FISH AND RADIOACTIVITY

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Introduction

With the advent of the atomic era there has been an increase in the number and in the quantity of radioisotopes that occur on the surface of the earth, both on land and in the sea. Naturally occurring radioisotopes always have been present in our environment; however, the detonation of nuclear devices and the operation of atomic reactors have added more and new radioisotopes to the environment. In this chapter, the consequences of adding radioisotopes to the aquatic environment will be discussed, especially in regard to the effects upon man of the uptake of radioisotopes by fish.

Radioisotopes in the sea can affect fish in two ways, first, as an external source of ionizing radiation as the fish swims about in radioactive water; and second, as an internal source of ionizing radiation from radioisotopes that have been taken into the body of the fish either directly from the water or indirectly through the food chain. Radioisotopes as an external source of radiation are called external emitters, but once within the

body they are known as internal emitters. The hazard from internal emitters is greater than for external emitters, and by a large factor, with the possible exception of the area in the immediate vicinity of and at the time of a nuclear explosion. Radioisotopes are more hazardous inside the body than outside because sources inside the body are in intimate contact with the body tissues, and they irradiate the body continuously until they are eliminated, which may be a very long time for some radioisotopes.

For practical purposes man's concern about radioisotopes in the sea and their effect upon fish is ultimately in the effect upon his welfare. There is considerable academic interest in the exposure of fish to radioactivity, as this information will contribute to the general knowledge of the biological effect of ionizing radiations on animals but there would be little concern if there were no effect upon man. A loss of a fisheries resource or the uptake of radioisotopes by man from fish are effects about which we are concerned. For example, if the numbers of fish that are killed result in a reduction in the harvest of the fishery, then this is an effect upon one of our resources and indirectly upon our welfare. Or, if fish as an item of food

are a means by which radioisotopes are transferred from the sea to man and the transfer results in a hazard to man's health, this is also an effect upon our welfare.

Of the two effects, the role of fish in transferring radioisotopes from the sea to man is of greater concern than the mortality that may occur to the fish. Whatever the contamination is in the sea, the transfer of radioisotopes from the sea to man will be of some concern, whereas the loss of fish would occur and be of concern only under unusual conditions of very great contamination. The condition that is most likely to be expected in the sea is that in which fish may acquire some radioisotope in an amount that is not lethal to the fish but when taken up by man by eating fish possibly could be concentrated to a level that is above the maximum permissible amount for man. Another reason that the indirect effect upon man is more important than the direct effect upon fish is that under conditions of equally heavy contamination of land and sea, man would succumb to radiation before fish, and therefore the mortality to fish would come too late to be of concern. Fish are less sensitive to external ionizing radiations than man, the lethal dose being about twice as great for

fish as for man. In addition, the radiation dose to fish in the sea would be less than the dose to man on land because of the greater dilution of the radioactive contaminants by the mixing processes of the sea.

For information on the lethal effect of radioisotopes in the sea upon fish, reference is made to observations at the Bikini-Eniwetok test site where the United States has tested its largest nuclear devices. The levels of radioactivity are considerably greater in the Bikini-Eniwetok area than elsewhere in the oceans, and therefore the effects would be expected to be more evident. Dead fish have been observed in the vicinity of the detonation of nuclear devices at Bikini-Eniwetok, and although the cause of death was not known for sure, it is reasonable to believe the cause was more likely to have been from blast effects or radiation released at the instant of the detonation than from the radioisotopes in water. In the absence of the effects of blast and heat, death to fish from ionizing radiations could be expected in the immediate vicinity of the detonation of a large device. However, if death resulted solely from radioactivity in the water, mortalities would be expected to occur over an extended period of time and area, but this condition has not been

observed. Although it is recognized that observations of dying fish may not be apparent because of the removal by predators of fish in a weakened condition, it is believed that the amount of radioactivity in water necessary to kill fish directly would have to be greater than the amount of radioactivity that has occurred in the water in the vicinity of Bikini-Eniwetok.

If fish survive exposure to ionizing radiations there still may be non-lethal effects in the form of pathological or genetic damage from either external or internal emitters. Again referring to the Bikini-Eniwetok area, thousands of fish have been examined for gross pathological and morphological changes but no obvious changes have been observed (Welander 1959). However, Gorbman and James (1959) found upon examining microscopically the thyroids of reef fish from an area close to a test site that the damage to the thyroid ranged from zero to 100 per cent. In those fish in which the thyroid was damaged, the fish upon superficial examination otherwise appeared to be normal. The cause of damage to the thyroids was undoubtedly radioisotopes of iodine, as internal emitters, that are present in relatively great abundance immediately after the detonation of a nuclear device. As the half life

of these radioisotopes of iodine is short (range from 2 hours to 8 days), the damage to the thyroids would be expected to occur soon after exposure.

Because the genetic effects of ionizing radiations occur in the progeny of the exposed individuals and may be subtle, it cannot be said that mutations have not occurred in the fish in the Bikini-Eniwetok area, although it can be said that there have been no recognizable mutations in the thousands of fish that have been observed. If mutations have occurred they are not the type that manifest themselves as morphological abnormalities. Laboratory experiments rather than field observations are needed to determine the genetic effects of low, chronic doses of ionizing radiation.

In the above discussion it has been pointed out that the indirect effect upon man of the uptake by fish of radioisotopes from the sea is of greater concern than the direct effect upon the fish themselves. Therefore, in the presentation to follow, principal consideration will be given to the role of fish in transferring radioisotopes from the sea to man. Because fish acquire radioisotopes either directly from the water, or indirectly by feeding

upon other radioactive organisms, fish as a food of man, like land plants or animals, can be a pathway by which radioisotopes are transferred from the environment to man. The presentation will include a discussion of the nature of radioactivity, the biological effects of ionizing radiations, the evaluation of hazard from internal emitters, the distribution of radioisotopes in the sea, and the uptake of radioisotopes by fish.

The Nature of Radioactivity

The biological effects to be expected from exposure to radioactivity can be explained, in part, by the nature of the energies that are released from the nucleus of an atom of a radioisotope. All atoms consist principally of a nucleus, in which there are protons and neutrons, and of electrons that orbit about the nucleus. Each chemical element is characterized by having a specific number of protons in the nucleus; however, the number of neutrons associated with any given number of protons may vary. Atoms with the same number of protons but with various numbers of neutrons are known as isotopes of the element. For example, in the nucleus of the iron atom there are 26 protons, but

there may be 26 to 34 neutrons, thereby forming 9 isotopes of iron. Certain ratios of neutrons to protons create an unstable condition in the nucleus. When in this condition the nucleus attempts to reach the stable state by the release of energy. Such an isotope is known as a radioisotope.

The energy is released from the nucleus as an alpha particle, a beta particle, a gamma photon or a neutron and each energy release is known as a disintegration. After one or sometimes more disintegrations, the nucleus reaches the stable state and is no longer radioactive. The rate at which a group of radioactive nuclei disintegrates varies from one radioisotope to another but occurs at an accurately predicted rate for any specific radioisotope. The time required for one-half of the nuclei in a group of atoms to disintegrate is known as the half life of the isotope.

The half lives of radioisotopes range from a fraction of a second to trillions of years. Almost all of the radioisotopes created by the fission process are short-lived. For the mixture of all fission products, radioactivity decreases tenfold for each sevenfold increase in time following the detonation in which the isotopes were

produced. At this rate the decrease in activity from one hour after to 343 hours after (approximately two weeks) is a thousandfold. The half life of some isotopes will be greater than, and of other isotopes, less than the average; therefore as time elapses, the rate of decline of the radioactivity of the mixture of fission products decreases. Radioisotopes cannot be destroyed by any means. Only by the natural process of decay can isotopes change from the radioactive to the stable state.

Radioactivity cannot be detected by the normal senses of sight, sound, touch or odor, either. However, nuclear radiations are capable of ionizing matter through which they pass, and as a result of this process radioactivity can be detected and measured by means of electronic devices. In the case of a nuclear radiation passing through a gas (or matter), the electrical field accompanying the passage of the alpha or beta particle or gamma photon dislodges an electron from the gas atom or molecule and thereby changes an electrically balanced atom or molecule into an ion pair -- a positive ion, the gas, and a negative ion, the electron. This process is known as ionization and is the most significant characteristic of nuclear radiations.

An instrument for measuring nuclear radiations designed on the principle of ion-pair formation consists of a gas-filled chamber with two electrodes, an electrical power supply to the electrodes and a meter for measuring electrical current. When an ionizing radiation enters the gas chamber at a time when a potential is applied to the electrodes, the ion-pairs that are formed will provide positive ions that will collect on the cathode and negative ions which collect on the anode and thereby create a pulse of current which will be indicated on the current meter. The amplitude of the pulse depends upon the number of ion-pairs produced and the applied voltage. Three common types of ionization instruments that are basically similar but differ in the applied voltage are the ionization chamber, the proportional counter and the Geiger-Müller counter.

Another type of instrument for measuring nuclear radiation is the scintillation counter. Certain materials -- phosphors -- when exposed to nuclear radiations will emit small flashes of light -- scintillations. The scintillations are received from the phosphor by the photocathode which responds by emitting electrons that are greatly increased by stages in the photomultiplier tube until a measurable

current pulse is produced. The pulses, recorded by a scaling unit, are a measure of the radioactivity of the sample.

Radioactivity is measured in terms of the number of disintegrations per unit of time. The disintegration rate of radium was arbitrarily selected as the standard. The unit is called a curie and is defined as the quantity of any radioactive materialhaving associated with it 3.7×10^{10} disintegrations per second (or 2.2×10^{12} disintegrations per minute). One gram of radium has an activity of one curie. Other radioisotopes disintegrate at different rates; therefore the number of curies per gram varies from isotope to isotope in proportion to the half life and the number of atoms per unit weight. One gram of strontium-90 (Sr^{90}) which has a half life of about 28 years has an activity of 147 curies. A radioisotope with a shorter half life and/or a lesser atomic weight, would have an even greater specific activity. It is important to note that the radioactivity per gram is large and that the amount of a radioisotope necessary to be of concern as a potential health hazard is essentially weightless. The amount is usually too small to be determined by gravimetric

methods and can be measured only by instruments that are designed to count nuclear radiations. For example, the amount of Sr^{90} that has fallen out of the stratosphere and onto the continental United States has been less than a pound and a half or approximately one gram per 5,000 square miles (calculated on the basis of 30 millicuries per square mile and 3×10^6 square miles in the continental United States). Sr^{90} from this pound and a half source can be found, by radiological methods, in milk, wheat, plant and bone samples collected throughout the nation, but the amount in any one sample is too small to be weighed even by the most sensitive balance.

In biological work, a curie is often too large an amount of radioactivity to be expressed simply, so the unit of radioactivity is often expressed as a fraction of a curie, e. g., a millicurie (mc), one one-thousandth of a curie; or a microcurie (μc), one one-millionth of a curie; or even a micro-microcurie (μμc), which is equal to 2.2 disintegrations per minute.

The curie is the rate at which energy is being released from nuclei of atoms regardless of whether the energy is being released as alpha or beta particles, gamma

photons, or neutrons. These forms of energy differ in their power of penetration and ability to ionize other materials, factors that are important in determining the biological effect of ionizing radiations upon tissue. To measure the energy absorbed by the tissue other terms are used.

The dose of radiation that is received by the tissue of an organism from any type of radioactive source is measured in terms of the energy absorbed per unit mass of the tissue. It is evident that the curie is not a measure of radiation dose. The unit of measurement of the radiation dose is the rad and is arbitrarily defined as the absorption of 100 ergs of energy per gram of tissue as measured in the tissue which is being irradiated. To give some idea of the size of this unit, 420,000 rads would raise the temperature of water by one degree centigrade, assuming that all the absorbed energy is converted to heat (Platzman 1959).

Another unit which has been used for many years to express the amount of radiation from X rays or gamma rays is the roentgen, which is a measure of the ionizations produced in air rather than of the energy absorbed in tissue. One roentgen produces about two billion each of positive and negative ions in one cubic centimeter of air at standard

conditions. Water or soft tissues exposed to one roentgen (r) receive an absorbed dose of very nearly one rad. As defined, the roentgen cannot be applied to radiations other than X rays or gamma rays.

To compare the effectiveness of absorbed doses of radiation delivered in different ways, the term relative biological effectiveness (RBE) is used. The unit of RBE is the rem. By definition, one rad of X rays, gamma rays or beta rays is equivalent to one rem but one rad of alpha rays is equivalent to about 10 rem. Dose in rad x RBE = dose in rem.

Biological Effects of Ionizing Radiations

The ionizations that occur when nuclear radiations pass through a gas or a phosphor make it possible for radioactivity to be detected and measured, and the same ionizations, when they occur in tissues of living organisms, are primarily responsible for the biological effects that occur as a result of irradiation. The answer to the question, "Why are radioisotopes harmful?", is that they emit ionizing radiations which are capable of injuring or destroying cells. However, precise information on the exact manner in which

the cell is affected is lacking but it is known that the primary site of the biological effects of radiation is in the cell.

The irradiation of single cells has resulted in a large variety of effects and has led to two concepts about the basic mechanism of radiation damage; one, that radiation striking a sensitive area of the cell has produced ion-pairs which react with neighboring molecules to form new substances that disturb the normal function of the cell; the other, that the ionizing event occurred in the water of the cell and produced highly oxidizing radicals and molecules, which in turn disturb the biochemistry of the cell.

The Report of the United Nations Committee on the Effects of Atomic Radiation (1958) also points out the lack of knowledge about the mechanism of radiation damage in the following statement.

> The effects of ionizing radiations on living matter are extremely complicated, and their exact mechanisms are still largely unknown. The initial disturbance is associated with ionization (and excitation) of molecules which lead to alterations in their properties. Many functions of the cell are thus affected

by radiation, and, although some specific effects may be caused by one or a few events in the cell, many are probably the combined results of numerous such events.

Even if the effect of ionizing radiations upon cells were known, it would not tell the entire story of the effect upon the whole animal. An organism such as a fish or a man is an organization of specialized and interdependent cells. As stated by Loutit (1959), "Radiation damage to one organ can disturb the functioning of another. The cooperative action of cells and tissues in a many-celled organism profoundly complicates the primary effects of radiation. Given time, this action can also mitigate or reverse some of the effects."

The biological effects of radiation depend upon the total dose received by the organism and the length of time in which the dose is administered. The effects range from death to physiological disturbances that are too slight to be observed. The damage that results from small doses is the kind of damage to be expected from the addition of radioisotopes to the sea.

Depending upon the type of cell absorbing the

radiation, the effects are classified as somatic or genetic. Somatic cells are those concerned with the maintenance and integrity of the individual and include cells in the bone marrow, blood, liver or nervous system; genetic cells are the reproductive cells of the gonads, which are responsible for the transfer of genetic information from generation to generation. Damage to somatic and genetic cells differs in that somatic effects are limited to the irradiated individual, whereas genetic effects may be passed on to the progeny of the irradiated individual.

The biological effects of radiation vary considerably between individuals of the same species but the difference between widely separated groups of organisms is even greater. In general the more complex the organism, the more vulnerable it is. The lethal dose, fifty per cent at thirty days (LD_{50}), is about 400 roentgens for man but is two to three times greater for fish. Values for other organisms are given in Figure 1 which is prepared from data reported by Donaldson and Foster (1957). Owing to the great variety of circumstances that existed in the experiments from which these data were obtained, the values in Figure 1 represent only orders of magnitude of effects.

Evaluation of Hazard from Internal Emitters

The use of the word "hazard" to denote radioactivity in a biological sample, regardless of kind or amount, is questionable and therefore is used here with reservation. "Hazard" implies a risk or danger, which may or may not be correct depending upon the point of view. One point of view is that all ionizing radiations are damaging and, therefore, a hazard exists even from one ionizing event. The other point of view is that there can be some repair of the damage caused by ionizing radiations, thus, if the radiation dose does not result in an observable change, a hazard does not exist. In other words, from this latter point of view, there is a threshold level of radiation below which there is no net effect.

A threshold level of ionizing radiation is more generally accepted for effects upon somatic tissue rather than upon genetic tissue. There is reliable evidence which demonstrates that the biological effect upon somatic tissues of two equal doses of radiation administered over unequal periods of time is less for the dose at the longer exposure. These results have been interpreted to mean that some repair has taken place and that for very low levels of exposure,

repair could be complete. For genetic tissue it is generally believed that there is no repair of damage once incurred, but in a recent experiment by Russell with mice, "...low dose rates of radiation turned out to be only one-fourth as effective in producing mutations as the same dose given at high dose rates." (Joint Committee on Atomic Energy 1959). At the present time experimental data on the biological effects from very low radiation doses are not available; therefore, it cannot be said positively that there is or is not a threshold dose.

Before considering the hazard from radioisotopes that have been added to our environment by man, consideration will be given to the ionizing radiations to which we are exposed in nature. Irradiation by natural sources is relatively constant in any one area but varies from area to area with local geological conditions. External emitters from natural sources include cosmic rays and the radioactive isotopes present in the crust of the earth and in the air. Cosmic rays account for about one quarter of the natural background radiations. Internal emitters include the radioisotopes K^{40} and C^{14}, which exist as a small percentage of these elements and are normal constituents of the body, and

other isotopes such as Ra^{226}, Th^{232} and their decay products.

A question commonly asked is what role have ionizing radiations from natural sources played in the evolutionary process. Radiation causes mutation; the mutant gene is the raw material of evolution; is radiation thereby an important factor in evolution? This line of reasoning also can be extended to inquiring if an increase in radiation in the environment from fallout may speed up the evolutionary process.

The answers to these questions are given by Crow (1959) who states that "...it is likely that ionizing radiation has played only a minor role in the recent evolutionary history of most organisms." For Drosophila the background radiation would have to be increased more than 10,000 times to account for the natural mutation rate. Crow also shows evidence that the reason for the slow rate of evolution in some groups is not from insufficient genetic variability. He observes also that the effects produced by mutations are of all sorts and are mostly harmful. The reason that mutations are mostly harmful is that genes duplicate complicated structures for which there may be only one or a few ways in which to build the structure correctly, but many ways in which to make it incorrectly.

Crow concludes by stating that "...ionizing radiation is probably not an important factor in animal and plant evolution. If it is important anywhere, it is probably in those species, such as man, that have a long life span, and at least for man it is harmful rather than a potentially beneficial factor."

Since the first nuclear explosion in 1945, radiation from artificial radioisotopes has been added to the environment but even today natural radioisotopes irradiate human beings far more intensely than man-made fallout (Arnold and Martell 1959). The estimated total radiation dose for an average resident of the United States for the first thirty years of life is 3 roentgens for background radiation, 3 roentgens for medical radiation to gonads, and 0.3 roentgens for fallout.

Recognizing that even low levels of ionizing radiations from fallout are potentially hazardous, questions arise such as, "At what levels of radioactivity in the environment should we take positive measures to reduce exposure to radiation?" The general answer is that society must make the decision as to what is or is not an acceptable risk. The reason is explained by Loutit (1959) to be as follows:

> Each advance in man's power over nature has brought with it an element of danger. Atomic energy is no exception. Consciously or unconsciously, we adopt a policy of acceptable risk in every facet of our lives. Society must decide what risk it will accept in the development of atomic energy; the scientist must make clear the potential gains and losses.

Balancing the risk against the reason for taking the risk is difficult because there is no common measure. How is the biological damage from ionizing radiations to be balanced with the economic or social gains from atomic energy, especially when some or all of the benefits may accrue to others than those taking the risk? The economic and social effects are called "bioeconomic" by Claus (1958), a term which is defined as the balance between using radiation to improve conditions of living and total health and using radiation in such a way that living conditions and total health deteriorate. Radiation effects are classified by Claus as somatic, the effect upon the exposed individual; genetic, the effect upon future generations; or bioeconomic, the effect on total well being.

food have been established upon the assumption that something less than a dose that produces no apparent damage is an acceptable risk. For example, if X amount of radiation produces no apparent damage, then 1/10 of X should be an acceptable risk. Maximum permissible dose is defined by the International Commission on Radiological Protection as "that dose accumulated over a long period of time or resulting from a single exposure, which in light of present knowledge carries a negligible probability of severe somatic or genetic injury."

The standards for body burden and for the concentration of radioisotopes in air, water and food that are used in this country are based upon recommendations made by the National Committee on Radiation Protection and Measurement and include recommendations by the International Commission on Radiological Protection, with whom the National Committee works closely. The recommendations have been published by the National Bureau of Standards, first in 1953 as Handbook 52 and later, in a revised and more complete version, in 1959 as Handbook 69. The title of Handbook 69 is, "Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air

and in Water for Occupational Exposure." The meaning of a maximum permissible value is explained by Dunham (1959) in the following statement:

> In a sense a maximum permissible value is similar to a speed limit. A speed equal to the speed limit is not an absolutely safe speed since many serious accidents occur at lower speeds. Nor is it extremely dangerous to drive at speeds somewhat greater than the speed limit. Neither a recommended maximum permissible dose nor a speed limit has any particular significance beyond marking the point at which an advisory group has agreed to draw the line in recommending a maximum degree of hazard appropriate under a given set of circumstances.

There are two types of maximum permissible values, dose and concentration. The maximum permissible dose is a limit for exposure to external emitters, and the recommended value is 0.3 rems per week. The maximum permissible concentration (MPC) is a limit for the concentration of radioisotopes in critical organs of the body. The MPC values for 240 radioisotopes are given in Handbook 69.

For fallout radioisotopes of biological interest, the MPC values are given in Table I.

The difficulty in estimating the maximum permissible concentration (MPC) is indicated by the number of factors which must be considered in calculating MPC values. There are ten factors which include, "quantities available," "initial body retention," "fraction going from blood to critical body tissue," "radiosensitivity of tissue," "size of critical organ," "essentiality of the critical organ to the proper function of the body," "biological half life," "radioactive half life," "energy of the radiation," and the "specific ionization and attenuation of energy in tissue." For lack of better information, some factors have been estimated with a large degree of uncertainty and, therefore, MPC values will need to be revised from time to time as more information becomes available. One of the most likely sources of error is in the extrapolation of the effects upon laboratory animals to the effects upon man, which is necessary though because the data on the biological effects of ionizing radiations on man are meager. The radioisotopes that present the greatest potential hazard are those for which there is a physiological need, which are abundant in a

biologically available form, are taken up by a critical tissue or organ of the body, are highly ionizing and have both a long physical and a long biological half life.

The MPC values for radioisotopes in drinking water, including water in foods, given in Handbook 69 assumes that there is no other source of contamination and that the daily water consumption is 2.2 liters (or kilograms) per day for a 50-year exposure period during which the level of radioactivity remains constant. The MPC values for drinking water can be used for fish or other foods when compensation is made for the difference between the calculated water intake and the actual food intake. If fish are the only source of radioisotopes in the diet and 0.22 kilogram (1/2 pound) of fish which is 1/10 of the water consumption upon which the MPC values were based is eaten every day for 50 years, the MPC values for fish would be obtained by multiplying the values given in Handbook 69 by a factor of 10. The assumption of one-half a pound of fish per day in the diet is nearly twenty times the average consumption per capita in the United States (Taylor 1951) but would be approximately the amount in the diet if fish were the only source of protein.

The permissible concentrations of radioisotopes in sea water -- not in drinking water -- were calculated by Carritt et al. in Publication 655 of the National Academy of Sciences - National Research Council (1959). The purpose of making the calculations was to establish a limit for the concentration of radioisotopes in sea water, so that the uptake of radioisotopes by fish living in the water would be less than the MPC value for fish. The permissible sea water concentration (PSC) for any one radioisotope was calculated by dividing the MPC value for fish by the factor by which the fish concentrated the isotope from sea water. . The ratio of water intake to fish intake that was used in the Carritt report was 10, the same as above. Although not stated in the report in this form, $PSC = \frac{(MPC)f}{\text{concentration factor}}$, where (MPC)f is the value for $(MPC)_w$ corrected for the difference between water and fish intake and is obtained by multiplying $(MPC)_w$ by the ratio of the calculated water intake (2.2 kilograms) to the actual fish intake. A partial permissible concentration value for seafood for radioisotopes to be expected as waste from the operation of nuclear-powered ships has been calculated by Revelle et al in

Publication 658 of the National Academy of Sciences - National Research Council (1959). The calculation is essentially as given above with the exception that it has been refined by taking into account the fraction of the maximum permissible dose alloted to nuclear-powered ships for various zone of the sea. If the ratio of water intake to food intake in the fish is the same as the concentration factor for the isotope from sea water the the PSC equals the $(MPC)_w$ as given in Handbook 69. The error in estimating PSC by this method is often great because of the limited information about the concentration of elements in the sea by fish.

The MPC values are not always an adequate estimate of hazard because the uptake of some radioisotopes is influenced by the presence of chemically similar isotopes. Strontium and calcium are chemically similar as are cesium and potassium. When calcium is abundant and in an available form, less Sr^{90} is taken up, and therefore a measure of only Sr^{90} is not a reliable estimate of hazard. For Sr^{90} a better estimate than the MPC value is the ratio of Sr^{90} to calcium. The ratio of one micro-microcurie of Sr^{90} to one gram of calcium is called a strontium unit, and for

the general population the permissible level is 100 strontium units; i.e., the amount of radioactivity from Sr^{90} in the diet should not exceed the ratio of 100 micro-microcuries (220 disintegrations per minute) of Sr^{90} for each gram of calcium. Often the strontium units that are determined for single items in the diet are used as the criteria for hazard, but more properly the strontium units should be determined from the Sr^{90} and calcium in the entire diet. Sr^{90} has been used here as an example only, because it will be shown later that very little Sr^{90} is found in marine organisms.

The MPC values for the radioisotopes listed in Tables I and VI vary by a large factor, but the range in values for the 240 isotopes in Handbook 69 is even greater, one million. This large range in MPC values is important to note because it is obvious that an evaluation of hazard cannot be based solely on the amount of radioactivity in the sample without chance of a great error. Proper evaluation of hazard depends upon the determination of the amount of radioactivity as well as the identification of the radioisotopes in the sample.

A concept of hazard is sometimes created, psycho-

logically, merely by the terms used for the units of measurement in describing the amount of radioactivity present. For example, to say that the amount of radioactivity from naturally occurring radioisotopes in the body of a 170-pound man is about one-half a million disintegrations per minute is more alarming to some people than to say that it is one-fourth of one one-millionth of a curie. As the units of measurement of radioactivity become better known, a better understanding of the hazard associated with radioisotopes can be expected.

The standards of radiation protection can be expected to change as more accurate information is acquired on the factors upon which the standards are based and as the opinions of those who have the responsibility for establishing the standards change with time. In evaluating hazard, Dunham (ibid) makes the following statements:

> The important considerations are that (1) we bring to bear on the problem all of the information that can be made available; (2) we take maximum advantage of the combined judgment of able and well-informed persons; and, (3) we, as a people, understand the general nature of any standards which may be recom-

mended and adopted.

Distribution of Radioisotopes in the Sea

The radioisotopes present in the sea are those that (1) occur naturally, (2) have been created by the detonation of atomic devices, or (3) have been disposed of as waste from the operation of atomic reactors or from research and clinical laboratories.

The naturally occurring radioisotopes in sea water are listed in Table II. These isotopes have been and will continue to be present in the ocean at the level of abundance given in the table, with the exception of C^{14} and H^{3}. Carbon-14 is formed naturally in the atmosphere by a two-stage process which involves the shattering of a nitrogen atom by a high-energy proton from space. Most C^{14} exists in the form of carbon dioxide for which the residence times are an average of five years in the atmosphere, five years in the surface layer of the ocean and 1,200 years in the deep waters. However, thermonuclear explosions also produce C^{14}. Since the detonation of the first thermonuclear device in 1952, a ton of C^{14} has been produced which equals about one per cent of the total

natural abundance. Because of the long half life of C^{14} (5,600 years) the one per cent increase of C^{14} from thermonuclear detonations is an amount that will be detectable in ocean water.

Tritium, H^3, is formed in nature by many processes but most often in a manner similar to C^{14}. Thermonuclear explosions also yield H^3 as a direct product of the fusion reaction. Since 1952 the total output of bomb-produced H^3 is estimated at 100 pounds, about five times the amount of natural H^3. Tritium, with a shorter half life (12.5 years) than C^{14}, does not travel as far as C^{14} before disintegrating but a significant amount is carried from the atmosphere as rain and eventually into the deep oceans before it decays. The addition of bomb-produced H^3 to the environment has obscured the picture of the natural distribution of H^3 and increased the amount of H^3 in the sea above the values given in Table II.

Natural radioisotopes in the sea come from the crust of the earth. More than a dozen long-lived isotopes have been found in our environment but K^{40}, Th^{232}, and U^{238} provide the bulk of natural radiation. Rocks such as granite are more radioactive than limestone or alkaline

basalts. Some of the radioisotopes are leached from the rocks by weathering and carried to sea. Their fate in the sea is described in the following statements by Arnold and Martell, ibid.

> Thorium washed into the ocean rapidly precipitates as insoluble compounds. Potassium remains in solution, but some of its ions become attached to particles of clay which fall to the bottom. Uranium precipitates more slowly than thorium. Its decay product, thorium-230, precipitates rapidly, but decays into radium, some of which dissolves back into the ocean.

The lesser amount of radioactivity in water than in rock and also the lesser exposure of organisms in water to cosmic rays result in a smaller environmental radiation dose to fish than to man. The radiation dose in terms of millirads per year has been calculated to be 207 for man living over granite rock at 10,000 feet elevation, 142 for man over granite rock at sea level, 75 for man over sedimentary rock at sea level, 64 for a large fish living near the surfact of the sea and 30 for a large fish living at a depth of 100 meters (Folsom and Harley 1957).

A great number of radioisotopes have been added to the oceans from the detonation of nuclear devices especially in the vicinity of Bikini and Eniwetok Atolls. The kinds of radioisotopes produced are determined largely by the type of detonation -- fission or fusion. Previous to 1952 nuclear detonations were exclusively of the fission type but since then there also have been detonations of the fusion and the fission-fusion-fission type. The greatest number of radioisotopes is produced by the detonation of a fission type device.

In the fission process the nuclei of atoms of U^{235} or Pu^{239} are split when struck by neutrons. In addition to the release of energy at the time of fission, the two parts of the original atom become isotopes of two new elements that are approximately one-half the weight of the original atom. The nuclei of the newly-formed isotopes are unstable usually because of an excess of neutrons, hence are radioactive. (Because the neutron-proton ratio in the nucleus of stable light elements is less than in heavy elements and, nearly all of the newly-formed isotopes retain the neutron-proton ratio of the original atom (U^{235} or Pu^{239}), there is generally an excess of neutrons in the nuclei of the newly-formed isotopes.)

There are about 200 isotopes of 35 elements that are created by fission, but many with half lives of seconds or minutes or hours decay rapidly and usually are not detected. An indication is given in Table I of the number of fission products to be expected in samples collected at various times after origin and the percentage contribution of individual fission products to the total activity of all the fission products of common origin. The importance of knowing the age of the fission products when attempting to identify the radioisotopes in a sample is apparent from inspection of the data in Table I. However, if the age is not known a reliable estimate often can be made from calculations of the ratios of certain observed radioisotopes. Since the ratios are constantly changing, a particular ratio will identify the age of the fission products, providing they are all of the same age and have not been differentially segregated by biological, physical or chemical processes.

In the fusion process the nuclei of two atoms are joined together to form the nucleus for one atom, just the opposite of the fission process. The nuclei of hydrogen atoms are joined to form the nucleus of a helium atom

(or a heavier isotope of hydrogen) and in the process energy is released as well as a vast number of neutrons. To accomplish fusion, temperatures of the order of a million degrees centigrade and heavy isotopes of hydrogen -- H^2, deuterium, or H^3, tritium -- are needed. The greatest amount of energy is produced when the ingredients are deuterium and tritium. Radioisotopes released to the environment from the detonation of a fusion device are tritium plus tritium and deuterium as debris and a varied assortment of induced radioisotopes. The induced radioisotopes were originally stable isotopes that captured a neutron released during the fusion process or were imparted energy upon being hit by a neutron. The kinds of radioisotopes produced by the detonation of a nuclear device depend upon the kinds of stable isotopes within range of the neutrons and the ability of the stable isotopes to capture a neutron, and therefore are not entirely predictable. The radioisotopes produced by the fusion process generally have a shorter half life and are less hazardous than fission products.

Following the development of the fission process and the fusion process, fission was used to provide the

high temperatures needed for fusion, but in turn, the high neutron flux from fusion was used to fission more material. This is the fission-fusion-fission process and is used in the largest of current nuclear devices. The radioisotopes released to the environment by this process include those that are created by both fission and fusion. Because the hazard per unit of energy released is less for the fusion process than for the fission process the term "clean device" or "clean bomb" has become associated with devices or bombs using the fusion process. Actually a "clean device" is one in which the ratio of fission to fusion is at a minimum, i.e., as little fission as possible in order to keep to a minimum the hazard from radioisotopes released to the environment.

A third source of radioisotopes in the sea is the waste disposed from the operation of nuclear reactors or from research and clinical laboratories. Little use is now being made of the oceans for disposal of radioactive wastes and, although the United States always may dispose of most of its waste by underground burial, other countries, by necessity, are expected to make use of the sea in the near future. Once the radioisotopes are deposited in the sea

they are the concern of all, thus it is essential to begin now to accumulate the information that will be needed to make wise recommendations about sea disposal of radioactive wastes.

In the United States something less than 6,000 curies of low level wastes were added to the water off the Atlantic coast between 1951 and 1958. The amount disposed of is a maximum estimate as it often was estimated from the quantity received by the user without correction for loss during use or for radioactive decay. The waste, which is contained in more than 8,400 drums of 55-gallon capacity to which concrete was added, has been deposited in more than seven localities in designated areas about 200 miles off the Atlantic coast in approximately 1,000 fathoms of water (NAS-NRC No. 655). It is doubtful if radioisotopes from these disposals can be detected in Atlantic coastal waters by conventional means.

A Committee of the National Academy of Sciences has recently considered the use of areas closer to shore than the present disposal sites for disposal of radioactive wastes. Carefully determined values for the concentration of radioisotopes in sea water have been established by the Committee (NAS-NRC No. 655) and it is their opinion that

certain coastal areas of the Atlantic Ocean and Gulf of Mexico can be used as receiving waters for the controlled disposal of packaged, low level, radioactive wastes under certain prescribed conditions.

One of the areas where radioactivity is being added continuously is the Irish Sea which receives low level liquid wastes from the British Atomic Energy Authority plant at Windscale.

Previous to the disposal of these liquid wastes by means of a pipe line to the shallow waters approximately one mile off the British coast, the problem had been studied in detail. The circulation of surface waters in the vicinity of the outflow was determined by a fluorescein dye dispersal method; the breeding and migratory habits of fish which were of commercial interest were studied for the purpose of estimating the uptake of radioisotopes; the uptake of radioiodie by seaweed, which is used in a porridge by local residents, was estimated; and the levels of radioactivity to which the people using the beaches would be exposed were calculated. When all of these factors were taken into consideration the discharge of 1,000 curies per month was authorized. As a result of monitoring studies

made during the build-up of the discharge level to 1,000 curies per month and a reassessment of the "safety factors" that were included in the original studies and recommendations, authorization has now been granted to discharge wastes at Windscale at the rate of 10,000 curies per month.

Another area where radioactivity is being added continuously is the Pacific Ocean at the mouth of the Columbia River which receives low level liquid wastes from the Hanford plutonium production reactors. The total amount of radioactivity entering the ocean at this point is about 1000 curies per day (U. S. Atomic Energy Commission, 1960).

The waters of the oceans cover 71 per cent of the earth's surface and the mean depth for all oceans is about 14,000 feet. The ocean can be conveniently divided into three domains: (1) the inshore area, including the intertidal zone, bays and harbors; (2) the shelf area, extending from the low tide line out to a depth of about 600 feet where the bottom slope increases rapidly towards the deep sea; and (3) the deep sea. The waters of the inshore area, the shelf and the uppermost part of the deep sea are thoroughly mixed by the winds and in the inshore areas by the tides. By contrast, the waters of the deep sea below the surface layer, which varies in depth but is usually about 200 feet thick, are stratified and slow moving. This means that both vertical and horizontal mixing is limited. The boundary between the surface and the deeper waters is identified by a rapid decrease in temperature with depth (thermocline) or a rapid increase in density with depth

(pycnocline). The deep sea comprises approximately 95 per cent of the ocean waters.

The principal current systems of the surface water in the northern hemisphere circulate in the ocean basins in a clockwise direction. In the southern hemisphere the circulation is counterclockwise. The waters move slowly, the average speed of the surface currents in the open ocean being one-half to one mile per hour, but the deeper currents move much more slowly. Below the relatively thin surface layer, often called the stirred layer, the waters are stratified into a series of layers of increasing density and slow movement, with little mixing between layers. The direction of flow of the deepest layers may be in counter direction to the flow of the surface current. With these conditions the exchange of deep water and surface water can be expected to be a slow process. At the present time the age of deep ocean water in the Atlantic is being determined by the C^{14} age-dating method. From the best information now at hand, the time required for the replacement of Atlantic bottom water with surface water is in the range of 200 to 500 years, but in the Pacific the time may be as great at 1,000 years (Schaefer 1958). An exception to this

statement are the areas of upwelling where surface water is being pushed away from a land mass by the wind and is being replaced by intermediate water. In an area of upwelling, vertical movement is still slow but may be one or two meters a day. Other areas of upwelling are found along current boundaries. Circulation of water in the deep ocean is also influenced by high submarine ridges which may be comparable in height to mountain ranges on land. The ridges restrict circulation and if circular in shape can create basins of isolated water. There is a need for more information on the circulation of the deep waters of the ocean in order that predictions of the fate of radioisotopes introduced into the sea may be made.

Distribution and biological uptake of fallout in the ocean are influenced by the size and solubility of the fallout particles which, in turn, are determined by the size of the detonation and the type of soils (or water) in the vicinity of the detonation.

A detonation may be classified, arbitrarily, as large or small, depending upon the height attained by the cloud formed by the detonation. A large detonation is one in which the cloud reaches the stratosphere and the

fallout is worldwide; in smaller detonations, the cloud remains in the troposphere and distribution of fallout is limited. The point of division between a large and a small detonation is about 200 KT (energy released equivalent to 200,000 tons of TNT). For all detonations there is a large amount of local fallout within a few hundred miles of ground zero, occurring within a day or two of the detonation. In the Bikini-Eniwetok area the radioactivity in the local fallout resides on particles of NaCl, CaO, $Ca(OH)_2$ and $CaCO_3$ made by the great heat of the fireball acting on the coral of the islands and the sea floor. For detonations over other types of soil, different compounds would be formed by the fireball and would occur in the fallout.

In the Atlantic Ocean fallout has been either tropospheric or stratospheric and measurable amounts are detectable if special techniques and sensitive counting methods are used. In the Pacific Ocean, local fallout in the vicinity of the test sites of the United States and the United Kingdom has contributed by far the greatest amount of radioactivity to the ocean. The movement of radioactivity from the U. S. test site at Bikini and Eniwetok Atolls has been estimated and the amount of radioactivity in the water has been measured by both United States and

Japanese scientists. The Russian ship "Vityaz" entered the area in 1958 during its 27th voyage (Kreps 1959) but radiological information about the voyage is not known at this time.

Following the Castle test series of March 1 to May 14, 1954 at Bikini-Eniwetok and during which time the Japanese fishing vessel, "Lucky Dragon," was caught in fallout of March 1, a survey was made in the Bikini area during the month of June by the Japanese vessel, "Shunkotsu Maru." Miyake et al (1956) reported that fallout was present in samples collected about 1,200 miles west of Bikini nearly four months after March 1, a rate of advance of about 10 miles per day. A second measurement of radioactivity in the ocean from the same test series was made by United States' scientists in March and April, 1955 (Harley 1956). About 400 days after March 1, 1955, fallout was detected in water samples from off the south coast of Japan, a distance estimated to be 3,300 miles from Bikini, a rate of advance of about 8 miles per day. Estimates made of the westward movement of radioactive waters from the area of Bikini-Eniwetok after the 1956 and 1958 test series, for periods of 43 days and 21 days res-

pectively, were approximately 7 miles per day (Seymour 1957 and Lowman 1960). The estimates, although similar, could be in error because the precise time or place of fallout into the ocean was not known, nor the center of the fallout area, nor the exact direction of the flow. Current movement as measured near Eniwetok Atoll in 1958 with a surface drogue was 17 miles per day. The rate of advance of radioisotopes in water would be expected to be slower than the surface current because of eddy diffusion and turbulence.

The amount of radioactivity in the water was also measured. For radioactivity produced in the Castle series, the maximum value reported by Miyake was 91,000 disintegrations per liter at a station 350 miles west of Bikini Atoll, 110 days after March 1, 1954; and the maximum reported by Harley was 570 d/m/l off the coast of Luzon, 2,600 miles from Bikini, about 400 days after March 1, 1954. For the 1956 test series (Redwing), the maximum value was 120,000 d/m/l (Donaldson et al. 1956). The sample was taken north of Bikini of fallout that was believed to be three weeks old. Maximum values per se are not meaningful unless related to the time and place of origin of the radioisotopes in the sample.

An idea of the amount and distribution of radioisotopes in the North Pacific Ocean at the present time -- a year and one half after the last test series at Bikini-Eniwetok -- can be gained by again referring to the report of Operation Troll by Harley. One year after the 1954 series a two-month survey was begun near Bikini and continued westward to the Philippine Islands, then northward to Japan. The course of the vessel between Bikini and the Philippines was southwest, northwest and again southwest, a course that three times cut across the westward flowing North Equatorial Current. From the Philippines the course was southward a short distance to Morotai, then northward to Japan. Analyses of the sea water and plankton samples indicated that the radioactivity was of low level and widespread. The radioactivity in water from other than naturally occurring radioisotopes ranged from 0 to 570 disintegrations per minute per liter and in plankton from 3 to 140 disintegrations per minute per gram of wet sample. At the station with the highest value twelve samples were taken between the surface and a depth of 653 meters and the average of the twelve samples was 190 d/m/l. By comparison, the radioactivity in sea water from naturally occurring K^{40} is 736 d/m/l. The low values were found to be east of Bikini Atoll at the beginning of the survey; at the outer edges of the North Equatorial Current, although this was difficult to

define; and near the end of the survey, off the coast of Japan. At the present time the radioisotopes may be distributed more thoroughly in the North Pacific circulation system than in 1954 but the levels of radioactivity would be generally comparable.

Recent determinations have been made of Sr^{90} in sea water from the western North Pacific and North Atlantic. For the Pacific, Miyake et al. (1960) reported 2 to 5 d/m/l for six samples collected in 1957, 1958 and 1959. The values for eleven samples collected in 1956 and 1957 in the Atlantic were about one-tenth the Pacific values and ranged from .06 to 0.3 d/m/l (Bowen and Sugihara 1958). Larger values in the Pacific were not surprising because of the contribution by local or close-in fallout from Bikini-Eniwetok. Measurement of Sr^{90} in waters of the eastern North Pacific have not been made; however, there has been time for the long-lived isotopes from local fallout at Bikini-Eniwetok to reach the eastern Pacific by ways of the current system. As a result of the surveys following the weapons tests at Bikini-Eniwetok in 1954, 1956 and 1958, it is known that the areas of contamination can be identified by measuring the radioactivity in the water or plankton but more easily in the plankton, and that the radioactivity moves westward from

Eniwetok with the North Equatorial Current at a rate of approximately 7 to 10 miles per day. Fallout areas in the ocean from particular detonations have not been identified as discrete areas of relatively high contamination within the ocean but rather as low levels of contamination spread over a wide area.

Radioisotopes in the ionic or colloidal form, if not taken up biologically, will move vertically at a slower rate than radioisotopes incorporated into particulate matter, because of gravitational force. An estimate of the movement of fallout materials through sea water was made at Eniwetok during the 1958 test series from samples of sea water taken 6, 28 and 48 hours after detonation. At six hours, the greatest amount of fallout as measured by the radioactivity of the water was at the surface and decreased rapidly to near zero at a depth of 200 feet; at 28 hours, the highest value was still at the surface, but there was only a moderate decrease down to 200 feet; at 48 hours, the values were low at the surface and down to 200 feet but increased rapidly from 200 feet to a maximum at 300 feet, which coincided with the thermocline (Lowman 1960). Thus, in 28 to 48 hours the largest fraction of the fallout materials had moved through the stirred or mixed water layer, the water above the thermo-

cline, at a rate of about 8 feet per hour. The distribution of radioactivity within the mixed layer was not homogeneous at the time of sampling. In three other surveys during 1956 and 1958 in which samples were collected up to six weeks after detonation, the radioactivity also was not distributed homogeneously in the mixed layer. However, one year after the 1954 test series, the radioactivity in the water above the thermocline was well mixed, from which it is concluded that the time required for fallout materials in the surface waters of the ocean to mix thoroughly is greater than six weeks and less than a year.

Below the thermocline, in the period from 28 to 48 hours after fallout, the radioactivity in the particulate form descended at more than 10 meters per hour, four times the rate of movement through the mixed water layer (Lowman 1960). The increased rate of movement is assumed to be the gravitational effect upon the heaviest fraction of the particulate matter.

The chemical and physical form of fallout materials as they enter the sea may change upon interaction with the salts and other materials in the sea. Fallout that enters the sea as particulate matter may go into solution, and material in the ionic form may change to the particulate

form by precipitation, co-precipitation with accompanying materials, or by adsorption to organic material or inorganic silts and clays. As in other solutions, precipitation will occur only when the solubility product of the least soluble compound of the element has been exceeded. Prediction of the physical state of fission products in sea water is difficult because most of them are elements that occur only rarely in the complex mixture of elements in the sea and little is known of their ionic activity.

Although water movement is the principal method of horizontal transport of fallout materials in the ocean and the physical and chemical forms of fallout are important to vertical transport, the uptake of radioisotopes by the biota temporarily removes some of the fallout material from the forces of gravity and also may be an important factor in the vertical transport of materials in the ionic form across water boundaries such as the thermocline and the stratified layers below the thermocline. Plankton and the organisms of the "deep scattering layer" commonly make diurnal migrations from deep water to near the surface. When fallout material first enters the water, plankton acquire radioisotopes in both the soluble and insoluble

form and evidently select the insoluble material somewhat indiscriminately as indicated by the fact that most of the fallout radioisotopes found in sea water are also found in plankton. Later, when the insoluble material is not available, only those isotopes for which there is an apparent physiological need are found in the plankton. Plankton concentrate radioisotopes from the sea, often by a factor of a thousand or more, and are good biological indicators of contamination. A sample of plankton would be the best source of information to find out quantitatively if radioisotopes are present in a particular area of the ocean.

The fallout radioisotopes in ocean water are often difficult to identify because of the great amount of salts present. Identification of the radioisotopes in a plankton sample is more precise than the analysis for water samples because the isotopes taken up by plankton are more abundant and the techniques of analysis are simpler. Where both plankton and water data are available for samples collected soon after fallout, the lists of isotopes are generally similar. The radioisotopes listed in Table III were determined for plankton samples collected soon after fallout but also are representative of the radioisotopes to be found in water from which the plankton samples were collected. The

radioisotopes in other plankton samples collected one to six weeks post shot, with fallout from more than one detonation, are given in Table V. In the two tables only those radioisotopes that were present in the amount of about one per cent or more of the total activity are listed. Undoubtedly other radioisotopes were present but the greater abundance of the isotopes listed overshadows the isotopes present in minute quantities.

Other information about radioisotopes in sea water is included in the discussion that follows on the uptake of radioisotopes by fish.

UPTAKE OF RADIOISOTOPES BY FISH

In the discussion of the uptake of radioisotopes by fish, principal consideration will be given to the isotopes that are most likely to be transferred to man. Maximum transfer from sea to man requires that the radioisotopes are in an available form and are elements of nutrition for fish and man. In Table IV certain elements present in sea water, sea foods and fish muscle and the concentration of the elements in fish muscle are given.

The elements selected for Table IV as being most likely to be transferred to man are the essential mineral

elements of nutrition, as well as the elements always present in man but of unknown function (Heinz Company 1958). These are not the only elements found in man, however, as at times barium, boron, cesium, lead, lithium, rubidium, strontium, tin and titanium also may be present. Some of these elements may be of importance to the well-being of the individual but the presence of most of them is probably fortuitous. Also listed in Table IV as part C are elements with radioisotopes that occur in fallout, but the elements of fallout -- such as zirconium, niobium, ruthenium, rhodium, praseodymium and promethium -- for which there is little or no data concerning their abundance in sea water or in fish muscle are excluded. Also excluded from Table IV, because of the uncertainty of their occurrence in sea water, are the radioisotopes that might be present from the operation of nuclear-powered ships or from the disposal of radioactive wastes. Isotopes that may be discharged to the sea from the operation of nuclear-powered ships in addition to those listed in Table IV are Cr^{51}, Cu^{64}, Hf^{175} and Ta^{182} (NAS-NRC 58). The radioisotopes that would be expected to be found in the sea from the disposal of radioactive waste and their maximum permissible concentration factors in fish are

given in NAS-NRC Publication 655.

The value of the information in Table IV is limited by the amount and the reliability of the data. The lack of information on the presence of elements of fission products, in a compilation by Vinogradov (1953) of data on the chemical composition of fish, is a good indication that little work has been done on these elements. Fukai and Meinke (1959) in a more recent publication, have reviewed the literature for data relative to the occurrence of trace elements in sea water and marine organisms including the soft parts of fishes. The information about trace elements in fish was either meager, of questionable reliability, or nonexistent. For marine plankton, valuable information about trace elements in ten species of marine zooplankton, based upon spectrographic analyses, has been published by Nicholls, Curl and Bowen (1959). From these beginning studies by Nicholls et al., it now appears that, "...for any given chemical element there will eventually be found at least one plankton species capable of spectacularly concentrating it." This is of significance to the consideration of trace elements in fish because many plankton organisms are preyed upon by fishes and therefore fish may have available to them trace elements in a concentrated

form.

The probable reason that the elements of fission products are not found in greater abundance in fish is that they are not physiologically important and, if present at all, are present in quantities that are difficult to detect by chemical methods. As the method of radioactivation analysis becomes more common, additional information on trace elements may be expected.

Further inspection of Table IV shows that the elements of the fallout radioisotopes are scarce in sea water -- 5 parts per hundred million or less, with the exception of strontium; also, that the quantitative uptake by fish of these elements is not great as compared to the uptake of the other mineral elements of nutrition. However, the fact that the amount of these elements in sea water is even less than in fish results in large concentration factors which are not indicative of the amount of a radioisotope that will be taken up (or of the hazard in terms of maximum permissible concentration) but only of the fact that there is an apparent physiological need for the element. It would be expected that radioisotopes of elements with the largest concentration factors would be taken up most readily from the sea.

The radioisotopes found in fish, clam, plankton and algae samples are given in Table V. The samples were collected in the vicinity of the test site at Bikini and Eniwetok Atolls within a month or two after the test series and were selected for their high level of radioactivity. Within the limits of the available data, the radioisotopes of elements with high concentration factors in Table IV were the radioisotopes taken up by fish as shown in Table V. Practically all of the radioisotopes found in fish were non-fission products -- those isotopes listed in Table V to the left of Sr^{89}. Either the fission products are not available to fish or fish do not have a physiological need for the elements of fission products. Also to be noted in Table V is that the amount of radioactivity in plankton is greater than in fish and that the species of isotopes are somewhat different in the two groups of organisms, although the samples are not strictly comparable in time and place. Contrary to some opinions, this observation indicates that although marine organisms concentrate certain radioisotopes, the concentration will not increase necessarily as the isotopes are transferred upward through the food chain.

To determine the hazard to man from fish and clam samples with high levels of radioactivity, the ratio of the

amount of radioactivity in the sample to the allowable amount was calculated. the samples were selected from collections that were made in the Bikini-Eniwetok area during or a few weeks after a test series and at a time when the levels of radioactivity in the environment were at a maximum (Table V). The allowable daily intake was calculated from the value for $(MPC)_w$ in Handbook 69. The ratio of the amount of radioactivity from each isotope in the sample to the allowable amount for that isotope was calculated; the value for the sample was the sum of the values for the individual isotopes. The values given in Handbook 69 are for various tissues and for occupational exposures of either 40 or 168 hours per week. In this and following calculations the values for the most sensitive tissue and for the 168-hour exposure have been used. Often a value of 1/10 that for the occupational exposure is used in the case of the population at large.

The results of the calculations are presented in Table VI. For "total" values less than one -- flying-fish muscle, flying-fish liver, bonito liver -- the radioactivity in the sample is less than the maximum permissible amount as determined by the National Committee on Radiation Protection. For the clam kidney, a value greater than one, the result is interpreted as meaning that it would not be safe to eat 22 grams of clam kidney with this amount of radioactivity every day for 50 years. However, the interpretation is not realistic because it is assumed in the calculations that there is a constant level of radioactivity,

at first, slower later, but always decreasing. Although it would be impractical to calculate MPC values on a basis other than a constant exposure level, the fact that the exposure level usually is changing needs to be considered in evaluating hazard from MPC values.

The specific radioactivity of the ocean waters decreases with distance from the Bikini-Eniwetok test site because of decay of the isotopes and dilution within the ocean, but because plankton and fish concentrate certain elements from the sea, radioactivity was detected in some fish caught by Japanese fishermen in waters to the westward of Bikini-Eniwetok Atolls. During the 1954 survey by the Japanese research ship, "Shunkotsu Maru," in the vicinity of the test site, radiological analyses were made of both fish and water samples. Also during the summer of 1954 radiological analyses were made of tuna and other fishes caught by Japanese fishermen during regular fishing operations in the central and western Pacific Ocean.

A radiological survey of fish at the ports of landing in Japan begain in March and continued until November. When the Japanese fishing vessel, "Fortunate Dragon," which had been caught in the fallout of March 1, 1954 from a test at Bikini Atoll, returned to Japan and it was found that

the fish aboard were radioactive, as well as fish caught by other vessels fishing in nearby areas, there was a serious disruption in the tuna fishing industry. The sale of tuna for the fresh fish market stopped, as well as the sale for export. As a consequence the Japanese arbitrarily established an acceptable level of radiation for fish and the sale of tuna resumed but at a depressed price for some time. A conference between American and Japanese scientists also helped to re-establish the tuna market. The acceptable level was arbitrarily set as a value less than 100 counts per minute as measured with an end window G-M counter placed 10 cm from the surface of the fish. Fish of higher counts were discarded and usually buried.

During the survey period 71,179 tons of fish from 2,152 boats were inspected at five designated ports. Of these boats 11 per cent had their catch discarded and a total of 358 tons or 0.5 per cent of the fish inspected were declared radioactive. The amount of radioactivity in the discarded fish was as follows: 64 per cent between 100 and 500 cpm; 19 per cent between 500 and 1,000 cpm; 14 per cent between 1,000 and 3,000 cpm; 1.8 per cent between 3,000 and 5,000 cpm; and 0.8 per cent over 5,000 cpm (Kawabata 1956a). The use of a field survey-type meter to monitor

the radioactivity of whole fish does not tell what isotopes are present or whether the radioactivity is on the outside surface of the fish or absorbed within the tissues, either edible or non-edible. Without this information there is some question as to the necessity for discarding the fish.

At the time the fish were being monitored at dock side, determinations of the gross beta activity of fish tissues were made of a limited number of samples in laboratories. Values as high as 48,000 cpm were obtained from a sample of skipjack liver in June 1954 (Kawabata 1956b) but the isotopes present were not identified. Three months later the count was one-sixth of the June value. In the same fish the radioactivity of the muscle was 160 to 180 cpm per gram, the highest value found for muscle. Later, the principal isotope in the muscle of another tuna, an albacore, was identified as Zn^{65}; other radioisotopes were present but not identified. Since 1954 some of the results of the analyses of fish for specific radioisotopes by Japanese scientists have been published in English (Hiyama 1957); these are summarized in Table VII. Other results have been published in Japanese but are soon to be published in English as part of a report on radioactivity in marine organisms which is being prepared by the United Nations

Scientific Committee on the Effects of Atomic Radiations (Hiyama 1960). The values in Table VII probably are not from random samples, as it was stated for part of the sample -- the 1954 yellow fin tuna -- that it was the most intensively contaminated individual in a lot of about 100. The values in Table VII appear to be well below the allowable daily intake, with the possible exception of the kidney of the big eye tuna caught in June 1956.

The distribution of Sr^{90} and Cs^{137} in the sea deserves special comment because these are the two fission products that are of greatest public concern. They are produced in substantial quantities (about 1-1/2 times as much Cs^{137} as Sr^{90}), have long half lives that are approximately the same (28 years), and are metabolized to some degree by man. However, in marine organisms these two radioisotopes are either absent or present in only very small amounts (Table V and Table VII). Some strontium is found in the bone of fishes but at much lower levels than in bones of terrestrial animals. The uptake of radioisotopes from the sea is inversely related to the quantity of available stable isotopes of the same element, or of

chemically similar elements. The abundance in sea water of strontium and calcium, which is chemically similar to strontium, is part of the reason that Sr^{90} is not abundant in marine organisms (Table IV). Another factor is the low specific activity for Sr^{90} in sea water due to mixing and dilution by ocean waters. The low level of Cs^{137} in marine organisms cannot be accounted for entirely for the same reasons, because the stable isotopes of cesium are scarce in the sea. However, potassium, an element chemically similar to cesium, is abundant (Table IV) and also, the specific activity for Cs^{137} in sea water is low.

The Sr^{90} in canned tuna fish and salmon has been determined by the Health and Safety Laboratory of the U.S. Atomic Energy Commission. In 48 analyses of samples received between April 20, 1956 and August 8, 1957, the highest value was ,.004 μμc/g, wet weight (U. S. Atomic Energy Commission report HASL 42 1958). Because the values were very low and there seemed to be no trend in time or location, the sampling was discontinued temporarily. The health hazard from Sr^{90} which is determined from the ratio of Sr^{90} to calcium is low for fish because

of the low uptake of Sr^{90} and normal values for calcium. For conditions of equal fallout, the Sr^{90}-calcium ratio is a good deal less for fish than for such calcium-rich terrestrial food products as milk (Schaefer 1957).

The principal radioisotopes found in fish are non-fission products, of which Zn^{65} appears at this time to present the greatest potential hazard (Table V). In samples from the western Pacific and from the United States, Zn^{65} was identified in tuna muscle (Kawabata 1956b; Hiyama 1957) and has been found in trace quantities in foods analyzed by Murthy et al. (1959). The values reported by Murthy et al. were higher for oysters and clams than for land crops and, in terms of μμc/kg were as follows: Chesapeake Bay oysters, January 1959, 178; Chesapeake Bay oysters, March 1958, 124; and East Coast hard-shelled clams, May 1958, 40. The MPC for Zn^{65}, 10^6 μμc/kg, as calculated from Handbook 69, is considerably greater than the present values for oysters and clams. The concentration of Zn^{65} was not unexpected as Chipman (1959) found that oysters, clams and scallops concentrate large amounts of zinc, thousands of times above its level in sea water.

Because the local fallout in the vicinity of the

Bikini-Eniwetok test site has provided a variety of radioisotopes that become biological tracers for field experiments in mineral metabolism, valuable information has been acquired on the distribution of many elements in the marine biota. However, to understand the transfer processes and the biological effects of radiation, experiments under controlled laboratory conditions are required. Conversely, results of laboratory experiments that are to be extrapolated to the natural environment require field testing.

Many of the observations by the staffs of the Laboratory of Radiation Biology, University of Washington, Scripps Institution of Oceanography, and the Naval Radiological Defense Laboratory on the uptake of radioisotopes by marine organisms in the natural environment have been verified and supplemented by the laboratory experiments of the Radiobiological Investigations of the Bureau of Commercial Fisheries and the Hawaii Marine Laboratory, University of Hawaii. For example, Lowman (1956) and Schaefer (1958) have reported from field observations and Chipman (1959) from laboratory experiments that zooplankton rapidly accumulate radioactive particles. Chipman states that when the radioactive particles are no longer available, the zooplankton soon lose their radioactivity. Oysters,

clams and scallops like zooplankton readily accumulate radionuclides in the form of particles but many of the radionuclides that occur in the digestive tract are not absorbed. In laboratory experiments, strontium radioisotopes are taken up and deposited in the shells of oysters, clams and scallops but not in the soft tissues (Chipman 1959). In both the field and the laboratory, it was found that marine fish absorb few of the fission products from the digestive tract, and do not concentrate strontium radioisotopes in muscle, but do absorb Zn^{65} very rapidly. Although little Cs^{137} has been found in the marine organisms from the Bikini-Eniwetok area, Cs^{137} has been taken up and concentrated in the muscle by fish and shellfish in laboratory experiments, and therefore is regarded as the fission product with the greatest potential hazard. The conditions that make for a higher concentration of Cs^{137} in the laboratory than in the field are not known. In the field the concentration of cesium in four types of plankton is less than in sea water, as determined by Ketchum and Bowen (1958), which indicates that there is not a great demand for cesium at least by some types of plankton. Lastly, in both the field and laboratory observations it

was found that zinc and cobalt are taken up rapidly and in high concentrations by plankton (Table V; Chipman 1959).

SUMMARY

The atomic era has added new radioisotopes to our environment. For the radioisotopes in the sea, the effect upon man as a health hazard is potentially greater than the effect upon the fish and shellfish that take up the radioisotopes.

Radioisotopes emit ionizing radiations that are biologically destructive. The primary site of damage is the cell, but the exact manner in which the cell is affected is not known. The relationship of radiation dose to biological effect is well known for large doses but not for small doese of the size that now occur in the ocean. A maximum permissible dose -- a dose for which there is a negligible probability of severe somatic or genetic injury -- has been established and is used to calculate the maximum permissible concentration of radioisotopes in air and water (food). Because the range in MPC values for various radioisotopes may be as great as one million, an evaluation of the hazard requires that the radioisotopes in the sample be known as well as the amount of radioactivity.

The radioisotopes in the ocean are those that occur from natural sources, from fallout, or from disposal of radioactive waste. The radioactivity in sea water from naturally occurring radioisotopes is about 750 disintegrations per minute per liter, of which K^{40} contributes ninety-seven per cent of the activity. The greatest amount of radioactivity added to the ocean has been in the area of local fallout near Bikini-Eniwetok Atolls. The amount of radioactivity from fallout in a liter of sea water, immediately following a detonation may be thousands of times greater than the amount from K^{40}; within one year, however, the amount is less than from K^{40}. From the Bikini-Eniwetok area, fallout is carried westward by the North Equatorial Current system at a rate of seven to ten miles per day.

Radioactive wastes may be the principal source of radioisotopes in the ocean in the near future but at the present time the contribution from this source has been negligible.

Biological effects from radioisotopes in the ocean should be especially evident in the Bikini-Eniwetok area; however, gross population changes or morphological abnormalities have not been observed in samples from the area,

although damage to the thyroid did occur in some fish from a reef close to a test site. Subtle genetic effects that would not be obvious from field observations also may have occurred.

The two fallout isotopes of greatest public concern, Sr^{90} and Cs^{137}, are not taken up by marine organisms to any great extent. Practically all radioisotopes found in fish are isotopes of the non-fission product elements -- zinc, iron, cobalt, and manganese. At the present time the man-made radioisotopes can be detected with sensitive instruments in many of the waters of the world, but their concentration in aquatic organisms is well below the MPC values.

Exposure to all ionizing radiations is to be avoided. The amount of risk, above that from exposure to natural sources of radiation, which will be acceptable in the development and use of atomic energy is a decision that society must make, based upon the combined judgment of all able and well-informed persons.

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Relative sensitivity of various organisms

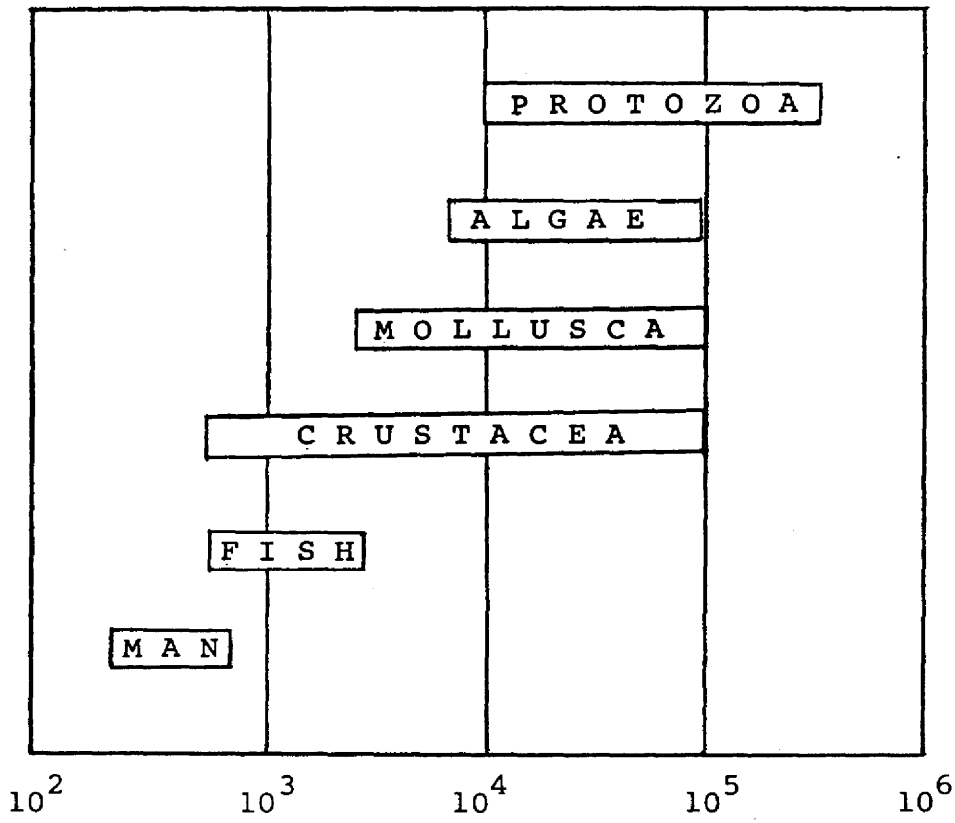

Fig. 1. X-rays or gamma rays required to kill 50 per cent of organisms. (Data from Donaldson and Foster, 1957).

Table 1. Per Cent Abundance and Relative Maximum Permissible Concentration of Fallout Radioisotopes of Biological Interest

Fission products	Per cent abundance at:[1] One week	One month	One year	Ten years	Half life[2]	Relative MPC values[3]
Sr^{89}	2.1	6.7	2.7	---	50 days	100
Sr^{90}	---	---	1.8	21.8	28 years	1
Y^{90}	---	---	1.8	21.8	64 hours	200
Y^{91}	2.4	7.6	3.9	---	58 days	300
Zr^{95}	2.5	8.2	7.3	---	65 days	600
Nb^{95}	---	4.1	15.0	---	35 days	1,000
Ru^{106}	---	---	2.4	---	365 days	100
Rh^{106}	---	---	2.4	---	2 hours	*
I^{131}	6.3	3.7	---	---	8 days	20
Cs^{137}	---	---	1.5	18.2	27 years	200
Ba^{137m}	---	---	1.5	18.2	3 mins.	*
Ba^{140}	8.8	10.8	---	---	13 days	300
La^{140}	9.4	12.5	---	---	40 hours	200
Ce^{144}	---	2.0	26.5	---	285 days	100
Pr^{144}	---	2.0	26.5	---	17 mins.	*
Pm^{147}	---	---	5.7	15.8	950 days	2,000
Non-fission products						
Mn^{54}					290 days	1,000
Fe^{55}					950 days	8,000
Fe^{59}					45 days	600
Co^{57}					270 days	5,000
Co^{58}					71 days	1,000
Co^{60}					1900 days	500
Zn^{65}					245 days	1,000

1) Hunter and Ballou (1951)
2) Strominger, Hollander and Seaborg (1958)
3) MPC values relative to Sr^{90}; based upon values from Handbook 69 for which MPC for Sr^{90} is 10^{-6} $\mu c/cc$
* A short-lived daughter for which hazard is negligible in comparison to hazard for parent

Table II. The Naturally Occurring Radioisotopes in Sea Water

Isotope	Total activity in ocean in megacuries[1]	Per cent of total activity	Radioactivity in sea water d/m/l[2]	Radioactivity in sea water μc/cc[3]	$(MPC)_w$[4] μc/cc
K^{40}	460,000	97.1	736	3×10^{-7}	*
Rb^{87}	8,400	1.77	13	6×10^{-9}	10^{-3}
U^{238}	3,800	.80	6	3×10^{-9}	2×10^{-4}
Ra^{226}	1,100	.23	2	10^{-9}	10^{-7}
C^{14}	270	.057	1	5×10^{-10}	8×10^{-3}
U^{235}	110	.023	1	"	3×10^{-4}
H^3	12	.003	1	"	.03
Th^{232}	8	.002	1	"	2×10^{-5}
Total	473,700	100.0	758	3×10^{-7}	

1) Revelle (1957)
2) Disintegrations per minute per liter based upon calculated value for K^{40}
3) Microcuries per cubic centimeter
4) Maximum permissible concentration for water consumed by man (Handbook 69, 1959)
* No value given; hazard negligible

Table III. Radioisotopes Present in Plankton Collected soon after Fallout

Isotope	Half life	Per cent of total radioactivity
Bomb debris		
Np^{239}	2.3 days	69
U^{237}	6.8 days	3
Fission products		
Mo^{99}-Tc^{99m}	66 hours	12
Te^{132}-I^{132}	77 hours	8
Ru^{103}-Rh^{103}	40 days	3
Ru^{105}-Rh^{105}	36 hours[2]	
Ru^{106}-Rh^{106}	1 year	
Ba^{140}-La^{140}	12.8 days	2
Ce^{141}-Pr^{141}	33 days	2
Zr^{95}-Nb^{95}	65 days	1
Ce^{144}-Pr^{144}	285 days	<1
		100+

1) Data from Lowman (1960); average value for six samples collected less than 48 hours post shot; fallout from one detonation; Rehoboth survey (1958)
2) Half life of daughter

Table IV. Mineral Elements of Nutrition and Other Elements of Interest and Their Occurrence in Sea Water and in Sea Foods

	Sea water[1] PPM	Sea foods,[2] edible portions PPM	Fish muscle[3] or soft parts PPM	Concentration in fish muscle, median value	Fallout radio-isotopes
A. Mineral elements of nutrition (man)[2]					
Chlorine	18,980		300 - 2,410	<1	
Sodium	10,561	480 - 10,000	300 - 1,680	<1	
Magnesium	1,272		100 - 4,520	2	
Sulfur	884		1,320 - 2,340	2	
Calcium	400	80 - 3,860	100 - 3,036	4	
Potassium	380	1,100 - 5,600	1,900 - 5,160	9	
Bromine	65		?	---	
Fluorine	1.4		4 - 34	14	
Iodine	.05		<1 - 27	270	$I^{131, 132}$
Phosphorous	.001 -.10	1,000 - 8,910	680 - 4,600	52,000	
Iron	.002 -.02	4 - 70	<1 - 56	2,500	$Fe^{55, 59}$
Copper	.001 -.01	<1 - 37	<1 - 37	3,700	
Manganese	.001 -.01		<1 - 252	25,000	$Mn^{54, 56}$
Zinc	.005		2 - 47	4,900	Zn^{65}
Cobalt	?		<1 - 1+	3,200*	$Co^{57, 58, 60}$

Table IV. - (continued)

	Sea water[1)] PPM	Sea foods, edible portions[2)] PPM	Fish muscle or soft parts[3)] PPM	Concentration in fish muscle, median value	Fallout radio-isotopes
B. Elements always present in man but of unknown function[2)]					
Silicon	.02 - 4.0		<1 - 40	10	
Aluminum	.5		<1 - 40	180	
Arsenic	.015		<1 - 12	490	
Nickel	.0001		?	---	
C. Other elements with isotopes of radiological interest					
Strontium	13.		+	< 1**	$Sr^{89, 90}$
Cesium	.002		?	35+**	Cs^{137}
Cerium	.0004		?		$Ce^{141, 144}$

1) Sverdrup et al. (1946)
2) Heinz Co. (1949 and 1958)
3) Vinogradov (1953)
* Fukai and Meinke (1959)
** Chipman (1958)

Table VI. Contribution of Selected Fish and Clam Samples to the Allowable Intake of Radioisotopes for Man[1)]

Sample	Daily[2)] intake μc	Fraction of Allowable Intake												
		Mn^{54}	Fe^{55}	Fe^{59}	Co^{57}	Co^{58}	Co^{60}	Zn^{65}	Sr^{90}	Y^{91}	Zr^{95} Nb^{95}	$Ru^{103-106}$ $Rh^{103-106}$	Ru^{106} Rh^{106}	Total
Flying fish muscle	.333	---	0	.003	.001	.017	.003	.127	0	---	<.002	<.001	---	.154
Flying fish liver	.524	---	0	.052	.003	.076	.010	.112	0	---	<.003	<.001	---	.257
Bonito liver	.500	.005	.016	<.004	.001	.002	.014	.080	0	0	<.003	---	0	.125*
Clam kidney	16.0	.145	.673	<.121	.145	.655	.291	0	<73.	.727	<.097	---	<.364	3.2+
Allowable daily intake, μc[3)]		2.2	17.6	1.32	11.0	2.2	1.1	2.2	.002	.66	1.32 2.2	1.76 .22 220. ---	.22 ---	

1) Samples from Table V, selected for high levels of radioactivity
2) Assumed to be 220 grams for fish muscle and 22 grams for liver or kidney; factor used for converting dry to wet weight was 4.5
3) $(MPC)_w$ from Handbook 69 x 2200

Table VII. Radioisotopes in Fish Caught by Japanese Fishermen, 1954-1957[1)]

Fish	Tissue	Date	Locality	$\mu\mu$c/g (wet tissue) Fe^{55}	Zn^{65}	Sr^{90}	Cd^{113m}	Cs^{137}
Big eye tuna	Muscle	6/56	So. Pacific	21;40	1.8			
" " "	Red muscle	" "	" "	1,200.	3.1			
" " "	Red muscle	10/56	" "	700.			120.	
" " "	Liver	" "	" "	5,000.	500.		12.	
" " "	Liver	6/56	" "	5,430.	36.			
" " "	Kidney	" "	" "	40,000.	531.			
" " "	Spleen	" "	" "	3,800.	60.6			
Yellow fin "	Kidney	6-7/54	" "		2,700.			
" " "	Muscle	" "	" "		33.	.0014		
" "	Liver	6/56	" "	1,000.	40.			
Carp	Muscle	6/57	Maebashi					.017
Cuttlefish		6/57	Pacific					.021
Skipjack	Muscle	5/57	Nojima					.016
Allowable daily intake[2)]				17.6×10^{6}	22×10^{5}	22×10^{2}	---	44×10^{4}

1) Data from Hiyama (1957)
2) Calculated from $(MPC)_w$ in Handbook 69

Relative Sensitivity of Various Organisms

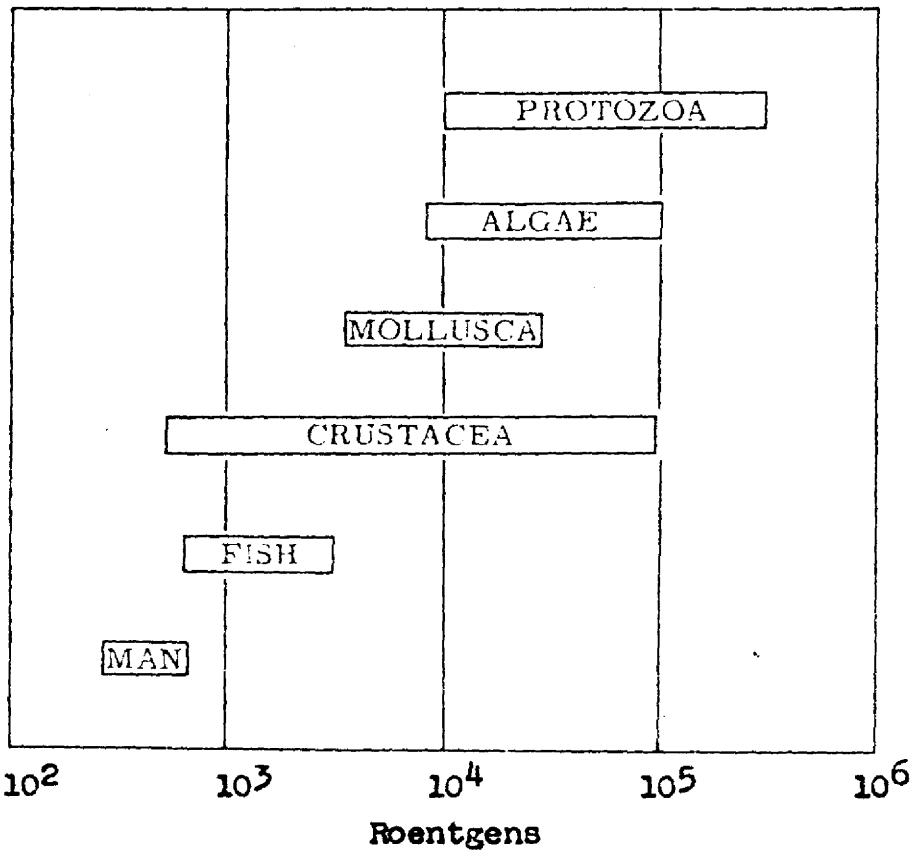

Fig. 1. X-Rays or Gamma Rays Required to Kill 50 Per Cent of Organisms.

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Zr^{95}	2.5	8.2	7.3	---	65 days	600
Nb^{95}	---	4.1	15.0	---	35 days	1,000
Ru^{106}	---	---	2.4	---	365 days	100
Rh^{106}	---	---	2.4	---	2 hours	*
I^{131}	6.3	3.7	---	---	8 days	20
Cs^{137}	---	---	1.5	18.2	27 years	200
Ba^{137m}	---	---	1.5	18.2	3 mins.	*

1) Hunter and Ballou (1951)

2) Strominger, Hollander and Seaborg (1958)

3) MPC values relative to Sr^{90}; based upon values from Handbook 69 for which MPC for Sr^{90} is 10^{-6} μc/cc

* A short-lived daughter for which hazard is negligible in comparison to hazard for parent

Table I. - (Continued)

Fission products	Per cent abundance at: [1] One week	One month	One year	Ten years	Half life[2]	Relative MPC values[3]
Ba^{140}	8.8	10.8	---	---	13 days	300
La^{140}	9.4	12.5	---	---	40 hours	200
Ce^{144}	---	2.0	26.5	---	285 days	100
Pr^{144}	---	2.0	26.5	---	17 mins.	*
Pm^{147}	---	---	5.7	15.8	950 days	2,000
Non-fission products						
Mn^{54}					290 days	1,000
Fe^{55}					950 days	8,000
Fe^{59}					45 days	600
Co^{57}					270 days	5,000
Co^{58}					71 days	1,000
Co^{60}					1900 days	500
Zn^{65}					245 days	1,000

1) Hunter and Ballou (1951)

2) Strominger, Hollander and Seaborg (1958)

3) MPC values relative to Sr^{90}; based upon values from Handbook 69 for which MPC for Sr^{90} is $10^{-6}\mu c/cc$

* A short-lived daughter for which hazard is negligible in comparison to hazard for parent

Table 1. Per Cent Abundance and Relative Maximum Permissible Concentration of Fallout Radioisotopes of Biological Interest

Fission products	Per cent abundance at:[1] One week	One month	One year	Ten years	Half life[2]	Relative MPC values[3]
Sr^{89}	2.1	6.7	2.7	---	50 days	100
Sr^{90}	---	---	1.8	21.8	28 years	1
Y^{90}	---	---	1.8	21.8	64 hours	200
Y^{91}	2.4	7.6	3.9	---	58 days	300
Zr^{95}	2.5	8.2	7.3	---	65 days	600
Nb^{95}	---	4.1	15.0	---	35 days	1,000
Ru^{106}	---	---	2.4	---	365 days	100
Rh^{106}	---	---	2.4	---	2 hours	*
I^{131}	6.3	3.7	---	---	8 days	20
Cs^{137}	---	---	1.5	18.2	27 years	200
Ba^{137m}	---	---	1.5	18.2	3 mins.	*

1) Hunter and Ballou (1951)

2) Strominger, Hollander and Seaborg (1958)

3) MPC values relative to Sr^{90}; based upon values from Handbook 69 for which MPC for Sr^{90} is 10^{-6} $\mu c/cc$

* A short-lived daughter for which hazard is negligible in comparison to hazard for parent

Table I. - (Continued)

Fission products	Per cent abundance at: [1] One week	One month	One year	Ten years	Half life[2]	Relative MPC values[3]
Ba^{140}	8.8	10.8	---	---	13 days	300
La^{140}	9.4	12.5	---	---	40 hours	200
Ce^{144}	---	2.0	26.5	---	285 days	100
Pr^{144}	---	2.0	26.5	---	17 mins.	*
Pm^{147}	---	---	5.7	15.8	950 days	2,000
Non-fission products						
Mn^{54}					290 days	1,000
Fe^{55}					950 days	8,000
Fe^{59}					45 days	600
Co^{57}					270 days	5,000
Co^{58}					71 days	1,000
Co^{60}					1900 days	500
Zn^{65}					245 days	1,000

1) Hunter and Ballou (1951)

2) Strominger, Hollander and Seaborg (1958)

3) MPC values relative to Sr^{90}; based upon values from Handbook 69 for which MPC for Sr^{90} is $10^{-6}\mu c/cc$

* A short-lived daughter for which hazard is negligible in comparison to hazard for parent

Table II. The Naturally Occurring Radioisotopes in Sea Water

Isotope	Total activity in ocean in megacuries[1]	Per cent of total activity	Radioactivity in sea water d/m/l[2]	Radioactivity in sea water $\mu c/cc$[3]	$(MPC)_w$[4] $\mu c/cc$
K^{40}	460,000	97.1	736	3×10^{-7}	*
Rb^{87}	8,400	1.77	13	6×10^{-9}	10^{-3}
U^{238}	3,800	.80	6	3×10^{-9}	2×10^{-4}
Ra^{226}	1,100	.23	2	10^{-9}	10^{-7}
C^{14}	270	.057	1	5×10^{-10}	8×10^{-3}
U^{235}	110	.023	1	"	3×10^{-4}
H^3	12	.003	1	"	.03
Th^{232}	8	.002	1	"	2×10^{-5}
Total	473,700	100.0	758	3×10^{-7}	

1) Revelle (1957)

2) Disintegrations per minute per liter based upon calculated value for K^{40}

3) Microcuries per cubic centimeter

4) Maximum permissible concentration for water consumed by man (Handbook 69, 1959)

* No value given, hazard negligible

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Rb^{87}	8,400	1.77	13	$6x10^{-9}$	10^{-3}
U^{238}	3,800	.80	6	$3x10^{-9}$	$2x10^{-4}$
Ra^{226}	1,100	.23	2	10^{-9}	10^{-7}
C^{14}	270	.057	1	$5x10^{-10}$	$8x10^{-3}$
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* No value given, hazard negligible

Table III. Radioisotopes Present in Plankton Collected soon after Fallout

Isotope	Half life	Per cent of total radioactivity
Bomb debris		
Np^{239}	2.3 days	69
U^{237}	6.8 days	3
Fission products		
Mo^{99}-Tc^{99m}	66 hours	12
Te^{132}-I^{132}	77 hours	8
Ru^{103}-Rh^{103}	40 days	3
Ru^{105}-Rh^{105}	36 hours[2)]	
Ru^{106}-Rh^{106}	1 year	
Ba^{140}-La^{140}	12.8 days	2
Ce^{141}-Pr^{141}	33 days	2
Zr^{95}-Nb^{95}	65 days	1
Ce^{144}-Pr^{144}	285 days	<1
		100+

1) Data from Lowman, (1960); average value for six samples collected less than 48 hours post shot; fallout from one detonation, Rehoboth survey, (1958)

2) Half life of daughter

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Ru^{105}-Rh^{105}	36 hours[2]	
Ru^{106}-Rh^{106}	1 year	
Ba^{140}-La^{140}	12.8 days	2
Ce^{141}-Pr^{141}	33 days	2
Zr^{95}-Nb^{95}	65 days	1
Ce^{144}-Pr^{144}	285 days	<1
		100+

1) Data from Lowman, (1960); average value for six samples collected less than 48 hours post shot; fallout from one detonation. Rehoboth survey, (1958)

2) Half life of daughter

Table IV. Mineral Elements of Nutrition and Other Elements of Interest and Their Occurrence in Sea Water and in Sea Foods

	Sea water[1]	Sea foods,[2] edible portions	Fish muscle[3] or soft parts	Concentration in fish muscle,	Fallout radio-
	PPM	PPM	PPM	median value	isotopes
A. Mineral elements of nutrition (man)[2]					
Chlorine	18,980		300 - 2,410	< 1	
Sodium	10,561	480 - 10,000	300 - 1,680	< 1	
Magnesium	1,272		100 - 4,520	2	
Sulfur	884		1,320 - 2,340	2	
Calcium	400	80 - 3,860	100 - 3,036	4	
Potassium	380	1,100 - 5,600	1,900 - 5,160	9	
Bromine	65		?	---	
Fluorine	1.4		4 - 34	14	
Iodine	.05		<1 - 27	270	$I^{131, 132}$

Table IV. - (Continued)

	Sea water[1]	Sea foods,[2] edible portions	Fish muscle[3] or soft parts	Concentration in fish muscle, median value	Fallout radio-isotopes
	PPM	PPM	PPM		
Phosphorous	.001 - .10	1,100 - 8,910	680 - 4,600	52,000	
Iron	.002 - .02	4 - 70	<1 - 56	2,500	$Fe^{55, 59}$
Copper	.001 - .01	<1 - 37	<1 - 37	3,700	
Manganese	.001 - .01		<1 - 252	25,000	$Mn^{54, 56}$
Zinc	.005		2 - 47	4,900	Zn^{65}
Cobalt	?		<1 - 1+	3,200*	$Co^{57, 58, 60}$
B. Elements always present in man but of unknown function[2]					
Silicon	.02 - 4.0		<1 - 40	10	
Aluminum	.5		<1 - 40	180	
Arsenic	.015		<1 - 12	490	
Nickel	.0001		?	---	

Table IV. - (Continued)

	Sea water[1]	Sea foods,[2] edible portions	Fish muscle[3] or soft parts	Concentration in fish muscle, median value	Fallout radio-isotopes
	PPM	PPM	PPM		
C. Other elements with isotopes of radiological interest					
Strontium	13.		+	<1**	$Sr^{89, 90}$
Cesium	.002		?	35+**	Cs^{137}
Cerium	.0004		?		$Ce^{141, 144}$

1) Sverdrup et al. (1946)

2) Heinz Co. (1949 and 1958)

3) Vinogradov (1953)

* Fukai and Meinke (1959)

** Chipman (1958)

Table VI. Contribution of Selected Fish and Clam Samples to the Allowable Intake of Radioisotopes for Man[1)]

Sample	Daily[2)] intake	Fraction of Allowable Intake												
	μc	Mn^{54}	Fe^{55}	Fe^{59}	Co^{57}	Co^{58}	Co^{60}	Zn^{65}	Sr^{90}	Y^{91}	Zr^{95} Nb^{95}	$Ru^{103-106}$ $Rh^{103-106}$	Ru^{106} Rh^{106}	Total
Flying fish														
muscle	.333	---	0	.003	.001	.017	.003	.127	0	---	<.002	<.001	---	.154
Flying fish														
liver	.524	---	0	.052	.003	.076	.010	.112	0	---	<.003	<.001	---	.257
Bonito														
liver	.500	.005	.016	<.004	.001	.002	.014	.080	0	0	<.003	---	0	.125
Clam														
kidney	16.0	.145	.673	<.121	.145	.655	.291	0	<73.	.727	<.097	---	<364	3.2+
Allowable daily intake, μc[3)]		2.2	17.6	1.32	11.0	2.2	1.1	2.2	.002	.66	1.32 2.2	1.76 .22 220. ---	.22 ---	

Table VI. - (continued)

1) Samples from Table V, selected for high levels of radioactivity

2) Assumed to be 220 grams for fish muscle and 22 grams for liver or kidney; factor used for converting dry to wet weight was 4.5

3) $(MPC)_w$ from Handbook 69 x 2200

* One per cent of total activity from unknown anions not included in total

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Flying fish muscle	.333	---	0	.003	.001	.017	.003	.127	0	---	< .002	.001	---	.15
Flying fish liver	.524	---	0	.052	.003	.076	.010	.112	0	---	< .003	.001	---	.25
Bonito liver	.500	.005	.016	< .004	.001	.002	.014	.080	0	0	< .003	---	0	.12
Clam kidney	16.0	.145	.673	< .121	.145	.655	.291	0	< .73.	.727	< .097	---	< .364	3.2+
Allowable daily intake, μc[3)]		2.2	17.6	1.32	11.0	2.2	1.1	2.2	.002	.66	1.32 2.2	1.76 220, 22 ---	.22 ---	

Table VI. - (continued)

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* One per cent of total activity from unknown anions not included in total

Table VII. Radioisotopes in Fish Caught by Japanese Fishermen, 1954-1957[1)]

Fish	Tissue	Date	Locality	$\mu\mu c/g$ (wet tissue) Fe^{55}	Zn^{65}	Sr^{90}	Cd^{113m}	Cs^{137}
Big eye tuna	Muscle	6/56	So. Pacific	21;40	1.8			
" " "	Red muscle	" "	" "	1,200.	3.1			
" " "	Red muscle	10/56	" "	700.			120.	
" " "	Liver	" "	" "	5,000.	500.		12.	
" " "	Liver	6/56	" "	5,430.	36.			
" " "	Kidney	" "	" "	40,000.	531.			
" " "	Spleen	" "	" "	3,800.	60.6			
Yellow fin "	Kidney	6-7/54	" "		2,700.			
" "	Muscle	" "	" "		33.	.0014		
" "	Liver	6/56	" "	1,000.	40.			
Carp	Muscle	6/57	Maebashi					.017

Table VII. - (continued)

				$\mu\mu$c/g (wet tissue)				
Fish	Tissue	Date	Locality	Fe^{55}	Zn^{65}	Sr^{90}	Cd^{113m}	Cs^{137}
Cuttlefish		6/57	Pacific					.021
Skipjack	Muscle	5/57	Nojima					.016
Allowable daily intake[2]				17.6×10^6	22×10^5	22×10^2	---	44×10^4

1) Data from Hiyama (1957)

2) Calculated from $(MPC)_w$ in Handbook 69

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Big eye tuna	Muscle	6/56	So. Pacific	21;40	1.8			
" " "	Red muscle	" "	" "	1,200.	3.1			
" " "	Red muscle	10/56	" "	700.			120.	
" " "	Liver	" "	" "	5,000.	500.		12.	
" " "	Liver	6/56	" "	5,430.	36.			
" " "	Kidney	" "	" "	40,000.	531.			
" " "	Spleen	" "	" "	3,800.	60.6			
Yellow fin "	Kidney	6-7/54	" "		2,700.			
" "	Muscle	" "	" "		33.	.0014		
" "	Liver	6/56	" "	1,000.	40.			
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