50

Assessment of Aquatic Organisms as Bioindicators of Historical Radionuclide Release to the Columbia River

D. D. Dauble

T. M. Poston

R. L. Newell

December 1988

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE MEMORIAL INSTITUTE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

NTIS Price Codes
Microfiche A01

Printed Copy

Pages	Price Codes
001-025	A02
026-050	A03
051-075	A04
076-100	A05
101-125	A06
126-150	A07
151-175	A08
176-200	A09
201-225	A10
226-250	A11
251-275	A12
276-300	A13

**ASSESSMENT OF AQUATIC ORGANISMS AS
BIOINDICATORS OF HISTORICAL RADIONUCLIDE
RELEASE TO THE COLUMBIA RIVER**

D. D. Dauble
T. M. Poston
R. L. Newell

December 1988

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

EXECUTIVE SUMMARY

This study examined the potential for using several aquatic organisms as biological indicators of historic levels of radionuclides released to the Hanford Reach of the Columbia River. The purpose of the study was to determine the types of environmental samples that could be collected to further our understanding of previous releases of radionuclides at Hanford.

Information was initially collected to determine the relative abundance and persistence of radionuclides historically released at Hanford. The potential for long-lived radionuclides to bioaccumulate in aquatic organisms was then assessed. The life history of several common aquatic organisms was examined to evaluate their use as potential bioindicators of radionuclides released to the Columbia River. Considerations for analyzing strontium (^{90}Sr) in biological samples were determined.

Based on our review of radionuclides released to the environment and their potential for bioaccumulation in aquatic organisms, strontium appears to be the only radionuclide suitable for further study. White sturgeon (*Acipenser transmontanus*) and the common mussel (*Margaritifera margaritifera*) are suitable candidates for developing dose reconstruction scenarios. Considerations for tissue analysis of radionuclide concentration in these species include potential for biological turnover and tissue mass.

ACKNOWLEDGMENTS

We thank J. K. Soldat for helpful comments on earlier versions of the manuscript and K. R. Price for assistance with the literature review. G. P. O'Connor provided essential editorial advice, and M. A. Eierdam typed the report.

CONTENTS

EXECUTIVE SUMMARY.....	iii
ACKNOWLEDGMENTS.....	v
INTRODUCTION.....	1
BACKGROUND.....	3
BIOACCUMULATION POTENTIAL OF RADIONUCLIDES IN AQUATIC ORGANISMS.....	7
DATA REVIEW.....	7
POTENTIAL FOR BIOACCUMULATION.....	8
CONCLUSIONS.....	13
AQUATIC ORGANISMS WITH POTENTIAL FOR RECONSTRUCTION OF HISTORIC DOSE.....	15
FISH SPECIES.....	15
White Sturgeon.....	15
Carp.....	16
Largescale Sucker.....	16
Northern Squawfish.....	17
AQUATIC INVERTEBRATE.....	18
Freshwater Mussel.....	18
CONCLUSIONS.....	18
EXPOSURE SCENARIO	21
ASSUMPTIONS	21
ANALYSIS	22
CONCLUSIONS.....	22
SUMMARY.....	25
REFERENCES.....	27

FIGURE

- 1 Relationship Between Reactor Operations and Whole-Body
Dose from Radionuclides Released to the Environment.....4

TABLES

- 1 Relative Abundance of Reactor Effluent Radionuclides
in the Columbia River8..
- 2 Total Reported Releases of Radionuclides to the Columbia
River from Hanford Reactors During 19749
- 3 Summary of Physical Half-Life, Biological Turnover, and
Bioaccumulation Potential for Long-Lived Radionuclides.....1 1
- 4 Biological Characteristics of Potential for Bioindicators in the Hanford Reach
of the Columbia River.....19

INTRODUCTION

The potential impacts to humans and the environment resulting from historic releases of radionuclides at Hanford have generated considerable public interest, in part because of the U.S. Department of Energy's (DOE) decision to release declassified documents regarding radiation releases over the last **40** years. As a result of this interest, scientists at Pacific Northwest Laboratory (PNL) are attempting to reconstruct historic radionuclide levels. The purpose of the study was to examine the possibility of using aquatic organisms as biological indicators of past radionuclide levels at Hanford.

In a preliminary analysis, available data were reviewed to determine the relative abundance of radionuclide concentrations in reactor effluents and to estimate the bioaccumulation potential for long-lived radionuclides. We examined the potential for using several aquatic organisms as biological indicators of historic levels of radionuclides present in the Hanford Reach of the Columbia River. A sample exposure scenario was then developed using ^{90}Sr as an indicator of historic radionuclide releases to the river.

Although the potential for transport of radionuclides to aquatic organisms has been studied to develop exposure pathway models, the application of these models has generally been limited because of lack of historic verification. Additional information on historic contamination of the environment could be provided if the relationship between time and tissue burden of radionuclides could be determined for long-lived species of aquatic organisms.

This report explains the data examined, assumptions made, and the calculations used to determine future research needs. The potential application of results to predictive models and to monitoring programs is outlined, and recommendations are made for areas of further study.

BACKGROUND

In 1944, the first plutonium production reactor at Hanford began discharging cooling water containing radionuclides into the Columbia River. Eventually, up to eight reactors with once-through cooling and the 100-N Reactor, with secondary cooling, were operating at one time. The total annual radiation dose to humans, resulting from the release of radionuclides to the environment, was related to the start-up and operation of these reactors (Figure 1). The last once-through-cooled reactor closed in January 1971, significantly reducing the release of radionuclides to the Hanford Reach of the Columbia River.

When radionuclides are released into the river, they disperse in solution, bind to organic and inorganic particles, or are taken up by aquatic biota. Eventually, the dissolved and particulate-bound radionuclides are transported downstream where they enter the ocean or are stored in sediments in the river or estuary. These processes of radionuclide flux between water, sediment, and organisms are highly dynamic. Availability of radionuclides to aquatic biota is related to the relative concentrations and chemical characteristics of radionuclides that were discharged, river flows (dilution), sediment load, and distance downstream of the reactors (Hauschild, Dempster, and Stevens 1975; Hauschild et al. 1971; Perkins, Nelson, and Hauschild 1966).

Concentration of contaminants in freshwater organisms is dependent on several factors, including 1) the physical-chemical properties of the contaminant; 2) quantity of the contaminant; 3) physical and chemical features of the aquatic system (i.e., water quality and temperature); 4) trophic relationships among organisms; and 5) the metabolic pathways within organisms. Radionuclides that enter the aquatic ecosystem may be assimilated by organisms following absorption through the integument, gills, or digestive tract. The significance of uptake through the food web depends on feeding habits and mechanisms of storage and elimination.

Radionuclide concentrations in higher trophic-level organisms tend to be lower than concentrations in their food. For example, uranium, thorium, and radium are apparently all discriminated against in freshwater trophic chains. Concentrations of these elements decreased with increasing trophic level.^(a) However, there may also be instances where tissue concentrations of higher trophic-level organisms exceed those found in the environment.

(a) Final report. 1980. Critical Biological Pathways for Transfer of Radioactive and Heavy Metal Contaminants to Biota and Man in Relation to Development of the Uranium Industry in British Columbia. Prepared for the Royal Commission of Inquiry into Uranium Mining, Battelle, Pacific Northwest Laboratories, Richland, Washington.

FIGURE 1. Relationship Between Reactor Operations and Whole-Body Dose from Radionuclides Released to the Environment. Doses up to 1974 based on environmental data; post-1974 doses based on effluent data. Whole-body doses are to the maximum individual from all pathways, including fallout.

The decline in tissue concentrations of radionuclides in selected Columbia River biota after shutdown of the production reactors has been described by Cushing et al. (1980). They found that the measurable concentration of fission-produced radionuclides was nearly undetectable 2 yr after radionuclide input from the reactors ceased. However, studies have not been attempted to reconstruct historic radionuclide levels for long-lived aquatic organisms that have resided in the Hanford Reach since reactor operations began.

BIOACCUMULATION POTENTIAL OF RADIONUCLIDES IN AQUATIC ORGANISMS

This section describes an initial analysis of radionuclide concentrations and their potential for uptake and storage by aquatic organisms. It was necessary to understand several conditions concerning radionuclide behavior in the environment before assessing which radionuclides might be useful in reconstruction of historic concentrations. Initial analysis was based on the following criteria:

1. The radionuclide of interest was released into the Columbia River in large enough quantities so that it is reasonable to assume that it will be found in environmental samples.
2. The radionuclide has a long enough physical half-life so that detectable concentrations still could be present.
3. The radionuclide has a slow turnover in biological tissue. In most cases, this limits biological samples to hard (calcified) tissue.
4. The radionuclide is representative of the total radioactive releases to the river from reactor operations (i.e., a radionuclide that was unique to only one aspect of Hanford operations would not be related to all operations).

DATA REVIEW

The first step in the analysis was to determine if historic data were available describing radionuclide concentrations in reactor effluents and Columbia River water. The main concern of the analysis was the period of time when reactors were operating (pre-1975). An examination of early radiological monitoring reports revealed that gross alpha and beta measurements were taken, but information on specific radionuclides was not available.

In 1957, with the development of the gamma spectrometer, specific radionuclide analyses were reported at Hanford. Subsequent monitoring reports typically noted the relative abundance of reactor effluent radionuclides from reactor cooling water (Table 1). Although ⁹⁰Sr was analyzed in Columbia River water from 1957-1975 (Healy et al. 1958; Anderson 1959; Junkins et al. 1960; Nelson 1961; Soldat and Essig 1966; Essig and Soldat 1967; Woolridge and Essig 1970; Woolridge 1962, 1965, 1969, 1973; Wilson 1963; Wilson 1970; Corley 1970; Bramson and Corley 1972; Nees and Corley 1974; Fix 1975; Blumer, Fix, and Speer 1976), data included mainly concentrations of relatively short-lived radionuclides (Table 2).

TABLE 1. Relative Abundance (as Percent of Activity) of Reactor Effluent Radionuclides in the Columbia River (Wilson 1963)^(a)

<u>Major (90%)</u>	<u>Minor (8%)</u>		<u>Trace (2%)</u>	
²⁴ Na	³ H	⁹⁰ Sr	¹¹¹ Ag	¹⁴⁹ Pm
³¹ Si	¹⁴ C	⁹¹ Sr	¹¹⁵ Cd	¹⁵¹ Pm
⁵¹ Cr	³² P	⁹² Sr	¹³¹ I	¹⁴⁷ Nd
⁵⁶ Mn	³⁵ S	⁹¹ Y	¹³² I	¹⁴⁹ Nd
⁶⁴ Cu	⁴⁵ Ca	⁹² Y	¹³⁷ Cs	¹⁵³ Sm
⁷⁶ As	⁴⁶ Sc	⁹³ Y	¹⁴⁰ Ba	¹⁵⁶ Sm
²³⁹ Np	⁴⁷ Sc	⁹⁵ Zr	¹⁴⁰ La	¹⁵² Eu
	⁵⁴ Mn	⁹⁷ Nb	¹⁴¹ La	¹⁵⁶ Eu
	⁵⁹ Fe	⁹⁹ Mo	¹⁴¹ Ce	¹⁵⁷ Eu
	⁶⁰ Co	¹⁰³ Ru	¹⁴³ Pr	¹⁶⁰ Tb
	⁹⁰ Sr	¹³³ I	¹⁴⁴ Ce-Pr	¹⁸⁷ W
	⁶⁵ Zn	¹³⁵ I	¹⁴² Pr	²¹⁰ Po
	^{69m} Zn	²³⁹ U	¹⁴³ Pr	²²⁷ Ac
	⁷² Ga		¹⁴⁵ Pr	²³⁸ U
	⁸⁵ Sr		¹⁴⁷ Pm	²³⁹ Pu

(a) Values are adjusted to 4-hr post-irradiation.

As described in these reports, ⁹⁰Sr concentrations were measured in Columbia River water taken from either the Pasco or Richland water supply systems. The average annual concentration from 1958-1975 varied from non-detectable to 1.0 pCi/L. Analysis of other long-lived radionuclides began in 1972 (Bramson and Corley 1973). In 1974, ²³⁹Pu and ¹⁵²Eu were also analyzed (Fix 1975). The average annual measured concentration of these radionuclides for 1974-1975 did not exceed 2.0 pCi/L.

This review of data supported the assumption that historic information on discharge of radionuclides from reactors was not detailed enough to reconstruct concentrations of radionuclides in the river.

POTENTIAL FOR BIOACCUMULATION

The next step in the analysis was to determine which radionuclides could persist in the environment long enough to still be detectable in tissues of aquatic organisms 20 to 40 yr after they were released.

TABLE 2. Total Reported Releases of Radionuclides to the Columbia River from Hanford Reactors during 1974 (Fix 1975)

<u>Radionuclide</u>	<u>Total Release (Ci)</u>
^3H (HTO)	190
^{24}Na	1.0
^{32}P	0.004
^{46}Sc	0.02
^{51}Cr	0.22
^{54}Mn	0.5
^{56}Mn	5.0
^{58}Co	0.02
^{59}Fe	0.18
^{60}Co	1.2
^{65}Zn	0.2
^{76}As	0.03
^{90}Sr	0.3
^{95}Nb	0.1
^{95}Zr	0.11
^{99}Mo	<0.6
^{99}Tc	<0.1
^{103}Ru	0.12
^{106}Ru	0.5
^{122}Sb	<0.01
^{124}Sb	<0.07
^{125}Sb	0.02
^{131}I	2.33
^{133}Xe	0.003
^{137}Cs	0.12
$^{140}\text{BaLa}$	0.8
^{141}Ce	0.05
^{144}Ce	0.103
^{187}W	<0.01
^{239}Np	0.02

Major classes of radionuclides were initially grouped according to their physical decay rates, using a radiological half-life of >10 yr as a lower limit (Table 3). Based on decay alone, such radionuclides released 20 yr ago would be present at $\leq 25\%$ of their historic level.

The retention time of radionuclides in biological tissue is another key variable that must be considered in determining historic radionuclide concentrations. Because little information is available on the turnover of radionuclides in specific tissues of aquatic organisms, values listed for human tissue in Napier et al. (1988) were used. Most long-lived radionuclides turn over fairly slowly in biological tissues (half-time >1 yr). However, even at this rate, little of the absorbed radionuclide would be retained 20-30 yr after initial uptake. The last column in Table 3 lists the concentration ratios (CR) reported for various aquatic organisms exposed to radionuclides in water (Poston and Klopfer 1986; Napier et al. 1988). No apparent relationship existed between the ability of an aquatic organism (mainly fish) to take up radionuclides from aqueous routes (as shown by the CR value) and either the physical half-life or the biological half-time (Table 3). This may be because many other chemical and physical variables exert a greater influence on the behavior of radionuclides in the environment and/or their bioavailability to aquatic organisms. However, the CR value provides a relative measure of the ability of aquatic organisms to absorb and store a particular element.

The list of potentially long-lived radionuclides was reduced by eliminating radionuclides that had both a short radiological half-life (<10 yr) and a short biological half-time (<1 yr). However, based on the radiological half-life (physical decay rate) and the biological half-time, only three radionuclides could be expected to be retained long enough in animal tissue to be analyzed 20 yr after exposure: ^{210}Pb , ^{90}Sr , and ^{226}Ra . Most other radionuclides (those with biological half-time <2 yr) would be expected to be present at <0.1% of the maximum tissue levels measured 20 yr ago. Thus, historic release levels and subsequent tissue burden would have to be 200-500 times greater for the short-lived radionuclides to have comparable tissue burdens today.

Radium-226 and ^{210}Pb are naturally occurring radionuclides that originate from the decay of ^{238}U . Releases of ^{238}U (half-life $>10^9$ yr) from Hanford reactors have not been high enough to increase levels of ^{226}Ra or ^{210}Pb in the Columbia River. Additionally, the concentration of ^{238}U in the river is high because of inputs from the Spokane River (Silker 1964). Thus, radionuclides of the uranium series are not suitable candidates for reconstructing historic river concentrations.

TABLE 3. Summary of Physical Half-Life Biological Turnover, and Bioaccumulation Potential for Long-Lived Radionuclides

<u>Element</u>	<u>Radionuclide</u>	<u>Physical Half-life (yr)</u>	<u>Tissue</u>	<u>Biological Half-time (yr)</u>	<u>Concentration Ratio (a)</u>
Nickel	^{59}Ni	75,000	Whole body	1.8	100
			Liver	1.4	
	^{63}Ni	96	Bone	2.2	
Strontium	^{90}Sr	29.1	Whole body	11.0	50
			Bone	11.0	
Zirconium	^{93}Zr	1,500,000	Whole body	1.2	200
			Kidney	2.5	
			Bone	2.7	
Niobium	$^{93\text{m}}\text{Nb}$	13.6	Whole body	2.1	200
	^{94}Nb	20,300	Kidney	2.1	
			Liver	2.3	
			Bone	2.7	
Samarium	^{151}Sm	90.0	Whole body	1.8	300
			Kidney	1.8	
			Bone	4.1	
Europium	^{152}Eu	13.3	Whole body	1.7	300
			Kidney	4.1	
			Bone	4.1	
Holmium	$^{166\text{m}}\text{Ho}$	12,000	Whole body	2.1	300
			Kidney	2.2	
			Liver	2.4	
			Bone	2.7	
Lead	^{210}Pb	22.3	Whole body	4.0	2000
			Liver	5.3	
			Bone	10.0	

TABLE 3. (contd)

<u>Element</u>	<u>Radionuclide</u>	<u>Physical Half-life (yr)</u>	<u>Tissue</u>	<u>Biological Half-time (yr)</u>	<u>Concentration Ratio (a)</u>
Radium	^{226}Ra	1,600	Whole body	22.2	50
			Bone	45	
Thorium	^{229}Th	7,340	Whole body	100	100
	^{230}Th	77,000	Kidney	60	
			Liver	160	
			Bone	100	
Protactinium	^{231}Pa	32,800	Whole body	110	11
			Kidney	140	
			Liver	160	
			Bone	100	
Neptunium	^{237}Np	2,100,000	Whole body	110	50-2500
			Kidney	180	
			Bone	100	
Plutonium	^{238}Pu	87.7	Whole body	180	250
	^{239}Pu	24,100	Kidney	88	
	^{240}Pu	6,540	Liver	40	
	^{241}Pu	14.4	Bone	100	
	^{242}Pu	376,000			
Americium	^{241}Am	432	Whole Body	55	800
	$^{242\text{m}}\text{Am}$	152	Kidney	74	
	^{243}Am	7,380	Liver	40	
			Bone	100	
Curium	^{243}Cm	28.5	Whole Body	66	50-2500
	^{244}Cm	18.1	Kidney	66	
	^{245}Cm	8500	Liver	40	
			Bone	100	

(a) Concentration Ratio = $\frac{\text{pCi/kg tissue}}{\text{pCi/L water}}$

CONCLUSIONS

Strontium-90 appears to be the only candidate radionuclide suitable for further study. However, it is important that the relative contribution of fallout ^{90}Sr to the total concentration measured in the river be considered. Plutonium and americium were not considered further because of their high analytical costs and extremely low concentrations.

AQUATIC ORGANISMS WITH POTENTIAL FOR RECONSTRUCTION OF HISTORIC DOSE

This section describes four fish species and one aquatic invertebrate that were assessed to determine their suitability for analysis of historic radionuclide uptake. Discussion was limited to aquatic organisms with known life spans that exceeded 15 yr, because it was assumed that levels of available radionuclides present in the Columbia River water and sediments after 1972 were too low to attempt reconstruction of historic dose. Only longer-lived organisms would be useful as potential bioindicators of river concentrations in the early days of reactor operation (i.e., 1940-1950s).

FISH SPECIES

White Sturgeon (*Acipenser transmontanus*)

White sturgeon are a long-lived species that reside year-round in the Hanford Reach. They are primarily bottom feeders and can take up radionuclides directly from the water by ingesting contaminated sediments and via the aquatic food chain. Concentrations of various radionuclides were determined for white sturgeon tissues during radiological studies conducted in 1966-67 (Watson et al. 1970). However, no attempt was made to determine relationships among fish age and radionuclide concentration in carcass or muscle tissue. Sturgeon have been known to accumulate persistent organochlorine compounds [i.e., polychlorinated biphenyls (PCBs) and pesticides] in tissues (Bosley and Gately 1981); residue levels generally increased in direct proportion to their age.

The migrational characteristics of white sturgeon relate to its use as an indicator of general environmental contamination in the Hanford Reach. Because sturgeon cannot move upstream through fish ladders, their movements since 1960 have been restricted to the area bound by Priest Rapids, McNary, and Ice Harbor dams. However, seasonal movement throughout the McNary Pool is known to occur (Haynes 1978), and migration range could have been greater for sturgeon that existed before construction of McNary and Priest Rapids dams. Thus, even for sturgeon captured near former production reactors, exposure may not be directly related to radionuclide concentrations present at that location.

White sturgeon have not been aged at Hanford. However, based on age-growth studies in the Columbia River and other river systems (Semakula and Larkin 1968; Galbreath 1979; Malm 1981) and known size of sturgeon in the Hanford Reach, individuals >25 yr of age probably occur in the reach. Thus, it is likely that a small percentage of the present-day white sturgeon population has existed in the Hanford

Reach before and during reactor operations. Sturgeon are aged by sectioning pectoral fins. This method can be employed without killing the fish.

White sturgeon are cartilaginous and therefore have different physiological characteristics than true bony fishes (teleosts). This difference could affect the retention and clearance of environmental contaminants. For example, sampling for radionuclides that are deposited mainly in calcified tissues could be limited. However, available tissue mass for analysis is much greater for sturgeon than for other fish species because of their large size.

Carp (*Cyprinus carpio*)

The common carp is a relatively abundant species that resides throughout the McNary Pool and Hanford Reach. Adult carp feed on a variety of items, including aquatic plants, bottom insects, and surface detritus. Carp generally prefer areas of low current velocity such as nearshore or backwater environments of the Hanford Reach. Carp have been used to determine the uptake of radionuclides from artificial stream systems. Juvenile carp exposed to experimental mixtures of radionuclides that originated from the N Reactor trench readily accumulated certain radionuclides (Cushing et al. 1988).

General patterns of movement and residence time of carp in the Hanford Reach are unknown. Dense populations of carp occur in sloughs during spawning. Limited information based on tag recaptures indicate that although carp may reside in the same areas during certain seasons, movement of up to 32 km occurs (Page, Dauble, and Neitzel 1982). Thus, carp would be expected to be exposed to highly variable concentrations of radionuclides in the river.

The normal life span of carp in natural environments is up to 20 yr (McCrimmon 1968), although longer life spans have been speculated for carp in captivity. No studies of the age and growth of Columbia River populations have been conducted. However, specimens as large as 60 cm long have been collected at Hanford, which suggests that ages to 20 yr may be possible. Carp may be aged by counting annuli formed on scales, pectoral spines, or vertebrae.

Largescale Sucker (*Catostomus macrocheilus*)

Largescale sucker are the most common species of sucker found in the Hanford Reach (Gray and Dauble 1977). They are also one of the most abundant fish species in the river; peak population estimates of up to 15,000 largescale sucker per kilometer were obtained in late spring 1981 near river mile 352 (Dauble 1987). Largescale sucker are omnivorous, consuming mainly benthic algae and insects. Higher tissue burdens might be expected for a bottom-feeding species like sucker because of their

greater likelihood to contact sediment-bound contaminants. Concentrations of biologically important radionuclides were determined for largescale sucker tissues during studies in 1966-1967 and again in 1971-1973, following reactor shutdown (Watson et al. 1970; Cushing et al. 1980). Data suggested that largescale sucker did not accumulate any more radionuclides than did other fish species sampled.

Largescale sucker show marked seasonal movement that may be related to spawning or feeding migrations (Dauble 1987). Sucker move throughout the Hanford Reach and migrate over Priest Rapids Dam, although some individuals may have more restricted, localized movement patterns. Thus, no evidence exists that long-term or continuous exposure to radionuclides would occur at a fixed location.

The maximum age of largescale sucker at Hanford has been estimated at about 15 yr by scale analysis (Dauble 1978). However, fin rays generally provide a more accurate measure of age past maturity (Beamish and Harvey 1969), and scale ages are frequently less than those obtained by examining fin rays. Thus, it is possible that some larger individuals in the Hanford Reach may be >15 yr old.

Northern Squawfish (*Ptychocheilus oregonensis*)

Squawfish are a wide-ranging species of cyprinid that are highly piscivorous as adults. Because they are higher on the aquatic food chain, squawfish may be more likely to accumulate certain persistent environmental contaminants than lower trophic-level organisms. Squawfish were also studied in radiological surveys conducted by Watson et al. (1970) and Cushing et al. (1980), but general trends in radionuclide concentrations for tissues were not markedly different than for any other fish species examined.

Squawfish are highly mobile and known to move throughout the Hanford Reach and between reservoir systems on the Columbia and Snake rivers. They have localized seasonal increases in abundance below dams that may be related to predation on downstream migrating juvenile salmonids (Nigro et al. 1985).

No definitive studies on the age and growth of squawfish have been conducted for the Hanford Reach. However, squawfish are a relatively long-lived species in other river and lake systems. Maximum age in Montana is reported to be 19 yr (Brown 1970). Based on relative lengths of squawfish collected at Hanford, maximum ages may be similar in the Hanford Reach. Age analysis by scales is the most common method used, but analysis of fin rays may be more accurate.

AQUATIC INVERTEBRATE

Freshwater Mussel (*Margaritifera margaritifera*)

The freshwater mussel burrows into the bottom substrate and feeds by filtering organic detritus from the water. These bivalves may occur as individuals, or they may be found in dense colonies, depending on hydrologic conditions. Although mussels are mobile, and downstream displacement may occur, older mussels are largely sessile (Vannote and Minshall 1982).

Because bivalves filter large quantities of water and detritus, they are considered good bioindicators of water quality changes in the environment (Mellinger 1972). Studies of radionuclide uptake by invertebrates at Hanford have been limited. Watson et al. (1970) reported seasonal trends of some radionuclides in limpet (*Fisherola nuttalli*) soft tissue and shell. This small univalve lives <5 yr. No comparative information is available on common bivalves in the Hanford Reach (i.e., *Anadonta* sp. and *Corbicula* sp.)

M. margaritifera is the longest-lived freshwater invertebrate known, with some individuals reported at more than 100 yr of age (Comfort 1957). The age distribution of mussels tends to be uniform within a single bed (Chatters 1987). Maximum age of individuals collected in the Columbia River system has been estimated to range from 50 to 100 yr. Thus, it may be possible to analyze levels of radionuclides in mussels that were present before, during, and after shutdown of Hanford production reactors.

CONCLUSIONS

Movement patterns of all four fish species studied are generally limited to the Hanford Reach and McNary Dam Pool. However, movement for some individuals may range up to 50 km during their life span, and hydroelectric dams represent a barrier only to white sturgeon. Older sturgeon that were present prior to construction of McNary Dam (i.e., 1954) and Priest Rapids Dam (i.e., 1960) may have migrated greater distances. The relationship between tissue burdens and exposure to radionuclides based on reactor discharge concentrations would be difficult to establish for any mobile aquatic species because of potentially wide differences in radionuclide concentration within a species range. Only limited numbers of carp, sucker, or squawfish that may have existed during historic radionuclide releases would still be present. However, sufficient numbers of white sturgeon should exist to determine whether a correlation is present between fish age and tissue concentration of long-lived radionuclides. Because of the affinity of some long-lived radionuclides to particulate material and bottom sediments, sturgeon and sucker would be most likely to have been exposed to greater concentrations in the environment. No advantages are

evident for any fish species based on differences in aging methods; techniques are available to accurately age all four species.

Evidence exists that historic populations of *M. margaritifera* occurred in the Hanford Reach, based on the presence of shell mounds. One site is located immediately downstream of several reactor outfalls. However, no surveys have been conducted to document the locations or to determine the abundance of present-day populations. Because freshwater mussel populations have not been located, and their basic life history is not well characterized, their usefulness as a biomonitor species could be limited. One advantage of this species, however, is that the biological half-life of radionuclides is probably infinite because the shell is not vascularized as is fish bone.

A summary of key biological characteristics that relate to the potential usefulness of these organisms as biomonitor species is provided in Table 4.

TABLE 4. Biological Characteristics of Potential Bioindicators in the Hanford Reach of the Columbia River

<u>Species</u>	<u>Expected Maximum Age (yr)</u>	<u>Feeding Strategy</u>	<u>Movement</u>
White sturgeon	25-50	Benthic omnivore	Mobile
Common carp	10-20	Omnivore	Mobile
Largescale sucker	10-20	Benthic omnivore	Mobile
Northern squawfish	10-20	Predator	Mobile
Common mussel	50-100	Filter-feeder	Sessile

EXPOSURE SCENARIO

This section presents a possible exposure scenario using strontium as an indicator of historic input of ^{90}Sr to the Columbia River. The objective of this exercise is to estimate the sample size (grams of tissue) that would be required to estimate historic uptake of strontium by organisms found in the river. It was assumed that the analytical approach would involve one of two methods. The first would collect a subsample of hard tissue from an organism that has inhabited the Columbia River for decades. This subsample would consist only of tissue laid down during the year or years of interest, e.g., rings (annuli) from fin rays, spines, vertebrae or clam shells. The ^{90}Sr concentration would then be determined for years of interest based on organism age. The second method would collect whole-body samples from organisms of differing ages. This method would require that a relationship between organism age and radiological tissue burden be established; yearly inputs of radionuclides would then be estimated from differences in hard tissue burdens between organisms of different ages.

Because of its valence state and chemical similarity to calcium, strontium is expected to accumulate in bone [apatite: $\text{Ca}_5(\text{OH})(\text{PO}_4)_3$] and mollusk shell (calcite or aragonite: CaCO_3) at levels inversely proportional to background concentrations of calcium and stable strontium in water. The radionuclide ^{90}Sr is a fission product with a half-life of 29.1 yr. It is a pure beta emitter, and analysis of prepared tissues involves liquid scintillation counting techniques.

ASSUMPTIONS

In conducting an initial analysis, we made several assumptions, as described below.

1. Biological half-time of strontium in bone and mollusk shell is infinite. This may be true for shell, but fish bone is vascularized, and biological half-time may play a critical role in the ability to detect ^{90}Sr in older fish bone (see Table 3 for biological half-time of radionuclides in human bone).
2. No allowance is made for physical half-life (radiological decay). Because the half-life of strontium is equal to or less than the time Hanford reactors operated, physical half-life would affect our ability to detect ^{90}Sr in older bone or shell tissue.
3. The relationship for bioaccumulation of strontium in fish bone developed by Vanderploeg et al. (1975) was used to estimate the concentration factor (CF) of strontium in fish bone and mollusk shell. This relationship is:

$$\text{CF} = e^{(9.59 - 1.15 \ln [\text{Ca}])}$$

where [Ca] is the calcium concentration in water expressed in mg/L. The calcium content in shell and fish bone is 40% by weight, and the accumulation of ^{90}Sr is inversely proportional to the water concentration of calcium. Consequently, the use of this equation provides a conservative estimate of the amount of ^{90}Sr in calcified tissue.

4. The concentration of calcium in Columbia River water is assumed constant over the last 40 plus years at 17.3 mg/L (value based on river water sample collected at the 300 Area intake).

ANALYSIS

The analysis tested two cases, as described below.

- Case 1 assumed a mean annual concentration of 2 pCi/L ^{90}Sr in Columbia River water. From the Vanderploeg et al. (1975) equation, and the concentration of 17.3 mg/L Ca in Columbia River water, it is concluded that the CR for ^{90}Sr is 551. Therefore, it is assumed that 1102 pCi ^{90}Sr /kg of hard tissue (1.1 pCi/g) is present. This amounts to about 2.5 dpm/g. Assuming 100% counting efficiency and a minimum detectable limit of 50 dpm, about 20 g of tissue would be necessary for analysis.
- Case 2 involved a clam or a fish living in 100-N springs, which has a ^{90}Sr concentration of 1036 pCi/L (Fruchter et al. 1984). With a CF of 551, it is expected that 570,800 pCi/kg (570.8 pCi/g) would be present in the hard tissue of either organism. This amounts to a requirement of 40 mg of tissue per sample.

CONCLUSIONS

It would be difficult to obtain adequate tissue for analysis using the annuli subsample method if environmental concentrations were similar to those described under Case 1. The environmental conditions under Case 2 provide a more reasonable sample mass if annuli subsample methods are desired, but the concentration of ^{90}Sr in the 100-N springs would never be approached in the Columbia River after dilution. As indicated earlier, the mean annual concentration of ^{90}Sr (1958-1975) ranged from less than detectable to 1 pCi/L. Consequently, tissue mass requirements are greater by several orders of magnitude than those indicated in Case 1.

The conservative assumptions discussed above are also structured to favor a high concentration of ^{90}Sr in hard tissue. If we deviated from these assumptions, i.e., if biological turnover was more rapid, the

amount of radionuclide in hard tissue would be decreased. The lower value would require more material for analysis than indicated.

SUMMARY

Only two aquatic organisms have potential for developing dose reconstruction scenarios. Further information on the turnover of ^{90}Sr in white sturgeon soft tissues and cartilage could be collected by examination of the literature, or by conducting laboratory studies. If biological turnover of long-lived radionuclides was found to be relatively slow, then sampling and analysis could be initiated for ^{90}Sr in target tissues of white sturgeon. Sturgeon should be collected from several locations, including a control group above Priest Rapids Dam, and sufficient individuals should be collected from each site to determine whether correlations between tissue burden and age/length exist.

The location, abundance, and age-structure of common mussel populations should be determined in the Hanford Reach. If enough old (>20 yr) individuals are found, analysis for ^{90}Sr could be initiated. Because of the sample size required for detection, whole-shell concentrations of ^{90}Sr , rather than concentrations found in annuli, would have to be used to establish whether a relationship exists between radionuclide concentration and age of the mussel.

Some information on tissue concentrations of ^{90}Sr for long-lived aquatic organisms at Hanford could be obtained from existing monitoring programs. Specific sampling procedures should be implemented where this information is lacking. Data obtained could then be used to develop a preliminary model to predict tissue concentrations in aquatic organisms. These data could also be used to predict potential exposure for organisms consumed by humans.

REFERENCES

- Anderson, B. V. 1959. Hanford Environmental Monitoring Annual Report 1958. HW-61676, Hanford Atomic Products Operation, General Electric, Richland, Washington.
- Beamish, R. J., and H. H. Harvey. 1969. "Age Determination in the White Sucker." Fish. Res. Bd. Canada. 26:633-638.
- Blumer, P. J., J. J. Fix, and D. R. Speer. 1976. Environmental Surveillance at Hanford for CY-1975 Data. BNWL-1980, Pacific Northwest Laboratory, Richland, Washington.
- Bosley, C. E., and G. F. Gately. 1981. Polychlorinated Biphenyls and Chlorinated Pesticides in Columbia River White Sturgeon (Acipenser transmontanus). U.S. Fish and Wildlife Service, Nordland, Washington.
- Bramson, P. E., and J. P. Corley. 1972. "Environmental Surveillance at Hanford for CY-1971," AEC Research and Development Report. BNWL-1683. Pacific Northwest Laboratory, Richland, Washington.
- Bramson, P. E., and J. P. Corley. 1973. "Environmental Surveillance at Hanford for CY-1972." AEC Research and Development Report. BNWL-1727, Pacific Northwest Laboratory, Richland, Washington.
- Brown, C. J. D. 1970. Fishes of Montana. Big Sky Books. Montana State University, Bozeman, Montana.
- Chatters, J. C. 1987. "Shell of *Margaritifera margaritifera falcata* as a Source of Paleoenvironmental and Cultural Data." In Archaeological Investigations at Lake Britton, California. Appendix F. Pacific Gas and Electric Company, San Francisco, California.
- Comfort, A. 1957. "The Duration of Life in Mollusca." Proc. Malacol. Soc. London. 32:219-241.
- Corley, J. P. 1970. Environmental Surveillance at Hanford For CY-1970. BNWL-1669, Pacific Northwest Laboratory, Richland, Washington.
- Cushing, C. E., D. G. Watson, A. J. Scott, and J. M. Gurtisen. 1980. Decline of Radionuclides in Columbia River Biota. PNL-3269, Pacific Northwest Laboratory, Richland, Washington.
- Cushing, C. E., W. H. Rickard, and D. G. Watson. 1988. Radionuclide Accumulation by Aquatic Biota Exposed to Contaminated Water Before and After its Passage Through the Ground. NUREG/CR-5047, PNL-5590, U.S. Nuclear Regulatory Commission, Washington, D.C.
- Dauble, D. D. 1978. "Comparative Ecology of Two Sympatric Catostomids, Catostomus macrocheilus and Catostomus columbianus in the Middle Columbia River." Masters Thesis, Washington State University, Pullman, Washington.
- Dauble, D. D. 1987. "Life History and Ecology of the Largescale Sucker (Catostomus macrocheilus) in the Columbia River." American Mid. Natural. 116:356-367.
- Essig, T. H., and J. K. Soldat, eds. 1967. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1966. BNWL-439, Pacific Northwest Laboratory, Richland, Washington.
- Fix, J. J. 1975. Environmental Surveillance at Hanford for CY-1974. BNWL-1910, Pacific Northwest Laboratory, Richland, Washington.

- Fruchter, J. S., C. E. Cowan, D. E. Robertson, D. C. Girvin, E. A. Jenne, A. P. Toste, and K. H. Abel. 1984. Radionuclide Migration in Groundwater. NUREG/CR-3712, PNL-5040. U.S. Nuclear Regulatory Commission, Washington, D.C.
- Galbreath, J. 1979. "Columbia River Colossus--The White Sturgeon." Oregon Wildl. 34:3-8.
- Gray, R. H., and D. D. Dauble. 1977. "Checklist and Relative Abundance of Fish Species from the Hanford Reach of the Columbia River." Northwest Sci. 51:208-215.
- Hauschild, W. L., G. R. Dempster, Jr., and H. H. Stevens, Jr. 1975. Distribution of Radionuclides in the Columbia River Streambed, Hanford Reservation to Longview, Washington. Geological Survey Professional Paper 433-0, U.S. Government Printing Office, Washington, D.C.
- Hauschild, W. L., H. H. Stevens, Jr., J. L. Nelson, and G. R. Dempster, Jr. 1971. Radionuclides in Transport in the Columbia River from Pasco to Vancouver, Washington. Open-file report, U.S. Department of the Interior Geological Survey, Portland, Oregon.
- Haynes, J. M. 1978. Movement and Habitat Studies of Chinook Salmon and White Sturgeon. PNL-2471, Pacific Northwest Laboratory, Richland, Washington.
- Healy, J. W., B. V. Anderson, H. V. Clukey, and J. K. Soldat. 1958. "Radiation Exposure to People in the Environs of a Major Production Atomic Energy Plant." In Proceedings of the Second United Nations International Conf. on the Peaceful Uses of Atomic Energy, pp. 309-318, Geneva.
- Junkins, R. L., E. C. Watson, I. C. Nelson, and R. C. Henle. 1960. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1959. HW-64371, Hanford Atomic Products Operation, General Electric, Richland, Washington.
- Malm, G. W. 1981. White Sturgeon (Acipenser transmontanus) Population Characteristics in the Bonneville Reservoir of the Columbia River (1976-1978). U.S. Fish and Wildlife Service, Vancouver, Washington.
- Mellinger, P. J. 1972. The Comparative Metabolism of Cadmium, Mercury, and Zinc as Environmental Contaminants in the Freshwater Mussel, Margaritifera margaritifera. Dissertation, Oregon State University, Corvallis, Oregon.
- McCrimmon, H. R. 1968. "Carp in Canada." Fish. Res. Bd. Canada Bull. 165:93.
- Napier, B. S., R. A. Peloquin, D. L. Strenge, and J. V. Ramsdell. 1988. GENII-The Hanford Environmental Radiation Dosimetry Software System. Volume 3: Code Maintenance Manual. PNL-6584 Vol. 3. Pacific Northwest Laboratory, Richland, Washington.
- Nelson, I. C., ed. 1961. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1960. HW-68435, Hanford Atomic Products Operation, General Electric, Richland, Washington.
- Nees, W. L., and J. P. Corley. 1974. Environmental Surveillance at Hanford for CY-1973. BNWL-1811, Pacific Northwest Laboratory, Richland, Washington.
- Nigro, A., R. C. Beamesderfer, J. C. Elliot, M. P. Faler, L. M. Miller, B. L. Uremovich, and D. L. Ward. 1985. Abundance and Distribution of Walleye Northern Squawfish, and Smallmouth Bass in John Day Reservoir, 1985. Annual Progress Report, Project No. 82-12, Oregon Department of Fish and Wildlife, Portland, Oregon.

- Page, T. L., D. D. Dauble, and D. A. Neitzel. 1982. Columbia River Aquatic Ecological Studies Near the Skagit/Hanford Nuclear Project. Prepared for Northwest Energy Services Company by Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Perkins, R. W., J. L. Nelson, and W. L. Hauschild. 1966. "Behavior and Transport of Radionuclides in the Columbia River Between Hanford and Vancouver, Washington." Limnol. Oceanogr. 11:235-248.
- Poston, T.M. and D.C. Klopfer. 1986. A Literature Review of the Concentration Ratios of Selected Radionuclides in Freshwater and Marine Fish. PNL-5484. Pacific Northwest Laboratory, Richland, Washington.
- Semakula, S. N., and P. A. Larkin. 1968. "Age, Growth, Food, and Yield of the White Sturgeon (*Acipenser transmontanus*) of the Fraser River, British Columbia." J. Fish. Res. Bd. Canada. 25:2589-2602.
- Silker, W. B. 1964. "Variation in Elemental Concentrations in the Columbia River." Limnol. Oceanogr. 9:540-549.
- Soldat, J. K., and T. H. Essig, eds. 1966. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1965. BNWL-316, Pacific Northwest Laboratory, Richland, Washington.
- Vanderploeg, H. A., D. C. Parzyck, W. H. Wilcox, J. R. Kerchner, and S. V. Kaye. 1975. Bioaccumulation Factors for Radionuclides in Freshwater Biota. ORNL-5002, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Vannote, R. L., and G. W. Minshall. 1982. "Fluvial Processes and Local Lighology Controlling Abundance, Structure, and Composition of Mussel Beds." Proceedings of the National Academy of Sciences, U.S.A. 79:4103-4107.
- Watson, D. G., C. E. Cushing, C. C. Coutant, and W. L. Templeton. 1970. Radioecological Studies on the Columbia River. BNWL-1377, Pacific Northwest Laboratory. Richland, Washington.
- Wilson, R. H., ed. 1963. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1962. HW-76526, Hanford Atomic Products Operation, General Electric, Richland, Washington.
- Wilson, C. B., ed. 1970. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1968. BNWL-1341, Pacific Northwest Laboratory, Richland, Washington.
- Woolridge, C. B., ed. 1969. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1967. BNWL-983, Pacific Northwest Laboratory, Richland, Washington.
- Woolridge, C. B., ed. 1962. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1961. HW-71999, Hanford Atomic Products Operation, General Electric, Richland, Washington.
- Woolridge, C. B., ed. 1964. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1963. HW-80991, Hanford Atomic Products Operation, General Electric, Richland, Washington.
- Woolridge, C. B., ed. 1965. "Evaluation of Radiological Conditions in the Vicinity of Hanford For 1964." AEC Research and Development Report. BNWL-90, Pacific Northwest Laboratory Richland, Washington,.
- Woolridge, C. B., ed., and T.H. Essig. 1970. Evaluation of Radiological Conditions in the Vicinity of Hanford For 1969. BNWL-1505, Pacific Northwest Laboratory, Richland, Washington.

Woolridge, C. B., ed. 1973. Environmental Surveillance at Hanford for CY-1972. BNWL-1727, Pacific Northwest Laboratory Richland, Washington.

DISTRIBUTION

No. of
Copies

No. of
Copies

OFFSITE

34 Pacific Northwest Laboratory

10 Office of Scientific and
Technical Information

L. L. Cadwell
D. D. Dauble (10)
R. L. Dirkes
J. W. Falco
R. H. Gray
H. A. Haerer
P. C. Hays
R. E. Jaquish
P. J. Mellinger
R. L. Newell
G. P. O'Connor
T. L. Page
T. M. Poston
K. R. Price
W. R. Rickard
R. G. Schreckhise
R. L. Skaggs
J. K. Soldat
R. K. Woodruff
Publishing Coordination
Technical Report Files (5)

ONSITE

DOE Richland Operations Office

R. F. Brich
E.C. Norman/D. L. Hoff
M. W. Tiernan (5)