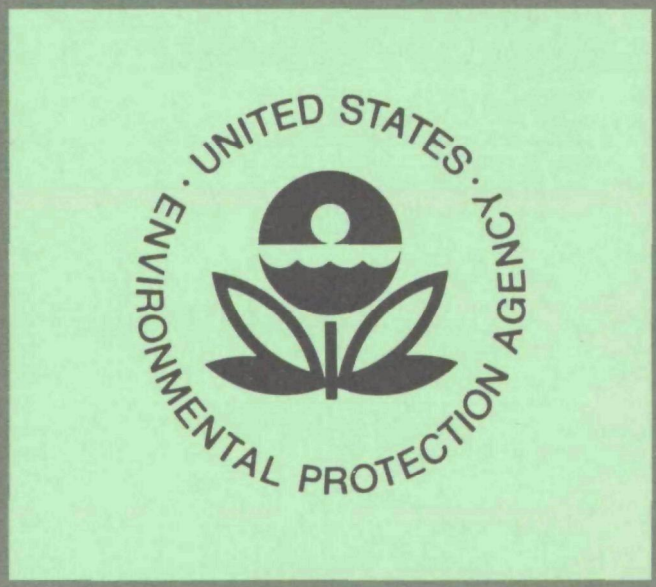
EPA-600/3-76-054
May 1976

Ecological Research Series

CONCENTRATION FACTORS AND TRANSPORT MODELS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS
A Literature Report



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EPA-600/3-76-054
May 1976

CONCENTRATION FACTORS AND TRANSPORT MODELS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS A LITERATURE REPORT

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Effective June 29, 1975, the National Environmental Research Center-Las Vegas (NERC-LV) was designated the Environmental Monitoring and Support Laboratory-Las Vegas (EMSL-LV). This Laboratory is one of three Environmental Monitoring and Support Laboratories of the Office of Monitoring and Technical Support in the U.S. Environmental Protection Agency's Office of Research and Development.

ABSTRACT

The relative risks to man from radionuclides released to the environment depend heavily on their accumulation or concentration by aquatic organisms. The organisms which accumulate those radionuclides present in the environment may be useful as indicators for environmental monitoring purposes. In addition, these organisms may be directly in food chain pathways to humans.

Literature is reviewed and summarized in regard to biological concentration of radionuclides in freshwater and marine environments. Concentration factors for elements found in organisms are tabulated for plants, invertebrates, and fish in marine and freshwater environs. Literature is also reviewed on models developed to calculate the possible radiation dose delivered to humans from radionuclides released into aquatic environments. The model approaches summarized range from simple generalized forms which, at best, give order of magnitude estimates to detailed models for a specific area which may be used to guide waste discharge practices.

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ACKNOWLEDGMENT

The technical assistance of Patricia L. Foster in summarizing information from the literature and in preparing this paper is greatly appreciated.

SECTION I
INTRODUCTION

The relative risks to man from radionuclides present in water depend heavily on their accumulation or concentration by organisms living in that water. Organisms which accumulate those radionuclides present in the environment may be useful as indicators for environmental monitoring purposes. In addition, these organisms may be directly or indirectly in food chain pathways to humans. The assessment of the potential human effects resulting from radionuclide releases to aquatic environments requires an understanding of the fate of the radionuclides in various environments. Of particular interest is the tendency for many organisms to extract and accumulate trace materials from their environment. This tendency is commonly measured in terms of a concentration factor (CF). The CF is defined as the ratio of the concentration of an element in an organism or its tissues to that found in ambient water. While this definition has some deficiencies, it provides a convenient means to compare the relative accumulation of substances in various organisms. Given the CF and other related parameters, mathematical models can be developed to provide estimates of the radiation dose to humans from the consumption of foods derived directly or indirectly from aquatic environments contaminated with radionuclides.

SECTION II

SUMMARY

Literature is reviewed and summarized in regard to biological concentration of radionuclides in freshwater and marine environments. Concentration factors for elements found in organisms are tabulated for plants, invertebrates, and fish in marine and freshwater environs. The range of concentration factors reported from environmental studies is given, and potential sources for variability in concentration factor determinations are discussed. Elements for which no data were found in the literature are identified.

The potential for using producer, primary consumer and higher order consumer organisms as biological indicators of environmental pollutant levels is discussed. Some research efforts in this area have been reported; however, no reliable pattern has emerged upon which to base a general monitoring program.

Considerable effort has been expended toward developing methods to calculate the possible radiation dose delivered to humans from radionuclides released into aquatic environments. Most of such work involves the use of mathematical models in which specific environmental information provides many of the required parameters. While many approaches are summarized in this report, most may be classed as either critical pathway or specific activity methods. The approaches summarized range from simple generalized forms which, at best, give order of magnitude estimates to detailed models for a specific area which may be used to guide waste discharge practices.

SECTION III

CONCLUSIONS

1. Concentration Factors

Variability in the concentration factors reported for similar organisms from different but similar environments appears to preclude the use of general concentration factors to assess the current or previous levels of radionuclides in any particular aquatic environment. Although many freshwater and marine organisms accumulate or concentrate radionuclides present in an aquatic environment, the concentration factors reported in the literature indicate that local conditions strongly affect concentration factors for organisms. Even for a given area the concentration factor for a given organism can be expected to vary with changing local conditions; for example, season, water temperature, and total biomass present.

2. Preliminary Estimates

The unit-rad contamination factors, discussed under RECOMMENDATIONS, page 5, are useful in making preliminary assessments of a situation. Further assessment will require field studies to evaluate the situation in aquatic environments where problems are anticipated. Each body of water has mixing characteristics which are unique in place and time. Moreover, radionuclides introduced into and diluted within aquatic environments can remain in solution or suspension, precipitate and settle on the bottom, or be reconcentrated by plants and animals. The species and population density of plant and animal life present in a body of water are also unique in place and time. In addition, the physical, chemical and biological interactions of a radioisotope are influenced by the presence or absence of other isotopes not only of that element but also of other elements.

3. Models for the Environment

Monitoring networks for aquatic environments can provide specific information required in models. Large numbers of environmental models

have been developed for use in calculating potential radiation doses to humans following release of radionuclides to aquatic environments. The radiation exposure of humans through environmental pathways has been studied extensively from almost the inception of the nuclear industry. Even the simplest of these models requires considerable information concerning the particular environment and population at risk in order to complete a risk or radiation dose assessment. Although simple or complex models may be written in generalized forms, the input parameters are specific for the environment of interest. The number and degree of complexity of models reported in the literature allow selection of a model to use for making calculations based upon the amount of information available on the environment of interest. The more extensive the data base, of course, the more reliable can be the information derived from the models.

4. Uses for Aquatic Organism Samples

Aquatic organisms can provide valuable information on radionuclide cycling in the environment. Because aquatic organisms concentrate radionuclides released to their environment to a considerable degree, even if variable, they provide valuable environmental monitoring samples which (1) indicate whether radioisotopes are concentrating within food chains leading to man; (2) delineate the geographical areas involved in a potential problem; (3) provide valuable information on whether additional control measures are needed; and (4) demonstrate environmental benefits gained when control measures are applied.

SECTION IV

RECOMMENDATIONS

1. Field Studies

This program was limited to a general literature review for data and models related to the behavior of radionuclides in aquatic environments. It was not within the program scope to evaluate the accuracy or reliability of information reported. The wide range in concentration factors found in the literature leads to the recommendation that, initially, field studies be conducted to evaluate the situation in each aquatic environment where problems are anticipated. A sufficient body of information exists to make preliminary evaluations of most environments; however, data from detailed field studies would supplement this information and, perhaps, provide a basis for recommending that less detailed studies be required to adequately assess hazards of radionuclides in other aquatic environments.

2. Preliminary Evaluations

It is recommended that the "unit-rad contamination factors" as published in Lawrence Livermore Laboratory (LLL) reports be used to make preliminary evaluations of radionuclide releases to aquatic environments. These factors are discussed under RESULTS. Areas where detailed field studies should be carried out can be selected on the basis of the preliminary evaluations. The LLL program is a continuing effort and revised information is incorporated into their system as more specific data become available. The reports contain a comprehensive listing of aquatic concentration factors and population dose estimates. The LLL program approaches environmental contamination by radionuclides and the resultant radiation dose to man from the standpoint of element specific activity--the biological-exchangeable-pool-of-elements concept. Using this approach, the passage of a radionuclide through the biosphere is presumed to be governed by the same factors that govern the distribution of the related stable element isotopes within the biological exchangeable pool. It is assumed that the radionuclide is biologically no more

available than the related stable isotopes within the environment. This approach should be valid for the types of radionuclide releases anticipated in the nuclear industry--low-level wastes diluted and dispersed in large bodies of air and water.

The product of the LLL program is a value for each isotope called the "unit-rad contamination in water (F_A)" for both freshwater and sea water. This factor is defined as the initial concentration of a radioisotope in water which would yield a 30-year integrated dose of 1 rad to a specific designated tissue of the standard man. In most cases F_A values for infants are also estimated. The assumptions in the many steps involved in reaching F_A values are basically conservative in nature, i.e., they are aimed toward obtaining F_A values for the worst situation that could develop.

3. Monitoring Network Design

Although considerable information is available for making generalized estimates of the radiation dose to people from radionuclides released to aquatic environments, information relevant to specific areas generally has not been collated. For example, seafood distribution and consumption patterns for an area are generally available through food distributors and dietary surveys, but have not been collated for making potential radiation dose estimates. Before such information is tabulated, however, a decision should be made on its desired accuracy and validity. The ultimate objective should be to obtain radiation dose estimates with a given degree of reliability. In order to obtain this objective, each data base for model parameters has a defined degree of reliability. Such considerations are part of monitoring network design criteria which are being developed at the EPA's Environmental Monitoring and Support Laboratory at Las Vegas. At present design criteria are being developed for air monitoring networks. These efforts should be extended to encompass design criteria for gathering information needed to assess the potential radiation dose to people from radionuclides released to aquatic environments.

4. Emphasis for Future Investigations

Some investigations on critical pathways indicate that the biomass and sediments may be neglected as a depot of radioactivity in a freshwater aquatic system, although as a rule of thumb it can be assumed that there is a thousandfold average concentration of all radionuclides in the biomass and that the bottom sediment has an average concentration a hundredfold greater than the water concentration. Apparently, the biomass and sediment have a small potential for transferring radioisotopes to people when compared with direct water intake. This concept, which has developed from tracer radioisotope studies in freshwater systems, has important implications in assessing freshwater pollutants of many types. The concept should be pursued in depth. Perhaps priority attention should be directed toward the oceans where the majority of all pollutants, however released, will ultimately reside. Some pollutants in the oceans will be returned to man through seafoods; however, the seafood yield of the world's oceans has been decreasing annually for many years. It appears that more emphasis should be placed on studying pollutant effects in ocean environments. Biological effects of released radioisotopes on ocean biota are not expected to be observable because of the low concentrations expected. However, radioisotopes already in the environment and those to be released in the future can provide valuable information on the rates of pollutant transfer from land sources to the ocean and on pollutant mixing patterns and fate in the oceans. Since man depends on the oceans for most of his oxygen, and, potentially, a large portion of his food, he has a vital stake in understanding and managing the productivity and well-being of the seas.

SECTION V

OBJECTIVES AND APPROACH

The objectives of this project were to survey literature on aquatic environments and:

1. Tabulate concentration factors for radioisotopes which may be released to the environment through man's activities and yield a significant radiation dose to man through aquatic pathways,
2. Identify indicator organism groups with high concentration factors for potential use in monitoring and assessment efforts, and
3. Survey environmental models used for calculating the potential radiation dose to man through aquatic pathways.

A concentration factor (CF) is defined as the ratio of the concentration of an element in an organism or its tissues to that found in ambient water. In reviewing and evaluating the literature, it was found that long-lived radioisotopes with CF's of about 10,000 or greater in biota were of primary importance for identifying indicator organisms and for estimating potential radiation doses to man. A CF of 10,000 or more is herein defined as the critical CF, and data on critical CF's are summarized in the following sections. The value of 10,000 was selected to concisely summarize data which are most pertinent to hazards assessment. Data on CF values less than 10,000 can be obtained from the referenced literature. In addition to isotopes with critical CF's, the CF's for several elements of current interest are included in the data summarized--even though the CF's are often relatively small. However, small amounts of some of these elements are considered to be much more dangerous to man than their CF's might appear to indicate. Furthermore, their potential biological effects on man warrant concern, vigilance and control. These elements include plutonium, strontium, cesium, uranium, and iodine. Since CF's for many isotopes were not identified in the

literature, and because the various isotopes of the same element generally exhibit the same chemical characteristics, the CF for one isotope is presumed to be applicable for other isotopes of the same element.

Indicator organisms may be useful in the following categories:

1. *Alert.* Indicate that radioisotopes are concentrating within food chains which could lead to man.
2. *Assess.* Evaluate the potential magnitude of intake by man and note trends in aquatic organism concentrations (increasing, decreasing, or stable).
3. *Delineate.* Show the geographical area involved in the potential problem.
4. *Control.* Decide whether control measures are needed and, if they are applied, demonstrate the environmental benefits gained.

Many models have been reported in the literature for calculating the potential radiation dose to man through aquatic pathways from radionuclides released to the environment. Several of these models are described in this report.

The approach was to survey literature on aquatic pathways and to summarize CF's for elements within organisms which live in various aquatic environments. This information, together with information on the radioisotopes released to the environment from nuclear activities, gives a data base to identify which radioisotopes may be important in various food chains leading to man, to identify indicator organisms with potential value in monitoring and assessment efforts, and to select appropriate models to make calculations of potential radiation doses to man. The type of data available on the environment in question determines which type of model is appropriate.

In addition to the critical CF criteria, the following guidelines were formulated to focus the literature review on information relevant to the objectives:

1. Primary attention is given to those radioisotopes which may yield a significant radiation dose to man through aquatic pathways.
2. Long-term and low-level releases of radioisotopes to the environment are of primary interest. Therefore, CF's for rather stable release rates and water concentrations are applicable. The nuclear industry guidelines for radioisotope discharges are of this nature. Concentration factors and environmental half-times for accidental releases (pulse labeling) are extremely dependent on local environmental conditions prevalent at the particular time of release and are not amenable to generalization. Therefore, time-dependent variables such as biological half-times and uptake rates are not identified in this report.
3. Certain types of data necessary for hazards assessment are beyond the scope of this report. The CF for a radioisotope bears little relationship to the potential hazard to man. Extensive data are available in the literature and in federal and international radiation standards on the relative hazards of radioisotopes based on decay emissions and energies, biological and physical half-lives, biological deposition sites, etc. These data are given for soluble and insoluble forms of the isotopes and for various routes of exposure. While such data may not be applicable for isotopes incorporated in unknown chemical forms in aquatic organisms, no attempt is made in this review to identify the chemical forms of radioisotopes within biota. The published data on biological and physical parameters for isotopes are not reiterated.
4. While certain radioisotopes are considered more hazardous than others, none were excluded from a search for concentration factors. The exclusion from such a search may lead to unwelcome uncertainties in the assessment of a situation

involving a mixture of radionuclides. In addition, it is difficult to predict the kinds and amounts of radionuclides to be released in the future from nuclear facilities, medical facilities, and various laboratories.

This report serves to present reported information on parameters needed to estimate radiation doses to man from various radionuclides which may be discharged to a given sector of an aquatic environment. Many parameters are extremely location-dependent and, therefore, are part of this literature survey only in that they are identified and evaluated in the dose computation models which are referenced. These parameters include such things as dilution factors, dispersal rates, mixing depths, biota prevalence, the organisms and their tissues directly consumed by man, and the indirect contributions from organisms in food chains.

SECTION VI

RESULTS

CONCENTRATION FACTORS

The term concentration factor (CF) has been defined (Thompson *et al.*, 1972) as "the ratio of the concentration of an element or radionuclide in an aquatic organism or its tissues to that in the surrounding water under equilibrium or steady-state conditions." This definition does not allow for the fact that aquatic organisms normally derive their nutrients from a variety of sources such as food, water, and suspended or deposited sediments. Polikarpov et al. (1966) noted this restriction in part by adding that "the capacity of an organism to accumulate radioactive substances is expressed by the ratio of its radioactivity to that of the aqueous medium or the preceding food link in which the radionuclide was concentrated."

This limitation is especially apparent with certain benthic organisms that live in bottom sediments and feed on debris (for example, some mollusca). Comparison of the trace element concentrations found in the tissues of these organisms with the concentrations found in open seawater is not a truly valid means to determine concentration factors. Yet, Lowman *et al.* (1971) describe many benthic organisms which are efficient accumulators of trace elements from water and are an important fraction of the total biomass in marine areas of economic importance.

Fewer problems are encountered in determining a CF for phytoplankton because there are no intervening trophic levels between water and their nutrient source. Data on marine plants (Thompson *et al.*, 1972) from the Lawrence Livermore Laboratory (LLL, formerly the Lawrence Radiation Laboratory) do not distinguish between phytoplankton and benthic or macrophytic algae, nor between zooplankton and macroinvertebrates. This ambiguity could lead to misinterpretation of what organisms are of most importance in a particular situation. Table 1 is a comparison of CF data from the LLL report to CF values for benthic algae and phytoplankton

reported by the National Academy of Science/National Research Council (NAS/NRC, 1971). In general, data from the latter report indicate that CF's for phytoplankton are higher (sometimes by 1 to 2 orders of magnitude) than those for benthic algae. Furthermore, the LLL average measured values tend to be lower than the NAS/NRC phytoplankton data, indicating that the LLL data are weighted towards benthic and/or macrophytic algae. On the basis of NAS/NRC data for phytoplankton, the CF's for aluminum, cerium, copper, silver, and zinc appear to be 10,000 or greater--values higher than the CF's listed in the LLL report (see Table 1).

TABLE 1. COMPARISON OF CRITICAL CONCENTRATION FACTORS FROM DIFFERENT SOURCES--MARINE PLANTS (VALUE LISTED X 10^3 = CF)

Element	Thompson *et al.* (1972) Marine Plants			NAS/NRC (1971)	
	Derived[a]	Maximum Measured[b]	Average Measured[c]	Benthic Algae	Phytoplankton
Sc	100.0	--	--	2.0	2.0
Mn	20.0	25.0	5.5	2.3	4.0
Fe	50.0	--	0.73	4.8	4.0-45.0
Y	10.0	--	--	0.48	1.0
Zr	10.0	--	--	2.2	60.0
P	2.8	22.0	3.0-4.0	10.0	3.0-34.0
Pb	1.0	--	5.0-10.0	0.7	40.0
Al	0.6	--	--	15.0	100.0
Ce	5.0	0.9	0.6-0.7	0.67	90.0
Cu	1.0	--	--	0.1	30.0
Ag	0.2	--	--	--	23.0
Zn	1.0	--	--	0.41	1.5-20.0
Pu	0.35	1.6-3.5	0.9-3.0	1.3	2.6

Dash (--) indicates no data in reference cited.

[a] Derived on the basis of stable element concentrations.

[b] Measured in radioisotope studies.

[c] Average of values from radioisotope studies.

In Table 2 marine animal data are listed in a similar manner from the the above LLL and NAS/NRC reports. Data from the former on marine invertebrates (crustaceans and mollusca) can be compared with values given for zooplankton, mollusc muscle and crustacean muscle in the NAS/NRC report. The latter data indicate two additional elements (ruthenium and aluminum) with critical CF's which were not shown in the LLL data.

TABLE 2. COMPARISON OF CRITICAL CONCENTRATION FACTORS FROM DIFFERENT SOURCES--MARINE ANIMALS (VALUE LISTED X 10^3 = CF)

Element	Thompson *et al.* (1972) Marine Invertebrates		NAS/NRC (1971)		
	Derived[a]	Average Measured[b]	Zooplankton	Mollusca	Crustaceans
P	28.6	30.0	13.0	6.0	24.0
Fe	20.0	--	25.0	9.6	2.4
Zn	100.00	50.0	8.0	11.0	2.0
Ge	16.7	--	--	--	--
Cd	250.0	--	--	--	--
Te	150.0	--	--	--	--
Hg	33.3	--	--	--	--
Sc	10.0	--	1.0	--	0.3
Mn	10.0	0.4	1.5	12.0	1.9
Ta	16.7	--	--	--	--
Tl	15.0	--	--	--	--
Po	20.0	3.0	--	--	--
Ru	2.0	0.1-1.0	34.0	0.003	0.1
Al	0.06	--	9.0	12.0	10.0

Dash (--) indicates no data in reference cited.

[a]Derived on the basis of stable element concentration.

[b]Average of values from radioisotope studies.

Critical CF's for marine organisms from the literature reviewed are listed in Tables 3, 4, and 5; those for freshwater organisms are listed in Tables 6, 7, and 8. Although much data exists in the literature, very little has come from environmental studies with radionuclides. The most comprehensive list of CF's found is contained in the LLL report (Thompson *et al.*, 1972). These data resulted from a multi-year effort which is continuing to update and extend the data base. This effort is part of a project which has the objective of assessing the radiation dose to people from radionuclides released to aquatic environments. The dose model being developed will be discussed later.

Harrison (1967) suggests that there are two classes of analytical techniques used to determine CF's, a distinction which was also incorporated into the LLL report by Thompson *et al.* (1972). These two classes are:

1. CF's obtained from measurements of stable element concentration in the organisms and water and
2. CF's obtained from measurements of the radionuclide content of the organisms and water.

Theoretically, the CF values obtained by these two methods should be identical, but in practice, both methods are subject to sampling and analytical errors. Jinks and Eisenbud (1972) describe several sources of variability in reported values for CF's:

TABLE 3. MARINE PLANTS--CRITICAL CONCENTRATION FACTORS (VALUE LISTED X 10^3 = CF)

Element	Chapman *et al.* (1968)	Thompson *et al.* (1972)				
				For Contaminated Environment[c]		
		Derived[a]	Measured[b]	Minimum	Maximum	Average
N	10.0	10.0	--	--	--	--
Sc	10.0	100.0	--	--	--	--
Mn	10.0	20.0	5.5	5.5	25.0	--
Fe	50.0	50.0	0.73	--	--	--
Y	10.0	10.0	--	--	--	--
Zr	10.0	10.0	--	--	--	--
In	100.0[d]	--	--	--	--	--
La[e]	10.0	--	--	--	--	--
Hf	10.0	--	--	--	--	--
Tl	100.0	--	--	--	--	--
Bi	--	--	--	--	--	--
P	--	--	--	0.21	22.0	4.0
Pb	--	--	--	--	--	5.0-10.0
Pu	1.0	0.35	1.0	0.77	3.5	3.0
Sr	0.013	0.013	0.01	0.009	0.2	0.025-0.04
Cs	0.02	0.02	0.05	0.01	0.17	0.04-0.07
U	1.0	0.067	--	--	--	--
I	4.0	4.0	1.0	--	--	1.0

Dash (--) indicates no data in reference cited.

[a] Derived on the basis of stable element concentrations.

[b] Measured in radioisotope studies.

[c] Measurements made in environmental studies.

[d] No data available, estimate based on worst possible case.

[e] For lanthanide series.

TABLE 4. MARINE INVERTEBRATES--CRITICAL CONCENTRATION FACTORS (VALUE LISTED X 10^3 = CF)

Element	Chapman *et al.* (1968)	Thompson *et al.* (1972)		Thompson *et al.* (1972) (For contaminated environment)[c]				
		Derived[a]	Measured[b]	Crustaceans	Mollusca			Crustacean & Mollusca
				Average	Minimum	Maximum	Average	weighted average
N	17.0	17.0	--	--	--	--	--	--
P	28.6	28.6	30.0	--	2.4	41.0	30.0	30.0
Fe	20.0	20.0	--	--	--	--	--	--
Zn	100.0	100.0	50.0	--	8.4	180.0	100.0	50.0
Ge	16.7	16.7	--	--	--	--	--	--
Cd	250.0	250.0	--	--	--	--	--	--
In	100.0	--	--	--	--	--	--	--
Sb	16.0	--	--	--	--	--	--	--
Te	--	100.0	--	--	--	--	--	--
Hg	100.0	33.3	--	--	--	--	--	--
Bi	--	--	--	--	--	--	--	--
Sc	1.0	10.0	--	--	--	--	--	--
Mn	5.0	10.0	0.4	0.00015	0.8	0.83	0.83	0.4
Ta	0.1	16.7	--	--	--	--	--	--
Tl	150.0	15.0	--	--	--	--	--	--
Po	0.4	20.0	3.0	--	--	--	5.0	5.0
Pu	0.286	0.1	0.2	--	0.1	0.38	0.28	0.2
Sr	0.006	0.006	0.02	0.03	0.005	0.017	0.003	0.02
Cs	0.02	0.02	0.025	0.02-0.04	0.009	0.15	0.037	0.025
U	0.33	0.01	--	--	--	--	--	--
I	0.05	0.05	--	--	--	--	--	--

Dash (--) indicates no data in reference cited.

[a] Derived on the basis of stable element concentrations.

[b] Measured in radioisotope experiments.

[c] Measurements made in environmental studies.

TABLE 5. MARINE FISH--CRITICAL CONCENTRATION FACTORS (VALUE LISTED X 10^3 = CF)

Element	Chapman *et al.* (1968)	Thompson *et al.* (1972)				
				For Contaminated Environment[c]		
		Derived[a]	Measured[b]	Minimum	Maximum	Average
N	60.0	60.0	--	--	--	--
P	28.6	28.6	--	--	--	--
Nb	30.0	30.0	--	--	--	--
In	100.0	30.0	--	--	--	--
Tl	100.0	--	--	--	--	--
Ta	0.1	30.0	--	--	--	--
Th	0.01	10.0	--	--	--	--
Pu	0.01	0.0035	0.003	0.001-0.003	0.005-4.5	0.003-0.013
Sr	0.0005	0.0005	0.002	0.003	0.004	0.003
Cs	0.03	0.03	0.04	0.02-0.03	0.13-0.45	0.1
U	0.01	0.01	--	--	--	--
I	0.01	0.01	--	--	--	--

Dash (--) indicates no data in reference cited.

[a] Derived on the basis of stable element concentrations.

[b] Measured in radioisotope experiments.

[c] Measurements made in environmental studies.

TABLE 6. FRESHWATER PLANTS--CRITICAL CONCENTRATION FACTORS (VALUE LISTED X 10^3 = CF)

Element	Chapman *et al.* (1968)	Thompson *et al.* (1972)				
				For Contaminated Environment[c]		
		Derived[a]	Measured[b]	Minimum	Maximum	Average
N	25.0	12.5	--	--	--	--
P	100.0	10.0	500.0	100.0	850.0	270.0
Sc	10.0	10.0	--	--	--	--
Mn	10.0	10.0	10.0	--	--	--
Y	10.0	5.0	--	--	--	--
Zr	10.0	1.0	--	--	--	--
In	100.0	--	--	--	--	--
La[d]	10.0	5.0	--	--	--	--
Hf	10.0	1.0	--	--	--	--
Tl	100.0	--	--	--	--	--
Sb	--	1.5	--	--	--	--
Zn	4.0	1.0	20.0	--	--	130.0
Ru	2.0	0.2	2.0	1.8	12.0	--
Ce	10.0	5.0	4.0	2.0	10.0	--
Pu	1.0	0.35	--	--	--	--
Sr	0.5	0.5	0.5	0.24-0.33	0.28-0.69	0.6
Cs	0.2	0.08	0.5	0.07-0.47	0.87-1.2	--
U	1.0	1.0	0.0005	0.0004	0.0007	0.0006
I	0.1	0.04	--	--	--	--

Dash (--) indicates no data in reference cited.

[a] Derived on the basis of stable element concentrations.

[b] Measured in radioisotope experiments.

[c] Measurements made in environmental studies.

[d] For lanthanide series.

1. Because of the higher concentration of stable mineral elements in seawater, the radionuclide CF's for most freshwater organisms are much higher than those for similar seawater biota.
2. Radionuclide accumulation and concentration in various tissues of an organism may differ considerably. Data illustrating this variability were published by Harvey (1967).
3. Error originates from the practice of analyzing the stable elemental content of biota and then calculating the CF from general literature data on the elemental concentrations in freshwater or seawater.
4. Physiological factors (same as those discussed by Harrison, 1967): species differences, age, size, reactions to changes in temperature,

and chemical characteristics of the environment vary. The concentration of an element in tissue may be controlled homeostatically. Such control of potassium and calcium concentrations in organisms may strongly affect CF's for their chemical congeners, cesium and strontium, respectively. While such effects are well-known for higher animals, they may occur in many simple organisms. For example, the concentration of manganese in Hudson River fish remains constant in spite of large variations in the water concentration of manganese.

5. A CF with respect to water may be influenced by the pathway through which the nuclide reaches the organism. Although an organism may not directly concentrate an element from water, some element in its food chain may have performed the concentrating function. Furthermore, the food source for an organism may vary with seasons and be affected by many environmental factors.
6. Short-term variations in the concentration of stable elements or radionuclides in environmental media may result in nonequilibrium conditions between organisms and the environment at the time of measurement. The concentrations of elements with long effective half-times in the tissue may reflect an earlier high concentration in the environment rather than that existing at the time of capture.

Additionally, some studies indicate an apparent selectivity by a few organisms for certain radionuclides over their stable counterparts. Presumably, this is related to differences in physio-chemical availability or to different pathways of uptake. For example, data from the LLL report (Thompson *et al.*, 1972) show that the following elements have experimentally measured CF values (based on radionuclide concentrations) that are greater than the derived values (based on stable element concentrations) by 1 or more orders of magnitude:

Biota	Elements with CF measured value >10 x derived value
Marine:	
Plants	Lead, Plutonium
Invertebrates	Strontium
Fish	Strontium
Freshwater:	
Plants	Phosphorus, Chromium, Zinc, Ruthenium, Cesium
Invertebrates	Sodium, Chronium
Fish	Sodium, Chronium, Strontium, Cesium, Polonium

TABLE 7. FRESHWATER INVERTEBRATES--CRITICAL CONCENTRATION FACTORS (VALUE LISTED X 10^3 = CF)

Element	Chapman *et al.* (1968)	Thompson *et al.* (1972)				
				For Contaminated Environment[c]		
		Derived[a]	Measured[b]	Crustacean	Mollusca	Average
N	42.5	150.0	--	--	--	--
P	100.0	100.0	20.0	10.0	20.0	20.0
Mn	40.0	40.0	90.0	--	93.0	90.0
Zn	40.0	10.0	10.0	4.0	20.0	10.0
Ge	16.7	--	--	--	--	--
In	100.0	--	--	--	--	--
Sb	16.0	--	--	--	--	--
Te	--	100.0	--	--	--	--
Hg	100.0	100.0	--	--	--	--
Tl	100.0	10.0	--	--	--	--
Po	0.4	20.0	--	--	--	--
Pu	0.29	0.1	--	--	--	--
Sr	0.7	0.1	--	--	--	--
Cs	1.0	0.1	--	--	--	--
U	0.3	0.1	0.06	0.06	--	0.06
I	0.025	0.005	--	--	--	--

Dash (--) indicates no data in reference cited.

[a] Derived on the basis of stable element concentrations.

[b] Measured in radioisotope experiments.

[c] Measurements made in environmental studies.

TABLE 8. FRESHWATER FISH--CRITICAL CONCENTRATION FACTORS (VALUE LISTED X 10^3 = CF)

Element	Chapman *et al.* (1968)	Thompson *et al.* (1972)				
				For Contaminated Environment[c]		
		Derived[a]	Measured[b]	Minimum	Maximum	Average
N	150.0	150.0	--	--	--	--
P	100.0	150.0	100.0	0.5-30.0	55.0-100.0	50.0
Nb	30.0	30.0	--	--	--	--
In	100.0	--	--	--	--	--
Tl	100.0	10.0	--	--	--	--
Np	100.0	--	--	--	--	--
Ta	0.1	30.0	--	--	--	--
Pu	0.01	0.003	--	--	--	--
Sr	0.04	0.005	0.03	0.003	0.17	0.058
Cs	0.001	0.4	2.0	0.39-1.6	1.1-4.7	2.4-3.9
U	0.01	0.01	0.002	--	--	0.002
I	0.001	0.015	--	--	--	--

Dash (--) indicates no data in reference cited.

[a]Derived on the basis of stable element concentrations.

[b]Measured in radioisotope experiments.

[c]Measurements made in environmental studies.

In general, the elements that are concentrated significantly in aquatic organisms (Tables 1 to 8) were grouped by Lowman *et al.* (1971) into at least one of five categories:

1. Structural elements: Carbon, Nitrogen, Phosphorus, Silicon, Calcium, Strontium.
2. Catalyst elements: Iron, Copper, Zinc, Manganese, Cobalt, Nickel, Chromium, Cadmium, Silver.
3. Elements easily hydrolyzed at seawater pH: Aluminum, Gallium, Scandium, Yttrium, Cerium, Plutonium, Titanium, Zirconium.
4. Heavy halogens: Bromine, Iodine.
5. Heavy divalent ions: Barium, Radium, Rubidium.

Supportive data on concentration factors for several elements in various biota have not been identified. For seawater these elements are indium, tellurium, and bismuth. In freshwater the elements with least known data are indium, thallium, bismuth, tellurium, and germanium for invertebrates and neptunium for fish.

INDICATOR ORGANISMS

Biological indicators are defined (Rice, 1965) as those organisms which concentrate relatively large amounts of specific radionuclides, thereby making it possible to detect the presence of those isotopes in the environment through an analysis of the organisms. Additional qualifying characteristics of "indicators" are listed by Feldt (1971). The organism must:

1. be readily attainable at all times,
2. have a sufficient CF for several radionuclides of interest, and
3. yield measurement results in a form which makes it possible to assess the radiation dose to man.

The concentration of elements in aquatic ecosystems is highly variable. CF's are related to the chemical content of the water, and it is impossible to draw general conclusions from elemental composition of organisms without simultaneous reference to data from their environment. In spite of this, however, Polikarpov (1966) states that it is generally recognized that CF's (taken in the equilibrium state) of each separate radionuclide for closely related species of marine plants and animals (i.e., same genus or family) do not differ significantly in different seas and oceans. There appears to be less acceptance of a second generalization attributed to Polikarpov: that CF's for radionuclides which are present in water in microquantities are similar for related marine and freshwater organisms. It has been more frequently observed that the freshwater CF for a particular isotope by a particular type of organism is higher than that in a marine environment (due to differential isotopic dilution in these two basic systems).

A logical, universal biological indicator of radioactivity in any aquatic system would appear to be mixed phytoplankton. Those elements that are concentrated by at least a factor of 1,000 by marine phytoplankton have been tabulated by the NAS/NRC (1971). Unfortunately, however, photoplankton do not accumulate nuclides in the same propor-

tions as, for example, fish. For mixed phytoplankton to meet the third qualifying characteristic previously noted, they would have to be a predictable component of a food web eventually leading to man. Even though phytoplankton may remove large amounts of radionuclides from water, the amount of radioactivity that may be passed up the food web will vary with cell mass, cell numbers, and specific concentrations, as well as with efficiency of utilization by ensuing trophic levels. Another complication with using mixed phytoplankton has been described by Feldt (1971): "It is especially difficult. . ." (in rivers, and presumably estuaries and coastal areas too) ". . . to separate the plankton from detritus." He concludes that while the concentration of certain nuclides in plankton would help to prove the presence of these nuclides, it would not give useful information on the risks these radionuclides present to man because they are not found in the same concentrations in edible aquatic products.

With these arguments in mind, it seems natural to fall back to the Polikarpov (1966) terminology -- that there are "biological indicators" (as previously defined) and there are "biological accumulators" (of which mixed phytoplankton would be an example). By the guidelines set forth for this report, the "accumulators" would be useful only in the *delineation* of the geographic area involved in a potential hazard to man. "Biological indicators" would be required for the other categories: *alert* (indicate that radioisotopes are concentrating within food chains that may lead to a hazard for man); *assess* (give indication of the potential magnitude of intake by man); or *control* (indicate whether or not control measures are effective).

In reviewing literature on biological indicators and accumulators, organisms were separated into the broad categories of producers and consumers.

Producers

In general, the literature reviewed does not present sufficient information for determining which specific producer organism is an accumulator or an indicator for a particular radioisotope. Most compilations of CF's have categorized producers very broadly and lump

together phytoplankton, littoral and benthic seaweeds and algae; submerged, floating, and emergent angiosperms; etc. The only generally accepted generalization concerning producers, according to the IAEA (1971), is that phytoplankton tend to concentrate activation products to a greater extent than fission products. This reference also notes that while the degree of concentration of activation products is highest in the primary producers, intermediate in the herbivores and lowest in the carnivores, the primary producer's concentration in comparison to fission products is more subject to variation and rapid fluctuation in response to changes in the ambient contamination level. The concentration responses in the higher trophic species are more sluggish and less predictable.

Primary Consumers

Few valid generalizations can be made from the literature about the indicator value of tropic levels. For example, filter feeding animals have been shown to concentrate different radionuclides even while they are living in the same microenvironment. (For example, see Table 9.) Also, the distribution of a radionuclide within a particular organism may vary greatly; e.g., even though scallops were found to be excellent accumulators of manganese-54, most of the activity was present in the kidney, which is not eaten by man, and relatively little was present in the muscle. Bryan *et al.* (1966) reported that CF's generally decrease from lower to higher trophic levels because of radioactive decay, mode of uptake, and turnover rates. A summary of the ranges reported for element CF's in marine organisms at various trophic levels is given in Table 10.

Higher-Order Consumers

In general, the same limitations that have already been noted apply to organisms in the higher trophic levels. Feldt (1971) argues that fish are excellent indicators in freshwater systems because of their availability, their measurable (although not usually maximal) CF's for a wide variety of isotopes, and their applicability to assessment of hazard to man. For the same reasons, he also supports the use of mussels as the indicator organism in river estuaries.

TABLE 9. RELATIVE ABUNDANCE OF RADIONUCLIDES IN FOUR SPECIES OF MOLLUSCA FROM THE SAME ENVIRONMENT(a)

	^{144}Ce	^{106}Ru	^{137}Cs	^{54}Mn	^{65}Zn
Scallops	4	3	2	1	5
Oysters	2	3	(b)	(c)	1
Clams	1	2	(b)	(c)	(c)
Mussels	1	2	(c)	(c)	(b)

(a) Data from Bryan *et al.* (1966).
(b) Present but not relatively abundant.
(c) Indication of presence

Studies on the uptake of radionuclides by various fish in the Columbia River (Foster and McCannon, 1962) have shown that there is a wide variation in uptake between individual specimens of the same species, thus requiring large samples for statistical validity. Uptake also changed rapidly with the season. The most pronounced variations were found with short-lived radionuclides that were necessarily acquired via food chains.

In summary, it appears that few generalizations can be made concerning indicator organisms. Most studies have identified accumulators with varying degrees of effectiveness, but still there appears to be no consistently reliable pattern. Relatively few studies have attempted to describe, much less quantify, entire aquatic ecosystems. Yet, at present this appears to be the only way to make intelligible recommendations concerning the identity of accumulator and/or indicator organisms.

ENVIRONMENTAL STUDIES

Those studies which have actually identified indicator organisms have been, at least initially, comprehensive ecosystem effects studies. Since the overwhelming complexity of environmental effects seriously hinders effective control, it is accepted practice to determine just which factors are critical (i.e., those which give rise to the greatest risk) and to

TABLE 10. RANGES OF ELEMENT CONCENTRATION FACTORS IN MARINE ORGANISMS[a]

	Algae				Grazers				Predators							
	Sessile			Plankton[b]			Plankton[c]			Shellfish			Plankton[d]			Fish			Squid		
Ag	100	—	1,000	< 100	—	220	<100	—	--	330_5	—	$20,000_6$	<45	—	900_4	--	—	--	900	—	3,000
Cd	11	—	20	< 350	—	6,000	<80	—	10^5	10^5	—	$2x10^6$	<300	—	10^4	--	—	>10	2,800	—	--
Ce	100	—	3,300	2,000	—	4,500	--	—	--	40	—	300	--	—	--	5	—	12	--	—	--
Co	15	—	740	75	—	1,000	<110	—	10^4	24_4	—	260_5	<70	—	1,300	28	—	560	<200	—	50,000
Cr	100	—	500	< 70	—	600	<15	—	10^4	$6x10^4$	—	$3x10^5$	<55	—	3,900	3	—	30	<70	—	--
Cs	16	—	50	16	—	22	6	—	15	3	—	15	--	—	--	6	—	10	--	—	--
Fe	1,000	—	5,000	750	—	7,000	440	—	60,000	$7x10^4$	—	$3x10^5$	3,000	—	30,000	400	—	3,000	1,000	—	3,000
I	160	—	7,000	--	—	--	--	—	--	40	—	70	--	—	--	10	—	--	--	—	--
Mo	10	—	200	< 3	—	17	2	—	175	30	—	90	<2	—	14	200	—	--	<10	—	--
Mn	20	—	20,000	300	—	7,000	21	—	4,000	3,000	—	60,000	270	—	1,600	95	—	10^5	1,000	—	--
Ni	50	—	1,000	25	—	300_6	2	—	$1,000_6$	17	—	90	17	—	90	--	—	--	30	—	80_5
Pb	8,000	—	20,000	1,000	—	$3x10^6$	3,000	—	$2x10^6$	200	—	60,000	200	—	60,000	5	—	10,000	100	—	$2x10^5$
Ru	100	—	1,200	< 200	—	--	10	—	6,000	10	—	2,400	10	—	2,400	10	—	--	400	—	2,100
Sr	0.1	—	90	0.9	—	54	1	—	85	1.2	—	10	1.2	—	10	4	—	5	0.9	—	1.2
Ti	200	—	30,000	600	—	10,000	28	—	30,000	110	—	20,000	110	—	20,000	--	—	--	300	—	3,000
Zn	80	—	3,000	200	—	1,300	125	—	500	50	—	--	50	—	--	280	—	20,000	2,500	—	--
Zr	200	—	3,000	< 1,000	—	20,000	360	—	30,000	<800	—	40,000	<800	—	40,000	5	—	--	20,000	—	--

Dash (--) indicates no data in reference cited. Long dash (—) means "to" in giving the ranges of CF's

(a) Data from the NAS/NRC (1971).

(b) Phytoplankton and Sargassum.

(c) Copepods, Pteropods, Salps and Doliolid.

(d) Euphausiids, Planktonic Amphipods, Shrimp (Acanthephyra, Palcomonetes).

exclude all other factors which make no significant contribution to the risk. For each instance of environmental contamination with radioactive materials, it is initially necessary to identify the critical radionuclides with reference to man. The physical and metabolic characteristics of these nuclides determine which ecological pathways or routes to man are critical. Since it is generally recognized that some members of the general population may be more affected than others, it is often necessary to further identify a critical population (Comar and Lengemann, 1966; IAEA, 1971; Straub, 1960).

Typical of this approach are studies (Foster and Soldat, 1966) of radionuclides discharged to the environment from the Hanford operations. The critical radionuclides and routes of exposure from effluents released into the Columbia River involve ^{32}P and ^{65}Zn in local fish and the produce from irrigated farms; ^{24}Na, ^{76}As, ^{239}Np and ^{131}I in drinking water; and the external exposure of swimmers to ^{24}Na in water. The critical population was determined to be persons who ate unusually large quantities of fish caught in the river immediately downstream from the reactors. For most of the population, however, drinking water provided the only significant source of waterborne radionuclides.

Parker (1964) maintains that only large-scale producers/users of radionuclides need to be considered in determining permissible discharge limits to freshwater environments. The use of river water for the dilution of low-level radioactive wastes is widely practiced. In addition to the studies on the Columbia River, other environmental studies in freshwater environments have generally indicated that local fish were the critical indicator organisms.

A study of the Clinch River by the Oak Ridge National Laboratory (Cowser *et al.*, 1963) showed that the major pathways of exposure were (1) consumption of contaminated water and fish; (2) consumption of agricultural produce irrigated with river water; (3) exposure to contaminated water and bottom sediments directly; and (4) exposure to the buildup of radionuclides in sludge and deposits in water systems which utilize the river water. The critical radionuclides identified were ^{90}Sr for bone, ^{137}Cs for total body, ^{106}Ru for the gastrointestinal tract, and ^{131}I for the thyroid.

Environmental studies of marine ecosystems generally have been undertaken in response to two types of situations: (1) to determine the effects of high-level discharges from nuclear detonations, and (2) to determine the effects of continuous low-level discharges from nuclear facilities. The latter situation is of primary interest, and studies of discharges from power reactors in England provide information that illustrates the variability of critical factors for different environments.

The Irish Sea coastal area adjacent to the Windscale reprocessing plant is one of the most important areas known with respect to the degree of radioactive contamination in the marine environment. The critical radionuclides released were determined to be ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{95}Nb, ^{144}Ce and ^{90}Sr. The most critical pathway is for ^{106}Ru in the seaweed, *porphyra*, which is used to make laverbread. A critical group of heavy consumers of this foodstuff was identified. For three nuclear power stations sited on England's open coastline, the exposures from ^{65}Zn, ^{60}Cs, and ^{124}Sb were the controlling factors in limiting waste discharges. At Hinkley, accumulation of these radionuclides in silt and fish flesh established the permissible discharge levels, whereas at Dungeness and Sizewell accumulation in fish flesh alone was definitive (Straub, 1964)

The potential damage to man and his environment is the limiting criterion on waste releases in most Western countries, whereas Russian interpretation requires that releases to the environment meet drinking water and breathing tolerances. The doses that cause injury to plants and animal life are, according to most Western authorities, much higher than would be permitted for human exposure. As an example, no effects were noted following the irradiation of chinook salmon at dose rates of up to 5.0 rem/day beginning immediately after fertilization of the egg. The possibility of synergism occurring with the combination of ionizing radiation and heated effluent temperatures also has been investigated; while no such synergism has been demonstrated, it has been shown that some organisms absorb radionuclides up to 50% faster due to increased metabolic rates in warmer waters (Eisenbud, 1973).

RADIATION PATHWAY AND DOSE MODELS

This section describes ways in which radionuclide releases to aquatic environments can be translated into estimates of the resulting potential radiation dose to people. Present radiation guides for permissible radionuclide concentrations in water are based upon direct consumption of the contaminated water by people. Although such direct intake does not take into account potential radionuclide transfer and subsequent consumption through aquatic food chains, this intake through the diet is recognized, and the basic radiation standards are based on permissible radiation dose rate and accumulated dose from all sources--exclusive of natural background and medical radiation (ICRP, 1959, 1962, 1966a and b, and 1968; USAEC, 1970; NAS/NRC, 1972; NCRP, 1971). The basis for and detailed descriptions of radiation standards are described and discussed extensively in the literature and are outside the scope of this report. However, a brief discussion of the models used in deriving air and water standards is helpful.

The International Commission on Radiological Protection (ICRP, 1959, 1962, 1966a and b, and 1968) has established recommended values for maximum permissible total body burdens (q), and for maximum permissible concentrations in air and water (MPC_a and MPC_w, respectively). Values are given for about 240 radionuclides. These values are based on two metabolic models: (1) an exponential or compartmental model, and (2) a power function model. In the exponential model each body organ is assigned a biological half-life, uptake fraction, etc., and the radiation dose to different organs can be calculated following a given radionuclide intake. Since considerable data indicate that for some radionuclides the fraction of the body burden excreted daily varies inversely with time, an alternative power function model is used to estimate the radiation dose for certain long-lived radionuclides (e.g., strontium, radium, plutonium and uranium). The permissible body burdens of these bone-seeking radionuclides are based on the comparison of the energy deposited in bone by the particular radionuclide with that deposited by 0.1 μCi of ^{226}Ra and its daughters. The derivations of these models and a listing of model parameters for

each radionuclide (with literature references) are given in the ICRP reports.

The ICRP recommendations have been basically incorporated into U.S. radiation standards. Such regulations start with a permissible radiation dose, from which permissible concentrations in air and water are calculated; these are based on the standard physiological factors for man, and would be expected to yield the permissible dose following the direct intake of such air and water. It must be emphasized that the basic criterion for the standards is radiation dose, and that the air and water concentration guides are derived values. The derived guides must take into account radionuclide exposure and intake from all sources. One of the important routes of radionuclide intake by people is through aquatic foods from environments which have received discharges of radioactive materials. As described in previous sections, many environments have unique characteristics, and generalizations about critical pathways for radionuclides are tenuous. As a result, there exists in the literature a multitude of models for such pathways in specific cases and for specific conditions. The general approaches used in pathway and dose modeling are herein described, together with a brief description of those modeling efforts which appear to be comprehensive and applicable to initial planning for radionuclide pathways assessment.

There are basically three methods of translating recommendations for the maximum radiation dose into guides for acceptable discharge rates for the various radionuclides. The first, which was described briefly above, involved maximum permissible concentrations in air and water for individual radionuclides or mixtures. If dilution, dispersion, and decay rates are known, then a discharge rate which meets these standards can be calculated. This approach, as mentioned above, considers only direct intake of contaminated air or water and neglects environmental pathways which may be more limiting.

A second approach, called the critical pathway method, is frequently used in assessing the potential dose to segments of the population through environmental pathways. Through investigation of environmental

transfer mechanisms for radionuclides anticipated to be present, critical pathways leading to the exposure of people to radiation are identified. The groups of people most likely to be exposed through critical pathways are identified; and the group most likely to receive the highest dose, in relation to a recommended permissible dose, is identified as the critical group. The isotope which provides the largest dose through the critical pathway may be called the critical radionuclide. The permissible environmental levels of the critical radionuclides are then calculated and frequently called Derived Working Limits (DWL). Although critical pathways, radionuclides, and population groups can be identified and may be used to calculate a DWL for each radionuclide, the basic limiting factor is total radiation dose from all radionuclides in all pathways--including direct intake from air and water. This approach, then, is a logical extension of the air and water MPC approach and integrates exposures from indirect environmental pathways.

The third approach, based on specific activity, is somewhat different. The use of specific activity, or radioactivity content per unit mass of an element in a medium, was first proposed by the NAS/NRC (1962) for establishing permissible levels of environmental radioactivity in regard to radioactive waste disposal into the Pacific coastal waters of the United States. In this approach it is presumed that the radionuclide specific activity in any organism, including man, would not exceed that in its basic environmental substrates. Although a radionuclide may be concentrated many orders of magnitude through physical or biological processes in the environment, radioactive nuclides of that element are always diluted by stable isotopes in the environment and the specific activity remains the same throughout various environmental media. Since the elemental composition of man and his various tissues is well documented in the literature (e.g., as summarized by the ICRP, 1959), the maximum dose to man which could result from a given specific activity in the environment can be calculated. For the case in which only a portion of the environment is contaminated, the calculated dose is modified by man's degree of involvement with that portion of the environment.

These three kinds of models are interrelated and require (NAS/NRC, 1971) a great deal of supplemental information concerning:

1. Kinds and quantities of radionuclides present.
2. Physical and chemical forms of the radionuclides.
3. Initial mechanism of dispersal.
4. Physical processes of dilution.
5. Availability to biota.*
6. Concentration factors and uptake rates.*
7. Consumption of marine products.
8. External exposure and exposure to other sources.

*Not used in specific activity approach.

In both the critical pathway and specific activity approaches, it is necessary to know, or to estimate (1) the kinds and quantities of radionuclides present; (2) the physical and chemical form of the nuclides; (3) the method of entry into the environment; (4) the extent of environmental transport and deposition through physical and chemical processes; and (5) the importance of biological processes in transport and concentration phenomena (Pritchard, 1961 and Russell, 1964).

CRITICAL PATHWAY APPROACHES

Parker (1959) has summarized major and moderate contributor pathways to total radiation dose from radioactive wastes discharged to surface waterways. The major pathways Parker evaluates are drinking water, immersion, biological chains, irrigation, waste treatment plants, and external exposure from proximity to radioisotopes.

Varying degrees of sophistication have been used in determining the allowable environmental contamination by the critical pathways approach. A simple method for seafood is described by Pritchard (1959) who used the following generalizations:

$$MPC_{sf} = 20 \cdot MPC_{w} \qquad (1)$$

where:

MPC_{sf} = maximum permissible concentration in seafood

MPC_w = maximum permissible concentration in water; as described by the ICRP (1959)

20 = factor derived on the basis of a diet in which 50% of the protein requirement is met by aquatic foods, and radionuclides in such foods constitute the only source of ingestion

$$MPC_{sf} = 4 \cdot MPC_w \quad (2)$$

where:

4 = 20% of the 20, above, on the assumption that 20% of the entire maximum permissible radionuclide intake is assigned to seafoods

$$MPC_{sw} = \frac{MPC_{sf}}{CF} \quad (3)$$

where:

MPC_{sw} = maximum permissible concentration in seawater derived from assumptions for either equation (2) or (3)

CF = concentration factor from water to organism of interest

This approach is simple and easily understood. However, the arbitrary assignment of the portion of the total radionuclide intake to be permitted from seafood is controversial. In addition, reliable information on CF's and dietary habits for most areas are not generally available.

As an example of a rather sophisticated critical pathway model, an outline of some general steps is given in Figure 1. Starting with an allowable dose rate to man, a maximum allowable discharge rate for radionuclides is calculated. The calculation may proceed along the following lines for the particular case of ingestion of a radionuclide in a contaminated seafood (NAS/NRC, 1959; Parker, 1959). Starting with the recommended maximum permissible concentration for water (MPC_w) a derived working limit for seafood, $(DWL)_{sf}$, can be calculated (Wolfe and Rice, 1968);

$$(DWL)_{sf} = \frac{(MPC)_w \ 2200 \ F}{I} \quad \mu Ci/g \quad (4)$$

where:

I	=	Rate of ingestion for seafood, g/day/person
2200	=	Rate of water intake, g/day/person
F	=	Fraction: an administrative number to apportion to seafood some fraction of the maximum permissible intake from water.

A derived working level for seawater, $(DWL)_{sw}$, can be obtained directly from $(DWL)_{sf}$ by dividing by the ratio of the elemental concentration in the seafood to that in the seawater, i.e., by the concentration factor, CF (Wolfe and Rice, 1968):

$$(DWL)_{sw} = \frac{(DWL)_{sf}}{CF} \tag{5}$$

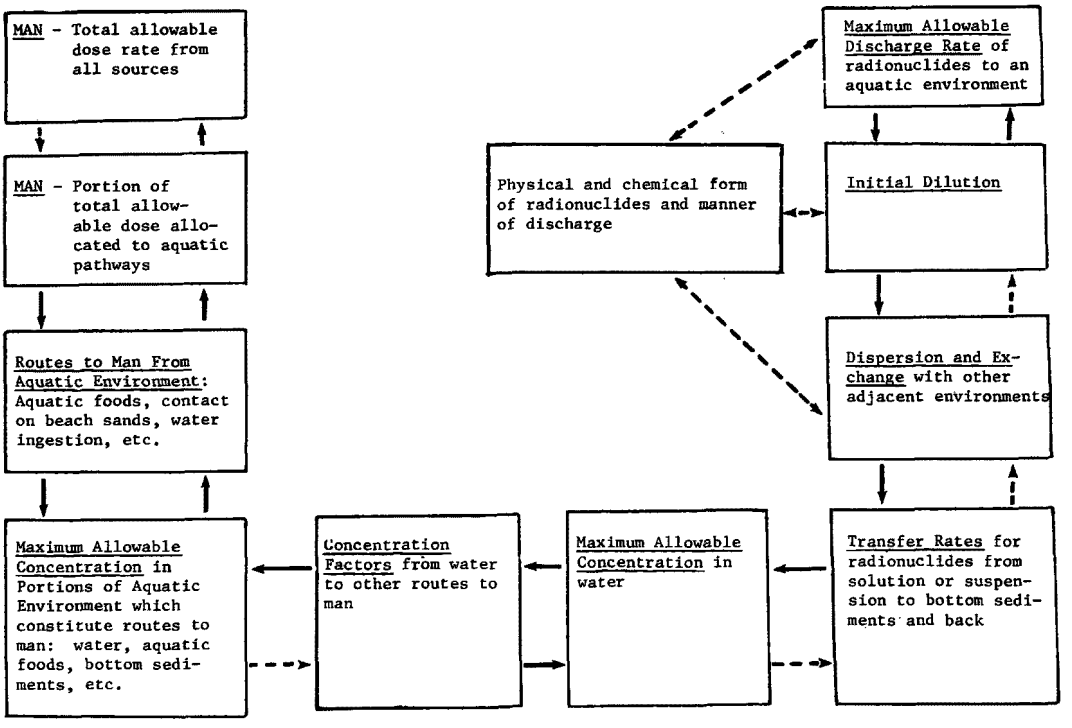

FIGURE 1. General steps in critical pathways evaluation for aquatic environments -- Solid arrows indicate direction of radionuclides, broken arrows that of calculations

Application of mixing rates, dilution factors, etc., to $(DWL)_{sw}$ allows an allowable rate of discharge to be calculated (Aten, 1961; Freke, 1967).

The rate of seafood ingestion, I in equation (4), has been estimated in various ways. These fall into two categories of data on food consumption patterns (Middleton, 1964). The first is a food balance sheet or estimate of the per capita supplies of various human foods available in a country, and the second is a survey of dietary habits for selected population groups. For example, in a typical U.S. diet it has been estimated (Harley, 1969) that marine food products contribute 0.01% of the radioisotope intake, while the contribution is 0.5% for a typical Japanese diet (Pritchard, 1961).

Marey and Saurov (1964) have developed relative indices which characterize the movement of an isotope from a water media into a food product. These indices, called accumulation multiples (AM), are very similar to the CF's discussed previously. An AM is calculated by dividing the activity per kilogram of the food product by the activity per liter of the water media. These factors, along with dietary information, can be used to establish the amount of a given isotope contributed to people by individual foods in their diet. For this purpose contribution coefficients (CC) were expressed:

$$CC = \frac{AM \cdot P}{V} \tag{6}$$

where:

AM = accumulation multiple for particular food

P = g/day intake of the food item

V = g/day intake of water

By summing the CC values for all food products and radioisotopes, a general CC is obtained which characterizes the ingestion by people of radioisotopes via aquatic foods in relation to the potential direct intake via the water media. This approach differs from the usual,

which utilizes CF's for edible aquatic organisms, in that radionuclides in actual food products are measured. The accumulation multiple is a form of CF, but only for processed food items. The contribution coefficient is a means to reduce accumulation multiple data for various foods and radionuclides to a form for intercomparison.

The amount of radioactivity in processed foods is of primary importance in determining the radiation dose to people (Harrison, 1972). Radionuclide losses may be significant during processing and marketing, e.g., from radioactive decay and the tissue selected for food. In addition, food transportation to areas remote from the harvest area leads to difficulties in accurate assessment of potential problems. World interest in the expanded use of fish protein concentrate (FPC) is growing. About three kilograms of whole fish yields about 500 grams of FPC, and the radionuclide (as well as other pollutants) content of the concentrate may be hazardous if it is used as a major dietary item. In one assessment (Beasley, 1971) based on a reasonable 10 gram per day intake of FPC, the ^{210}Pb-^{210}Po accumulation was found to be hazardous, the cobalt intake was increased by a factor of 2 to 3, and the silver intake was increased by about a factor of 1.3.

The consideration of radionuclide ingestion alone is useful in assessing pathways to man and relative hazards; however, the dose to man depends on many other factors such as summarized from ICRP reports by Harrison (1972). These factors include:

1. Quantity of radionuclides ingested.
2. Fraction of ingested quantity which is absorbed and deposited in tissues.
3. Energy absorbed by tissues.
4. Effective half-life of the radionuclide in tissue.

Since aquatic environments may become contaminated from a variety of sources (e.g., fallout, reactors, reprocessing plants, and nuclear ships), efforts have been made by Templeton (1964) to measure the relative contributions of these various sources of natural and artificial

radionuclides. These data can be used for establishing allowable discharge rates for the various sources. However, the establishment of source-oriented MPC_{sw} values from MPC_{sf} data would require both estimations of the seafood ingestion rates and distribution patterns and an arbitrary allocation of some fraction of the MPC_{sw} that could come from, for example, nuclear ships.

The NAS/NRC (1959) recognized that the MPC for aquatic environments was generally interpreted to be the MPC_w, and that this was generally sufficient when food organisms in question derive their radionuclides directly from water. Where the food organisms derive radionuclides from other sources (such as bottom debris or via food chains), then the CF's specific for biota of interest should be used in calculations. Usually such data are unknown.

Eventually, it may be possible to incorporate dynamic food chain models into the derivation of CF's. A simple food chain simulation model has been described by Eberhardt and Nakatani (1969) in which a tracer substance is released at an exponentially declining rate, and a fraction is transferred to the first compartment, a producer. Two hypothetical animals are included in this model, one with a single internal compartment and the other with two compartments in parallel. This type of compartmentalized food-chain model could be expanded to describe complex ecological webs. A similar source-pathway-receptor model is described by Reichle (1970).

Another possible refinement of CF determinations may occur if interspecies variability can be predicted. The uptake and retention of many substances is proportional to a fractional power of body weight. When this relation is extended to interspecies comparisons, the resulting power coefficient is much lower than that expected on the basis of relationships between size and metabolic rate. It was proposed by Thomas and Eberhardt (1969) that interspecies comparisons be based on "similarity ratios" that depend on the proportionality coefficient in the equation:

$$y = a\,W^{b} \tag{7}$$

where:

y = retention time

a = proportionality coefficient

W = body weight

b = 0.75

Eberhardt (1969) has successfully related the long-component half-life for ^{137}Cs in humans to body weight with a power coefficient of 0.66.

Further modifications to calculations can be made by either broadening or narrowing the scope of dietary assumptions. Rice (1963) points out that the organisms which concentrate a radionuclide to the highest degree are not necessarily the controlling factor in allowable radioactive waste disposal rates. Rather, the organisms which concentrate to the highest degree in relation to the amount ingested by man are critical. He has tabulated CF's for the following major organism classifications: algae, mollusca and fish. Freke (1967) used a similar approach, but included crustacea as a major food subdivision. In other concentration factor listings, Aten (1958 and 1961) considered only fish as critical, while Polikarpov (1966) lumped together "marine organisms" and "freshwater organisms".

In an International symposium report, Hiyama (1960) adds yet another suggestion by pointing out that since man eats numerous kinds and quantities of marine foods a derived working level should not be based merely on the CF for one food product. He recommends the use of a "seawater equivalent for daily human intake" (W_d). Alternatively, when the specific activity approach is used, he recommends a "seawater equivalent for the whole body" (W_b), or for the critical organ (W_{bc}). The following formulas are derived:

$$W_d = M \cdot I/S \text{ liters/day/person} \tag{8}$$

where:

I = average daily intake of element, g/day/person

M = fraction of I that originates from seawater marine organisms

S = concentration of that element in seawater, g/l

W_d = seawater equivalent for human intake

The ICRP (1959) listings for maximum permissible daily intake of certain radionuclides (MPDI in μCi/day/person) can be used to derive the maximum permissible concentrations for seawater (MPC_{sw}):

$$MPC_{sw} = MPDI/W_d \quad \mu Ci/l \tag{9}$$

If little is known about I, but the average amount of the element in the whole body or in the critical organ is known, then the following seawater equivalents can be derived:

$$W_b = M \cdot B/S \text{ liters} \tag{10}$$

or

$$W_{bc} = M \cdot B_c/S \text{ liters} \tag{11}$$

where:

W_b = seawater equivalent for the human body (liters)

W_{bc} = seawater equivalent for the critical organ (liters)

B = average amount of element in the human body (grams)

B_c = average amount of element in the critical organ (grams)

S = concentration of element in seawater (gram/liter)

M = fraction of B or B_c that originates from seawater or marine organisms

The ICRP (1959) listings for the maximum permissible amounts of radioisotopes in the total body (q, μCi) and in the critical organ (q_c, μCi) also can be used to derive a maximum permissible concentration for seawater (MPC_{sw}):

$$MPC_{sw} = q/W_b \quad \mu Ci/l \tag{12}$$

$$= q_c/W_{bc} \quad \mu Ci/l \tag{13}$$

Where radioactive contamination extends to terrestial food, any such MPC_{sw} must be reduced proportionally. If the isotopic dilution ratios are similar in both terrestrial foods and seafoods, then "M" is not required for the above equations.

So far, the foregoing discussion illustrates that the scope of the terms MPC_{sf} and MPC_{sw} can range from very generalized figures that, at best, give order of magnitude estimates, to more specific derived working limits for particular areas which may be used to guide waste disposal practices. An additional interpretation (Miller and Inclan-Suarez, 1970) of allowable discharge rates or MPC values has been applied at least once by a State regulatory agency with review authority for nuclear power plants. This interpretation sets MPC values as those attained by the lowest technologically-feasible point-source radioactive waste discharge.

It should be noted that MPC values are intended for limiting human exposure risks. Relatively little is known about the tolerance of aquatic biota to chronic radioactive contamination. There appears to be considerable species differences and variability, as well as significant variability within the same species in regard to different developmental stages and food intake. Between species it has been generally observed that for acute radiation doses the least specialized forms are also the most resistant.

There appears to be no generally applicable approach to describe the transport of radionuclides introduced into surface waters. Each stream, river, lake, bay, estuary, and sea has mixing characteristics unique in place and time. Moreover, introduced radionuclides can remain in solution or suspension, precipitate and settle on the bottom, or be concentrated by plants and animals (Rice, 1965). For those nuclides that remain in solution, mixing by physical processes of diffusion, turbulence, etc., is generally defined (Eisenbud, 1973) by a combination of theoretical calculations and measurements that are applicable only to the specific locality. An example of an investigation in which many factors were incorporated is the Delaware River/Estuary Study

(Parker *et al.*, 1961). Data on convection and diffusion were obtained from a scale model with tracer dyes. Calculations based on these data were modified for radioactive decay. Since they are somewhat concentration dependent, sorption and sedimentation reactions were expressed mathematically as exponential decay.

The marine environment has been subdivided descriptively by several investigators. From the viewpoint of characteristic physical and biological processes which may return radioactive materials to man, there are three major divisions (Schaefer, 1961; NAS/NRC, 1959): (1) near shore areas--harbors, estuaries, and coast out 2 miles from shore; (2) continental shelf area--subdivided into an inner shelf 2 to 12 miles from shore and an outer shelf from 12 miles to the 200 fathoms contour; and (3) open ocean--more than 12 miles from shore and 200 fathoms deep. The latter two areas can be subdivided further into commercial fishing and noncontributory areas.

Rice (1965) compares estuarine and oceanic habitats. He points out that radionuclides released in the open ocean tend to be rapidly diluted and dispersed, whereas in an estuary there is more chance of biological and physico-chemical concentration. Due to the shallow depth, sediment and the benthic community involved, radioisotope exchanges are relatively important in estuaries, but not in open seas. A third difference is that oceanic food chains are simple in comparison with those of an estuary.

Berglin (1960) has proposed a method for determining mixing in the Woronara Estuary, Australia, which may be useful for studying other estuarine situations. Typically, freshwater discharged to the estuary moves seawards by mass flow with turbulent mixing resulting from tidal motion. If the freshwater inflow rate is monitored, the mean dilution which can be expected within a particular section (e.g., 1/4 mile) can be determined by measuring the ratio of seawater to freshwater in the section. Then, multiplying the maximum acceptable activity levels in a section by the mean dilution factor gives the allowable mean input concentrations for the section. This method would have limited usefulness

in situations where freshwater overrides the saltwater or where freshwater plumes extend out to sea.

Pritchard (1960) reviews several methods of calculating mixing and transport in different aquatic habitats. Included are formulas to estimate dispersal in a continental shelf area so that a radionuclide concentration at an input area boundary can be calculated. In another model, the dispersal of activity from a deep-sea bottom is estimated using conservative estimates of the vertical diffusion and by neglecting the loss to sediment. The formulas he presents yield estimates of the amounts of activity which, if released annually in a deep-sea segment, would produce at the bottom of the ocean layer harvested by man an equilibrium concentration which would not exceed the allowable values for the isotopes involved.

One of the least known components of aquatic ecosystems is the exchange between water and bottom sediments. The NAS/NRC (1971) has even suggested that it may be impossible to generalize these sorption and sedimentation reactions. However, Lerman (1961) has developed a generalized sediment transport model which defines several mechanisms for entry of radioisotopes into the sediment. These are:

1. Deposition of suspended particles of inorganic or organic origin;
2. Diffusion from the overlying water into the sediment interstitial water, followed by adsorption; and
3. Production from parent radionuclides in sediments or release from decomposition of organic matter.

The effects of these various factors on the concentration of a radionuclide in sediment is summarized in a series of equations which relate these factors to the rate of change of the radionuclide concentration in interstitial water.

A paper by Reynolds (1963) on the sorption and release of radionuclides from sediments contained theoretical dispersion formulas based on a simple compartmental stream model. He developed an equation for a sediment distribution coefficient which was derived from a

mass-action equilibrium equation with the assumption that the exchanged ions were at very low concentrations.

A study of the Clinch River provides some surprising results in regard to river transport characteristics. It was found that sloughs did not have an appreciably greater buildup of radionuclides than the main river channel. Parker (1967) stated that "possibly the most important outcome of the Clinch River Study is the successful application of mass-balance techniques to entire river complexes". This study showed that the water-borne load of radionuclides was almost the entire amount discharged to the river. The sediment load was small (2 to 5% maximum); the maximum inventory possible in the biomass was exceedingly small and could be neglected. These results support earlier work reported by Polikarpov et al. (1966) in which it is postulated that the biomass may be neglected as a significant depot of activity in a reservoir or lake system. This is true, they concluded, even though the average CF's in the biomass and sediments may be 1,000 and 100, respectively, because the relative biomass is small as compared to other system components.

Armstrong and Gloyna (1967) developed a general equation to describe radionuclide transport in terms of hydraulic dispersion and convection in detention systems (compartments) which sorb and release. For non-conservative substances, such as radionuclides, a sink must be included to account for the uptake of material from solution. A generalized form of the equation for a radionuclide is:

$$\Delta C = D - V - U \qquad (14)$$

where:

ΔC = change in water concentration

D = dispersion term

V = convection term

U = uptake term

They point out that the uptake term is very poorly defined. It includes such variables as sorption of radionuclides by suspended solids, sediment and plants. They also proposed a general equation for uptake reactions in which the uptake is a summation of sorption on various substrates. The rates of

sorption and desorption are dependent on the concentration gradient between a transfer substrate and the sorbent. A linear transfer function is used to correlate sediment specific activity to that of water, although they point out the possibility of using an alternative nonlinear function. The study showed that suspended solids, sediment, and plants *(vallisneria)* sorb according to a nonlinear (Freundlich isotherm) function. However, attempts to use this equation in the reaction term of the one-dimensional (e.g., a river) dispersion equation forces it into a nonlinear form and makes analytical solution impossible. A by-product of the transport equation (using a linear transfer function) is the specific activity of radionuclides in the biological system. It is feasible to use these results to determine the passage (transfer coefficients) through aquatic food chains by using the general equation for uptake reactions.

Kaye and Ball (1967) proposed the application of systems analysis techniques to ecosystem models describing radionuclide transport. Their approach uses input and loss relationships which allow calculation of the radionuclide concentration in any environmental compartment. Two important kinds of required information are (1) an accurate compartmentalized representation of the environment under review; and (2) the rate constants which quantify the intercompartmental transfer of nuclides. This technique was developed into a comprehensive model for predicting transport of radionuclides from an underground nuclear explosion through the seawater--fish--man pathway by Bloom (1971). It is apparent that further modifications of this technique may allow the development of models to deal with chronic releases of radionuclides from various sources.

SPECIFIC ACTIVITY APPROACHES

The specific activity approach is attractively simple in concept and is very useful in assessing long-term problems concerning the radioisotopes of elements which (1) are relatively abundant in nature, (2) are rapidly dispersed throughout the environment, and (3) are normal constituents of organisms (e.g., ^{3}H, ^{14}C, ^{32}P). The NAS/NRC (1962)

in developing recommendations for ocean disposal of radioactive wastes stated the principle: ". . . if the specific activities (that is, the radioactive proportions of the elements) of the chemical elements in the sea in the environment of human food organisms are maintained below the allowable specific activities for those elements in the human body or human food, no person can obtain more than an allowable amount of radioactivity from the sea, regardless of his habits." For trace elements and those radioisotopes which are close chemical congeners of another element, the specific activity approach lacks consistency. A method recently developed at LLL (Pratt, 1970; Tamplin *et al.*, 1968 and 1969; and Thompson *et al.*, 1972) is based on specific activity and is similar to the NAS/NRC approach except that it relates to tissue dose rather than to critical organ dose.

The specific activity approach, as developed by the NAS/NRC (1962), was for calculation of derived working limits for radioactive waste disposal into Pacific coastal waters. The approach is based on two major assumptions:

1. that a radioisotope introduced into the environment readily equilibrates with the stable isotope(s) of the same element, and
2. that the quantity of each stable element in each body organ is fixed and does not fluctuate with the intake of that element.

It is known that the conditions of the first assumption are not always met. Known exceptions have been tritium, carbon, sulfur, vanadium, iron, cobalt, copper, and zinc. These elements may be introduced as stable organic complexes and, hence, are not diluted by the common abundant chemical form of the stable elements. The NAS/NRC (1962) recommends that for these elements the safety factor for modifying figures from occupational worker dose to general public dose be increased from a factor of 10 to 100.

The maximum permissible specific activity (MPSA) for any radioisotope with a critical organ other than the GI tract is readily available from data on standard man (ICRP, 1959).

$$MPSA = q/mC \quad \mu Ci/g \tag{15}$$

where:

MPSA = maximum permissible specific activity ($\mu Ci/g$)

q = maximum permissible burden of radioisotope in critical organ (μCi)

m = mass of the critical organ (g)

C = concentration of stable element in critical organ (g/g)

The MPSA can then be converted to derived working limits for a media. For example, the values for seawater (DWL_{sw}) or seafood (DWL_{sf}) are calculated (NAS/NRS, 1962):

$$DWL_{sw} = K_w \text{ (MPSA)} \quad \mu Ci/cm^3 \text{ seawater} \tag{16}$$

$$DWL_{sf} = K_f \text{ (MPSA)} \quad \mu Ci/g \text{ seafood} \tag{17}$$

where:

K_w = grams of stable element/cm^3 seawater

K_f = grams of stable element/g seafood

A DWL derived in this fashion is independent of CF's and the consumption rates for seafoods.

For those isotopes that are only slightly absorbed by the body and mainly affect the gastrointestinal tract, the MPC in water is converted to a MPSA of the stable species in seafood by utilizing the abundance of that element in seafood organisms which results in the most restrictive requirement. Although developed specifically for pathways from the sea, this approach can be extended to freshwater and terrestrial environmental pathways (NAS/NRC, 1962, and Bryant, 1970).

An alternate method has been proposed by the NAS/NRC (1959) for computing allowable environmental levels of certain radionuclides for which a known human discrimation factor exists. For example, the ratio of ^{90}Sr to calcium in the total body should not exceed 0.1 $\mu Ci/kg$. Man physiologically discriminates against strontium in a strontium-calcium mixture by a ratio of 8:1. If man receives his total protein allowance from fish which contain ^{90}Sr, then the MPC for fish is

defined as 0.8 μCi of ^{90}Sr/kg calcium. Further, since the calcium concentration of seawater is about 0.4 g/kg seawater, the seawater MPC is about 0.8 μCi/2,500 kg or about 3 X 10^{-2} μCi/ml of seawater. The occupational MPC for continuous exposure in drinking water is 10^{-10} μCi/ml (ICRP, 1959).

Odum (1963) has extended this idea of using the known ratio of a radionuclide to a stable physiological element in a theoretical "element ratio method". Basically, he proposes that if the ratios of the minor elements to carbon are known, then the measurement of carbon metabolism can be used to predict cycling of the minor elements.

Bloom (1971) suggests that two screening operations be performed before detailed calculations are made. The first uses a simple two-compartment specific-activity model in which the specific activity of each radionuclide in man is assumed to be the same as in the environmental sink before dilution. Then, using ICRP data on radionuclides, standard man, etc., the infinite-time internal dose is calculated for (1) the gastrointestinal tract and (2) all other organs. For radioisotopes found potentially important in this initial screening, he recommends an eight-compartment transport model which requires source term data and transfer coefficients for the radionuclides identified in screening. Solution of a complex set of equations provides estimates of the radionuclide concentration in each of the eight compartments as well as of the internal radiation dose to man as a function of time.

A modified specific activity method for determining safe discharge rates for radionuclides into aquatic systems is being developed at the Lawrence Livermore Laboratory (Chapman *et al.*, 1968; Harrison, 1972; Tamplin, 1967, 1968, 1969; Burton, 1968; Ng *et al.*, 1966, 1968; Thompson *et al.*, 1972). The approach is similar to that of the NAS/NRC (1962), except it uses a tissue dose concept rather than a critical organ dose concept. The method approaches environmental contamination by radionuclides and the resultant radiation dose to man from the standpoint of the specific activities of ingested elements; this is also known as the biological

exchangeable pool of elements concept (Ng and Thompson, 1966). The passage of a radionuclide through the biosphere is presumed to be governed by the same factors that govern the distribution of the related stable element isotopes within the biological exchangeable pool. It is also presumed that the radionuclide is biologically no more (or less) available than the related stable isotopes within the environment.

The basic product of this effort is a value for each isotope called the unit-rad contamination in water (F_A) in $\mu Ci/m^3/rad$. Values have been derived for freshwater and seawater. This factor is defined as the initial concentration of a radioisotope in water which would yield a 30-year integrated dose of 1 rad to a specific designated tissue of standard man. In most cases F_A values for infants are also estimated, since values for adults are usually less restrictive than for infants. The values for F_A are derived with several simplifying assumptions, almost the same as those used by the NAS/NRC (1962), which must be clearly understood in applying the F_A factors to environmental data. These assumptions yield very conservative values for F_A, for the worst situation that could develop, and are designed in this manner so that basic information on the environment and population at risk can be used to develop modifying factors. The simplifying assumptions include:

1. Man exists on a diet of totally aquatic origin, and
2. Initial water concentrations decrease only through radioactive decay. (Dilution of the system by uncontaminated water and dilution beyond the area of initial rapid mixing are not accounted for.)

The basic calculation process is, briefly, to start with the ppm values for stable elements in seawater and freshwater and to calculate an uptake by man from various routes. CF's derived from the literature for items in man's food chain are used to obtain stable isotope intake. Various data, e.g., terrestrial and aquatic, are evaluated to obtain the most critical pathways for calculating F_A. The introduction of radioisotopes into aquatic environments and the subsequent intake by man are evaluated on the basis of stable element pathways. The

radiation dose to man from radioisotopes is calculated using GI uptake fractions, daily ingestion amounts, energy absorbed in tissue per disintegration, effective half-lives in the environmental media and man's tissue, distribution in man's tissues, etc. In a LLL report (Ng *et al.*, 1968), all of these input parameters for calculations are listed for radionuclides with half-lives greater than 12 hours. The derived values for F_A are also listed, and some methods are given to modify these basic values to those more representative of a given situation. For example, the calculations are described for obtaining modified F_A values for a population on a mixed aquatic and terrestrial diet. The modified values obtained include corrections for both the dietary mix and the ratio of isotope concentrations in aquatic and terrestrial foods.

The estimated maximum radiation dose to a tissue (EDA) from a particular isotope is obtained by dividing the aquatic environment concentration by F_A as follows:

$$EDA = ECA/F_A \text{ rad}$$

where:

EDA = 30-year integrated dose to a specific tissue of standard man (or infant when designated value of F_A is used) from an aquatic diet and for a specific isotope

ECA = contamination level of the aquatic environment in $\mu Ci/m^3$

F_A = unit-rad contamination factor in $\mu Ci/m^3/rad$

The total dose to a specific tissue is the sum of the doses from individual radionuclides.

Pratt (1970) has used much of the above information in deriving unit-dose-rate water concentration values. These are water concentration values for isotopes which could yield an equilibrium dose rate of 1 rad per year to an adult through aquatic foods. Values are also derived for infants. Since these concentrations can be scaled to appropriate maximum allowable dose rates, the maximum allowable concentrations in an aquatic environment can be calculated. The maximum allowable rates of release can

then be calculated using appropriate dilution and dispersion factors. The maximum allowable dose to people through aquatic foods still must be designated in this approach if firm regulatory guides are to be promulgated. In general, regulatory bodies have avoided fragmenting the total allowable dose into portions related to specific exposure routes. The basic criteria is dose from all sources, and the fractional contribution from a specific route varies with each particular situation.

In general, the specific activity approach gives less stringent standards for radioactive waste disposal in seawater than are derived through critical pathways models. The latter tend to be more restrictive, apparently because conservative values are generally used for unknown concentration factors. The specific activity approach has the following limitations (Wolfe and Rice, 1968):

1. It is not valid when considering radiation dose to the gastrointestinal (GI) tract.
2. As the maximum permissible body burden for radioactivity is approached in humans, the total activity in the GI tract becomes significant, regardless of what the specific activity may be.
3. Radionuclides introduced into the environment may be more (or less) readily available for bioaccumulators than the corresponding stable elements.

In the final transition of adapting allowable dose rates into maximum permissible concentrations for radionuclides in air, food, and water, and subsequently into directives for operating practice, there is practical value in an easily understood index of hazard. Such an index has been proposed by Rohwer and Struxness (1972) and is termed a "cumulative exposure index". This is a numerical guide indicating relative significance (dose estimate/dose limit) of measured environmental radioactivity on the basis of the total dose to man from all radionuclides and exposure modes of importance.

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TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA-600/3-76-054		

4. TITLE AND SUBTITLE	5. REPORT DATE
CONCENTRATION FACTORS AND TRANSPORT MODELS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS A Literature Report	May 1976
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
R. G. Patzer	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Environmental Monitoring and Support Laboratory Office of Research and Development P. O. Box 15027 Las Vegas, NV 89114	1FA083
	11. CONTRACT/GRANT NO.
	n/a
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Same as above	Final - FY75
	14. SPONSORING AGENCY CODE
	EPA-ORD, Office of Health and Ecological Effects

15. SUPPLEMENTARY NOTES

16. ABSTRACT

The relative risks to man from radionuclides released to the environment depend heavily on their accumulation or concentration by aquatic organisms. The organisms which accumulate those radionuclides present in the environment may be useful as indicators for environmental monitoring purposes. In addition, these organisms may be directly in food chain pathways to humans.

Literature is reviewed and summarized in regard to biological concentration of radionuclides in freshwater and marine environments. Concentration factors for elements found in organisms are tabulated for plants, invertebrates, and fish in marine and freshwater environs. Literature is also reveiwed on models developed to calculate the possible radiation dose delivered to humans from radionuclides released into aquatic environments. The model approaches summarized range from simple generalized forms which, at best, give order of magnitude estimates to detailed models for a specific area which may be used to guide waste discharge practices.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Concentration	Biological concentration	06R
Isotopes*	Concentration factors*	07E
Mathematical Models*	Transport models*	08H
Radiation Monitors		18H
Radiation Protection		

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	64
	20. SECURITY CLASS *(This page)*	22. PRICE
	UNCLASSIFIED	

EPA Form 2220-1 (9-73)

☆GPO 691-217-1976