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C. E. CUSHING JR.
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Radioactive Materials in Aquatic and Terrestrial Organisms Exposed to Reactor Effluent Water

By J. J. Davis, R. W. Perkins, R. F. Palmer, W. C. Hanson and J. F. Cline*

Water is commonly used as a coolant for nuclear reactors. Some are cooled with water which is pumped directly through the reactor. Others use the water in heat exchangers to control the temperature of a secondary coolant which is contained in a closed system that passes through the reactor. Water which has been forced through a reactor contains small amounts of many different radioisotopes from neutron-activation of dissolved minerals during normal operations. Fission elements may be produced if uranium is present in the water. Additional amounts of fission products may be present if ruptures occur in the coverings of fuel elements. Water discharged from the second type of cooling system does not contain radioactive materials except when leaks occur within the heat exchanger. Direct water cooling systems are used for reactors at the Hanford Atomics Products Operation. The Columbia River is the water source for these reactors and it is also the receptor of the reactor effluent.

Recently developed analytical techniques, based on gamma-ray and coincidence gamma-ray spectrometric methods of analyses[1] were used to qualitatively and quantitatively measure the radioisotopes in Columbia River water and in organisms which are exposed to it. These methods permitted the precise, sensitive, and rapid measurement of complex mixtures of radioisotopes without chemical separation in biotic samples. It was thus possible to measure the uptake and distribution of trace amounts of short-lived and long-lived radioisotopes in many organisms.

RADIOISOTOPES IN COLUMBIA RIVER WATER

More than 60 different radioisotopes have been identified in the effluent from Hanford reactors. Many of these isotopes have very short half-lives and are not detectable in the effluent at the time it is discharged into the river. Others which are initially scarce in the effluent are not detectable in the river water because of the tremendous dilution. Table 1 shows the concentrations of dominant radioisotopes in water collected during an average river flow period at a site known as Dot Island Station which is about 1.6 kilometers below the outfall of the reactor located farthest downstream.

Table 1. Radioisotopes in Columbia River Water[a]

Isotope	*Half-life*	*μc/ml*
Na^{24}	15.06 hours	8.6×10^{-6}
P^{32}	14.2 days	2.4×10^{-7}
Cr^{51}	27.8 days	2.0×10^{-6}
Mn^{56}	2.58 hours	7.4×10^{-6}
Cu^{64}	12.8 hours	1.7×10^{-5}
Zn^{65}	245 days	8.9×10^{-8}
As^{76}	26.8 hours	1.9×10^{-6}
Np^{239}	2.3 days	9.7×10^{-6}

[a] Collected at Dot Island Station, September 1957. Filtered through millipore filter. Raw water turbidity 4.0 ppm.

ACCUMULATION OF ISOTOPES BY INGESTION OF EFFLUENT

The Columbia River is a source of domestic water for several towns and cities located downstream from the Hanford reservation. In all cases the water is processed through standard water treatment plants before it is available to the public. The urban water pumping plant which is closest to the Hanford reservation is 61.6 kilometers downstream from the outfall of a reactor. The flow time between the outfall and the pumping plant varies during the year from 8 to 30 hours. Much of the radioactive material which is discharged into the river is lost by radioactive decay during this time.

The potential hazards from drinking Columbia River water that contains trace amounts of radioisotopes have been determined both by theoretical evaluation (using the standards established by the International Commission on Radiological Protection, ICRP) and by laboratory experiments. The uptake and retention of the longer-lived radioisotopes in reactor effluent were studied with rats which were maintained with concentrated effluent water as their sole source of drinking water for periods up to one year. The average amounts of the radioisotopes measured in the bone and combined soft tissues of these animals, and estimates of the amounts of isotopes consumed by the animals, are shown in Table 2.

As shown in Table 2, the amount of each isotope accumulated was only a small fraction of the maximum permissible body concentration, even though the consumed water contained concentrations of the isotopes that were thousands of times greater than the Columbia River water. The highest fraction of MPC

* Biology Operation, Hanford Laboratories, General Electric Company, Richland, Washington.

Table 2. Accumulation of Radioisotopes from Drinking Water by Rats

Isotope	*Total amount consumed* (μc)	*Concentration in soft tissues*[a] ($\mu c/g$)	*Concentration in bone* ($\mu c/g$)	*Fraction of MPC for bone*[b]
P^{32}	1.2	1.6×10^{-5}	8.5×10^{-5}	0.06
Sr^{89}	5.2×10^{-2}		1.9×10^{-6}	0.01
Sr^{90}	2.2×10^{-3}		8×10^{-7}	0.008
$Ba^{140}La^{140}$	$< 2.2 \times 10^{-1}$	10^{-7}	2.5×10^{-6}	0.02
Cs^{137}		3.5×10^{-6}	4×10^{-6}	
Zn^{65}	2.1[c]	1.1×10^{-4}	6.2×10^{-4}	0.07
Rare earths	4.4	7×10^{-7}	1.4×10^{-5}	0.13[d]

[a] Pelt and gastrointestinal tract not included.
[b] Maximum Permissible Concentrations (MPC) as established by ICRP for occupational exposure.
[c] Animals maintained on effluent for 347 days; other isotopes measured from animals which were maintained on same kind of water for 368 days.
[d] Total rare earth component assumed to be Ce^{144}–Pr^{144} for purposes of calculation.

reported for the rare earth component is undoubtedly an exaggeration. This component was assumed to consist entirely of Ce^{144}–Pr^{144} because its actual isotopic composition was not known. This pair of radioisotopes which were chosen for calculations would produce a higher build up in the bone than any other combination of rare earth isotopes.

ACCUMULATION OF ISOTOPES BY TERRESTRIAL PLANTS

Of the thousands of square kilometers of land that are irrigated with water from the Columbia River, only a few receive water which is drawn from the river downstream from the Hanford reservation. The closest irrigation project pumps water from a site 46 kilometers downstream from the Hanford reactors.

Zinc-65 was found in all farm produce that was sampled from this land and small amounts of Cr^{51} and Sc^{46} were found in a few samples of vegetation. Examples of the concentrations of Zn^{65} that were found are shown in Table 3.

The accumulation of Zn^{65} is undoubtedly associated with its long half-life and to the natural zinc deficiency of the soil in which the crops were grown. The comparatively high concentration of contaminants in the pasture grass was probably related to the manner of irrigation. The pasture land was watered by routine flooding which submerged the foliage and the garden crops were irrigated by rills. The concentration of radiozinc in milk is of particular interest. During mid winter, when the cow's diet did not include any foodstuffs that were grown on land of this irrigation project, the concentration of Zn^{65} in milk was about 10% of the value in Table 3. The concentrations reported for beef steak and fat samples were from an animal which was butchered in late winter. The animal had been fed on radiozinc-free foodstuffs for four months prior to the time of butchering.

Table 3. Zinc–65 in Farm Produce[a]

Sample	$\mu c/g$	*Concentration factor* ($\mu c/g$ *wet wt of produce*) / ($\mu c/ml$ *of water*)
Pasture grass	8.29×10^{-5}	440
Beef steak	5.23×10^{-6}	28
Beef fat	1.48×10^{-6}	7.9
Milk	4.88×10^{-6}	26
Blackeyed peas	5.49×10^{-7}	2.9
Tomatoes	4.58×10^{-7}	2.4
Okra	3.94×10^{-7}	2.1
String beans	2.90×10^{-7}	1.5
Corn	1.56×10^{-7}	0.83
Grapes	8.9×10^{-8}	0.47
Irrigation water[b]	1.88×10^{-7}	

[a] Samples collected during summer of 1957.
[b] Sample not filtered.

The uptake and translocation of radioisotopes by bean plants from undiluted reactor effluent water was studied by laboratory experiments. Culturing methods and nutrients used were as described by Rediske and Selders.[2] Table 4 summarizes data for 8 of the 14 radioisotopes that were observed in plants which were grown in effluent solution for 14 days.

Crops of barley have been grown on the same plots irrigated with undiluted reactor effluent for six years. Grain harvested from the plots was used to replant the same plot the following year. There have been no effects from the radioisotopes in the effluent upon the productivity of the crops or the germination of the grain which was produced.

ACCUMULATION OF RADIOISOTOPES BY AQUATIC ORGANISMS

All aquatic organisms which inhabit the Columbia River downstream from the Hanford reactors contain concentrations of radioisotopes. Differences in the concentrations of total beta emitters and of a few specific isotopes in different species of river organisms have been reported by Foster and Davis[3] and Davis and Foster.[4] The kinds and amounts of radioisotopes recently have been measured by better methods in many different kinds of aquatic organisms. The radioisotopic concentrations found in a few representative type of samples which were collected at Dot Island are given in Table 5.

The tremendous bioaccumulation of different radioisotopes in organisms is demonstrated by comparison of the values in Tables 1 and 5. Many of the isotopes found in the organisms were not measured in the river water because they occurred in amounts below detectable limits in the two-liter samples of water that were tested.

Comparison of the isotopic content of the various

Table 4. Radioisotopes in Bean Plants Cultured for 14 Days in Reactor Effluent Containing Plant Nutrients

Isotope	*Half-life in days*	*Ave. conc.*[a] *in solution (μc/ml)*	*Per cent of*[b] *added isotope in leaves*	*Per cent of*[b] *added isotope in stems*	*Per cent of*[b] *added isotope in roots*
P^{32}	14.2	1.26×10^{-5}	2.4	1.1	24.3
Sc^{46}	85		0.11	0.05	67.4
Cr^{51}	27.8	3.33×10^{-4}	0.11	0.08	29.8
Mn^{54}	310	4.6×10^{-7}		1.24	84.2
Co^{60}	1934.5	2.0×10^{-8}	1.0	0.26	19.7
Zn^{65}	245	1.28×10^{-5}	3.3	1.5	66.2
Ba^{140}	12.8	8.8×10^{-5}	1.7	0.61	55.3
Np^{239}	2.3	4.29×10^{-5}	0.13	0.01	33.1

[a] Each experimental group was grown in six liters of effluent solution. Fresh solution was supplied three times during the experiment.
[b] The average fresh weight total yields for leaves, stems and roots were 60, 29 and 75 grams respectively.

Table 5. Radioisotopes in River Organisms Collected near Hanford Reactor[a]

Isotope	*Physical half-life*	*μc/g wet weight*					
		Green[b] *algae*	*Sponge*[c]	*Insect*[d] *larvae*	*Snail*[e]	*Crayfish*[f]	*Minnows*[g]
Na^{24}	15.06 hours	5.74×10^{-4}	7.28×10^{-4}	7.09×10^{-4}	1.77×10^{-3}	1.95×10^{-3}	1.13×10^{-3}
P^{32}	14.2 days	-6.60×10^{-2}	4.46×10^{-2}	2.43×10^{-2}	1.44×10^{-2}	3.04×10^{-2}	2.40×10^{-2}
Sc^{46}	85 days	1.73×10^{-3}	9.47×10^{-5}	7.06×10^{-5}	2.88×10^{-5}		7.06×10^{-7}
Cr^{51}	27.8 days	7.90×10^{-2}	4.58×10^{-3}	6.00×10^{-2}	5.07×10^{-3}	4.82×10^{-4}	3.72×10^{-4}
Mn^{54}	310 days	1.03×10^{-3}		7.91×10^{-5}			
Mn^{56}	2.57 hours	8.22×10^{-2}	2.38×10^{-2}				1.87×10^{-4}
Fe^{59}	45.1 days	1.64×10^{-3}					
Co^{60}	5.3 years	1.55×10^{-4}	1.16×10^{-5}	1.72×10^{-6}	1.77×10^{-5}		
Cu^{64}	12.8 hours	1.13×10^{-1}	1.66×10^{-2}	7.10×10^{-3}	9.60×10^{-3}	5.14×10^{-4}	1.56×10^{-4}
Zn^{65}	245 days	-1.23×10^{-2}	1.46×10^{-2}	1.98×10^{-3}	1.53×10^{-3}	3.65×10^{-4}	7.62×10^{-4}
As^{76}	26.8 days	6.85×10^{-3}	6.32×10^{-3}	5.17×10^{-4}	1.00×10^{-4}	6.48×10^{-5}	1.73×10^{-4}
Zr^{95}–Nb^{95}	65.35 days	1.79×10^{-3}		6.63×10^{-5}			
Ru^{103}	39.8 days	1.22×10^{-3}		1.06×10^{-4}			
Ba^{140}	12.8 days	9.01×10^{-4}		4.22×10^{-5}	2.90×10^{-5}	1.88×10^{-5}	
La^{140}	40.0 hours	3.27×10^{-3}	1.23×10^{-3}	3.47×10^{-4}			
Ce^{141}	33.1 days	1.86×10^{-3}					
Np^{239}	2.3 days	2.69×10^{-3}	4.01×10^{-4}	3.11×10^{-4}			
Sr^{90}	28.0 years	2.07×10^{-4}	5.74×10^{-5}	1.21×10^{-5}			1.60×10^{-5}

[a] Organisms collected at Dot Island Station, August 1957.
[b] *Stigeoclonium lubricum* (Dillwyn) Kuetzing.
[c] *Spongilla lacustris* (Linnaeus).
[d] Trichoptera: *Hydropsyche cockerelli* Banks.
[e] *Stagnicola nuttailiana* (Lea). Shell not included.
[f] *Astacus trowbridgii* Stimpson.
[g] *Richardsonius balteatus* (Richardson). Contents of alimentary canal not included.

Table 6. Mineral Composition of Columbia River Organisms[a]

Element	*ppm of ash*					
	Green algae	*Sponge*	*Insect larvae*	*Snail soft tissues*	*Crayfish*	*Minnows*
Na	10,000	2,000	36,000	10,000	10,000	10,000
P	2,000	10,000	20,000	10,000	10,000	24,000
Cr	5	10	20	20	5	2
Mn	50	100	500	20	50	2
Fe	50,000	10,000–20,000	10,000–20,000	10,000	200	20
Co	<5	<5	<5	<5	<5	<5
Cu	100	1,000	500	200	500	100
Zn	<50	<50	<50	<50	<50	<50
Rare earths	<20	<20	<20	<20	<20	<20
Ru	<20	<20	<20	<20	<20	<20
Ba	500	20	200	20	20	<5
Ce	<50	<50	<50	<50	<50	<50

[a] Species of organisms same as listed for Table 5.

organisms from the same habitat indicated the effects of food habits, physiology and perhaps morphology upon the accumulation of radioactive contaminants by different species of plants and animals.

The algae, which derive their nutrients directly from the water, contained the most kinds and largest amounts of most isotopes that were accumulated by biological organisms. A considerable amount of the radioactive contamination which was measured from sessile algae was undoubtedly from materials adsorbed

on their extensive surface areas. Similar concentrations were accumulated by phytoplankton. Extensive surface area probably also accounts, in part, for the large amounts of Mn^{56}, As^{76}, La^{140}, and Np^{239} that were associated with the sponge.

The effects of biological demand are indicated by the efficiency of the transfer of Na^{24} and P^{32} to the higher animals. Large amounts of both of these elements are metabolized by the organisms as is indicated by their mineral composition in Table 6.

Stable iron is abundant in all of the organisms. The radioactive isotope, Fe^{59}, which has a relatively long half-life occurred in substantial amounts in plankton and sessile algae. It seldom was found, however, in measurable amounts in organisms from higher trophic levels in the food web. Zinc is biologically essential as a trace element. Although its abundance in the organisms is much less than phosphorus, sodium or iron, the Zn^{65} was readily transferred through the food web and occurred in relatively large concentrations in almost all organisms sampled.

The radioisotopic concentrations of different tissues from adult Catostomid fish collected at the Dot Island Station during the month when highest contaminations occurred are compared in Table 7. The concentrations of different radioisotopes in specific tissues of the fish are associated with the stable isotopic composition and physiological functions of the respective tissues. Radioactive phosphorus was the dominant radioisotope in all tissues. It was most abundant in calcareous tissues, including scales and bone, which are high in phosphates and it is lowest in muscle and fat. Significant concentrations of Zn^{65} occurred in all tissues, with values for the retina and iris of the eye three or more times greater than in any other tissue. The avidity of Zn^{65} by these parts of the eye are undoubtedly associated with the high stable zinc content of these tissues.[5] The radiozinc content of the lens was comparable to its concentration in fat. The gut contained high concentrations of a melange of isotopes as would be expected from its digestive and absorptive functions.

The importance of the food web upon the accumulation of radioisotopes by organisms is demonstrated in Fig. 1 which compares representative tissues from adult fish of different consumer or trophic levels. Inasmuch as all of these fish were collected at Dot Island Station at the same time, the differences in their radioactive contamination must result from their food habits and physiology. The suckers are the most radioactive species of large fish in the Columbia River. Their position in the food web is the second trophic level because they feed directly upon sessile algae. Whitefish represent the third trophic level. Their food consists primarily of immature herbivorous aquatic insects. Carnivorous squawfish belong to the fourth trophic level. Fish and crayfish are their main food items. The sturgeon is mainly a scavenger upon bottom debris.

The seasonal cycles of gross beta emitters in Columbia River fish have been discussed by Foster and

Table 7. Radioisotopes in Tissues of Fish from Hanford[a]

Tissue	Na^{24}	P^{32}	Sc^{46}	Cr^{51}	Mn^{54}	Fe^{59}	Co^{60}	Cu^{64}	Zn^{65}	Cs^{137}	Sr^{89}, Sr^{90}
	$\mu c/g$ wet weight										
Bone[b]	2.06×10^{-3}	4.64×10^{-3}			3.35×10^{-5}			2.28×10^{-4}	3.13×10^{-4}		7.7×10^{-6}
Scales[b]	1.16×10^{-3}	2.88×10^{-3}		4.37×10^{-4}	2.08×10^{-5}		1.46×10^{-8}	6.96×10^{-5}	7.74×10^{-4}		2.2×10^{-6}
Spleen	1.25×10^{-3}	1.57×10^{-3}	2.02×10^{-6}	3.64×10^{-4}			1.18×10^{-5}	2.79×10^{-4}	4.37×10^{-4}	—	—
Liver	2.85×10^{-4}	1.56×10^{-3}		4.02×10^{-4}	9.29×10^{-6}		1.71×10^{-8}	4.36×10^{-4}	2.79×10^{-4}	—	—
Kidney	9.47×10^{-4}	1.24×10^{-3}		3.75×10^{-4}	1.06×10^{-5}		6.70×10^{-8}	3.73×10^{-4}	4.33×10^{-4}	—	—
Gut	3.17×10^{-4}	7.34×10^{-3}	2.02×10^{-6}	9.54×10^{-6}	1.43×10^{-5}	2.59×10^{-5}	3.91×10^{-8}	1.13×10^{-3}	2.99×10^{-4}	—	—
Heart	4.02×10^{-4}	7.18×10^{-3}		1.72×10^{-4}				8.39×10^{-5}	1.33×10^{-4}	—	—
Ovaries	5.82×10^{-4}	6.81×10^{-3}						1.45×10^{-4}	4.39×10^{-4}	—	—
Retina	1.37×10^{-3}	5.97×10^{-3}						5.75×10^{-4}	2.81×10^{-3}	—	—
Blood	1.61×10^{-3}	5.62×10^{-3}	2.32×10^{-6}	5.39×10^{-4}					2.87×10^{-4}	—	—
Brain	1.06×10^{-3}	5.46×10^{-3}		1.01×10^{-4}				5.0×10^{-5}	8.99×10^{-5}	—	—
Muscle	2.59×10^{-4}	2.82×10^{-3}	3.02×10^{-7}					5.54×10^{-5}	2.81×10^{-3}	9.0×10^{-7}	—
Skin	3.31×10^{-4}	2.80×10^{-3}	2.96×10^{-7}	6.28×10^{-5}				1.12×10^{-4}	8.51×10^{-4}	—	—
Fat	2.55×10^{-4}	1.79×10^{-3}	1.29×10^{-6}	1.86×10^{-4}	3.36×10^{-6}		2.65×10^{-7}	1.93×10^{-4}	8.50×10^{-5}	—	—
Lens		1.79×10^{-3}							8.26×10^{-5}	—	—

[a] Suckers, *Catostomus macrocheilus* Girard, collected at Dot Island Station, August 1957

[b] Bone and scales only tissue analyzed for Sr^{89} and Sr^{90}.

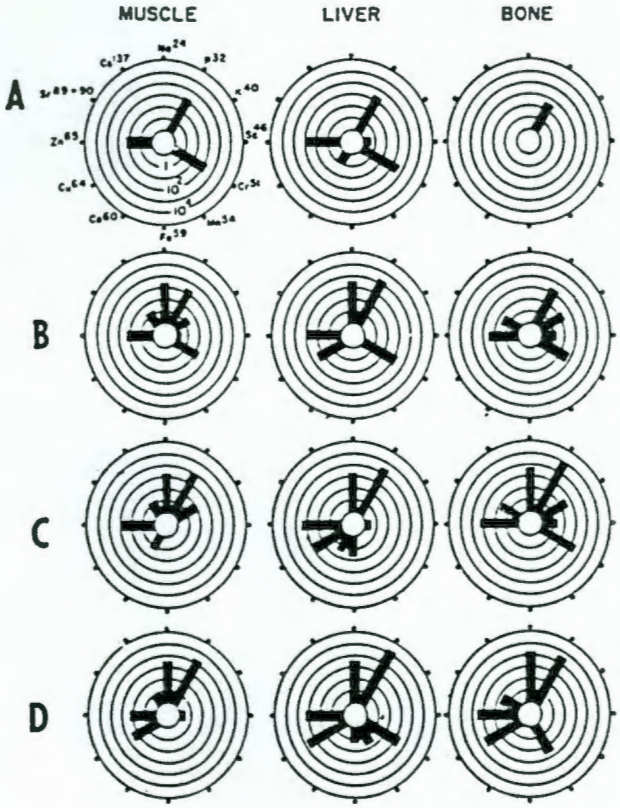

Figure 1. Radioisotopes in different kinds of fish from the Columbia River

A. Sturgeon, *Acipenser transmontanus* (Richardson)
B. Squawfish, *Ptychocheilus oregonensis* (Richardson)
C. Whitefish, *Prosopium williansoni* (Girard)
D. Sucker, *Catostonus macrocheilus* (Girard)

Davis.[3] In brief, the contamination levels coincide with the water temperatures which, in turn, control metabolic rate. Comparison of data from fish collected at the Dot Island Station indicated that the concentrations of radiosodium and radiophosphorus decreased greatly during the winter but that the concentrations of radiozinc which has a long radioactive half-life remained relatively constant throughout the year.

ACCUMULATION OF RADIOISOTOPES BY WATERFOWL

The radioisotopic content in waterfowl which frequent the Columbia River below the Hanford reactors depends upon the food habits of the birds and the geographical areas of the river in which they feed. Contamination levels in tissues from a species of duck which usually possessed higher contamination than other associated species are shown in Table 8. This species feeds almost exclusively on aquatic insect larvae and other invertebrates from the river bottom of the main river channel. The radioisotopic content of the various tissues are generally comparable to those of fish which consume similar food organisms with the exception of the proportionally much higher concentration of P^{32} in flesh of the ducks. The samples of duck flesh reported were pectoral muscles which contain a high concentration and a high turnover rate of phosphorus. The abundance of phosphorus is associated with the large energy requirements of these muscles that provide the muscular work for flight. Hanson and Kornberg[6] reported that the greatest concentrations of P^{32} in juvenile waterfowl occurred in the pancreas and skeleton but that in adult birds comparable concentrations occurred in the muscle. The flesh sampled for these studies was also pectoral muscle. Leg muscle of ducks contained about 67% as much P^{32} as the pectoral muscles. The reason for the comparatively lower concentration of Zn^{65} in the iris and retina of ducks than in fish is unknown. The relatively high concentration of radiophosphorus in the pancreas is comparable to that in pyloric caeca which contain functionally similar tissue in the fish.

CONCLUSIONS

Of the many radioisotopes that have been measured in Columbia River water, none were present in quantities near the recommended maximum limits for drinking water by the International Commission on Radiological Protection. Studies in which rats were maintained on reactor effluent that contained concentrations of radioisotopes several thousand times the concentrations in the river water indicated that the danger of assimilating harmful levels of radioisotopes

Table 8. Radioisotopes in Tissues of Ducks[a]

Tissue	μc/g wet weight						
	Na^{24}	P^{32}	Sc^{46}	Cr^{51}	Co^{60}	Cu^{64}	Zn^{65}
Bone	3.74×10^{-5}	1.36×10^{-2}					
Pancreas	1.03×10^{-4}	7.27×10^{-3}			1.93×10^{-6}		5.16×10^{-4}
Liver	4.19×10^{-5}	7.20×10^{-3}	2.18×10^{-6}		1.50×10^{-6}		5.63×10^{-4}
Spleen		6.32×10^{-3}			1.87×10^{-6}		1.92×10^{-4}
Testes		5.46×10^{-3}					1.70×10^{-4}
Flesh	2.31×10^{-5}	4.41×10^{-3}					6.84×10^{-5}
Kidney	8.44×10^{-5}	3.80×10^{-3}	1.03×10^{-6}		1.69×10^{-6}	1.04×10^{-4}	1.90×10^{-4}
Heart	7.01×10^{-5}	2.97×10^{-3}			2.66×10^{-7}		1.32×10^{-4}
Gut		2.56×10^{-3}	1.25×10^{-6}	1.85×10^{-4}	2.82×10^{-6}	2.01×10^{-4}	1.20×10^{-4}
Lungs		2.28×10^{-3}		3.00×10^{-5}	1.27×10^{-6}	8.06×10^{-5}	7.43×10^{-5}
Blood	2.51×10^{-5}	1.74×10^{-3}	1.01×10^{-6}	5.39×10^{-5}	1.61×10^{-6}	3.94×10^{-5}	4.17×10^{-5}
Fat		9.28×10^{-4}					2.83×10^{-5}
Feathers		7.65×10^{-5}	4.47×10^{-6}				

[a] Barrow Goldeneye, *Bucephala islandica* (Gmelin). Collected at Dot Island Station, November 1957.

by drinking Columbia River water was negligible. Significant quantities of various isotopes were concentrated, however, in the bodies of most organisms which had access to the contaminants via the natural food web. It is apparent that food organisms of man could accumulate hazardous levels of certain radioisotopes from water which contained concentrations of the contaminants that were well within the permissible limits for drinking water. Representative organisms are routinely sampled from natural environments which are exposed to dilution of effluent from the Hanford reactors to ascertain that the amounts of radioactive contamination are well below hazardous levels.

The data presented emphasizes the value of the need for radioecological research to further elucidate and evaluate environmental relationships that pertain to radioactive waste disposal.

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