiency for Food Conversion (%)	Reference
Calanus finmarchicus	50–80	Marshall and Orr, 1955
Calanus finmarchicus	60–78	Marshall and Orr, 1956
Artemia	10–55	Gibor, 1957
Calanus	14	Cushing, 1955
Euphausia pacifica	10–70	Lasker, 1960
Calanus helgolandicus	74–91	Corner, 1961
Temora longicornis	50–98	Berner, 1962
Calanus hyperboreus	15–20	Conover, 1962
Artemia	9–20	Mason, 1963
Artemia	up to 60	Reeve, 1963
Calanus	10–99	Mullin, 1963

In the genus *Calanus*, the conversion efficiencies varied from 10 to 99 percent with an average value of about 50 percent for the six experiments. The low conversion values reported for *Artemia* by Mason (1963) were postulated by Reeve (1963) to be partly due to the animals not being offered the optimum amounts of food necessary for maximum growth; very small or very large amounts of available food reduce the utilization efficiencies. At a given low level of phytoplankton, the cell concentration passing through the gut of a zooplankter would be sufficient only to maintain metabolic function with no excess left for building of new tissue. At high concentrations of phytoplankton, the pressure from incoming food would move the gut contents through at rates greater than those allowing efficient digestion and assimilation (Reeve, 1963).

Although the average conversion efficiency in nature may be well below 50 percent, laboratory data do not confirm this view. In the present considerations, a conversion of 50 percent of the ingested phytoplankton into second trophic level matter is assumed. This would tend to emphasize the role of direct biological transport by vertical migration of zooplankters from a surface contaminated area in comparison with transport downward by fecal pellet production if the conversion efficiencies are indeed lower than 50 percent under field conditions.

A conversion factor of 50 percent for organic material (based on carbon) does not necessarily indicate an equal conversion factor for all of the trace elements and radionuclides incorporated into, or associated with, food. In many cases, the conversion factors in the zooplankters for elements that have no known biological functions and for the catalyst elements are lower than those for carbon. This results in lower concentration factors for zooplankton compared with the amounts of the element or nuclide in the water. It also results in increased amounts of these elements in the excretory products of the zooplankton, relative to carbon and nitrogen compounds. Silica is an example of an element strongly accumulated by phytoplankton but not by zooplankton.

Concentration factors in marine phytoplankton and zooplankton may be used to provide estimates of the efficiency of utilization of different elements by zooplankton. Conversion efficiencies may be obtained by relating the concentration factors of the other elements to those for carbon in the two trophic levels. If the conversion factor for phytoplankton carbon is 50 percent in actively feeding zooplankton, then elements with the same ratios of concentration factors in the two trophic levels will also be utilized by the zooplankton at an efficiency of 50 percent, under equilibrium conditions. Thus

$$\text{Conv.}_{elz}(\%) = \frac{CF_{cp}}{CF_{cz}} \times 0.5 \times 100 \times \frac{CF_{elz}}{CF_{elp}},$$

where $\text{Conv.}_{elz}(\%)$ is the conversion factor in percent for the element by zooplankton, CF_{cp} is the concentration factor for carbon in phytoplankton, CF_{cz} is the concentration factor for carbon in zooplankton, CF_{elp} is the concentration factor for the element in phytoplankton, and CF_{elz} is the concentration factor for the element in zooplankton.

The major weakness in this method of calculating conversion factors for a variety of elements is the uncertainty of the accuracy of the concentration factors for phytoplankton and zooplankton. Relatively few analyses have been made on phytoplankton and zooplankton collected from one area. Vinogradova and Koval'skiy (1962) did analyses for 25 elements in 4 species of phytoplankton and 7 species of zooplankton collected in the Black Sea. The present calculations for relative concentration factors in the two trophic levels are based upon these and other measurements for individual elements. Concentration factors may be calculated by two methods: (a) comparison of direct analyses of the stable element in the organism and in the water and (b) comparison of the amount of a radioactive tracer accumulated by an organism at equilibrium with the amount of the radionuclide in an equal weight of water. Both methods are subject to sampling and analytical errors. In addition, variations in species characteristics, environmental factors, biological condition of the experimental organisms, and changes in population densities, especially for phytoplankton in the exponential growth phase, may introduce further errors.

Estimates for the utilization efficiencies for several elements by zooplankton feeding on phytoplankton are shown in Table 5. The conversion factors are based upon an assumed 50 percent conversion efficiency for carbon.

Conversion factors in zooplankton are low for barium (3 percent), radium (1 percent), yttrium (7 percent), cerium (3 percent), silicon (1 percent), tin (5 percent), and lead (5 percent). On the basis of these estimates, only small amounts of the seven elements would be incorporated into the biomass of the second trophic level, and almost all of the ingested elements would be excreted in fecal pellets and other excretory products. Fecal pellets appear to provide the major transport mechanism for these trace elements and their radionuclides out of the mixed layer into the deeper waters. This agrees with the observations that those elements with low conversion factors, including barium, cerium, silicon, and ^{210}Pb, have been shown to occur in larger amounts in deeper waters than in surface layers of the sea (Chow and Goldberg, 1960; Hogdahl *et al.*, 1968; Rama *et al.*, 1961; Goldberg and Koide, 1963).

Zooplankton have intermediate efficiencies of utilization for the following elements: zinc (20 percent), scandium (30 percent), copper (13 percent), zirconium (30 percent), phosphorus (25 percent), silver (25 percent), manganese (20 percent), chromium (28 percent), titanium (17 percent), and cobalt (30 percent). The zooplankton would excrete or void 70 to 90 percent of these elements. Of the elements with intermediate conversion factors, silicon and phosphorus show large increases in the water with depth in the open sea. Skeletal silicon tests of diatoms are known to pass intact through the gut tracts of zooplankton and to be voided in the feces; thus, this element would be subject to efficient transport to deeper waters with limited dissolution on the way down. Some phosphorus would be transported to deeper waters through the sinking of dead and moribund phytoplankton and zooplankton. In addition, part, at least, of the 75 percent excreted or voided phosphorus would be combined in the fecal pellets as insoluble phosphates of calcium, strontium, barium, radium, yttrium, cerium, and chromium and would be carried to deeper waters by gravity.

On the basis of the estimates shown in Table 5, more than 40 percent of the ingested nitrogen, strontium, iron, aluminum, gallium, and zinc are converted into biomass by zooplankton. This is true for carbon and nitrogen; however, the high values for iron, aluminum, and gallium are probably due largely to adsorption of hydrated hydroxides of these elements onto the surfaces of zooplankton, with an ensuing low rate of loss. The relatively high values (30 percent) for the apparent conversion of scandium and zirconium, elements with no known biological functions, are probably also due to this mechanism. Excreted fecal material, as well as the surfaces of zooplankton, would also be expected to provide adsorptive surfaces for these elements, with resulting transport downward.

TABLE 5 Estimates of Efficiency of Retention for Zooplankton Feeding on Phytoplankton[a]

Chemical Group	Element	Concentration Factor Phytoplankton	Concentration Factor Zooplankton	Conversion Factor, Phytoplankton to Zooplankton (%)	Concentration, Deep Water/Surface Water
IB	Ag	23,000	9,000	25	>1[b]
	Sr	21	28	85	1
IIA	Ba	17,000	900	3	4
	Ra	12,000	190	1	~4
IIB	Zn	26,000	8,000	20	1.8[c]
IIIA	Al	100,000	100,000	60	–
	Ga	8,000	7,000	60	–
IIIB	Sc	2,000	1,000	30	–
	Y	1,000	105	7	–
	Ce	4,000		3	~4
IVA	Si	17,000	300	1	>100
	Sn	6,000	450	5	–
	Pb	40,000	3,000	5	~2.5
IVB	Ti	25,000	6,800	17	–
	Zr	60,000	25,000	30	–
VA	N[d]	36,000	24,000	43	>10
	P	34,000	13,000	25	>100
VIB	Cr	4,400	1,900	28	1
VIIB	Mn	4,000	1,500	20	1
VIII	Fe	45,000	25,000	40	2.3[c]
	Co	1,500	700	30	>1
	Cu	30,000	6,000	13	1.3[c]
	Ni	5,000	3,000	39	2.7[c]

[a] Values based on an average conversion factor of 50 percent for carbon and published concentration factors of phytoplankton and zooplankton; calculations based on the assumption that the total element is derived from food.

[b] Schutz and Turekian (1965a) (area of high production).

[c] Highest reported value; other reports indicate no change with depth.

[d] Structural elements.

Relative Uptake of Radionuclides by Zooplankton from Food and Water

The calculation of conversion factors in zooplankton from concentration factors is based on the assumption that the zooplankton receive a major part of their trace elements and corresponding radionuclides from food. Not all investigators agree with this assumption. According to Polikarpov (1966),

> The amount of radionuclide transmitted from phytoplankton to zooplankton is a function of phytoplankton biomass. When this biomass is many orders in excess of the natural levels, the zooplankton may concentrate more radionuclide from the phytoplankton than directly from sea water. With the normal biomasses characteristic of seas, the quantity of radionuclides concentrated from the water is far in excess of the same radionuclides absorbed from food.

Polikarpov further wrote,

> The pattern that emerges of the ways in which marine animals concentrate chemical elements is, therefore, as follows. The animals satisfy their requirements for most elements by direct absorption from the surrounding water. Intricate organic substances, on the other hand, are usually derived by the animals in the process of heterotrophic nutrition, i.e., at the expense of other organisms. The main source of carbon for the animals is, apparently, to be found in the food links. A certain amount of phosphorus and trace elements incorporated in specific biologically active molecules is also apparently derived in this manner, although it is also possible that there may be direct utilization of such molecules present in sea water as a result of their release by living marine plants. The greater part of the chemical mineral substances and, therefore, by far the greater part of the corresponding radionuclides are consequently accumulated by marine animals other than with their food.

The statement that marine animals reject trace elements in their food but accumulate carbon compounds from this source while taking trace elements through the integument from a relatively dilute solution is not consistent with the normal metabolic characteristics of animals studied thus far. Osterberg *et al.* (1963), in field studies, observed that surface adsorption played a relatively insignificant part in the accumulation of ^{65}Zn, ^{95}Zr, ^{103}Ru, ^{106}Ru, ^{51}Cr, and ^{144}Ce in copepods, euphausids, and salps. In laboratory studies, Rice (1963) observed that *Artemia salina* concentrated approximately seven times as much ^{65}Zn from food as from water when the concentration of introduced ^{65}Zn was the same in each experiment. The degree to which a trace element or radionuclide is accumulated from the food or from the surrounding water appears to depend upon the relative concentrations of the element in the two sources. Food in which an element is concentrated only slightly over the concentration in seawater supplies a relatively low fraction of the element.* However, when the element is highly concentrated in the food, compared to seawater, a major fraction of the element may be derived from the food through the gut.

It is significant that all of the experiments quoted by Polikarpov supporting the concept of direct uptake of radionuclides from seawater (in which the radionuclides were identified) were concerned with uptake of elements for which low concentration factors exist in the food organism. Uptake of this group of elements directly from the water could, indeed, occur. In the case of ^{90}Sr, an average concentration factor of about 20 in phytoplankton and 30 in zooplankton may be derived (Vinogradova and Koval'skiy, 1962; Chipman *et al.*, 1958). If a zooplankter consumed daily an amount of food equal to 0.3 times its body weight and utilized 85 percent of the contained ^{90}Sr (Table 5) it would have to extract all of the dissolved ^{90}Sr in a volume of water about 5 times ($20 \times 0.3 \times 0.85$) its body volume every 24 hours to obtain an equal amount of ^{90}Sr from the water. A medium-sized copepod (*Calanus*) 2 mm in length can graze the phytoplankton from a volume of water about 500 times its body volume (Cushing, 1959). Its oxygen requirements may be met, however, by the extraction of the gas from a volume of water 20 to 25 times its body volume.† Direct uptake of oxygen and ions from the water probably occurs mainly through mucous absorption, and the volume of water stripped of ^{90}Sr would more nearly approximate that stripped of oxygen rather than of food. Under these conditions, 80 percent or more of the total ^{90}Sr accumulated by the zooplankton would be accumulated directly from the water, and 20 percent or less, from the food.

Those elements that are highly concentrated by both marine phytoplankton and zooplankton are not, however, accumulated directly from seawater but, rather, are incorporated from ingested food. The accumulation of ^{95}Zr by phytoplankton and its transfer to zooplankton may be used as an example. The average concentration factor for ^{95}Zr in phytoplankton is about 60,000, and in zooplankton, about 25,000. Zooplankton appear to convert about 30 percent of the ^{95}Zr associated with the phytoplankton eaten for food. If a zooplankter consumed daily an amount of food equal to 0.3 times its body weight and converted about 30 percent of the contained ^{95}Zr, it would have to extract all of the radionuclide present in a volume of water about 5,500 times its body volume ($60{,}000 \times 0.3 \times 0.3$) each day to obtain an equal amount of ^{95}Zr from the water. If the zooplankter is capable of extracting daily the element from a volume of water only 20 to 25 times its body volume, then 99 percent, or more, of the ^{95}Zr would be accumulated from the food.

Transport by Zooplankton, Fecal Pellets, Moults, and Dead Organisms

Calculations may be made to determine the relative roles of diurnal vertical migration, fecal pellet production, moulting, and death upon the transport of radionuclides. These may be made by using the data of Kuenzler (1965, 1969) on plankton distribution, turnover rates, and percent particulate excretion, along with other estimates on biological parameters. These include average feeding rates (0.3 × body weight/day), food utilization (50 percent of carbon intake), and the conversion factors, relative to carbon, for the trace elements shown in Table 5.

A simplified block diagram for downward transport of iron by biological mechanisms is shown in Figure 4. About 92 percent of ^{55}Fe or ^{59}Fe transported through the thermocline by biological mechanisms would be transferred by the influence of gravity on fecal pellets, moults, and dead organisms. Only 8 percent of the element would be transported by vertical migration of the animals. It is difficult to distinguish between direct biological transfer, resulting from exchange of surface-adsorbed material plus excretion of soluble and particulate products of metabolism, and indirect transfer, resulting from fecal pellets and moults. Varying amounts of excreted metabolic by-products may be associated with voided pellets, and surface-adsorbed matter is lost from the organisms through moulted exoskeletons (Jerde and Lasker, 1966; Fowler and Small, 1968).

After equilibration of differences in chemical and physi-

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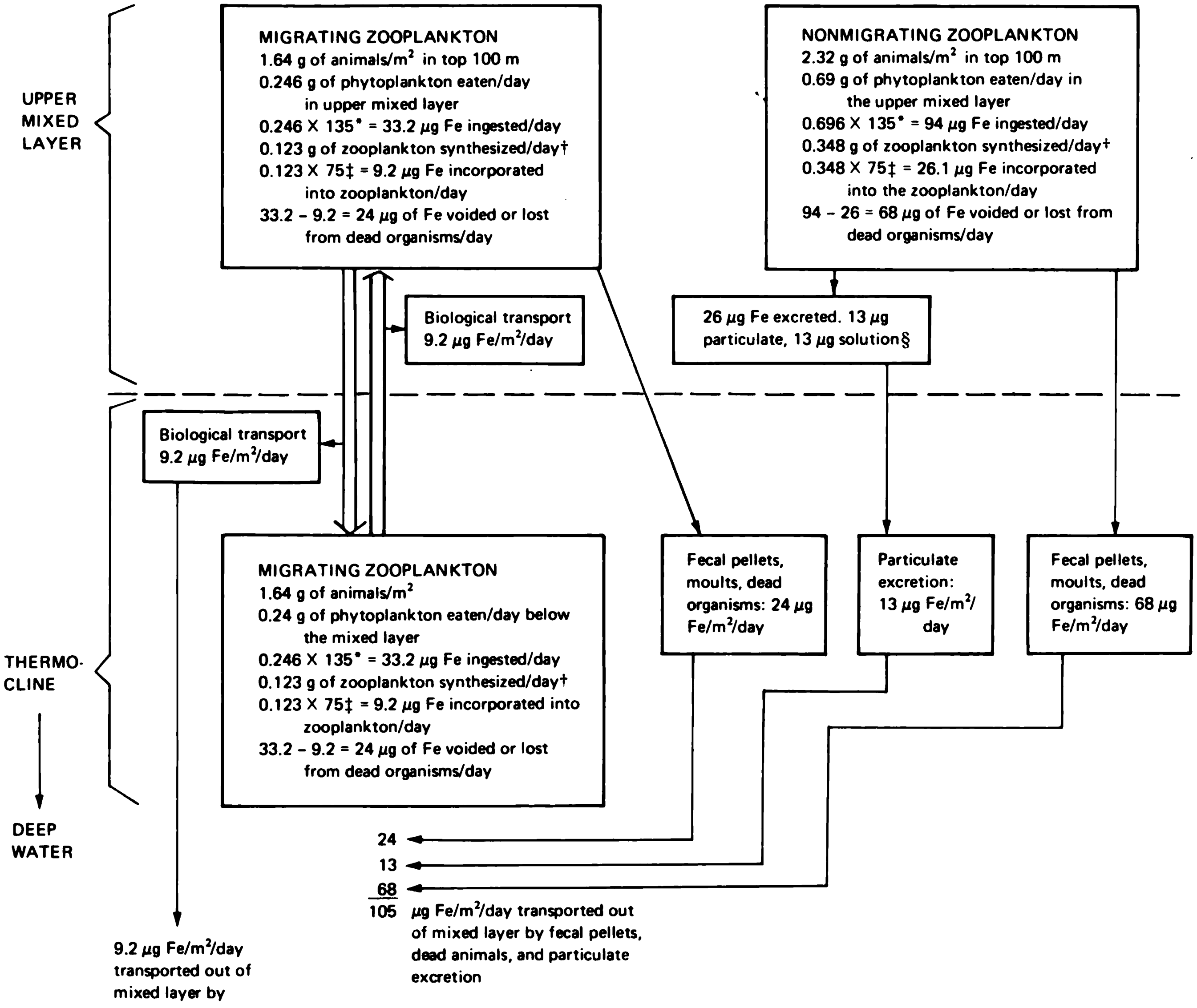
*Food organisms may have high concentration factors for a radionuclide yet contain small amounts at any given time because they are not at equilibrium with the radionuclide in the water. If the food organisms live in an area of rapid dilution or if they continue to grow rapidly after a single exposure to the radionuclide, they will contain a relatively small amount of the contaminant.

†A *Calanus* 2 mm in length has a body volume of about 0.16 mm^3. One zooplankter requires about 0.8 μl of O_2 per hour, or about 19 μl per day (Marshall and Orr, 1955). At 20°C, this amount of oxygen could be extracted from about 3.6 mm^3 of air-saturated water per day, a volume 20 to 25 times the volume of the organism.



*135 µg Fe/g phytoplankton. †Based on 50% conversion of food by zooplankton. ‡75 µg Fe/g zooplankton.
§50% of excreted iron particulate, 50% soluble (Kuenzler, 1965).

FIGURE 4 Block diagram showing the downward transport of iron by biological mechanisms in phytoplankton and zooplankton in the northeastern Pacific (plankton populations based on Kuenzler, 1965).

Iron content in seawater = $3\times10^3\ \mu g/m^3 = 3\times10^5\ \mu g/100\ m^3$.

Transport of iron out of mixed layer by vertical migration = $\dfrac{3\times10^5}{9.2} = 3.26\times10^4$ days = 89 yr.

Transport of iron out of mixed layer by fecal pellets, excretion, and dead organisms = $\dfrac{3\times10^5}{105} =$ 2.86×10^3 days = 7.8 yr.

Transport of iron out of mixed layer by all biological processes = $\dfrac{3\times10^5}{114.2} = 2.63\times10^3$ days = 7.2 yr.

The mean residence time of ^{90}Sr in upper 300 m of the Pacific is 2.7 years (Saiki, 1968). Strontium-90 is not biologically active, and main transport downward is by physical processes. Transport down by biological mechanism is less than by physical factors.

Physical half-life of ^{55}Fe is 2.9 yr. In the time required to transport ^{55}Fe out of the mixed layer by all biological mechanisms (7.2 yr), the ^{55}Fe would decay to about 18 percent of its original activity.

TABLE 6 Time Required to Transport Radionuclides Added into the Surface Waters of the Sea out of the Upper Mixed Layer by Diurnal Vertical Migration, by Fecal Pellets and Dead Animals, and by All Biological Transport

			Years Required To Transport Radionuclides								
	Concentration Factor		Eastern North Pacific[a]			Coastal Areas[a]			Upwelling Areas[a]		
Radionuclide	Phytoplankton	Zooplankton	Diurnal Vertical Migration	Fecal Pellets; Dead Animals	Total Biological Action	Diurnal Vertical Migration	Fecal Pellets; Dead Animals	Total Biological Action	Diurnal Vertical Migration	Fecal Pellets; Dead Animals	Total Biological Action
^{54}Mn	4×10^3	1.5×10^3	1,500	78	74	140	7	7	60	3	3
^{55,59}Fe	4.5×10^4	2.5×10^4	90	7.8	7.2	8	0.7	0.7	3.6	0.3	0.3
^{57,58,60}Co	1.5×10^3	7×10^2	3,200	240	220	290	22	20	130	9.6	8.8
^{65}Zn	2.6×10^4	8×10^3	280	13	12	25	1.2	1.1	11	0.5	0.5
^{95}Zr	6×10^4	2.5×10^4	90	5.7	5.4	8	0.5	0.5	3.6	0.2	0.2
^{210}Pb	4×10^4	3×10^3	2,200	7.3	7.3[b]	200	0.7	0.7[b]	90	0.3	0.3[b]

[a] Zooplankton biomass: eastern North Pacific, 0.04 cc/m^3 (Kuenzler, 1965; Ketchum, 1957); coastal areas, 0.44 cc/m^3 (Riley *et al.*, 1949); upwelling areas, 1.0 cc/m^3 (Ketchum, 1957).

[b] Rama *et al.* (1961) reported a residence time of less than 2 years for ^{210}Pb in the upper mixed layer and attributed the removal to biological mechanisms.

cal form between the introduced material and that already present in the water, radionuclides of iron would be transported downward by biological processes at the same rate as the normally occurring stable element. The total rate of biological transport out of the mixed layer would be 114 μg/day/m^2 under the specified conditions. The total amount of iron in the mixed layer is 3×10^5 μg/100 m^3 (3×10^5 μg/m^2 for a layer 100 m thick). Transport of iron out of the mixed layer by biological mechanisms would require $3 \times 10^5/114 + 2.6 \times 10^3$ days, or 7.2 years. This calculation is based on the assumption that a shear zone exists at the upper edge of the thermocline so that the radionuclides are not transported back into the mixed layer again. Maximum biological transfer is considered to occur, since the assumption is made that redissolution of the radionuclides from pellets or dead organisms does not take place in the mixed layer.

The time of 7.2 years for biological transport of ^{55}Fe or ^{59}Fe from the surface waters appears to be relatively long in comparison with physical transport. Strontium is subject to little biological transport, and the residence time for ^{90}Sr in the top 300 m of the Pacific Ocean is about 2.7 years (Saiki, 1968). Rama *et al.* (1961) reported a residence time for ^{210}Pb in the upper mixed layer of less than 2 years and proposed that the short residence time was due to vertical biological transport. Goldberg (1965) later reported a residence time for lead of 7 years in the upper mixed layer, based on a nonsteady state for stable lead. In the present calculations, the length of time required for ^{210}Pb (or stable lead) to be transported out of the mixed layer by biological activity is about 7.3 years (Table 6)—about the same as for iron. These values are for a relatively productive area of the sea, however, and the average for the entire Pacific Ocean should be less.

The calculations made for biological transport of iron from the upper mixed layer in the eastern North Pacific may be repeated for other elements and radionuclides and for other areas of the sea. The calculations for the times, in years, required to transport ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{95}Zr, and ^{210}Pb from the mixed layer are shown in Table 6 for the eastern North Pacific, coastal areas, and upwelling sites. The areas of the sea considered here represent a wide range of biological productivity with a relatively low standing crop of zooplankton in the eastern North Pacific (0.04 cc/m^3), intermediate amounts in the coastal regions (0.44 cc/m^3), and high amounts in areas of upwelling (1 cc/m^3). According to these data, biological activity would have little effect upon vertical distribution in the open sea. However, the total biological activity of zooplankton could increase the transport of the radionuclides of iron, zirconium, lead, and perhaps zinc into the deeper waters from the surface downcurrent from areas of high coastal productivity and areas of upwelling.

Manganese-54, ^{55}Fe, ^{59}Fe, ^{57}Co, ^{58}Fe, ^{60}Fe, and ^{65}Zn represent elements that are biologically important as constituents of enzymes, vitamins, and molecules that participate in oxygen and electron transport. Of these radionuclides, those of cobalt appear to be subjected to the least biological transport. This is due mainly to relatively low concentration factors in both the phytoplankton and zooplankton. According to the present calculations, the time required for all biological mechanisms to remove the cobalt radioisotopes from the surface mixed layer into the deeper waters of the eastern North Pacific is 240 years, a relatively long time in comparison with the physical half-life of ^{60}Co (5.26 years)—the longest half-life for any of the cobalt radionuclides. Kuenzler (1965) calculated that biological processes would require about 350 years for transport out of the mixed layer but proposed that this transport time might be high by a factor of at least ten as a result of several biological factors. In the present work, estimates for the removal of cobalt radionuclides from the mixed layer by total biological action of the zooplankton—for coastal areas 20 years and for areas of upwelling 8.8 years (Table 6)—more nearly agree with the low value of 35 years calculated by Kuenzler.

Conversion factors for Ag, Zn, Sc, Y, Ce, Si, Sn, Pb, Zr, P, Cr, Mn, and Fe are 40 percent or less, and these elements (or their radionuclides) would be excreted in higher proportions than the organic matter. Those elements that are relatively insoluble in seawater or form insoluble salts would be retained to a high degree in the fecal pellets.

The proposal here that the effect of gravity on fecal pellets constitutes a major biological mechanism for the transport of some radionuclides and trace elements to the deeper waters from the surface is not new. Rex and Goldberg (1958) suggested that the accumulation of micron-size particles of terrigenous quartz into fecal pellets by filter-feeding zooplankton might provide a mechanism for the rapid transport of this material to the bottom sediments. Goldberg and Arrhenius (1958) found barite (barium sulfate) in bottom aggregates that they assumed to be fecal pellets of benthic organisms, and Chow and Goldberg (1960) proposed that the increase in barium with depth in the sea resulted from a high concentration of sulfate ions resulting from bacterial decomposition of detrital organic material, which caused barium sulfate to precipitate. Goldberg (1965) stated "barytes crystals, incorporated in, and sinking with, the decomposing organic matter, may either dissolve in deeper waters during the destruction of their carbonaceous matrix or be incorporated in the sediments on the sea floor." Such a mechanism explains not only the depth profiles of barium, but also the observation that sediments below zones of high organic productivity are enriched with barium. This is also true for deposits containing large numbers of diatom frustules (Goldberg, 1958). Brongersma-Sanders (1967) demonstrated that at least two species of diatoms concentrate large amounts of barium.

The siliceous frustules of these species are not preserved in the sediments, but they may act as transport packages to deeper water for barium (Turekian, 1968) and silica, especially if incorporated into fecal pellets which contain relatively large amounts of sulfate.

Goldberg (1965) proposed that two biological mechanisms for transport downward of Hg, rare earths, and ^{210}Pb were likely to exert significant effects. One of these depended upon the enrichment of the elements in the biosphere with subsequent sinking and continued oxidation of dead organisms and metabolic waste products (including fecal pellets). The other process involved the adsorption of the dissolved metals onto disorganized masses of organic matter with a resulting net transfer from the surface to deeper waters. In both processes, the fecal pellets could play an important role, since they are produced in relatively large amounts, contain significant fractions of the ingested elements, and provide reactive surfaces. Conover (1964) suggested that *Calanus hyperboreus* contributed largely to the food supply of deeper dwelling animals by producing fecal pellets containing at least 50 percent of the energy originally available in the phytoplankton. Johannes and Satomi (1966) proposed that a significant fraction of the energy contained in marine communities was channeled into the production of fecal pellets and that the quantitative sinking of this material out of the euphotic zone would result in rapid transport of large quantities of phosphate into the deeper waters, provided the element was not rapidly redissolved from the pellets. They also postulated that the rate of downward transport of phosphate would be reduced if fecal pellets were utilized by other zooplankton. If this is correct, however, the original fecal pellets would be relatively enriched in phosphorus in comparison with the phytoplankton food (based on transfer of carbon) and the fecal pellets of the omnivores would be further enriched in phosphorus, since the conversion factor for phosphorus in zooplankton is about one half that for carbon (Table 5).

Osterberg *et al.* (1963) presented convincing evidence that some artificial radionuclides are removed from surface waters of the sea to the bottom by fecal pellet production. Samples of sea cucumbers were collected from the bottom at 200-m and 2,800-m depths off the Oregon coast and analyzed for radionuclides by gamma spectrometry. The relative amounts of ^{144}Ce-^{144}Pr, ^{95}Zr, ^{65}Zn, and ^{40}K in the samples from the two depths were used to calculate that the ^{144}Ce-^{144}Pr and ^{95}Zr-^{95}Nb were transported to a depth of 2,800 m in 7 to 12 days but that ^{65}Zn was subject to only limited downward movement below 200 m. Osterberg and his associates noted that the particle sizes of particulate fallout radionuclides were too small to be transported *per se* to the bottom at the observed rates but that these nuclides were known to be accumulated by herbivorous zooplankton from contaminated phytoplankton. The conversion rates for ^{144}Ce-^{144}Pr and ^{95}Zr-^{95}Nb are low in zooplankton, and the authors proposed that the unassimilated radionuclides would be released in fecal pellets that would sink rapidly because of their relatively large size. The fecal pellets of *Euphausia pacifica*, produced by feeding the diatom *Skeletonema costatum* to the euphausid, were found by one of the authors to sink at a rate of 43 m per day. Radionuclides associated with fecal pellets sinking at this rate would be carried to a depth of 2,800 m in about 65 days.

Elements incorporated onto or into fecal material would be efficiently transported downward and released into the deep waters or the bottom sediments only if they were released from the pellets at a slow rate during descent. In general, elements must remain in particulate or colloidal form for continued association with fecal pellets. Radionuclides that would be expected to remain with the pellets would include those that

Form relatively insoluble salts or hydroxides in seawater and are accompanied by carrier amounts of similar or the corresponding stable element

Form strong complexes with biological surfaces

Coprecipitate with inorganic scavengers

Are incorporated into biological structural compounds, i.e., the incorporation of carbon, hydrogen, nitrogen, sulfur, phosphorus, calcium, and silica into skeletal material, lipids, proteins, and polymerized carbohydrates.

Vertical Distributions of Trace Elements and Radionuclides

Distribution patterns with depth of water for several elements and radionuclides determined by different authors are shown in Figure 5. Of these elements, silicon, phosphorus, nitrate, barium, radium, lead, cerium, neodymium, and total rare earths have been observed to increase in amount with depth even in areas of low biological productivity. Silver, in contrast, shows increased amounts with depth only in areas of relatively high productivity.

The structural elements P, N, and Si increase more or less continuously with depth, even in areas of low productivity, down to about 1,000 m, below which little or no increase occurs to 4,000 m. The break at 1,000 m for silicon is not as pronounced as for phosphorus and nitrate, however, and the shape of the distribution curve for silicon appears to be intermediate between the structural elements and those in the next group. The elements radium, lead (^{210}Pb), barium, cerium, neodymium, and the total rare earths* increase slowly in amount down to a depth of about 600 m, with a rapid increase in amounts between 800 m and 1,000 m.

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*Only two values were given by Goldberg and Koide (1963) for the total rare earths (surface and 4,000 m); however, these points both fall near the curve for barium.

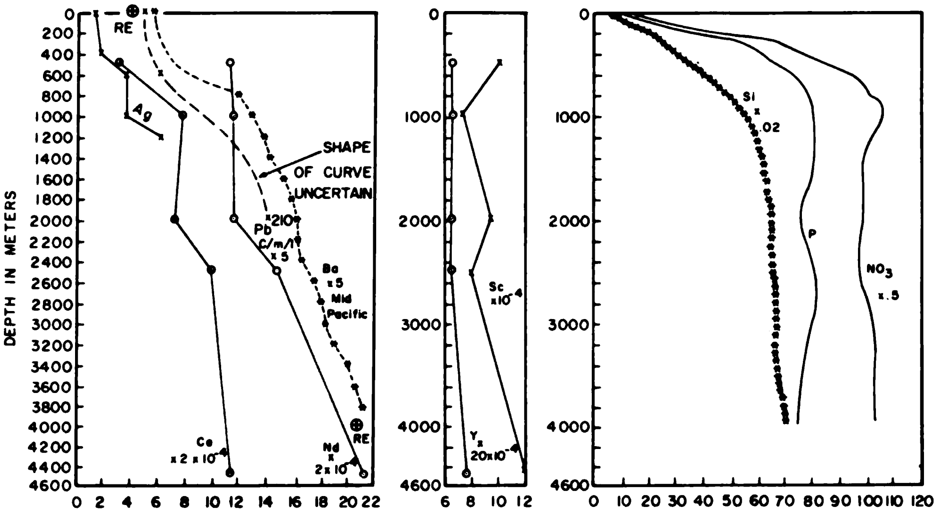

FIGURE 5 Distribution of Si, P, N, Ag, Ce, Ra, Nd, ^{210}Pb, Ba, Y, and Sc in water with depth. Ba—Chow and Goldberg (1960); RE (rare earth)—Goldberg and Koide (1963); ^{210}Pb—Rama *et al.* (1961); Ag—Turekian and Schutz (1965a); Ce, Nd, Y, and Sc—Hogdahl *et al.* (1968); Si, P, and NO_3, average of Pacific, Atlantic and Indian Oceans—Sverdrup *et al.* (1942).

Below 1,000 m, the amounts of the elements in the water increase slowly down to a depth of at least 4,000 m at about the same rate as that above 800 m.

All of the elements and radionuclides in which transport by fecal pellets, moults, and dead organisms (Table 7) constitutes 99 percent or more of the total biological transport show strong increases in amounts in the water with increased depth. These include silicon, barium, rare earths, and lead. Silver, on the basis of concentration factors in phytoplankton and zooplankton, would be transported only about 91 percent by fecal pellets, moults, and dead organisms, yet it appears to have a vertical distribution pattern in areas of upwelling similar to the elements listed above. Schutz and Turekian (1965a) attributed the increase in silver with depth to downward biological transport and regeneration of silver in the deeper waters. This metal is known to be concentrated highly in the digestive glands of crustacea (Palumbo, personal communication, 1959; Seymour, 1963; Folsom and Young, 1965), although it is concentrated only slightly in other tissues of these organisms (Zesenko, 1965; Lowman *et al.*, 1967b).

Although cerium is transported 99 percent or more by fecal pellets, moults, and dead organisms, its distribution pattern does not conform to that expected from the calculated time (2.9 years) to transport the element out of the upper mixed layer by biological action in areas of high productivity. The vertical distribution of the rare earths (Goldberg and Koide, 1963) in the open sea and the observations of Osterberg *et al.* (1963) on the presence of ^{144}Ce-^{144}Pr in sea cucumbers at 2,800 m off the coast of Oregon indicate that the rate of downward transport is much greater than that calculated. The long transport time reported here appears to result from the low concentration factor used for phytoplankton (4,000), based on laboratory uptake experiments (Chipman, 1958). If this concentration factor occurs under field conditions, then the zooplankton must ingest significant amounts of cerium from sources other than phytoplankton, or stable and radioactive cerium must be adsorbed directly onto fecal pellets and other biological debris, including dead organisms, in amounts greater than those accumulated by the phytoplankton. Data to resolve this question are not available at the present time, although it is known that cerium shows a marked propensity for adsorbing onto surfaces, both organic and inorganic.

The similarity in vertical distribution patterns of phosphorus and nitrate is probably due to their being incorporated into structural compounds of phytoplankton with approximately equal concentration factors and to both being released from structural biological material through breakdown by bacteria. Although bacteria would also decompose the protective layer covering the frustules of diatoms (Cooper, 1952; Lewin, 1961), the release of silica, *per se*, would result from ordinary inorganic re-solution of the silica tests and not from decomposition by bacterial oxidation.

Iron, copper, zinc, cobalt, manganese, and, perhaps, nickel are associated with organisms as constituents of reactive biological molecules. These catalyst elements occur both in plants and animals, and, except zinc, all are members of the first series of transition elements, which form essentially covalent linkages as well as ionic complexes.

In general, the catalyst elements are not as firmly incorporated into biological structural material as are the structural elements carbon, hydrogen, nitrogen, and phosphorus. Some chelates of copper and iron exist as hemes, however, which are extremely stable, although they are associated with free erythrocytes or are in solution in the blood of marine animals. Cobalt is an integral part of vitamin B_{12} that appears to be extremely stable in the marine environment. Other chelate complexes with transition elements are not as stable as the hemes and B_{12}, and in some enzymes the metal-protein complex is less stable than the metal-free protein (Lehninger, 1950).

In addition to their being incorporated into biological catalysts, the transition elements and zinc are also capable of forming complexes with biological surfaces so that appreciable amounts of these elements associated with marine organisms may not be incorporated into the organisms but only adsorbed onto surfaces. Thus, Wolfe (1970) estimated that as much as 96 percent of the total zinc in the oyster *Crassostrea virginica* was not incorporated into physiologically important molecules but was loosely bound to cellular components from which it could be removed by dialysis without impeding the zinc-dependent enzyme action of alkaline phosphatase. As a consequence of the lower degree of stability of complexes between such loosely bound elements and the biota, compared to incorporated structural elements, they would be expected to be remineralized rapidly from organic detritus, including fecal pellets and moults (Fowler and Small, 1968) and from dead organisms more rapidly than the integrally combined structural elements or those elements forming insoluble salts or hydroxides in seawater.

Conflicting observations have been made on the depth distribution of iron, nickel, copper, and zinc in the sea. Corcoran and Alexander (1964) measured the amounts of nickel, iron, and copper in the Atlantic Ocean off Florida and observed increases with depth. However, in a later study of the area, they found a reverse distribution for copper, with the metal occurring in slightly greater amounts in the surface waters than at depths down to 800 m (Alexander and Corcoran, 1967). No correlation was found between phytoplankton (based on chlorophyll A) and copper levels. Schutz and Turekian (1965a) observed increases in concentration with depth for silver and cobalt in three out of four areas of high productivity (upwelling) but did not find significant increases in nickel. Morita (1955) found that the deeper waters of two deep bays in Japan were slightly enriched in copper and zinc, although the differences from surface values were not great. Slowey (1966) reported that the distribution patterns of copper and zinc were similar off the Pacific Coast of South America but were different from that of manganese. The rates of biological transport for zinc and copper are similar and are about six times that for manganese (Table 7); thus, the difference in distribution pattern would be expected. Along the west coast of South America, Slowey (1966) observed that the greatest amounts of copper and zinc were found in areas of upwelling and high biological productivity. Upwelling areas may provide sinks for biologically important elements. Because surface currents transport phytoplankton and zooplankton downwind from

TABLE 7 Calculations of Transport of Trace Elements and Radionuclides Downward by Vertical Diurnal Migration of Zooplankton and by Sinking of Fecal Pellets, Moults, and Dead Animals and Time Required for Zooplankton to Transport Radionuclides, Introduced at the Surface, out of the Upper Mixed Layer[a]

	Percent of Vertical Transport by Zooplankton		Time in Years to Transport Element out of the Mixed Zone by Biological Processes[b]	
Radionuclide	Diurnal Vertical Migration	Fecal Pellets, Moults, and Dead Animals	Eastern North Pacific	Upwelling Areas
^{14}C	12	88	94	3.8
^{32}P	6	94	9.6	0.4
Si	<1	>99	17	0.7
^{54}Mn	5	95	74	3
^{55}Fe	8	92	7.2	0.3
^{60}Co	7	93	220	8.8
Ni	9	91	66	2.6
^{64}Cu	3	97	11	0.4
^{65}Zn	4	96	12	0.5
^{95}Zr	6	94	5.4	0.2
^{110}Ag	9	91	11	0.4
^{140}Ba	<1	>99	17	0.7
^{144}Ce	<1	>99	73	2.9
^{210}Pb	<1	>99	7.3	0.3

[a] See Figure 4 for method of calculation.
[b] Does not include transport by sinking of dead phytoplankton.

the areas of high productivity, a continuing downward rain of organic debris would pass out of the upper mixed layer through a shear zone into the deeper layers of water returning toward the areas of upwelling. With the trapping of the biologically incorporated elements in the system and the continued addition from incoming deeper water, the elements may build up to levels several times those found in the open sea.

Menzel and Spaeth (1962) were unable to demonstrate that more iron was present in the deeper waters of the Sargasso Sea than in the surface layers. In areas of high biological productivity, however, the distribution patterns of iron, nickel, copper, zinc, and silver appear to be related to biological activity. Menzel and Ryther (1961) showed that iron was a limiting factor in the growth rate of phytoplankton in the Sargasso Sea, an area of relatively low productivity, although the plants did not effect a change in vertical distribution of the element. Menzel and Spaeth (1962) also did not find a seasonal cycle for iron in the Sargasso Sea, but Thompson and Bremner (1935) and Armstrong (1957) found marked seasonal variations of iron in Puget Sound and the English Channel, respectively. Seiwell (1935) observed total depletion of iron in the surface waters of the Gulf of Maine during August, although the metal was present in the waters below a depth of 40 m. Raymont (1963) stated that the reduction of iron in the upper layers might, at times, become temporarily limiting.

Thus, the vertical distribution patterns of the catalyst elements appear to be significantly influenced by biological action only in areas of high productivity and high density of organisms—usually near land masses with upwelling or addition of nutrients from runoff of surface water. Biological influence upon phosphorus, nitrogen, silica, radium, lead, barium, and the rare earths would also be greatest in these regions. The radionuclides considered in Tables 2 and 3 whose distribution patterns may be influenced by biological activity in marine areas of high productivity include ^{144}Ce-^{144}Pr, ^{210}Pb, ^{110}Ag, ^{65}Zn, ^{55}Fe, and ^{59}Fe. The radionuclides ^{32}P (half-life 14.3 days) and ^{95}Zr (half-life 65 days) would not be subject to significant biological transfer because of their short physical half-lives. Another radionuclide, ^{226}Ra, occurs naturally and has a long physical half-life. Although this element is used in medicine, industry, and research, its large-scale introduction into the sea by man does not appear likely.

ESTUARIES

Characteristics

Estuaries, salt marshes, bays, sounds, and shore lagoons lie geographically, geochemically, and biologically between the terrestrial and marine hydrospheres. They have highly diverse physical, chemical, and biological characteristics that affect the distribution and transport of trace elements and radionuclides. These characteristics include depth; width and shape of submerged areas; seasonal and daily changes in sunlight and water temperature; rate of evaporation; height of tidal excursion and strength of tidal bores; geological substrates of neighboring watersheds and rate of addition of river colloids, dissolved ions, and suspended sediments; wind direction and velocity; strength and direction of adjacent ocean currents; and the rate of exchange of estuarine water with that of the open sea. Estuaries may be broadly divided into marine-dominated, river-dominated, and evaporate categories (Wood, 1965). Because of the diversity exhibited by estuarine environments, all types cannot be considered individually in this discussion; however, the more important characteristics of estuaries that influence the interaction of the biota with added trace elements or radionuclides will be reviewed.

Influence of Sedimentation upon Biological Availability of Radionuclides

Estuarine areas differ from the open sea in several features that alter the relative influence of the water, organisms, and bottom sediments upon trace element and radionuclide transport and distribution in the two environments. In the sea, limited sedimentation processes are slow, and the depth–solubility relationships for elements in the deeper waters plus the length of time required for particles to sink to the bottom result in the return of most elements to the water as ions or colloids before they reach the bottom. In contrast, relatively high rates of sedimentation occur in estuarine areas.

Estuarine nutrients are supplied mainly by surface runoff, which leaches organic and inorganic materials from the watersheds. The nutrients are transported by rivers, primarily in solution or in colloidal form, although some rivers of high velocity may contain large amounts of suspended sediments, especially during flood conditions. Variable amounts of biologically important elements, including iron, manganese, cobalt, zinc, copper, molybdenum, and phosphate, are supplied by the rivers, usually in great excess of the amounts the photosynthetic plants are capable of utilizing or even accumulating. Nitrate is also supplied by rivers, but, unlike the elements listed above, it is usually introduced in small amounts and often may be a limiting factor for organic productivity in estuarine areas (Ketchum and Bowen, 1958). Other elements with no known biological function may also be supplied in amounts exceeding their solubility in seawater. These include scandium, silver, zirconium, lead, and cerium (Carpenter and Grant, 1967).

When rivers enter estuaries, current velocities decrease, and, as a result, much of the suspended sediments sink at

TABLE 8 Distribution of Total Mass, Cobalt, Iron, Manganese, and Zinc in the Water Sediments and Biota of a Texas Salt Marsh[a]

Sample	Weight[b]		Co		Fe		Mn		Zn	
	g/m^2	%	mg/m^2	%	mg/m^2	%	mg/m^2	%	mg/m^2	%
Bay water (1 m)	1×10^6	96.7	0.5	3.1	30	0.1	5	0.2	8	0.7
Sediment (3 cm)	3×10^4	2.9	12.0	73.6	30,000	95.2	2,740	79.5	900	71.5
Plants (*Thalassia* and *Diplanthera*)	3.6×10^3	0.4	3.8	23.3	1,496	4.7	700	20.3	350	27.8
Animals (Mullet, pinfish, crab, brown shrimp)	2.4	0.01	0.0012	0.01	0.414	0.01	0.075	0.01	0.173	0.01
Total	–	100	16.3012	100	31,526	100	3,445	100	1,258	100
$\frac{\text{Water}}{\text{Sediment}}$	33		$\frac{1}{24}$		$\frac{1}{1{,}000}$		$\frac{1}{550}$		$\frac{1}{110}$	

[a] Reprinted with permission from Parker *et al.*, 1963.
[b] Wet weight for water; dry weight for other samples.

rates dependent on the size and mass of the particles. Upon mixing of the river water with the saline waters of the estuaries, iron, aluminum, gallium, titanium, zirconium, scandium, and dissolved silica precipitate into hydrous gels, and the coagulation of colloidal clay particles occurs because of the increase in pH and electrolyte content of the water. Simultaneously with precipitation of these colloids, magnesium and calcium may exchange with some, but not all, of the cations adsorbed to the suspended particles of river sediment. Zinc, at least, is not accumulated by Columbia River sediments through a simple ion-exchange reaction. Johnson *et al.* (1967) observed that only an insignificant fraction of ^{65}Zn (0 to 3.3 percent; average 0.76 percent) was removed from freshwater Columbia River sediments by artificial seawater. In contrast, 0.05 M $CuSO_4$ solution removed 33 to 54 percent of the radionuclide. In nine samples of Columbia River sediment, about 40 percent of the ^{54}Mn and none of the ^{51}Cr or ^{46}Sc could be removed by the action of artificial seawater. The authors attributed the behavior of the ^{65}Zn to "specific absorption" (Tiller and Hodgson, 1962), in which the metal sorbed to the sediment cannot be displaced by alkali or alkaline earth metals but may be displaced by other transition elements. The general order of displacement from sediment or soils is $Cu^{+2} > Co^{+2} > Zn^{+2} > Mn^{+2}$.

Precipitation and coprecipitation of river materials is primarily a physicochemical process. The ensuing association of the precipitates with suspended and bottom sediments, detritus, and biota does not result entirely, however, from inorganic mechanisms. For some elements, at least, the bacteria and other microorganisms associated with the surface of the sediments, detritus, and organisms appear to play a significant role in the adsorption and desorption of fine precipitates and ions from and to estuarine waters. In addition, benthic filter feeders may accelerate sedimentary processes in some areas by producing fecal particulates from ingested colloids and fine particulates (Duke *et al.*, 1966).

Estuarine sediments are of primary importance in determining the distribution and availability of added trace elements and radionuclides to the water organisms. This is true because the sediments present the greatest total reactive surface of the estuarine components, contain large amounts of adsorbed periphyton, and are subject to gravity. Sedimentation in estuarine regions is highly efficient, with rapid removal of most nonconservative elements from the water to the bottom, so that only a minor fraction of this material, introduced by rivers, reaches the open sea. Thus, the sediments retain added trace elements and radionuclides in estuaries so that they are available to organisms for longer periods of time than if the trace elements remained in solution or were suspended in the water.

Parker (1962, 1963) and his associates calculated the mass, per square meter, of the water, upper sediments, and organisms in a Texas salt marsh and determined the distribution of available cobalt, iron, manganese, and zinc under equilibrium conditions in the system. With an average water depth of 1 m and a 3-cm depth of sediment available for exchange reactions, the water accounted for almost 97 percent of the total mass, the sediments about 3 percent, and the total biomass about 0.35 percent (Table 8). Although the water represented most of the mass, it contained only minor amounts of the four elements, accounting for about 3.1 percent of the cobalt and less than 1 percent of the iron, manganese, or zinc. In the sediments, only extractable or biologically available element contents were considered. Even with this restriction, 70 to 95 percent of the four elements were associated with the sediments. The water-to-sediment ratios were 1/1,000 for iron, 1/550 for manganese, 1/110 for zinc, and 1/24 for cobalt, suggesting that iron and manganese were more strongly influenced by sedimentation processes in this type of estuary than were zinc and cobalt.

Turekian and his coworkers (Carr and Turekian, 1961; Turekian and Schutz, 1965; Schutz and Turekian, 1965a and b; Turekian, 1965,1968; Kharkar *et al.*, 1968) considered in detail the removal of trace elements from rivers and seawater in nearshore areas and emphasized the action of the biota in removing added cobalt, nickel, and silver, supplied by streams, to the sediments. Kharkar *et al.* (1968) studied the adsorption of several elements onto ferric hydroxide, manganese dioxide, and clays under conditions that simulated the river environment, followed by desorption in seawater. In experiments with freshly precipitated ferric hydroxide, 79 percent of cobalt, 50 percent of silver, and 74 percent of selenium originally dissolved in fresh water remained associated with ferric hydroxide precipitated under estuarine conditions. In nature, this reaction could result in significant sedimentation of these three elements. In similar experiments with clay, 27 to 55 percent of cobalt, 14 to 25 percent of silver, and 15 to 35 percent of selenium supplied by the streams would be carried to the bottom sediments by settling clay particles in the marine environment. Manganese hydroxide, in general, was least effective in coprecipitating several trace elements, with 20 percent of total stream cobalt, 4 percent of total stream silver, and 34 percent of total stream selenium remaining associated with the particles in salt water.

Jones (1960), in laboratory experiments with radioruthenium, observed that the uptake of nitrosyl ruthenium by sediments was directly proportional to their surface areas and that, once adsorbed, less than 7 percent of the ruthenium could be removed from the marine sedimentary particles by seawater even at pH values of 6, 4, or 2. He observed further that ferric hydroxide was capable of interacting with the radioruthenium and the sediments, enhancing the uptake and retention of the element. Jones reported

> Where rivers flow to the sea via iron ore deposits, iron is brought to the sea in relatively large quantities particularly as colloidal particles. . . . When ferric ions are added to sea water containing (Amersham) nitrosyl Ru^{106} and shaken

> for six hours, there results an insoluble complex between the iron and ruthenium which can be removed by centrifugation. In such an experiment . . . some 70 percent of the Ru^{106} was removed at an iron concentration of 1 ppm.

Jones observed that by doubling the amount of iron normally adsorbed onto the sand fraction of the marine sediment (1.4 mg Fe/g sand to 3.1 mg Fe/g sand), the uptake of ^{106}Ru from seawater also doubled when the sand was subsequently placed in the contaminated seawater. In field studies of the accumulation of nitrosyl ruthenium by fine particles and marine organisms near the outflow of the Windscale Works on the Irish Sea, Jones attributed the major uptake of radioruthenium to physical adsorption. Templeton and Preston (1966) reported that concentration factors for ^{106}Ru in British nearshore sediments, or for suspended matter that later settled out, were largely a function of particle size and ranged from 10^2 for coarse sands to 10^4 for finely divided silts. Even in productive areas the top centimeter of bottom sediments may provide a mass three times that of the total biomass (Parker, 1962), and minimum concentration factors of 3×10^2 to 3×10^4 in the biota would be required to equal physical adsorption by the top centimeter of sediment. Frequent resuspension of the top several centimeters of bottom sediments by wind action and turbulence from tidal currents would increase even more the relative adsorption efficiency of the sediments.

Hampson (1967) measured the dispersion of ^{95}Zr and ^{95}Nb in estuarine areas. These elements adsorb strongly to surfaces, in comparison with ^{106}Ru, which represents radionuclides of medium adsorptive characteristics. Concentration factors for ^{95}Zr in the sediments varied from 4×10^4 to 1×10^5 and for ^{95}Nb, from 6×10^5 to 8×10^5, compared with the average activities of the two radionuclides in the water over a 4-week period. These concentration factors were 100 to 200 times as great as those measured in the benthic alga *Porphyra* for the same radionuclides (CF ^{95}Zr, 410; CF ^{95}Nb, 430), and the higher values measured in the sediments are as great as or greater than any reported for any marine organisms, for ^{95}Zr or ^{95}Nb. According to Hampson, "The high concentrations of ^{95}Zr and ^{95}Nb found on sediments at Dounreay indicate a strong scavenging of these two nuclides from the sea water by siliceous and detrital material."*

In addition to the rapid removal of iron, aluminum, silver, cobalt, ruthenium, manganese, scandium, zirconium, and niobium to suspended and shallow bottom sediments, other elements, including zinc and phosphorus, are also concentrated by the sediments. Orr (1947) and Marshall and Orr (1948) observed that large amounts of phosphate disappeared from a semi-isolated shallow marine basin at rates much greater than could result from phytoplankton production. Bachmann (1963) showed that oxidized sediments could remove 96 percent of zinc added to saline waters in 22 days, and Seymour and Lewis (1964) found that 69 percent of ^{65}Zn in the water off the Washington coast in the shallow but open sea areas was associated with particles that were stopped by a 0.45-μ filter. Duke *et al.* (1966) studied the movement of ^{65}Zn (added to the water in solution) through an estuarine ecosystem contained in a walled pond. At the end of 100 days, 99.4 percent of the remaining zinc was in the sediments and the associated periphyton, and 0.6 percent was in the plants and animals.

Evidence that other elements also precipitate from river water added to shallow areas of the sea has been observed by several investigators. The average scandium content of the leaves of sugar cane is about 4.3×10^{-2} $\mu g/g$ of living leaf. The average scandium content of washed leaf detritus from sugar cane in the top layer of nearshore marine sediments in one bay of Puerto Rico is about 5.2 μg of scandium per gram of wet leaf fragment—an increase of about 120 times that of the content in living material. The iron content in the detritus was about 210 times, nickel 3 times, and manganese 2.2 times the amounts of the same elements in the living leaves (D. K. Phelps, personal communication). This is in agreement with the observation of Hood and Slowey (1963) that a large portion of the manganese in the water of the Brazos River of Texas was deposited quickly in the vicinity of the fresh water–seawater interface and that the manganese was not associated with clay. Manganese is known to be coprecipitated with iron under estuarine conditions.

In most cases, the amounts of iron and other easily hydrolyzed elements in seawater probably do not form precipitates large enough to sink from gravity. A large fraction of these precipitates appear to adsorb rapidly and firmly to the surfaces of mineral and clay particles, to phytoplankton and zooplankton, and to particulate organic detritus. These larger particles are usually subject to sedimentation, the rates dependent upon their size, shape, and specific gravity. In addition, particles as small as bacteria, at least, are subject to efficient removal from the water by benthic molluscs, which are capable of essentially complete removal of particulates from volumes of water two orders of magnitude greater than that of the animal volume in a few hours (Lund, 1957a and b).

Effects of Epiphyton on Uptake and Loss of Radionuclides by Sediments

Relatively few investigations have been made to compare the role of bacteria and of inorganic adsorption processes in the uptake of trace elements and radionuclides by near-

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*The conditions quoted by Hampson are for Dounreay. Peculiar to the Dounreay situation is the siliceous and detrital material. According to W. L. Templeton (personal communication), these conditions do not exist in the Irish Sea.

shore suspended and bottom sediments. Microbial epiphyton show a marked propensity for association with particulate material and for the accumulation of ionic and colloidal trace elements. They thrive in estuarine areas supplied with organic food and other nutrients from river runoff in which an abundance of surfaces upon which the microbes may flourish are present, in comparison with the situation in the open sea. Marine bacteria appear to require these surfaces for continued growth and division.

Rubentschik *et al.* (1936) reported that bacteria adsorbed readily onto positively charged mud particles in saline sediments and that 99.2 percent of all the bacteria were adsorbed to the sediments at an optimum concentration of about 2 cells per microgram of mud.* Wood (1953) observed that optimum bacterial adsorption occurred on particles 1 to 2 μ in diameter and that up to 99.8 percent of the cells were adsorbed. The association of periphyton with sediments appears to be directly related to the available adsorptive surface area. Thus, greater numbers of microorganisms are associated with smaller particles. Wood (1965) showed that the amounts of nitrogen in estuarine sediments increased rapidly with decreased particle size. A significant fraction of total nitrogen in some estuarine regions may be associated with the epiphyton in sediments. According to Wood, the microbiota associated with these fine particles are important in food chains because many animals, including mullets, are capable of selecting for fine particles.

For the large amounts of epiphyton in sediments to significantly influence the distribution of added trace elements or radionuclides, these organisms must be capable of accumulating the contaminants at concentration factors comparable to those of inorganic adsorptive mechanisms that operate on the sediment surfaces.

Marine periphyton are known to accumulate several trace elements efficiently. Chipman and Schommers (1968) cleaned the marine clam *Tapes* of bacteria and kept it in sterilized seawater. The shells accumulated 1/22 to 1/36 as much ^{54}Mn as the shells with bacteria, although metabolic uptake into the soft parts of the sterilized and unsterilized clams was the same. Lowman *et al.* (1956) noted that the accumulation of radionuclides by benthic algae at the Eniwetok test site was directly related to surface area of the plants. Several radionuclides, including ^{65}Zn, become rapidly adsorbed to the walls of plastic aquaria containing marine bacteria, although little or no adsorption occurs under sterile conditions. Zobell (1946) pointed out that placing seawater in a container increased the number of bacteria sharply and unnaturally in comparison with natural conditions. This characteristic of aquarium environments has plagued many investigators studying the uptake of a wide variety of radionuclides by marine organisms. In both fresh and marine water, the periphyton exhibit marked ability to accumulate a variety of biologically important radionuclides. However, quantitative measures of concentration factors in these organisms are difficult to determine because of their extremely small biomass and their intimate association with the substrate (Kevern *et al.*, 1966).

Pomeroy *et al.* (1967) observed that relatively large amounts of phosphate and trace metals were adsorbed onto the sediments in salt marshes of the eastern United States. According to Pomeroy *et al.*, there is little doubt that the bacteria adsorbed to the sediment particles provided an important link between the adsorbed material on the sediments and filter-feeding and deposit-feeding benthic organisms. Pomeroy and his associates found that ^{65}Zn and ^{32}P, introduced into the estuarine regions, were adsorbed quickly and locally near the sites of introduction and were not transported appreciably by water during short periods of time.

Because most of the ^{65}Zn and ^{32}P became adsorbed onto the particles of sediment and detritus, the filter-feeding benthic organisms were the first to show detectable amounts of the radionuclides and to reach peak levels of activity. The turnover times in the filter feeders were long, and these organisms provided a relatively large biomass in the area investigated. Pomeroy *et al.* postulated that the benthic filter feeders thus provided a pool for slowing the movements of the radionuclides through the system.

The ^{65}Zn and ^{32}P reached equilibrium between the water and the sediments within 24 hours. With continued additions of radionuclides, the amounts in the sediments would increase until daily loss equaled daily accumulation in the sediments. At equilibrium the amounts in the sediments would greatly exceed the amounts in the water and the amount added each day. Pomeroy *et al.* did find, however, that continuous exchange occurred between the water and the sediments and that the radionuclides became more widely dispersed with time. Pomeroy *et al.* (1965) reported that the exchange of phosphate between the water and the sediment was controlled by two mechanisms, one an inorganic sorption reaction and the other controlled by biological exchange, probably between adsorbed microorganisms and the water. In surface fractions of the sediments poisoned by formalin, the rate of inorganic exchange of phosphorus was only one half to two thirds the rate of inorganic plus biological exchange for sediments with living microorganisms. If the same fraction of total exchange holds for other elements as for phosphorus, then the microorganisms may exert a profound effect upon trace element distribution in sediments. Concentration factors in sediments for several nuclides, based on micrograms of exchangeable iron, manganese, phosphorus, calcium, and strontium per gram in sediment divided by micrograms of element per gram in water (or on activity of radionuclides of zirconium, niobium, and ruthenium per gram in sediment divided by the activity per gram in water), have been reported as follows:

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*At this concentration, the sediment/bacteria ratio would be approximately 300/1.

Nuclide	Concentration Factor	Reference
Fe	2.9×10^7	Ganapathy *et al.* (1968)
Mn	2.8×10^6	Ganapathy *et al.* (1968)
Nb	6×10^4 to 8×10^5	Hampson (1967)
Zr	4×10^4 to 1×10^5	Hampson (1967)
P	7.5×10^4	Ganapathy *et al.* (1968)
Ru	1.0×10^4	Templeton and Preston (1966)
Ca	70	Ganapathy *et al.* (1968)
Sr	33	Ganapathy *et al.* (1968)

In the case of iron and manganese, the extremely high concentration factors are most probably due to inorganic precipitation of iron and coprecipitation of manganese. For the other elements, however, a significant fraction of them and their radionuclides may be accumulated in the sediments through biological activity. Even in the case of coral reefs, in which essentially all of the bottom is calcium carbonate, only a small fraction of the calcium or strontium in the overlying water is sedimented by biological mechanisms.

Oxidizing Bacteria and Reducing Sediments

The combination of precipitation of colloids, physical adsorption to sediments, and uptake by periphyton result in large stores of exchangeable nutrients in the bottom sediments. In these shallow areas of high productivity, large populations of oxidizing bacteria often produce reducing environments containing varying amounts of hydrogen sulfide, especially in areas of limited current and wave action.

Ito and Imai (1955) reported that the bottom sediments 300 m distant from Japanese oyster farms contained an average of 0.7 percent total sulfide but that directly under the oyster rafts the sulfide amounted to 4 percent of the total sediments. According to these investigators, "when the bottom sediment is stirred by turbulence, the hydrogen sulfide in the sediment is liberated into sea water. If the oxygen content of the sea water is high, the liberated sulfide may be left unoxidized." The release of hydrogen sulfide into the water by resuspension of sediments precipitates some of the trace elements introduced into estuaries or sounds (Schutz and Turekian, 1965a). In shallow areas, nickel, copper, zinc, silver, and lead could react with biogenous hydrogen sulfide in the bottom sediments and be precipitated as sulfides.

Evidence that reducing environments and production of hydrogen sulfide are effective in fixing of trace elements in bottom sediments is provided by fossil sediments. Carbonate deposits formed in shallow water rich in organic material often are enriched in vanadium, nickel, copper, zinc, germanium, arsenic, bromine, molybdenum, iodine, and lead (Chilingar *et al.*, 1967). Keith and Degens (1959) have reviewed the similarity of trace element assemblages in sedimentary pyrites and those in organisms and the possibility that the elements were collected by organic material. Arrhenius (1959) discussed the enhanced amounts of zinc, copper, lead, tin, and silver in biogenous apatite, and several authors have reported positive relationships between biogenic carbon and trace elements in shales (Twenhofel, 1932; Mason, 1958).

Sedimentation by Benthic Organisms in Estuaries

In some estuarine regions, benthic filter feeders may exert profound effects upon the rates of sedimentation. Some of the highest concentration factors for trace elements in marine organisms have been reported for benthic filter feeders. Oysters have been reported to concentrate ^{65}Zn up to 250,000 times the amounts present in water (Chipman *et al.*, 1958; Preston, 1966); however, field experiments of Seymour (1966) showed concentration factors about one order of magnitude lower. These differences in concentration factors are probably due to the fact that uptake in oysters is determined primarily by uptake of particulates from the water and not from the element in solution. Analyses of oyster feces and pseudofeces were reported by Haven and Morales-Alamo (1966a), who showed that 70 to 90 percent of the material consisted of illite, chlorite, and mixed-layer clays. Lund (1957a) reported that oysters could clear turbid suspensions of yeast, milk, kaolin, carbon black, fuller's earth, yolk of hen's egg, soluble starch, calcium carbonate particles, isolated chloroplasts, *Euglena*, *Chlorella*, diatoms, and protozoa. Haven and Morales-Alamo (1966b) observed that the ranges of particle size in feces, pseudofeces, and control sediment particulates were essentially the same, with at least 80 percent of the particles under 2 μ and 95 percent less than 3 μ. The mineral compositions were similar, with contents of 70 to 90 percent illite, chlorite, and mixed-layer clay. During a 9-month period, the organic content (assuming 40 percent carbon content) averaged 16 percent in the feces, 12 percent in the pseudofeces, and 10 percent in the control sediments. The particulates in the water filtered by the oysters included about equal amounts of bioseston and abioseston. Although some discrimination against the inorganic particulates occurred, the composition of the excreta suggest that large amounts of suspended sediments were ingested by the oysters. Sediments, with their associated microbiota, were shown above to be capable of concentrating several trace elements at concentrations 10^4 to 10^7 over the amounts of trace elements already preconcentrated in the sediments. In addition, bioseston are capable of adsorbing large amounts of hydrous colloids of iron and aluminum along with their suites of coprecipitated metals, and the utilization of these plants for food would contribute greatly to the trace element accumulation in oysters and other filter feeders.

In some estuaries, the molluscs and possibly the barnacles, tunicates, and zooplankton are more efficient in removing phytoplankton, organic detritus, and other small particulates from the water than are the effects of gravity. Many of these organisms select for the smaller particle sizes. Jǿrgensen and Goldberg (1953) demonstrated that *Crassostrea virginica* retained 2- to 3-μ particles, but that a majority of particles smaller than 1 to 2 μ passed through the gills and were not utilized. Most of the large particles transported by rivers are dropped near the outflows in which few, if any, filter feeders dwell. Sedimentation of larger sand and silt particles is due almost entirely to gravity. Sedimentation of smaller particles in the 1-to-3-μ range often occurs, to a large degree, from the action of filter-feeding benthic organisms—if large numbers of these animals are present. Lund (1957b) showed that the amounts of small particulates in seawater that were sedimented through the feces and pseudofeces of oysters in laboratory experiments were eight times greater than those deposited on the bottom by gravity. Haven and Morales-Alamo (1966b) in similar experiments arrived at a ratio of 7 to 1 for biodeposition to inorganic sedimentation. Of course, relative sedimentation rates by organisms and by gravity are dependent upon, among other things, the ratio of the volume of water to the volume of organisms. However, several field observations and experimental measurements indicate that in the natural environment, too, biological deposition may sometimes equal or exceed inorganic sedimentation from gravity.

In Japan, Ito and Imai (1955) reported that an oyster weighing 90 g (9 g of soft parts) was capable of producing daily a minimum amount of feces equal to 3.3 percent of the live animal weight. At this rate the oyster would produce feces equal to more than 10 times its live body weight per year. These authors calculated that a raft of oysters of 60 m^2 would produce annually 0.6 to 1.0 metric tons (dry weight) of fecal pellets, an amount equal to 10 to 16 kg (dry weight) of fecal pellets per m^2. The top centimeter of bottom sediment in a square meter weighs about 15 kg; thus, a physical sedimentation rate of about 1 cm per year would be required to equal the biological sedimentation from fecal pellets under the oyster rafts.

In a study of biodeposition in the laboratory, Lund (1957b) calculated that oysters covering an acre of estuary bottom would deposit about 7.6 metric tons of fecal material (dry weight) in 11 days. Assuming that this represents a reasonable average for 9 months of the year, an annual deposition of 190 metric tons would result. An acre contains about 4.1×10^3 m^2. The annual deposit of fecal pellets would thus be 46,500 g/m^2. A physical sedimentation rate of about 3 cm per year would be required to equal the biological deposition rate of oysters completely covering the bottom. Oysters could not survive under these conditions, and it appears that the population density of oysters over large areas may be controlled, in part at least, by the production of excretory products. Even in areas where oysters covered only a third of the bottom, the biological sedimentation rates would be high.

Haven and Morales-Alamo (1966b) conducted studies on biodeposition by the oyster *Crassostrea virginica* throughout the year and related variation in fecal production to season. Below 2.8°C during the winter months, measurable amounts of excretory products were not produced. Maximum amounts of feces and pseudofeces were produced during September. These authors stated

> In the lower York River, commercial oyster growers frequently plant to an acre about 250,000 small oysters similar in size to those used in the trough study. From April through October these would deposit about 405 kg (dry weight)/week of solids with a maximum of 981 kg/week; larger oysters would produce greater quantities.

During the interval April through October the young oysters would deposit about 3×10^3 g of excretory products per square meter, equal to about 2 mm of sedimentation per year. Haven and Morales-Alamo noted that

> Biodeposition rates for other common species of invertebrates may equal or exceed that of the oysters, and when the abundance of these animals is considered, the magnitude of the process becomes evident. Barnacles literally cover many wharfs and pilings in the intertidal zone as well as rocks and shells on the bottom. Tunicates compete for space on the same objects and many hundreds may be found in 0.1 m^2. Soft clams and ribbed mussels occur in the shallow intertidal zone and their densities may be as high as several hundred on a square meter.

Biological Productivity and Radionuclide Distribution in Estuaries

Photosynthesis in estuarine areas is accomplished by both phytoplankton and benthic plants. The character of the estuarine region determines the relative importance of the different primary producers. In estuaries near Sapelo Island, Georgia, Ragotzkie (1959) reported that phytoplankton provided little or no production but that marsh grass accounted for about 80 percent and benthic algae 20 percent of the total. In estuaries near Beaufort, North Carolina, Williams and Murdoch (1966, 1969) reported that phytoplankton, marsh grass, and eel grass and benthic algae each accounted for about a third of the total productivity. In other estuarine areas, phytoplankton apparently provide almost all of the photosynthetic activity (Ketchum, 1967).

In shallow, turbid estuarine areas, a major source of trace elements and radionuclides for higher trophic levels appears to be suspended particles of both organic and inorganic origin (Phelps, 1967; Haven and Morales-Alamo, 1966a and b). Thus, in estuarine regions, food webs are probably not as important as they are in the open sea. Significant processes governing the transport and distribution of introduced contaminants in estuarine regions may be divided into physicochemical and biological categories:

Physicochemical

Settling of large particles, with "selectively sorbed" radionuclides, as a result of reduced current flow of rivers

Precipitation and coprecipitation by hydrous gels

Adsorption of gels and coprecipitates onto suspended abioseston, seston, and bottom sediments

Biological

Adsorption of precipitated colloids and selective accumulation of ions by periphyton

Sedimentation by planktonic and benthic filter feeders

Sulfide precipitation and regeneration by reducing muds from bacterial action

Quantitative measurements are not available from which the relative influence of physical and biological mechanisms can be determined for individual radionuclides, although ^{32}P, ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{58}Co, ^{60}Co, ^{65}Zn, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{46}Sc, ^{110}Ag, and ^{144}Ce-^{144}Pr would be rapidly sedimented by precipitation, coprecipitation, and biological activity. Iron and phosphorus would be partially regenerated, at least, under reducing conditions.

Importance of Estuarine Regions to Man

More detailed and quantitative data are needed to evaluate the physical and biological mechanisms that control the distribution and transport of radionuclides introduced into estuarine areas. Many of these areas are of critical interest to man.

The most populated areas of the world are contained in a strip 250 miles wide around the Pacific, Atlantic, and Indian Oceans. Most of the large cities, with their associated industries, are located close to the sea or on rivers that are navigable to the sea. Thus, man's major point of contact with the ocean has been and will continue to be in nearshore areas, especially estuaries. The use and incidental production of radionuclides will continue to increase; some of these isotopes will never reach the open sea but will accumulate instead in estuaries. These regions are important to commercial fisheries since they serve as nursery grounds for larval and immature forms. About 65 percent of all the commercial fish and shellfish harvested in the United States consists of species that occupy estuarine areas during some phase of their life cycle.

Molluscs provide the third most economically valuable fishery in the United States. Because these animals ingest phytoplankton, detritus, and sediment, they are capable of greatly concentrating some radionuclides. They thus may pass relatively large amounts of short-lived nuclides to man, in comparison to the activity that would be accumulated through other food chains or webs in which the food organisms are more selective in their eating habits.

The most significant role of estuarine organisms in contaminated areas is probably that of transporting the radioactive contaminants from the shallow water areas to man. This redistribution of radionuclides may not be significant in terms of the total amounts released into marine areas, but it may occasionally result in the ingestion of undesirable amounts of these nuclides by some individuals eating large amounts of seafood from limited areas over long periods of time. That the return of radionuclides to man does not represent a significant proportion of the total released has been demonstrated by Templeton and Preston (1966). They state that surveys of the seaweed *Porphyra umbilicolis*, used as food by man, show that the annual quantities of ^{106}Ru reaching the South Wales alga *Porphyra* represent less than one millionth of the ^{106}Ru discharged. At the present time, no use of marine food is restricted because of contamination by man-made radionuclides. Restrictions upon food items that may be utilized by man from estuarine regions because of contaminants should not be condoned. Rather, an adequate knowledge of the mechanisms controlling the movements and distributions of radioactive and other contaminants in marine regions must be used to restrict the introduction of these materials to levels that may be safely tolerated in these vital and ecologically sensitive areas.

SUMMARY

A major fraction of the mass and surface area of the biota in the sea is provided by the lower trophic levels of food webs. Biological transport of some trace elements and radionuclides has been attributed to these organisms. One reason is that they are capable of concentrating several elements to levels much greater than their concentration levels in the water; in addition, many zooplankton undergo vertical migration and produce detritus in the form of fecal pellets, moults, and carcasses, which sink because of the influence of gravity. Elements that may be significantly concentrated by marine organisms include structural, catalytic, and heavy divalent elements; heavy halogens; and elements easily hydrolyzed at seawater pH. The amounts of the nutrient elements—phosphate, nitrate, and silicate—increase significantly with increased depth in the sea, in contrast to the

amounts of the conservative elements, which change only with salinity.

The heavy divalent ions—barium and lead, the rare earths, yttrium, scandium, and silver—follow the distribution patterns of the nutrient elements to varying degrees.

Considerations related to biomass, feeding rates, conversion efficiencies, and migratory habits of zooplankton, as well as the chemical characteristics of the elements of interest, suggest that the major downward transport of these elements and radionuclides is effected through the influence of gravity on fecal pellets, moults, and carcasses, with direct biological transport accounting for 10 percent or less of the total movement toward the bottom of the sea.

In estuarine and other nearshore marine regions, the bottom sediments are close to the sites of photosynthesis and to the sites of the introduction of fallout and terrestrial additions of radionuclides. In these marine regions, the bottom sediments and their associated epiphyton often significantly influence the distribution of added radionuclides. In areas containing large populations of sessile filter feeders, these organisms may exert profound effects upon the rates of sedimentation of added trace elements and radionuclides.

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Chapter Eight

ECOLOGICAL INTERACTIONS OF MARINE RADIOACTIVITY

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Ecology considers organisms not as individuals in isolation but as components of systems that include other organisms—plants, animals, and the host of microorganisms—and in which the interrelatedness of the lives of the component species, and of all with the environment, is the primary problem for exploration. Clearly, the dichotomy between the approaches of this chapter and those of Chapter 7 is artificial at best and unavoidably vague at many points; it has been, however, a curious aspect of the development of radioecology (Morgan, 1960; Schultz and Klement, 1963; Polikarpov, 1966) that it has concentrated on the direct effects of radioactivity on organisms and, in their environments, on the effects of organisms on the distributions of radionuclides, to the very considerable neglect of comparable questions in respect to ecosystems. It is our hope in the present chapter to move toward redressing this imbalance, at least to the extent of pointing out problems deserving attention.

It should be clearly understood that no parts of the argument to follow are directed toward the view that levels of radioactivity now in the ocean present hazards to any marine species or ecosystems. Furthermore, present licensing procedures will permit the growth of nuclear power for many years to come without approaching the levels of radioactivity introduced into the oceans by weapons testing. The purely homocentric approach to decisions about safe levels of radionuclide concentrations in the oceans is an incomplete one; nevertheless, we know of no evidence that it has not provided for the protection of man and of the food chains of the sea on which he depends for sustenance.

We believe that the probably large gene pools of most oceanic species, and their efficient recruitment from oceanic areas, provide oceanic ecosystems with considerable resistance in the face of local introductions of radioactivity. Measurable damage to open-ocean ecosystems may well not be encountered except at higher concentrations of radioactivity than man could tolerate. Certainly, experience to date suggests that other factors are of far greater concern. The well-being of man, however, depends on the vigor of the complex ecosystems of the sea; prudence, therefore, demands our continuing study of oceanic radioecology to search for possible weak links in these chains, where radiation stress could be a problem even at comparatively low levels. It is an essential part of marine radioecology research to study the concentration patterns of radionuclides in marine food elements in order to determine how radioactivity can be returned to man. Such studies should always bear in mind the parallel search for problem areas not for man but for the ecosystems themselves.

The oceans and seas are characterized by a physical and chemical stability quite unusual elsewhere in the biosphere. An ecological consequence of this stability has been, apparently, that ecosystems of the open ocean are responsive to smaller changes in environmental variables than are typically of significance elsewhere. In such a situation, continuous measurement of fundamental ecological parameters, e.g., temperature, salinity, pH, turbidity, insolation, O_2-tension, and the like, would be an extremely valuable, as well as obvious, approach. It is, however, extremely difficult to avoid influencing the biological system through the very act of making the desired measurements. An instrument array suspended from a float can cause interesting changes in the ecosystem. Fish are attracted to the float, perhaps initially by the shade. Algae and various sessile organisms with planktonic stages settle on the hard surfaces of the instrument package. Birds landing on the float add nutrients to the system, enriching it. In response to increased primary productivity, the grazers move in, followed by the predators. Thus, the mere presence of an instrument array alters the community structure of the system being studied. There may indeed be operating in ecology a parallel of the "uncertainty principle" in physics. At least the great demand in the ocean for surfaces upon which to settle is satisfied in part and temporarily by the introduction of instruments, bringing to the community new members who previously found the site an unsatisfactory place to reside. At rates often significant in terms of "station time," comparable and related changes are introduced by the presence of a ship or other platform for scientist observers.

In view of this situation, one may hope that the neutrality of computer modeling will permit insights to marine ecosystems that are not susceptible to direct observation, much less to experimental manipulation.

In the jargon of today, ecology might be viewed as the "systems analysis" approach to the living world; as such, it appears to lend itself well to computer modeling as an analytical tool. Modeling as an analytical technique is relatively new and by no means universally accepted or even understood, but we feel it is especially promising for study of marine ecology.

ROLES OF MODELS IN THE FUTURE OF MARINE ECOLOGY

The preparation of Chapters 11 and 12 of *The Effects of Atomic Radiation on Oceanography and Fisheries* (National Academy of Sciences-National Research Council, 1957) and the Geneva conferences (United Nations, 1958) stimulated thinking about some of the large-scale problems of the nuclear age in terms of rather simple mathematical models. It was never expected that models of four mixed "boxes" would encompass all that has been learned about the hydrosphere, atmosphere, and biosphere in the decade just past. The current chapters on physical circulation, chemistry, and biology all have suggestions for improvement. But revisions might not have come so quickly if a provocative model of some generality had not been set forth, e.g., by Craig (1957a, b; 1963).

A commentary on modeling and abstraction as part of general scientific method (Levins, 1966) reminds us that we cannot hope to gain maximum generality, reality, and precision at the same time. Fallout from nuclear tests provided a distinct opportunity for gaining more reality in modeling the atmosphere and hydrosphere—for instance, in permitting recognition of the subcompartmentalization of the troposphere and the stratosphere, in forcing attention to the actual scales and modes of interchanges both within and between the atmosphere and hydrosphere, or in providing direct evidence of the existence of short-circuited pathways for movement of surface water to intermediate depths (Reid, 1965; this volume, Chapter 4). The stage for more precision awaits refined instrumentation and analysis and better sampling, designed to answer questions about the mechanism and biological significance of physical redistribution of nuclides. An irritating and often perplexing aspect of the "refined instrumentation and better sampling" question is that these qualifications are based—or appear to be based—on interaction between the model and the experimenter; selection of improved versions is one part of the continuing process of revision of model as well as of observations.

In a recent meeting on modeling techniques in biological oceanography (Banse and Paulik, 1969), there was general agreement that construction of simulation models has special promise for marine ecology. Although enthusiastic reference was made to the success of models in fishery population dynamics (where models have been built for tuna, halibut, salmon, and marine mammals, among others, and have been of great importance in resource management), few other examples could be found of successful use of complex ecological models in biological oceanography. This impression, that "practical science" is well ahead in the use of models, is strengthened by Watt's (1968) exposition of the potentialities and realities of ecological modeling in resource management. In more theoretical ecology, however, it seems that in the use of models that actively involve organic matter of the biosphere, living and dead, research may be in a stage comparable with that of box models for the physical world 10 years or more ago. In order to avoid mistakes of expecting too much or too little from the modeling of a very complex process, we may note several valuable rules and phases of abstraction.

Early models are very tentative, but should provide a framework for relating many fragments of present knowledge. They are broad enough to encompass a variety of special cases and suggest possibilities for relating these cases to one another, e.g., in an order governed by one or more parameters. The general models may have too few parts to be very realistic, but specific cases may then become differentiated by adding (or subtracting) some components and specifying reactions that are more comprehensive (e.g., nonlinear) than those first tried.

Successive approximations are naturally used to focus attention on parameters (transfer rates) that have relatively greatest direct or indirect impact on the rest of the system or on a component of interest (e.g., man, whales). This sensitivity analysis often shows that some coefficients can vary widely with relatively unimportant impact, while smaller changes in a key factor have crucial effects. Such findings thus guide continuing research to put more experimental or field effort on certain variables than before. They spur us to pursue aspects of the problem that might have been overlooked or at least postponed.

Later refinements, in some cases at least, will permit sound predictions of the effects of disturbances—and of the fate of the disturbing substances—such as from contamination that has not yet occurred and that one hopes may never occur (e.g., thermonuclear war or massive accidental releases of stored wastes). Where releases of radioactivity are anticipated (as in waste-disposal activities or in some technological uses of nuclear explosives), we expect the prerelease predictions to improve the effectiveness of the operation itself, of the postrelease monitoring, and of further scientific research on both effects and dispersal. We discuss below (p. 207) that although stable element ("analytical") concentration ratios must be considered, they offer guides only to the equilibrium distribution of any radionuclides whose release may be of concern. In most predictive modeling, the transient conditions indicated by kinetic models will be of much greater value. (For further discussion, see Chapter 10.)

In each stage of modeling it is of the greatest importance that the model structure be examined for uniqueness. In a report highly pertinent to our discussion of species diversity, Cohen (1968) points out that two models, his own (1966) and MacArthur's (1957), lead to exactly the same numerical predictions; he then proceeds to outline a third model also leading to the same numerical predictions. As Cohen points out, the moral is that the justification of the model simply by its accounting for available data cannot be an acceptable conclusion: "It is necessary to ask what other explanations are available, to determine how these other explanations differ in their observable implications, and to search for data which could discriminate among the explanations."

This caveat applies equally well to examination of the various relations that make up a model: In an attempt to construct a stochastic model of nickel distribution in the Atlantic Ocean, Spencer (1968) found that observed vertical profiles of nickel in the South Atlantic could be predicted by his model only by an approximate doubling of the transfer efficiency assigned to organisms of the southern ocean. The trenchancy of this observation is reduced, however, by the observation that much the same effect on the vertical profile would be produced, after examination of the flux of nickel at the water-rock interfaces on the sea floor, by using a different value and sign for the transfer of nickel between water column and bottom. Since writing this, we have seen the suggestion by Boström *et al.* (1969) that nickel is locally supplied at the sea floor.

Another and more dangerous aspect of model-making is psychological: that the elegance of a model will so overwhelm its creator as to induce him to assume that any data from the real world not encompassed by his model's predictions must necessarily be spurious. Few examples of this can be cited without introduction of invidious overtones; one reasonably neutral example is that of the practice, routine among hydrographers until recently, of drawing smooth curves through vertical profiles of temperature or salinity and of discarding as bad data any points lying well off the smooth curves, on the not unreasonable assumption that sharp discontinuities of density-determining properties of seawater would not occur. In fact, however, the introduction of devices capable of measuring salinity or temperature as continuous functions of depth has revealed many cases of just such sharp discontinuities—many persisting for considerable periods (Wooster and Jones, 1966)—and it is now clear that many of the "bad points" of former years were good points. Too slavish adherence to the "smooth-curve" hydrographic model delayed for some years its testing and the discovery that it is a model only partly applicable to the real ocean.

Numerous comparable cases could be cited, pointing up how dangerous a bond is created between a model and its progenitor. In fact, as indicated above, a model exists only for the purpose of being changed, and only in this process are models fully used to clarify our understanding of nature.

MARINE ECOSYSTEMS

Three major classes of marine ecosystems are clearly recognizable: pelagic, benthic, and near-shore. Although each is so complex as to be subject to almost infinite subclassification, each major class has properties that lead to distinctive responses to introduced radioactivity as well as to more basic ecological factors. Some of these properties are summarized in Table 1.

The major divisions of the marine environment, pelagic and benthic, have subdivisions that are largely a function of water depth. Light intensity decreases rapidly with depth so that most photosynthetic plants are found in near-surface waters in the photic zone. Below the level of light penetration, roughly 50 to 200 m, photosynthesis is not possible. Animals of the aphotic zone must rely on food derived, either directly or indirectly, from primary production in the overlying photic zone.

Pelagic animals, both nekton and plankton, are associated with the water, maintaining buoyancy by flotation mechanisms or by swimming. Benthic animals are associated with the ocean bottom. The benthic and pelagic regions merge at the edge of the sea, forming the near-shore environments

TABLE 1 Properties of Classes of Marine Ecosystems Relevant to Their Responses to Introduced Radioactivity

Property	Pelagic: Open Ocean	Benthic: Open Ocean	Near-Shore: Benthic and Pelagic
Adsorptive surfaces	Largely biological	Bottom and resuspended sediment	Bottom, resuspended sediment, and terrigenous detritus
Physicochemical stability	Quite stable	Very stable	Maximum variability
Productivity	Only in euphotic-zone plankton	None	Euphotic zone plankton and attached to bottom–generally high
Primary nutrients supply	Largely by water mixing	Not important	Largely from land and bottom
Size range of primary producers	Mostly microscopic	–	Small to very large
Size range of herbivores	Mostly small	–	Moderate to large
Food chain length range	Mostly long	Moderate	Short
Population mobility	Maximum–plankton by currents, nekton by swimming	Very low–most by larval travel	Both high and low
Accessibility to man, both exploitation and effects	Intermediate to minimal	Minimal	Maximal

that combine with properties of the other two regions a number of new properties related to proximity to the land surface.

In addition to the implications of these properties for the reactions of the various ecosystems to introduced radioactivity, the availability of radioactivity to them may be expected—and has been demonstrated—to be different. Availability is dependent upon two factors: the foci of introduction of radioactivity and the physicochemical environment into which it is introduced. It is possible that the physical and chemical characteristics of radionuclides introduced near shore are systematically different from those introduced to the pelagic or to the benthic environments.

With the exception of worldwide fallout, radioactive byproducts of man's activities have generally been introduced near shore and will probably continue to be; this has obvious economic bases. Even fallout nuclides not strongly fixed by the land surface—as Slowey *et al.* (1965) have suggested for antimony-125—must be significantly enriched in near-shore waters by land runoff. With the exception of tragic accidents like those of the nuclear submarines *Thresher* and *Scorpion*, the open-ocean benthic environments receive radionuclides only as result of geochemical, biological, or hydrodynamic transport through the other environments. Direct waste-disposal to the deep ocean has thus far involved trivial amounts of radioactivity, although the scale of such disposal may increase as near-shore marine areas (such as the North Sea), or land areas, become no longer able to accept the increasing volumes of waste produced. In terms of exposure to introductions of artificial radioactivity, then, the rank order of the three environment classes is as follows: near-shore, most frequent and severe; pelagic, next; and benthic, least frequent and least severe.

Both benthic and near-shore environments differ from the pelagic in respect to the amounts and kinds of available surfaces. In the benthic and near-shore environments the sediment-water interface plays a major role, and resuspended bottom materials in both, supplemented by run-off terrigenous materials in the second, ensure that in terms of surfaces of absorption the biota have considerable competition. This is not so in the open-ocean pelagic environment, where, in the illuminated zone, most available surfaces must be biological; at greater depths, the competition must be largely between organisms and biogenous detritus, which is, in turn, food for the organisms. Menzel (1967) has described unexpectedly uniform concentrations (in place and in time) of organic detritus particles in the deep ocean; this has not been confirmed, however, by work of Hobson (1967) or of others. Both Menzel and Hobson tend to conclude that sinking of organic detritus particles is almost negligibly slow, leading to the conclusion that their removal by biological processes must be more significant than removal by sinking. This suggests that recycling of organic material is of major importance in the pelagic environment; in other contexts this has been suggested by Ketchum and Bowen (1958) and by Bowen and Sugihara (1965). For surface-active radionuclide waste or debris, then, the fraction immediately associated with the biota may be expected to be highest in the pelagic, lower in the benthic, and lowest in the near-shore environments; clearly, however, the feeding habits of the biota quickly modify the effect of this immediate distribution, and the detritus feeders and "surface scrapers" prevalent in the near-shore and benthic environments may wholly reverse this order of exposure. That this is an important set of variables in considering effects of radionuclide introductions is emphasized especially by the studies of Mauchline and others (Mauchline, 1963) and of Chesselet and coworkers (Chesselet and Lalou, 1964a, b; 1965a, b) concerning the association of fallout or of radionuclide waste with marine suspensoids.

Beyond the differences in basic exposure outlined above, one can discern a variety of differences in detail among the various ecosystem classes that will further distinguish their treatment of, and responses to, introduced radioactivity. These differences are discussed in the next three sections.

The Pelagic Environment

Nektonic animals such as tuna and whales make long migrations through different water masses. Since they are strong swimmers, their travels are not dependent on current systems, and they may encounter different levels of radioactivity in the water, depending on fallout patterns or on local sources. Thus, they act as averagers, and the levels of radioactivity they contain depend somewhat on their individual histories. It is known that tuna, salmon, and whales, for example, accumulate ^{65}Zn in passing through waters off the Oregon–Washington coast. A part of the ^{65}Zn, introduced into the Northeast Pacific by the Columbia River, will be carried in the animals, at least for a time, as they travel into less radioactive parts of the ocean. Local sources of radioactivity offer some promise as tags to use in determining migratory patterns of pelagic animals (Osterberg, 1964; Forster, 1968; Kujala *et al.*, 1969; Jennings, 1968).

Because strength and size are needed to swim against or through ocean currents, nekton generally have a high ratio of mass to surface area and are therefore relatively inefficient at taking up radionuclides directly from the water. Data are not available for evaluation of the importance to nekton of radionuclide uptake through the gill-surfaces or by drinking. As discussed below (see page 215), Polikarpov's (1966) lines of argument lead one to conclude that for predators or grazers, a more important source of radioactivity is the smaller organisms in their diet.

Plankton, on the other hand, are generally regarded as "drifters"—a part of the water mass in which they reside. They may travel long distances by passively moving with a rapid current, or, where water motion is weak, their travels may be quite limited. Certain macroplankton, such as euphausiids, are, indeed, fairly capable swimmers, migrating vertically into surface waters at night and into deeper waters during daylight. Thus, the euphausiids may drift in one direction at night and in a different direction during the day.

Larger plankton, e.g., euphausiids and salps, effectively filter large volumes of water while feeding. Because of their small size, it seems unlikely that fallout particles would be filterable; nevertheless, salps and euphausiids both responded quickly to increased amounts of ^{95}Zr-^{95}Nb after the 1961–1962 atmospheric nuclear tests (Osterberg, 1962; Osterberg *et al.*, 1963b). Either the particles of ^{95}Zr-^{95}Nb were adsorbed on mucous, were directly filtered, or became attached to phytoplankton and smaller zooplankton that were eaten. The last seems most probable, although mucous adsorption is also a possibility.

Since plankton tend to move with the surface waters and equilibrate with them, they can often be used as biological monitors of the radioactivity in their environments. Smaller plankton are particularly quick to equilibrate. Foster and Davis (1955) reported that plankton algae in the Columbia River attained maximum radioactivity within little more than an hour. Probably, as hinted by some of Lowman's data (1960), marine phytoplankton respond equally rapidly, providing an input of radioactivity from the photic zone directly into the herbivores.

In general, among organisms inhabiting surface waters exposed to fallout, the smaller ones are the first to reach maximum radioactivity. A time lag occurs before the maximum level of radioactivity is reached by larger organisms farther up the food chain. Animals from deeper waters take even longer (Seymour and Lewis, 1964). For the majority of midwater pelagic animals, however, the delay in reaching maximum radioactivity may not be large, because of vertical migrations of the organisms or of their food; unfortunately, few data bearing on this point are available.

The data of Osterberg *et al.* (1963a) imply that transport of some fallout radionuclides to the bottom is rapid. In April 1963, they found ^{95}Zr-^{95}Nb in sea cucumbers (*Paelopatides*) from a depth of 2,800 m. Because of the small size of fallout particles, their settling rates would be too slow to account for the radioactivity of the sea cucumbers at this great depth. The authors therefore attributed the increased sinking rates to the processing of fallout radionuclides into fecal pellets by zooplankton, thereby increasing the particle size and the sinking rate.

Pearcy and Osterberg (1967) reported that micronekton and macroplankton from middle depths (to 500 m) show the same seasonal cycle of ^{65}Zn as those from surface waters; this cycle is related to the seasonal change in position of the Columbia River plume. Since no apparent lag occurs with depth, they believe that vertical transport of ^{65}Zn is quite rapid. However, only a portion of the ^{65}Zn reaches deep water.

Near-Shore Environment

The near-shore environment is considerably different from that of the open ocean. Near-shore areas are generally more productive. In shallow seas over the continental shelf, a large fraction of the water column receives sufficient light to support photosynthesis. Nutrients are abundant, being confined to the system by the proximity of the bottom. The bottom itself is especially rich in biota, nourished by the photic zone just above it.

Radioactivity introduced into this environment will be found later in the water, sediment, and biota. The relative abundance of the radionuclide in each compartment depends both on the properties of the sediment and the biota and on the specific radionuclide chemistry. Data presented by Seymour and Lewis (1964) and by Osterberg *et al.* (1963a) show that for demersal fish and some benthic invertebrates from near-shore locations, the content of some radionuclides was smaller the greater the depth at which the organism was caught; this effect may be stronger for ^{65}Zn in Columbia River outflow than for several of the abundant nuclides in worldwide fallout. It was noted by Seymour and Lewis, however, that the fish data could be interpreted as showing differences in physiology at different pressures rather than in radionuclide availability. Even in the unusually well surveyed areas of the Columbia River outflow, we rarely, if ever, have enough data to allow full evaluation of all the complexities of ecosystem responses.

The behavior of radionuclides is especially complex in near-shore environments for a number of reasons. The affinity of the sediment for the radionuclide depends to a large extent on the particle size of the sediment, the amount of organic material present, and the degree of mixing (see Chapter 6). Biological concentration factors of radioisotopes are influenced by the concentrations of their stable counterparts, and these levels, too, can be highly variable in waters affected by adjacent land masses. In short, the near-shore area is subject to many environmental variables.

Benthic Environment

The benthic community in deep waters is only slightly less complex. Many animals live on or slightly below the sediment–water interface. A radionuclide introduced into this environment would be subject to sorption on sediment surfaces or to biological uptake. Chesselet and Lalou (1964a, b; 1965a, b) showed that, at least for some elements, the sediments quickly bind most of the radioisotopes. Benthic organisms that ingest sediment particles may or may not remove the radionuclides from the sediments, depending in part on the pH of their digestive systems. It seems clear that unless plants and animals associated with the sediment-water interface can remove radioactive trace elements from sediment particles, most of the radionuclides introduced into the ocean will eventually end up in the sediments. Several aspects of nonbiological processes resulting in element—and by inference, radionuclide—recycling from bottom sediments to water column are discussed in Chapter 6; that there are purely biological vectors cannot be doubted, but we know little of them. Both predation by deep-water nekton on benthic organisms and redistribution as floating eggs or planktonic larvae of benthic organisms might well prove to be significant processes.

ORGANISM INTERRELATIONSHIPS IN MARINE ECOSYSTEMS

The marine environment is a continuum in a sense not approached by either the land or fresh waters. In addition to the significantly freer movement available to individuals or species that float or swim, the environment ensures that, subject only to their physicochemical stability, the exometabolites of all marine organisms are available to all the others. It is therefore not surprising that, as Connell and Orias (1964) concluded, interaction with other species should appear to be an extremely important factor in the determination of marine ecological niches. Below, we discuss the possibility that this excessive closeness of species in marine ecosystems may render such systems unusually sensitive to radiation; it seems worthwhile also to consider whether there may be aspects of the mechanisms of species interaction that affect the radiation exposure of the individual organisms. The interaction mechanisms that appear relevant in this connection may be classified as follows:

1. Species interactions not mediated by food
    a. Interactions mediated by exometabolites
        i. Exometabolites as attractants, repellents, or antibiotics
        ii. Exometabolites as complex-formers, competing for inorganic nutrients
    b. Interactions not mediated by exometabolites
        i. Indirect stimulation of growth of other species, by increase in rate of regeneration of nutrients
        ii. Indirect effect on the growth of other species, by altering the physical environment
2. Species interactions along food chains

Interactions Not Mediated by Food

Among those interactions not mediated by food, the actions of exometabolites of marine organisms as attractants, repellents, or antibiotics, while of considerable general interest, appear to be essentially neutral with respect to the radionuclide metabolism of organisms. Only specific attractants that are either ingested or adsorbed and are characterized by radionuclide constituents could become significant contributors to the radiation exposure of the attracted organisms. No such examples have been described.

The reaction of exometabolites as complex-formers is, however, clearly of importance in controlling the balance of distribution of adsorbable radioelements between neutral surfaces and organisms. Even excreted orthophosphate may be expected to significantly resolubilize lanthanides attached to sediment surfaces, and such large molecules as the phosphate compounds released by marine plankton algae (Bowen and Campbell, unpublished experiments) or the amino acids known from zooplankters (reviewed by Stephens, 1968) could be expected to be more effective. Such tightly linked species associations as we infer for marine ecosystems can be expected, then, to be unusually efficient in recycling adsorbed elements, whether physiological or not, and consequently to be more subject to radiation exposure from introduced nuclides.

A third aspect of species interlinkage in the oceans is the provision it gives for recycling of specific plant-nutrient elements. The evolutionary value, to an ecosystem, of linkage of this sort is obvious, but such recycling cannot be limited only to nutrients in demand. It was suggested by Ketchum and Bowen (1958) that in some situations and in some environments the same relationships responsible for nutrient recycling should result in long-term retention of nonphysiological elements, or radionuclides, in the euphotic zone. Bowen and Sugihara (1965) described a situation in which this mechanism appeared to have operated for fallout ^{144}Ce. Again, the very aspect of the close-linked system that gave it evolutionary advantage may sometimes act to increase its exposure to radiation.

Especially in near-shore areas, but also in other marine environments, an important aspect of species interlinkage is clearly the physical effect of one species in making the environment more suitable for another, whether in producing shade, in preconditioning—or providing—a settling surface, or in mechanically damping out the force of water movements. It is not evident, however, that the special nature of any of these interactions should increase the radiation sensitivity of the ecosystems in which they operate.

It appears then, that of the various ways in which marine species are especially closely linked, other than as food, some may, by virtue of their special properties, increase the potential for radiation exposure of the ecosystem, while others appear to be essentially neutral. We have not discerned any that appear to have contributed to decreased radiation exposures.

Interactions Mediated by Food or Feeding

"Perhaps the most challenging and complex problem in the ocean is the question of how organic matter is formed and how it moves through the food web of marine organisms."*

Ecologists have found it useful in attempting to define regularities in this problem to erect a variety of hierarchies based on feeding relationships—food chains, food webs, or feeding cycles. In most of these hierarchies, the concept of *trophic level* appears, as an indication of how far removed a species is from direct feeding on the primary producers—photosynthesizing plants. Usually, trophic level I refers to the photosynthesizing plants, level II to the grazers or herbivores, level III to predators on herbivores, level IV to predators on level III predators, and so on. There is general recognition of the factors that blur the distinctions between trophic levels—i.e., that few species have simple feeding habits, that for many species the food emphasis shifts during the life cycle, and that for others it may shift even seasonally. The concept is still useful and has suggested a variety of theoretical and model studies of natural populations, even though in such practice, fractional trophic levels often are required (tuna, for instance, are best viewed as occupying trophic level III½, according to Schaefer, personal communication).

An interesting aspect of the marine, as opposed to terrestrial, food chains is the generally small range of sizes of herbivores; in the open ocean, this appears to be a clear consequence of the generally tiny size of the primary producers, but even in the kelp forests, no herbivores of sizes comparable to the large terrestrial mammals have appeared. It is likely that this size factor is chiefly responsible for the longer food chains of the oceans. The continuity of the physical environment, however, has another important consequence: Other than the efficiency factor, there is no bar to short-circuiting of their food chains by higher trophic levels. A salp or euphausiid is as accessible to a tuna as is a herring—quite unlike otherwise comparable situations on land. And, because of the size differences and consequent increase of the surface-to-volume ratio, such short-circuiting may always be assumed to lead to an increase in exposure of the higher level to adsorbable radioactivity in the environment.

Another possibly important result of this combination of environmental continuity and prevalent small size of prey has been the repeated evolution, by marine organisms, of feeding by means of mucous threads or sheets. It is assumed that these are "intended" principally as traps for small organisms and nutritive detritus particles. We do not know whether there has been positive selection for mucous having strong ion-exchange properties and consequent good efficiency at trapping dissolved organic molecules, or whether this property has developed only as a by-product in the evolution of suitable mechanical properties of the mucous. It is clear, however, that many mucopolysaccharides, especially those with strong sulfuric acid residues, have powerful ion-exchange properties. Goldberg (1957) emphasized this phenomenon in both selective and nonselective bioaccumulation of elements from seawater. Since mucous feeding is usually mediated by actual ingestion of the mucous and its digestion along with the load of trapped food [often on a regular and

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*National Academy of Sciences–National Research Council (1967, p. 52).

rapid cycle (Prosser and Brown, 1961, p. 107)], this habit must greatly increase an organism's exposure to radionuclides in his environment, even those that are wholly nonphysiological and represented by negligible concentration factors.

The generalization has been made that only about 10 percent transfer efficiency per food-chain step is typical for many elements, as it seems to be for energy (Slobodkin, 1968; Schaefer, 1968). When they are studied with radionuclide tracers, many elements have appeared subject to discrimination, or a reduction in mass concentration through each step in a trophic series (see Baptist and Lewis, 1969, for data and a review). Odum (Odum and Golley, 1963), in fact, was so impressed by the parallelism in this respect between transmittal of some elements and energy along food chains that he proposed the use of radionuclides of zinc or phosphorus as "indices of energy flow" in ecosystems. It has been frequently concluded that man, in drawing little food from marine primary producers, only moderate amounts from marine grazers, and most from predators at fairly high trophic levels, places an effective barrier between himself and radioactive nuclides introduced to the oceans (National Academy of Sciences–National Research Council, 1962, p. 52). Examination of the large amount of data now available shows clearly that the situation is by no means so simple; this is, in fact, an example of the "unrealistic safety factors" referred to by Revelle (1959).

The use of stable-element "concentration factors"—derived from chemical analysis of both the medium and the organisms—has been frequently proposed and used to predict both the places of major entry of introduced radionuclides into food webs and the concentrations to be expected in the various organisms involved. The use of such data is basic to the "specific activity approach" (National Academy of Sciences–National Research Council, 1962), but they have proved useful in other contexts as well. Some of the reservations to be applied to their use, however, are serious, as has already been discussed in the consideration of the uses of concentration factors (Chapter 1).

Briefly, we may note here two of the most salient reservations; they stem from our uncertainty of the time constants of element uptake or excretion and also from our uncertainty of the physical and chemical environment in which uptake occurs:

Time Constants of Uptake or Excretion Strictly speaking, one can expect the stable-element and radionuclide concentration factors to be the same only when the specific activity of the radionuclide in the medium has remained about constant over several biological half-lives of the nuclide in the organism. This implies uniform labeling over time as well as space. It is, however, very unusual for us to know the biological half-life of an element in any marine organism; as Kuenzler (1969a, b) has shown, such measurements as can be made are in most cases of questionable generality. The ideal situation must, of course, be approached as the life cycles of the organisms considered are shorter, and the least favorable situation must be approached in those large organisms that immobilize much of a given element metabolite, whether in massive skeletons, in stored reserves, or in stored excreta. Conversely, predictability must be favored in situations where a nearly uniform flux of radionuclide is maintained over long periods, in a relatively restricted environment, as in the Columbia River outflow under constant operating levels at Hanford.

The Physicochemical State of Radionuclides and Stable Element in the Environment Even in the open ocean, we have inadequate data on the physicochemical states of elements or of their nuclides and on the rates of equilibration among these (see Chapter 5). As discussed by Jones (1960), among others, this ignorance has made radioruthenium release behavior essentially unpredictable. Bernhard and Zattera (1969) have presented data illustrating uncertainty of the fractions of seawater zinc available as carrier for introduced ^{65}Zn; data on ^{55}Fe in Pacific salmon may be used to raise the same sorts of question (Jennings, 1968). With respect to benthic organisms, especially those that feed largely within the sediment, there seems to us to be complete uncertainty about what might be taken as the "environmental" concentration of any trace elements; this is a severe bar to using such data as those of Phelps *et al.* (1969) in a predictive way.

Unfortunately, however, the literature dealing with the uptake of radionuclides released to environments appears just as difficult to use for prediction. Either the physicochemical constants of the local environment are too little known to support extrapolation of data to new situations or the local time-constants for exposure levels are comparably little known, or both. This situation is well illustrated in a massive study (Seymour and Lewis, 1964) of the Columbia River effluent contaminations of marine organisms, sediment, and water from 1961 through 1963. In spite of the enormous amount of data collected, often on the same species from several different locations, very little generalization was possible, largely, one feels, because of uncertainty about precise exposure times or levels and because of local variation in stable nuclide diluents. Jennings (1968) points out data (his and those of Kujala *et al.*, 1969) that indicate that "available" levels of several elements (notably Mn, Zn, and Fe) are higher inshore than offshore, and may be subject to local variation as well.

In view of this, we feel the best approach to prediction of radionuclide behavior in ecosystems is still by use of stable-element analytical data, suitably qualified by examination of such tracer studies as seem applicable. Accordingly, we have summarized a considerable number of analyses of marine organisms, mostly of the open ocean; species have been chosen about which some trophic-level conclusions could be drawn.

In Table 2 are set out, from the literature or from data of Bowen *et al.* (to be published), the ranges of concentration factors, for a variety of elements, observed in a variety of primary producers and grazers as well as predators at various trophic levels in the marine environment. These data by no means represent a complete summary of all available data, but they appear to be reasonably consistent as a body and to illustrate the complexity of biological concentration processes. It must be noted, however, that no data are now available to show the extent of seasonal variability of element composition in most marine groups. For several bivalve grazers, such variability is known to be appreciable (Hobden, 1967; Shimizu, 1967), as it is for many attached algae (Black and Mitchell, 1952); variation in some organic components of Calanus (Comita *et al.*, 1966) shows that it is likely that plankton species also exhibit seasonal variations of inorganic composition. This is clearly an important area in which more research would be of great value. In Figure 1 we have summarized the data of Table 2 for easier visualization.

A direct reduction of about an order of magnitude in concentration factor at each higher trophic level is indeed seen in tracer studies of cerium, ruthenium, and zirconium, as Baptist and Lewis (1969) reported for ^{65}Zn; Osterberg *et al.* (1964) reported even greater decreases from grazers to predators in respect to ^{95}Zr–^{95}Nb and ^{141}Ce, but this may have represented 1.5 to 2 trophic-level steps. Of the stable elements, however, only iodine exhibits a systematic decrease in concentration factor at each higher trophic level; the other stable trace elements exhibit great variation in behavior. An arbitrary, but we think useful, indicator of radionuclide behavior in ecosystems is the occurrence of concentration factors as high as 10^4 in the range exhibited by the groups tabulated. Applying this criterion, the grazers clearly show the greatest prevalence of high concentration factors: 10^4 appears in the range of six elements in the planktonic and of seven in the benthic grazers, whereas it appears for only three elements in the benthic and four in the planktonic algae, and for five, three, and three elements in the three groups of predators tabulated. As noted above, we do not know precisely how data on benthic predators might be compared to these values. We believe that examination in detail of the data summarized in Table 2 and in other summaries pertinent to this question shows that this is a real property of aquatic trophic networks and that of those elements that are ever *strongly* concentrated by organisms, the chances are that the highest concentration factors and the most frequent occurrence of high concentration will be among grazing organisms. Cs, Mo, Ce, or Ru, never strongly concentrated by organisms, are clear exceptions, but Sr, strongly concentrated only in the skeletons of Acantharia, falls within this generalization.

It hardly seems necessary to emphasize that in any particular situation, such a probabilistic statement may be expected not to hold. The number of exceptions, already noted, shows clearly that there is no substitute for the evaluation in detail of each exploited food web known to have been contaminated (as advocated in Chapter 10); however, since man is continually faced with the problems of making predictions in the absence of such evaluations, we feel that such a summary of the statistics available provides a useful guide.

THE POSSIBLE RADIOSENSITIVITY OF MARINE ECOSYSTEMS

In earlier discussions (National Academy of Sciences–National Research Council, 1957, p. 32), it was emphasized that planktonic or pelagic marine organisms are exposed to levels of background radiation lower than those experienced by any other populations except those in freshwater lakes living at depths below 100 m but not near the bottom. That report, however, considered exposure only to cosmic rays or to beta or gamma radiation from ^{40}K. More recently, Polikarpov (1966), Cherry (1964), Yermolayeva-Makovskaya *et al.* (1968), and Beasley (1969) have presented evidence that alpha-particle doses from ^{226}Ra or ^{210}Po contained in marine organisms may be very high and that the low background levels in the marine environments, cited by the National Academy of Sciences–National Research Council (1957) may be gross underestimates. That these high alpha exposures apply generally to marine organisms is not yet well established, while the low cosmic ray and ^{40}K backgrounds rest on much data.

We are also inclined to believe that the world ocean represents the oldest environment continuously available to living organisms. It must be possible, then, that in their long history of evolution, possibly under exceptionally low radiation levels, the organisms of the oceans have selected against those genes responsible for radiation resistance, and may consequently be consistently more radiation sensitive than are the better known species of the land. This is Polikarpov's view (1966). The very small amount of direct evidence bearing on this question is considered in Chapter 9 of this report. As ecologists especially conscious of the diversity of species of marine organisms and of the very small number that have thus far been maintained in culture (Mullin and Brooks, 1967; Conover, 1968), we must caution against overly broad generalization from currently available data concerning the radioresistance of marine organisms.*

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*In fact, Angelovic and Engel (1970) have recently summarized data showing that such common marine organisms as the grass shrimp (*Palaemonetes pugio*) and the mummichog (*Fundulus heteroclitus*)

Such data have derived only from the "toughest" species, since only these are maintainable for experiment. Although it is by no means clear that any correspondence exists between sensitivity to radiation and to laboratory manipulation, caution is indicated. Furthermore, many fundamental marine species—ecologically speaking—undergo periodic population minima in response, presumably, to a seasonally inimical environment. They consequently also undergo periodic "population explosions" as the environment becomes more favorable. We do not know of any studies of the radiosensitivity of marine organisms under an ecologically meaningful range of conditions, both of environment and of growth rate.

We must also consider whether the ecosystems of the open ocean may be themselves intrinsically more sensitive to increases of incident radiation.

Radiation effects may be expressed in two ways: (a) directly, by the irradiated organism, in a series grading from prompt lethality to reduced vigor, shortened life-span, and diminished reproductive rate; and (b) genetically, by the transmission to offspring of radiation-altered genes. Such altered genes are most commonly recessive and most commonly disadvantageous to their carriers. It has been shown for *Drosophila*, however (Wallace and King, 1951; Wallace, 1956, 1958; Buzzati-Traverso, 1960), that heterosis produced by accumulated radiation-produced mutations leads to significantly increased individual viability, even up to levels of accumulated mutations that result in a considerable frequency of genetic death. Rugh and Wolff (1958) have reported that radiation-induced mutations in mice may produce increased resistance to radiation, again by heterosis. Comparable studies of several groups of microorganisms were noted by Buzzati-Traverso (1960), who emphasized that complex populations living competitively under radiation fields have not shown selection in favor of radioresistance genes.

Styron (1969) recently studied two isolated populations of an aquatic isopod, one in an area of high natural gamma-ray background from extensive exposure of granite (Folsom and Harley, 1957); the population evolving under high radiation backgrounds ("several times the average for aquatic ecosystems") proved to have a 7.9 times greater tolerance to ionizing radiation, but the author did not claim this as evidence of selection for genes enhancing radiation resistance.

Our ignorance of the genetic diversity within marine species still prevents any useful attempt to consider the genetic effects of increased radiation on pelagic ecosystems, although consideration of the histories of radiation exposure of these ecosystems leads to the expectation that new sources of increase in the available gene pool should have beneficial effects for at least some species.

Recent studies by Wallace (1966) and by Hoenigsberg (1968) emphasize the importance in this connection of the effective size of the gene pool. Hoenigsberg has shown that, in *Drosophila* species under wild conditions, gene pools may extend only over physical distances of 100 m or so and be represented by comparably small numbers of organisms. This clearly has very important implications for the frequency of elimination, by homozygosis, of recessive lethal mutations. We have emphasized the mobility of all pelagic populations and of a large fraction of the near-shore populations as well. Studies by Scheltema (1966, 1967, 1968) have shown that the dispersal of planktonic larvae of many benthic species is an important source of recruitment of populations severely localized as adults and is also responsible for continual interchange of genetic material among separated local populations. The gene pools of marine species may, then, be expected to be characterized by large size, in terms of both physical extent and number of organisms, and consequently by a slow rate of elimination of lethal genes. Such a situation would appear to optimize the beneficial effects of radiation-increased heterosis.

There are two possible exceptions to this generalization: (a) those benthic organisms, both open-ocean and near-shore, that do not produce planktonic larvae and may be expected to move only over very small distances; and (b) those planktonic species that undergo great seasonal fluctuations in abundance, with a consequent great reduction in genetic variability at each population minimum. Although this class of plankton appears to include a very large number of the most important pelagic primary producers (Hulburt, 1966; Menzel and Ryther, 1960), we do not appear to have enough information to estimate the minimum population at any season or to judge to what degree physical mixing processes may be able to produce new genetic intermixing during the periods of rapid population increase. This would appear to be an important and fertile field for future study, both theoretical and observational; its importance for our understanding of the radioresistance of oceanic ecosystems is clearly large.

We know of no data concerning the radiosensitivity of marine ecosystems as compared to that of the individuals within them. Fontaine (1960) pointed out that it is to be expected that the first effect on an ecosystem of a damaging radiation response by the most radiosensitive species in that ecosystem would be disequilibrium in "its community, a disequilibrium that clearly may have repercussions on human life outside the field of radioactivity." We know of no research that appears to have been directly stimulated by this suggestion. In fact, as discussed in Chapters 9 and 10 of this volume, the usual criteria for "acceptable" levels of radionuclide release, or of environmental radionuclide concentra-

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have long-term LD_{50}'s for ^{60}Co irradiation of 215 and 300 rads, respectively—values quite like those exhibited by man. Examination of this question, they found, was further complicated, in the cases of *Fundulus* and of *Artemia salina*, by a strong inverse correlation between environmental salinity and radiation tolerance. See also Angelovic *et al.* (1969).

TABLE 2 Ranges of Element Concentration Factors[a] in Marine Organisms at Various Trophic Levels[b]

Element	Algae		Grazers		Predators		
	Sessile	Plankton (Phytoplankton and Sargassum)	Plankton (Copepods, Pteropods, Salps, Doliolid)	Shellfish	Plankton [Euphausiids, Planktonic Amphipods, Shrimp (Acanthephyra, Paleomonetes)]	Fish	Squid
	(18)	(1)	(1)	(2)	(1)		(1)
Ag	100–1,000	<100–220	<100	330–2×10^4	<45–900		900–3,000
	(12, 13)	(1)	(1)	(2)	(1)	(12)	(1)
Cd	11–20	<350–6,000	<80–10^5	10^5–2×10^6	<300–10^4	>10	2,800
	(14)	(4)		(14)		(14)	
Ce	100–3,300[c]	2,000–4,500[c]		40–300[c]		5–12[c]	
	(13, 14, 16)	(1, 13, 17)	(1)	(7)	(1)	(14)	(1)
Co	15–740	75–1,000	<110–10^4	24–260	<70–1,300	28–560	<200–5×10^4
				(14)			
				18[c]			
	(14)	(1)	(1)	(2)	(1)	(8)	(1)
Cr	100–500	<70–600	<15–10^4	6×10^4–3×10^5	<55–3,900	3–30	<70
	(14)	(14)	(3)	(14)		(14)	
Cs	16–50	16–22	6–15[c]	3–15[c]		6–10	
	(14)	(1)	(1)	(2)	(1)	(14)	(1)
Fe	10^3–5×10^3	750–7×10^4	440–6×10^4	7×10^4–3×10^5	3×10^3–3×10^4	400–3×10^3	10^3–3×10^3
	(14)			(14)		(14)	
I	160–7×10^3			40–70		10	
	(9, 16)	(1)	(1)	(2)	(1)	(9)	(1)
Mo	10–200	<3–17	2–175	30–90	<2–14	~200	<10
	(14)	(1, 14)	(1)	(2, 14)	(1)	(14)	(1)
Mn	20–2×10^4	300–7×10^3	21–4×10^3	3×10^3–6×10^4	270–1,600	95–10^5	10^3
	(13, 16)	(1)	(1)	(2)	(1)		(1)

Ni	50–10^3	25–300	2–10^3	4×10^3–10^4	17–90		30–80
Pb	(13) 8×10^3–2×10^4	(1, 17) $<10^3$–3×10^6	(1) 3×10^3–2×10^6	(2, 6) 39–5×10^3	(1, 15) 200–6×10^4	(10) 5–10^4	(1,15) 100–2×10^5
Ru	(14) 100–10^{3c}			(14) 1–16^c	(11) 10^c	(14) 10^c	
	(18) <200–1,200	(1) <200	(1) <10–6×10^3		(1) <160–2,400		(1) <400–2,100
Sr	(14) 0.1–90	(1) 0.9–54	(1) 1–85	(5) ~50^c	(1) 1.2–10	(14) 4–5	(1) 0.9–1.2
Ti	(18) 200–3×10^4	(1) 600–10^4	(1) 28–>3×10^4		(1) 110–2×10^4		(1) 300–3,000
Zn	(14, 16) 80–3,000	(1) 200–1,300	(1) 125–500	(2, 14) 1,400–10^5	(17) ~50	(14) 280–2×10^4	(1) 2,500
Zr	(14) 200–$3{,}000^c$	(1) <1,000–2×10^4	(1) 360–3×10^4	(14) 8–36^c	(1) <800–4×10^4	(14) 5^c	(1) 2×10^4

[a] Concentration in whole, fresh organism versus concentration in seawater (see Chapter 5).

[b] Literature references are shown in parentheses in upper left of box and listed below. No attempt has been made to achieve completeness; the ranges of concentration factors are for illustration, but are believed to be representative.

[c] Concentration from radionuclide tracer experiments.

(1) Bowen *et al.*, unpublished; some data from Nicholls *et al.*, 1959.
(2) Brooks and Rumsby, 1965.
(3) Bryan, 1963.
(4) Chipman, 1958.
(5) Cigna *et al.*, 1963.
(6) Costa and Molina, 1957.
(7) Fukai, 1968.
(8) Fukai and Broquet, 1965.
(9) Fukai and Meinke, 1962.
(10) Goldberg, 1962.
(11) Hiyama and Khan, 1964.
(12) Hiyama and Shimizu, 1964.
(13) Ishibashi *et al.*, 1964.
(14) Polikarpov, 1966.
(15) Tamotsu *et al.*, 1964.
(16) Young and Langille, 1958.
(17) Vinogradova and Kovalskiy, 1962.
(18) Black and Mitchell, 1952.

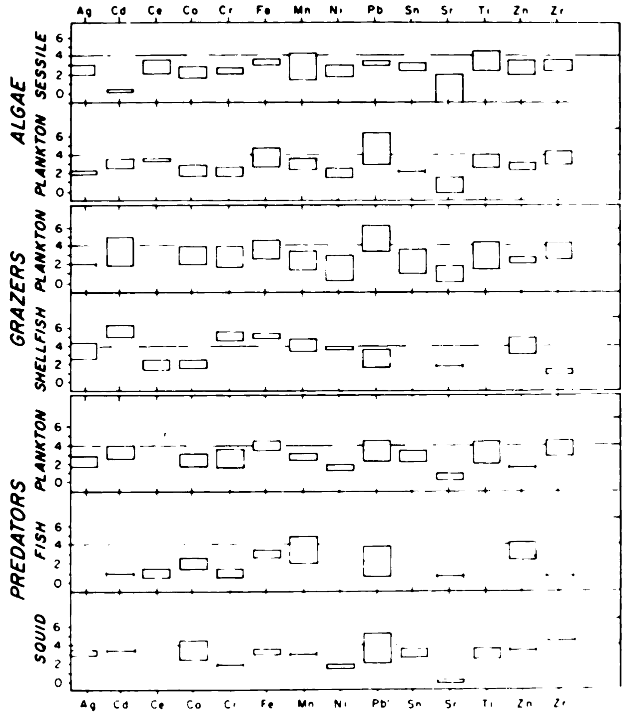

FIGURE 1 Ranges of element concentration factors in marine organisms at various trophic levels (data from Table 2). Log scale: 10^{-1} to 10^6; 10^4 marked by horizontal limit line.

tion, are clearly homocentric, oriented toward questions concerning direct radiation exposure of man or of his present food organisms. It is concluded in Chapter 10 that

> a continuation of the policies and practices concerning the control of low-level waste disposal . . . established during these formative years should assure that radioactive contamination of the marine environment will not reach unacceptable levels.

While it would be purely serendipitous if, in attempting to protect only man and the organisms he now exploits, the right decisions have been made with respect to protecting the ecology of the oceans as a whole, this appears in fact to have been the case. Man is among the most radiosensitive organisms, and his greatest vulnerability to radiation lies in his genetic apparatus. Because we cherish the life of every human being but are interested in preserving other organisms as populations rather than as individuals, our standards for the direct protection of man have also protected the pyramid of life in the sea, on which man ultimately depends. This conclusion is discussed further in Chapter 9.

We believe, however, that information is available upon which to build a qualitative picture of the radiosensitivity of marine ecosystems; we believe further that our attempt in the following pages to build such a picture is appropriate to the present discussion.

Klopfer (1959) suggested that four variables are principally involved in determining the number of taxa in a given area: the time elapsed since colonization; the area's geographic extent; its topographic variability; and its climatic variability. Of these, the first two would be expected, in the world ocean, to have favored development of a very large number of marine taxa. We believe that life originated in the oceans and that consequently the time for marine speciation has been the maximum available on earth; and, as we emphasized earlier, the world oceans represent the longest environmental continuum along which organisms can spread and differentiate. On the other hand, in ocean environments, the range of variability, either topographic or climatic, is probably small, compared to that in most large land areas. Contrary to Thorson's (1957) generalization, Hutchinson (1959) has noted that, in fact, marine environments (once account has been taken of the nonrepresentation therein of Insecta, Amphibia, and most orders of mammals) are characterized by greater species diversity than are those of land or fresh water. In spite of the enormous area represented by the oceans and in spite of the enormous time available for evolution within this area, such extensive diversity seems surprising in view of the very modest ranges of most of the environmental variables involved in niche determination. It appears that the diversity of marine species has been achieved by a long, slow evolution that has divided the environment into a series of niches probably definable by variations much more subtle than those that are critical in either land or freshwater determination.

This conclusion appears to have significant implications with respect to the mechanisms of marine population control. In a stimulating but controversial contribution, Hairston *et al.* (1960) concluded that

> (1) populations of producers, carnivores, and decomposers are limited by their respective resources in the classical density-dependent fashion, (2) interspecific competition must necessarily exist among the members of each of these three trophic levels, (3) herbivores are seldom food limited, appear most often to be predator-limited, and therefore are not likely to compete for common resources.

In a related analysis, less simply theoretical and strictly marine, Connell and Orias (1964) emphasized the role of available energy in encouraging species diversity. They also concluded that the "instability of the medium" must discourage oceanic creatures from speciation into niches characterized by very rigorous physical, or presumably chemical, specifications. As noted above, we believe that this argument does not apply to open-ocean plankton or benthon; we believe that even in the upper layers of water—those of maximum "instability"—the *position* in the environment of the various physically or chemically delimited niches must change, but that very few such niches would be seasonally or randomly obliterated.

Some other conclusions reached by Connell and Orias are especially relevant, however, in the present context:

> Although some niches are determined by physical variations in the environment, most of the dimensions of the niche are a result of interaction between organisms. For this reason, it is impossible to predict the number of niches (and therefore species) from environmental complexity alone. We also discard the idea that rigorousness *per se* limits diversity.

If these conclusions apply to the diversity of marine species, as we believe they do, the members of marine ecosystems might be expected to be more dependent on each other's productions and interactions than is true for members of land or freshwater ecosystems; the presence or products of other species may be the resources most commonly competed for. Equally, the need for specific products of a competitor species may be the factor preventing competition from resulting in complete displacement of one species by another. It seems likely to us that such considerations may be more important to planktonic than to benthic organisms. Kohn (1967) has shown that for *Conus*—a near-shore benthic gastropod genus—environmental complexity is the main source of species diversity.

Radioactivity introduced into systems like these in damaging amounts may be viewed either as a "stress," which appears analogous to environmental "rigorousness" as considered by Connell and Orias (1964), or as an analogue of predation; the latter seems to be a more useful line to consider. As noted above, Hairston *et al.* (1960) concluded that only herbivores, as an ecological class, can be regarded as usually predator-limited. Murdoch (1966) has strongly criticized this view, and certainly it is more usual, we believe, to consider the aquatic herbivores as "resource-limited," with respect to food. Should herbivores, in fact, already be predator-limited, the introduction of a new "predator"—radiation damage—would directly reduce this population, and presumably as directly increase the fraction of primary productivity available for "decomposers," short-circuiting the food chain. At the same time, reduction of the herbivore population would, according to the argument of Hairston *et al.*, reduce in parallel the resource-limited predator populations. Of those trophic levels readily exploitable by man, only the detritus-feeders might be expected to profit from such a change. Of course, as the intensity of radiation predation increased, an increasing number of species would become predation-limited, and radiation resistance would finally become the major criterion of interspecific competition.

Following Slobodkin (1968), one may inquire whether, in this context, radiation damage is a "good" predator. Slobodkin considers "goodness" of predator behavior only from the predator's point of view—and, incidentally, concludes that predators generally "act as if they were behaving prudently." We have tried, however, to develop from his argument an analogous "goodness" in terms of the biosphere: The yield of each species, in terms of conversion of its special set of raw materials to usable commodities, should be maximized, and the size of no species' population should be so reduced as to seriously diminish its ability to respond to disadvantageous fluctuations of the environment. From the viewpoint of the biosphere, the over-all "commodity" of the oceans might be taken as the maintenance of optimal O_2 and CO_2 levels in the atmosphere; this is clearly in the hands of the "primary producer" plants and of the carbonate skeleton formers. All other species, in this view, would exist simply to ensure a smooth flow of nutrients to and catabolites from the primary producers and the carbonate formers and, following the argument of Connell and Orias (1964), to contribute to the "interspecific" fitness of their environment.

From such a point of view, radiation damage is an exceptionally poor predator; it is not density-dependent and thus has no tendency to diminish its effect as a species approaches the level of "reduced resilience." It tends to affect most seriously the young, fast-growing stages of the population—those of lowest "population efficiency," in Slobodkin's terms—whose loss both affects the resilience of the population and results in the poorest use of energy. Furthermore, unlike any other form of predation, it should tend finally to emphasize selection only for a variable of no other selection value at all, radiation resistance. It may be further argued that radiation predation would be especially dangerous for the planktonic species of the open sea: Most of these species undergo cyclic reductions to very low abundances, at which the population may not be able to withstand further predation; these periods of low abundance are followed by periods of very rapid multiplication during which the radiosensitivity of individuals—by analogy with all experimental organisms so far studied—may be expected to increase in proportion to the rate of cell division.

It appears possible also to argue that the more closely interdependent are the species of an ecosystem—and we believe that those of marine environments are especially closely linked, and to a greater diversity of mutually interdependent species—the more especially radiosensitive will be the system. This argument critically depends on a wholly untested hypothesis—that the species interrelationships of marine ecosystems are specifically inflexible and that few (or no) participants in such an interrelationship can be replaced by others. If this is so, then the group, when exposed to damaging radiation, would be limited by the response of its most sensitive members. This hypothesis, we believe, is supported by the types of marine species interactions listed above (p. 205), as well as by the arguments of Connell and Orias (1964); it was also advanced by Fontaine (1960), on other grounds, some time ago. Support is also provided for this view by Imbrie's (personal communication) success in analyzing species abundance relationships in marine Foraminifera populations (from sediments and from plankton), using a statistical procedure that required that the relative abundances of the individual species must always be the same in order to appear to vary as a group. If the relationship "species A needs species B needs species C needs species D" is quite inflexible, it would not be surprising to find, over considerable ranges of environment and of population abundance, that it could be quantitated as "10 individuals of species A need 6 of species B need 3 of species C need 2 of species D," and the relative abundance ranking of the species in the group would be maintained as the absolute abundances varied. On the other hand, if the relationship were "species A needs either species B_1, B_2, or B_3," then we would not expect B_1, B_2, or B_3 to maintain a constant abundance relative to A as the absolute abundances vary. Techniques to explore whether this latter situation is at all common are presently being sought, but the fact that analysis based on the simpler situation has had success appears to confirm some cases of specifically inflexible interdependences.

It is clear that this sort of specifically inflexible interdependence would not act to increase stability of a community or ecosystem, contrary to the suggestions of MacArthur (1955) and Watt (1964) that increased stability would result from increase in the number of interspecies links. Paine (1969) has suggested that in quite different sorts of situations—in ecosystems of moderate complexity (the intertidal community of the North American Pacific coast) or even of great complexity (the Great Barrier Reef off Australia)—the removal of one specific carnivore (among, in both cases, many other large carnivores) has resulted in major, even drastic (in the latter case), changes in the species composition of the whole system. Clearly, Paine's (1969) suggestions relate closely to that of Fontaine (1960), noted above, of the dependence of a linked ecosystem on the radiosensitivity of its most sensitive member.

In conclusion, it appears that the patterns of cyclic abundance of many marine species are such as to make radiation damage especially serious when viewed as a form of predation; it appears further that the patterns of interspecific mutual dependence inferred for marine ecosystems may have specifically reduced the ability of individual species to profit from higher levels of radioresistance. It does not appear to us possible even to erect chains of argument that might relate this set of properties of marine ecosystems to those discussed earlier as probably making marine species more likely to show the stimulating effects of radiation-induced heterosis. The *directions* that should be taken by research in these areas are quite clear—as indeed they were when noted by Fontaine (1960) and Buzzati-Traverso (1960) at Monaco in 1959—but the actual experimental details are mostly either obscure or of frightening complexity. Our hope in this discussion has been to point out how important these problems are and to imply how unlikely it is that their practical solutions will be found in purely man-centered regulations. Again, however, let us reiterate that no support is intended for the view that present levels of ocean radioactivity, or any levels now contemplated, constitute significant ecological variables.

HUMAN ECOLOGY AND THE MARINE ENVIRONMENT

As man is an organism interacting with the oceans, his ecology is as suitable a subject for consideration here as that of any wholly marine species. Perhaps the most salient aspect of human ecology today, the "population explosion," clearly implies that there must be significant changes in man's relationships with the biosphere, as well as with his fellow man. Much recent discussion (Polikarpov, 1966, p. 233; President's Science Advisory Committee, 1966, Ch. 2; Butler and Holston, 1966; Iversen, 1967; Emery and Iselin, 1967; *Hydrospace*, 1968; President's Science Advisory Committee, 1967; Schaefer, 1968) has dealt with the idea that increasing human needs for food can and will be met partially by expanded and more efficient exploitation of the organisms of the oceans.

Such a change has clear and pertinent implications for our present consideration:

Increasing the efficiency of exploitation of pelagic biota necessarily implies—if anything like an order-of-magnitude increase of efficiency is to be sought—that lower and lower trophic levels must be exploited; such exploitation carries with it the prospect of changes in the radionuclide exposure of the exploiters.

Efficiency of exploitation can be greatly extended with safety only after significant growth of our understanding of the oceans as ecosystems. Marine radioactivity has been and is now a basic tool in extending this understanding, and the regulation of releases of radioactive materials to the marine environment should continue to be designed with this use in mind.

As ecosystems, the oceans are continua to an extent not exhibited by any other systems subjected to human exploitation save the atmosphere, and like the atmosphere, the world ocean in something like its present ecological-geochemical dynamism is essential to life on earth. Consideration of the physical and biological histories of many areas in which the application of intensive agriculture predated reasonable knowledge of soil conservation—the deserts of

Persia and North Africa or the semideserts of Spain and much of China, among others—shows how vital it is that exploitation of the marine biota be intensified only hand in hand with full understanding of the importance to man of the intact ecosystem and of those variables critical to its continued integrity.

These three problems are considered in greater detail in the following sections.

Use of Lower-Trophic-Level Organisms in the Oceans as Food for Man

As the earth's population increases, more attention will be directed toward the oceans as a source of human food. In terms of the efficiency of energy conversion and of deliberate attempts to cultivate and harvest the seas, the primary producers and the grazers become more and more appealing. Thus, grazers on plankton may become important foods for human consumption, supplementing the predaceous fish now largely in use, as well as the lower-trophic-level crustaceans and shellfish. This change of emphasis is a consideration of special force in aquatic environments, where food chains are typically of much greater length than are those on land.

Polikarpov (1966, Ch. 12) concluded that in a uniformly labeled aquatic environment, radionuclides are more generally accumulated directly from the water than indirectly by way of the food chain; he appears to have been strongly influenced in this view by studies of such radioelements as strontium and cesium. In studies of manganese metabolism by the lobster, Bryan and Ward (1965) have presented data indicating that most uptake must be from the animal's food; most of its zinc and copper, however, appears to be absorbed directly from the water (Bryan, 1964). Studies of this kind have been made of few other organisms, and it does not appear to us that the available data support any firm generalization about paths of radioelement uptake. As Polikarpov points out, however, in a nonuniformly labeled environment, where organisms—either prey or predators—migrate from higher to lower levels of radioisotope contamination, the food chain may be expected to be a major variable in an organism's accumulation of radioactivity. Data presented in Chapter 3 show that over very long periods of time after any radionuclide is introduced, and invariably for short-lived nuclides, nonuniform labeling of the marine environment is precisely what will exist.

As we have pointed out above, consideration of the energetics of ecosystems indicates that large increases in man's efficiency of exploitation of the marine biosphere for food can be made only by emphasizing increased use of grazers. If, as noted by Butler and Holston (1966), the emphasis is on the oceans as a source of protein food—and lack of protein is notoriously the great dietary deficiency—then the grazers in the sea, as on land, are the most promising level for exploitation. Or if, as noted by Emery and Iselin (1967), man is approaching the point of diminishing returns in "gathering" or "hunting" food in the sea and must face the prospect of sea-farming—or, in their terminology, "herding" of marine animals—there appears again more hope from exploitation of grazers. As Iversen (1967) pointed out, we know how to increase the food available to impounded crustaceans or bivalves, as well as to "those fishes that feed low on the food web," but we do not know how to provide, cheaply or efficiently, food for impounded carnivorous fishes. Clearly, in this sort of situation, the position of scavenger species that can exploit human wastes also deserves detailed consideration.

In general, all roads to conclusions about large increases in "food from the sea" lead to increased use of grazing organisms. And as the data of Table 2 suggest, this increase will be accompanied by increased intake of a large number of elements having important fission products or activation-produced radionuclides. If there is progressively increasing human use of marine grazing animals for food, accompanied by widespread marine disposal of radionuclide wastes, the problems of maintaining human exposure to radiation at low levels would be intensified. Use of the "specific-activity approach" to these problems (described in Chapter 10) depends on the assumption that man's present element concentration levels would be maintained even in the face of significant changes in dietary concentration levels. For very few elements do we have good data in support of this assumption; on the other hand, we know of several (As, Sb, Te, Pb, to name a few) that man is unable to regulate; some implications of this uncertainty were considered in the 1962 National Academy of Sciences–National Research Council publication, *Disposal of Low-Level Radioactive Wastes into Pacific Coastal Waters*. Furthermore, the history of regulatory activities is one of very slow response to increased hazards resulting from technological change. This is well illustrated by consideration of the histories of the use of lead in interior paints, of automobile design and traffic control, of the use of fossil fuels in urban environments, and so on. That the Windscale and other British releases of radioactive waste have been constantly, and thoughtfully, re-evaluated and controlled (Preston and Jefferies, 1969) is a great credit to the agencies responsible, but this is only a small ray of hope in the largely gloomy history of industrial pollution.

Whatever the approach used—"specific activity" or "critical pathway" (see Chapter 10)—there will exist a clear opportunity for neglect of those changes in regulations implied by alteration of man's dietary emphases. In the specific-activity approach, the probability that much of the change in exposure to man may be in increased gut-content levels is coupled with the explicit neglect of synergism between exposures to the gut and to the "critical organ" estimated from stable element distribution. In the critical-pathway approach, the likelihood that new critical pathways will appear for each radionuclide as human food emphasis shifts to lower trophic levels puts on the regulatory agency an onus of constant resurvey of each disposal situation. It appears clear to us that at any given level of radioactivity in the oceans, both the hazards to man and the problems of guarding populations against these hazards will necessarily be greater, and possibly a good deal greater, if he is eating grazing organisms than if he is eating predators. The statistics of published information on critical-pathway control situations are in agreement with this view.

Two other consequences of importance in this context would follow man's placing greater reliance on grazers or first-stage predators drawn from the pelagic environment for food: (a) the organisms eaten would, in general, be of smaller size and consequently of greater surface : volume ratio than most of those now exploited; and (b) even beyond this purely geometrical increase in surface : volume ratio, the organisms of the plankton have tended to evolve bodies having extra-large surfaces, whether to enhance gas exchange, to resist sinking, or as an adjunct to food gathering. Moreover, as food-organism size diminishes, man's tendency to consume the whole organism increases, for instance, sardines, fish meal, or even the often-advocated plankton paste. From foods of this sort, man would be efficiently exposed not only to the amount of radionuclide concentrated by the organism, as indicated by its analytical concentration factors, but also to the amounts held in its gut from recent meals and to that adsorbed on its external surfaces. Studies summarized by Lowman (1960) have shown that many radioactive nuclides appear at higher concentration ratios in gut contents or adsorbed on surfaces than they do in the metabolic cycles of plankton organisms.

Already, both dietary preference and economic factors have led large fractions of the world population to the consumption of whole marine organisms, including shell; the small dried shrimp used widely in the Orient and in Brazil and the shrimp-flour chips that have spread from Indonesia to very wide use in South America and elsewhere offer examples of this.

An amusing aspect of the variability of human ecology, in the sense of feeding habits, appears as a consequence of Ward's (1966) study of the uptake and distribution of plutonium in the lobster. Her conclusion is, implicitly, that because 90 percent of the ^{239}Pu taken up by *Homarus vulgaris* is deposited in the calcified shell, the lobster as a dietary vector of Pu can be taken as minimized. This is clearly reasonable for those who ingest the organisms in cocktails, salads, or otherwise from the shell; it is not so, however, for those gourmets addicted to lobster bisque—a soup thickened by a paste of pounded lobster shell. Since one can reasonably predict similar Pu concentration in the shells of other edible crustaceans, and since crab bisque, shrimp bisque, and crayfish bisque are of comparable elegance, one can envision a dichotomy of radionuclide hazard based on gastronomic sophistication.

Since we are here considering human ecology, it is appropriate to mention the very extensive literature—largely in Japanese—concerning the use of drastically "modified" marine organisms as food, in the forms of fish pastes, "cheeses," sausage, and the like. Much of this work appears to have been motivated by a desire to retrieve materials stored too long rather than by a striving for gastronomic elegance. However, the modification procedures point toward ways of producing edible protein of much lower radionuclide content, as well as toward lowering the content of toxic or unpalatable decomposition products, toward which this work was aimed. That such processing would result in increased costs appears self-evident, but that it could be done without unbearable economic results in case of persistent need for radionuclide decontamination is as likely as that it would—as it has in Japan—prove cheaper to reprocess fish than to store them at low temperatures. Man's technological dexterity and his ability to find its results acceptable are among his most salient adaptive traits.

Considerable interest attaches to the time scale of man's increasing demand for food. From Butler and Holston (1966), it appears that the United Nations is thinking in terms of supplying from the sea about 20 percent of the world demand for animal protein—estimated values vary from 14 g per capita per day (Butler and Holston, 1966) to 15-20 g per capita per day (Schaefer, 1968, and personal communication), a maximum requirement for 3×10^9 people of 22×10^9 kg of animal protein per year. In 1964, the world ocean yielded (Emery and Iselin, 1967) 28.6×10^6 tons of animals for human food—roughly equivalent to 5×10^9 kg of animal protein; this represented, then, almost 25 percent of the estimated maximum requirement for a world population of three billion, if properly distributed. As both Emery and Iselin and Butler and Holston imply, the ocean's yield of human food could have been approximately doubled simply by using the marine food now fed to domestic animals or used as fertilizer. Butler and Holston concluded that present annual harvests of marine animals could be increased fivefold to tenfold by fairly straightforward improvements of present harvesting, but that this represents a ceiling above which further increase in harvesting can be achieved only by an emphasis on trophic levels lower than those now exploited. Schaefer (1968), however, pointed out that as much as 40 percent of the 1964-1968 world fishery harvest was represented by "herring-like fishes" feeding at the first or second trophic level above the planktonic plants. Although this did not offer encouragement that large gains in fishery yield would come from transfers of effort to still lower trophic levels, Schaefer's calculations of "probable potential yield" of the world ocean (based on estimates of total net primary productivity of the oceans, and 10 to 20 percent per trophic-level transfer coefficient) indicate that the marine fishery should be capable of supplying (without change in trophic-level emphasis) one to six times the total protein requirement of a world population of six billion people—double that of the present. Schaefer further estimated that, based on technical considerations, about a fivefold increase of the world fishery harvest could be achieved "with no radical developments, such as fish farming." It may then be concluded that while there are basic disagreements about the reasons, there is some agreement about the conclusion that by the time a fivefold to tenfold increase has been achieved in the protein harvest of the oceans, either trophic-level considerations or considerations of harvesting technique will cause greater emphasis to be placed on the use as food of lower-trophic-level fishes and invertebrates (Butler and Holston, 1966) or on those marine animals susceptible to economic "sea farming" [lower-trophic-level fishes and invertebrates again, as Iversen (1967) concluded]. This appears from both Butler and Holston and Schaefer to be projectable to the last decades of the present millenium—1980 to 2000. It is, then, not too soon to engage in speculation on the consequences of massive (or of more massive) exploitation of marine grazers.

Man's present low level of dependence on marine sources of food, combined with his present practice of using principally the higher trophic levels in his diet, has given added assurance that the permissible concentrations of radionuclides in ocean water (as given by the National Academy of Sciences–National Research Council, 1962, for example) are conservative. As human populations grow, with rapidly increasing demands for animal protein, it will become desirable to significantly increase our exploitation of marine biota. Consideration either of the energetics of food webs or of the technology of animal "aquiculture" has led us to conclude that sizable increases in marine food supply must be made by utilizing grazing animals, as has been true on land for the same reasons. Analytical data show, however, that marine grazers tend to have larger concentrations of radionuclides than do the marine predators now largely exploited. Thus, the future seems to indicate that man's increasing need for protein will tend to reduce his emphasis on one or two links in the food chain that may have provided an extra degree of insulation from the radioactivity in the sea. As the National Academy of Sciences–National Research Council (1962) noted, much of the basic data concerning the elemental composition of marine food organisms and man's assimilation and retention of elements from such food was lacking. This is still true.

We hasten to add that either the specific-activity approach or the critical-pathway approach, when properly applied and continually adjusted as the environmental situation changes, will ensure that the concentrations of radionuclides in our diet, regardless of what seafood we eat, will be below permissible levels. Extensive long-term studies of food organisms from the Irish Sea near Windscale and from the northeast Pacific Ocean off the Columbia River indicate that the health and safety of human beings has been a primary consideration in release limits of radionuclides.

Marine Radioactivity as a Basic Research Tool in Ecology

The tracer experiment provided to scientists by the two periods of intense atmospheric testing of nuclear explosives offered a unique opportunity to explore details, especially of rates of transfer, of marine geochemistry and hydrography. These tracers are discussed both in Chapter 3 and in sections concerned with circulatory processes in Chapter 4. However, even after fifteen years we still have not learned how this tracer experiment may best be exploited. It must be emphasized that the "fallout tracer experiment" itself seriously affected our ability to profit from other tracer experiments. Tritium, for example, is a "natural" radionuclide in addition to being a major product of thermonuclear devices. It is continuously being produced, at low but measurable levels, by cosmic ray processes in the atmosphere. The existence of this natural tritium as a geochemical and meteorological tracer was predicted by Libby in 1946; publication of data confirming this promise began in 1951 (Grosse *et al.*, 1951). In 1952, the United States carried out its first thermonuclear explosion, and by 1954, the Castle series of weapons tests had introduced artificial tritium to levels that overwhelmed the natural levels. By contrast, the first subsurface seawater samples were collected for tritium analysis in September 1954 (Begemann and Libby, 1957); the only pre-Castle surface-water samples were collected in late 1952, and these were very few in number. Not merely has our ability to use the data resulting from the introductions of thermonuclear tritium been diminished by our ignorance of pre-Castle tritium concentrations, but our opportunity to observe the profiles of equilibrium concentration levels of natural tritium was destroyed before it could be taken. Extremely valuable oceanographic information could have been obtained through estimation of the speed at which the bomb-introduced tritium approached the equilibrium profile of natural concentrations. The same test series also disrupted our use of natural carbon-14 as a long-lived tracer for study of many of the same processes as those in which tritium is involved.

In fairness, however, it should be stated that the development of sensitive counting equipment essential to tracer studies was greatly accelerated by the nuclear testing program.

The oceans must be studied as a single intact ecosystem. Above (p. 200), we noted one type of bar to their easy exploration, but there are many others. Large-scale tracer experiments may be the only way by which some parameters can be evaluated well enough to support the kind of model-system studies that we believe are required to evaluate plans for new ways to exploit the marine environment. Our use of such large-scale tracer experiments to achieve *understanding* of the marine ecosystem could be hampered if radioactivity concentrations in seawater are very much lower than the levels that would affect our present ability to exploit the system. Plans for any massive new introductions of radioactivity into the oceans must be evaluated in the light of, among other things, the possibility of premature interruption of the tracer experiments now in progress.

The Significance to Man of the Oceans as a Functioning Ecosystem

It has been common for man to assume that any space is relatively large compared to his own dwelling—and this seems to apply as well to air-shafts in slum apartments as to streams, rivers, lakes, marshes, the atmosphere, and the oceans—and can be used as a waste depository. The hazards of this approach have become clear in the case of streams, rivers, and lakes, and locally in the case of the atmosphere; it has even become clear that each ecosystem may have a characteristic half-time for recovery from such abuse and that this interval is longer as the system in question is larger—i.e., longer for a river than for a stream, or for a lake than for a pond—and longer as it is more sluggish—i.e., longer for a pond than for a stream or for a lake than a river. All our knowledge of the oceans promises that, once seriously insulted in an ecological sense, their recovery would be slow indeed. It is most fortunate that the philosophy of nuclear waste disposal has so far been enlightened in this respect. It is important to emphasize, among the many reasons for continued adherence to today's enlightened philosophy, the fact that the oceans represent the fraction of the earth's surface exhibiting a positive O_2 balance versus the atmosphere and controlling the CO_2 balance. Man must maintain the world ocean as a functioning ecosystem for this reason, even if there were no others.

SUMMARY

Lower trophic levels of the seas are likely to have greater concentrations of radioactivity than higher trophic levels. If the population explosion forces man to use these lower trophic levels as food sources, then the capacity of the seas to safely accept waste radioactivity will decrease.

If, as suggested, marine species are both more closely linked than those of other ecosystems, and if unique—or very limited—species interdependences are here more common, then the radiation resistance of marine ecosystems will often be strongly limited by that of the least resistant species. No marine radioactivity levels have yet approached possible danger points, but this is argued as one of several aspects of the oceans needing fuller consideration in these contexts.

In order to assess properly the consequences to man of radioactivity in the marine environment, continued research is needed into the physics, chemistry, and biology of the oceans and into the interdisciplinary "hyphenated" offspring of these basic sciences. Of particular importance is increased research into the radiosensitivity of marine ecosystems. It is hoped that with the aid of modern high-speed digital computers we will attain new and more profound understandings of how the oceans will respond to the presence of this new contaminant—man-made radioactivity.

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Chapter Nine

RADIATION EFFECTS

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Under the present expansion in the development of nuclear energy, it is inevitable that the marine environment will continue to receive an increasing burden of radiation. We must therefore be concerned with the amounts and kinds of radiation the individual organism, the population, and the ecosystem can tolerate without significantly changing the "balance of nature." This balance is not static; it responds to a multiplicity of factors, both natural and man-made. Radiation is but one of these factors. However, irradiation is not a recent introduction to the marine environment, since low levels of radiation from environmental and cosmic sources have been present to varying degrees throughout geological time. Our knowledge of the responses of ecosystems to small changes in their components is extremely limited; and, furthermore, the interactions of elements of the natural environment have yet to be explored.

All types of ionizing radiations produce changes in living cells, and some changes, at least, are regarded as deleterious. Until we learn otherwise, it is assumed that any radiation exposure in excess of the "natural" level might produce deleterious changes. The biological consequences of low-level irradiation have grown, therefore, into a subject of importance in all environments. Our knowledge of this subject with respect to the marine environment is still extremely limited and is mainly concerned with somatic effects.

The potential hazards to man of radiation exposure have been and are being studied very extensively. In general, the conclusion from these studies is that any significant increase in radiation levels is detrimental from the genetic point of view. However, for man, this acceptance rests on ethical considerations with respect to the individual and on the fact that genetic anomalies are not reparable in an individual. When considering the marine environment, however, we are not primarily concerned with individual organisms, but with populations, and at the population level genetic damage is reparable by natural selection.

This conclusion is summed up clearly in a paper by Purdom (1966).

> It would seem likely that the genetic response of populations is relatively unimportant and that general mortality and infertility would be the limiting factors in the extent to which populations may overcome radiation exposure. This certainly seems true for animals which have been studied extensively—*Drosophila*, the mouse, and domesticated farm animals. Provided that marine organisms are not more sensitive genetically than these other organisms, genetic damage will probably have negligible effects, even under the maximum radiation exposures that seem possible from present day practice.

In Chapter 2 are described those sources that release radionuclides into the environment. In Chapters 3 through 8 the physical, chemical, and biological factors that determine the distribution of the radionuclides are discussed. Some radionuclides remain in solution or suspension in the seawater, and some find their way into sediments, both sources contributing to the dose to organisms. Other nuclides will be

TABLE 1 Radiation Dose Rates in mrad/Week in Various Tissues of Cod (*Gadus callarias*) and Haddock (*Gadus aeglefinus*), from the Decay of ^{40}K and ^{226}Ra[a]

Radionuclide		Cod				Haddock			
		Muscle	Skin	Bone	Fins	Muscle	Skin	Bone	Fins
^{40}K	β	0.43	0.25	0.28	0.16	0.36	0.25	0.32	0.16
	γ	0.03	0.01	0.01	0.01	0.03	0.01	0.02	0.01
^{226}Ra		–	0.020	–	–	–	0.040	–	–

[a] After Fedorov (1965).

TABLE 2 Estimates of Natural-Radiation Dose Rates to Plaice (*Pleuronectes platessa*) in the Irish Sea[a]

Source	Type of Radiation	Dose in mR/week
Natural activity in seawater (^{40}K only)	γ	0.07
	β	0.18
Natural activity in seabed (estimate)	$\beta\gamma$	1.00
Cosmic radiation at 20 m		0.10
^{40}K in fish	γ	0.01
	β	0.22
Total		1.58 mR/week or 0.082 R/year

[a] Data from Woodhead (personal communication).

absorbed by organisms or adsorbed on their surfaces; these nuclides will contribute to the internally delivered dose to a greater or lesser extent, depending on the size and complexity of the organism and the energy of the emission. In addition, any of the parameters discussed in Chapter 7 or any group of them influences the exposure of organisms to radiation.

Laboratory studies of radiation effects on individuals have been made with a variety of techniques. The basic studies of sensitivity to acute doses of radiation have mainly employed x rays, while studies attempting to reflect the environmental conditions have used ^{60}Co and ^{137}Cs sources or radionuclides in solution. Environmental and population studies have been more limited, however.

Our intention in this chapter is to consider only that research pertinent to the prediction of what might occur in the marine environment; therefore, we have not considered all of the studies that have been carried out using marine organisms in radiobiological research.

NATURAL RADIOACTIVITY

The sources contributing to the natural background radiation dose of organisms in the marine environment are cosmic rays and the natural radioactivity in the earth's crust, present in seawater sediments and biota (Folsom and Harley, 1957). More than 60 radionuclides have been identified within the marine ecosystem (Chapter 2), and, based on these measurements, some calculations have been made of the dose rates to which the biota is exposed.

Cherry (1964), in studies with phytoplankton from the open sea, shows that dose rates from total alpha activity in organisms could range from 230 mR/yr to 2.8 R/yr. Fedorov (1965) has calculated the tissue dose from ^{40}K and ^{226}Ra in cod, *Gadus callarias*, and in haddock, *Gadus aeglefinus* (Table 1). Estimates have been made (Ministry of Agriculture, Fisheries and Food, 1967) of the beta and gamma dose rate from the environment to the predominantly bottom-living plaice, *Pleuronectes platessa*, in the Irish Sea (Table 2). The major source of radiation as far as this species of fish is concerned is the seabed.

Recent measurements of ^{210}Po in marine organisms suggest that radiation from natural alpha emitters contributes significantly to the radiation dose to organisms (Beasley, 1968).

MORTALITY INDUCED BY ACUTE RADIATION EXPOSURE

Lethal amounts of acute radiation differ widely among organisms because of biological variations related to such factors as species, age, physiological state, and body size. In the aquatic environment, these variations are further complicated by the interaction of environmental factors such as temperature, dissolved oxygen, chemical composition, and salinity. Nevertheless, exclusive of the eggs and larvae of invertebrates and fish, most of the freshwater and marine organisms for which data exist are relatively radioresistant.

Marine species differ little in radiation tolerances from freshwater species.

Values of LD_{50} (lethal dose resulting in 50 percent mortality) for acute irradiation of aquatic organisms have been listed by Donaldson and Foster (1957) and by Polikarpov (1966). In general, there is a relationship between radioresistance and the phylogeny and ontogeny of the organism. Primitive forms are more resistant than the complex vertebrates, and older organisms are more resistant than the young. Bacteria and algae may tolerate doses of thousands of roentgens, but freshwater fish, the most sensitive group listed by Donaldson and Foster (1957), were affected by considerably lower doses. The LD_{50} for adult rainbow trout, *Salmo gairdneri*, ranged from 300 to 3,000 R; and for the most sensitive stage of a developing trout egg, the LD_{50} value was as low as 16 R.

Despite research into lethal effects of radiation for over 50 years, surprisingly few LD_{50} values have been determined for marine organisms. Polikarpov (1966) lists LD_{50} values for 50 species of aquatic organisms, but the majority of these are freshwater organisms. White and Angelovic (1967) provided some LD_{50} values for several marine species irradiated with a ^{60}Co source. For six species of marine adult fish, the $LD_{50/30}$ (lethal dose for 50 percent mortality in 30 days) ranged from 1,050 R to 5,550 R, similar to values for freshwater fishes.

White and Angelovic (1965, 1966) point to the need to describe radiation tolerances in terms of time curves for mean lethal dose rather than in terms of the usual $LD_{50/30}$. Figures 1 and 2 show the time curves for mean lethal dose for 14 marine fishes and invertebrates. Figure 2 shows, for example, that for the first 25 days, the oyster *Crassostrea virginica* is much more resistant to radiation than the clam *Mercenaria mercenaria*; after 80 days, however, the LD_{50} for the clam was greater than for the oyster.

LD_{50} curves are needed, especially for the dominant economically important marine species in different ecosystems. However, because the levels of radiation required to kill marine organisms are so high, actual kills by radiation in the environment are extremely unlikely. These experimental LD_{50} curves will, however, keep in proper perspective our view of the likelihood of mortality in marine organisms from acute radiation.

CHRONIC EXPOSURE

External Radiation

Donaldson and Bonham (1964, 1966) have taken advantage of the migratory habit and the fecundity of chinook salmon, *Oncorhynchus tshawystcha*, to make a continuing long-term study of the effect on a population of chronic low-level irradiation from a ^{60}Co source during embryonic development. Eggs were first irradiated at 0.5 R/day from shortly

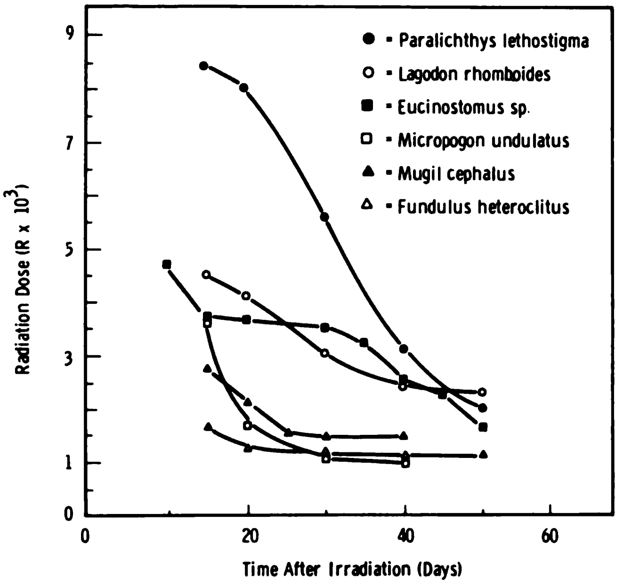

FIGURE 1 Mean lethal dose time curves for several species of marine vertebrates showing the dose-time combination at which 50 percent of the experimental animals died. (Reprinted with permission from White and Angelovic, 1966.)

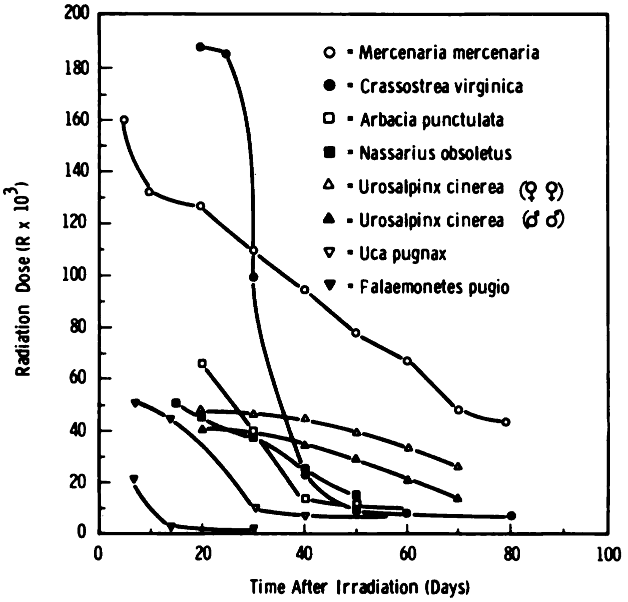

FIGURE 2 Mean lethal dose time curves for several species of marine invertebrates showing the dose-time combinations at which 50 percent of the experimental animals died. (Reprinted with permission from White and Angelovic, 1966.)

after fertilization until feeding commenced. The total dose was 33–40 R. The fingerlings were reared and then allowed to migrate to sea. Those that returned to the hatchery during the second year were precocious males; during the third and fourth years following irradiation, both male and female adults returned. Various crosses were made, some of which were reirradiated at 1.3 R/day to a total dose of 95 R. The initial dose chosen was about 40 times the calculated maximum dose that the germ cells and young salmon could receive in the Columbia River before migration to the sea. The radiation level used in the experiment was appreciable, amounting to 10^3 times the normal background, or 0.02 mR/hr.

These series of long-term experiments involving large numbers of organisms (96,000 to 256,000 fingerlings were released per experiment) indicate that irradiation at 0.5 R/day from the fertilization stage to the feeding stage produced no damage to the stock sufficient to reduce the reproductive capability over a period of slightly more than one generation. Although abnormalities in young fish were increased by irradiation, the number of adults returning was not affected. On the contrary, the irradiated stock returned in greater numbers and produced a greater total of viable eggs than the control stock.

Brown and Templeton (1964) and Templeton (1966) report on a series of experiments in which the eggs of plaice, *P. platessa*, were irradiated with a ^{137}Cs source. Total doses ranging from 0.6 to 500 R were used, at rates of 10 mR/hr to 1 R/hr from fertilization until hatching. No significant differences at hatching were observed in the survival or in the number of abnormal larvae produced.

Engel (1967) reports on studies of the effects of chronic low-level irradiation on the growth and survival of young blue crabs (*Callinectes spidus*). Single acute exposures had indicated that the sensitivity of these crabs is similar to that observed for other marine invertebrates. In the chronic irradiation experiment, the crabs were irradiated at an average of 22.5 hr/day at rates of 3.2, 7.3, or 29.0 rads/hr. The total radiation doses received over 70 days were 5,105, 11,502 and 45,693 rads. A significant number of deaths due to radiation occurred only among the crabs that received the highest radiation dose. The death rate of the crabs that received 3.2 and 7.3 rads/hr was similar to that of the controls. All irradiated and control crabs molted at least once. The numbers of second and third molts were affected by the radiation dose. The crabs that were irradiated at 29.0 rads/hr molted least, and none had three successful molts. Crabs that received 3.2 and 7.3 rads/hr underwent more second and third molts than did the controls, although this difference was not significant.

Radionuclides in the medium do not constitute the only radiation source to which pelagic organisms are exposed in the environment. A high degree of sorption of radionuclides into or onto the egg, for example, could give rise to a radiation dose within, or in the immediate vicinity of, the developing embryo that would be greater than that arising from the medium alone (Polikarpov and Ivanov, 1961, 1962; V. M. Brown, 1962; Brown and Templeton, 1964; Polikarpov, 1966). In addition, radiation from radionuclides incorporated into the developing tissues of an embryo may also be more effective in causing damage than external radiation alone (Polikarpov, 1966).

Hibiya and Yagi (1956) and Mikami *et al.* (1956), using fallout ash and rainwater residues from weapons tests, reported on the effects of these materials on the development of fish eggs. Concentrations in excess of 2×10^{-9} Ci/liter were found to be lethal, and abnormalities and delay in hatching were observed in concentrations down to 4×10^{-10} Ci/liter. The interpretation of these data must remain doubtful, however, since the radionuclide concentrations and chemical compositions of the ash and residues were not determined.

Polikarpov and Ivanov (1961, 1962), Ivanov (1965), and Polikarpov (1966) reported on the effect of ^{90}Sr-^{90}Y at low concentrations in seawater on the development of eggs of Black Sea fishes. Polikarpov and his co-workers, who pioneered the studies in this field, have reported on extensive studies with eggs of a large number of marine and freshwater species over the concentration range 10^{-14} to 10^{-4} Ci/liter of ^{90}Sr-^{90}Y. Reduced hatching of the larvae and early mortality were seen at concentrations of 10^{-7} Ci/liter and above, and the number of abnormalities increased significantly and with remarkable consistency at concentrations of 10^{-10} Ci/liter and above (Figure 3).

V. M. Brown (1962), Brown and Templeton (1964), and Templeton (1966) conducted similar experiments using eggs of the brown trout (*Salmo trutta*) and of plaice (*Pleuronectes platessa*), maintained from immediately after fertilization until hatching in water contaminated with ^{90}Sr-^{90}Y over the concentration range 10^{-10} Ci/liter to 10^{-4} Ci/liter. They did not observe any significant increase in mortality or in the production of abnormal larvae. Fedorov *et al.* (1964) reported that the eggs of plaice from the Barents Sea were sensitive to low concentrations of ^{90}Sr-^{90}Y in seawater. White and Angelovic (1966) report on the effects of chronic exposure to low levels of ^{137}Cs on the developing eggs and larvae of mummichogs (*Fundulus heteroclitus*). The concentrations of ^{137}Cs used, 3×10^{-7}, 3×10^{-6}, and 3×10^{-5} Ci/liter, produced no visible abnormalities. On the fourth day after fertilization, however, a general retardation in the rate of development was evident for those subjected to the highest concentration levels. On the nineteenth day after fertilization (approximately 50 percent hatch), the groups in the highest and the lowest concentrations had a slight reduction in the number of fish hatching, but this difference was no longer observable by the time hatching was completed.

Neustroev and Podymakhin (1966a), in similar studies with the eggs of the Atlantic salmon, *Salmo salar*, found

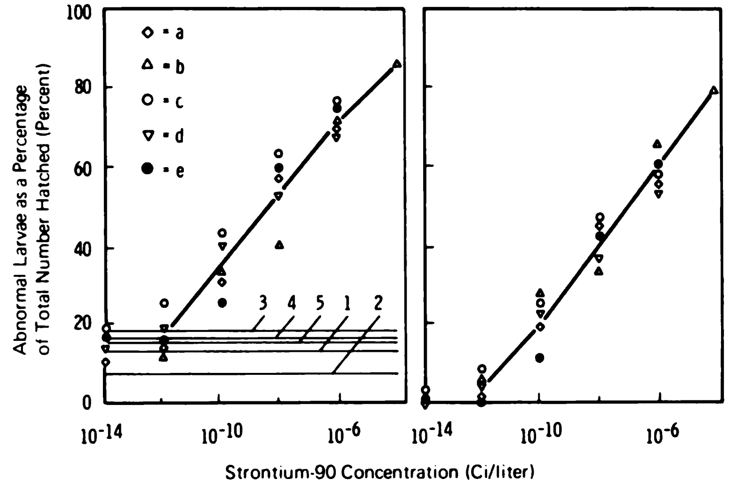

FIGURE 3 Dependence of proportion of abnormal larvae of Black Sea Fishes on strontium-90 concentration in surrounding seawater. (1) and (a) = abnormal mullet larvae; (2) and (b) = green wrasse (Labridae); (3) and (c) = horsemackerel; (4) and (d) = anchovies; (5) and (e) = from a mixture of pelagic eggs. (The figures denote the proportion of abnormalities in controls of the various species of fishes.) Left-hand graph without allowance for control, right-hand graph with allowance for control. (Reprinted with permission from Polikarpov, 1966.)

that at 10^{-10} Ci/liter ^{90}Sr-^{90}Y, the rate of development of the egg was the same as the control up to the stage of one-third development of the yolk sac. Subsequently, the rate of development in the contaminated aquaria was more rapid than in the control. However, the mortality and number of deformities did not differ from those of the controls. At 10^{-8} and 10^{-6} Ci/liter, the rate of development was the same as that observed in the 10^{-10} Ci/liter experiment. Mortality and deformities were increased only at the higher level of radioactivity.

Studies by Kulikov *et al.* (1966) of the effects of ^{90}Sr-^{90}Y on the development of the eggs of the freshwater mollusc *Limnaea stagnalis* L. show that morphological abnormalities, delay in development, and mortality were significant only at concentrations greater than 10^{-4} Ci/liter.

Nelson (1968) studied the effects of radionuclides on Pacific oyster larvae, *Crassostrea gigas*. The larvae were reared for 48 hr following spawning in seawater containing either ^{65}Zn, ^{51}Cr, or ^{90}Sr-^{90}Y. The concentrations of each radionuclide range from 10^{-2} Ci/liter to 10^{-8} Ci/liter. Significant increases in abnormal larvae were detectable at the following minimal concentrations: ^{65}Zn (carrier free), 10^{-4} Ci/liter; ^{51}Cr, 10^{-4} Ci/liter; ^{90}Sr-^{90}Y, 10^{-3} Ci/liter.

The effects of tritiated seawater on the germination of the asexually produced spores of *Padina japonica* have been studied. A reduced percentage of germination was observed in those subjected to a concentration of tritium of 3×10^{-2} Ci/liter. This concentration of tritium also had a marked effect on the subsequent growth of the embryos, causing a decrease in the number and length of the rhizoids as well as in the number of cells in the multicellular filamentous thallus (Buggeln and Held, 1968).

All of these studies have assessed radiation effects in terms of observable gross effects. Ivanov (1967) has considered more sensitive parameters and reports on the effects of ^{90}Sr-^{90}Y in seawater on the mitotic activity and production of chromosome aberrations on the dividing cells of eggs of the Black Sea scorpionfish *Scorpaena porans*. As the concentration increased from 10^{-10} Ci/liter to 10^{-5} Ci/liter, the mitotic activity of the cells decreased. At the same time, the percentage of chromosome aberrations increased, with statistical significance when the concentration exceeded 10^{-9} Ci/liter. The types of aberrations were varied, with chromosomal and chromatid bridges and fragments observed most frequently. At the highest concentrations, abnormal mitoses were observed.

Experimental data concerning the radiation effects on developing embryos maintained under laboratory conditions in contaminated media are conflicting. In all these studies, the embryos were maintained under highly artificial conditions in the laboratory, including artificial fertilization. The developmental temperatures varied with species and experiments, ranging from 6°C to 24°C. In some experiments, antibiotics were used to reduce bacterial contamination. In addition, experimental incubation containers varied in capacity from 60-ml petri dishes to 40-liter plastic containers, with experimental lots ranging from 30 to thousands of individuals. The successful maintenance of marine eggs and embryos under controlled environmental conditions is yet to be developed for many marine organisms. In addition, more data are required on the effects of other environmental parameters before significant radiation effects can be demonstrated.

Of particular significance in the work from the Soviet Union is the unique concentration effect response (Polikarpov, 1966). An increase in concentration of ^{90}Sr-^{90}Y of six orders of magnitude (2×10^{-10} to 2×10^{-4} Ci/liter) no more than triples the abnormality production rate (Figure 3), and increases mortality only fivefold. If the radiation dose received by the developing eggs is proportional to the concentration and all other factors are equal, one would expect a more marked dose-effect response. The results are inconsistent with the linear hypothesis of dose response as well as with data from many radiobiological and

TABLE 3 Mortality, Growth, and Radionuclide Concentration in Chinook Salmon, *Oncorhynchus tshawytscha*, Reared under Various Reactor Effluent Conditions, December 1965-April 1966[a]

Treatment groups[b] (% effluent)	% Mortality (5 mo)	Mean Weight (g)	Concentration (pCi/g wet weight)		
			^{24}Na	^{51}Cr	^{65}Zn
0	18	0.70	77	19	6.8
2	10	0.82	750	38	20
4	13	1.05	1,390	53	36
6	13	1.20	2,210	65	45

[a] After Olson (1967).
[b] At least 1,000 fish in each group.

toxicological investigations. Although our knowledge of the dose-effect response at low levels of irradiation is limited, present knowledge suggests that perhaps factors other than radiation need to be considered and evaluated.

The biological effects of the effluent from production reactors at the Hanford plant have been monitored for more than 20 years by rearing salmonids in diluted effluent. The three main factors in Hanford reactor effluent considered to have a potential effect are the thermal increment, radioactivity, and chemical toxicity, the last from the hexavalent chromium used as a corrosion inhibitor (Olson and Nakatani, 1965; Nakatani and Foster, 1966; Olson, 1967).

Freshly fertilized eggs of salmonids have been incubated, and the fish reared in various concentrations of effluent until they reached migrant-sized fingerlings. The mortality, growth rate, and radionuclide concentration in chinook salmon reared under various effluent concentrations for 5 months during the 1965-1966 season are shown in Table 3. No significant lethality occurred in an effluent concentration of 6 percent, far above the existing levels in the river. The concentrations in the fish of the three gamma emitters, ^{24}Na, ^{51}Cr, and ^{65}Zn, are approximately proportional to the effluent concentration. The body burdens at these levels have produced no demonstrable damage in chinook salmon.

Internal Emitters

The effects on rainbow trout, *Salmo gairdneri*, of ingestion of some biologically important radionuclides have been studied. Yearling trout were force-fed radionuclides in capsule form over extended periods to determine what quantity of radionuclides must be ingested to produce radiation syndrome and damage. All of these experiments used very high levels of radionuclides, which would not normally be experienced in a contaminated aquatic environment, as the controlled disposal of radionuclides is limited to lower levels by man's use of the environment. However, the data from these ingestion experiments help to keep observations of the body burdens of radioactivity in field-contaminated fish in proper perspective with respect to potential radiation damage in fish in general.

In a series of feeding experiments, either ^{32}P, ^{90}Sr-^{90}Y, or ^{65}Zn was fed daily to rainbow trout at levels from 0.005 μCi/g/fish to high levels of about 10 μCi/g/fish (Watson *et al.*, 1959; Nakatani and Foster, 1963; Nakatani, 1966). Table 4 summarizes the results. The hematopoietic tissues of trout were found to be the most radiosensitive, as in mammals. Leukopenia was an early indicator of radiation damage following the ingestion of beta-emitting ^{32}P or ^{90}Sr-^{90}Y and gamma-emitting ^{65}Zn. Depression of growth also indicated radiation damage for the rapidly growing yearling trout. The radiation syndrome, including leukopenia, anorexia, loss of scales, lethargy, and growth depression, was very pronounced for fish fed ^{32}P at the higher levels and to a lesser extent for fish fed ^{90}Sr-^{90}Y. Because much of the energy from the gamma-emitting ^{65}Zn is not absorbed, trout were able to ingest without observable effect much higher levels of ^{65}Zn than of ^{32}P or ^{90}Sr-^{90}Y on a μCi/g/fish basis. In addition to the differences in the radiation characteristics of the isotopes, the rates at which the fish metabolize and the deposition sites for each radionuclide also differ, although all three are "bone-seekers."

Columbia River fish captured in the Hanford environs contain small amounts of virtually all of the radionuclides present in the water, but the only nuclides that accumulate in the fish flesh in significant amounts are ^{32}P and ^{65}Zn (Foster and Soldat, 1966). In 1965, the average concentration in muscle tissue of whitefish, *Prosopium williamsoni*, was 200 pCi/g and 27 pCi/g for ^{32}P and ^{65}Zn, respectively. Substantial seasonal variations may occur, with a tenfold difference for ^{32}P and a fourfold difference for ^{65}Zn. In terms of radiation damage to fish, however, the concentrations are far below those levels for which demonstrable damage to fish can be expected. Laboratory trout with a body burden of ^{65}Zn 10,000 times greater than that of river fish showed no detrimental effects. Similarly, trout with a body burden of ^{32}P 100 times greater than that of river fish showed no effect, although leukopenia occurred if the body burden was 1,000 times greater.

The determination of the actual dose absorbed by organisms and tissues is fundamental to the assessment of the potential effects of radiation in aquatic studies. Such determinations have been reported in only a few studies, however.

Although no dosimetric measurements were made for rainbow trout fed ^{65}Zn, Erickson (1966) calculated the dose from data on body burdens of serially killed fish fed ^{65}Zn daily over a period of 16–18 weeks. He estimated a total absorbed dose of 25 R for trout fed 0.01 μCi/g of body weight and 1,200 R for trout fed 1.0 μCi/g. Watson *et al.* (1959) also calculated a dose to the bone, gut, and muscle of approximately 6,000 R, 1,600 R, and 800 R, respectively, in trout fed 0.06 μCi of ^{32}P/g daily for 17 weeks. At this level, the growth rate of the fish was reduced. No deaths were attributable to radiation damage, and there appeared to be an increase in growth rate after cessation of isotope feeding.

In some of the experiments involving chronic dosages to marine fish eggs, absorbed doses were measured and calculated (Table 5) (V. M. Brown, 1962; Brown and Templeton, 1964). Absorbed doses calculated for anchovy eggs from the basic data of Polikarpov and Ivanov (1962) indicate that at the concentration where abnormalities become significant (10^{-10} Ci/liter), the dose absorbed was 0.16 mR/day. The total dose accumulated by the eggs was the same, since the period from fertilization to hatching was 20–24 hr. The calculated dose rate from the ^{40}K in the seawater and in the egg was about 0.02 mR/day. However, consideration must also be given to the additional background dose within the laboratory, which would be about 0.3 mR/day, or approxi-

TABLE 4 Effect of Chronic Ingestion of ^{32}P, ^{90}Sr-^{90}Y, or ^{65}Zn on Yearling Rainbow Trout (*Salmo gairdneri*)[a]

Treatment (μCi/g fish/day)	Duration of Feeding (wk)	Growth Depression	Significant Mortality	Leukopenia	Gut Damage	Concentration at End of Feeding (μCi/g wet weight)	
						Bone	Muscle
^{32}P							
0.006	25	no	no	no	no	–	–
0.06	25	wk 17	no	4 mo	no	1.8	0.23
0.60	25	wk 11	yes	17 days	yes	–	–
^{90}Sr-^{90}Y							
0.005	21	no	no	no	no	2.1	0.0022
0.05	21	no	no	no	no	28	0.078
0.50	21	wk 12	wk 15	wk 15	yes	248	0.27
^{65}Zn							
0.01	17	no	no	no	no	–	–
0.10	17	no	no	no	no	–	–
1.0	17	no	no	no	no	4.0	0.35
10.0	10	no	no	wk 10	no	–	–

[a] After Watson *et al.* (1959); Nakatani and Foster (1963); Nakatani (1966).

TABLE 5 Calculated and Measured Radiation Dose Rates from Contaminated Media to Fish Eggs under Experimental Conditions[a]

Fish	Concentration Factor	Nuclide	Water Concentration (pCi/liter)	Range of Dose Rate to Egg (per day)	Dose Rate from ^{40}K in Water and Egg (mR/day)	Approximate Background Dose Rate in Laboratory (mR/day)
Salmon (*Salmo salar*)	30	^{90}Sr-^{90}Y	10^3	1–10 mR	0.017	0.3
Brown trout (*Salmo trutta*)	25	^{90}Sr-^{90}Y	10^5-10^{-7}	60 mR–3.0 R	0.017	0.3
Plaice (*Pleuronectes platessa*)	0.35 10.0	^{90}Sr ^{90}Y	10^2-10^8	<0.1 mR–13.0 R	0.02	0.3

[a] After Brown (1962) and Brown and Templeton (1964).

mately twice that calculated to be delivered to the egg from the ^{90}Sr-^{90}Y.

Calculations of the dose rates from radionuclides absorbed by or adsorbed on the organism in the laboratory and in the natural environment are difficult to compute accurately for several reasons. The radionuclide is not uniformly distributed throughout the organisms or even throughout the volume of a single organ or cell. One organ or tissue may contribute to the dose of an adjacent organ, the concentration may change with time, and the structures of organisms are of varying size and shape. A further complexity is the wide variation in the energy of the emissions. Calculations for the assessment of the dose to fish eggs arising from immersion in contaminated seawater have been developed by Fedorov (1965) and Adams (1968).

A major advance in the determination of dose is the development of thermoluminescent dosimeters (TLD's). These dosimeters are commercially available in a variety of forms, most commonly as LiF powder in sachets or as LiF extrusions. The lower limits of detection in routine use are of the order of 20 mR, with an error of about 10 percent, and the techniques are rapidly being improved. The small size of the dosimeters allows them to be conveniently emplaced on or in living organisms to provide the best practical means of direct measurement of the radiation dose received by the organisms in their natural habitat. These dosimeters can also be used to measure the radiation dose in sediments directly, and they can be used in water with inexpensive buoyline-anchor support systems.

The performance of TLD's is very good. They are little affected by wide variations in environmental conditions and can therefore be left unattended for long periods. They are nearly energy-independent over a wide range of energies and can be reused.

The principal limitations of TLD's, however, are the same as those of other dosimeters that measure accumulated dose—only the total dose can be determined without knowledge of the rate at any time during the exposure period or of the radionuclides contributing to the dose.

Thermoluminescent dosimeters have been successfully used to measure the dose to fish in the Windscale area (Ministry of Agriculture, Fisheries and Food, 1967, 1968) and to aquatic organisms of the Columbia River in the vicinity of the Hanford Works (Watson, personal communication). Tests of TLD's in the tropical marine environment using a ^{60}Co source indicate that 0.7-MeV gamma emitters at concentrations of approximately 10^{-3} μCi/ml in seawater could be detected after 48-hr exposures (Baltzo and Held, 1968). It should be expected that the results of more such studies, relating concentration to radiation dose in the aquatic environment, will appear in the literature.

With the wider application of this technique, the results of experimental studies and field studies, and, it is hoped, of the predictions of effect, will become more meaningful in the future.

INFLUENCE OF ENVIRONMENTAL FACTORS ON RADIATION EFFECTS

The potential ecological interactions and the assessment of the radiosensitivity of marine ecosystems have been discussed in a previous chapter. The influence of two major environmental factors—temperature and salinity—on radiation effects on marine organisms will be briefly reviewed here. The effect of oxygen on radiosensitivity of organisms has been studied extensively (Bacq and Alexander, 1961) and will not be reviewed here. The literature shows little experimental work on the interactions of temperature and salinity with radiation effects, but research into this subject becomes increasingly important with greater use of nuclear energy to meet man's needs for power and for fresh water from the sea. Nuclear power and desalination plants imply not only some release of radioactive materials to the sea but also release of heat and highly saline water.

As the metabolism of poikilotherms is dependent on temperature, it is easy to understand the greater radiosensitivity observed following elevation of the temperature during or after radiation exposure. After exposure to 28,000 R, fecundity of a freshwater snail, *Physa acuta*, was reduced to 30 percent of that of the control snails in 20°C water and to 12 percent in 30°C water (Ravera, 1966). The hatchability of *Artemia* eggs exposed to irradiation by x rays was significantly reduced in warm water, but when cooler water was used, no significant reduction occurred (Iwasaki, 1964; Cervini and Giavelli, 1965). Goldfish, *Carassius auratus*, exposed to 8,000 R all died within 10 days when maintained at 22°C, but similarly irradiated fish survived for more than 100 days at 4°C (Hyodo, 1965). Gros *et al.* (1958) postulated that low temperatures (7°C) would protect the crucian carp, *Carassius carassius*, against radiation damage and that radiation, on the other hand, would protect the fish against cold by lowering the thermal tolerance.

White *et al.* (1967) reported on the combined effects of ionizing radiation, salinity, and temperature on the estuarine fish *Fundulus heteroclitus*. In a factorial experiment, fish were subjected to four levels of acute radiation (500, 1,000, 2,000, and 2,500 rads), three levels of salinity (5, 15, and 25$^{\circ}/_{\circ\circ}$), and four levels of temperature (12°, 17°, 22°, and 27°C). They found that different combinations of levels of temperature and salinity yield different LD_{50} values. The estimated LD_{50} values for different experimental conditions ranged from 300-350 rads to more than 2,500 rads. The significance of the experiments is the demonstration of the importance of the environmental factors on the radiosensitivity of aquatic organisms. In other words, the effects of radiation on aquatic organisms can be evaluated only along with the effects of other major environmental factors.

The investigations by Egami and Etoh (1966) and Etoh and Egami (1967) of the effect of temperature on the rate of damage accumulation and recovery in the fish *Oryzias latipes* showed that the processes of recovery from damage induced by external irradiations (x rays or ^{60}Co γ rays) are active to some extent at 23°C but remain almost undeveloped at 11°C. For poikilotherms, it seems likely that low temperature not only delays the development of radiation-induced damage but also decreases the rate of recovery (Egami *et al.*, 1967).

Fish with body burdens of ^{90}Sr and ^{131}I were subjected to thermal shock of 13°C from an acclimation temperature of 25°C. These body burdens did not impair their ability to withstand lethal temperatures. Indeed, there was an indication that their survival time was increased. The approximate beta dose to the bone and thyroid tissues was calculated for the fish containing maximum concentrations of ^{90}Sr and ^{131}I. These were found to be at least 10^4 rads to bone tissue and 10^5 rads to thyroid tissues (Ophel and Judd, 1966). Blaylock and Mitchell (1969) determined that temperature was an important factor in the $LD_{50/30}$ for *Gambusia affinis affinis*, since a difference of only 5°C resulted in a significantly different $LD_{50/30}$.

Few experimental data are available to explain how temperature, salinity, and other physicochemical environmental factors interact to affect the radiosensitivity of different aquatic organisms. It is recognized, however, that all organisms in nature have always been subject to different environmental stresses, including radiation, in varying degrees. The possibilities for damage to aquatic organisms will most probably arise through the combination and interaction of different environmental factors, since survival is rarely dependent on a single environmental factor.

BEHAVIOR AND METABOLIC STIMULATION

Scattered reports describe the use of behavioral criteria to determine the effects of radiation on aquatic organisms. These studies suggest that aquatic organisms apparently detect ionizing radiation, although the receptors have not been identified. In experiments with fish, particularly for high levels of external radiation, it has not been established whether the fish are responding directly to a radiation source or indirectly to induced products of water hydrolysis. O'Brien and Fujihara (1963) observed that larvae of the freshwater cichlid fish *Aequidens portalegrensis* became hyperactive during x-ray irradiation. Doses of 50 R stimulated activity, and larvae remained active for 90 minutes after irradiation. These larvae are not normally free-swimming but remain in clumps on the bottom. O'Brien and Fujihara also observed the influence of a ^{137}Cs source in the water, emitting less than 5 rads per hour at a distance of 1 in., on the behavior of 2-day-old cichlid larvae. The control larvae were scattered randomly around a sham source, but the experimental larvae avoided a ^{137}Cs source. Pravdina (1965) reports that carp avoided x-ray sources providing a dose rate of 2.5 rad/min.

Hyperactivity in fish has also been reported by Scarborough and Addison (1962). They observed definite periods of hyperactivity in the golden shiner, *Notemigonus crysoleucas*, during irradiation at doses of 7,200-18,000-R x rays given for periods of 6-12 min. Tsypin and Kholodov (1964) used γ rays from ^{60}Co at a dose rate of 0.1-0.5 rad/sec for a duration of 5-10 sec as a conditioned stimulus, and they used electric shock as the unconditioned stimulus on fish. Four of thirteen trials resulted in a motor response to the γ rays alone. A modification of the locomotor orientation of the turbellarian *Dugesia dorotocephala* has been reported under low-level chronic irradiation in the range 26 μR/hr to 240 μR/hr (Brown, 1962; Brown and Park, 1964). Field *et al.* (1964) compared the swimming activity of rainbow trout tagged with 16-μCi ^{60}Co wire tags and with nonradioactive tags. Tests made 16 hours after tagging showed a significantly higher swimming activity in ^{60}Co-tagged fish, which persisted for at least 26 hours.

Apparent stimulation of growth by radiation in aquatic organisms is reported for periphyton, certain marine invertebrates, young blue crabs, and rainbow trout. The growth of periphyton during 5 months in aquaria containing mixed fission products (3×10^{-6} to 6×10^{-7} Ci/liter) resulted in a greater biomass and greater species diversity than in control aquaria (Timofeyeva-Resovskaya, 1958). Glass plates with ^{90}Y on the surface at a concentration of 3.3 Ci/cm^2, with a calculated surface dose of 40 rads/day, exposed in the sea, enhanced the development of barnacles, bryozoans, calcareous worms, and small mussels (Dolgopolskaya *et al.*, 1959).

Engel (1967) reports that exposure of young blue crabs, *Callinectes sapidus*, to ^{60}Co at 3.2 rads/hr resulted in significantly more rapid growth than that of crabs irradiated at 7.3 and 29.0 rads/hr or of control crabs. Since molting is associated with growth, the percentage increase in width of molted carapaces was monitored; it was found to be related to the radiation dose rate. Crabs receiving 29.0 rads/hr, the highest dose rate, had the smallest percentage of increase in carapace width, and those that received 3.2 rads/hr had the largest increase in width, compared with the control crabs.

Metabolic activity of salmon (*Salmo salar*) eggs and fry was increased by the presence of ^{137}Cs at a concentration of 1 μCi/liter. Oxygen consumption increased gradually throughout development and was always higher in the ^{137}Cs-treated eggs than in the controls. Upon hatching of the eggs, the oxygen consumption increased by 50 percent in the controls and 200 percent in the treated aquaria (Neustroev and Podymakhin, 1966b). Although insufficient replicate groups were maintained to provide a rigorous statistical test, rainbow trout fed 0.01, 0.1, and 1.0 μCi ^{65}Zn/g of fish daily for 17 weeks showed more rapid growth than the control group (Nakatani, 1966).

Other examples of possible stimulation of organisms by radiation under laboratory conditions can be found; however, little or no knowledge exists about either the mechanisms involved or the significance of radiation as a possible stimulus to individuals, populations, or ecosystems.

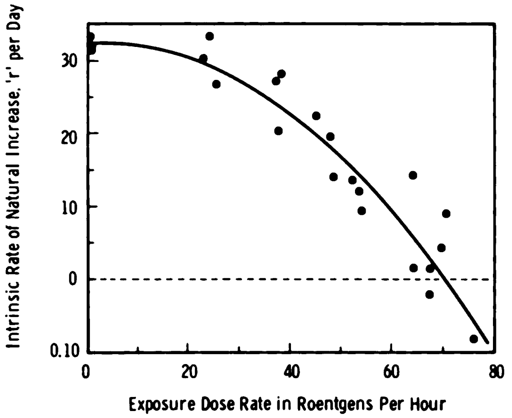

FIGURE 4 The effects of continuous gamma radiation on the intrinsic rate of natural increase of *Daphnia pulex*. (Reprinted with permission from Marshall, 1962.)

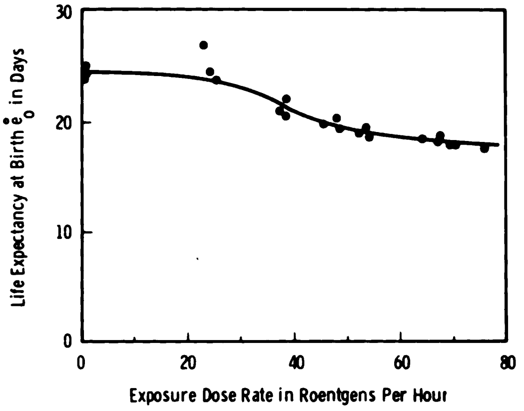

FIGURE 5 The effects of continuous gamma radiation on the life expectancy at birth, $\mathring{e}_0$, or average life-span, of *Daphnia pulex*. (Reprinted with permission from Marshall, 1962.)

RADIATION EFFECTS ON POPULATIONS

Ultimately, we are concerned with radiation effects on populations and ecosystems in the marine environment rather than with the demise of individuals. Marshall's (1962) work with *Daphnia* is indicative of a line study that is particularly significant. He investigated the effects of gamma radiation on the intrinsic rate of natural increase, *r*. The dose rates used ranged from 25 to 75 R/hr, and the cultures were exposed for about 19 hr/day.

Reproduction was entirely parthenogenetic. There was a continuous decrease of *r* as a nonlinear function of the dose rate (Figure 4). The decrease was almost entirely caused by a falling birth rate, which in turn resulted from direct effects of radiation on the ovaries. Average life-span was not greatly shortened, even at the highest dose (Figure 5). Interestingly, growth in length of individuals increased with increasing dose rate, which was interpreted as due to the utilization for growth of energy ordinarily spent in egg production.

Grosch (1962, 1966) followed the reproductive capacity of mass cultures of *Artemia* for 8 years. Cultures were repeatedly contaminated with sublethal amounts of ^{65}Zn or ^{32}P (~7-30 μCi/liter). The following are some of Grosch's conclusions:

> Although the number of adults seen in mass cultures may be equivalent, subcultures of control and experimental strains react differently to radioisotope additions. Strains descended from ancestors exposed to ^{32}P do not necessarily survive a second dose even though total dosage does not exceed the extinction dose given as a single addition. A period of recovery involving generations must intervene. Most notable is the consistent demonstration that the number of adults can be identical in different mass cultures, but that the reproductive potential of populations with different ancestral histories differs considerably. On the basis of pair mating tests, maintenance of mass cultures at an observed level of 300 adults per three liters requires only 0.2% of the reproductive potential of controls. Cultures of experimental origin utilize 1% or more of their potential to maintain the same total.

Grosch has a detailed discussion in which he compares his conclusions with those derived from *Drosophila* populations (Wallace, 1956) and *Tribolium* (Crenshaw, 1965), which showed increased genetic fitness following the irradiation of inbred strains.

ENVIRONMENTAL STUDIES

Pacific Proving Grounds

The first large-scale introduction of man-made radionuclides into a marine environment was at Bikini Atoll in 1946. Two 20-kiloton devices were detonated, the first an air burst and the second an underwater detonation in the 250-mi^2 lagoon, which has maximum depth of about 60 m. In succeeding years, through and including 1958, Bikini and Eniwetok became the Pacific Proving Ground. During that time, nuclear and thermonuclear devices with a total yield of many megatons were detonated at the atolls. Certainly, these atolls represent the most radioactively contaminated marine environment in the world, as far as is known from public announcements. And yet today, more than 23 years since the initial contamination of the atolls, a statement by Schultz in a 1947 report on the observed biological effects of the nuclear weapon test "Operation Crossroads" still holds true:

> Undoubtedly, countless animal individuals have perished at Bikini because of the atomic bomb experiments and still others may perish. But, this destruction of life in a large

> atoll like Bikini amounts to only an extremely small percentage of the total animal life. The overall picture of life on the reefs has changed little because beneath this surface layer, and from extensive adjoining unaffected areas, individuals have come forth to repopulate and occupy the reefs. The pressure of population from all sides into the damaged areas is very great and soon replaces the losses. Thus, nature begins the repopulation cycle, and, if given sufficient time, the wounded reefs will be cleansed of their contamination, biological equilibrium will be reached; and life will establish itself as in past millenniums—similar to that before man released the greatest destructive force in his history.

It is inconceivable that there were no radiation effects at the test sites. Evidently, where the prompt radiation at the moment of detonation was sufficiently intense to produce immediate visible effects, the concomitant effects of blast and heat virtually eliminated the populations. Furthermore, those individuals suffering sufficient injury from the residual radiation to be readily recognized are soon eliminated. In other words, gross radiation injury in marine organisms has not been seen at Bikini and Eniwetok because seriously injured individuals do not survive the natural rigors of the environment, and the more subtle injuries are exceedingly difficult to detect.

Bikini and Eniwetok were intensively studied before and following the test series (Revelle, 1954; Hines, 1962), including extensive studies of fish (Schultz *et al.*, 1953, 1960; Hiatt and Strassburg, 1960; Welander, 1957; Welander *et al.*, 1967), corals (Wells, 1954), and algae (Taylor, 1950; Dawson, 1957), and yet in none of the reports on marine organisms is there reference to anomalous individuals.

However, Gorbman and James (1963) studied the thyroid histologically and observed thyroid tissue damage in fish collected at Eniwetok Atoll 30 days to 8 months following nuclear detonations. Although no radioiodine was detectable in the fish at the time of examination, it seems clear from indirect evidence that the observed anomalies were caused by radioiodine. Blinks (1952) examined physiological functions of sessile algae at Bikini a year after the first atomic tests; at that time, the dose rate was estimated to average 20 to 30 mR/day. He concluded that there was "no noticeable alteration of many normal somatic functions."

The land plants and animals were subjected to greater intensities of radiation both from external sources and from internally deposited radionuclides, but even here, lasting effects of radiation on populations or on the ecosystem are not apparent.

Jackson (1967) has reconstructed the story of the survival of rat populations at Eniwetok Atoll. Engebi Islet, with an area of 260 acres, was subjected to radiation in 1948, 1952, 1954, 1956, and 1958. The accumulated dose a year after contamination was 44 R (1948), 11,000 R (1952), and 118 R (1954). Prior to the nuclear tests, probably only the Polynesian rat (*Rattus exulans*) was present at Engebi, but early in the testing period the roof rat (*R. rattus*) was unintentionally introduced. The former makes a nest under surface vegetation and debris, while the roof rat often burrows. Rats were killed following the detonations in 1948, 1952, and 1954, but a sufficient number of individuals survived to re-establish the population. In summary, Jackson states:

> The Eniwetok story is made difficult by taxonomic confusion and lack of specimens. At best, a hypothetical reconstruction can be attempted. Early in the test program at Engebi Islet, the Polynesian rat (*Rattus exulans*) was exterminated, probably by heavy surface radiation. Prior to the detonation of a thermonuclear detonation in 1952 [14 megatons detonated approximately 3 mi from Engebi], the roof rat (*R. rattus*) had become established on Engebi; and a nucleus survived the heavy initial radiation by being deep in burrows. Calculations show that repopulation by 1954 and 1955 was theoretically possible. The decline of the Engebi rat population from its high density in the mid-fifties was a result, probably, of a change in the carrying capacity of the environment.

Cole (1951) reported that the insects of Bikini were studied for structural anomalies in 1947 and that none were found. He further stated that "continuing studies of *Drosophila* cultures, taken in the living form at Bikini Island, have not thus far [1947-1951] revealed genetic abnormalities in excess of normal variability." However, more detailed studies following further contamination did reveal probable genetic effects in *Drosophila*. Stone and his co-workers (Stone *et al.*, 1957; Stone and Wilson, 1958, 1959; Stone *et al.*, 1962) made extensive genetic studies of wild populations of *Drosophila* subjected to fallout in 1954 at Bikini, Rongelap, and Rongerik Atolls, comparing them with populations of uncontaminated islands. The estimated gamma dose to the Bikini *Drosophila* population was 3,000-8,000 R, but the absorbed dose is not known. The conclusion was reached that the genetic load of detrimental and lethal factors of the irradiated *Drosophila* populations had been increased but was returning to a normal range. These tests were possible only because of the extensive and sophisticated genetic techniques for *Drosophila*. Similar established techniques are not now available for the marine organisms; hence, we can only speculate that they, too, might have had an increased deleterious genetic load but have recovered or are continuing to recover.

Rongelap Atoll was heavily contaminated with radioactive fallout in 1954 (Glasstone, 1962; Dunning, 1957). From observations in 1956, Fosberg (1959a and b) described the condition of plants at islets where the integrated dose was approximately 3,360 R. He suggested that the situation observed might be primarily due to the radiation. Held (1963a), following further studies at Rongelap, suggested that other environmental factors might be more important than radiation in this case. The question is not resolved, nor is it likely to be without controlled experiments with the species involved under varying conditions.

In a more intensive study, Palumbo (1962) documented the recovery of land plants at a site 2½ miles from a multimegaton detonation at Eniwetok. The vegetation was destroyed by heat and blast, and the estimated external dose to the vegetation was 400 R in 200 days. Palumbo noted that regrowth of the vegetation occurred in 6 months and described two minor anomalies. Palumbo discusses many of the factors influencing plant growth in the island environment and reviews the anomalies in plants reported by others at the test site and in other areas of the Central Pacific. He points out that similar anomalies have been observed in areas of heavy fallout and in uncontaminated areas. Again, there can be no doubt that there was damage and even the destruction of individual plants by radiation; but the specific examples of damage or killing caused by radiation or by other factors, or in combination with other factors, cannot be sorted out. The important point is that although individuals may be debilitated or destroyed, the ecosystem recovers.

The land-dwelling hermit crab, *Coenobita* sp., and coconut crab, *Birgus latro*, are subject to higher levels of chronic radiation from internally deposited radionuclides than any other organism studied at the atolls. The levels of ^{90}Sr and ^{137}Cs were found to remain virtually constant at 4,500 pCi of ^{90}Sr per gram of skeleton and 450 pCi of ^{137}Cs per gram of muscle in *Coenobita* sp. at Eniwetok over a period of two years (Held, 1960).

Parallel studies of *Birgus* sp. at Rongelap Atoll showed that the crabs contained more than 700 pCi ^{90}Sr per gram of skeleton and 100 pCi of ^{137}Cs per gram of muscle over a period of 10 years, 1954-1963. No gross anomalies were noted among these crabs, and no obvious population changes were noted during this fallout period; however, population studies as such were not made.

Observations at Bikini Atoll in 1964 (Welander *et al.*, 1967) showed that vegetation nearly covered the islet in a mass that was impenetrable without cutting. The vegetation, exclusive of roots, taken from an area 10 m in diameter in a *Scaevola* sp. community in 1967 yielded 108 kg/hectare (wet weight), and no gross anomalies were seen on the islet (Held, 1967, unpublished field notes). The abundance and size of fish and of spiny lobsters and coconut crabs at Bikini Atoll appear to be greater than ever, which does not, of course, reflect a beneficial effect of radiation but presumably results from the absence of predation by man.

Irish Sea

Apart from the Pacific Proving Grounds, the Irish Sea coastal area adjacent to the Windscale reprocessing plant of the United Kingdom Atomic Energy Authority probably represents the most important area known with respect to the degree of contamination in the marine environment (Mauchline and Templeton, 1964).

Studies of the relationships between radionuclides and man have been of prime importance; some studies, though limited, have considered effects of radiation in the environment. Morgan (1960) reported that effects of radiation had been sought in plaice (*Pleuronectes platessa*) caught in the region of the Windscale discharge, but none has been established so far. The area, which had been carefully surveyed before and after discharges began, showed that there were no changes in bottom organisms that could be ascribed to the effects of the discharge.

Calculations (Dunster *et al.*, 1964) of the external radiation dose rate to benthic organisms that would result if the seabed were at the derived working limit of 0.1 μCi/g total beta activity are about 45 mR/hr, or 1 R/day, mainly of beta radiation.

In a series of experiments concerned with the uptake by plaice eggs of six radionuclides, ^{85}Sr, ^{137}Cs, ^{144}Ce, ^{103}Ru, ^{88}Y, and ^{95}Zr/^{95}Nb, data were obtained on the rates of accumulation and concentration factors (Woodhead, 1970). The concentration of these radionuclides in the egg can now be calculated for any concentration of the radionuclides in seawater to which they may be exposed. Tentative conclusions have been drawn that the dose rate to the eggs of plaice from the contaminant activity in the spawning areas off St. Bees Head, near Windscale, was 9.1×10^{-2} μrad/hr compared with 7.0×10^{-1} μrad/hr from the natural ^{40}K in the seawater.

Studies have also been made of the radiation dose received by fish in the Windscale discharge area (Ministry of Agriculture, Fisheries and Food, 1967). The plaice, *Pleuronectes platessa*, is a seabed resident and is known to spend its early life, up to the beginning of its third year, inshore. Calculations were made of the potential dose that a fish could receive on the basis of data from measurements of seawater, seabed, and the fish themselves over the previous 5 years at a point 2 miles south of the outfall and 1½ miles offshore from the pipeline. The radionuclides ^{137}Cs, ^{106}Ru, ^{95}Zr/^{95}Nb, ^{144}Ce, and ^{90}Sr were considered, since these represent the major contributions to dose in terms of the various known factors of concentration in the major segments of the environment. In the calculations, allowance was made for fish movement and type of bottom, and these indicated that an annual dose of 7.3 rads could be accumulated, with most of the dose contributed by the seabed (Woodhead, 1968 personal communication). The seawater and internal radionuclides would only contribute about 1 percent of the total expected dose (Table 6). The annual dose might increase to 40-50 rads if the fish spent the whole year close to the pipeline outlet on a silty bottom.

This calculation suggested that the use of thermoluminescent dosimetry was warranted, and 2,500 marked plaice were released in the vicinity of the outfall in 1967 (Ministry

TABLE 6 Estimates of Dose Rates to Plaice from Fission Products at Windscale[a]

Source	Type of Radiation	mR/week
Seawater	γ	0.01
	β	0.01
Seabed	γ	48.8
	β	91.4
Radionuclides in muscle	γ	0.013
	β	0.057
		140.3[b]

[a] Data from Woodhead (1968, personal communication).

[b] Or 7.3 R/year.

of Agriculture, Fisheries and Food, 1968). Each fish carried two lithium fluoride dosimeters, incorporated in a Petersen fish tag, one to measure the accumulated dose on the upper surface and one to measure the dose on the under surface. In the first few months, over 243 marked fish were recovered, and preliminary data indicated an integrated exposure of 4.5 R for the bottom dosimeter. This was equivalent to a dose of 10 R per year. The ratio of top to bottom dose was 0.73. The differential response of dosimeters placed on the top and the underside of the fish indicated the expected response to beta radiation. The agreement between calculated and measured dose is extremely good.

Gamma dose rates measured 1 m above the Ravenglass mud flats, near Windscale, from January 1965 to June 1967, averaged about 140 μR/hr, compared with 10-15 μR/hr background. Over sandy beaches, dose rates ranged from 11-16 μR/hr, compared with a background rate of 8-12 μR/hr. The mean dose rate over mud, from $^{106}Ru/^{106}Rh$, $^{144}Ce/^{144}Pr$, and $^{95}Zr/^{95}Nb$, has been calculated as about 1 mR/hr, or 8 R/yr (Jeffries, 1968).

Thermoluminescent dosimetry measurements in 1965, using LiF powder in sachets implanted in the mud at a point where $^{106}Ru/^{106}Rh + ^{144}Ce/^{144}Pr$ levels were about half those reported by Jeffries, gave values of 100 mR/wk in the top 1 in. of mud, 50 mR/wk at a depth of 3 in., and <20 mR/wk at depths to 17 in. Organisms in the top 3 in. might then be exposed to a radiation dose of 4.0 R/yr, comparable to that received by fish near the outlet (Templeton, 1968, personal communication).

Oak Ridge

Very few studies have been made of natural populations exposed to chronic radiation higher than background. The salivary chromosomes of the larvae of *Chironomus tentans*, which inhabit the contaminated bottom sediments of White Oak Creek and White Oak Lake at Oak Ridge National Laboratory, were analyzed for 5 years for chromosomal aberrations (Blaylock, 1966). Calculations and measurements of the adsorbed dose for the larvae living in the sediments gave values of 230-240 R/yr, or 1,000 times background for that area. More than 130 generations had been exposed to this or greater dose rates over the previous 22 years. The conclusion was that the ionizing radiation from the contaminated environment was increasing the frequency of new chromosomal aberrations in the irradiated population, but that the new aberrations were eliminated by natural selection. Also, the present level of chronic irradiation has not affected the frequency of the endemic inversions.

Blaylock (1969) also studied the fecundity of a natural population of fish, *Gambusia affinis affinis*, that had been exposed to chronic irradiation in White Oak Creek for many generations, compared with a control population. The calculated dose rate from the bottom sediments was 10.9 rads/day. A significantly larger brood size occurred in the irradiated than in the nonirradiated population, although significantly more dead embryos and abnormalities were observed in the irradiated broods. These results suggest that an increased fecundity is a means by which a natural population having a relatively short life cycle and producing a large number of progeny can adjust rapidly to an increased environmental stress caused by radiation.

We return then to a concept that has recurred in this report: Man, as an individual, is the critical biological target in predicting the consequences of introducing radioactive materials into marine environments. If the radionuclides are present in concentrations acceptable for man, the individual, then it is difficult to conceive that there will be more than subtle effects on ecosystems—perturbations that would probably be indistinguishable from those due to causes other than radiation. On the other hand, it is not only conceivable, but probable, that with increasing uses of atomic energy, accidents will occur that will result in damaging concentrations of radionuclides. In preparation for these contingencies, there is a pressing need to increase the sensitivity of methods for studying the response to radiation of populations and ecosystems in the marine environment.

EFFECTS ON RESOURCES

The extrapolation of the results of laboratory experiments into the practical terms of their effects on marine resources must be made with care since, without evaluation of the natural variations related to changes in fecundity, mortality, and recruitment, quite erroneous conclusions can be reached. Polikarpov (1966) suggests "inhibition and degeneration of a number of food fishes and other radio-sensitive organisms" and "rapid development and multiplication of bacteria, microphytes and radio-resistant forms of invertebrates" as a result of radiation modification of the marine environment.

Zaytsev and Polikarpov (1964) and Polikarpov (1966), based on their previously discussed laboratory data, have calculated the time needed to halve the stocks of various species of Black Sea fishes in relation to the proportion of the eggs damaged by radiation. These workers conclude that there is reason to assume that the fisheries will be adversely affected, and may, in many instances, cease to function if fish stocks are reduced to one half or less. They consider that from the commercial standpoint, radiation damage must not affect more than 10 percent of the eggs, though even at this level, catches will be perceptibly reduced.

Garrod (1966) points out the fallacy of this argument. For marine mammals or for certain fish species with low fecundity, a 50 percent mortality of the young or of the eggs will be reflected as a 50 percent reduction in recruits to the stocks, though this does not generally occur in the marine environment. In the general case of organisms with high fecundity, reduction in the spawning stock does not necessarily result in a decrease in the number of recruits. The capacity of the environment to support young fish is limited, and this capacity can be satisfied by the eggs from a small number of adults, the superfluous eggs and larvae being reduced by density-dependent mortality.

The magnitude of the mortality is also pertinent in the evaluation of the effects of radiation on marine resources. Garrod indicates that for stocks of arctic cod with high fecundity, survival is 1 in 10^4 and can be as low as 1 in 10^5. The addition of 50 percent mortality in the egg stage due to the effects of radioactive contamination of the environment would not noticeably alter the existing mortality rate of 99.99 percent. This would also be true of all highly fecund species.

While it may be true that the fecundity of both the individual and the population could be reduced by radiation effects, it is extremely doubtful that these would influence stock size beyond the normal range related to environmental changes, except in very heavily exploited stocks. It is certain, however, that with the techniques of assessment now available, it would not be possible to obtain an unbiased measure of the effect attributable to radiation alone.

RESEARCH NEEDS

- Further studies are required concerning the concentrations of naturally occurring radionuclides and natural background radiation doses in the environment as a baseline for studies of the effects of radiation.
- Time curves for acute exposures of lethal doses should be extended, with particular attention to exposure as a function of age.
- Because of the conflicting nature of the present experimental data on the effects of low-level chronic irradiation on developing embryos, more studies should be established under rigorous, controlled experimental conditions. The effects of other environmental "stress" factors, such as salinity, temperature, oxygen, and pollutants, must be studied and expanded to include the interaction of these factors with radiation effects.
- More sensitive parameters of radiation effects on individuals, populations, and communities should be developed. Long-term studies, in which factors such as rates of growth, morphological abnormalities, onset of maturity, and reproductive capacity are considered, for example, should be emphasized. Chromosomal studies under experimental and field conditions should be extended, and the somatic and genetic consequences of such changes on populations and ecosystems should be evaluated.
- Measurements of absorbed dose using microdosimeters should become standard practice in experimental work. In areas subject to radioactive contamination, studies should be initiated to determine the radiation regime in the environment. The data obtained should be closely correlated with measurements of concentration of radionuclides in order that the historical radiation regime can be determined.

SUMMARY

Radiation is not a recent introduction to the marine environment, since low levels from environmental and cosmic sources have been present throughout geological time. Lethal amounts of acute radiation differ widely among marine organisms and are related to variations such as species, age, physiological status, and body size. These variations are further complicated by the interaction of environmental factors such as temperature and salinity. Exclusive of the eggs of fish and larvae of invertebrates and fish, most marine organisms for which data exist are relatively radioresistant.

Limited studies on the effects of chronic exposure have been conducted. These have been limited to selected developmental stages and indicate that, with the possible exception of some Russian data, the dose necessary to evoke an unequivocally detectable biological response is considerably above that of concentrations of radionuclides in the environment as a result of controlled waste disposal operations.

Studies on the genetic consequences of radiation exposure to population indicate that, despite larger numbers of mutations, increased utilization of reproductive capacity maintains a population at preradiation density.

Field studies on the effects of radiation indicate that our best technologies and methods cannot demonstrate effects on marine ecosystems, at prevailing dose rates, that are clearly and uniquely attributable to ionizing radiation.

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Chapter Ten

EVALUATION OF HUMAN RADIATION EXPOSURE

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The presence of artificial radioactive materials in the marine environment must be considered in relation to radiation exposure of people who use the sea and its products. Some exposure is inevitable and, indeed, has always existed from naturally occurring radionuclides. To this natural background exposure must now be added some additional radiation dose as a consequence of man's entry into the age of atomic energy.

The magnitude of exposure that is likely to result from artificially produced radionuclides depends upon many complex relationships. These relationships involve how, where, what kinds, and in what amounts radioactive materials are introduced into the seas, and the ways and extent to which the seas and their resources are used by man. It is the purpose of this chapter to identify the relationships that are now recognized as important in assessing human radiation exposure, to discuss ways in which the presence of radioactive materials can be translated into estimates of radiation dose to people, to provide perspectives on the magnitude of exposure from marine sources in relation to acceptable dose limits, and to summarize the status of radioactive contamination in the seas after the first quarter century of use of atomic energy.

USE OF THE SEA IN RELATION TO RADIATION EXPOSURE

Influx of Radioactive Materials

The seas have been the intended or casual recipient of significant quantities of artificial radionuclides since 1945, when the fissioning of ^{235}U and ^{239}Pu was first undertaken on a massive scale. The earliest additions consisted mostly of short-lived neutron activation products present in the cooling water of the plutonium-producing reactors at the Hanford Works in the State of Washington. This radioactive material reached the North Pacific Ocean via the Columbia River. Only a few months after the startup of the reactors, atomic weapons were detonated in the atmosphere, and the oceans of the world began to receive the fallout of fission products and other debris. In the following year, atomic devices were exploded underwater at Bikini Atoll.

In a general sense, the ways in which radioactive materials were added to the marine environment in the first year and a half of the atomic age characterize the types of additions with which we are still concerned:

Chronic discharges of low-level wastes from operating reactors and fuel processing plants. Such discharges are planned and monitored and can be controlled at the source. They result in a gradual buildup of longer-lived isotopes to some equilibrium level, and the area of greatest concern is ordinarily in the region close to the discharge point.

Worldwide fallout from detonations that inject debris into the atmosphere. In this case, the source is widespread, fluctuates greatly in intensity at different times, is not under control, and results in low-level, but worldwide, contamination in the oceans.

Acute releases from single incidents at or below the sea or ground surface. Such releases may result in high levels of contamination in local regions—contamination that decreases with time but spreads in area. In the early years, acute releases occurred from weapons tests. In future years, they may result from accidents at shore installations, to ships, or to isotopic power sources. Several large-scale engineering projects that have been proposed under the Plowshare Program of the U.S. Atomic Energy Commission (AEC) could also cause acute releases. These differ from the accident cases in that the place, time, and nature of the incident is under control. The magnitude of the release is likely to be substantially greater than in most accidents, however.

Additions of radioactive materials to the sea that are more characteristic of contemporary uses of atomic energy also include

The disposal of low-level packaged waste and intermittent releases from nuclear-powered ships. Such disposal represents an intermediate case between the chronic discharge of liquid wastes from fixed installations and the acute releases from single incidents. Repetitive disposal into designated dump areas requires administration comparable with that for chronic discharge.

TABLE 1 Consumption of Fish and Shellfish

Country or Area	Direct Consumption (g per capita per day) Area Average	Specific Locality Surveys
Oceania (Cook Is.)	260[a]	–
Southern Nigeria	244[a]	–
Norway	105[a]	–
Portugal	100[b]	–
Japan	60[b]	–
United Kingdom	30[b]	807[c]
Finland	30[b]	308[d]
United States	15[b]	390[e]
India	5[b]	180[f]

[a] Rao (1962).
[b] Food and Agriculture Organization of the United Nations (1960).
[c] Preston (1966).
[d] Jokelainen (1967).
[e] Hanson *et al.* (1967).
[f] Bhat *et al.* (1967).

The use of sealed radioisotope power sources for such purposes as navigational lights and space satellites, which increases the potential inventory of radioisotopes in the marine environment; however, these radioisotopes are not biologically available except after accidents.

Radiation exposure of man as a result of the introduction of artificial radioactive contaminants into the marine environment may occur by one or more of several pathways, and only careful evaluation can determine the most important or critical pathways in particular situations. The following pathways are the ones that normally merit consideration, however.

Pathways of Exposure

FOODSTUFFS

The consumption of contaminated marine foodstuffs, including fish, seaweeds, and manufactured products, is ordinarily the exposure pathway of greatest importance. This is particularly true of the heavily populated northern hemisphere, where sea fisheries produce over 70 percent of the world's sea harvest. Closely related is the use of the sea as a source of fresh water, a use that is expected to increase with the application of nuclear desalination. Desalination processes tend to exclude radioactive materials from the finished water. On the other hand, waste water from nuclear desalination plants will contribute to the burden of radioactive contaminants in the marine environment.

The levels of contamination in the edible portions of marine plants and animals may be many times higher than that in the seawater because of biological reconcentration processes. The levels that are acceptable for any specific situation depend upon the rates of consumption of locally derived marine foods, and these rates vary widely. Statistics for countries as a whole are usually of little relevance with respect to specific regions, and only surveys of local consumption habits can provide the data required to determine permissible concentrations in particular dietary items. The data in Table 1 illustrate the wide range of consumption rates. The rate for a specific locality within a country may be 30 times that of the national average, and the national average for some countries is 50 times that of others.

SAND AND SEDIMENTS

Some radioactive debris enters the sea as particulates, and some radionuclides in solution are easily adsorbed to the surface of particles. The result is radioactive contamination of sand and sediments. If the contamination results principally from surface adsorption, then finely divided sediments, which present the greater surface area per unit weight, will exhibit the highest degrees of contamination. This mode of radiation exposure is most apt to be of significance in silted estuaries or coastal water areas with soft bottoms. Operational experience in the United Kingdom (Mitchell, 1967b; Jefferies, 1968) has shown that open sandy beaches in the vicinity of effluent discharge points are not likely to be a problem, but silty estuaries up to distances of several miles from a discharge point may be of concern (Jefferies, 1968). Radioactive particulates discharged to estuaries and coastal waters, especially gamma-emitting radionuclides, may lead to quite significant exposure rates (Preston and Dutton, 1967; Dutton and Steele, 1966).

The use of coastal waters as recreational areas has increased very greatly in the last decade among the affluent nations of the world. Since these nations are also the likely contributors to radioactive contamination of the same areas, the radiological implications of this use should be considered. However, discharge restrictions based on the contamination of marine foodstuffs will, in general, also limit exposure from swimming and from the beaches to negligible proportions.

FISHING GEAR

Fishing gear—more particularly, commercial fishing gear—may become contaminated directly by adsorption of radioactive materials from the water or indirectly from radioactive particulates if the gear is operated on the seabed or in very silty waters. This is more likely to be of concern with static fishing gears, such as the salmon bag-nets used on the Caithness coastline in Scotland near the discharges from the Dounreay Experimental Reactor Establishment (Mitchell, 1967b; Morgan, 1967). Experience at the U.K. Atomic Energy Authority's Windscale Works has shown that trawl gear operated in the outfall area, even over soft bottoms containing up to 10^4 pCi of fission product radionuclides per gram of dry sediment, does not become substantially contaminated.

OTHER PATHWAYS OF EXPOSURE

Other pathways that may lead to very minor degrees of human radiation exposure include

Seaspray driven onto land or beaches—particularly spume, which scavenges fine particulates as it travels over the water surface, leaving patches of finely divided material of increased radionuclide content after stranding and collapsing (United Kingdom Atomic Energy Authority, 1965).

Seaweed, trash fish, and spoiled fish used as fertilizer or animal feed.

Periodic inundation of pastureland with seawater that contains radioactive contaminants from nearby nuclear plants.

The dredging of harbors and the maintenance of buoys in areas where radioactive wastes have been discharged.

The sea as a mineral resource is already receiving considerable attention, particularly in continental shelf areas, with respect to the extraction of salts and drilling for oil and natural gas. The technology for the economical recovery of manganese and other valuable minerals from the bed of the deep sea may also be developed soon. Such mining of the sea might require prior consideration of possible radioactive contamination if the mineral resources are in areas where radioactive materials have been discharged.

Future undersea exploitation involving underwater residence by man may require advance consideration of the proximity to radioactive waste disposal areas; at present, however, radioactive contamination of the water or seabed hardly seems a credible restriction on such uses.

ACCEPTABLE DOSE

The Concept

Exposure of man to ionizing radiation entails a risk of deleterious biological effects, and if man wishes to benefit from the use of atomic energy, some risk will be involved. Radiation doses must therefore be limited to a level at which the risks involved are acceptable to the individual and to society as a whole; such doses may be termed "acceptable doses."

Recommendations with respect to acceptable doses are made from time to time by the International Commission on Radiological Protection (ICRP) on the basis of the conservative assumption that "down to the lowest levels of dose, the risk of inducing disease or disability increases with the dose accumulated by the individual," (International Commission on Radiological Protection, 1959) and that this increased risk of developing biological effects has a linear relation to dose. This concept assumes that there is no threshold dose below which no deleterious effect is produced. In light of these concepts and many years of practical experience with radiations of various kinds, both with man and with experimental animals, acceptable doses are set so that there is a very low probability of biological effect. These limits, which are for continuous exposure, are not to be regarded as absolute upper limits, but rather as guides. It will normally be quite practicable to keep the actual levels of irradiation very much lower, and the Commission states that every effort should be made to do so, compatible with economic and social considerations. Undue importance should not, however, be attached to occasional exposures that exceed the limits.

The Commission's "permissible limits" (ICRP, 1966b) distinguish between occupationally exposed persons, members of the general public, and populations, and in the context of radioactive contamination of the marine environment, it is the recommendations in relation to individual members of the public and of populations that will, of course, apply. These recommendations are intended to restrict the risk to the individual of developing deleterious somatic effects. The limits for members of the public correspond to one tenth of the recommended maximum permissible dose rates for occupationally exposed persons. The lower limit on dose for the public is deemed desirable in order to protect children and, among other things, in recognition of the absence of some controls that apply to radiation workers, such as employee selection, supervision, and monitoring. The limitations imposed on exposure of populations are based on considerations of heredity (genetic effects) and will be determined by the magnitude of the dose to individuals as well as by the number of persons exposed. No specific recommendations are made for a maximum somatically significant population dose, but the limits for individuals ensure that the frequency of somatic injury in a population will remain at a very low level.

TABLE 2 Recommended Dose Limits

Part of Body	ICRP Dose Limit[a]
For individuals	
Gonads, red bone marrow, whole body (uniformly irradiated)	0.5 rem/yr
Skin, bone	3.0 rem/yr
Thyroid	3.0 rem/yr (1.5 rem/yr children under 16)
Hands, forearms, feet, ankles	7.5 rem/yr
Other single organs (including the gastrointestinal tract)	1.5 rem/yr
For populations	
Whole body	5.0 rem/person for 30 yr

[a] Acceptable dose limits for members of the public recommended by the International Commission on Radiological Protection (1959). Similar limitations are recommended by the Federal Radiation Council (1960, 1961).

General Application

For both individuals and populations, the exposures under consideration here are those that are above natural background and in addition to exposures received by a patient during medical treatment. Exposure may result from either internal or external sources, and the recommended dose limits, given in Table 2, are intended to include radiation from both sources.

For radioactive materials deposited in the body, dose rates are related to a permissible body (or organ) burden for specific radionuclides, and these in turn are related to a permissible continuous daily intake sufficient to establish and maintain that body burden. The ICRP has expressed these permissible daily intakes in terms of maximum permissible concentrations in air and in drinking water (ICRP, 1959). In the case of long half-life radionuclides with slow turnover rates, the permissible body burdens and the resulting dose rates may not be achieved for tens of years. Consequently, early exposure history related to these intakes, or any body burdens derived from them, will result in only a small fraction of the acceptable dose.

The establishment of such a model for exposure from internally deposited nuclides and the derivation of the associated control criteria imply assumptions about the average anatomy, physiology, and behavior of man. To this end, the ICRP has defined a "standard man," representing a typical occupationally exposed adult. Individual variations related to size, sex, age, physical condition, eating habits, and so on are not taken into account in defining maximum permissible body burden and associated parameters.

The general application of acceptable doses is intended for planning purposes and is normal for situations resulting from controlled operations. If the source of exposure is uncontrolled, as it might be following a reactor accident or a nuclear explosion, abnormal exposures may arise that call for remedial action. It is unwise to stipulate action levels at which remedial measures should automatically be invoked, since the measures themselves may involve a degree of risk. However, the ICRP regards such measures as mandatory if whole-body exposures of 100 rads are likely to result from an uncontrolled situation. Some guidance concerning suitable emergency exposures may be obtained from recommendations of the United Kingdom Medical Research Council (1960) and the Federal Radiation Council (FRC) of the United States (1964, 1965). Both reports suggest upper limits on the contamination levels that would result in annual doses to the critical organs broadly equivalent to the annual exposure permitted radiation workers. Such exposures are considered acceptable if they are experienced by the same population only once or twice, with some years between exposures, and if the size of this population is small compared with the total population of the country or area concerned.

Application to Radioactive Contamination of the Marine Environment

The Brynielssen committee of the International Atomic Energy Agency (IAEA) (1961), convened in 1958 to consider the problems of radioactive waste disposal into the sea, endorsed the ICRP recommendations and gave as its collective opinion that radiation exposure resulting from disposals would probably be limited to small fractions of the population. It follows, therefore, that genetic exposure should not be significant and that the critical control should be based on exposure of individuals. Thus, limitations of radiation exposure from artificial radionuclides in the marine environment may be based on the recommended ICRP dose limits for individual members of the public. The use of somatic rather than genetic criteria has been adopted by, among others, the Federal Radiation Council (1961) and the United Kingdom Minister of Housing and Local Government, Minister of Welsh Affairs, and Secretary of State for Scotland (1960).

In applying these dose limits through the derived criteria, such as maximum permissible concentrations in drinking water, it must always be remembered that the actual doses received by individuals will vary, depending on variation in parameters such as age, size, and sex. Such variations can, to a large extent, be allowed for by careful selection of "critical groups" that are reasonably homogeneous with respect to age, diet, and other factors affecting dose (ICRP, 1966a), and the Commission states that "it will be reasonable to apply the appropriate Dose Limit to the mean dose of this group." Some individuals may therefore receive doses higher than the dose limit, but since at the levels of risk implied such infringements will be minor in character, this approach is, in general, satisfactory.

Where a critical group cannot be defined, a safety factor should be applied to the derived concentration limits. It is recognized that the value of such a factor will vary according to particular circumstances, but in the United Kingdom, for example, a safety factor of 10 is applied to preoperational assessments of waste disposal to the marine environment, and, if after some years of operation it is still not possible to accurately define a critical group, the dose limitations are applied to the habits of the most exceptional individual found through a survey of local habits. This approach has been endorsed in principle by the authorities in the United Kingdom (United Kingdom, Minister of Housing and Local Government, Minister for Welsh Affairs, and Secretary of State for Scotland, 1960), who recognize that surveys include but a sample of the entire population, and consequently that there may exist some person with such exceptional habits that his exposure is greater than the dose limit. Again, at the levels of risk involved, this is considered to be a satisfactory approach.

The final stages in applying acceptable dose limits depend upon the results of surveys of local habits that have as their objective the identification of those individuals who will be subjected to the greatest radiation exposure. This unusually high exposure may result from their activities in the local environment or, in some cases, in an environment some distance from the contaminated area. Their behavior needs to be assessed in quantitative terms—for example, in grams of marine foodstuff consumed per day or in hours per year spent on a beach or in working contaminated fishing gear. Such terms are needed to convert the ICRP maximum permissible daily intakes, or dose rates, into derived working limits for the environment in question.

After some operational experience has been gained for a particular situation, it should be possible to confirm the validity of the critical pathways predicted by the above procedure and to identify the radionuclides contributing the most significant fraction of the dose limit. Field measurements should also permit confirmation of the critical organ or organs involved in the irradiation. When this stage has been reached, it should be practicable to pursue more detailed inquiries directed toward isolating the critical group of individuals. Finally, isolation of this group will permit the determination of average behavior patterns in the group and, if necessary, suitable adjustment of the provisional derived working limits.

In some situations, levels of contamination are so low that they are virtually indistinguishable from natural background. If calculation of absolute upper limits of exposure shows that it is far below the exposure considered as acceptable, then detailed studies need not be undertaken. In such cases, it may be possible to dispense with the routine environmental monitoring operations normally established for the areas of significant contamination.

METHODS OF EVALUATION

In order to put a particular level of radioactive contamination in the marine environment in proper perspective, estimates must be made of the radiation dose to man that may result. It is now rather standard practice to make such estimates prior to the planned introduction of radioactive materials into the sea, not only to determine whether the proposed operation is in fact feasible (safe), but also to establish preliminary guides concerning the maximum amount of such materials that might be introduced at selected sites over appropriate intervals of time. In making such predictions, careful consideration must be given to the exposure pathways that are most likely to result in the highest doses and to the particular individuals who will receive these doses. Early identification of the critical pathways and critical population groups is also an important prerequisite to the design of an efficient and effective environmental surveillance program.

Following the actual introduction of potentially significant quantities of radioactive materials into the marine environment, a re-evaluation of the probable dose to man can and should be made. As pointed out in the preceding section, the evaluation made after an installation has been in operation for some months is of value not only in providing a refined estimate of the actual doses received by critical populations in relation to acceptable dose limits but also in confirming the validity of the assumptions used in making the preoperational estimates. Several cases are now available where comparisons of preoperational predictions can be made with "postoperational" experience measured after startup. These will be discussed in the section on disposal of radioactive wastes (p. 253). Usually, the preoperational predictions are shown to have been highly pessimistic and to have resulted in tentative discharge guides that were more restrictive than necessary to maintain the radiation dose to people within the prescribed limits. This was the intent, of course, and is most desirable. Nevertheless, it must be borne in mind that when postoperational assessments are made, the basic limit is the acceptable dose to man and not the tentative permissible discharge rate or permissible concentration in seawater calculated as a part of the preoperational evaluation. Another incentive for updating guides derived during preoperational evaluations is that a particular exposure pathway predicted at the outset to be critical may ultimately be shown to be of less importance than some other pathway.

Evaluations made both before and after the planned discharge of radioactive materials should serve not only to provide a technical basis for prudent control of the releases but also to inform both the lay and the scientific communities of the degree of risk associated with planned and actual discharges. Much of the public's wariness of radioactive contamination of the environment is justified because the true risks to man have not been published in either a timely or an easily understood manner.

Dose Prediction

Predictions of the radiation dose that may be received by people as a result of radioactive contamination of the marine environment ordinarily begin with a comprehensive review of the uses that man makes of the sea in the region of interest and with the selection of a few postulated pathways by which internal and external exposures of greatest magnitude appear most likely to occur. Comprehensive evaluation requires that the separate contributions from all pathways of significance be added together so that the combined dose can be compared with the ICRP or FRC recommendations. This consolidation of dose contributions should include artificial radioactive contaminants of the terrestrial and freshwater environments as well as contaminants of the marine environment.

In practice, it has been found that one, or a very few, pathways are so dominant that the multitude of alternate routes that can be conceived contribute only relatively insignificant doses. The pathways requiring detailed evaluations invariably include the possible contamination of edible products harvested from the sea in the region of interest and, where appropriate, may include the potential external exposure from beaches, fishing gear, and other pathways.

Fundamentally, the mechanics of predicting the dose that may be received by people has four parts:

Estimating the concentrations of the contaminants that will exist in the seawater at places and times of interest.

Estimating the relationships that will exist between the concentrations in the water and in seafoods, sediments, beaches, fishing gear, and other materials that are used directly by man.

Estimating the rates of consumption of particular seafoods by critical population groups and the extent (time and distance) of exposure to materials that can deliver an external dose.

Converting the estimated intakes of radionuclides and the intensity of the deposited contaminants into estimates of internal and external dose (rem), consolidating these estimates with doses received from other environmental sources, and comparing them with recommended limits.

In the last decade, the Committee on Oceanography of the National Research Council has had several opportunities to consider and develop in detail procedures for predicting acceptable rates of discharge of radioactive materials into the sea (National Academy of Sciences-National Research Council, 1959a, 1959b, 1962). The International Atomic Energy Agency (1961) has likewise published definitive guidance for this practice, and several nuclear installations have used these basic procedures to establish tentative limits for discharges to the sea or to provide evidence that anticipated releases will be well below the levels that would result in dose rates of concern.

The first evaluation made by the Committee on Oceanography was in response to a request by the U.S. Atomic Energy Commission that questions concerning the disposal of packaged low-level waste into coastal waters of the Atlantic Ocean and the Gulf of Mexico be examined. The recommendations of the working group assigned to this study were published in 1959 (National Academy of Sciences-National Research Council, 1959a). This working group identified the potential concentration of radioisotopes in edible marine products as the factor that would most limit waste disposal. Maximum permissible concentrations in seafoods were calculated for several radioisotopes on the basis of intake rates recommended by the National Committee on Radiation Protection (NCRP), which at that time did not distinguish between exposure limits for radiation workers and members of the public at large (NCRP, 1953). In essence, these were the NCRP recommendations for maximum permissible concentrations in drinking water adjusted for the rate of intake of seafoods and for concentration of the radioisotopes by the marine organisms.

The assumption was made that individuals eating the seafoods obtained all of their protein from this source. This is a very conservative assumption in relation to the food habits of North Americans. Other assumptions used conservative, but best available, data on the extent to which fish, shellfish, and other edible marine products might concentrate specific isotopes from the seawater, and on the rate of dispersion of the contaminants from packaged radioactive waste. The report specified maximum permissible concentrations for several radioisotopes in seawater and the associated permissible rate of disposal of wastes—i.e., the disposal rate that would not result in greater concentrations in the seawater within disposal areas 2 miles in diameter.

In June 1958, the AEC made an additional request of the Committee on Oceanography to undertake a study of the problems of disposal of radioactive wastes from nuclear-powered ships (National Academy of Sciences–National Research Council, 1959b). The working group appointed for this new project used basically the same approach as the Atlantic and Gulf Coast group—i.e., consideration of the rate of dispersion by ocean currents, concentration of isotopes from the seawater by edible marine products, consumption of the seafoods by humans, and maximum permissible rates of intake recommended by the NCRP. The nuclear ships study dealt with a more complex set of disposal conditions, however. The marine environment was classified into four zones: inshore waters within 2 mi of the coastline; the coastal area, between 2 and 12 mi from the shore; the outer continental shelf, beyond 12 mi from shore; and the open sea. The most stringent limits were assigned to the inshore zone because some individuals might obtain all of their protein requirements from fish harvested there, and because dispersion would be relatively poor in this zone. This report also recognized that people may receive radiation exposure from other environmental sources, e.g., the atmosphere and the land, and an allocation was made for the fraction of the total that should be assigned to the sea. This allocation permitted radionuclide intakes one third as large as those recommended by the NCRP (1953).

The International Atomic Energy Agency *ad hoc* panel on radioactive waste disposal into the sea has presented a reasonably detailed description of the step-by-step procedure that may be used in arriving at maximum permissible rates of introduction of radioactive materials into particular marine locales (IAEA, 1961), and many of these details are not repeated here. All of the studies cited above use a method that is now referred to as the "critical pathway approach"—a procedure that is recommended by Committee 4 (Application of Recommendations) of the ICRP (1966a).

The Critical Pathway Approach

The critical pathway approach involves evaluation of a sequence of events through which radioactive material introduced into the marine environment is diluted, perhaps reconcentrated, and ultimately reaches man either in food or via material with which he comes in contact. Prediction of the radiation dose that man will receive from an introduction of radioactive material (or the reverse procedure, the calculation of tentative guides for acceptable rates of introduction) not only requires a number of source terms but also requires the use of several mathematical simulation models, transfer coefficients, and basic assumptions. Beginning at the point of introduction, the significant parameters are considered in the following sections.

KINDS AND QUANTITIES OF RADIOACTIVE MATERIALS INTRODUCED

This source data must be available in terms of rates of introduction (e.g., Ci/mo) of specific radionuclides. Data presented in terms of concentrations (e.g., μCi/liter) is not very useful unless accompanied by information on the volume (flow rate) of the water involved. Further, data expressed only in terms of gross beta or alpha activity is of little value unless the calculation is being carried out solely to demonstrate that the amount of radioactive material involved is insignificant in relation to acceptable dose limits.

PHYSICAL AND CHEMICAL FORM OF RADIONUCLIDES AT TIME OF INTRODUCTION

These characteristics determine whether the material will be dispersed promptly in the area of introduction more or less as a solution or whether it will settle to the bottom. Once deposited on the seabed, the radioactive material may become a significant source of external exposure, as well as a continuing source, by dissolution, to the seawater and to marine organisms. In the absence of direct measurements on such deposited material, assumptions must be used to define the rate of dissolution. The mean residence time on the bottom is important in relation to the radioactive decay rates (half-lives) of the nuclides.

INITIAL MECHANICAL DILUTION

Initial dilution by this means depends upon the manner in which the material is introduced into the sea. It is important for the elimination of differences in densities that might delay dilution by advection and turbulent diffusion. Inadequate initial dilution could lead to unnecessarily high concentrations in the vicinity of the point of introduction.

DILUTION BY NATURAL MIXING PROCESSES OF THE SEA

These processes (advection and turbulent diffusion) are of paramount importance in accomplishing the required dilution of the radionuclides within the time and distance constraints peculiar to the disposal site. Mathematical models are ordinarily used to predict the manner in which the concentrations of the nuclides diminish in space and time. These processes are discussed in detail in Chapter 4.

For each disposal site, there will be some minimum volume of seawater that will be of interest, and even though small regions within the "critical volume" will have higher concentrations, this will not affect the final outcome of the prediction. One method of identifying the size of the critical volume is to relate productivity of the critical food organism of the area to its rate of consumption by a critical population. For example, if a local population of 100 people subsists largely on fish produced in the vicinity of the site of introduction of the radionuclides, and if the quantity of fish consumed is 50,000 lb/yr, then the smallest area of interest is that required to produce an annual harvestable crop of the species of interest of 50,000 lb. This might be an area of 1,000 acres. The critical volume is, then, the mass of water available in relation to this acreage. The rate of replacement of the water in the critical volume then determines the flow (i.e., m^3/day) available for dilution of the radioactive materials. This critical volume approach was used by a working group of the National Research Council to evaluate the potential hazard of a sealed nuclear power source to marine fisheries.

AVAILABILITY OF INTRODUCED MATERIAL TO MARINE BIOTA

Most predictive calculations assume that radioisotopes added to seawater will be adsorbed or absorbed by marine organisms in a manner identical with that of their stable counterparts already present in solution in the seawater. It is on this basis that concentration factors are usually derived. It is also on this basis that the "specific activity approach" (to be described later) is derived. In actual practice, however, freshly introduced radioisotopes cannot be expected always to have the same availability as the stable isotope of the same element in seawater because the freshly introduced material is initially apt to be in a different physical and chemical state. For specific situations, it may be possible to predict whether the radioisotope will be more or less available to organisms of interest.

CONCENTRATION FACTORS

Perhaps the greatest uncertainty in predictive calculation is the selection of an appropriate concentration factor whereby the estimated concentrations of specific radionuclides in the seawater can be translated into the concentrations that will result in the marine food products of interest. This general topic is discussed in several other chapters of this volume, and several publications are now available that record values deemed appropriate by various authors (IAEA, 1961; National Academy of Sciences–National Research Council, 1962; Mauchline and Templeton, 1964; Bryan *et al.*, 1966: Polikarpov, 1966).

The wide variations that have been observed among different environments and even among closely related species in the same environment emphasize the need for careful consideration of the specific characteristics of each site. In dealing with radioisotopes with relatively short half-lives (days or weeks), it is also important to bear in mind that their accumulation by fish to a level that is in equilibrium with the environment requires a significant period of time and that radioactive decay will reduce the quantity present to levels substantially below those anticipated from the use of unmodified concentration factors.

CONSUMPTION OF MARINE PRODUCTS

It is now conventional to select or hypothesize a small population group, or even individuals, that eat extremely large quantities of the seafoods that are expected to become contaminated through the introduction of radionuclides. Such an approach is endorsed by both the ICRP (1966a), which uses the term "Critical Group," and the FRC (1961), which refers to "the exposed population." In the absence of factual data derived from dietary surveys, predictive calculations have usually employed obviously safe assumptions, i.e., that the entire protein requirement is derived from the seafood of concern.

Desired refinements in the estimated seafood consumption by the critical group include not only the true proportion of the diet that is made up of the seafood of interest, but also the proportion of this food that actually originates in the critical area (market dilution) and the lag time that ordinarily exists between the harvest of the food and actual consumption. This lag time can allow substantial radioactive decay of short-lived nuclides.

STANDARD MAN

The ICRP and the FRC have computed the daily rates of intake of radionuclides that will eventually lead to the allowable radiation dose rates in various organs of the body. In making these calculations, it was necessary to fix a number of physiological and anatomical characteristics that are implicit in the calculations. These "standard man" parameters include such things as body and organ sizes, chemical composition, metabolic rates, and intake and excretion rates (ICRP, 1959). To the extent that individuals differ from the standard man, the dose estimates that are associated with specified intakes of particular radionuclides will also differ. The greatest differences will obviously occur for infants and children, and the ICRP is therefore developing an additional set of parameters associated with the "standard child." For predictions of radiation dose from marine foods to critical population groups, there appears to be no need for concern that deviations from the standard man assumptions will introduce serious errors, except perhaps when some foods are fed to infants in quantities approaching or exceeding those eaten by adults. The capability of accurately calculating the dose received from seafoods by the critical population group improves as the identity of the group, and thus the physical and metabolic characteristics of its members, becomes better defined.

EXTERNAL EXPOSURE

In addition to the reconcentration of radioactive materials in food chains leading to man, accumulations or reconcentrations may also occur by physical, chemical, and biological processes that will lead to external exposure. Such accumulations can result from the deposition of undissolved particulate material on the seabed and beaches, the adsorption of ions from solution onto sediments or suspended matter that will later settle out, and the adsorption or "growth" of contaminated materials onto fishing gear and other equipment immersed in the sea. Prediction of the magnitude of the external exposure that will result from these sources involves not only estimation of the concentration factors that will occur on the sources of exposure but also the amount of time that members of the critical population group will be exposed (e.g., the number of hours during a season that a fisherman handles his nets).

APPLICATIONS

The critical pathway approach to evaluating the dose to critical population groups has been used for several radioactive waste disposal operations throughout the world:

Nation	Use
United States	Radioactive waste disposal into Atlantic and Gulf coastal waters
	Radioactive waste disposal from nuclear-powered ships
	Hanford (now Richland) plutonium production plant discharge to Columbia River
	Oak Ridge National Laboratory discharge to the Clinch River
United Kingdom	Atomic Energy Authority sites
	Windscale (Irish Sea)
	Dounreay (north coast of Scotland)
	Winfrith (southern coast of Britain)
	Civil nuclear power stations (total of 9)
	Royal Navy nuclear submarine base (Chatham)
	U.S. Navy nuclear submarine base (Holy Loch)
	Nuclear shipping
France	Commissariat à l'Energie Atomique site (Cap de la Hague)
Sweden	A.B. Atomenergi Studsvik
India	Bhabba Atomic Research Center
	Tarapur Atomic Power Station
	Rajasthan Atomic Power Station

The Specific Activity Approach

The National Research Council's Committee on Oceanography was asked by the AEC in 1958 to consider the problems of disposal of low-level radioactive wastes in the Pacific Ocean off the North American coast (National Academy of Sciences–National Research Council, 1962). The approach used by the group set up for this study was basically different from that used by others. Rather than using the NCRP recommendations on rates of intake of radioisotopes, they used the NCRP (1959) values for maximum permissible body burdens of individual radioisotopes in combination with expected amounts of the nonradioactive isotopes of the same elements in the body. This resulted in a set of maximum permissible "specific activities," e.g., microcuries of radioactive ^{90}Sr per gram of stable ^{88}Sr ordinarily present in the body. They pointed out that, if the maximum permissible specific activity were not exceeded in the environment where foods were grown, there was no mechanism by which the specific activity in the foods, or in the humans who ate the foods, could exceed the limit. Therefore, maximum permissible concentrations of radioisotopes in seawater could be calculated on the basis of the specific activities in the seawater.

The advantage of the specific activity approach is that it eliminates the need for speculation on the extent to which marine organisms might concentrate isotopes from the water, and it eliminates the need for apportioning some fraction of the total intake of radionuclides to marine foods. Such an approach can be used to evaluate the disposal of radioisotopes into the sea because the chemical content of the seawater is quite uniform.

The specific activity approach is very conservative, especially for North Americans, because it assumes that an individual's entire food supply is derived from the sea. It also has limitations for elements that are not common in the human body, and the working group found it necessary to use "stand-ins" for a number of elements for which human data were unavailable. They also postulated that some elements, such as zinc, cobalt, iron, and copper, might enter the sea as complexes that would not permit complete isotopic dilution with the element in the seawater prior to uptake by marine organisms. To allow for this, an additional safety factor of 10 was introduced.

Another limitation of the specific activity approach is that it is not applicable when the gastrointestinal tract is the critical organ. Here, the principal exposure is from food that passes through the gut rather than from the radioisotope after deposition in the body. The working group resolved this problem by reverting to the critical pathway approach of estimating the concentration that might occur between the seawater and the marine organisms (i.e., as used by the Committee on Oceanography working group for nuclear-powered ships) and assuming a generous consumption rate for the seafoods.

In making a predictive calculation using the specific activity approach, many of the same parameters used in the critical pathway approach must still be applied. These are

Detailed knowledge of the kinds and quantities of radioactive materials introduced
Knowledge of or assumptions about the physical and chemical form of the radionuclides introduced
Calculation of the initial dilution by mechanical mixing
Calculation of the dilution that will result from natural mixing processes of the sea
Assumptions about the relative availability of the introduced material to the marine biota

Parameters used in the critical pathway approach that are not needed with the specific activity approach are

Assumptions about the concentration of radionuclides by marine organisms from the seawater
Knowledge of the kinds and quantities of marine products used as food. (It is assumed that the entire food supply has its origin in the marine environment of interest.)

Follow-up Evaluations

Estimation of the mode and magnitude of radiation exposure to people that might result from the introduction of radioactive materials into the marine environment is prerequisite to the deliberate discharge of waste and to the assessment of risks that may be associated with large-scale accidents. It must be remembered, however, that the actual exposures received by people after a new radioactive waste disposal operation has begun, or after the occurrence of an accident, may differ substantially from early predictions, both in terms of the critical pathways of exposure and in terms of the magnitude of the dose. Once the introduction of radioactive materials into the sea from a new installation has started, the preoperational predictions will, in large measure, have served their purpose, and attention should be shifted to a determination of the actual concentrations of radionuclides that are accumulating. This means that provisional limits placed on the rates of introduction of contaminants will very likely need to be revised, and it may be possible to dismiss safety factors that were added solely because of uncertainties in rates of dispersion and of biological reconcentration.

Follow-up evaluations should, insofar as practical, be based on measurements of the materials directly responsible for human exposure. This means the species of fish, shellfish, and seaweed actually consumed by the public, the beaches used by the public, and the gear handled by the fishermen. Measurements made of the concentrations of contaminants in seawater, in plankton, and in the sediments of the seabed are useful as reference points from which interrelationships can be developed, but they have only limited use in evaluating radiation exposure to people.

Once the critical exposure pathways and the critical populations have been confirmed by measurements made after waste disposal has begun, estimates of the dose received can be refined by comprehensive investigation of seasonal trends in the levels of contamination and in the use of the marine products and zones of interest. Greater attention can also be given to the size, age composition, and habits of the critical population. If gamma emitters are involved, it may be possible to measure the actual body burdens in individuals that make up the critical population. This technique is now being used at the Hanford plant to determine the quantities of ^{65}Zn acquired by people who drink water and eat fish from the Columbia River or who eat oysters from the coasts of Washington and Oregon (Foster and Soldat, 1966).

The determination of actual body burdens of radionuclides acquired from environmental sources represents something of an ultimate technique for evaluating the radiation dose from internally deposited sources. When such methods are feasible, they should be favored over the estimation of dose from the sampling and analysis of water and foodstuffs. However, practical considerations will ordinarily dictate that the measurements be made on the water and foodstuffs and, thus, that dose estimates be made using assumed rates of ingestion and the ICRP metabolic models. Where the concentrations of radionuclides in foods are so low that radiochemical analyses are difficult and imprecise, it may be desirable to sample some other organism or group of organisms (e.g., plankton) that concentrate the radionuclides of interest to levels that are easily detected. Such organisms are often termed "indicator species" because they can be used to indicate the relative levels of contamination in the environment (including the concentration in edible species) and because they can show increasing or decreasing trends in these levels. In order to use the concentrations of radioactive materials observed in indicator species to estimate dose to people, it is first necessary to establish the relationship between concentrations in the indicator species and in species actually used by man. In situations where the dose to man is of any real significance in respect to limits, it will be possible to make the measurements directly without the use of indicator species.

The Windscale Evaluation: An Example of Acceptable Dose in Relation to the Radioactive Contamination of Coastal Waters

The preoperational evaluation at Windscale, on the Cumberland coast of Britain, was the first detailed evaluation of radioactive material in the marine environment using the critical pathway approach. It involved an assessment of the rate of dilution of activity, its reconcentration in materials previously identified as playing an important role in man's use of the local environment, and a carefully controlled period of experimental discharges of radioactivity during which a safety factor of 10 was introduced between the calculated maximum rates of discharge and those employed in the study (Seligman, 1956; Dunster, 1956; Fair and McLean, 1956; Dunster, 1959).

The results of these studies completely substantiated the preoperational estimates of discharge rates and confirmed the major pathways by which radioactivity is returned to man. The results of subsequent surveillance programs at the site have continued to demonstrate the acceptability of the Windscale sea discharges in terms of the criteria discussed in earlier parts of this chapter (Mitchell, 1967b; Dunster, 1959; Morgan, 1964; Dunster *et al.*, 1964; Morgan, 1965; Langley and Templeton, 1965; Templeton and Preston, 1966; Howells, 1966; Preston and Jefferies, 1967).

The consumption of laverbread (an edible product manufactured from seaweeds of the genus *Porphyra*) is the limiting route or critical pathway by which radionuclides discharged by the Windscale plant (particularly ^{106}Ru) return to man. This critical pathway for human radiation exposure is described in detail by Preston and Jefferies (1967, 1969). The maximum rate of consumption of laverbread, found in a sample of consumers obtained by local survey, was 75 g per day, but the distribution of observations indicated the possible existence of a few consumers who might be eating laverbread at a greater rate than this.

A recent survey (Preston and Jefferies, 1969) of laverbread consumption rates identified the heavy consumers more accurately. In the sample obtained, rates of consumption ranged between 75 g and 388 g per day. The median rate of consumption of this group, based on the distribution of observations in the sample, is 160 g per day, or approximately twice the previous maximum rate. The group is composed of adults of both sexes and is estimated to comprise 170 individuals in a total laverbread-consuming population of about 26,000 persons.

The survey also reviewed the supply, distribution, and processing of the seaweed, and confirmed that the concentration of ^{106}Ru in the laverbread was only about half that in the seaweed because of the incorporation of extra water during processing (Preston and Jefferies, 1967). Parallel investigations of market dilution based on market sampling of the processed foodstuff revealed an average ^{106}Ru dilution factor of 5 between the activity as measured in seaweed in the vicinity of the outfall and the concentration of the radionuclide in laverbread (Preston and Jefferies, 1969).

The calculations that follow are based on the results of the recent surveys but ignore the market dilution factor, since this varies from year to year with the particular market situation and has in recent years tended to decrease in value as Windscale seaweed has come to occupy a larger fraction of the market. The calculations that follow are designed to illustrate the degree of somatic exposure experienced by the critical group and to compare this exposure with the genetic exposure received by the South Wales population.

CRITICAL SOMATIC EXPOSURE: ESTIMATED DOSE TO LOWER LARGE INTESTINE

The average annual dose to the gastrointestinal tract (lower large intestine) between 1959 and 1965 was estimated by Preston and Jefferies (1967) at 0.35 rem. This estimate was based on a consumption rate of 75 g of laverbread per day. Adopting the new median consumption rate of 160 g per day for the critical group, this estimate becomes 0.74 rem per year, compared with the ICRP recommended dose limit of 1.5 rem; the range of estimated doses experienced by the critical group as a whole was 0.35 to 1.78 rems (Preston and Jefferies, 1969). (See Figure 1.)

GENETIC EXPOSURE IN THE SOUTH WALES POPULATION

The mean acceptable annual genetically significant dose is calculated as follows: The size of the laverbread-consuming population based on the 1962 (Preston and Jefferies, 1967) and 1967 (Preston and Jefferies, 1969) laverbread surveys is

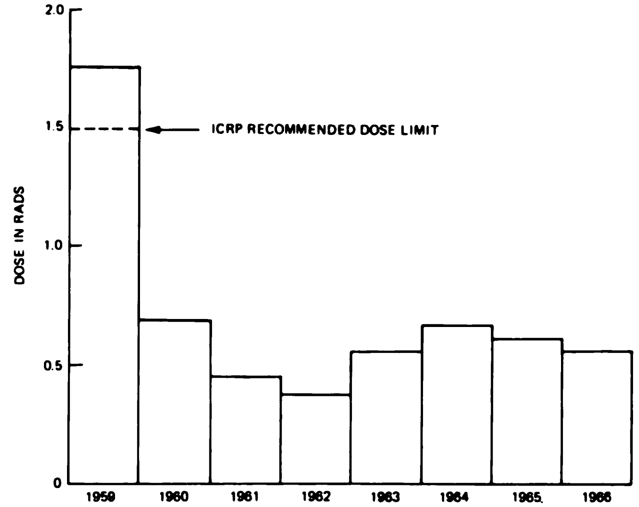

FIGURE 1 Estimated annual dose (rads) to the lower large intestine of adults eating 160 g/day of laverbread manufactured from Cumberland *Porphyra*. (Modified from Figure 4 of Preston and Jefferies, 1967.)

2.6×10^4 persons. The population of South Wales, in the laverbread area, is 1×10^6 persons. Therefore, on the basis of ICRP recommendations, the mean acceptable annual genetically significant dose, D, is

$$2.6 \times 10^4 / 1 \times 10^6 \times 30 \text{ yr} \times D = 0.1 \text{ rem}$$

$$D = 0.13 \text{ rem}$$

This calculation carries the conservative assumption that South Wales has a semiclosed population, with restricted interchange of genetic material with the rest of the U.K. population, and allots 1/10 of the U.K. recommended genetic exposure rate from radioactive waste disposal (U.K., Minister of Housing and Local Government, Minister for Welsh Affairs, and Secretary of State for Scotland, 1960) to the Windscale sea disposal operation.

The genetically significant dose actually received by the whole laverbread-eating population is computed as follows: From above, the average annual gastrointestinal tract dose for the critical group consuming 160 g of laverbread per day is 0.74 rem. The average rate of consumption of the whole laverbread-eating population is 14.6 g per day. By ratio, the average gastrointestinal tract dose for the population is 0.065 rem. From the ratio of ICRP permissible intakes, the gonad receives a dose of 1/600 of that to the lower large intestine. Hence, average gonad dose is $0.065 \times 1/600 =$ 0.0001 rem.

SOMATIC AND GENETIC DOSES IN RELATION TO RECOMMENDATIONS

From the above calculations, the ratio of the dose estimated for the gastrointestinal tract of the critical population to the ICRP limit is 0.74 rem/1.5 rem = 1/2, and for the genetic dose is 0.0001/0.13 = 1/1,000.

CONCLUSIONS

Without allowing for dilution of Windscale-contaminated seaweed with seaweed from other sources entering the South Wales laverbread market, the average annual dose to the critical organ (gastrointestinal tract) of laverbread eaters over the years 1959–1965 was 50 percent of the recommended dose limit. The somatic dose is 500 times more restrictive than is the genetic dose, even though calculations for the genetic case were based on very conservative assumptions for the mixing of the South Wales population with the total U.K. population.

HUMAN EXPOSURE FROM RADIONUCLIDES IN THE SEA

Natural Radioactivity

Compared to the land, the sea presents a relatively friendly radiation environment. In the sea, natural radionuclides are present in lower concentrations than in most rocks and soils (Folsom and Harley, 1957), and the only ones of significance in seawater that enter food chains leading to man are ^{40}K and ^{87}Rb. The dose from ^{87}Rb is insignificant, and the dose from the burden of ^{40}K in the body will be about 17 mrems per year regardless of whether it is acquired from seafood or from food of terrestrial origin.

Worldwide Fallout

Some fraction of the radiation exposure received by man from fallout radionuclides results from his use of the sea and of food from the sea. For most of the world's population, this marine-derived exposure constitutes only a small fraction of the total exposure from fallout. The major fraction of internally deposited fallout radionuclides is derived from terrestrial food chains (United Nations Scientific Committee on the Effects of Atomic Radiation, 1964).

Calculations by the Food and Agriculture Organization (FAO) of the United Nations (1960) for ^{90}Sr indicate that fish contribute less than 0.5 percent of the total daily intake of this radionuclide. This small uptake applies even in Japan, where marine fish are an important part of the total diet (Table 1). Because the ^{90}Sr/Ca ratio in marine fish is much lower than in other items in the human diet (FAO, 1960), the marine contribution to ^{90}Sr actually deposited in human bone would be much less than 0.5 percent.

The marine contribution to ^{137}Cs intake by people is also small—probably considerably less than 10 percent (United Nations Scientific Committee on the Effects of Atomic Radiation, 1962). The ^{137}Cs/K ratio in marine fish is much lower than that in milk and meat, so that the percentage contribution to retained ^{137}Cs (and hence to the internal radiation dose) is even less.

From the dose commitment calculations made by the United Nations Scientific Committee on the Effects of Atomic Radiation (1966), the worldwide average dose from ^{90}Sr to human bone (actually to "cells lining bone surfaces") up to the year 2000 is almost exactly 5 mrem per year. The marine-derived portion of this dose can then be approximated as less than 0.5 percent of 5 mrem per year, or an average of less than 0.025 mrem per year. A similar calculation for ^{137}Cs whole-body dose results in less than 10 percent of 0.5 mrem per year, or an average of less than 0.05 mrem per year.

The above calculations refer to average doses for the world population. In some parts of the world, there are small population groups that depend on fish and other marine organisms for most of their food. An approximate upper limit for the marine-derived ^{90}Sr and ^{137}Cs dose rate to such people can be calculated by using the maximum fish-consumption rate (807 g/day) found by Preston (1966) for English fishermen together with the concentrations of these radionuclides measured in the flesh of commercial fish (0.04 pCi ^{137}Cs/g wet weight; 0.007 pCi ^{90}Sr/g wet weight) (Mitchell, 1967b). These lead to possible daily intakes of 32 pCi ^{137}Cs/day and 6 pCi ^{90}Sr/day. Using conservative biological parameters, these intakes could result in dose rates as high as 0.36 mrem/year to the whole body and 30 mrem/year to parts of the skeletal bone.

In addition to ^{90}Sr and ^{137}Cs, many other fallout radionuclides reach the sea; these include ^{22}Na, ^{54}Mn, ^{60}Co, ^{65}Zn, ^{95}Zr-^{95}Nb, ^{103}Ru, ^{106}Ru, ^{131}I, ^{141}Ce, and ^{144}Ce-^{144}Pr. Most of these have short half-lives, and their concentrations in the marine biota that are used for human food are transitory and usually very low (Morgan, 1965).

One exception is the radionuclide ^{55}Fe (half-life of 2.6 years), which by 1965 had been recorded in humans, marine fish, and plants (Palmer and Beasley, 1967). Concentrations found in marine fish flesh are much higher than those in terrestrial animals. Consequently, small population groups that have a high proportion of fish in their diet have been found to have body burdens of ^{55}Fe approaching 1,000 nCi. The resulting internal radiation dose to the red blood cells in these individuals (if they retain this body burden) has been estimated to be about 30 mrem/year (Palmer and Beasley, 1967).

The other marine radionuclides mentioned above contribute only a small fraction to the external dose received by the world's population. Although the exact fraction cannot be calculated at this time, from our knowledge of the world population distribution and habits, we know it is small.

Local Fallout

Many studies of radionuclide concentrations found in marine organisms as a result of local fallout from nuclear explosions have been published (e.g., Japan Society for the Promotion of Science, 1956; Hines, 1962, p. 341 et seq.). Much of the information gathered is not directly usable for the estimation of the marine-derived fraction of the total radiation dose received by persons living in the local fallout zone. However, some generalizations can be made with regard to the relative importance of fallout radionuclides to man.

In the edible portions of marine fish, the major radionuclides found (Lowman, 1960, 1963) have been ^{65}Zn, ^{55}Fe, ^{57}Co, ^{58}Co, ^{60}Co, and ^{54}Mn. None of these elements are fission products. They presumably result from neutron activation of materials in and around the device. Fission-product concentrations in fish are very low in comparison with the above radionuclides even some years after the contaminating event (Lowman, 1960).

Other edible marine organisms, such as shellfish, contain much the same spectrum of radionuclides as fish in which the cobalt radioisotopes predominate. The fission products ^{106}Ru + ^{106}Rh and ^{95}Zr + ^{95}Nb have also been found in clams (Lowman, 1960).

One example is available in which it was possible to determine the dose contribution from marine food in a population exposed to "close-in" or local fallout from a nuclear explosion. In this case, a group of people from Rongelap Atoll were returned to their homes several years after the atoll had been heavily contaminated by local fallout. From data given by Cohn *et al.* (1960), it is possible to estimate that about 10 percent of the ^{90}Sr bone dose to these people was 0.36 μCi (Cohn *et al.*, 1960), which would deliver approximately 30 mrem per year internal whole-body irra- to include the land crabs in their diet, this would reduce the marine food contribution of ^{90}Sr to 2 percent of the total ^{90}Sr intake.

For whole-body radiation, the marine contribution was somewhat greater. Virtually all of the body burden of ^{65}Zn was contributed by fish in the diet. This ^{65}Zn body burden was 0.36 μCi (Cohn *et al.*, 1960), which would deliver approximately 30 mrem per year internal whole-body irradiation. However, terrestrial food chains contributed

0.68 μCi of ^{137}Cs, resulting in a dose of about 110 mrem per year. Consequently, the marine contribution would amount to some 20 percent of the total internal whole-body dose. Since this small native population had a relatively high proportion of seafood (including fish viscera) in its diet, these percentage contributions to internal radiation dose are probably near a maximum for similar occurrences.

The contribution of marine radioactivity to external dose was very small, the gamma dose rates over the sea being much lower than those over land (Dunning, 1957).

Plowshare Program

Calculations of the marine-derived radiation exposure to humans are being made for proposed nuclear excavation projects associated with the Plowshare Program of the U.S. Atomic Energy Commission (Martin, 1969).

Disposal of Radioactive Wastes

There are a considerable number of locations in the world where radioactive wastes are discharged into the sea, or where such disposals are planned. Most of these discharges are associated with experimental or power reactors, but the best-documented case is the multifacility U.K. Windscale site. Fuel-processing wastes are included in the effluent from Windscale, which is discharged directly into the coastal waters. The discharge of reactor cooling water from the Hanford plant in the State of Washington is also well-documented. The Hanford reactor effluent enters the Columbia River about 360 miles upstream from the Pacific Ocean, but some nuclides persist and can be detected in the sea beyond the mouth of the river. (For information on the kinds and quantities of radioactive waste discharged to the marine environment, see Chapter 2.)

Waste discharges differ from worldwide fallout in a number of important characteristics:

Waste discharges are planned and controlled with respect to both time of disposal and amounts of materials.

Wastes are made up of relatively concentrated solutions and are discharged from point sources, usually into coastal waters.

A smaller spectrum of radionuclides is present in the wastes, and, except for fuel-processing plants, neutron activation products often predominate (at least qualitatively) over fission product radionuclides.

Other chemicals or heat present in the waste discharge may have a greater biological effect than the waste radionuclides.

The number of people exposed to measurable amounts of radiation as a consequence of waste discharge is a very small fraction of the total population exposed to worldwide fallout.

Radiation exposure of humans has been accurately assessed in relation to the discharge of radionuclides from several atomic energy installations, and the experience at these sites is summarized in the following paragraphs.

LARGE NUCLEAR INSTALLATIONS

Windscale An assessment of radiation exposure resulting from discharges of the U.K. Atomic Energy Authority factory at Windscale, Cumberland, was detailed earlier. The radionuclide in the effluent of this fuel-processing plant that most restricts the total discharge of radioactive waste is a fission product, ^{106}Ru. The critical pathway that limits the amount of ^{106}Ru discharged is the consumption by a small population in a distant area of contaminated seaweed (*Porphyra*) collected near Windscale (Figure 2).

Discharge rates of radionuclides are accurately recorded (U.K. Atomic Energy Authority, 1965, 1966, 1967), and regular assessments are made of radiation doses received by members of the public (Preston and Jefferies, 1967, 1969). Disposal rates in the years 1964–1966 of 2,000 Ci/mo of ^{106}Ru via the effluent pipeline resulted in intestinal doses, as a result of eating *Porphyra*, of 0.67, 0.61, and 0.57 rad/yr (i.e., about 40 percent of the recommended dose limit) in the three respective years. (Dose rates for other years are shown in Figure 1.)

Other routes of exposure and other radionuclides present in the discharged wastes result in much lower fractions of recommended dose limits for members of the public. For example, the average external gamma dose rate on mud flats near Windscale was 140 μR/hr during 1964–1967, and this resulted largely from the discharge of ^{95}Zr–^{95}Nb (Jefferies, 1968). Allowing for occupancy factors, this would result in a whole-body dose to some people (fishermen) of 10 percent of that recommended by the ICRP. These are, of course, not the same people who receive internal radiation from eating *Porphyra*.

Dounreay Limited amounts of radionuclides are discharged from the Dounreay Experimental Reactor Establishment on the coast of Scotland (Morgan, 1967). Because of the kinds of radionuclides of major significance in the discharge (^{141}Ce, ^{144}Ce, ^{103}Ru, ^{106}Ru, ^{95}Zr, and ^{95}Nb) and the absence of seaweed harvesting, it has been found that the major source of radiation exposure to man is the handling of fixed nets by fishermen in nearby bays.

Discharge rates from the offshore pipeline varied from 600 to 2,000 Ci (total radionuclides) a month in 1965–1966 (U.K. Atomic Energy Authority, 1966, 1967). Beta radiation dose rates measured at experimental nets during

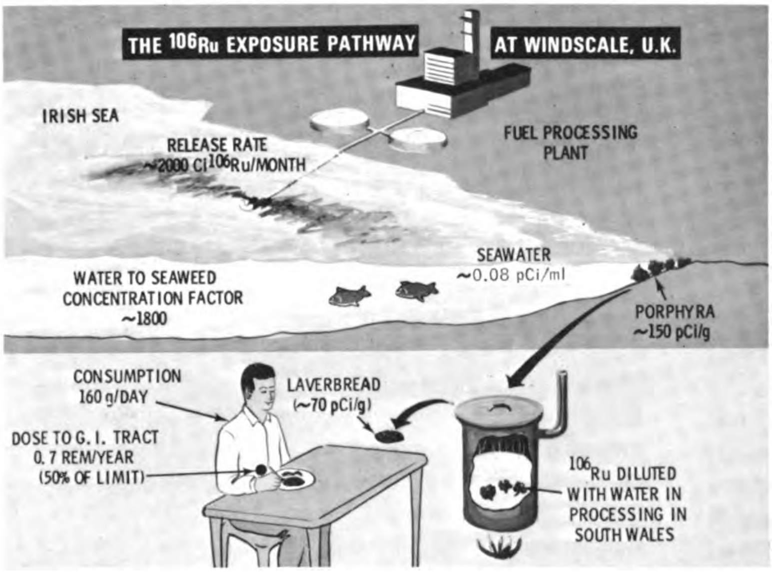

FIGURE 2 The critical exposure pathway associated with the U.K. atomic energy plant at Windscale is the release of ^{106}Ru to the Irish Sea, its accumulation by seaweed, and the consumption of the seaweed by man.

this period (Mitchell, 1967b) were less than 150 μR/hr (i.e., less than 2.5 percent of the acceptable dose rate when handling times for commercial fishing gear are considered).

Hanford Project The Hanford Project (now known as Richland Operations) discharges substantial quantities of radionuclides into the Columbia River. Some of these radionuclides reach the mouth of the river and enter the Pacific Ocean (Soldat and Essig, 1966). Of those radionuclides that reach the sea, only ^{65}Zn and ^{32}P warrant consideration as sources of exposure to humans in nearby communities. Oysters grown in a bay several miles north of the river mouth contain higher concentrations of these radionuclides than other common seafoods. Estimated human dose rates from an assumed consumption rate of 230 g of oysters per week are 5 mrem per year to the intestinal tract and 3 mrem per year to the whole body (Soldat and Essig, 1966). These represent 0.3 percent and 0.6 percent, respectively, of the acceptable dose rate to individual members of the public.

Other marine fish and shellfish in the vicinity also contain measurable concentrations of ^{65}Zn, but human exposure from these sources is much less than from oyster consumption.

Within the Columbia River, near the point of discharge of the effluent from the reactors, the uptake of ^{32}P by fish is the critical pathway. This is illustrated in Figure 3.

OTHER INSTALLATIONS

Radionuclides are discharged into the sea at a number of other research centers, including Trombay, India (Pillai and Ganguly, 1961); Studsvik, Sweden (Agnedal and Bergström, 1966); and Petten, Netherlands (Van Dam and Davids, 1966). Discharges at all these locations have been small in amount, and the critical pathways for human radiation exposure have been evaluated only as a preoperational exercise.

At Trombay, the critical radionuclide is ^{32}P, and the critical pathway is expected to be the eating of marine fish. At Studsvik, the composition of the discharged effluent is uncertain. Present indications are that external exposure from ^{60}Co on fishing gear will be the critical pathway for neutron activation products, and internal exposure from ^{106}Ru in fish will be the critical pathway for fission products. At Petten, the critical pathways are assumed to be fish and shellfish consumption. No critical radionuclides have been identified.

NUCLEAR POWER REACTORS

Bradwell The nuclear power station at Bradwell, England, discharges radionuclides into a river estuary. Here, the critical pathway is the consumption of contaminated oyster flesh, and the critical radionuclide in the effluent is ^{65}Zn (Preston, 1967) (Figure 4). Preoperational and follow-up assessments have been made (Table 3). The radiation dose received by the small critical population group is 0.17 percent of the acceptable dose rate for the total body and 0.08 percent of that for the intestinal tract (Preston, 1968).

Other U.K. Power Reactors At five other power reactor sites in the United Kingdom that began operation in 1968, preoperational assessments had been made of the critical pathways and critical radionuclides (Table 4). Actual discharges at Berkeley and Hinkley have been so low in relation to the maximum rate permissible for the site that only very small concentrations of radionuclides have been detected in the environment. At Berkeley, ^{137}Cs has been measured ex-

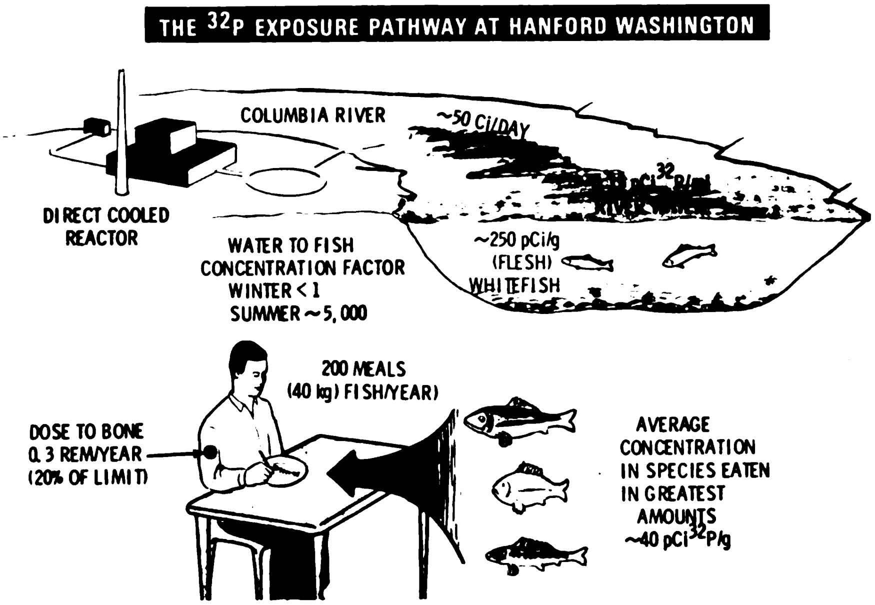

FIGURE 3 The critical exposure pathway associated with the Hanford plutonium-producing reactors is the release of ^{32}P to the Columbia River, its uptake by fish, and the consumption of the fish by man.

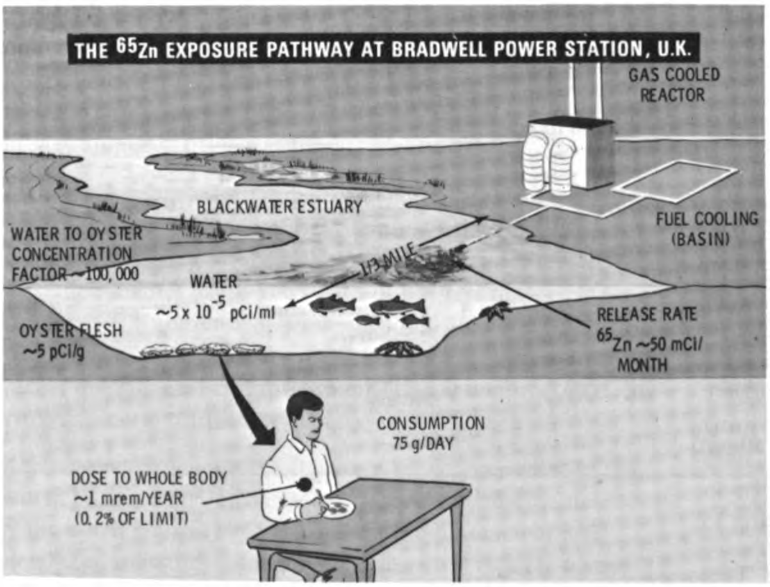

FIGURE 4 The critical exposure pathway associated with the nuclear power station at Bradwell, U.K., is the release of ^{65}Zn to the Blackwater Estuary, its uptake by oysters, and the consumption of the oysters by man.

tensively in silt (1-2 pCi/g dry) taken from the Severn Estuary, but at levels that have made no measurable contribution to gamma whole-body dose rates—the controlling feature of this type of contamination—and at Hinkley, ^{65}Zn has been detected in seaweed taken from the vicinity of the outfall (0.1-0.5 pCi/g wet). Two of the other three stations have only recently become operational. The critical pathway for human exposure at two of the sites is predicted to be external radiation from silt contaminated by activated corrosion products. At the three other sites, the critical pathway is irradiation of the human intestinal tract by neutron activation products concentrated in fish flesh (Preston, 1966).

Humboldt Bay The nuclear power station on Humboldt Bay in northern California began operating in 1962. The quantities of radionuclides discharged to the estuary each year with the cooling water have been substantially below the authorized limit of 20 Ci (0.1 pCi/ml). The nuclides of principal interest in the discharge are ^{65}Zn, ^{60}Co, ^{54}Mn, and ^{137}Cs. A preoperational survey and assessment indi-

TABLE 3 Permissible Discharge Rate of ^{65}Zn from the Bradwell Power Station to the Blackwater Estuary[a]

Preoperational Estimate		Follow-up Assessment	
Estimated concentration of ^{65}Zn in estuary per curie released per day	1.5×10^{-7} μCi/ml	Average ^{65}Zn discharge per month	16.6 mCi
Concentration factor–oyster flesh/seawater	100,000	Average ^{65}Zn concentration in oyster flesh at nearest commercial bed	3.4 pCi/g
		ICRP maximum permissible daily intake	0.22 μCi
Daily intake ^{65}Zn in 75 g of oysters	1.1 μCi	Maximum daily intake of oyster flesh	75 g/day
ICRP maximum permissible daily intake	0.22 μCi	Maximum permissible concentration of ^{65}Zn in oyster flesh	2,900 pCi/g
Estimated maximum daily discharge rate	0.2 Ci	Calculated maximum daily discharge rate	0.5 Ci

[a]Adapted from Preston (1967).

TABLE 4 Comparison of Critical Parameters for Five Civil Nuclear Power Station Sites[a]

	Central Electricity Generating Board Site				
Parameters	Berkeley	Hinkley	Dungeness	Sizewell	Wylfa
Site dilution factor	10	10^2	10^3	10	10^2
Water–μCi/ml/Ci of discharge/day	10^{-7}	10^{-8}	10^{-9}	10^{-7}	10^{-8}
Nuclides	^{65}Zn, ^{60}Co	^{65}Zn, ^{60}Co	^{65}Zn, ^{60}Co	^{65}Zn, ^{60}Co	^{60}Co, ^{65}Zn, ^{32}P
Material	Silt and salmon flesh	Silt and fish flesh	Fish flesh	Fish flesh	Fish and shellfish
Concentration factor	10^4	10^4	10^4	10^4	10^4
Population size	10	10	10	10	10
Critical organ	Total body	Total body	Gastrointestinal tract	Gastrointestinal tract	Bone, gastrointestinal tract
Ingestion rate (g/day)	23	195	807	130	205
Exposure (hr/yr)	340 (silt)	750 (silt)	–	–	–

[a]Adapted from Preston (1966, p. 735) and Mitchell (1967b).

cated that the critical exposure pathway would be via the accumulation of ^{65}Zn in oysters that are grown commercially in Humboldt Bay. Postoperational surveys have never shown levels of radionuclides in oysters of the Bay that were significantly above background. Under experimental conditions, oysters have been held directly in the plant discharge water, and at one time, their concentration of ^{65}Zn reached 175 pCi/g. The level has since declined (Salo, 1968). Inasmuch as the shellfish consumed by the public have only background amounts of the radionuclides, the radiation dose is negligible.

Tarapur A preoperational assessment of critical pathways and radionuclides has been carried out for the Indian power station at Tarapur (Kamath *et al.*, 1966). Neutron activation products (mainly ^{65}Zn) in marine fish consumed by fishermen and families are expected to contribute most to human exposure.

NUCLEAR-POWERED VESSELS

Studies have been made of the amounts and the fate of waste radionuclides discharged to coastal waters by U.S. Navy nuclear-powered submarines and ships at 13 U.S. docking facilities (Vaughan and Miles, 1966) and of conditions in harbors used by the civilian ship NS *Savannah* (Flora and Wukasch, 1966). (Discharges from the U.S. Naval ships are listed in Chapter 2.) No measurable transfer to man of the discharged radionuclides has occurred, and external radiation dose rates near the facilities have shown no measurable increase.

The accidental loss of the *Thresher* in 1964 was followed by extensive radiation monitoring in the vicinity of the hulk. No detectable escape of radionuclides into the sea was found, and no human exposure resulted.

SOLID-WASTE DISPOSAL

United States Sea disposal of contaminated solid wastes was carried out along both the Pacific and Atlantic coasts of the United States during the years 1946–1963. The total quantity disposed in the Pacific was approximately 15,000 Ci, and in the Atlantic, 46,000 Ci (Belter, 1965). Radiation monitoring of the two areas (Pneumo Dynamics Corporation, 1961; Brown *et al.*, 1962) has been carried out. No detectable concentrations of waste radionuclides were found in seawater or marine organisms.

United Kingdom Contaminated solid wastes containing approximately 1,500 Ci of beta-gamma radionuclides are disposed of yearly by the U.K. Atomic Energy Authority into the Atlantic deeps at a depth of not less than 1,500 fathoms. The amounts of waste disposed are considered to be too small to have an appreciable effect on the environment; therefore no radiation monitoring is undertaken (U.K. Atomic Energy Authority, 1966).

Asia Some radioactive wastes have been disposed of in the North Pacific near Japan. One of several measurements made by Akiyama (1965) suggested the presence of artificial contamination near the bottom in the vicinity of the disposal site. The measurement was only slightly above the expected background level, and one would not expect it to have any radiological implications.

CONCLUSIONS

Over the past decade, the use of atomic energy for peaceful purposes, and especially the production of electric power, has moved from the stage of planning and technical development to the stage of demonstration and competitive application. This application has been accompanied by the discharge of low-level radioactive waste into rivers and directly into the sea. In every case, the proposed discharge has received careful study in advance of operations, and prudent restrictions have been specified on the kinds and amounts of radioactive materials that could be released. Follow-up surveys show that the restrictions have been entirely adequate to keep human exposure well within the guidelines specified by the ICRP and FRC. A continuation of the policies and practices concerning the control of low-level waste disposal that have been established during these formative years should assure that radioactive contamination of the marine environment will not reach unacceptable levels.

SUMMARY

The addition of artificial radioactive materials to the marine environment results in some added radiation exposure to people who use the sea and its products. The magnitude of the additional exposure that results depends upon many complex relationships, however, and involves the kinds and quantities of radioactive materials added and the manner and place of their introduction.

Over the past quarter century, the major sources of artificial radioactive materials to the sea have been worldwide fallout from the testing of nuclear devices in the atmosphere and the chronic discharge of low-level wastes from operating reactors and fuel processing plants. Much less significant additions have resulted from nuclear detonations below the surface of the sea, from the disposal of low-level waste in packages, and from the inadvertent loss of radioactive materials.

Radioactive contaminants can follow a variety of pathways in the sea that may bring them into contact with man and contribute to the radiation exposure that he receives from the environment as a whole. Only careful evaluation of each individual situation can determine the most important or critical pathway, but the following ones are likely to be of greatest importance:

The accumulation of certain radionuclides in fish, shellfish, and seaweeds that are eaten by man in substantial quantities

The deposition of radioactive particles on the seabed in places where people may contact them either in the course of their occupations or in pursuit of recreation

The adsorption of certain radionuclides on fishing gear.

A large number of other pathways have been identified or postulated, but at this point in time their contribution to the radiation exposure of people has been much smaller than the ones listed above.

The significance of the radiation exposure contributed by all of the pathways is dependent on the intensity (concentration) of the source, on the frequency and length of time that people are exposed to the source, and on the number of people that may be exposed. It is generally held that any exposure to ionizing radiation entails at least some small risk of a deleterious biological effect. Therefore, the radiation dose must be held to such a low level that the very small risk involved is acceptable to society as a whole in relation to the benefits derived from the use of atomic energy. The Federal Radiation Council, the National Council on Radiation Protection and Measurements, the International Commission on Radiological Protection, and other authoritative groups have recommended doses that they believe should be considered as the maximum acceptable for continuous exposure. In all cases, they recommend restricting exposure to the lowest practicable level compatible with economic and social considerations. For radioactive materials deposited in the body, dose rates are related to a permissible body (or organ) burden for specific radionuclides, and these in turn are related to a permissible continuous daily intake sufficient to establish and maintain that body burden.

When an introduction of radioactive material into the marine environment is being considered, it is now conventional to identify the probable critical pathway of exposure and the individuals or critical population likely to receive the greatest exposure. Preoperational guides are then calculated that establish the maximum allowable rates of introduction of specific radionuclides. The preoperational guides are based on the critical exposure pathways and on some fraction of the internationally recognized dose limits. Following the actual introduction of potentially significant quantities of radioactive materials, a re-evaluation of the probable dose to man can and should be made on the basis of actual measurements of the concentrations of radionuclides in the environment. Follow-up evaluations should be based on measurements of the materials directly responsible for human exposure—the species of fish, shellfish, and seaweed actually consumed by the public, the beaches used by the public, and the gear handled by the fishermen. The follow-up evaluations have almost always shown that the preoperational predictions were highly pessimistic and thus that the tentative discharge guides were much more restrictive than actually necessary to maintain the radiation dose to people within the prescribed limits.

The radiation dose received by people from worldwide fallout via food from the sea is very small both in magnitude and in relation to that received from the terrestrial environment. Small population groups that depend on fish and other marine organisms could have received at most about 0.4 mrem per year to the whole body and 30 mrem per year to parts of the skeleton.

The large nuclear installations of the U.K. Atomic Energy Authority at Windscale on the Irish Sea and of the U.S. Atomic Energy Commission at Richland on the Columbia River discharge far greater quantities of radionuclides into the sea than do nuclear power reactors. The critical exposure pathway for the Windscale plant is the accumulation of ^{106}Ru by seaweed that is eaten in quantity by a small population group in a distant area. The resulting radiation dose amounts to about 40 percent of the recommended limit. The critical exposure pathway for the Richland plant (Hanford) is the concentration of ^{32}P and ^{65}Zn by fish in the Columbia River and oysters grown near the mouth of the river. The marine pathway leads to a dose to people who eat oysters of less than 1 percent of the recommended limit.

Discharges to the sea from power-producing reactors have resulted in doses to the public of much less than 1 percent of the recommended limit. At many sites, the concentrations of reactor-produced nuclides in the marine environment are so low that they can be measured only with sophisticated techniques. The radionuclides from power reactors that appear to be of greatest interest in relation to human exposure are the neutron activation products ^{65}Zn, ^{60}Co, and ^{32}P, because of their accumulation in marine organisms used for food.

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