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# Produced Water Radionuclide Hazard/Risk Assessment Phase I

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## Produced Water Radionuclide Hazard/Risk Assessment — Phase I

## Health and Environmental Sciences Department

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#### ABSTRACT

Petroleum production may be accompanied by the production of saline water, called "produced water". Produced water discharged into freshwater streams, estuaries, coastal and outer continental shelf waters can contain enhanced levels This document reports on the first phase of a study to of radium isotopes. estimate the risk to human health and the environment from radium discharged in produced water. The study involved five major steps: 1) evaluate the usefulness of available produced water outfall data for developing estimates of radium environmental concentrations: 2) review the literature on the bioaccumulation of radium by aquatic organisms; 3) review the literature on the effects of radiation on aquatic organisms; 4) review the information available concerning the human health risks associated with exposure to Ra-226 and Ra-228 and 5) perform a conservative, screening-level assessment of the health and environmental risks posed by Ra-226 and Ra-228 discharged in produced waters. A screening-level analysis was performed to determine whether radium discharged to coastal Louisiana in produced waters presents potential health or environmental risks requiring further study. This conservative assessment suggested that no detectable impact on populations of fish, molluscs or crustaceans from radium discharged in produced waters is likely. The analysis also suggested that there is a potential for risk were an individual to ingest a large amount of seafood harvested near a produced water discharge point over a lifetime. The number of excess cancers predicted per year under a conservative scenario is comparable to those expected to result from background concentrations of radium.

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#### EXECUTIVE SUMMARY

#### Introduction

Petroleum production may be accompanied by the production of saline water, called "produced water". Produced water discharged into fresh water streams, estuarine, coastal and outer continental shelf waters can contain enhanced levels of radium isotopes. This document reports on the first phase of a study to estimate the risk to human health and the environment from radium isotopes discharged in produced water. The study involved five major steps:

- evaluate the usefulness of available outfall data for developing estimates of radium environmental concentrations;
- review the literature on the bioaccumulation of radium by aquatic organisms;
- review the literature on the effects of radiation on aquatic organisms;
- 4) review the information available concerning the human health risks associated with exposure to Ra-226 and Ra-228 and
- 5) perform a conservative, screening-level assessment of the health and environmental risks posed by Ra-226 and Ra-228 discharged in produced waters.

In addition to reviewing the relevant literature and collecting the available data needed to perform a risk assessment, a screening-level assessment was performed. The results of this screening-level analysis will be used to determine whether radium in produced waters presents a potential health or environmental risk requiring further study.

In this screening-level analysis, exposures to aquatic biota and to man from Ra-226 and Ra-228 were estimated for background concentrations of 0.1 and 1.0 pCi/l, produced water discharge concentrations of 30, 500 and 2,000 pCi/l, and for the levels measured in organisms in the Continental Shelf Associates study (CSA, 1991). Doses to fish, molluscs and crustaceans were estimated as described in

IAEA (1976). The major pathway resulting in exposure to man is expected to be the consumption of contaminated fish and shellfish. Maximum individual risks to people consuming fish and shellfish harvested near a produced water outfall were estimated using USEPA risk factors for a range of water concentrations, concentration factors and intake rates. The number of excess cancers expected from the ingestion of radium in fish and shellfish was estimated using simple conservative assumptions.

#### Chemistry and Fate of Radium in Produced Water

When a produced water is discharged into a body of water, the resulting distribution of radium is controlled by a number of physical and chemical processes. The most important of these processes are mixing and dilution by turbulence, advection and dispersion; adsorption/desorption interactions with sediments and suspended solids, and coprecipitation of soluble salts. Radium discharged to nearshore, low energy environments will not be diluted as rapidly as offshore discharges. Radium in water exists primarily as the divalent ion Ra and has chemical properties similar to calcium, barium and strontium.

Field data and model simulations demonstrate that rapid dilution occurs in both nearshore and offshore environments, with dilution factors of 30 - 1500 within 50 - 100 feet of an outfall. Offshore discharges are diluted more rapidly than nearshore discharges.

#### Concentration Factors

Concentration factors are commonly used in dose assessments to estimate the levels of radionuclides in aquatic organisms. The concentration factor (CF) is a function of the concentration in the water or sediment (C) and the equilibrium concentration (on a wet weight basis) in the organism (Q).

It is usually assumed that there is a linear relationship between C and Q and that the concentration factor is independent of the

concentration in the environment. Concentration factors can be used to estimate the concentration of radium in aquatic organisms, based on the concentration in water. These factors are affected by many variables, including the species and the concentration of radium in water.

The concentration factors commonly used in dose assessment studies (IAEA, 1982) are appropriate when water concentrations are low, but are probably over-estimates for the relatively high concentrations that occur near produced water outfalls. Concentration Factors derived from the CSA data set (CSA, 1991) were smaller than the generic factors commonly used in assessments.

## Screening-Level Analysis for Effects on Aquatic Biota

In this screening-level assessment, conservative assumptions were made to develop estimates of dose to aquatic animals resulting from internal and external exposure to radium discharged in produced waters. This analysis used discharge and dilution data typical of nearshore coastal discharges. Offshore discharges will result in smaller doses because of the increased dilution and reduced chance for uptake by aquatic organisms.

Dose estimates were calculated for fish, molluscs and crustaceans using the simple models described in IAEA (1976). Estimates were calculated for two scenarios:

- 1) Background water concentrations of 0.1 and 1.0 pCi/1 Ra-226 and Ra-228; discharge concentrations of 30, 500 and 2,000 pCi/1 Ra-226 and Ra-228, a dilution factor of 100; and conservative, generic IAEA concentration factors.
- 2) The water, sediment and organism concentrations measured at the three sites in the Continental Shelf Associates study (CSA, 1991).

Even using conservative assumptions (IAEA concentration factors, IAEA, 1982), the estimated dose rates were below those expected to result in deleterious effects (1-10 mGy/day [.1-1 rad/day] for individuals, >10 mGy/day [1 rad/day] for natural populations).

This conservative, screening-level assessment of the risk presented by radium discharged in Louisiana coastal waters suggested that no detectable impact on fish, molluscs or crustaceans is likely. Offshore discharges will be diluted to a greater extent than nearshore discharges, and the chance for exposure of aquatic organisms to radium from produced waters will be smaller. No impact to aquatic biota from offshore discharges of radium is likely.

## Screening-Level Analysis for Effects on People

In this screening-level assessment, conservative assumptions were made to develop estimates of individual lifetime risk resulting from ingestion of radium from produced waters. This analysis used discharge and dilution data typical of nearshore coastal discharges. Offshore discharges will result in smaller risks because of the increased dilution and reduced chance for uptake by aquatic organisms.

The conservative EPA risk factors were used in this assessment, and it was assumed that half of an individual's seafood consumption comes from animals harvested near a produced water outfall. Intake levels used in the analysis included those for the individual eating the most seafood.

Estimates were made for actual site data (CSA, 1991), for two potential background concentrations of Ra-226 and Ra-228 (0.1, 1.0 pCi/l), and for three potential discharge scenarios (30, 500 and 2,000 pCi/l Ra-226 and Ra-228), using IAEA concentration factors and a dilution factor of 100. The potential scenarios were included in the assessment because data was available only for three impacted sites, and the range of water concentrations for these sites did not cover the range of concentrations encountered in produced water. The calculations based on 0.1 and 1 pCi/l also allowed an estimate of the potential risks presented by background radium levels.

The risk estimates were based on the following assumptions:

- 1. Radium discharged in nearshore produced water is reduced by a factor of 100 before coming in contact with fish and shellfish consumed by people.
- 2. An individual gets one-half of their yearly fish and shellfish from near (i.e. where the dilution factor reaches 100; probably within 100 feet) a nearshore produced water outfall for their entire lifetime.
- 3. The use of conservative IAEA concentration factors in estimating the concentration of radium in fish and shellfish.
- 4. The use of conservative EPA risk factors.

This conservative, screening-level assessment of the risk presented by radium discharged to Louisiana coastal waters suggested the potential for a level of risk that could be considered significant (greater than 1 x 10<sup>-5</sup>) for 1) an individual who ingests a large amount of seafood harvested near (where the dilution factor reaches 100) a nearshore produced water discharge point over his lifetime and 2) a person consuming an average amount of seafood (over a lifetime) harvested near a nearshore produced water outfall discharging a large amount of radium (500 pCi/1 Ra-226 and 500 pCi/1 Ra-228). The number of excess cancers predicted per year is comparable to the number expected to result from background concentrations of radium. It should be emphasized that the screening-level analysis presented here is a simple, conservative analysis that will necessarily overestimate the risks associated with the discharge of radium in produced water.

Because of the many conservative assumptions incorporated into this screening-level analysis it can be concluded that the risk associated with the discharge of produced water to coastal Louisiana is small. The results of this study do, however, support the need for a more detailed analysis of the potential risk to humans consuming seafood harvested close to nearshore produced water discharge points.

Radium discharged offshore will be diluted more rapidly than radium discharged to nearshore waters. Organisms living offshore will have a smaller chance of coming into contact with discharged radium because of the large water volumes involved and the rapid dilution that occurs. A person is also unlikely to harvest a significant amount of his yearly seafood close to an offshore outfall. Because of the additional reductions in the radium concentration in water and aquatic biota expected near offshore outfalls as compared to nearshore discharges, it can be concluded that the risks associated with offshore discharges will be extremely small.

## Uncertainties and Conservatisms

The major uncertainties and conservatisms in this screening-level analysis are:

1. The concentration of radium in water and the geographic distribution of contaminated shellfish. The analysis of individual risk and dose to aquatic biota used concentrations likely to be measured near a produced water discharge, assuming a dilution factor of 100. Considerable dilution occurs with distance from the discharge point, and offshore discharges will be diluted faster than nearshore discharges. A reduction in fish and shellfish concentration will also occur with increasing distance from an outfall. The concentration of radium in the water in which fish and shellfish are harvested is critical to the estimation of risk.

For the calculation of population risk, a simple box model was used that assumed complete mixing of all discharged radium, and a resultant "average" concentration of radium in fish and shellfish harvested from the region. In fact, radium concentrations in water and in fish and shellfish are variable over the area.

- 2. The concentration factor used in calculating the concentration of radium in fish, molluscs and crustaceans. Commonly used concentration factors (IAEA, 1982) are higher than those derived from the CSA data set. The concentration factor used in the analysis has a large effect on the resulting risk estimates. The IAEA concentration factors do not consider the effect of varying water concentrations on the extent of uptake, or the fact that radium tends to concentrate in the inedible portions of fish and shellfish (bone, shell, exoskeleton).

  Conservative IAEA concentration factors were used in this assessment, which probably resulted in an overestimate of the concentration of radium in food.
- 3. The distribution of intake rates and the percent of consumed fish and shellfish that is radium contaminated. This analysis assumed that the maximally exposed individual harvested one-half of his seafood from near a produced water outfall. The distribution of seafood intake among the population, and the percentage of the seafood consumed that is contaminated with radium is uncertain.
- 4. The risk factors for radium. This screening-level analysis used the conservative EPA risk factors for Ra-226 and Ra-228. Central estimates of the risk factors would predict smaller risks and fewer cancers associated with produced water discharges.

Based on the results of this screening-level assessment, a more comprehensive analysis can be performed to produce more realistic estimates of health and environmental risk.

# Section 1 INTRODUCTION

Petroleum production may be accompanied by the production of saline water, called "produced water". Produced water discharged into fresh water streams, estuarine, coastal and outer continental shelf waters can contain enhanced levels of radium isotopes. This document reports on the first phase of a study to estimate the risk to human health and the environment from radium isotopes discharged in produced water. The study involved five major steps:

- 1) evaluate the usefulness of available data for developing estimates of environmental radium concentrations resulting from produced water discharges to coastal and offshore Louisiana;
- 2) review the literature on the bioaccumulation of radium by aquatic organisms;
- 3) review the literature on the effects of radiation on aquatic organisms;
- 4) review the information available concerning the human health risks associated with exposure to Ra-226 and Ra-228 and
- 5) perform a screening-level assessment of the health and environmental risks posed by Ra-226 and Ra-228 discharged to coastal Louisiana in produced waters.

A screening-level assessment was needed to determine whether radium in produced waters represents a potential health risk which warrants more detailed study, and to identify gaps in required data and information. Information and data gathered in this Phase I screening study also provide the basis for the exposure and risk estimates that would be needed in a more detailed analysis of outfalls in Louisiana and for assessments in other regions of the United States. It should be emphasized that the screening-level analysis presented here is a simple, conservative analysis that will necessarily overestimate the risks associated with the discharge of radium in produced water.

A review of the risk assessment process, and the steps required in performing a risk assessment study for radium discharged in produced water is described in Section 2. The fate of radium discharged into surface waters is reviewed in Section 3, and the data available to estimate source terms and resulting environmental concentrations of radium are described in Section 4. Information available regarding the concentration factor of radium in fish, molluscs and crustaceans is discussed in Section 5. Section 6 reviews the literature on the effects of radiation on aquatic biota to support an assessment of the potential environmental effects resulting from discharges of radium. The data and models used to estimate the human health effects of radium-226 and radium-228 are critically reviewed in Section 7.

In addition to reviewing the relevant', literature and collecting data needed to perform a risk assessment, a screening-level assessment was performed. The results of this screening-level analysis will be used to determine whether radium in produced waters presents a potential health or environmental risk requiring further study. These analyses are presented in Sections 8 and 9.

Appendix A gives a brief discussion of the quantities and units used in the measurement of radionuclide activity and dose.

# Section 2 RISK ASSESSMENT AND SCREENING-LEVEL ANALYSIS

#### 2.1 BACKGROUND

#### Risk

There are several definitions of risk. In the risk analysis literature, risk is often defined as the possibility of suffering harm from a hazard (Cohrssen and Covello, 1989). An analysis of risk describes (1) a hazard; (2) the event or events that create the possibility of harm; and (3) an estimate of the likelihood that the harm will occur (Cohrssen and Covello, 1989).

#### Risk Assessment

The goal of a risk assessment study is to estimate the relationship between the source term and the potential resulting effects on human health and the environment. The process follows the discharge of a pollutant through its transport in air, soil, water and food to man. Health and environmental risks are then calculated based on data and models that relate exposures to risk (Till and Meyer, 1983).

The risk assessment process consists of four major steps (Cohrssen and Covello, 1989):

- 1. Source/Release Assessment: Estimate the amount, rate and location of the contaminant's release to the environment.
- 2. Exposure Assessment: Identify the populations or ecosystems at risk, estimate concentrations of the contaminant at exposure points, and determine the duration and mode of exposure.

- 3. Dose-Response Assessment: Determine the dose received by the exposed populations and estimate the relationship between different doses and the magnitude of their effects.
- 4. Risk Characterization: Integrate the data and information derived from the previous steps into estimates of risk.

#### Risk Estimates

A risk estimate is an estimate of the likelihood, or statistical probability, that harm will occur as a result of exposure to the risk agent (Cohrssen and Covello, 1989). Risk estimates are the major outcome of a risk assessment. For carcinogens, a risk estimate describes the probability of cancer associated with an exposure. For non-carcinogens, the risk estimate describes the probability of acute or chronic toxic effects.

There are several measures that can be used to describe the probability that harm will result from exposure to a risk agent. These include (Cohrssen and Covello, 1989):

- 1. Individual lifetime risk: The increase in probability that an individual will experience a specific adverse effect as a result of a continuous lifetime exposure to the risk agent.
- 2. Population risk: The number of cases resulting from one year of exposure, or the number of cases occurring in one year. It is usually calculated as individual risk times the number of people exposed.

#### Risk Management and Regulation

Risk management uses the information developed in a risk assessment along with information about resources and social and economic values to determine what action to take to reduce or eliminate risk (Cohrssen and Covello, 1989). Agencies responsible for promulgating regulations and standards to protect the public from environmental pollutants are engaged in risk management.

In an analysis of the use of cancer risk estimates in the Federal regulatory process (132 regulatory decisions), Travis et al. (1987) found a consistent pattern in the level of cancer risk that triggers regulation. Every chemical with an individual lifetime risk above 4 x  $10^{-3}$  was regulated. Except for one FDA decision, no action was taken to reduce individual lifetime risk levels that were below 1 x  $10^{-6}$ .

It is generally agreed that a one-in-a-million (1  $\times$  10<sup>-6</sup>) lifetime risk is an acceptable rate for a widely distributed carcinogen when the population at risk is very large. It is also generally accepted that for smaller populations a higher rate of individual risk is acceptable (Milvy, 1986).

In the analysis by Travis et al. (1987), regulatory agencies always acted to reduce risks in small populations when the individual lifetime risk was approximately  $4 \times 10^{-3}$ , and always acted to reduce risk in large populations (populations the size of the entire United States) when the individual lifetime risk was about  $3 \times 10^{-4}$ . For small populations, regulatory action was never taken for lifetime individual risk levels below  $1 \times 10^{-4}$ . For large populations the level of acceptable risk dropped to  $1 \times 10^{-6}$ .

Regulatory decisions not to regulate were usually based on insufficient population risk (Travis et al., 1987). Other factors included a lack of available control technology. In the area between the level of risk that always triggered regulation and the level of risk that never triggered regulation, cost-effectiveness was the primary determinant of regulation. In this region, substances with risk reduction costs of less than \$2 million per life were regulated, and substances that cost more were not regulated (unless they were above the level always acted upon).

### Risk Perception and Risk Communication

People perceive risks differently. Most people do not judge the risks presented by a hazard solely on the likelihood of its presenting adverse effects. Other factors are important in the way people define and perceive risks. Public fears are often not well-correlated with expert assessments. In the past, this discrepancy was seen as perceptual distortion on the part of the public, but the risk assessment community has begun to accept that the concept of "risk" means more than the probability of harm (as defined above). The following list identifies some of the characteristics other than mortality that factor into people's perception of risk (Sandman, 1986).

#### LESS RISKY

voluntary
familiar
controllable
controlled by self
fair
not memorable
no dread
chronic
diffuse in time/space
nonfatal
immediate
natural
individual mitigation
possible

## RISKY

involuntary
unfamiliar
uncontrollable
controlled by others
unfair
memorable
dread
acute
focused in time/space
fatal
delayed
artificial
individual mitigation
not possible

When explaining the risk associated with a hazard, these factors must be considered and acknowledged rather than dismissed as irrational. A common way to describe the risks associated with a hazard is by comparison with other, better known risks. However, because risk means more than simply the probability of harm, comparing a voluntary risk like smoking to an involuntary risk like living next to a nuclear power plant is likely to provoke anger and mistrust. People are also angered when the risk comparisons are used not just to explain how large risks are, but to minimize the magnitude of a hazard or to dictate what level of risk should be acceptable (e.g. the risk of living next to a nuclear

power plant is much less than the risk of smoking; since you choose to smoke you must also find the risk of living next to a nuclear power plant acceptable). Covello et al. (1988) developed a manual advising plant managers on how to present risk comparisons.

## Risks in Context -- Some Comparisons

The one-in-a-million  $(10^{-6})$  lifetime risk that is generally considered acceptable for a large population at risk is commonly used as a point of comparison in risk assessment studies. A  $10^{-6}$  lifetime risk of cancer for the entire United States population (243.4 million in 1987; Department of Health and Human Services, 1990) would result in 243.4 premature cancer deaths over a lifetime (70 years), or 3.5 deaths per year. These excess deaths are in addition to the 2.1 million deaths from all causes and the 477,000 cancer deaths that occurred in the United States in 1987 (Department of Health and Human Services, 1990). A 1 x  $10^{-4}$  lifetime risk for the entire population of the United States would result in 347 excess cancer deaths per year.

Table 2-1 gives the annual risk of death in the United States for a number of causes. This table is modified from Covello et al. (1988).

#### 2.2 RISK ASSESSMENT FOR RADIUM IN PRODUCED WATER

To perform a risk assessment for radium in produced water the major transport pathways, exposure routes and receptors of concern must be identified. Figure 2-1 summarizes the potentially important transport pathways and exposure routes for the two types of receptors that may be at risk from radium discharged in produced water -- people and aquatic biota.

The major steps in the assessment process for radium in produced water are outlined in Figure 2-2. The first step involves development

Table 2-1. Annual Risk of Death in the United States.\*

Hazard	Total Number of Deaths	Risk per Million Persons	Risk
All causes	1,973,003	0 0000	10.3
Heart Disease	757,075	3400.0	3.4 v 10 <sup>-3</sup>
Cancer	351,055	1600.0	1.6 × 10 <sup>-3</sup>
Motor vehicle accidents	46,200	210.0	2.1 x 10 <sup>-4</sup>
Work accidents	13,400	150.0	$1.5 \times 10^{-4}$
Homicides	20,465	93.0	$9.3 \times 10^{-5}$
Falls	16,300	74.0	7.4 × 10 <sup>-5</sup>
Drowning	8,100	37.0	$3.7 \times 10^{-5}$
Fires, burns		30.0	3.0 × 10 <sup>-5</sup>
Poisoning by solids or liquids		17.0	$1.7 \times 10^{-5}$
Suffocation, ingested objects	2,900	13.0	$1.3 \times 10^{-5}$
Firearms, sporting	2,400	11.0	$1.1 \times 10^{-5}$
Railroads	1,989	6.0	$9.0 \times 10^{-7}$
Civil aviation	1,757	8.0	$8.0 \times 10^{-7}$
Water Transport	1,725	0.7	$7.0 \times 10^{-7}$
Poisoning by gases	1,700	0.7	
Pleasure boating	1,446	9.0	7-01 × 0 9
Lightning	124	0.5	
Hurricanes	93	0.4	4.0 x 10 <sup>-7</sup>
Tornadoes	16	0.4	$4.0 \times 10^{-7}$
Bites and Stings	87	0.2	$2.0 \times 10^{-7}$
			9

\* Table modified from Covello et al. (1988); original in Atallah (1980).

adeorption/

Sediment

contact

uptake

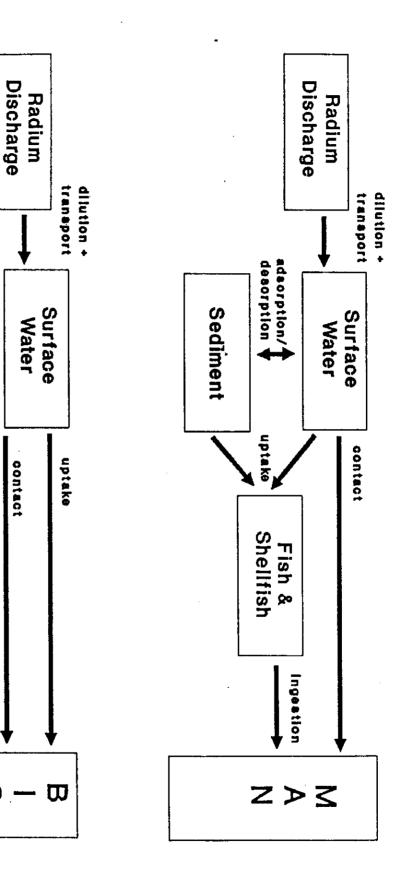
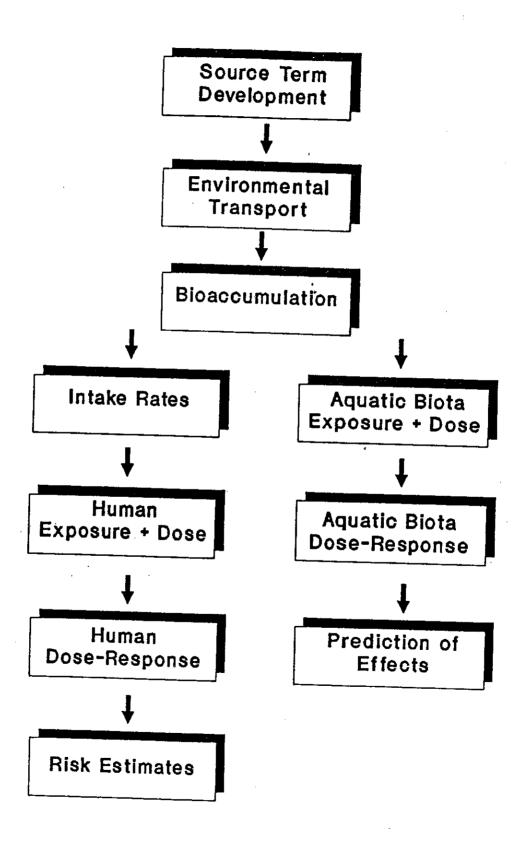


Figure 2-1. Major Exposure Pathways From Radium Discharged in Produced Water.

Figure 2-2. Steps in the Assessment Process.



of the source term. The quantity of radium discharged in produced water per unit time must be estimated.

The second step is to predict the fate of the discharged radium in the environment. This step involves use of models to simulate the physical transport of radium in the environment. Application of environmental transport models can produce an estimate of the concentration of the radionuclide which reaches people directly through secondary contact with water, or indirectly through consumption of contaminated fish and shellfish.

The third step in the process is to estimate the extent to which radium accumulates in fish and shellfish. The goal is to predict the concentration of radium in the edible portions of aquatic foods in terms of activity per unit mass. The fourth step involves development of intake rates for affected foods. To estimate exposures, the amount of contaminated food consumed must be estimated.

The next step is to calculate the absorbed dose associated with various intake rates for the radionuclide. The resulting risk to an individual or to a population can then be estimated through application of risk factors derived from models and epidemiological data. Because potential effects on aquatic biota are of concern, the concentrations of contaminants to which organisms are exposed must be estimated (both internal and external) and combined with available dose response information to predict potential effects.

A risk assessment for radium discharged in produced waters must be completed at two levels of possible impact. The impact of individual outfalls (for example, the outfall with the largest rate of radium discharge, or the outfall closest to a leased shellfishing area) and the potential risk to the individual harvesting fish and shellfish nearby must be assessed following the steps outlined above. To estimate population risks, the impact on the whole region must also be assessed.

## 2.3 SCREENING-LEVEL ASSESSMENT

This report represents the first phase of a risk assessment for radium discharged in produced waters. In addition to reviewing the relevant literature and collecting the data needed to perform a risk assessment, an initial screening-level risk assessment was performed.

In a screening-level analysis, complex environmental transport and dosimetric models are not necessary or appropriate. Simple models are used, along with conservative assumptions, to produce estimates of dose and risk. The results of this screening-level analysis are used to determine whether radium in produced waters presents a potential health or environmental risk requiring further study. The analysis presented in this report does not use state-of-the art'models or analytical techniques, but should be viewed as a screening exercise to determine if there is a potential for significant risk.

In this screening-level analysis, exposures to aquatic biota and to man from Ra-226 and Ra-228 were estimated for varying water concentrations of radium representative of produced water outfalls and for the levels measured in organisms in a study performed by Continental Shelf Associates (1991) for the Mid-Continent Oil and Gas Association (see Section 5). Doses to fish, molluscs and crustaceans were estimated as described in IAEA (1976) (Section 8). The major pathway resulting in exposure to man is expected to be consumption of contaminated fish and shellfish. Individual lifetime risks to people consuming fish and shellfish harvested near a produced water outfall were estimated using United States Environmental Protection Agency (USEPA) risk factors for a range of water concentrations, concentration factors and intake rates (Section 9). The total number of excess cancers per year due to radium discharged to coastal Louisiana in produced water was estimated using simple conservative assumptions to provide a value (Section 9).

Based on the results of this screening-level assessment, a more comprehensive and realistic analysis can be performed to produce more realistic estimates of health and environmental risk.

# Section 3 RADIUM

## 3.1 RADIUM ISOTOPES

Two isotopes of radium (Ra-226 and Ra-228) are the radionuclides of most concern in produced water. Radium isotopes probably make up the bulk of nuclide activity in produced water (Snavely, 1989). These isotopes of radium are important because of their high solubility, long half-lives and tendency to bioaccumulate in food organisms. Ra-226 and Ra-228 arise during the radioactive decay of their naturally occurring parent radionuclides. Radium-226, created from thorium-230, is a member of the uranium-238 decay series (Table 3-1). Ra-226 has a half-life of 1602 years and decays to radon-222. Radon is a gas with low solubility in water, and does not remain in surface waters. Ra-228 is part of the thorium decay series (Table 3-2), and its parent radionuclide is thorium-232.

## 3.2 BACKGROUND ENVIRONMENTAL RADIUM CONCENTRATIONS

Ra-226 and Ra-228 are naturally occurring radionuclides, and are present in the earth's crust, in fresh water rivers and lakes, and in coastal waters, oceans and sediments. Table 3-3 summarizes some of the literature describing the background concentrations of radium in coastal and oceanic waters and sediments. As a point of comparison, the United States Environmental Protection Agency (USEPA) interim drinking water standard for combined Ra-226 and Ra-228 is 5.0 pCi/l.

The concentration of Ra-226 in the ocean ranges from 0.024 to 0.182 pCi/l, and the average was estimated to be about 0.1 pCi/l (Cherry and Shannon, 1974). The concentration of Ra-228 in the ocean ranges from 0.0001 to 0.1 pCi/l. Radium levels are generally higher in the deep ocean than in surface ocean water. Surface water concentrations

Table 3-1. Uranium Decay Series<sup>1</sup>.

<del></del>		
NUCLIDE	DECAY MODE	HALF-LIFE
238 <sub>U</sub>	α	4.51 x 10 <sup>9</sup> years
$^{234}\mathrm{Th}$	$oldsymbol{eta}$ , $oldsymbol{\gamma}$	24.1 days
234 <sub>Pa</sub>	Ъ,с	1.17 minutes
234 <sub>U</sub>	α	2.47 x 10 <sup>3</sup> years
230 <sub>Th</sub>	$\alpha_1 \gamma$	$8.0 \times 10^4$ years
226 <sub>Ra</sub>	$\alpha, \gamma$	1602 years
222 <sub>Rn</sub>	α	3.823 days
218 <sub>Po</sub>	α	3.05 minutes
214 <sub>Pb</sub>	$eta$ , $\gamma$	26.8 minutes
214 <sub>Bi</sub>	β,γ	19.7 minutes
214 <sub>Po</sub>	α	164 µseconds
210 <sub>Pb</sub>	β,γ	21 years
210 <sub>Bi</sub>	ß	5.01 days
210 <sub>Po</sub>	α	138.4 days
206 <sub>Pb</sub>		STABLE

<sup>1:</sup> from Bureau of Radiological Health, 1970.

Table 3-2. Thorium Decay Series 1.

NUCLIDE	DECAY MODE	HALF-LIFE
232 <sub>Th</sub>	α	$1.4 \times 10^{10}$ years
228 <sub>Ra</sub>	β	6.7 years
228 <sub>Ac</sub>	β,γ	16.13 hours
228 <sub>Th</sub>	$\alpha, \gamma$	1.91 years
224 <sub>Ra</sub>	$\alpha, \gamma$	3.64 days
220 <sub>Rn</sub>	α .	55 seconds
216 <sub>Po</sub>	α	0.15 seconds
212 <sub>Pb</sub>	β,γ	10.64 hours
212 <sub>Bi</sub>	$lpha$ , $eta$ , $\gamma$	60.6 minutes
212 <sub>Po</sub>	α	304 nseconds
208 <sub>T1</sub>	$eta$ , $\gamma$	3.10 minutes
208 <sub>Pb</sub>		STABLE

<sup>1:</sup> from Bureau of Radiological Health, 1970.

Table 3-3. Background Concentrations of Radium.

	Ra-226 (PC1/1)	Ra-228 (pCi/l)	Source
Coastal Waters	0.01 - 0.59	( 1	Cherry and Shannon, 1974
La Coastal Water	0.2 - 0.7	0 - 10.3	CSA, 1991 <sup>1</sup>
La Coastal Water	0.58	:	Hanan, 1981
Ocean Surface	0.04 - 0.08	!	Cherry and Shannon, 1974
Ocean Surface	0.032	0.0135	Pentreath, 1984
Ocean Surface	0.034 - 0.083	0.001 - 0.1	Woodhead, 1984
Deep Ocean	0.08 - 0.16	;	Pentreath, 1984
Deep Ocean	0.154	0.0019	Woodhead, 1984
Fresh Water	0.1 - 0.5		Hess et al. 1985
Ground Water	0 - 81	0.3 - 32	Lucas, 1985
La Coastal Sed (pCi/g)	0.1 - 0.9	9.0 - 0	CSA, 1991 <sup>1</sup>
La Coastal Sed (pCi/g) 1.4	1.45	P.	Hanan, 1981
1 GSA (1991) study, Section	tion 4.4		

range from 0.032 to 0.083 pCi/l for Ra-226 and from 0.001 to 0.1 pCi/l for Ra-228. Deep ocean concentrations are about 0.154 pCi/l for Ra-226 and 0.0019 for Ra-228.

Radium concentrations in coastal waters are generally higher than in the ocean. Reported Ra-226 concentrations in coastal waters range from 0.01 to 0.70 pCi/l. Sediment concentrations of Ra-226 along the coast of Louisiana range from 0 to 1.45 pCi/gram (Hanan, 1981; CSA, 1991, Section 4).

#### 3.3 RADIUM CONCENTRATIONS IN PRODUCED WATER

Ra-226 and Ra-228 are found in elevated concentrations in some oil field produced waters. Table 3-4 shows the ranges of Ra-226 and Ra-228 concentrations in produced waters found by several investigators. Concentrations of Ra-226 in produced water range from 0 to 1620 pCi/l. Ra-228 concentrations range from 0 to 928 pCi/l.

#### 3.4 CHEMISTRY AND FATE OF RADIUM IN PRODUCED WATER

When a produced water is discharged into a body of water, the resulting distribution of radium is controlled by a number of physical and chemical processes. The most important of these are mixing and dilution by turbulence, advection and dispersion; adsorption/desorption interactions with sediments and suspended solids, and coprecipitation of soluble salts. Radium in water exists primarily as the divalent ion  ${\rm Ra}^{2+}$  and has chemical properties similar to calcium, barium and strontium.

Table 3-4 Radium in Produced Water (modified from Snavely, 1989)

Location	Ra-226	97	Ra-228	88
	pC1/1	#samples	pCi/l	#samples
Oklahoma <sup>l</sup>	10 - 1620	10	75 - 240	4
Texas Panhandle <sup>2</sup>	3 - 1560	75	i i	:
Gulf Coast <sup>3</sup>	1.3 - 437	10	204 - 575	9
Coastal La <sup>4</sup>	0 - 930	405	. 0 - 928	405
Offshore La <sup>5</sup>	4 - 584	42	18 - 586	42

1 Armbrust and Kuroda, 1955

2 Pierce, Mytton and Gott, 1964 3 Kraemer and Reid, 1984

4 State of Louisiana database, see section 4.4 5 Offshore Operators Committee database, see section 4.4

#### Dilution

Radium in produced water undergoes rapid mixing and dilution when discharged to surface water. Dilution of radium occurs in two stages -- near-field mixing and far-field mixing.

Near-field mixing is based on the turbulence produced by the discharge momentum and discharge buoyancy (Till and Meyer, 1983). Near-field mixing is rapid, occurs over a short distance, and may result in large dilutions (Till and Meyer, 1983). Factors that affect the amount of dilution achieved in the near-field mixing stage include momentum and buoyancy of the effluent, depth and current of receiving water, and outfall location and configuration.

Far-field mixing includes the ambient advection and diffusion processes which take place after initial dilution at the outfall. These processes are slow compared to the near-field mixing processes, and occur over a larger area. For estuaries and coastal seas, the mixing processes are strongly influenced by tides.

Model Predictions. Radium discharged into coastal waters is diluted very rapidly. Dilution is faster for deep offshore discharges than for shallow nearshore outfalls. The dilution expected under different conditions can be predicted using a surface water transport model. The Offshore Operators' Committee model (OOC) was used to predict the steady state concentration of radium (and the level of dilution achieved) at varying distances from nearshore and offshore produced water outfalls discharging 1000 pCi/l radium. The OOC model is described in more detail in Section 4.2.

Five simulations were run; two shallow water cases representative of discharges to canals, and three deep water cases representative of offshore discharges. The discharged radium was treated as a tracer and did not decay or adsorb to sediments in the model. The model assumes a unidirectional and constant current, which resulted in predictions that underestimate dilution. The input parameters used in the simulations are summarized in Table 3-5.

For each case, the minimum dilution factor (at the centerline of the plume) was calculated at various distances from the outfall:

dilution factor = initial conc. of radium 
$$(1000 \text{ pCi/l})$$
 (3-1)  
concentration of radium  $(\text{pCi/l})$ 

Figure 3-1 shows how the minimum dilution factors (calculated along the centerline of the plume) vary with distance from the outfall for the two nearshore cases (300 and 800 bbl/day, 8 foot depth). Within 100 feet of the discharge point, a dilution factor greater than 30 is achieved for the 800 bbl/day case. The 300 bbl/day discharge is diluted by a factor greater than 60 within 100 feet.

Figure 3-2 shows the dilution factors for the three offshore cases (5,000, 10,000 and 20,000 bbl/day, 220 foot depth). Dilution is more rapid for the offshore outfalls, and a dilution factor of greater than 300 is reached within 100 feet of the outfall. Similar dilutions are achieved for offshore outfalls at depths down to about 70 feet. When the outfall is located in shallower water (less than 70 feet) there is a potential for a decrease in dilution due to interactions with the bottom.

Table 3-5. Parameters for Nearshore and Offshore 00C Model Runs.

	NEARSHORE	)RE		OFFSHORE	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1
	CASE 1	CASE 2	CASE 1	CASE 2	CASE 3
water depth	8 ft	8 ft	220 ft	220 ft	220 ft
pipe depth 1	1 ft	1 ft	30 ft	30 ft	30 ft
pw salinity	100  ppt	100 ppt	100 ppt	100 ppt	100 ppt
velocity - top	0.1  ft/s	0.1 ft/s	0.3 ft/s	0.3 ft/s	0.3 ft/s
velocity - botto	m 0.02 ft/s	0.02 ft/s	0.001 ft/s	0.001 ft/s	0.001 ft/s
pipe diameter	2 inch	3 inch	12 inch	12 inch	12 inch
discharge rate	300 bb1/d	800. pp1/q	5,000 bb1/d	10,000 bb1/d	20,000 bbl/d
radium conc.	1,000 pci/l	1,000 pci/l	1,000 pci/1	1,000 pci/1	1,000 pci/1

# TRACER DILUTION WITH DISTANCE NEARSHORE

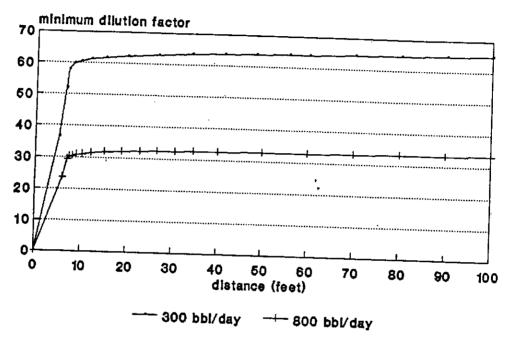


Figure 3-1. Predicted Dilution Factors For Nearshore Discharges Within 100 feet of an Outfall; 300 and 800 bbl/day, 8 foot depth.

## TRACER DILUTION WITH DISTANCE OFFSHORE

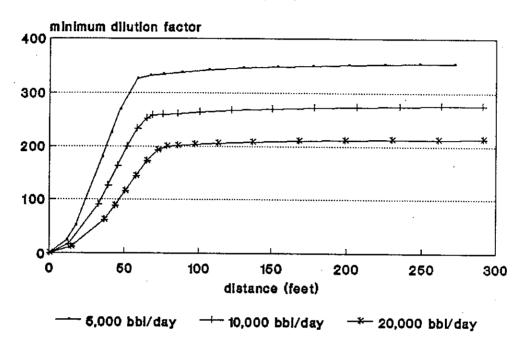


Figure 3-2. Predicted Dilution Factors For Offshore Discharges Within 300 feet of an Outfall; 5,000, 10,000 and 20,000 bbl/day, 220 foot depth.

<u>Field Data.</u> Field data can also be used to demonstrate the extent to which radium concentrations are reduced at varying distances from an outfall. A field study conducted by Continental Shelf Associates (CSA, 1991) for the Mid-Continent Oil and Gas Association provides useful data.

Three stations were included in the study; Ra-226 and Ra-228 were measured in water at the discharge point, and at 25 and 50 feet from the outfalls. Each of the three outfalls were located in canals along the Louisiana coast. Figure 3-3 shows the sampling configuration used in the study. Characteristics of the three outfalls are given in Table 3-6.

Figure 3-4 shows the average dilution factor for Ra-226 measured at 25 and 50 feet from the three outfalls. Water concentrations of Ra-226 were reduced by factors of 148 to 1526 at 50 feet from the outfalls.

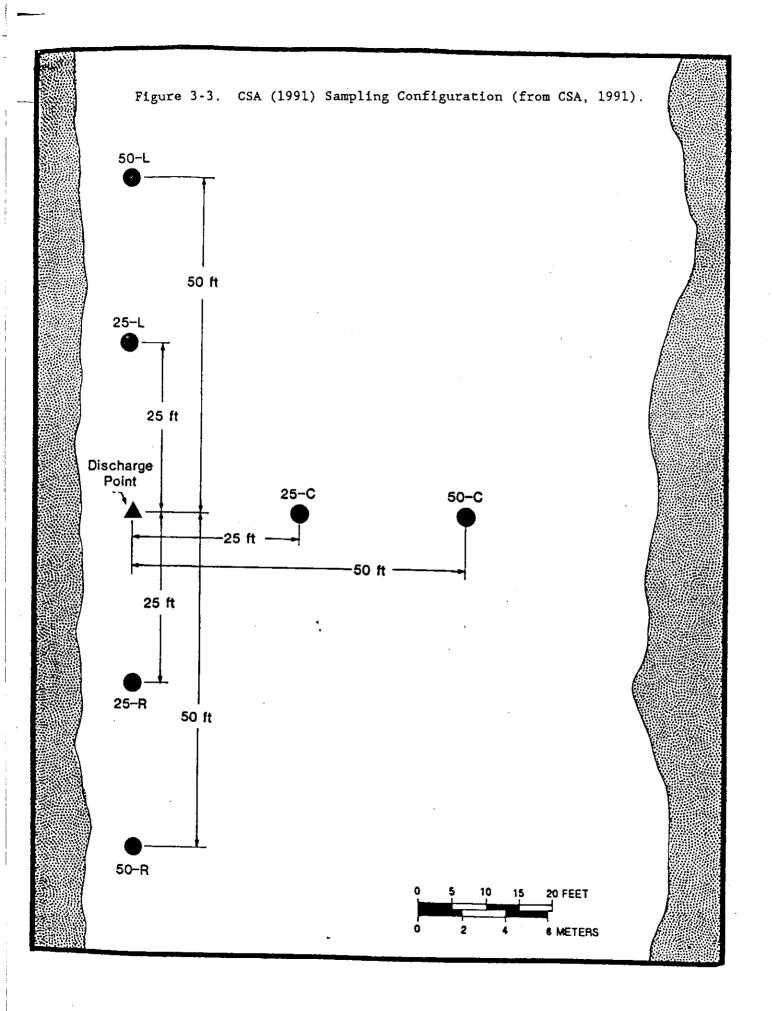
The dilution factors calculated from the CSA (1991) data are larger than those predicted by the OOC model for the two nearshore cases analyzed. Both sets of information suggest that significant dilution occurs within 50-100 feet of nearshore produced water outfalls (dilution factors of 50 - 1500). Dilution of produced water discharged offshore is more rapid (dilution factor of 200 within 100 feet).

#### Adsorption and TDS

The concentration of radium in water is usually controlled through adsorption-desorption reactions at the solid-liquid interface.

The extent to which radium becomes associated with sediments or suspended solids can be described by the distribution coefficient  $K_{\mathbf{d}}$ :

$$K_d$$
 (1/g) = activity in solid (pCi/g) (3-2) activity in water (pCi/1)



Characteristics of Three Nearshore Outfalls in CSA (1991) Study. Table 3-6.

SITE 3	1 - 37.5 ft No data 0.05-0.4 ft/s 5,076 bbl/d 251.9 pci/l
SITE · 2	1 - 8 ft 61.41 ppt 0.05-1.2 ft/s 3,775 bb1/d 110.6 pCt/1
SITE 1	1 - 5.5 ft 92.46 ppt 0.1-0.5 ft/s 4,000 bb1/d 228.9 pC1/1
	water depth pw salinity ambient flow rates discharge rate discharge Ra-226

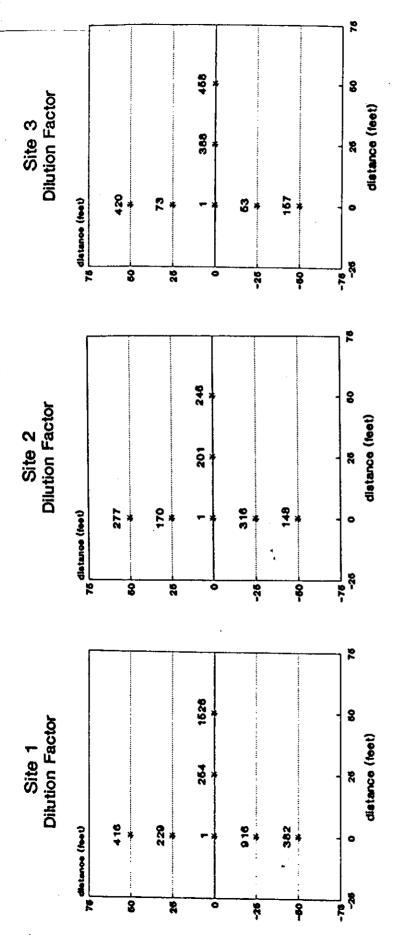


Figure 3-4. Dilution Factors for the Three CSA (1991) Sites.

The  $K_{\rm d}$  for radium depends on the type of solid (clay adsorbs more radium than sand), the salinity and pH of the water, and the presence of other ions in solution. The  $K_{\rm d}$  for radium can range from less than 10 to over 1500 1/kg (Snavely, 1989).

The adsorption of radium is subject to competitive interactions with other ions in solution for adsorption sites. If other cations are present in solution (such as  $Na^+$ ,  $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Sr^{2+}$  or  $Ba^{2+}$ ), they also adsorb to available absorption sites and decrease the capacity of the sediment or suspended particles for adsorbing radium. Consequently,  $Ra^{2+}$  is more mobile in waters with a high total dissolved solids (TDS) content. In fresh water environments, most radium is associated with sediments and suspended particles. In waters with high salinity, most radium is in solution.

Produced waters with high radium levels have high levels of total dissolved solids (TDS) which increase the mobility of the  $\mathrm{Ra}^{2+}$  ions. Kraemer and Reid (1984) found a linear relationship between the radium concentration of produced waters and their TDS. This relationship does not always hold, however, because a source of radium must exist in the reservoir to cause elevated levels of radium (Snavely, 1989).

Most produced waters are highly saline, and most radium is in solution as  $Ra^{2+}$ . Table 3-7 presents a summary of the distribution coefficients for waters of various total dissolved solids. Seawater has a TDS of approximately 30,000 mg/l. Figure 3-5 is a plot of these data.

When brine is discharged into a body of fresh or brackish water, the radium enrichment zone in the underlying sediments may be quite widespread due to salinity controlled limits on sorption in sediments near the discharge point (Landa and Reid, 1982). In the case of a discharge of brine into seawater the enrichment zone could be even more dispersed. Because the mobility of radium is enhanced by high TDS water, the zone of radium adsorption and deposition is likely to be large, and levels of radium in sediments will not be high even at the discharge point. In coastal and offshore areas influenced by tides,—

Table 3-7. Ra-226 Distribution Coefficients for Waters with Varying TDS (modified from Snavely, 1989).

	TDS	Water (pC1/1)	Sediment (pCi/g)	K <sub>d</sub> (1/g)	Source
Produced Water	90,000	133.	0.47	0.0035	Landa and Reid, 1983
Diluted Prod Water	30,000*	8.0	0.12	0.015	Landa and Reid, 1983
Diluted Prod Water	000'6	0.33	0.015	0.045	Landa and Reid, 1983
Estuary	3,800	0.024	0.14	5.6	Li et al., 1977
Marsh	200	0.58	1.45	2,5	Hanan, 1981
Hudson River	200	0.007	8.8	120.	Li et al., 1977
	-				

\* Seawater has a TDS of approximately 30,000 mg/l.

## Effect of TDS on Radium Distribution Coefficient

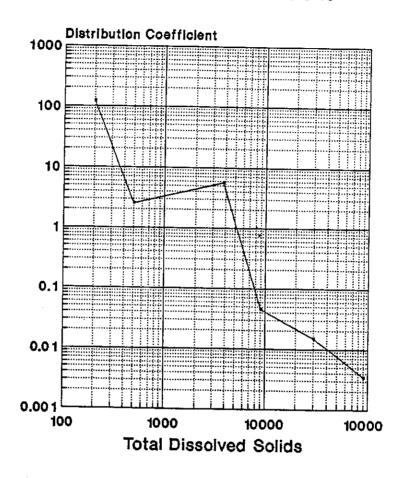


Figure 3-5. Effect of Total Dissolved Solids (TDS) on Radium Distribution Coefficient.

desorption and flushing of the radium will occur with seasonal and daily increases in salinity.

Data collected in the CSA (1991) study can be used to demonstrate the levels of sediment contamination likely to be found at varying distances from a produced water outfall. For the three outfalls included in the study, Figure 3-6 shows the average concentration of Ra-226 in sediment directly underneath the outfall and at 25 and 50 feet.

#### Coprecipitation

The concentration of radium in solution may also be affected by coprecipitation with barium, calcium and strontium salts. The solubility product  $K_{\mbox{sp}}$  describes the solubility of soluble salts:

$$K_{sp}$$
 (salt) = [cation conc.] [anion conc.] (3-3)

If the product of the cation concentration times the anion concentration exceeds the  $K_{\rm sp}$  the salt will precipitate. The solubility of radium sulfate is about 1.0 mg/l, and radium will not precipitate by itself because the concentration of  ${\rm Ra}^{2+}$  is so low in seawater (Snavely, 1989). During precipitation of a salt containing barium, calcium or sulfate, radium is coprecipitated and removed from solution along with the other ions.

Produced waters can contain barium, calcium and strontium ions. Seawater contains about 2700 mg/l of sulfate and precipitates can occur when the water is discharged. Barium sulfate is the most likely precipitate because its solubility is the smallest (Snavely, 1989):

$$K_{sp} = 1.39 = [Ba^{2+}mg/1] [SO_4^{2-}mg/1]$$
 (3-4)

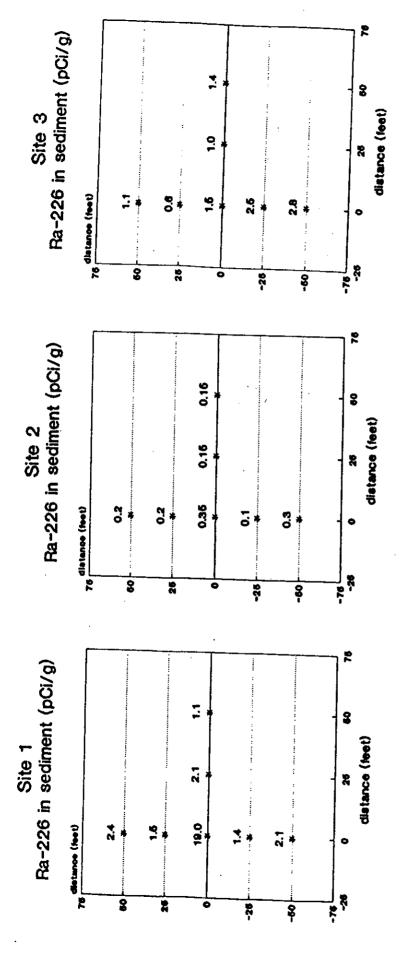


Figure 3-6. Ra-226 Sediment Concentrations Measured in the CSA (1991) Study.

Seawater with a sulfate concentration of 2700 mg/l can contain in solution 0.0005 mg/l Ba $^{2+}$  (Snavely, 1989). Radium can coprecipitate with barium sulfate, removing a large percentage of the radium in solution. For a produced water with 1100 mg/l Ba $^{2+}$  (saturated concentration), diluted by a factor of 100 (11 mg/l Ba $^{2+}$ ), BaSO $_4$  would precipitate. Gulf of Mexico produced water can contain high concentrations of barium, and coprecipitation of radium may be significant.

If precipitation of barium, calcium or strontium sulfate occurs, some of the radium in solution may be precipitated and deposited near the discharge point.

## Section 4 PREDICTION OF ENVIRONMENTAL RADIUM CONCENTRATIONS

#### 4.1 INTRODUCTION

Five data sets were reviewed to assess their usefulness for developing the exposure estimates needed in a risk assessment study. The data sets were examined and compared to the information needed to estimate environmental concentrations of radium resulting from the discharge of produced water.

### 4.2 MODELS TO ESTIMATE ENVIRONMENTAL RADIUM CONCENTRATIONS

There are a number of models available that can be used to estimate the concentration of radium in surface water that would result from the discharge of produced water. These models predict the concentrations of contaminants in surface water as a result of dilution, dispersion and adsorption onto sediment. Predictions may be given as a function of space and time. Two models appropriate for use in assessing the fate of radium discharged in produced water, along with their data requirements, are described below. The two models discussed represent the simplest models available, and the more complex models which make fewer simplifying assumptions but require more data. The level of complexity required in modeling the environmental transport of radium will depend on the characteristics of the receiving water body, the potential exposure pathways, how accurate the resultant dose estimates need to be (NCRP, 1984) and the data that is reasonably available for the analysis.

#### Box Model

The simplest model that can be used to estimate the concentration of radium in surface water resulting from produced water discharges is a

box model. The model assumes a constant input of radium and complete mixing within a given volume. Radium is removed from this volume by a combination of water exchange, radioactive decay and sediment interactions (IAEA, 1982). The steady state concentration of radium in water can be estimated as:

$$C = Q / K_e V$$
 (4-1)

where

C = concentration of radium in water

Q = input rate of radium into the mixed volume

V = the mixed volume of the receiving water

Ke - removal constant

The removal constant Ke is estimated as:

$$K_e = \rho + r/V \tag{4-2}$$

where

 $\rho$  = the decay constant r/V = the fractional loss rate of water from the mixed volume.

In tidally influenced coastal seas, V may be described by the tidal excursion, and r/V by the net water movement (IAEA, 1982).  $K_{\rm e}$  can also include the loss of radium to the sediments, based on the distribution coefficient  $K_{\rm d}$ .

Data required by the box model include:

- (1) Input rate of radium into mixed volume
- (2) Mixed volume of the receiving water
- (3) Decay constant
- (4) Fractional loss rate of water from the mixed volume
- (5) Distribution coefficient

#### OOC Model

A model was developed by the Offshore Operators Committee (OOC) and Exxon Production Research Company to predict the initial fate of drilling mud and cuttings discharged to the marine environment (Brandsma and Sauer, 1983a, 1983b; O'Reilly et al., 1988). The model was modified to allow prediction of the initial dynamics and passive diffusion of

produced waters. The OOC model is a modification of simpler models (Koh and Chang, 1973; Brandsma and Divoky, 1976), which may be appropriate for use in modeling radium transport when the data required by the OOC model are not available.

The OOC model simulates the descent of the jet of discharged material through the water column, dynamic collapse as the material spreads out on the bottom or within the water column, and passive diffusion (Brandsma and Sauer, 1983). The passive diffusion phase begins when the transport and spreading of the plume are determined more by ambient currents than by the dynamic character of the plume. The diffusion portion of the model is of Langranian formulation -- groups of particles leaving the dynamic portion of the plume are represented by many small clouds of material. Each cloud is independently advected, diffused and settled according to local conditions (Brandsma and Sauer, 1983a).

Currents can be variable in three dimensions, density profiles can change with time, and the model can incorporate variable depths and land boundaries (Brandsma, 1983). The model requires the following data (Brandsma and Sauer, 1983):

- (1) Mud/contaminant characteristics
  - (a) Bulk density
  - (b) Number of discrete particle classes
  - (c) Volume concentration, density and settling velocity for each particle class
- (2) Discharge characteristics
  - (a) Rate
  - (b) Duration
  - (c) Radius and orientation of discharge nozzle
  - (d) Position of the rig within global coordinate system
- (3) Ambient characteristics
  - (a) Times when ambient data are available
  - (b) Density profile(s)
  - (c) Current velocity distribution(s)
  - (d) Wave height(s) and period(s)

#### 4.3 DATA NEEDED TO ESTIMATE ENVIRONMENTAL RADIUM CONCENTRATIONS

To assess the total impact of the many (greater than 400) produced water discharges in the coastal zone of Louisiana some application of the simple box model described above will be needed. Simple box models for estimating the environmental concentration of radionuclides discharged into surface waters require information on the concentration of contaminant in the discharge, the flow rate of the discharge, the volume of the receiving water being modeled, and the fractional loss rate of water from the mixed volume. More complex models (such as the OOC model described above) may be needed to assess the effects of radium discharged from individual outfalls.

These more complex models may require data on the concentration of contaminants in the discharge, the density of the discharge, the rate of discharge, the depth of the discharge and of the receiving water, tidal data, and a description of the geometry of the outfall location in relation to land formations.

To estimate the loss of a contaminant from the water column to the sediment, additional data are needed. Models which estimate the adsorption of a contaminant to sediments require data on the amount of suspended sediment and distribution coefficients  $(K_d)$  for the contaminant. Since the  $K_d$  for radium is affected by the Total Dissolved Solids content (TDS) of the water, the relationship between TDS and  $K_d$  is also required.

To predict the extent to which radionuclides may be lost to the sediment through coprecipitation with barium, calcium or strontium sulfate, the concentration of these ions in produced water and in receiving water is needed.

#### 4.4 AVAILABLE DATA

Relevant data for developing exposure estimates for radium discharged in coastal and offshore Louisiana are available in five data sets. The information and data fields in each of these data sets are listed in Table 4-1. Each data set is described in more detail below.

#### MOGA Database

The Louisiana Mid-Continent Oil and Gas Association (MOGA) data base contains data for 267 stations, with geographic coordinates for 254. The locations of these stations are plotted in Figure 4-1.

For each station, the produced water discharge rate (bbl/d) and concentration (pCi/l) of Ra-226 and Ra-228 in the discharge water have been measured. The flow rate of the receiving water is not available in this data set.

Ra-226 concentrations varied from 0 to 792 pCi/l, with an average concentration of 181.6 pCi/l. Ra-228 concentrations varied from 0 to 928 pCi/l, with an average of 219.7 pCi/l. The concentration of Ra-226 is correlated with the concentration of Ra-228 (Figure 4-2). Ra-228 concentrations in the discharge were generally about equal to the concentration of Ra-226, although this relationship did not hold at every station. Figure 4-3 gives the frequency distributions of the Ra-226 and Ra-228 concentrations in the discharge for these stations.

Table 4-1. Data Available for Predicting Environmental Concentrations of Radium Discharged in Produced Water in Louisiana.

### Mid-Continent Oil and Gas Association (MOGA) database

- o 267 stations
- o station location (254 stations)
- o water discharge rate
- o Ra-226 and Ra-228 conc. in discharge
- o no flow rate for receiving water
- o no radium concentration at distance from outfall

## State of Louisiana database (includes MOGA stations)

- o 447 stations
- o no station location
- o Ra-226 and Ra-228 conc. in discharge
- o water discharge rate (412 stations)
- o no flow rate for receiving water
- o no radium concentration at distance from outfall

#### Offshore Operator's Committee database

- o 42 outfalls
- o Ra-226, Ra-228 conc. in discharge
- o water discharge rates
- o no radium conc. at distance from outfall
- o no receiving water flow rates

## Louisiana Wetlands Study (Steimle & Associates, Inc.)

- o 38 stations
- o station location
- o water discharge rate (from MOGA database)
- o Ra-226 and Ra-228 conc. in discharge (from MOGA database)
- o flow rate of receiving water
- o salinity of receiving water at distances from outfall
- o Ra-226 and Ra-228 conc. at distances from outfall
- (6 stations)
- o Ra-226 and Ra-228 conc. in sediment (6 stations)

## Continental Shelf Associates (CSA, 1991) Study

- o 3 outfall stations, 6 reference stations
- o Ra-226, Ra-228 conc. at outfall
- o Ra-226, Ra-228 conc. at distances from outfall
- o Ra-226 conc. in sediments
- o Ra-226 conc. in fish, molluscs and crustaceans
- o flow and water discharge rates

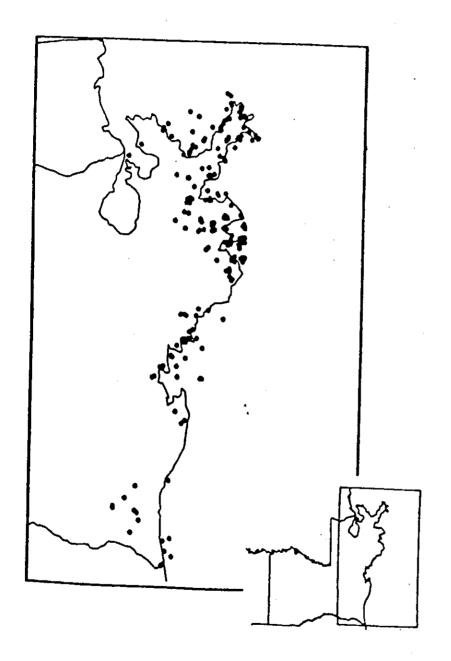


Figure 4-1. Location of Stations in the MOGA database

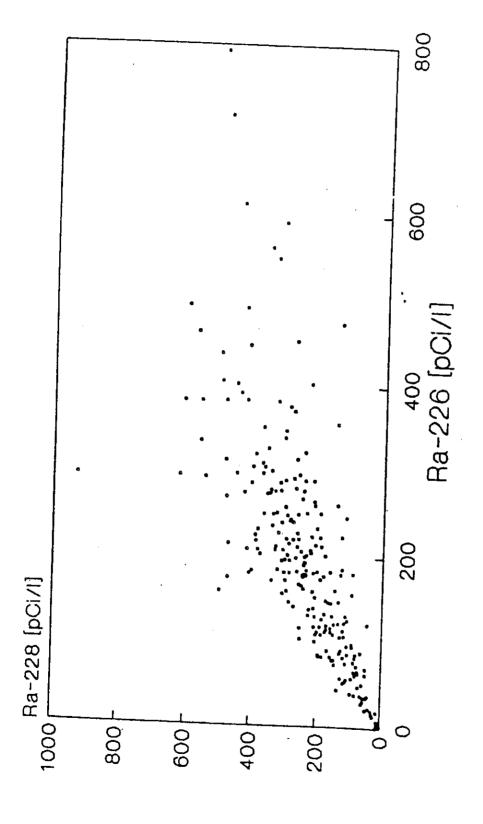
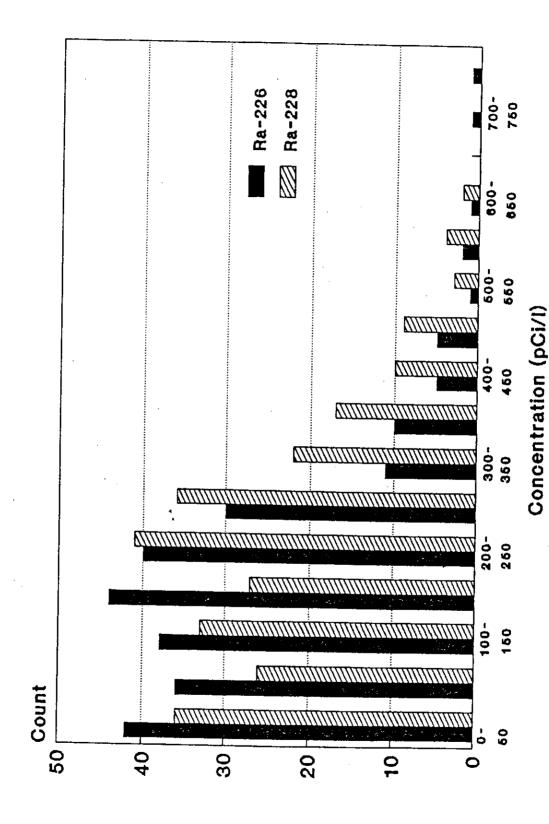


Figure 4-2. Concentration of Ra-226 vs. Ra-228 (pCi/l) for Stations in the MOGA Database.

Figure 4-3. Frequency Distributions for Ra-226 and Ra-228 Concentrations for Stations in the MOGA database.



The discharge rate in pCi/day of Ra-226 and Ra-228 was calculated for each station as: [Water discharge rate (bbl/day) x 159 (1/bbl) x radium concentration (pCi/l)]. Ra-226 discharge rates varied from 0.0 to 5.3 x  $10^9$  pCi/day. Ra-228 discharge rates varied from 0.0 to 7.3 x  $10^9$  pCi/day. The frequency distributions of the radium discharge rates (pCi/day) for Ra-226 and Ra-228 are presented in Figure 4-4.

#### State of Louisiana Database

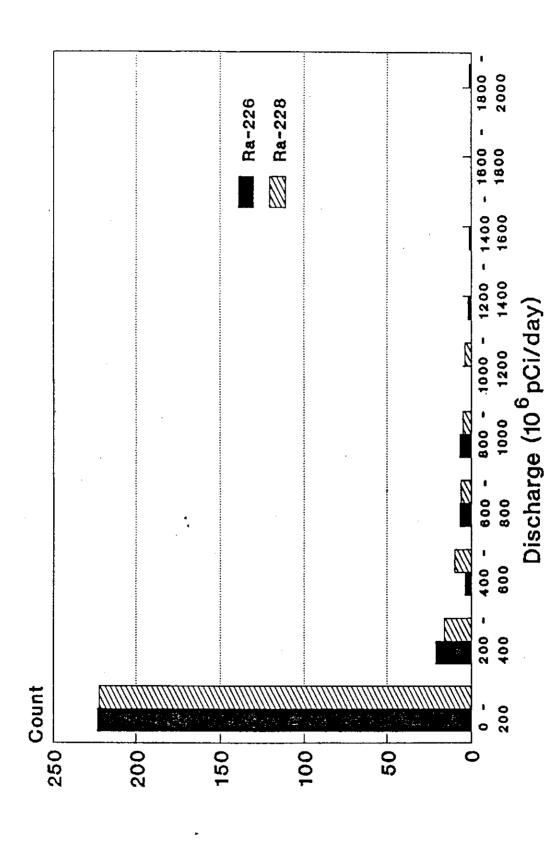
The State of Louisiana database contains essentially the same data elements as the MOGA database, and includes 447 stations (including the 267 MOGA stations). For 405 stations, the produced water discharge rate and concentration of Ra-226 and Ra-228 in the discharge water were measured. The location of the stations and the flow rate of the receiving water are not available in this data set.

Ra-226 concentrations varied from 0 to 930 pCi/l, with an average concentration of 159.2 pCi/l. Ra-228 concentrations varied from 0 to 928 pCi/l, with an average of 164.5 pCi/l. Discharge rates of Ra-226 varied from 0.0 to 5.2 x  $10^9$  pCi/day. Ra-228 discharge rates varied from 0.0 to 7.3 x  $10^9$  pCi/day. Figure 4-5 gives the frequency distributions of the Ra-226 and Ra-228 discharge rates for these stations.

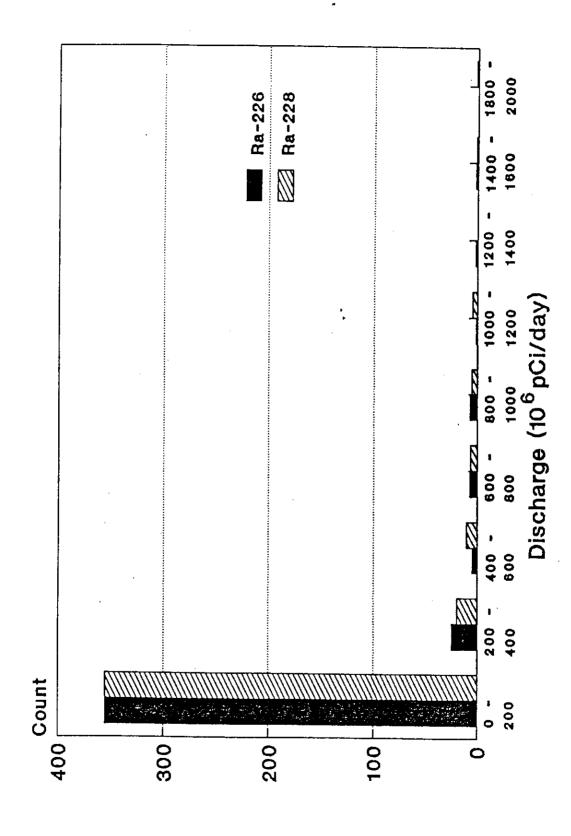
## Offshore Operators Committee Database

This data set contains information for 42 offshore outfalls, including Ra-226 and Ra-228 concentrations in the discharge, and the produced water discharge rate for each station. Station locations are available in the form of lease block designations. The salinity of the discharge was measured at 25 of these stations. Salinity ranged from 12695 to 203000 mg/l, with an average salinity of 112322.5 mg/l.

For Stations in the MOGA database (Several large discharge points are not included in the Figure; for Ra-226: 1 point at 2067 x 10<sup>6</sup> pCi/day, 1 point at 5317 x 10<sup>6</sup> pCi/day. For Ra-228: 1 point at 2068 x 10<sup>6</sup> pCi/day, 1 point at 2449 x 10<sup>6</sup> pCi/day, 1 point at 7371 x 10<sup>6</sup> pCi/day). Figure 4-4. Frequency Distributions for Ra-226 and Ra-228 Discharges (pCi/day)



for Stations in the State of Louisiana database (Several large discharge points are not included in the Figure; for Ra-226: 1 point at 2067 x 10<sup>6</sup> pCi/day, 1 point at 5317 x 10<sup>6</sup> pCi/day. For Ra-228: 1 point at 2068 x 10<sup>6</sup> pCi/day, 1 point at 2449 x 10<sup>6</sup> pCi/day, 1 point at 7371 x 10<sup>6</sup> pCi/day). Figure 4-5. Frequency Distributions for Ra-226 and Ra-228 Discharges (pCi/day)



The concentration of Ra-226 at these outfalls varied from 4.0 to 584.0 pCi/l with an average of 262.3 pCi/l. Ra-228 varied from 18.0 to 586.0 pCi/l, with an average concentration of 276.7 pCi/l. Discharge rates for Ra-226 varied from 1.18 x  $10^5$  to 1.39 x  $10^9$  pCi/day, with an average of 2.97 x  $10^8$  pCi/day. Ra-228 discharge rates varied from 1.44 x  $10^5$  to 1.43 x  $10^9$  pCi/day with an average discharge rate of 2.97 x  $10^8$  pCi/day. The frequency distributions of the Ra-226 and Ra-228 discharge rates for these outfalls are given in Figure 4-6.

#### Louisiana Wetlands Study

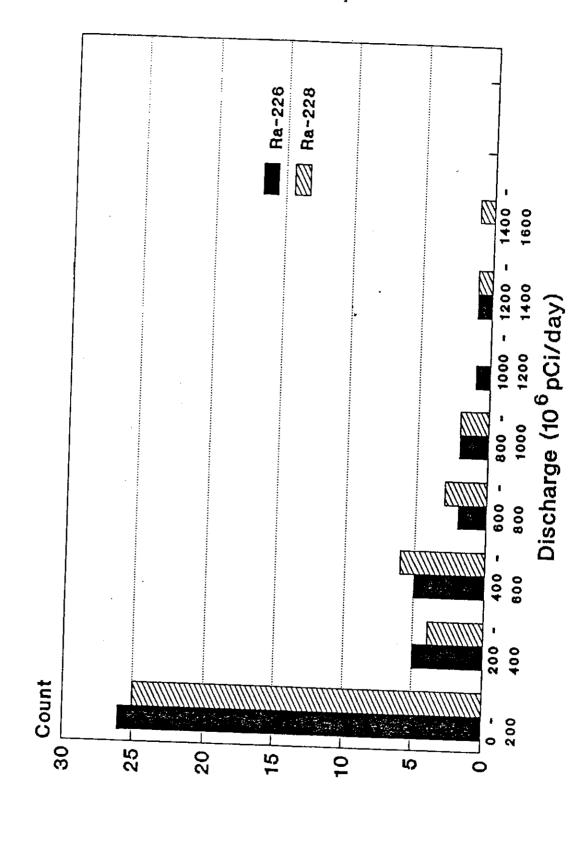
These data were collected as part of a study for the American Petroleum Institute (Steimle & Associates, 1990). The data set contains information collected for a subset of the stations in the MOGA database (Section 4.3). For 38 stations, salinity and flow rate was measured at various distances from the outfall. The locations of these stations are plotted in Figure 4-7. For six stations, data are available for Ra-226 and Ra-228 concentrations at various distances from the outfall in water and in the sediment.

#### Continental Shelf Associates (CSA, 1991) Study

This data set contains data for three outfall stations and six reference stations. For each station, Ra-226 and Ra-228 concentrations were measured in the discharge water, and at 25 and 50 feet from the outfall. Ra-226 and Ra-228 concentrations were also measured in sediment, fish, molluscs and crustaceans. Conductivity, salinity, temperature, dissolved oxygen and flow were measured at all stations.

Ra-226 concentrations in the discharge water varied from 110.5 to 251.85 pCi/l, and Ra-228 concentrations varied from 244.4 to 383.0 pCi/l. Average Ra-226 concentrations in sediment directly below the discharge point ranged from 0.35 to 19.0 pCi/g, and average Ra-228 sediment concentrations at the outfall ranged from 0 to 2.5 pCi/g.

Figure 4-6. Frequency Distributions for Ra-226 and Ra-228 Discharges (PCI/day) for Stations in the Offshore Operators Committee database.



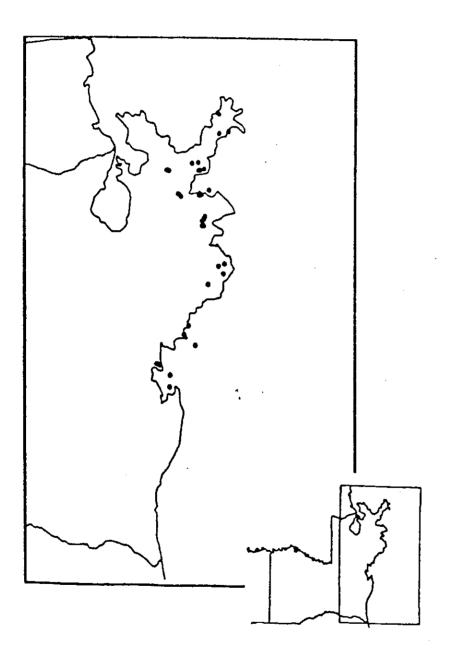


Figure 4-7. Location of Stations in the Louisiana Wetlands Study.

Biological samples were taken within 50 feet of the produced water outfalls. Average Ra-226 concentrations ranged from 0.0 to 0.041 pCi/g in whole fish, 0.0065 to 0.0075 pCi/g in mollusc tissue and 0.0675 to 0.125 pCi/g in whole crustaceans. Average Ra-228 concentrations ranged from 0.005 to 0.022 pCi/g in whole fish, 0.003 to 0.011 pCi/g in mollusc tissue and 0.025 to 0.243 pCi/g in whole crustaceans. These data are described in more detail in Section 5.5.

## 4.5 ASSESSMENT OF AVAILABLE DATA

The available data can be assessed in terms of their usefulness in estimating environmental radium concentrations using models similar to the ones described above. All of the available data sets are limited in that they include only single samples from the produced water outfalls represented. The extent to which discharge rates, radium concentrations, salinity and flow rates vary over time cannot be assessed. Any analysis using these data sets must assume that changes over time at each outfall are minimal, and that the discharge has been occurring over several years, allowing a steady state to be reached.

## For Assessing Impact of Individual Outfalls

To estimate the impact of individual produced water outfalls (e.g. those with the largest radium discharge rate, or those closest to leased shellfishing beds) the concentration of radium as a function of distance from the outfalls must be calculated. These calculations require the use of a transport model, and an assessment of the importance of adsorption and coprecipitation in determining surface water concentrations of radium.

<u>Transport Calculations.</u> Using a simple surface water dispersion model, the data from the Louisiana Wetlands Study (38 stations) can be used to calculate the environmental concentrations of radium resulting from the

discharge of produced water at these stations. The concentration of Ra-226 and Ra-228 in water at varying distances from the outfall was measured for six of these stations. These data can be used to calibrate or validate the model used. The three CSA (1991) Study sites can also be analyzed in this way.

The State of Louisiana data set contains discharge rates and concentrations but no receiving water flow rates or geographic coordinates. If the flow rates can be determined or estimated (possibly from models developed specifically for the Gulf Coast of Louisiana) then environmental concentrations can be estimated for these stations. The State of Louisiana data set would be more useful if geographic coordinates for each station were available.

The Offshore Operator's Committee data set contains discharge rates and can be used to calculate resulting environmental radium concentrations from offshore produced water discharges if a flow rate for the receiving water can be determined.

Some of these data sets may have adequate data for the OOC model, but most will have to be assessed using simpler dispersion models with fewer data requirements. The detailed data required by more sophisticated surface water models such as the OOC model are not readily available for most stations. Such models and data may, however, have been developed specifically for the Gulf Coast of Louisiana, and this possibility should be investigated further.

Adsorption to Sediment. Reports in the literature suggest that highly saline produced waters will not contribute significantly to the concentration of radium in sediments because the radium remains in solution (Section 3). Sediment radium concentrations were measured for six stations in the Louisiana Wetlands Study and the three outfall locations included in the CSA (1991) Study. These data can be used to investigate the importance of the adsorption of radium to sediments in the vicinity of produced water outfalls.

The distribution coefficient  $(K_d)$  for radium varies with the total dissolved solids (TDS) content of the water (Section 3). If radium adsorption to sediments is included in the modeling of radium transport, the salinity of both the produced water and the receiving water must be known. Salinity data are available for the 38 stations sampled for the Louisiana Wetlands Study, for 25 of the 42 outfalls in the Offshore Operator's Committee database, and for the three stations sampled in the CSA (1991) study.

Distribution coefficients for radium in waters of varying salinity are available (Section 3). The adsorption of radium to sediments can be estimated for stations for which salinity was measured. It may also be possible to estimate the salinity of the receiving waters based on other data sources. For stations with sediment and water concentration measurements at varying distances from the outfall, the simple models used to calculate radium transport can be calibrated or validated.

Coprecipitation. It has been suggested that large amounts of radium may be removed from a produced water discharge through coprecipitation of the radium with barium, calcium or strontium sulfate (Section 3). Gulf of Mexico produced water can contain high levels of barium, and coprecipitation of radium near produced water outfalls in the region could be significant. None of the available data sets contain data describing the concentration of these elements in produced water.

The likelihood of large sediment concentrations resulting from the co-precipitation of radium with the sulfate salts of barium, calcium or strontium will be difficult to estimate because of a lack of data. A more through literature search may produce some estimates of the concentrations of these elements in produced waters in Louisiana. However, Snavely (1989) concluded that "there are no published studies on the dilution of  $\mathrm{Ra}^{2+}$  containing produced waters by seawater, the activity level of the precipitates and the amount of  $\mathrm{Ra}^{2+}$  remaining in solution".

# For Assessing Total Impact to Region

To assess the total impact of radium discharged in produced water, the coastal zone of Louisiana must be analyzed using some form of the box model described above.

To estimate the overall impacts, geographic coordinates for the stations in the State of Louisiana data set must be identified, and the extent to which these data represent all produced water outfalls in the region determined. The total amount of radium discharged per day by the stations in the State of Louisiana data set is available (Ra-226: 4.39 x  $10^{10}$  pCi/day; Ra-228: 4.83 x  $10^{10}$  pCi/day). The mixed volume and loss rate can be determined from NOAA charts and tidal compilations for coastal Louisiana, and from other studies done in the region.

# Section 5 CONCENTRATION FACTORS

## 5.1 INTRODUCTION

Concentration factors are commonly used in dose assessments to estimate the levels of radionuclides in aquatic organisms. The concentration factor (CF) is a function of the concentration in the water or sediment (C) and the equilibrium concentration (on a wet weight basis) in the organism (Q).

$$CF = Q / C$$
 (5-1)

It is usually assumed that there is a linear relationship between C and Q and that the concentration factor is independent of the concentration in the environment. In general, this would only be valid for relatively small environmental concentrations. This assumption, along with the assumption that groups of similar organisms have similar concentration factors for a specific radionuclide, allows the use of generic concentration factors in dose assessment models. These assumptions may not always be justified, and generic factors can only be used in a preliminary assessment as a first order estimate of bioaccumulation. Site and organism specific factors are desirable for such a study, but can be difficult and expensive to obtain.

Concentration factors are important parameters in a risk assessment. These factors are needed to estimate exposure to man from ingestion of fish and shellfish and to estimate the dose to aquatic organisms from internal exposure. The following sections describe the factors which can influence the CF, and present CFs from a number of field and laboratory studies. CFs were also derived from the site specific data collected as part of the CSA (1991) Study. CFs are presented only for fish, crustaceans and molluscs, because they represent the most important exposure pathways to man. Generic

concentration factors are also presented, and their usefulness in dose assessment is discussed.

# 5.2 VARIABLES AFFECTING THE CONCENTRATION FACTOR (CF)

The values reported in the literature for the concentration factor have a considerable range. There are a number of factors which contribute to this variation, and they should be considered when using generic values in risk assessment studies. Important influences on the concentration factor include inter-specific differences, differences between organs, the effects of environmental factors, the extent to which the organism is in equilibrium with its surroundings, the relationship between the concentration factor and the concentration of the radionuclide in the water, and the presence of chemically similar elements. These factors contribute to the variation in reported concentration factors and are discussed below.

# Species and Trophic Level Differences

Radium uptake varies among groups of organisms (fish, molluscs, crustaceans) and among species in a group. Differences in habitat, food preference and position in the food web account for some of the differences in concentration factors noted between species. In general, the concentration factors of radionuclides decrease at higher trophic levels.

Swanson (1983) studied the levels of radionuclides in fish from lakes affected by effluents from a uranium mine and mill. Concentration factors for radium varied among species in the same lake (Table 5-1). Small bottom-feeding forage fish (trout-perch, nine spine stickleback, spottail shiner and lake chub) generally had greater radium levels than the larger species. Cisco, a planktivorous species, accumulated smaller amounts of radium than the bottom-feeding foragers. Among the larger fish (white sucker, lake whitefish and lake trout), radium levels were highest in the bottom feeding white suckers, moderate in the

Table 5-1. Concentration Factors for Whole Fish from Beaverlodge Lake (1.5 pCi/1; 0.056 Bq/1) [Source: Swanson, 1983]

Species	Concentration Factor	-
nine spine stickleback	387	
trout-perch	1473	
cisco	53	
spottail shiner	986	
lake chub	187	
white sucker	287	
lake whitefish	33	
lake trout	13	

omnivorous lake whitefish, and lowest in the lake trout feeding mainly on cisco. The inter-specific differences in concentration factors suggest that use of a single concentration factor to represent all species in a group may not always be appropriate.

#### Differences Between Organs

Certain radionuclides preferentially concentrate in particular organs of aquatic organisms. Radium is chemically similar to calcium and concentrates in bone and shell. Table 5-2 shows the differences in the concentration factor for radium in various parts of three species of fish from a lake contaminated by effluent from a uranium mill and mine (Swanson, 1983). In all three species, bone accumulated the most radium, flesh the least.

Table 5-3 gives the concentration factors for radium in marine molluscs (Iyengar, 1984). In all cases, the shell accumulated more radium than the soft edible parts of the animal. These studies all suggest that concentration factors based on the radium content of the whole organism can overestimate the level of radium in the edible portion.

#### Equilibrium with Surroundings

The concentration factor method assumes that the organism is in chemical equilibrium with its surroundings. The time required for equilibrium to be attained depends on the half-life of the radionuclide and the biological half-life of the element in the organism (Till and Meyer, 1983). For fresh water mussels, Jeffree observed that the radium concentration in flesh continued to increase with age in mature animals from a single location (Jeffree, cited in Williams, 1984). 28-day experiments in which radium uptake was induced also failed to reach equilibrium (Jeffree, cited in Williams, 1984). These studies all suggest that for situations in which the environmental concentrations change over time, organisms will not reach complete equilibrium with -----

Table 5-2. Concentration Factors for Various Parts of Fish from Beaverlodge Lake (1.5 pCi/l; 0.056 Bq/l) [Source: Swanson, 1983].

Species	Portion	Concentration Factor	
1.5.	flesh	12	
white sucker	skin	31	
	bone	1793	
	whole	287	
lake whitefish	flesh	3	
	skin	93	,
	bone	360	
	whole	33	
lake trout	flesh	1	
	skin	20	
	bone	100	
	whole	13	

Table 5-3. Concentration Factors for Radium in Marine Molluscs (0.041 pCi/1; 0.0015 Bq/l) [Source: Iyengar, 1984]

Species	Portion	Concentration Factor
oyster	soft parts	50
(Ostrea sp.)	shell	500
green mussel	soft parts	46
( <u>Perna viridis</u> )	shell	419
snail	soft parts	63
( <u>Thais</u> sp.)	shell	156
snail	soft parts	44
( <u>Petalla radiate</u> )	shell	256

their surroundings. Mobile organisms such as fish may also fail to reach equilibrium.

## Major Ion Effects

The presence of major ions in solution influences the biological uptake of non-nutrient trace substances because chemically analogous substances compete for uptake and retention by organisms. Radium and calcium are chemically similar, and the amount of calcium available can have an effect on the rate of radium uptake.

De Bortoli and Gaglione (1972) found that for three of four lakes studied in Italy, the concentration of calcium in the water was inversely related to the concentration of radium in fish.

Jeffree found that uptake of radium by the fresh water mussel (<u>Velesunio angasi</u>) was suppressed in water containing 5 mg/l calcium, but not in water containing 0.5 mg/l calcium (cited in Williams, 1984).

Barium is also chemically similar to radium, and was found to reduce the uptake of radium by unicellular algae (Sebesta et al., cited in Williams, 1984).

#### Other Environmental Factors

Environmental factors, including temperature, salinity and pH affect the growth and metabolism of organisms, and consequently the uptake of radium and other radionuclides. Small increases in temperature tend to increase biological activity and the uptake and excretion of radionuclides (Till and Meyer, 1983). In a study of zinc-65 uptake by shellfish, Duke et al. (1969) found that the primary factors affecting the concentration factor were salinity and temperature.

# Relationship Between the Concentration in Water and in Organism

Most applications of the concentration factor method in dose assessment assume a linear relationship between C (the concentration of the radionuclide in water) and Q (the concentration of the radionuclide in the organism). This assumption allows use of a single concentration factor (CF = Q/C) over a range of environmental concentrations. Available data suggest that this assumption is not valid for all groups or species of organisms.

Williams (1984) investigated the relationship between C and Q for several groups of organisms. He found a linear relationship for insects and fresh water mussels. The data for fresh water mussels were for a single species (<u>Velesumio angasi</u>), but the animals had not reached equilibrium with the surrounding water (Jeffree, cited in Williams, 1984).

Williams (1984) pooled data from a number of studies to test the linearity of the relationship between C and Q for fish. This analysis resulted in a highly nonlinear model:

$$CF = 13 C^{-0.32}$$
 (5-2)

Because of the importance of the factors described in this section (e.g. inter-specific differences), it may not be appropriate to pool data for different species of fish. Other studies also suggest that use of a single concentration factor over a range of water concentrations is not appropriate. In the studies by Swanson (1983, 1985), the concentration factor for radium in several species of fish generally decreased with an increase in water concentration.

Data from the CSA (1991) study were used to derive site-specific concentration factors for radium (Section 5.5). These data also suggest a non-linear relationship between C and Q for fish, molluscs and crustaceans.

#### 5.3 GENERIC VALUES AND THEIR APPLICATION

Generic or average values for concentration factors were suggested based on surveys of published data (Thompson et al. 1972, Cherry and Shannon 1974, IAEA 1982). These factors are usually based on the higher values found in the literature. Generic factors are meant for use in radiological assessment models for estimating the dose to man from a number of pathways. Commonly used models contain default concentration factors for a number of radionuclides and groups of organisms. The generic factors suggested by IAEA (1982) are used by many authors and models and are given in Table 5-4.

Site and species specific concentration factors are not usually available, and developing them for a specific site is expensive and time consuming. The variation within and between species, and the many other variables which can affect the concentration factor make using generic factors problematic, but also, in most cases, necessary. In a preliminary risk assessment study the use of generic factors is appropriate. If the resulting estimates are high compared to background or to an acceptable level of risk, detailed site specific data will be needed. The following sections summarize the concentration factors available in the literature and derived from the CSA (1991) study.

#### 5.4 CFS AVAILABLE IN THE LITERATURE

Concentration factors are available for fresh water and marine organisms in a number of published studies. These studies are reviewed here.

#### Fish

Swanson (1983, 1985) studied the concentration of radionuclides in fish in lakes impacted by a uranium mine and mill (Table 5-5). Ra-226

Table 5-4 IAEA Generic Concentration Factors for Radium.

	Concentration Factor
Fresh Water Fish	50
Fresh Water Invertebrates	300
Marine Fish	100
Marine Invertebrates	100
Marine Crustaceans	100

Table 5-5. Ra-226 in Fresh Water Fish (Canada) [Source: Swanson 1983, 1985]

Species	Organ	Conc.in Water Bq/l (pCi/l)	Concentration Factor
nine-spine stickleback	whole	0.056 (1.5)	387
trout perch	whole	0.056 (1.5)	1473
cisco	whole	0.056 (1.5)	53
long nose sucker	whole	0.804 (21.7)	200
spottail shiner	whole	0.056 (1.5) 0.296 (8.0)	986 270
lake chub	whole	0.056 (1.5) 0.296 (8.0) 0.804 (21.7)	187 83 81
lake trout	whole skin bone flesh skin + flesh	0.056 (1.5) 0.056 (1.5) 0.056 (1.5) 0.056 (1.5) 0.056 (1.5)	13 20 100 1 3
lake whitefish	whole skin bone flesh skin + flesh	0.056 (1.5) 0.056 (1.5) 0.056 (1.5) 0.056 (1.5) 0.056 (1.5)	33 93 360 3 20
	skin flesh bone	0.01 (0.27) 0.01 (0.27) 0.01 (0.27)	6 20 390
	skin flesh bone	0.02 (0.54) 0.02 (0.54) 0.02 (0.54)	40 20 200
	skin flesh bone	0.06 (1.62) 0.06 (1.62) 0.06 (1.62)	83 3.3 500

Table 5-5. cont. Ra-226 in Fresh Water Fish

Species	Organ	Conc.in Water Bq/l (pCi/l)	Concentration Factor
white sucker	whole	0.056 (1.5)	280-287
WILL CO DUCKEL	skin	0.056 (1.5)	31
	bone	0.056 (1.5)	1793
	flesh	0.056 (1.5)	12
	skin + flesh	0.056 (1.5)	100
	whole	0.296 (8.0)	318
	whole	0.804 (21.7)	104
	skin	0.01 (0.27)	70
	flesh	0.01 (0.27)	10
	bone	0.01 (0.27)	200
	skin	0.06 (1.62)	400
	flesh	0.06 (1.62)	18
	bone	0.06 (1.62)	1333

levels and concentration factors varied among lakes, species and tissues. Small bottom-feeding fish (trout-perch, nine spine stickleback, longnose sucker, spottail shiner and lake chub) had higher concentration factors for Ra-226 than did larger fish.

Concentration factors in whole fish ranged from 13 in lake trout to 1473 in trout-perch (both in a lake with Ra-226 water concentrations of 0.056 Bq/1; 1.5 pCi/l). Concentration factors were highest for bone (100 to 1793), lowest for flesh (1 to 20).

A study in Australia designed to observe the occurrence of Ra-226 in fish under pre- and post- mining conditions found a wide range of concentration factors (Iyengar, 1984, Table 5-6). In barramundi the concentration factor in tissue ranged from 11 to 646 (water concentration range: 0.0063 to 0.1095 Bq/l; 0.17 to 3 pCi/l). In black bream the range was from 72 to 450 (water concentration range: 0.0074 to 0.041 Bq/l; 0.2 to 1.1 pCi/l). The concentration factor generally declined with an increase in water concentration.

De Bortoli and Gaglione (1972) also reported a wide range of concentration factors for Ra-226 in fresh water fish. In a study of four lakes in Italy, the concentration factor for Ra-226 in Perca fluviatilis ranged from 70 to 228 (Table 5-7). This study also demonstrated the role of water calcium concentrations in radium uptake.

A study in a fresh water stream in India found a much smaller range for the radium concentration factor (Iyengar, 1984). Fish (Ophicephalus sp.) were confined to a section of the stream for one month (average water concentration 0.518 Bq/; 14.0 pCi/l). The concentration factor in muscle ranged from 5 to 15, and in bone from 40-77 (Table 5-8). The small range in concentration factors observed may result from the fish being in equilibrium with the water.

Table 5-9 presents concentration factors for Ra-226 and Ra-228 in marine fish taken off the coast of India (Iyengar et al., 1980). The concentration factor for Ra-226 in muscle ranged from 55 to 130, and

Table 5-6. Ra-226 in Tissue of Fresh Water Fish (Australia) [Source: Iyengar, 1984]

Caraios	Cone	in water	Concentration
Species	Bq/l	(pCi/l)	Factor
barramundi	0.0063	(0.17)	646, 59,59
Darramunur	0.0074	(0.2)	350
	0,0089	(0.24)	249
	0.01	(0.27)	19
	0.0103	(0.28)	36
	0.0136	(0.37)	68
	0.017	(0.46)	152
	0.0172	(0.46)	43
	0.0294	(0.8)	440
	0.034	(0.92)	11
	0.0888	(2.4)	50
	0.1095	(3.0)	78
til la lawaam	0.0074	(0.2)	450
black bream	0.0296	(0.8)	125
	0.03	(0.81)	123
	0.0405	(1.1)	73
	0.041	(1.1)	72

Table 5-7. Ra-226 in Lake Water Fish, <u>Perca fluviatilis L.</u> (Italy) (de Bortoli and Gaglione, 1972)

Lake	Water Cond	centration (pCi/1)	Concentration Factor
1	0.00052	(0.014)	228
2	0.00074	(0.02)	70
3	0.00056	(0.015)	139
4	0.00052	(0.014)	128

Table 5-8. Ra-226 Distribution in Ophiocephalus sp., (Fresh Water, India). [Source: Iyengar, 1984].

Catch No.	Water Concentration Bq/l pCi/l	Concentra MUSCLE	ation Factor BONE
	nd/I boi/I	MOSCLE	PONE
1	0.518 (14.0)	5	40
2	0.518 (14.0)	11	70
3	0.518 (14.0)	11	65
4	0.518 (14.0)	15	77
5	0.518 (14.0)	15	72
6	0.518 (14.0)	11	66

Table 5-9. Concentration Factors for Ra-226 and Ra-228 in Marine Fish (India) [Source: Iyengar et al., 1980].

Species	Portion	Ra-226 Water bq/l (pCi/l)	Ra-226 CF	Ra-228 Water bq/l (pC1/l)	Ra-228 CF
sole	Muscle Bone	0.0015 (0.04)	55 370	0.0119 (0.32) 0.0119 (0.32)	21 160
mackerel	Muscle Bone	0.0015 (0.04)	120	0.0119 (0.32) 0.0119 (0.32)	63 700
ray	Muscle Bone Liver	0.0015 (0.04) 0.0015 (0.04) 0.0015 (0.04)	65 -	0.0019 (0.32) 0.0019 (0.32) 0.0019 (0.32)	- 98 72
oil sardine	Muscle Bone	0.0015 (0.04)	130 610	0.0019 (0.32) 0.0019 (0.32)	180

from 65 to 610 in bone (water concentration: 0.0015 Bq/l; 0.04 pCi/l). Concentration factors for Ra-228 ranged from 21 to 63 in muscle and from 98 to 700 in bone (water concentration: 0.0119 Bq/l; 0.32 pCi/l). The concentration factors were generally higher for Ra-226 than Ra-228 which had a higher concentration in water.

Table 5-10 summarizes the range of concentration factors for fish in the reviewed literature.

## Molluscs and Crustaceans

Davy and Conway (1974) studied the concentration of Ra-226 in fresh water mussels in Australia. The concentration factor was extremely variable (10 to 12100) (Table 5-11), possibly because the animals do not reach equilibrium (Jeffree, cited in Williams, 1984).

Concentration factors for Ra-226 in marine molluscs were published by Iyengar (1984). The range of concentration factors in these animals was small (Table 5-12). The concentration factor for Ra-226 in the soft parts of the molluscs ranged from 44 to 50, and in the shell from 156 to 500 (water concentration: 0.0056 - 0.016 Bq/1; 0.15 - 0.4 pCi/1).

Table 5-13 presents concentration factors for both Ra-226 and Ra-228 in marine molluscs (Iyengar et al., 1980). The concentration factor for Ra-226 in the soft parts of the shellfish ranged from 73 to 140 (water concentration: 0.0015 Bq/1; 0.04 pCi/1). The concentration factor for Ra-228 ranged from 130 to 170 (water concentration: 0.0119 Bq/1; 0.32 pCi/1).

Marine crustaceans were also studied by Iyengar et al. (1980). For Ra-226 the concentration factor in the muscle of crustaceans ranged from 80 to 210, and in the exoskeleton from 220 to 830 (Table 5-14). The range for Ra-228 was from 35 to 360 in muscle and from 230 to 800 in exoskeleton. Table 5-15 summarizes the range of concentration factors for molluscs and crustaceans in the reviewed literature.

Table 5-10. Fish . Ranges of Concentration Factors in the Reviewed Literature.

	Water Bq/l (pCi/l)	Whole	Flesh	Bone
Fresh Water -Canada [Swanson 1983, 1985]	0.01-0.06(0.27-1.6)	13 - 1473	1 - 20	100 - 1793
Fresh Water -Australia [Iyengar, 1984]	0.0063-0.1095(0.17-3.0)	:	11 - 646	:
Fresh Water · Italy [de Bortoli and Gaglione, 1972]	0.00052-0.00074(0.014-0.02)	70 - 228	;	;
Fresh Water - India [Iyengar, 1984]	0.518 (14.0)	:	5 - 15	40 - 77
Marine - India [Iyengar et al., 1980] (Ra-226)	0.0015(0.04)	;	55 - 130	65 - 610
Marine - India [Iyengar et al., 1980] (Ra-228)	0.0119(0.32)	1 .	21 - 63	98-700
Overall Range	0.00052-0.518(0.014-14)	13 - 1473	1 - 646	40 - 1793

Table 5-11. Ra-226 in Fresh Water Mussels (Alligator Rivers Area, Australia) [Source: Davy and Conway, 1974]

	Conc in v	ater	Concentration
	Bq/l	(pCi/1)	Factor
	0.0096	(0.26)	230
	0.0137	(0.37)	12100
	0.0144	(0.47)	1040
	0.0178	(0.48)	685
	0.037	(1.0)	1530
	0.0389	(1.05)	85
	0.0681	(1.8)	38
	0.111	(3.0)	10
•	0.2257	(6.1)	23
	0.3127	(8.4)	227

Table 5-12. Concentration Factors in Marine Molluscs (India) [Source: Iyengar, 1984]

Species	Organ/ body part	Conc in t		CF
		Bq/1	pCi/l	
oyster	soft parts	0.0016	(0.043)	50
( <u>Ostrea sp</u> .)	shell	0.0016	(0.043)	500
green mussel	soft parts	0.0016	(0.043)	46
( <u>Perna viridis</u> )	shell	0.0016	(0.043)	419
clam	soft parts	0.0016	(0.043)	-
( <u>Meretrix sp</u> .)	shell	0.0016	(0.043)	200
snail	soft parts	0.0056	(0.15)	63
( <u>Thais sp</u> .)	shell	0.0056	(0.15)	156
snail	soft parts	0.0016	(0.043)	44
( <u>Petalla radiate</u> )	shell	0.0016	(0.043)	256

Table 5-13. Concentration Factors for Ra-226 and Ra-228 in Marine Molluscs (India) [Source: Iyengar et al., 1980].

(0.32) (0.32) (0.32)	PORTION Ra-226 Ra-226 Ra-228 Water Uater GF Water Bq/l (pCi/l) Bq/l (p	PORTION         Ra-226 Water (GF Water Bq/l (pCi/l))         Ra-226 Water Bq/l (pCi/l)           soft parts         0.0015 (0.04)         140         0.0119 (0.32)           soft parts         0.0015 (0.04)         73         0.0119 (0.32)           shell         0.0015 (0.04)         -         0.0119 (0.32)	•					
soft parts       0.0015 (0.04)       140       0.0119 (0.32)         soft parts       0.0015 (0.04)       73       0.0119 (0.32)         shell       0.0015 (0.04)       -       0.0119 (0.32)	soft parts 0.0015 (0.04) 140 0.0119 (0.32) soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) -	soft parts 0.0015 (0.04) 140 0.0119 (0.32) soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) - 0.0119 (0.32)	SPECIES	PORTION	Ra-226 Water Bq/l (pCi/l)	Ra-226 CF	Ra-228 Water Bq/l (pCi/l)	Ra-228 CF
soft parts       0.0015 (0.04)       140       0.0119 (0.32)         soft parts       0.0015 (0.04)       73       0.0119 (0.32)         shell       0.0015 (0.04)       -       0.0119 (0.32)	soft parts 0.0015 (0.04) 140 0.0119 (0.32) soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) - 0.0119 (0.32)	soft parts 0.0015 (0.04) 140 0.0119 (0.32) soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) - 0.0119 (0.32)						
soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) - 0.0119 (0.32)	soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) - 0.0119 (0.32)	soft parts 0.0015 (0.04) 73 0.0119 (0.32) shell 0.0015 (0.04) - 0.0119 (0.32)	clam	soft parts	0.0015 (0.04)	140	0.0119 (0.32)	170
solution 0.0015 (0.04) - 0.0119 (0.32)	shell 0.0015 (0.04) - 0.0119 (0.32)	shell 0.0015 (0.04) - 0.0119 (0.32)	) (	the trace	0.0015 (0.04)	73	0.0119 (0.32)	130
			oystei	sort parts shell	0.0015 (0.04)	1	0.0119 (0.32)	100

Table 5-14. Concentration Factors for Ra-226 and Ra-228 in Marine Crustaceans (India) [Source: Iyengar et al., 1980].

)					
SPECIES	PORTION	Ra-226 Water Bq/l (pCi/l)	Ra-226 CF	Ra-228 Water Bq/l (pCi/l)	Ra-228 CF
crab ( <u>Scylla serrata</u> )	Muscle Gills	0.0015 (0.04)	110 340	0.0119 (0.32) 0.0119 (0.32)	180 160
crab ( <u>Neptanus</u> sp.)	Muscle Gills Exoskel	0.0015 (0.04) 0.0015 (0.04) 0.0015 (0.04)	- 11000 830	0.0019 (0.32) 0.0019 (0.32) 0.0019 (0.32)	35 12000 800
mole crab	Muscle Exoskel	0.0015 (0.04) 0.0015 (0.04)	210 220	0.0019 (0.32) 0.0019 (0.32)	68 230
prawn	Muscle Muscle Exoskel	0.0015 (0.04) 0.0015 (0.04) 0.0015 (0.04)	80	0.0019 (0.32) 0.0019 (0.32) 0.0019 (0.32)	360 56 300

Table 5-15. Molluscs and Crustaceans - Ranges of Concentration Factors in the Reviewed Literature.

Mollison Daret H	Bq/l (pCi/l)	Joic Farts/Muscle	Shell/Exoskeleton
Australia [Davy and Conway, 1974]	0.0096-0.3127 (0.26-8.4)	10 - 12100	
Molluscs - Marine India [Iyengar, 1984]	0.016 (0.43)	44 - 63	156 - 500
Molluscs - Marine India (Ra-226) [Iyengar et al., 1980]	0.0015 (0.04)	73 - 140	
Molluscs - Marine India (Ra-228) [Iyengar et al., 1980]	0.0119 (0.32)	130 - 170	1001
Crustaceans - Marine India (Ra-226) [Iyengar et al., 1980]	0.0015 (0.04)	80 - 210	220 - 830
Crustaceans - Marine India (Ra-228) [Iyengar et al., 1980]	0.0119 (0.32)	35 - 360	230 - 800
Molluscs - Overall Range	0.0015 - 0.313 (0.04-8.4)	$10 - 12100^2$	100 - 500
Crustaceans - Overall Range 0	0.0015 - 0.0119 (0.04-0.32)	35 - 360	
1: only one species 2: 12100 is for fresh water mus molluscs.	mussels in Australia; excluding these mussels gives a range of 1-360 for	these mussels gives a	range of 1-360 for

#### 5.5 CONCENTRATION FACTORS DERIVED FROM THE CSA (1991) FIELD STUDY

Data collected included Ra-226 and Ra-228 concentrations in water, sediment, fish, shellfish and crustaceans at three produced water outfalls in Louisiana.

To provide a comparison with background levels, samples were also taken from two reference stations for each site, located 1/2 mile from the outfall. Biological samples were taken within 50 feet of the discharge point. The outfalls at Sites 2 and 3 discharge directly into a canal. At low tide Station 1 may discharge onto dry land, and then into a canal. All mollusc samples were oyster (Crassostrea virginica), and all fish samples were seatrout (Cynoscion sp.). Crustaceans sampled at Site 2 were crabs (Callinectes sp.) and at Sites 1 and 3 were shrimp (Penaeus sp.). Oysters were removed from the shell before analysis. Fish and crustacean samples were whole-body samples, including bone, skin and exoskeleton.

Concentration factors were calculated for each site/organism combination:

Tables 5-16 and 5-17 give the concentration data and concentration factors for Ra-226 at the three sites and the six reference stations. Tables 5-18 and 5-19 present the same data for Ra-228. The concentration factor was calculated for both the concentration at the outfall, and for the average concentration at 50 feet because the biological samples were taken within (not at) 50 feet of the discharge.

Measured Ra-226 Concentrations in Water and Organisms Reported in CSA (1991) Study. Table 5-16.

Station	Discharge (pC1/1)	Conc. at 50 feet (pG1/1)	Fish (pCi/g)	Oysters (pCi/g)	Crustaceans (pCi/g)
Site 1	228.9	0.43	0.014	0.007	0.07
Reference 1-1	0.25	1 1 1	ND	0.001	0.014
Reference 1-2	0.2	!	0.005	0.004	0.024
Site 2	110.6	0.53	0	0.008	0.07
Reference 2-1	0.7	;	0.002	0.004	ND
Reference 2-2	0.5	1 1 1	0.031	0.001	0.07
Site 3	251.9	0.92	0 041	Ş	2010
Reference 3-1				GN	0.016
Reference 3-2	0.7	1	0.03	ND	0.077

ND: no data : average of measured concentration at three stations, each 50 feet from discharge

Calculated Ra-226 Concentration Factors $^1$  for Organisms Sampled in CSA (1991) Study. Table 5-17.

Station	Discharge (pCi/l)	Conc. at 50 feet (pCi/1) <sup>2</sup>	Fish CF	Oysters CF	Crustaceans CF
Site 1	228.9	0.43	(0.06-32.6)	(0.03-16.3)	(0.31-163)
Reference 1-1	0.25	3 2 1	ND	4.0	96.0
Reference 1-2	0.2	!	25.0	20.0	120.0
Site 2	110.6	0 53	c	11 21 20 07	7000
Reference 2-1	0.7		2.9	(0.0/-13.1)	(O.0-132) ND
Reference 2-2	0.5	:	62.0	2.0	140.0
Site 3	251.9	0.92	(0.16-44.6)	ND	(0.5-135.9)
Reference 3-1	0.7		25.7	ND	22.9
Reference 3-2	0.7	1 1	42.9	ND	110.0

ND: no data 1. concentration factors for sites were calculated for both the discharge concentration and the gverage concentration 50 feet from the discharge : average of measured concentration at three stations, each 50 feet from discharge

Measured Ra-228 Concentrations in Water and Organisms Reported in CSA (1991) Study. Crustaceans (PCi/g) 0.094 0.004 0.025 0.025 0.033 0.243 0.058 0.045 ND Oysters (pCi/g) 0.003 0.036 0.011 0.011 0.003 0.01 ON-g Fish (pC1/g) 0.005 0.021 0.022 0.012 0.058 0.017 0.03 0.13 ND Conc. at 50 feet (pCi/l)<sup>1</sup> 0.93 0.2 4.0 Discharge (pCi/l) 383.0 244.4 254.9 0.15 10.3 3.3 Reference 1-1 Reference 1-2 Reference 2-1 Reference 2-2 Reference 3-1 Reference 3-2 Table 5-18. Station Site 1 Site 2 Site 3

ND: no data l: average of measured concentration at three stations, each 50 feet from discharge

Calculated Ra-228 Concentration Factors for Organisms Sampled in CSA (1991) Study. Table 5-19.

Station	Discharge (pCi/l)	Conc. at 50 feet (pCi/l) <sup>2</sup>	F <b>ish</b> CF	Oysters CF	Crustaceans CF
Site 1	383.0	0.93	(0.013-5.4)	(0.008-3.2)	(0.25-101)
Reference 1-1	0.15	; ;	ND	240	26.7
Reference 1-2	0	1 1	NC	NC	NC
		4			
Site 2	244.4	0.2	(0.09-110)	(0.045-55)	(0.102-125)
Reference 2-1	0	1 2	NC	NC	ND
Reference 2-2	0	i i	NC	NC	NC
Site 3	254.9	4.0	(0.05-3.0)	ND	(0.95-60.8)
Reference 3-1	3,3	1 1	17.6	ND .	17.6
Reference 3-2	10.3	1 1 1	1.7	UN	4.4

ND: no data

NC: not calculated, water concentration — 0 pCi/l !: concentration factors for sites were calculated for both the discharge concentration and the gverage concentration 50 feet from the discharge : average of measured concentration at three stations, each 50 feet from discharge

# 5.6 SUMMARY AND SUGGESTED CFS

The generic concentration factors suggested by IAEA (1976) (100 for marine fish, molluscs and crustaceans) are consistent with the ranges of concentration factors found in the literature. However, the values reported in the literature are based on samples taken from water with relatively low levels of Ra-226 (0.00052 - 0.518 Bq/1; 0.014 - 14.0 pCi/l).

Based on data available in the literature (Section 5.4) and an independent analysis of data collected as part of the CSA study, it can be concluded that the concentration factor for fish, molluscs, and crustaceans is affected by the concentration of radium in the water. The concentration factors calculated for the reference sites in the CSA (1991) study (Ra-226: 0.2 to 0.7 pCi/l, Ra-228: 0.0 to 10.3 pCi/l) were smaller than the IAEA concentration factors of 100 for salt water fish, molluscs and crustaceans. The concentration factors calculated for the CSA (1991) data suggest that at the relatively high concentrations encountered in produced water discharge, the generic IAEA concentration factors may be over-estimates. The concentration factors reported for fish and crustaceans in Tables 5-17 and 5-19 are also over-estimates because they include bone and exoskeleton, which concentrate radium, and which people do not eat.

The CSA data set does not allow development of an empirical model for calculation of the concentration of radium in organisms given the concentration in water because biological samples were not taken exactly at water sampling points (biological samples were taken within 50 feet of the discharge). The concentration factor for radium does vary with water concentration, and concentration factors are smaller for higher water concentrations of radium. Many factors influence the concentration factor for radium in fish, molluscs and crustaceans. The IAEA factors are conservative and overestimate the concentration of radium in fish and shellfish because they do not consider the dependence

of the concentration factor on water concentration, and because they do not separately consider radium in bone, shell and exoskeleton. Because of the complexity of these factors, generic CFs are only appropriate for use in a screening-level assessment. In this case, some site-specific data are available, and the screening level assessment will use both conservative, generic CFs and the actual concentrations of radium measured in fish and shellfish measured at the three CSA sites. More data relating radium concentrations in water to the resulting levels in food organisms are needed.

# Section 6 EFFECTS OF RADIATION ON AQUATIC ORGANISMS

#### 6.1 RADIATION EFFECTS

Radiation standards for people were developed to protect the individual. The principal concern for aquatic organisms is the protection of the population and the ecosystem, rather than the individual animal.

A distinction is made between stochastic and non-stochastic effects. These are defined as follows (ICRP, 1979):

"Stochastic" effects are those for which the probability of an effect occurring, rather than its severity, is regarded as a function of dose, without threshold. "Non-stochastic" effects are those for which the severity of the effect varies with the dose, and for which a threshold may occur. At the dose range involved in radiation protection, hereditary effects are regarded as being stochastic. Some somatic effects are stochastic; of these, carcinogenesis is considered to be the chief somatic risk of irradiation at low doses and therefore the main problem in radiation protection."

There is little information available concerning induction of cancer and genetic effects in aquatic organisms. Most studies are concerned with induction of non-stochastic, somatic effects. These effects include increases in mortality, and pathophysiological, developmental and reproductive effects. These are the effects emphasized in this review. There are a few studies of stochastic, genetic effects in aquatic organisms.

Most studies were performed in the laboratory, and describe the effects of irradiation on individual animals. The few studies of effects in natural populations, along with a consideration of the natural control mechanisms of populations are discussed in Section 6.4.

Most available studies of radiation effects deal with acute exposures of organisms to ionizing radiation. This is because of the difficulties in detecting radiation effects at low-dose rates. However, the effects of exposure to radium isotopes in produced water are likely to result from chronic, low level exposure (see estimated doses, Section 8). This review briefly mentions acute effects but concentrates on the effects of chronic, low-level exposure. Anderson and Harrison (1986) have recently reviewed the literature on the effects of radiation on aquatic organisms, and the following text and tables draw heavily from that document.

The biological effect of a radionuclide is related to the absorbed dose (or the dose rate). The absorbed dose is the amount of energy imparted to matter. An absorbed dose of 100 erg/gm is called 1 rad. In the SI system the absorbed dose unit is 1 Joule per kilogram (J/kg), and 1 J/kg is called the Gray (Gy). An absorbed dose of 1 rad is equal to 0.01 Gy (1 Gy = 100 rads). Harmful levels of radiation doses are generally expressed in terms of rads. For example, over a hundred rads must be imparted in a short period of time to a substantial portion of the body before most individuals will show significant clinical symptoms (Saenger, 1963). Occupational doses are not allowed to exceed a few mrad per hour (1 mrad =  $1 \times 10^{-3} \text{ rad}$ ).

The absorbed dose associated with the concentration of a radionuclide in water is dependent on a number of factors, including the amount of water taken up by an organism, the distribution of the radionuclide in tissue, and the energy of the particles emitted during decay.

## 6.2 ACUTE EFFECTS

Acute effects are usually observed in studies which deal with survival of organisms after a single exposure to relatively high doses. End-points other than mortality which were used to indicate acute radiation damage include growth rate, reproduction, fecundity, and

physiological and behavioral reactions. Anderson and Harrison (1986) have reviewed the effects of acute radiation exposure on aquatic organisms.

Mortality is usually expressed in terms of the  $LD_{50/30}$ , which is the dose killing 50% of the exposed organisms within 30 days. Anderson and Harrison (1986) compiled data on the range of lethal doses for fish and invertebrates. The range of lethal doses for adult fish is 375 to 55,000 rad. 50% mortality of fish embryos was demonstrated for exposures as low as 16 R (silver salmon, 1 cell stage, Bornham and Welander, 1963). For invertebrates, lethal doses range from 210 rad to above 50,000 rad.

The 30-day time period was chosen based on the survival rates of small mammals, and may not be long enough to allow the observation of effects in aquatic organisms (Anderson and Harrison, 1986; Woodhead, 1984).

 $LD_{50}$  values are also difficult to compare and evaluate because they may be strongly influenced by factors such as temperature, salinity, lifestage, dose rate and species (Anderson and Harrison, 1986). The  $LD_{50}$  for fish, for example, has a range of over three orders of magnitude and depends strongly on the lifestage of the organism.

#### 6.3 EFFECTS OF CHRONIC EXPOSURE

#### Mortality

Table 6-1 presents a summary of studies of effects of chronic radiation exposure on mortality in fish and invertebrates.

Donaldson and Bonham (1964) found no significant increase in mortality in salmon (Oncorhynchus tschawytscha) exposed to 0.54 R/d (equivalent to 5.4 mGy/d) for approximately 20 days (up to release of smolts). Irradiated females returned to spawn in greater numbers than

Induction of Mortality in Fish and Invertebrates From Chronic Exposure to Radiation 1. Table 6-1

		•	
Organism/lifestage	Radiation regime <sup>a</sup>	Comments	References
FISH Oncorhynchus tschawytsca embryos (Chinook salmon)	5 mGy/d (0.5 R/d) for 61 - 69 days (Co-60)	No significant differences in mortality and greater return of females	Donaldson and Bonham, 1964, 1970
Oncorhyncus tschawytsca (Chinook salmon)	5-475 mGy for 71 - 86 days (Co-60)	Lower return of spawning adults at dose rates > 95 mGy/d	Hershberger et al.,1978; Woodhead, 1984
Poecilia reticulata 1-week old juveniles (guppy)	50-1000 mCi/l 240-4700 rad (tritium)	No significant increase in mortality	Erickson, 1973
Poecilia reticulata neonates to adult (guppy)	40.8 - 305 mGy/d (Co-60)	No effect on survival of offspring	Woodhead, 1977
INVERTEBRATES Callinectes sapidus adults (blue crab)	3.2-29.0 rad/h 70 d (Co-60)	Decreased survival only at highest dose rate	Engel, 1967
<u>Mercenaria mercenaria</u> juvenile (clam)	0.06 - 370 mGy/hr (0.006 - 37 rad/h) 14 months (Co-60)	Deleterious effects on growth Baptist et al., and survival only at highest 1976 dose rate (16-37 rad/hr)	Baptist et al., 1976
Agopecten irradians juveniles (scallop)	0.06-370 mGy/hr (0.006 - 37rad/h) 3 months (Co-60)	No deleterious effects on growth and survival	Baptist et al., 1976
1 - Table modified from Ande	Table modified from Anderson and Harrison 1986		

1 - Table modified from Anderson and Harrison, 1986 an - anadromous species, fw - fresh water species, ma - marine species

controls. In later studies, lower returns of spawning adults were observed after exposure to >10R/d (equivalent to >95 mGy/d) (Hershberger et al., 1978, Woodhead, 1984).

Exposure of embryos of the guppy <u>Poecilia reticulata</u> to 0.05 to 1 mCi/ml of tritium (total dose 340 - 4700 rad) resulted in no increase in mortality (Erickson, 1973). Lifetime exposures to the guppy (<u>Poecilia reticulata</u> of 40.8 to 305 mGy/d (4.08-30.5 rad/d) resulted in no effect on the survival of offspring (Woodhead, 1977).

Juveniles of the clam <u>Mercenaria mercenaria</u> exposed to 0.06 to 370 mGy/hr (0.006 to 37.0 rad/hour) for fourteen months exhibited decreases in growth and survival only at the highest dose rate (160 - 370 mGy/hr; 16-37 rad/hr) with 10% survival (Baptist et al., 1976). Juveniles of the scallop <u>Agopecten irradians</u> showed no effects after exposure to the same dose rates for three months (Baptist et al., 1976).

## Pathophysiological Effects

Table 6-2 summarizes the pathophysiological effects of chronic radiation exposure in fish. No data are available describing such effects in invertebrates.

In juvenile and yearling trout exposed as embryos to water with tritium concentrations of 37 and 370 Bq/l (1 and 10  $\mu$ Ci/l) antibody synthesis against the bacterium <u>Chondrococcus columnaris</u> was reduced (Strand et al., 1973a).

Mosquitofish (<u>Gambusia affinis</u>) exposed to dose rates of 360 to 720 mGy/d (36-72 rad/d) for 128 days showed mild hemopoietic atrophy in kidneys and spleen in some fish. No damage was observed at dose rates of 120-1300 mGy/d (12-130 rad/d) for 37 days (Cosgrove et al., 1975).

Table 6-2 Induction of Pathophysiological Changes in Fish from Chronic Exposure to Radiation<sup>1</sup>.

Endpoint	Organism/lifestage	Radiation regime	Comments	References
Immune Response: antibody synthesis against Chondrococcus	Salmo gairderii <sup>fw</sup> embryos (rainbow trout)	1,10 mCi/l 20 d (tritium)	Developing embryos demonstrated depressed immune response	Strand et al.,1973a
Hemopoieses	<u>Gambusia affinis</u> adults (mosquitofish)	0.5-5.43 rad/h 336-9216 rad (Co-60)	No demonstrable Gos damage to hemopoetic ettissues after 37 days at 5.43 rad/hr; mild hemopoietic atrophy in kidney and spleen in some fish after 128 d at 1.5 and 3.0 rad/h	Gosgrove et al., 1975 t me and

<sup>1 -</sup> Table modified from Anderson and Harrison, 1986. fw - fresh water species

#### Reproductive Effects

Effects on fertility have been observed at relatively low dose rates in fish and aquatic invertebrates. The lowest dose rate at which effects of chronic radiation exposure on fertility on aquatic organisms has been demonstrated is between 0.59 R/day (Trabalka and Allen, 1977) and 10 R/day (Bonham and Donaldson, 1972) (Anderson and Harrison, 1986). Table 6-3 presents a summary of the reproductive effects of chronic radiation in fish and invertebrates.

Natural populations of the mosquitofish (<u>Gambusia affinis</u>) in White Oak Lake at Oak Ridge National Laboratory were studied (Trabalka and Allen, 1977; Blaylock and Mitchell, 1969). The population was exposed to chronic radiation for at least 60 generations with the dose rates varying from  $2.5 \times 10^{-2}$  to  $>4.0 \times 10^{-1}$  mGy/h (0.06 to >1 rad/d). The population was found to have an increased incidence of dead embryos as well as an increase in fecundity over the control population.

Woodhead (1977) studied the fecundity of the guppy (<u>Poecilia</u> reticulata) at 6, 12 and 40.8 rad/d and found a reduction in total fecundity at all dose rates. Purdom and Woodhead (1973) exposed the guppy to similar dose rates and found an increase in sterility at 12 rad/d and total sterility at 40.8 rad/d. The fecundity of the pairs remaining fertile was not affected.

Hyodo-Taguchi (1980) observed an increased frequency of unfertilized eggs and sterile individuals in <u>Oryzias latipes</u> exposed to 6.8 R/d for 60 days. No effects were observed at 2.9 R/d.

Embryos of the Chinook salmon (<u>Onchorhynchus tschawytscha</u>) were exposed to 0.5 to 50 R/d for 80 days (Bonham and Donaldson, 1972). Gonad development was retarded in smolts receiving 10 or more R/day.

Table 6-3 Induction of Reproductive Changes in Fish and Invertebrates From Chronic Exposure to  ${
m Radiation}^1.$ 

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Organism/lifestage	Radiation regime <sup>a</sup>	Comments Ref	References
FISH			
Gambusia affinis all stages (mosquitofish)	White Oak Lake N. 0.06->1 rad/d . lifetime 0.59 rad/day sample	No decrease in fecundity observed - increased embryo mortality le	Trabalka and Allen, 1977
Poecilia reticulata 0-3 d embryos to adult (guppy)	4.08-30.5 rad/d, number of days varied (Cs-137)	Total fecundity was markedly reduced at all dose rates because of decrease in mean actual brood size and an increase in temporary and permanent infertility	Woodhead, 1977
Poecilia reticulata 0-3 d embryos to adult (guppy)	6.0,12.0,40.8 rad/d, number number of days varied (Cs-137)	One pair out of 6 and 5 out of 7 were sterile at 6 and 12 rad/d; total doses were 4000 and 8000 rad; at 40.8 rad/d, sterility occurred at 5000 rad; fecundity was not affected in pairs remaining fertile	Purdom and Woodhead, 1973
Oryzias latipes adult males (medaka)	1.3-84.3 R/d 60 - 120 days (Cs-137)	408 R delivered at 6.8 R/d increased sterility and number of unfertilized eggs	Hyodo- Taguchi, 1980
Onchorhynchus tschawytscha <sup>an</sup> embryos (Chinook salmon)	0.5-50 R/d 80 days, (Co-60)	Gonadal development was retarded at 10 R/d or more	Bonham and Donaldson, 1972

Table 6-3 (cont.)

Organism/lifestage	Radiation regime <sup>a</sup>	Comments	References
INVERTEBRATES			
<u>Physa heterostropha</u> all stages (aquatic snail)	White Oak Lake 0.65 rad/d	Egg capsule production reduced, increased number of eggs per capsule total fecundity similar to controls	Gooley, 1973b
<u>Physa heterostropha</u> adults	24 - 600 rad/d 168 days	Egg and capsule production reduced at at 240 rad/d, but not at 24 rad/d	Cooley and Miller, 1971
Physa heterostropha adults	240-1200 mgy/d for 98 days	Egg production reduced at 240 mGy/d, egg capsule production reduced at >240 mGy/d at 25 C.	Cooley, 1973a

- Table modified from Anderson and Harrison, 1986.

an - anadromous species fw - fresh water species ma - marine species

A natural population of the aquatic snail <u>Physa heterostropha</u> in White Oak Lake exhibited a lower frequency of egg capsule production and an increased number of eggs per capsule at an estimated dose rate of 6.5 mGy/d (0.65 rad/d) (Cooley, 1973b). The overall fecundity of the population was similar to that of the control.

A laboratory study of <u>Physa heterostropha</u> at 25 C showed decreases in egg capsule production and an increase in sterility at 240 rad/d but not at 24 rad/d (Cooley and Miller, 1971). In another laboratory study, effects occurred at dose rates > 24 rad/d at 25 C.

#### Developmental Effects

Different criteria have been used to evaluate the effect of radiation on developing embryos including hatching success, mortality and the frequency of abnormal embryos and larvae (Anderson and Harrison, 1986). The developmental stage at which the dose is received is a major determinant of what the effect will be. Several investigators have demonstrated a trend of decreasing radiosensitivity with increasing development (Woodhead, 1984). Only a few studies are available which use solid sources (or tritium in water) to assess the effect of chronic radiation on the development of aquatic organisms (Table 6-4; Anderson and Harrison, 1986). Experiments which involved exposing the organism to the radionuclide in water are subject to question, because of the potential for bioaccumulation (Woodhead, 1984).

Table 6-4 presents a summary of developmental effects of chronic radiation in fish and invertebrates. Donaldson and Bonham (1964) found a significant increase in opercular defects of smolt at exposures of 33 to 40 R given at 0.5 R/day from fertilization.

Table 6-4 Induction of Developmental Changes in Fish and Invertebrates from Chronic Exposure to  ${
m Radiation}^1.$ 

Organism/lifestage	Radiation regime	Comments	References
Oncorhynchus tschawytscha embryos (Chinook salmon) Oncorhynchus kisutch embryos (coho salmon)	0.5 R/d 33-40 R (Co-60)	Significant increase in opercular defects	Donaldson and Bonham, 1964
Oncorhynchus tschawytscha embryos (Chinook salmon) Oncorhynchus kisutch <sup>an</sup> embryos (coho salmon)	0.5 R/d 33-40 R (Co-60)	In some phases, growth was superior in the irradiated group, greater average weight in the irradiated group	Donaldson and Bonham, 1964
Poecilia reticulata embryos and young (guppy)	0.05- 1 mCi/ml 340-4700 rad for young fish, 90-2500 rad for embryos (tritium)	Doses of 3400 and 4700 rad Erickso for young fish and 300 rad for embryos resulted in lower weights; significant increase in weight in males exposed to 2500 rads as embryos	Erickson, 1973 r ghts; t in embryos
<u>Salmo gairdnerii</u> <sup>Fw</sup> embryos (rainbow trout)	0.01,0.1,1.0, and 10 uCi/ml 25 d (tritium)	No significant decrease in hatching, no significant increase in malformations	Strand et al. 1973b

Table 6-4 (cont.)

Organism/lifestage	Radiation regime	Comments	References
<pre>Gasterosteus aculeatus fw embryos (stickleback)</pre>	0.5,1.0,2.0 mCi/ml 980-3920 rad (tritium)	Significant reduction in mean Walden, 1973 eye diameter in 1.0 and 2.0 mCi/ml water	ın Walden, 1973
Physa heterostropha adults and embryos (aquatic snail)	1,10,25 rad/h 25 weeks (Co-60)	Significant decrease in \$ hatching at 10 rad/hr; no hatching at 25 rad/hr; growth of adults was significantly increased by 10 and 25 rad/h	Cooley and Miller, 1971

<sup>1 -</sup> Table modified from Anderson and Harrison, 1986 an - anadromous species; fw - fresh water species

#### Genetic Effects

The principal genetic effect of low-level radiation in somatic tissue is cancer induction. Potential effects on germ cells are associated with the induction of an increased frequency of heritable disease. This may occur as a result of mutation, chromosomal breakage, chromosomal rearrangement or faulty separation of chromosomes at metaphase causing anueploidy.

The study of cytogenetic effects in aquatic organisms is difficult and studies on the induction of specific locus mutations are limited by lack of knowledge about the genetics of most aquatic organisms and the time and expense involved in conducting these experiments (Anderson and Harrison, 1986). There have been very few genetic studies on aquatic organisms, and data on mutation rates is limited.

Anderson and Harrison (1986) have reviewed the literature describing the genetic effects of radiation on aquatic organisms. Genotoxic effects have been observed in fish and invertebrates at low dose levels. An increased frequency of chromosomal aberrations in cultured fish (<u>Umbra limi</u>) were observed after 50 R exposures (Suyama and Etoh, 1983). Significant increases in sister chromatid exchange and the frequency of chromosomal aberrations have been observed at 60 rad and 200 rad (0.60 and 2.0 Gy) in the marine worm <u>Neanthes</u> arenaceodentata (Harrison at al., 1985).

Most studies involving the induction of specific locus and dominant lethal mutations have used relatively high exposures (500-1000 R) (Anderson and Harrison, 1986). The lowest effect level demonstrated in these studies involved the irradiation of eggs and spermatozoa of the rainbow trout <u>Salmo gairdneri</u> (McGregor and Newcombe, 1972a). A significant increase in the rate of major eye malformations was observed at acute doses of 25 rad (0.25 Gy).

Genetic effects of radiation may also have effects on aquatic populations. Recessive lethal mutations and deleterious genes will accumulate in the gene pool and be expressed in future generations. Effects of radiation on natural populations, including genetic effects are discussed in Section 6.4.

# 6.4 EFFECTS ON AQUATIC POPULATIONS

Most studies reported in the literature were performed in the laboratory and describe the effects of radiation on individual organisms (Section 6.3). For the most part, the effects of radiation exposure on natural populations must be extrapolated from these laboratory experiments.

Effects in aquatic systems at the ecosystem level have been demonstrated only for the large doses received at Eniwetok and Bikini atolls in the Pacific Proving Grounds (Templeton et al. 1971).

There are a few studies of the effects of low-level chronic irradiation on natural populations. Some of these studies were already discussed in Section 6.3, but are mentioned below because they represent the few studies of chronic radiation exposure on natural populations. The major somatic effects of concern are effects on fertility and fecundity. Enhanced mortality of eggs and larvae can affect recruitment and several authors have reviewed the data available on the natural regulatory mechanisms of marine fish to help predict the effects of exposure to radiation. Radiation can also affect the fitness of a population through the induction of genetic effects, and studies demonstrating such effects in natural populations are described here.

# Regulatory Mechanisms of Natural Populations

Since available data suggest that developing gametes, fertilized eggs, and larvae of fish are the most sensitive components of aquatic

ecosystems, there is a concern that chronic, low doses of radiation could adversely affect commercial fisheries. IAEA (1976) and Templeton (1980) examined the possible effects of chronic low level radiation on recruitment, fertility, fecundity and mortality by considering the known regulatory mechanisms of natural populations.

Recruitment for highly fecund species is not directly related to spawning stock size and the mortality rate operating on eggs and larvae varies from year to year. Survival of eggs and larvae depend to a large degree on the availability of food, and a large number of eggs are produced at each spawning  $(10^3 \text{ to } 10^6 \text{ per female}$ , Templeton, 1980). Density dependent mortality reduces fish larvae populations to the level that can be supported by the available food. IAEA (1976) concluded that if mortality is enhanced by low levels of radiation, recruitment to the stocks of highly fecund fish is not likely to be affected, unless the stocks are already at risk due to over-exploitation. The mechanisms controlling recruitment in invertebrates appear similar, except that environmental factors may be more important (Templeton, 1980).

For species with low fecundity (such as sharks and marine mammals), recruitment is closely related to parent stock size. It is not possible to predict the effects on recruitment for these species. However, at low dose rates, it is reasonable to assume that effects will be small compared to fishing pressure (IAEA, 1976).

For commercial fisheries, rates of exploitation as high as 50% are common. In addition, there is mortality due to natural causes. A heavily exploited stock may be subjected to total mortality rates of over 60% per year, yet is still able to replace itself (Templeton, 1980). Any mortality caused by low levels of radiation would probably not be detectable as such (IAEA, 1976).

Other aquatic species have not been studied in the same detail as commercially important fish. There is evidence that other aquatic species also have density dependent regulatory mechanisms (IAEA, 1976)

and a small increase in mortality caused by chronic radiation is unlikely to affect unexploited populations.

# Observations of Effects in Natural Populations

A number of studies were conducted in White Oak Lake, a retention pond for low level waste at Oak Ridge National Laboratory. Three aquatic species were studied in this contaminated environment: the midge, <u>Chironomus tentans</u>, the mosquitofish <u>Gambusia affinis</u>, and the snail <u>Physa hetersotropha</u>.

An increased frequency of chromosome aberrations was found in midge larvae (Chironomus tentans) at dose rates of approximately 6.3 mGy/d (0.63 rad/day). Ten years later the dose rate decreased to 0.3 mGy/d (0.03 rad/d) and an increased frequency of chromosome aberrations was not detected (Blaylock 1966a, 1966b, 1973).

The mosquitofish (<u>Gambusia affinis</u>) was found to have an increased incidence of dead embryos after the population had been exposed to dose rates varying from  $2.5 \times 10^{-2}$  to >4.0 x  $10^{-1}$  mGy/hr (0.06 to >1 rad/d) (Trabalka and Allen, 1977, Blaylock and Mitchell, 1969). The increased frequency of dead embryos and abnormalities was attributed to an increased frequency of recessive lethal and deleterious genes. The irradiated population also had an increase in fecundity over the control population.

The fecundity of the snail population in White Oak Lake (Physa heterostropha) was also studied (Cooley and Nelson, 1970; Cooley and Miller, 1974; Cooley, 1973b). The irradiated population received an estimated dose rate of 6.5 mGy/d (0.65 rad/d), and showed a reduction in the frequency of egg capsule production compared to a control population. However, the egg production by the two populations was similar because the irradiated population produced an increased number of eggs per capsule.

In the North East Irish Sea, at the discharge point for the Windscale fuel reprocessing plant, average long-term exposures of 2.1 x  $10^{-3}$  mGy/hr (maximum of 1.5 x  $10^{-2}$  mGy/hr) had no detectable effects on the population of plaice (<u>Pleuronectes platossa</u>) (Woodhead, 1984).

Several reviews (Blaylock and Trabalka, 1978; IAEA, 1976; NRCC, 1983) have concluded that the increased mutation rate from an exposure of 1 rad/day (10 mGy/day) or less will not have a significant deleterious effect at the population level.

# 6.5 SUMMARY - PREDICTION OF EFFECTS ON AQUATIC ORGANISMS AND POPULATIONS

The available dose-response data describing the effects of chronic low-level radiation on aquatic organisms is variable and hard to summarize. The most sensitive life-stage appears to be fish-fry. Effects on individual aquatic organisms have been detected at dose rates in the range of 1-10 mGy/day (0.1-1.0) rad/day (Anderson and Harrison, 1986).

Natural populations are affected by many factors and effects of low-level chronic radiation are not likely to be detectable. Dose rates less than 10 mGy/day (1 rad/day) are not expected to have significant deleterious effects at the population level.

#### Section 7

A REVIEW OF SELECTED DOSE-RESPONSE MODELS FOR THE EVALUATION OF HUMAN HEALTH EFFECTS OF RADIUM-226 AND RADIUM-228 INGESTION

## 7.1 INTRODUCTION

Ingestion by humans of large amounts of the radionuclides radium-226 and/or radium-228 cause cancer, notably bone sarcomas and possibly other kinds of neoplasms. Radium is a chemical analogue of calcium and is distributed and deposited in the body in a similar manner. Alpha particles, which result from radioactive decay of radium and its daughter nuclides, deposit energy via ionization in the immediate vicinity of the decaying nucleus. Less importantly, energy is also deposited throughout the body by gamma rays from radium and daughter decays. It is the direct or indirect disruption of DNA by the radiation which is implicated in excess cancer risk.

## Models Presented

To assess quantitatively the human health effects of radium ingestion, some sort of mathematical model is necessary. In the following sections, three such models are reviewed. The first two, due to the International Commission on Radiological Protection and the U.S. Environmental Protection Agency, are essentially identical in approach and differ only in detail. They provide almost identical results. Both models depend on indirect evidence. A complicated metabolic model is used to compute body burdens of radium as a function of intake level and then tissue doses are calculated. To obtain excess cancers from dose, coefficients based on atomic bomb survivor data are used. These people were exposed to high levels of gamma radiation, a low linear energy transfer (low-LET) type. Since radium, like calcium, seeks bone, one might expect the largest effects predicted to be bone sarcoma and

leukemia. This is true for ICRP but not for EPA where the greatest risk is predicted in soft tissue. All effects are assumed to vary linearly with intake.

The third model presented uses direct evidence. Using original studies and the recent report by the National Academy of Sciences' Committee on the Biological Effects of Ionizing Radiation (BEIR IV; NAS, 1987), dose-response coefficients are given based on observations of people who suffered large intake of radium-226 and/or radium-228. The results differ importantly from those of the other two models. Primarily, bone cancer risk at low doses is much lower and varies quadratically with intake level, and excess leukemias are not observed.

#### Length of Exposure

Risk assessments of ingesting food or water contaminated with radionuclides usually assume that all of the intake is from the contaminated source and that consumption takes place over all of an individual's lifetime. In the case of a large population exposed to wide contamination, these may be prudent but conservative assumptions, because some people may not leave the area of contamination in their daily movements or during their lifetimes.

In the case of an isolated, low volume contamination this is not the case. Children and working adults especially can be expected to be away from the contamination much of the day. A given individual cannot be expected to live out his entire life using the contaminated sources. Thus individual risks would be lower than those usually calculated. (Collective risks depend simply on the amount of contamination consumed and, under the assumption of no-threshold, linear dose-response, would not depend on the distribution of exposure in the population. As discussed below, however, the major effect from radium ingestion probably has a threshold and/or is not linear.)

Thus the assumption of lifetime exposure for each individual is unrealistic for a generic risk assessment. A twenty to thirty-year exposure would be a more appropriate upper-bound. If the contamination source and target population are clearly identified, then the best assessment would use site-specific information: level of use, possible treatment, characteristics and lifestyles of the people exposed.

Since the age distribution and length of exposure are not known  $\underline{a}$  priori, lifetime exposure will be used for median risks derived in this report. Results will also be stated as average annual risk for use when exposure duration is known but age is not.

#### 7.2 RADIUM-226

#### Absorption factor

The gut uptake factor (f1) adopted by the ICRP for compounds of radium radionuclides is 0.2 (ICRP 1979, p. 98). The same value is used by EPA in its risk assessments. This choice is based on the range of 0.15 to 0.21 given earlier by the ICRP (ICRP 1973, p. 64). These endpoints are taken from only two human studies with a small number of participants.

In deriving alternative, median risk estimates, this report will also conservatively use a gut uptake factor of 0.2 (20%). Ingestion of 1 pCi/day then corresponds to a systemic intake of 73 pCi/year, or 7.3 E-5  $\mu$ Ci/year, or 2.7 Bq/year.

#### ICRP Assessment Methodology

The metric of risk is effective dose equivalent. "Effective" in this context means that individual target organ doses were weighted and summed to obtain a single dose figure. The weight is meant to reflect the relative radiosensitivity of each organ. Actual risk, which in the

ICRP method is cancer mortality plus genetic effects in the first two generations, is meant to be strictly proportional to effective dose equivalent, regardless of target organs. This is also linear, nothreshold by implication. Both the weighting factors and the risk coefficients are based predominantly on studies of A-bomb survivors.

Table 7-1. ICRP Assessment of Ra-226 Ingestion Risk

Target Organ Ed	Dose quivalent (Sv/Bq)	Weight	Effective Dose Equivalent (Sv/Bq)	
Gonads	9.2E-08	0.25	2.3E-08	
Red Bone Marrow	6.0E-07	0.12	7.2E-08	
Bone Surface	6.8E-06	0.03	2.0E-07	
Sum			3.0E-07	Sv/Bq
			1.1E-06	rem/pC
Risk at 1.65E-04	per rem		1.8E-10	/pCi
Risk for 1 pCi/d	lay for 70	years	4.6E-06	, . · ·

#### EPA Assessment Methodology

EPA estimates an excess lifetime risk of 8.8 per million persons exposed per pCi/l of radium-226 in drinking water (EPA 1985a, p. VIII-10; EPA 1986, p. 34859f). Since EPA assumes a daily drinking water consumption of two liters, the excess lifetime risk from ingesting one pCi every day for life is 4.4 per million people.

EPA considers its estimate of 4.4 per million persons per pCi/day to be a central value, not a conservative upper bound. It is assumed to be the geometric mean of a log-normal probability distribution with a geometric standard deviation of 2 (EPA 1986, p. 34815). The uncertainty is attributed to the dose part of the risk calculation, not the dose-response coefficient. Thus the 95% confidence interval (2.5% to 97.5%) is 1.1 to 18 per million per pCi/day.

The general method followed by EPA in calculating adverse health risk per unit activity intake of a radionuclide is that of the International Commission on Radiological Protection (ICRP 1977).

For calculating organ doses per unit activity of ingested radionuclides, EPA uses the results of a model similar to those tabulated by the ICRP (ICRP 1979). This model uses a quality factor of 20 for alpha particles. Quality factor is the only factor currently used by the ICRP in converting dose to dose equivalent.

The assessment of health effects made by EPA differs from the methods contained in ICRP 1977 and ICRP 1979 in the following respects:

- (a) The column headings in Table D (risk and effective dose equivalent rate per pCi/l) of the advanced notice (EPA 1986, pp. 34859-34860) imply that a 70-year dose commitment period is being used rather than the 50-year period used in ICRP 1979. The effect is to increase calculated organ doses slightly.
- (b) EPA uses a different set of organ weighting factors. The relative effect is to redistribute attributable risk among the organs. The absolute effect depends on the magnitude of the risk coefficient and precisely which organs receive significant dose. The EPA weighting factors are lower for bone surfaces and the urinary tract and higher for red bone marrow and other organs, in general. In terms of annual effective dose equivalent for radium nuclides, the EPA and ICRP results are about the same.
- (c) All organs for which a non-zero dose has been calculated are included by EPA whereas the ICRP does not include organs that have dose equivalents significantly lower than others.
- (d) The linear, no-threshold dose-response (risk) coefficient adopted by EPA is slightly higher than the one used by the ICRP (200 per million rem versus 165 per million rem). (The ICRP coefficient includes genetic risk as well as excess cancer mortality.) The risk calculated by EPA in Table D of the advanced notice on an organ by organ basis is not proportional to effective dose equivalent as it should be using the methods of the ICRP. It is therefore not clear how EPA has actually calculated risk from effective dose equivalent.

Table 7-2 shows the relative contribution by target organ to total effective dose and total risk in the EPA model. It is derived from Table D.1 of the advance notice (EPA 1986, pp. 34859f).

Table 7-2. Effective Dose Equivalent and Risk by Target Organ for Ra-226. (1 pCi/l radium-226; 2 liter/day; lifelong consumption)

	Red Bone Marrow	Endosteal Bone	Soft Tissue	Total
Effective Dose Equivalent				
(mrem/y)	0.27	0.18	0.21	0.65
(percent)	41%	28%	31%	
Lifetime Risk				
(per million persons)	3.74	0.88	4.18	8.8
(percent)	43ቄ	10%	47%	

#### Scientific Basis

Neither the EPA nor ICRP models is based on evidence of adverse health effects, in humans or animals, induced by ingestion of radium-226. Risk is indirectly inferred from excess cancers among A-bomb survivors who received substantial instantaneous, low-LET, external irradiation. The models are a complicated procedure to calculate essentially a dose which is "effectively equivalent" to that experienced by the survivors and would possess the same dose-response coefficient. In addition, extrapolation to much lower absolute dose levels are involved.

The results of the models contradict direct human evidence. The most reliable data come from studies of female radium dial painters.

The discrepancies involve primarily the distribution of excess cancers by organ and the shape of the dose-response function.

In its recent review, the BEIR IV Committee (NAS, 1987) summarized current understanding of the health risks of internally deposited alphaemitters. The most striking feature of the BEIR IV report is that leukemia induced by ingestion of Ra-226 (or Ra-228) is absent (NAS 1987, p. 228). Leukemia is a large effect predicted by the EPA and ICRP models. Excess soft-tissue tumors are absent from the radium dial painters. Soft-tissue tumors as a group would exceed leukemia in the EPA risk assessment. The BEIR IV Committee also reviewed three studies of radium in drinking water. One found a correlation with malignant neoplasm involving bone, the second found correlation with incidence of bladder and lung cancer in males and lung and breast cancer in females, and the last found correlation with elevated leukemia. However, none of these studies demonstrated dose response, they are in disagreement with each other, and all are in disagreement with long-term studies involving known, higher level intake of radium.

In summary, with respect to leukemia and soft-tissue tumors which constitute 90% of EPA's calculated risk, the EPA assessment incorrectly predicts adverse health effects.

For radium ingestion, quantitative results for bone sarcoma and "head" carcinoma are based on the work of Rowland et al. (e.g., Rowland 1983). The subjects were radium dial painters for whom radium body burden measurements have been made. Thus lifetime radium intake is relatively well established.

The dose-response function for bone cancer induced by ingestion of Ra-226 or Ra-228 either has a threshold at very large dose to the skeleton, well above even the worst environmental exposures, or is essentially purely quadratic resulting in effectively no excess cancers at lower doses.

By far the best fit for bone sarcoma dose-response was quadratic in form with an exponential "cell-killing" factor linear in radium uptake. A more general functional form including a linear term resulted in a negative value for the linear coefficient. Only when the linear coefficient is constrained can an "acceptable" fit be obtained. The value of the linear coefficient at p=0.05 is 1.3 E-5 excess sarcomas per person-year at risk following a 5-year latency per microcurie of systemic intake (Rowland et al. 1983, p. 21). Among the 824 cases with measured systemic intakes in the range 0.25 to 100  $\mu$ Ci of radium, this coefficient would predict 4 excess sarcomas whereas none have been observed.

The dose-squared exponential function was used to predict observed bone sarcomas in radium cases other than dial workers (Rowland et al. 1983, p. 24). Agreement was good for the total number observed for other female cases (17.3 predicted versus 15 observed) but not for males (10.9 predicted versus 3 observed).

In order to remove a possible bias in the full sample of cases with radium intake measured, Rowland et al. removed any years at risk before the date of first measurement of intake (Rowland et al. 1983, p. 20). The number of bone sarcomas went from 42 down to 13 and the total number of person-years at risk from 58701 to 11770. Linear and quadratic fits could not be distinguished in this subsample probably due to very poor statistical power.

Excess carcinomas of the paranasal sinuses and mastoid air cells ("head carcinomas") were also observed (Rowland et at. 1978). The total number observed thus far is seventeen. The best fit to the data was simple linear dose-response (no quadratic term nor cell-killing factor.) These carcinomas were generally attributed to radon-222, the immediate daughter of radium-226. No excess head carcinomas are associated with radium-228 or radium-224 intake. The radon daughters of these radionuclides have much shorter half-lives than radon-222 and have no opportunity to migrate within the body before they decay. The value

derived for the linear coefficient is 1.6 E-5 per person-year at risk using a 10-year latency per microcurie of systemic intake.

If one applied EPA's methods (lifetable analysis, lifetime exposure, 2 liters per day of drinking water, 0.2 gut absorption factor, etc.) with this linear coefficient, 4.3 head carcinomas are predicted per million people exposed per pCi/l of radium-226 (2 pCi per day). This is roughly one-half the total risk predicted by EPA but involves a cancer site not considered by EPA.

The annual rate in a population of 200 million per pCi/l is thus about 12.3 (4.3 x 200 / 70 year life). Scaling to the estimate of 2.3 pCi/day intake for ICRP Reference Man (ICRP 1975, p. 404) gives 14 deaths per year from carcinomas of the paranasal sinuses and mastoid air cells. The excess of deaths in the United States in 1977 from cancers of the auditory tube, middle ear, and mastoid air cells is estimated to be 15 with a comparable number for the ethnoid, frontal, and sphenoid sinuses (NAS 1987, p. 217). Thus, if the simple linear dose-response derived from radium dial painters were correct, one would need to attribute most if not all carcinomas of the paranasal sinuses and mastoid air cells to radium-226 ingestion.

# Central Risk Estimators

Realistic, central risk estimators can now be derived for radium-226 intake. The analysis uses a lifetable approach with a constant systemic intake of 1  $\mu$ Ci per year at each age for life. The results may then be used to calculate risks at other levels of intake taking due care if dose-response is not linear, no-threshold. The male and female lifetables used are for the US population, 1980-1984 (NAS 1987, p. 54). (Separate male and female calculations are averaged.)

For bone sarcomas, the best fit to the full sample of dial painters with measured intake is used, namely a pure quadratic function with a coefficient of 7.0 E-8 per  $\mu$ Ci squared per person-year at risk

following a latency of 5 years. Cell-killing is not used because the results on a per  $\mu$ Ci basis will be extrapolated to much lower environmental conditions for which the cell-killing term has no effect.

For "head" carcinomas, the linear fit coefficient of 1.6 E-5 is conservatively used here recognizing that its statistical basis is poor, and that it would predict a suspiciously large number of carcinomas due to background radium-226 intake.

The best estimate of risk for leukemias and soft-tissue cancers is zero. A non-zero, linear coefficient is conservatively derived here in recognition of the fact that a non-zero value is not excluded by the available data - although it is unlikely. Assuming a Poisson process, the probability of observing zero excess cancers is 50% if the actual expected number is 0.7. Since the number of  $\mu$ Ci person-years at risk lagged by 10 years reported for dial painters was 2.2 E+6 (Rowland et al. 1983, p. 20), the 50 percentile value of 0.7 would correspond to a risk coefficient of 3.1 E-7 per  $\mu$ Ci per person-year at risk following a 10-year latency. This value is used conservatively for leukemias and soft-tissue cancers separately.

Table 7-3. Central Estimate of Lifetime Cancer Mortality Risk from Systemic Intake of 1  $\mu$ Ci Radium-226 per Year for Life

Туре	Male	Female	Average
Bone Sarcoma	7.89 E-3	1.04 E-2	9.16 E-3
Head Carcinoma	3.19 E-2	3.94 E-2	3.57 E-2
Leukemia	6.18 E-4	7.64 E-4	6.91 E-4
Soft-tissue	6.18 E-4	7.64 E-4	6.91 E-4

Table 7-4. Central Estimate of Lifetime Cancer Mortality Risk from Ingestion of 1 pGi/day of Ra-226 (f1 = 0.2)

Туре	Lifetime Risk	Average Annual Risk
Bone Sarcoma	4.8 E-11	7.0 E-13
Head Carcinoma	2.6 E-6	3.7 E-8
Leukemia	5.0 E-8	7.2 E-10
Soft-tissue	5.0 E-8	7.2 E-10
	2.6 E-6	3.7 E-8

#### 7.3 RADIUM-228

## Absorption factor

The absorptions factor is assumed to be the same as that for radium-226. See the relevant section above.

#### ICRP Assessment Methodology

The ICRP methodology for Ra-228 risk assessment is identical to that for Ra-226 described above.

Table 7-5. ICRP Assessment of Ra-228 Ingestion Risk

Target Organ Eq	Dose uivalent (Sv/Bq)	Weight	Effective Dose Equivalent (Sv/Bq)	
Gonads	1.6E-07	0.25	4.0E-08	
Breast	1.6E-07	0.15	2.4E-08	
Red Bone Marrow	6.5E-07	0.12	7.8E-08	
Lung	1.6E-07	0.12	1.9E-08	
Bone Surface	5.8E-06	0.03	1.7E-07	
Sum			3.4E-07	Sv/Bq
			1.2E-06	rem/p
Risk at 1.65E-04	per rem		2.0E-10	/pCi
Risk for 1 pCi/d	-	years	5.1E-06	

# EPA Assessment Methodology

EPA estimates an excess lifetime risk of 6.6 per million persons exposed per pCi/l of radium-228 in drinking water (EPA 1985a, p. VIII-11; EPA 1986, p. 34860). Since EPA assumes a daily drinking water consumption of 2 liters, the excess lifetime risk from ingesting one pCi every day for life is 3.3 per million people.

EPA considers its estimate of 3.3 per million persons per pCi/day to be a central value, not a conservative upper bound. It is assumed to be the geometric mean of a log-normal probability distribution with a geometric standard deviation of 2 (EPA 1986, p. 34815). The uncertainty is attributed to the dose part of the risk calculation, not the risk coefficient. Thus the 95% confidence interval (2.5% to 97.5%) is 0.9 to 13 per million per pCi/day.

The ICRP and EPA methodologies parallel those used for the assessment of radium-226 ingestion risk.

Table 7-6 shows the relative contribution by target organ to total effective dose and total risk in the EPA model. It is derived from Table D.2 of the advance notice (EPA 1986, pp. 34860). The difference in distribution of dose and effects by site relative to radium-226 is due to the shorter half-life of radium-228 (5.7 years versus 1600 years).

Table 7-6. Effective Dose Equivalent and Risk by Target Organ for Ra-228. (1 pCi/l radium-228; 2 liter/day; lifelong consumption)

	Red Bone Marrow	Endosteal Bone	Soft Tissue	Total
Effective Dose Equivalent		·	<del></del>	
(mrem/y)	0.114	0.063	0.20	0.38
(percent)	30%	17%	53%	
Lifetime Risk				
(per million persons)	1.96	0.39	4.25	6.6
(percent)	30%	6%	64%	

# Scientific Basis

The results obtained from radium dial painter studies for radium-228 differ from those for radium-226 in only two respects:

- (1) For induction of bone sarcomas, a systemic intake of one  $\mu \text{Ci}$  of radium-228 had the same effect as an intake of 2.5  $\mu \text{Ci}$  of radium-226.
- (2) No head carcinomas are observed, as is the case with radium-224 as well. This is attributed to the fact that radon gas produced by these radionuclides are short lived and can not migrate to the sinuses and mastoid air cells.

## Central Risk Estimators

The same risk estimators are used for radium-228 as for radium-226 except that bone sarcoma effects are multiplied by 6.25 (the square of 2.5) and "head" carcinoma risk is set to zero.

Table 7-7. Central Estimate of Lifetime Cancer Mortality Risk from Systemic Intake of 1  $\mu\rm{Ci}$  Radíum-228 per Year for Life

Туре	Male	Female	Average
Bone Sarcoma	4.93 E-2	6.50 E-2	5.72 E-2
Leukemia	6.18 E-4	7.64 E-4	6.91 E-4
Soft-tissue	6.18 E-4	7.64 E-4	6.91 E-4

Table 7-8. Central Estimate of Cancer Mortality Risk from Ingestion of 1 pCi/day of Ra-228 (f1 = 0.2)

Туре	Lifetime Risk	Average Annual Risk
Bone Sarcoma Leukemia Soft-tissue	3.0 E-10 5.0 E-8 5.0 E-8	4.4 E-12 7.2 E-10 7.2 E-10
	1.0 E-7	1.4 E-9

# Section 8

# SCREENING-LEVEL ASSESSMENT - DOSE TO AQUATIC ORGANISMS

#### 8.1 APPROACH

In this screening-level assessment, conservative assumptions were made to develop estimates of dose to aquatic animals resulting from internal and external exposure to radium discharged in produced waters. These estimates are based on assumed discharges to coastal Louisiana. The effects of radium discharged to offshore waters will be significantly smaller because of the increased dilution and reduced potential for uptake by fish and shellfish.

Estimates were made for the field data reported in CSA (1991), and for five potential discharge and water concentration scenarios (background concentrations of 0.1 and 1.0 pCi/l; discharge of 30, 500 and 2,000 pCi/l). The potential scenarios were included in the analysis to provide an assessment of the risk associated with the range of radium water concentrations that may result from produced water discharges. Dose estimates were calculated for fish, molluscs and crustaceans using the simple models described in IAEA (1976).

The calculation of dose to aquatic biota involves the following steps:

- 1. Estimate the concentration of Ra-226 and Ra-228 in water.
- 2. Estimate the concentration of Ra-226 and Ra-228 in fish and shellfish.
- 3. Calculate the internal and external dose to aquatic biota associated with these concentrations.

These steps are described in the following sections.

#### 8.2 ESTIMATION OF RADIUM CONCENTRATIONS IN WATER AND AQUATIC BIOTA

#### Radium Concentration in Water

For the data collected by CSA (1991), the concentration of Ra-226 and Ra-228 in water was measured. These concentrations are given in Table 8-1.

The five potential scenarios used in the analysis require the estimation of water concentrations. For all of these analyses, it was assumed that the concentration of Ra-226 was equal to the concentration of Ra-228 in each discharge. To represent background conditions, two zero discharge scenarios were included, with an assumed Ra-226 (and Ra-228) concentration of 0.1 and 1.0 pCi/l. The three discharge scenarios included in the assessment were 30 pCi/l, 500 pCi/l and 2,000 pCi/l. The 2,000 pCi/l discharge represents the very few high discharge concentration outfalls in coastal Louisiana, while the 500 and 30 pCi/l discharges are more typical.

The concentration in water resulting from these three discharge concentrations was calculated assuming a dilution factor of 100. The radium in produced water discharges is diluted very rapidly and the concentration is diluted by a factor greater than 100 within 100 feet of the outfall for all of the modeled offshore situations and nearshore field data reviewed in Section 3.3.2. Because of the toxic effects of hydrocarbons discharged in produced water, fish and shellfish are not likely to be living close to an outfall. The use of a dilution factor of 100 is a reasonable, but conservative assumption in this analysis, because most fish and shellfish will be exposed to radium in water further than 100 feet from a discharge point.

A dilution factor of 100 applied to the three discharge scenarios described above (30, 500 and 2,000 pCi/l) results in water concentration

(of both Ra-226 and Ra-228) of 0.3, 5 and 20 pCi/l. The water concentrations used in the analysis are summarized in Table 8-1.

## Concentration of Radium in Aquatic Organisms

The CSA (1991) data set includes measurements of Ra-226 and Ra-228 concentrations in fish, oysters and crustaceans. These data are given in Table 8-1.

The concentration of Ra-226 and Ra-228 in fish, molluscs and crustaceans were calculated for the five potential scenarios described above (water concentrations of 0.1, 1, 0.3, 5 and 20 pCi/l) using the concentration factor method:

Conc. in seafood = Conc. in water x Concentration Factor (8-1) (pCi/g) 
$$\frac{}{1000 \text{ (g/l)}}$$

For the purposes of this screening-level assessment, the conservative IAEA Concentration Factors (100 for fish, molluscs and crustaceans) were used. The estimated concentrations of Ra-226 and Ra-228 in fish, molluscs and crustaceans for the five potential discharge/water concentration scenarios are given in Table 8-1.

#### 8.3 CALCULATION OF DOSE - IAEA METHOD

The method described in IAEA (1976) is used here to estimate dose rates to aquatic organisms. A more detailed risk assessment study would use more complex models. The IAEA method uses simple, idealized geometries to model organisms. For all of these calculations, Ra-226 and Ra-228 were assumed to be in equilibrium with their short-lived daughters.

Table 8-1. Values Used in Calculation of Dose to Aquatic Biota - Ra-226 and Ra-228, IAEA Concentration Factors and CSA (1991) Site Data.

	WATER pCi/1	SEDIMENT pCi/g	FISH CF	pCi/g	MOLLI	MOLLUSCS GF pCi/g	CRUS	CRUSTACEANS CF nc1/n	
Discharge <sup>1</sup>							,	For/ B	
02 . 02	0.1		100	0.01 0.1	100	0.01	100	0.01	
500 pci/1 2000 pci/1	5.0 5.0 20.0	:::	100 100 100	0.03 0.5 2.0	100	0.03 0.5	100	0.1 0.03 0.5	
GSA Site 1					) }	, ,	700	2.0	
Ra-226 Ra-228	228.9 383.0	19.0 2.5		0.014		0.007		0.07	
CSA Site 2								60.0	
Ra-226 Ra-228	110.5 244.4	0.35		0.022		0.008		0.07	
CSA Site 3						110.0		0.025	
Ra-226 Ra-228	251.9 254.8	1.5		0.041 0.012		NS NS		0.125	
						2		0.243	

 $^1$ : for these scenarios, the concentration of Ra-226 and Ra-228 were assumed equal  $^2$ : these two water concentrations (0.1, 1 pCi/l represent the range of background concentrations)

# Dose Rates From Incorporated Radionuclides

Molluscs, crustaceans and fish are large in relation to the range of  $\alpha$  and  $\beta$  particles. It is assumed that no significant portion of the total energy emitted by incorporated radionuclides in the form of  $\alpha$  and  $\beta$  particles is dissipated in the surrounding water. The dose rate from  $\alpha$  and  $\beta$  particles closely approach the dose rate in an infinite volume, uniformly contaminated with the radionuclide.

$$D_{(\infty)} = 2.13 \text{ E C } \mu \text{rad/hour}$$
 (8-2)

where:

 $D_{(\infty)}$  is the dose rate  $(\alpha \text{ or } \beta)$  in an infinite volume, E is the average energy  $(\alpha \text{ or } \beta)$  MeV per disintegration, and C is the activity of the radionuclide in the organism (pCi/gram).

To estimate the dose from  $\gamma$  rays the dimensions and geometry of the organism are needed. The average dose rate from internal gamma radiation is given by:

$$D_{\gamma} = \Gamma C \rho g \times 10^{-3} \mu rad/hour$$
 (8-3)

where:

 $\Gamma$  is the specific  $\gamma$  ray constant in cm<sup>2</sup> . rad/hr . mCi<sup>-1</sup> C is the specific activity in the organism (pCi/gram)  $\rho$  is the density of the organism g is the mean geometrical factor in cm (Loevinger et al. 1956)

The idealized dimensions and geometrical factors used in IAEA (1976) are as follows:

Molluscs: flat cylinder, 1 cm high, 4 cm in diameter. g=10 cm Crustaceans: cylinder, 15 cm long, 6 cm in diameter. g=25 cm Fish: cylinder, 50 cm long and 10 cm diameter. g=41 cm

The tissue density is assumed to be 1 and the activity is assumed to be uniformly distributed throughout the volume. This assumption may result in an underestimate of the dose to some tissue, since radium tends to accumulate in bone and shell.

#### Dose Rates From Radionuclides in Water

For molluscs, crustaceans and fish IAEA (1976) assumes that external  $\alpha$  and  $\beta$  radiation from the sea water contributes negligible amounts to the average dose within the animals. The  $\gamma$  ray dose is taken to be D  $(\infty)$ .

$$D_{(\infty)} = 2.13 \text{ E C } \mu \text{rad/hour}$$
 (8-4)

#### Dose Rates From Radionuclides in Sediment

The  $\gamma$  and  $\beta$  radiation dose rate above sediments has been taken to be approximately equal to 0.5  $D_{(\infty)}$ . The dose from  $\alpha$  radiation is assumed to be negligible.

#### 8.4 DOSE ESTIMATES

Table 8-2 gives the estimated dose rates to fish, molluscs and crustaceans for the two scenarios.

Dose rates calculated for the potential scenarios (0.1, 1, 0.3, 5, and 20 pCi/l) using IAEA concentration factors ranged from 1.3  $\mu$ rad/hour (3.1 x 10<sup>-5</sup> rad/day; .1 pCi/l, fish, molluscs and crustaceans) to 264.6  $\mu$ rad/hour (0.006 rad/day; 2,000 pCi/l, fish). Dose rates for the three CSA sites ranged from 7.4  $\mu$ rad/hour (1.8 x 10<sup>-4</sup> rad/day; Site 2, fish, molluscs) to 272.5  $\mu$ rad/hour (0.007 rad/day; Site 1, crustaceans).

Estimated Doses to Fish, Molluscs and Crustaceans From Ra-226 and Ra-228 -- IAEA Concentration Factor and CSA (1991) Site Data Table 8-2.

******	THE CONTRACTOR OF THE PARTY OF		
	FISH µ rad/hour	MOLLUSCS µ rad/hour	CRUSTACEANS µ rad/hour
Discharge			
0 1 nGi /1	1.3	1.3	1.3
1 nGi/1	13.2	13.2	13.2
30 nGi /1	4.0	4.0	4.0
500 pCi/1	66.2	0.99	0.99
2000 pci/1	264.6	263.7	264.1
Site 1	260.8	260.3	272.5
Site 2	7.4	7.4	23.0
Site 3	20.7	;	31.7

# 8.5 COMPARE TO NO EFFECT LEVELS

Even using conservative assumptions (IAEA concentration factors), the estimated doses are below those expected to result in deleterious effects (1-10 mGy/day [.1-1 rad/day] for individuals, >10 mGy/day [l rad/day] for natural populations; Section 6).

# Section 9 SCREENING-LEVEL ASSESSMENT - RISKS TO HUMAN HEALTH

#### 9.1 APPROACH

In this screening-level assessment of the risks to human health, conservative assumptions were made to develop estimates of the individual lifetime risk that could result from ingestion of radium discharged in produced waters. These estimates are based on assumed discharges to coastal Louisiana. The effects of radium discharged to offshore waters will be significantly smaller because of the increased dilution and reduced potential for uptake by fish and shellfish.

Estimates were made for the field data reported in CSA (1991), and for five potential discharge and water concentration scenarios. The potential scenarios were included in the analysis to provide an assessment of the risk associated with the range of radium water concentrations that may result from produced water discharges.

Conservative risk factors were used in this assessment (EPA risk factors, Section 7), and it was assumed that one-half of an individual's seafood consumption comes from animals harvested near a produced water outfall (i.e. at the point where a 100-fold dilution occurs). Intake levels used in the analysis include those for the individual eating the most seafood.

This screening-level assessment is a conservative analysis, involving a number of simplifying assumptions that necessarily result in an over-estimate of the risk associated with the discharge of radium in produced water.

#### 9.2 INDIVIDUAL LIFETIME RISK

The calculation of the individual lifetime cancer risk associated with the discharge of radium in produced water involves the following steps:

- 1. Estimate the concentration of Ra-226 and Ra-228 in water.
- 2. Estimate the concentration of Ra-226 and Ra-228 in fish and shellfish.
- 3. Estimate the amount of fish and shellfish consumed by individuals.
- 4. Calculate the amount of Ra-226 and Ra-228 consumed by individuals.
- 5. Calculate the individual lifetime risk associated with the estimated radium intake in fish and shellfish.

These steps in the risk assessment are described in the following sections.

#### Radium Water Concentrations

For the data collected by CSA (1991), this step is unnecessary because the concentration of Ra-226 and Ra-228 in fish and shellfish was measured directly. The concentrations in water were also measured, and are given in Table 9-1 for the three CSA (1991) sites.

The five potential scenarios used in the analysis do require the estimation of water concentrations. For all of these analyses, it was assumed that the concentration of Ra-226 was equal to the concentration of Ra-228 in each discharge. To represent background conditions, two zero discharge scenarios were included, with an assumed Ra-226 (and Ra-228) concentration of 0.1 and 1.0 pCi/l. The three discharge scenarios included in the assessment were 30 pCi/l, 500 pCi/l and 2,000 pCi/l. The 2,000 pCi/l discharge represents the very few high discharge concentration outfalls in coastal Louisiana, while the 500 and 30 pCi/l discharges are more typical.

The concentration in water resulting from these three discharge concentrations was calculated assuming a dilution factor of 100. The radium in produced water discharges is diluted very rapidly, and the concentration is diluted by a factor greater than 100 within 100 feet of the outfall for all of the modeled situations and field data reviewed in Section 3.3.2. Because of the nearfield toxic effects of hydrocarbons discharged in produced water, fish and shellfish are not likely to be harvested close to an outfall. The use of a dilution factor of 100 is a reasonable, but conservative assumption in this analysis, because most fish and shellfish will be harvested further than 100 feet from a discharge point.

A dilution factor of 100 applied to the three discharge scenarios described above (30, 500 and 2,000 pCi/l) results in water concentration (of both Ra-226 and Ra-228) of 0.3, 5 and 20 pCi/l.

# Concentrations in Aquatic Organisms Used For Food

The data set extracted from CSA (1991) includes direct measurements of Ra-226 and Ra-228 concentrations in fish, oysters and crustaceans. The concentrations of radium reported for oysters are the levels measured in edible tissue. Fish and crustacean samples included skin, bone and exoskeleton, and overestimate the amount of radium that would be ingested by people in fish and crustaceans. These data are given in Table 9-1.

The concentrations of Ra-226 and Ra-228 in fish, molluscs and crustaceans were calculated for the five potential scenarios described above (water concentrations of 0.1, 1, 0.3, 5 and 20 pCi/l) using the concentration factor method:

Conc. in seafood = Conc. in water x Concentration Factor (9-1) (pCi/g) (pCi/l) 
$$\frac{}{1000 \text{ (g/l)}}$$

Table 9-1. Values Used in Calculation of Individual Risk for Five Potential Discharge/Water Concentration Scenarios and Three CSA (1991) Sites.

	pCi/l	CF	pCi/g	CF	MULLUSCS CF pCi/g	CRUS CF	CRUSTACEANS CF pCi/e
Discharges							
0 0 pCi/l	0.1	100	.0.01	100	0.01	100	0.01
30 pci/l 500 pci/l 2000 pci/l	0.3 5.0 20.0	100	0.03 0.5 2.0	100 100 100	0.1 0.03 0.5 2.0	100 100 100	0.1 0.03 0.5 2.0
<b>Site 1</b> Ra-226 Ra-228	228.9 383.0		0.014 0.005		0.007		0.07
<b>Site 2</b> Ra-226 Ra-228	110.5 244.4		0 0.022		0.008 0.011		0.07 0.025
<b>Site</b> 3 Ra-226 Ra-228	251.9 254.8		0.041 0.012		NS NS	·	.0.125 0.243

For the purposes of this screening-level assessment, the conservative IAEA Concentration Factors (100 for fish, molluscs and crustaceans) were used. The estimated concentrations of Ra-226 and Ra-228 in fish, molluscs and crustaceans for the five potential discharge/water concentration scenarios are given in Table 9-1.

# Intake Rates

To calculate risk from ingestion of radium in seafood, the amount of seafood consumed per year is needed. Food consumption patterns vary among individuals and with age and geographic location. This study used intake rates from a survey commissioned by the National Marine Fisheries Service in 1973-1974 and described in Rupp et al. (1980). Intake rates for people living in the West South Central Region of the United States (Arkansas, Louisiana, Oklahoma and Texas) were used in this assessment.

Rupp et al. (1980) report consumption rates for three age groups (1-11 years, 12-18 years, and 19-98 years) and for three types of seafood (fresh water fish, salt water fish and shellfish). A regional distribution of shellfish consumed by species (i.e. percent of the total of each species that is consumed by each U.S. region) is presented in Rupp et al. (1980). These data were used to estimate the percent of shellfish consumption represented by molluscs (23.76%) and by crustaceans (76.24%) for the West South Central Region (Table 9-2).

Table 9-3 presents the intake rates (kg/year) for salt water fish and shellfish for the three age groups. Intake rates for molluscs and crustaceans were estimated by dividing the rates for shellfish consumption by the percentages derived from Table 9-2. Values at the 50, 90 and 99 percentile are reported, along with the maximum intake value for each category. Two "average" values are also presented -- the average intake for people who consume fish or shellfish, and the average for the entire population, including people who eat no fish or shellfish.

Table 9-2. Distribution of Shellfish Consumption By Type in Arkansas, Louisiana, Oklahoma and Texas. I (West South Central Region).

Species	<pre>% Contribution To U.S. Total</pre>	Total # of Observations	# of Obs in W.S. Gentral Region	
abalone clam oyster scallop squid/octopi	0 3.7 12.5 3.6 2.2	51 2243 1241 527 45	0 83 155 19 1	
crab lobster shrimp crustaceans	12.1 2.3 11.1	1299 1028 5832	157 24 647 	
Other	3.7	54	2	

Data from Rupp et al., 1980.

Table 9-3. Fish and Shellfish Consumption in Arkansas, Louisiana, Oklahoma and Texas  $({
m Kg/year})^{*1}$ 

Age Group	Type	I	ercent	Percentiles	l			
(years)		50%	806	866	Maximum Consumer	Consumer Average	Population Average	
1-11 (n=510)	sw fish	0.95	3.55	8.60	15.12	1.77	1.52	85.9
(010.11)	molluscs	00	0.47	5.66 1.34	11.51	1.93	0.58	30.0
	crust.	0	1.51	4.32	8.78	1.47	0.44	
12-18	sw fish	1.43	4.92	15.62	28.62	2.76	2,40	1 78
(n=1.94)	shellfish	0	1.88.	9.83	14.12	2.38	0.70	70 00
	molluscs	0	0.45	2.34	3,35	0.56	0.17	•
	crust.	0	1.43	7.49	10.77	1.82	0.53	
19-98	sw fish	2.34	7.97	21.92	31.20	4.03	3.57	7 88
(n-1424)	shellfish	0	97.4	14.90	24.95	3.55	1,40	39.4
	molluscs	0	1.06	3.54	5.93	0.84	0.33	•
	crust.	0	3.40	11.36	19.02	2.71	1.07	

\* Shellfish divided into molluscs and crustaceans by percentages derived from data in Table 10-3. [23.76% molluscs, 76.24% crustaceans).

Data derived from Rupp et al., 1980.

### Individual Lifetime Risk Estimates

Conservative, upper bound estimates of individual lifetime risk were calculated using the EPA risk factors described in Section 7.

Ra-226:  $4.4 \times 10^{-6}$  per pCi/day lifetime intake Ra-228:  $3.3 \times 10^{-6}$  per pCi/day lifetime intake

Intake levels for adults (19-98 years) were used in the analysis. Three intake rates were used in this preliminary assessment: the maximum intake rate, the average intake rate for fish/shellfish consumers, and the average intake rate for the whole population (Table 9-3). In all cases it was conservatively assumed that 50% of a person's entire seafood intake (for a lifetime) was harvested near a produced water outfall (i.e. where the dilution factor reaches 100). Risk estimates were calculated separately for Ra-226 and Ra-228 and then summed for each exposure scenario.

Individual lifetime risk estimates were calculated as follows:

consumption rate = food conc. x 1000 x intake rate x 
$$1/365$$
 (9-2) (pCi/day) (pCi/g) (g/kg) (kg/year) (years/day)

Lifetime Risk = consumption rate (pCi/day) x Risk Factor (9-3)

Table 9-4 presents the estimates of individual lifetime risk for ingestion of Ra-226 and Ra-228 in seafood for the three CSA (1991) sites and the five potential concentration scenarios.

These risk estimates are based on the following assumptions:

- 1. Radium discharged in produced water is reduced by a factor of 100 before fish and shellfish consumed by people comes in contact with it.
- 2. An individual gets one-half of their yearly fish and shellfish from near (i.e. where the dilution factor reaches 100; probably within 100 feet) a produced water outfall for their entire lifetime.

Harvested From Waters With Enhanced Levels of Radium - Five Potential Discharge/Water Concentration Table 9-4. Individual Lifetime Risk Estimates for Consumption of Fish, Molluscs and Crustaceans Scenarios and Three CSA (1991) Sites.

	Water Gonc. Ra-226 pCi/l	Ra-228 pCi/l	Maximum Consumer	Lifetime Consumer Average	Lifetime Risk
Discharge				)	
0	0.1	0.1	5.92E-6	8.01E-7	5.29E-7
0	Н	1,	5.92E-5	8.01E-6	5.29E-6
30 pCi/1	0.3	0.3	1.78E-5	2.40E-6	1.59E-6
500 pci/1	5.0	5.0	2.96E-4	4.01E-5	2.65E.5
2000 pci/l	20.0	20.0	1.18E-3	1.60E-4	1.06E-4
Sité l	228.9	383.0	1.94E-5	2.73E-6	1,30E-6
Site 2	110.6	244.4	1.38E-5	1.94E-6	9,68E-7
Site $3^1$	251.9	254.9	4.46E-5	6.25E-6	3.08E-6

 $^1$  No mollusc samples at station 3, estimates based on concentrations of radium in fish and crustaceans

- 3. The use of conservative IAEA concentration factors in estimating the concentration of radium in fish and shellfish.
- 4. The use of conservative EPA risk factors.

Using the conservative assumptions outlined above, a level of risk which could be considered significant (greater than  $1 \times 10^{-5}$ ) is estimated for: 1) the person consuming the most seafood under all modeled discharge scenarios for a lifetime, and 2) a person consuming an average amount of seafood (for a lifetime) harvested within 100 feet of a high concentration discharge (500 pCi/l).

The highest individual lifetime risk estimated using the IAEA concentration factors is for the person consuming the maximum amount of seafood, harvested near an outfall discharging 2000 pCi/l (1.18 x  $10^{-3}$  lifetime risk).

The risks estimated for the three CSA sites were based on measured concentrations of radium-226 and radium-228 in fish and shellfish. The estimates based on the maximum consumption rate exceeded 1 x  $10^{-5}$  at all three sites (1.94 x  $10^{-5}$ , 1.38 x  $10^{-5}$  and 4.46 x  $10^{-5}$  individual lifetime risk). The estimates based on the average consumer consumption rate were all less than 1 x  $10^{-6}$ . These values are overestimates for the risk associated with the consumption of radium in fish and crustaceans because they include radium in skin, bones and exoskeleton.

# 9.3 PREDICTION OF EXCESS CANCERS

A conservative, upper-bound estimate of the population risk associated with the ingestion of radium from produced waters was calculated using simple models and assumptions. Only discharges into the coastal zone of Louisiana were included in this analysis. To estimate total population risk several calculations were needed -- the "average" increase in radium concentrations in coastal Louisiana; the resulting "average" concentration of radium in fish and shellfish; and the amount of fish and shellfish harvested (and ingested) from the area.

### Box Model

A simple steady state box model was used to estimate the increase in Ra-226 and Ra-228 concentrations in the coastal zone of Louisiana resulting from the discharge of radium in produced water (IAEA, 1982; Section 4). All radium discharged into the coastal zone was assumed to remain in the water column, resulting in an overestimate of resulting radium concentratuions.

$$C = Q / K_e V$$
 (9-4)

where: C =the change in radium water concentration (pCi/1)

Q = the radium discharge rate (pCi/day)

V = the volume of water in the Louisiana coastal zone

 $K_e$  = the fractional loss rate of water from the mixed volume  $(day^{-1})$ 

A conservative estimate (i.e. small) of the volume of water in the Louisiana coastal zone was developed based on the area of surface water in the coastal zone of Louisiana (2631 km $^2$ , Department of Commerce, 1984), and an assumed depth of 3 meters. These assumptions result in a calculated volume of 7.9 x  $10^{12}$  liters. The fractional loss rate used was 0.015. This value was derived from studies of the Barataria and Terrebonne-Timbalier Basins reported in Boesch and Rabalais (1989).

### Rate of Radium Discharge

The rate of Ra-226 and Ra-228 discharged into the coastal zone of Louisiana (Q) was estimated by summing the discharges (pCi/day) for all of the stations in the State of Louisiana database (Section 4.4). These totals were:

Ra-226:  $4.39 \times 10^{10} \text{ pCi/day}$ Ra-228:  $4.83 \times 10^{10} \text{ pCi/day}$ 

### Increase in Radium Concentrations

The increase in Ra-226 and Ra-228 concentrations due to produced water discharges (C) was calculated using the box model and parameters described above. The resulting increases in radium concentrations were:

Ra-226: 0.37 pCi/l Ra-228: 0.41 pCi/l

### Concentration of Radium in Fish and Shellfish

The "average" concentration of Ra-226 and Ra-228 in fish and shellfish harvested in coastal Louisiana was estimated using the concentration factor (CF) method (Section 5). The conservative IAEA values (100 for both fish and shellfish) were used in this analysis. The "average" radium concentrations in fish and shellfish were calculated for 1) the estimated increase in water concentration due to produced water discharges (Ra-226: 0.37 pCi/l, Ra-228: 0.41 pCi/l); 2) a low estimate of background radium levels (0.1 pCi/l for both Ra-226 and Ra-228) and 3) a high estimate of background radium concentrations (1.0 pCi/l for both Ra-226 and Ra-228).

The estimated radium concentrations in fish and shellfish were calculated as follows:

The resulting calculated concentrations in seafood were:

	Ra-22	.6	Ra-22	.8
	water (pCi/1)	food (pCi/g)	water (pCi/l)	food (pCi/g)
Prod Water	0.37	0.037	0.41	0.041
Background	0.1	0.01	0.1	0.01
Background	1.0	0.1	1.0	0.1

### Amount of Fish and Shellfish Harvested

To estimate the number of excess cancers resulting from ingestion of radium, the amount of fish and shellfish harvested (and ingested) per year from coastal Louisiana is needed. The commercial live weight catch for Louisiana in 1989 was  $1.23 \times 10^9$  pounds (Department of Commerce, 1990). Recreational fishermen also harvest fish in Louisiana, but the total amount is small compared to the commercial catch (approximately  $1.6 \times 10^5$  pounds for the entire Gulf Coast region (calculated from Tables in Department of Commerce, 1990).

Some of the commercial catch was from offshore -- for the entire U.S. catch in 1989, 37% was harvested inshore (0-3 miles) (Department of Commerce, 1990). This analysis assumed that 50% of the reported catch for Louisiana was harvested nearshore because of the importance of shellfish harvested in coastal waters. In 1989, 73.3% of the fish and shellfish harvested in the United States was used for human food (Department of Commerce, 1990). Not all of the fish and shellfish catch is consumed -- the value for Louisiana reported above includes fish bones and skin but not clam or oyster shells. In this analysis, it was assumed that 75% of the fish and shellfish used for human food was actually consumed.

The amount of fish and shellfish consumed per day over a year was calculated as follows:

$$(1.23 \times 10^9 \text{ lbs/year}) \times (0.5 \text{ caught inshore}) \times (9-6)$$
  
 $(0.73 \text{ used for food}) \times (0.75 \text{ actually eaten}) \times (453.6 \text{ g/lb}) \times (1/365 \text{ year/days}) = 4.2 \times 10^8 \text{ g/day}$ 

## Calculation of Predicted Excess Cancers

The total amount of radium ingested in fish and shellfish per day (pCi/day) was calculated by multiplying the amount ingested in the region over a year  $(4.2 \times 10^8 \text{ grams/day})$  times the "average"

concentration of Ra-226 and Ra-228 in fish and shellfish (pCi/g). The resulting estimates are given below.

		226	Ra-	228
	water (pCi/l)	total (pCi/d)	water (pCi/1)	total (pCi/d)
Prod Water	0.37	$1.6 \times 10^{7}$	0.41	1.7 x 10 <sup>7</sup>
Background	0.1	$4.2 \times 10^6$	0.1	4.2 x 10 <sup>6</sup>
Background	1.0	$4.2 \times 10^{7}$	1.0	4.2 x 10 <sup>7</sup>

Conservative, upper bound estimates of population risk were calculated using the EPA risk factors described in Section 7:

Ra-226:  $4.4E \times 10^{-6}$  per pCi/day lifetime (70 years) intake (6.3 x  $10^{-8}$  per pCi/day, 1 year intake)

Ra-228: 3.3E x  $10^{-6}$  per pCi/day lifetime (70 years) intake (4.7 x  $10^{-8}$  per pCi/day, 1 year intake)

The number of excess cancers expected per year is calculated as:

Total Risk = [consumption rate (pCi/day)] x [Risk Factor] (9-7) (per pCi/day)

The resulting estimates of excess cancers per year of operation are given in Table 9-5.

Based on a simple, conservative analysis, the total number of excess cancers expected per year from Ra-226 and Ra-228 discharged to coastal Louisiana waters is 1.8. A similar analysis, using a range of possible background concentrations (0.1 - 1.0 pCi/l) predicted from 0.5 - 5.0 excess cancers per year from background radium in the region. The risk to the individual associated with the increase in radium predicted by this analysis (0.37 pCi/l Ra-226; 0.41 pCi/l Ra-228) is  $4.6 \times 10^{-5}$  assuming the maximum consumption rate (Table 9-4) and  $6.2 \times 10^{-6}$  assuming the average population consumption rate.

Table 9-5. Estimated Excess Cancers per Year From Radium in Coastal Louisiana Waters  $^{\star}$ .

	Ra-	-226	Ra	-228	Total
	pCi/l	cancers	pCi/l	cancers	
Produced Water	0.37	1.0	0.41	0.8	1.8
Low Background Estimate	0.1	0.3	0.1	0.2	0.5
Realistic Backgr Estimate	ound	3.0	1.0	2.0	5.0

<sup>\*</sup> at current levels of produced water discharge

### 9.4 DISCUSSION

These conservative analyses suggest that a potential for a risk that could be considered significant (greater than  $1 \times 10^{-5}$ ) exists for the maximally exposed individual and for a person consuming an average amount of seafood (for a lifetime) harvested within 100 feet of a high concentration nearshore discharge (500 pCi/l). The number of excess cancers predicted per year is comparable to the number expected to result from background concentrations of radium. These risk estimates are based on the following assumptions:

- 1. Radium discharged in produced water is reduced by a factor of 100 before it comes in contact with fish and shellfish consumed by people.
- 2. An individual gets one-half of their yearly fish and shellfish from near (i.e. where the dilution factor reaches 100; probably within 100 feet) a produced water outfall for their entire lifetime.
- 3. The use of conservative IAEA concentration factors in estimating the concentration of radium in fish and shellfish.
- The use of conservative EPA risk factors.

Because of the many conservative assumptions incorporated into this screening-level analysis, it can be concluded that the risks associated with the discharge of produced water to coastal Louisiana are small.

Radium discharged offshore will be diluted more rapidly than radium discharged to nearshore waters. Organisms living offshore will have a smaller chance of coming into contact with discharged radium because of the large water volumes involved and the rapid dilution that occurs. An individual person is also not likely to harvest a significant amount of his yearly seafood close to an offshore outfall. Because of the additional reductions in the radium concentration in water and aquatic biota expected near offshore outfalls as compared to nearshore discharges, it can be concluded that the risks associated with offshore discharges will be extremely small.

# Section 10 SUMMARY AND CONCLUSIONS

### 10.1 QUANTITIES AND UNITS OF RADIONUCLIDE ACTIVITY AND DOSE

Radioactivity is quantified in terms of the number of spontaneous energy emitting transformations per unit time -- a quantity known as activity. An example of a transformation is the decay of a radium-226 nucleus into a radon-222 nucleus, an alpha particle, and gamma rays. The unit of activity has historically been the curie (Ci). One curie is equal to  $3.7 \times 10^{10}$  disintegrations per second. In the International System of units (SI), the basic unit of activity has been redefined as one disintegration per second, known as the becquerel (Bq). One curie is equal to  $3.7 \times 10^{10}$  Bq. Concentrations of radium in water are usually expressed in units of pCi/1 or Bq/1.

The biological effect of a radionuclide is related to the absorbed dose (or the dose rate). The absorbed dose is the amount of energy imparted to matter. An absorbed dose of 100 erg/gm is called 1 rad. In the SI system the absorbed dose unit is 1 Joule per kilogram (J/kg), and 1 J/kg is called the Gray (Gy). An absorbed dose of 1 rad is equal to 0.01 Gy (1 Gy = 100 rads). Harmful levels of radiation doses are generally expressed in terms of rads. For example, over a hundred rads must be imparted in a short period of time to a substantial portion of the body before most individuals will show significant clinical symptoms (Saenger, 1963). Occupational doses are not allowed to exceed a few mrad per hour  $(1 \text{ mrad} = 1 \times 10^{-3} \text{ rad})$ .

The absorbed dose associated with the concentration of a radionuclide in water is dependent on a number of factors, including the amount of water taken up by an organism, the distribution of the radionuclide in tissue, and the energy of the particles emitted during decay.

### 10.2 ASSESSMENT OF AVAILABLE DATA

The data available are adequate for assessing the impact of specific produced water outfalls, using simple dilution models. More sophisticated analyses can be performed if additional data can be gathered. These additional data include geographic coordinates, receiving water flow rates, water depth and salinity.

The total impact to the region can be assessed if the following additional data can be gathered: 1) geographic coordinates for all of the outfalls represented in the State of Louisiana data set; 2) A determination of the percent of all produced water outfalls represented in the State of Louisiana data set and 3) Information describing the depth, volume, tidal excursion and fractional loss rate for coastal Louisiana.

### 10.3 CONCENTRATION FACTORS

Concentration factors can be used to calculate the concentration of radium in aquatic organisms, based on the concentration in water. These factors are affected by many variables, including the species, the concentration of radium in water and the portion of the animal analyzed.

The IAEA concentration factors commonly used in dose assessment studies are appropriate when water concentrations are relatively low, but are probably over-estimates for the relatively high concentrations that occur near produced water outfalls. These concentration factors also overestimate the concentration of radium in ingested fish and shellfish because they do not distinguish between the concentration in edible parts from the higher concentrations in bone, shell and exoskeleton.

# 10.4 EFFECTS OF RADIATION ON AQUATIC ORGANISMS

The available dose-response data describing the effects of chronic low-level radiation on aquatic organisms is variable and hard to summarize. The most sensitive life-stage appears to be fish-fry. An estimated no-effect level for individuals is 1-10 mGy/day (0.1-1.0) rad/day.

Natural populations are affected by many factors and effects of low-level chronic radiation are not likely to be detectable. Levels below 10 mGy/day (1 rad/day) are not expected to significantly effect natural populations of aquatic organisms.

# 10.5 HUMAN HEALTH EFFECTS

The EPA risk factors for radium-226 (4.4  $\times$  10<sup>-6</sup> per pCi/day lifetime intake) and radium-228 (3.3  $\times$  10<sup>-6</sup> per pCi per day lifetime intake) are based on conservative models and assumptions. More realistic, central estimates of the risk factors were derived (Ra-226: 2.6  $\times$  10<sup>-6</sup> Ra-228: 1.0  $\times$  10<sup>-7</sup>).

# 10.6 SCREENING-LEVEL ASSESSMENT

A conservative, screening-level assessment of the risk presented by radium discharged in Louisiana coastal waters suggests a level of risk which could be considered significant (greater than  $1 \times 10^{-5}$ ) for: 1) the person consuming the most seafood under all modeled discharge scenarios for a lifetime, and 2) a person consuming an average amount of seafood (for a lifetime) harvested within 100 feet of a high concentration discharge (500 pCi/l). These risk estimates are based on the following assumptions:

- 1. Radium discharged in produced water is reduced by a factor of 100 before it comes in contact with fish and shellfish consumed by people.
- 2. An individual gets one-half of their yearly fish and shellfish from near (i.e. where the dilution factor reaches 100; probably within 100 feet) a produced water outfall for their entire lifetime.
- 3. The use of conservative IAEA concentration factors in estimating the concentration of radium in fish and shellfish.
- 4. The use of conservative EPA risk factors.

A similarly conservative assessment of the potential for harm to aquatic biota suggests that no detectable impact on fish, molluscs or crustaceans is likely. The number of excess cancers predicted per year is comparable to the number expected to result from background concentrations

of radium. Because of the many conservative assumptions incorporated into this screening-level analysis, it can be concluded that the risks associated with the discharge of produced water to coastal Louisiana is small. The results of this study do, however, suggest the need for a more detailed analysis of the potential risks to individuals consuming seafood harvested near a produced water outfall.

Radium discharged offshore will be diluted more rapidly than radium discharged to nearshore waters. Organisms living offshore will have a smaller chance of coming into contact with discharged radium because of the large water volumes involved and the rapid dilution that occurs. An individual person is also not likely to harvest a significant amount of his yearly seafood close to an offshore outfall. Because of the additional reductions in the radium concentration in water and aquatic biota expected near offshore outfalls as compared to nearshore discharges, it can be concluded that the risks associated with offshore discharges will be extremely small.

### 10.7 UNCERTAINTIES AND CONSERVATISMS

The major uncertainties and conservatisms in this screening-level analysis are:

1. The concentration of radium in water and the geographic distribution of contaminated shellfish. The analysis of individual risk and dose to aquatic biota used concentrations likely to be measured at the outfall of a produced water discharge. In fact, considerable dilution occurs with distance from the discharge point. A reduction in fish and shellfish concentration also occurs with increasing distance. The extent to which contaminant plumes from neighboring outfalls intersect is also uncertain. The concentration of radium in the water in which fish and shellfish are harvested is critical to the estimation of risk.

For the calculation of population risk, a simple box model was used that assumed complete mixing of all discharged radium, and a resultant "average" concentration of radium in fish and shellfish harvested from the region. In fact, radium concentrations in water and in fish and shellfish are variable over the area.

- 2. The concentration factor used in calculating the concentration of radium in fish, molluscs and crustaceans. Commonly used concentration factors are higher than those derived from the CSA (1991) data set. The concentration factor used in the analysis has a large effect on the resulting risk estimates. Conservative IAEA concentration factors were used in this assessment, which probably resulted in an overestimate of the concentration of radium in food. These concentration factors also overestimate the concentration of radium in ingested fish and shellfish because they do not distinguish between the concentration in edible parts from the higher concentrations in bone, shell and exoskeleton.
- 3. The distribution of intake rates and the percent of consumed fish and shellfish that is radium contaminated. This analysis assumed that the maximally exposed individual harvested one-half of his seafood from

near a produced water outfall. The distribution of seafood intake among the population, and the percentage of the seafood consumed that is contaminated with radium is uncertain.

4. The risk factors for radium. This screening-level analysis used the conservative EPA risk factors for Ra-226 and Ra-228. Central estimates of the risk factors would predict smaller risks and fewer cancers associated with produced water discharges.

#### Section 11

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### APPENDIX A

# QUANTITIES AND UNITS OF RADIONUCLIDE ACTIVITY AND DOSE

### A.1 ACTIVITY

Radioactivity is quantified in terms of the number of spontaneous energy emitting transformations per unit time -- a quantity known as activity. An example of a transformation is the decay of a radium-226 nucleus into a radon-222 nucleus, an alpha particle, and gamma rays. The unit of activity has historically been the curie (Ci). One curie is equal to 3.7 x  $10^{10}$  disintegrations per second. In the International System of units (SI), the basic unit of activity has been redefined as one disintegration per second, known as the becquerel (Bq). One curie is equal to 3.7 x  $10^{10}$  Bq.

# A.2 EXPOSURE AND ABSORBED DOSE

Radiation exposure can be quantified by measuring the ionization produced in air by radiation. The quantity that expresses the ionization produced by x rays in a volume of air is known as exposure. Exposure is expressed in quantities known as roentgens (R). The roentgen is the amount of x ray or gamma radiation which produces  $2.58 \times 10^{-4}$  coloumb per kilogram (C/kg).

Exposure describes the ionization produced in air. The absorbed dose is the amount of energy imparted to matter. An absorbed dose of 100 erg/gm is called 1 rad. In the SI system the absorbed dose unit is 1 Joule per kilogram (J/kg), and 1 J/kg is called the Gray (Gy). An absorbed dose of 1 rad is equal to 0.01 Gy (1 Gy = 100 rads). Harmful levels of radiation doses are generally expressed in terms of rads. For example, over a hundred rads must be imparted in a short period of time to a substantial portion of the body before most individuals will show

significant clinical symptoms (Saenger, 1963). Occupational doses are not allowed to exceed a few mrad per hour (1 mrad =  $1 \times 10^{-3}$  rad).

An exposure of 1 roentgen of electromagnetic radiation in the energy range 0.1-3 Mev results in an absorbed dose of 0.96 rad in tissue (ICRU, 1962). For most purposes, values of exposures in roentgens can be considered essentially equal to the value of the absorbed dose in rads to tissue.

The absorbed dose associated with the concentration of a radionuclide in water is dependent on a number of factors, including the amount of water taken up by an organism, the distribution of the radionuclide in tissue, and the energy of the particles emitted during decay.

# A.3 QUALITY FACTORS AND DOSE EQUIVALENT

Some types of particles produce greater biological effects than others for the same absorbed dose. The relative effectiveness in producing cancer and severe genetic effects from equal absorbed doses of different radiation types has been found to correlate with the relative density of the ionizing events. The higher the linear energy transfer (LET), the greater the probability of producing cancer or genetic effects for a given absorbed dose.

The Quality Factor (QF) expresses the relative effectiveness of a particle based on its linear energy transfer. Values for the QF were derived from the literature on the effects of radiation on a variety of biological end-points (ICRP, 1977). The quality factor for gamma ( $\gamma$ ) and beta ( $\beta$ ) particles has been assigned to be 1. The quality factor for alpha ( $\alpha$ ) particles is set to 20 for assessment purposes (ICRP, 1977).

The absorbed dose modified by the quality factor is known as the dose equivalent and is expressed in quantities called rems.

Dose equivalent (rem) = Absorbed dose (rad) x QF

In the SI system, the dose equivalent is expressed in sieverts (Sv). One sievert is equal to  $100\ \mathrm{rem}$ .

Table A-1 gives the quantities, names and units for activity, exposure, absorbed dose and dose-equivalent. Table A-2 gives the prefixes commonly applied to these units.

Table A-1. Table of Radiological Quantities, Names and Units

Quantity	Traditi Name	aditional	Name	Unit	Conversion Factor
Activity	curie (Ci)	3.7 x 10 <sup>10</sup> /sec	becquerel (Bq)	1 sec -1	1 Bq = 2.7 x 10-11 Gi
Exposure	roentgen (R)	2.58 x 10 <sup>-4</sup> c/Kg		C/kg	10d /7   hr 1
Absorbed Dose	rad (rad)	100 erg/gm	gray (Gy)	l J/Kg	1 Gy = 100 rad
Dose equivalent	rem (rem)	100 erg/gm	sievert (Sv)	1 J/Kg	1 Sv = 100 rem

Table A-2. Table of Prefixes

tera (T) - $10^{12}$	giga (G) - 10 <sup>9</sup>	$mega (M) = 10^6$	kilo $(k) = 10^3$
milli (m) - 10 <sup>-3</sup>	micro (u) - 10 <sup>-6</sup>	nano (n) = $10^{-9}$	pico (p) $-10^{-12}$

#### APPENDIX B

### Glossary

Absorbed Dose: The amount of energy imparted to matter. Historical unit is the rad. The unit in the International System of Units is the Gray.

Alpha ( $\alpha$ ) Particle: A helium nucleus, consisting of two neutrons and two protons. Commonly emitted by the heaviest nuclides in the periodic table.

Beta  $(\beta)$  Particle: Electrons emitted by a nucleus as a result of energy released in a radioactive decay process involving the transformation of a neutron into a proton.

Becquerel (Bq): The unit for activity in the International System of Units, replacing the curie (Ci).

Concentration Factor: A function of the concentration of a contaminant in water or sediment, and the concentration in an organism. Used to estimate the concentration of a contaminant in organisms from the measured concentration in water.

Curie (Ci): The historical unit of radionuclide activity. Activity is the number of spontaneous energy emitting transformations per unit time.

Dose: See absorbed dose.

Dose Equivalent: The dose equivalent is the absorbed dose modified by the quality factor. The historical unit for dose equivalent is the rem. The new unit is the sievert.

Exposure: The ionization produced in air. Described in units called roentgens (R).

Gamma  $(\gamma)$  Ray: Electromagnetic radiations emitted by radioactive nuclei as packets of energy, called photons, and often accompany the emission of Beta particles from the same nuclei.

Gray (Gy): The unit for absorbed dose in the International System of Units. Replaces the rad.

Individual Lifetime Risk: The increase in probability that an individual will experience a specific adverse effect as a result of a continuous lifetime exposure to a risk agent.

Population Risk: The number of cancer (or other effect) cases resulting from one year of exposure.

Produced Water: The saline water that can accompany petroleum production.

Quality Factor: Used to express the relative effectiveness of a particle based on its linear energy transfer (LET). Radiation types with higher LETs have a greater probability of causing harm, and have a higher quality factor.

Rad: The historical unit for absorbed dose. Replaced by the Gray.

Rem: Historical unit for dose equivalent. Replaced in the International System of Units by the Sievert.

Risk: The possibility of suffering harm from a hazard.

Risk Assessment: Process that estimates the relationship between a source term (e.g. a contaminant discharge) and the potential resulting effects on human health and the environment.

Roentgen (R): The unit which describes radiation exposure.

Sievert (Sv): The unit for dose equivalent in the International System of Units. Replaces the rem.