

---

## **Decline of Radionuclides in Columbia River Biota**

**C. E. Cushing  
D. G. Watson  
A. J. Scott  
J. M. Gurtisen**

---

**March 1980**

**Prepared for the U.S. Department of Energy  
under Contract EY-76-C-06-1830**

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





## NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express 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.

The views, opinions and conclusions contained in this report are those of the contractor and do not necessarily represent those of the United States Government or the United States Department of Energy.

PACIFIC NORTHWEST LABORATORY  
*operated by*  
BATTELLE  
*for the*  
UNITED STATES DEPARTMENT OF ENERGY  
*Under Contract EY-76-C-06-1830*

Printed in the United States of America  
Available from  
National Technical Information Service  
United States Department of Commerce  
5285 Port Royal Road  
Springfield, Virginia 22151

Price: Printed Copy \$ \_\_\_\_\*; Microfiche \$3.00

*Pages	NTIS Selling Price
001-025	\$4.00
026-050	\$4.50
051-075	\$5.25
076-100	\$6.00
101-125	\$6.50
126-150	\$7.25
151-175	\$8.00
176-200	\$9.00
201-225	\$9.25
226-250	\$9.50
251-275	\$10.75
276-300	\$11.00

3 3679 00054 4793

DECLINE OF RADIONUCLIDES IN  
COLUMBIA RIVER BIOTA

C. E. Cushing  
D. G. Watson  
A. J. Scott  
J. M. Gurtisen

March 1980

Prepared for  
the U.S. Department of Energy  
under Contract EY-76-C-06-1830

Pacific Northwest Laboratory  
Richland, Washington 99352

## ACKNOWLEDGMENTS

We would like to thank R. L. Burdette and others for their capable assistance in the field and laboratory. The work was performed under Contract No. EY-76-C-06-1830 between the Department of Energy and Pacific Northwest Laboratory, operated by Battelle Memorial Institute. Funding for publication was provided by the Radioecology of Nuclear Fuel Cycles Project supported by the Office of Health and Environmental Research, Department of Energy.

## ABSTRACT

In January 1971, the last of nine plutonium production reactors using direct discharge of once-through cooling waters into the Columbia River was closed. Sampling was initiated at three stations on the Columbia River to document the decline of the radionuclide body burdens in the biota of the Columbia River ecosystem.

Concentrations of  $^{46}\text{Sc}$  decreased to unmeasurable levels in most biota by spring of 1972; large fish averaged about 0.03 pCi/g dry weight (DW) and also had unmeasurable levels after one year. Body burdens of  $^{54}\text{Mn}$  in the lower trophic levels were essentially unmeasurable after one year. In suckers, body burdens declined to fairly constant levels of <1 pCi/g DW at White Bluffs, a value similar to those found at all times at McNary Reservoir and Bonneville Reservoir. Values in squawfish became unmeasurable in the last year of the study. Concentrations of  $^{60}\text{Co}$  in seston, periphyton, and invertebrates did not decrease to the degree that the other radionuclides did and this was related to the seepage of  $^{60}\text{Co}$  into the river from a disposal trench near the operating N Reactor. Levels of  $^{60}\text{Co}$  in suckers and squawfish showed some decreases, but obvious trends were not present. Zinc-65 was present in the biota in highest concentrations. Concentrations in seston and periphyton decreased rapidly and did not become unmeasurable until the spring of 1973. Zinc-65 in caddisfly larvae was not measurable by February 1973, but concentrations in McNary chironomids fluctuated between undetectable levels to 24 pCi/g DW and this was related to ingestion of contaminated sediments rather than larval body burdens. In suckers and squawfish,  $^{65}\text{Zn}$  decreased to fairly low, constant levels of 1 and 3 pCi/g DW, respectively. Levels of  $^{137}\text{Cs}$  were measurable initially in periphyton, but measurable levels in the lower trophic levels were mostly sporadic. Cesium-137 levels in suckers fluctuated markedly with no long-term decrease. In squawfish,  $^{137}\text{Cs}$  levels fluctuated at White Bluffs but were constant at McNary and Bonneville.

The data show that in a river-reservoir complex, the measurable body burden of fission-produced radionuclides decreased to essentially undetectable levels within 18 to 24 mo after cessation of discharge of once-through cooling water into the river. On the basis of data from the free-flowing station, we believe that this decrease would be even more rapid in an unimpounded river.

## CONTENTS

ACKNOWLEDGMENTS . . . . .	ii
ABSTRACT . . . . .	iii
FIGURES . . . . .	vi
TABLES . . . . .	vi
INTRODUCTION . . . . .	1
SAMPLING LOCATIONS . . . . .	3
METHODS AND MATERIALS . . . . .	5
BIOTA SAMPLED . . . . .	5
FIELD COLLECTION . . . . .	6
LABORATORY PROCESSING . . . . .	6
RESULTS AND DISCUSSION . . . . .	9
SCANDIUM-46 . . . . .	9
MANGANESE-54 . . . . .	9
COBALT-60 . . . . .	12
ZINC-65 . . . . .	14
CESIUM-137 . . . . .	17
CONCLUSIONS . . . . .	19
REFERENCES . . . . .	21

## FIGURES

1	Locations (Dots) of Sampling Stations . . . . .	3
2	Concentrations of $^{46}\text{Sc}$ in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Chironomids . . . . .	10
3	Concentrations of $^{54}\text{Mn}$ in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Suckers . . . . .	11
4	Concentrations of $^{60}\text{Co}$ in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Chironomids, d) Suckers, e) Squawfish . . . . .	13
5	Concentrations of $^{65}\text{Zn}$ in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Chironomids, d) Suckers, e) Squawfish . . . . .	15
6	Concentrations of $^{137}\text{Cs}$ in Selected Columbia River Biota; a) Periphyton, b) Suckers, c) Squawfish . . . . .	18

## TABLES

1	Biota Sampled, Sampling Periods, and Sampling Locations . . . . .	5
2	Radioactivity in Selected Columbia River Biota, 1966 and 1967 . . . . .	7

## INTRODUCTION

In 1944, the first plutonium production reactor on the Atomic Energy Commission's Hanford Reservation (now the Department of Energy's Hanford Site) began discharging radionuclides into the Columbia River via the cooling water effluents. In succeeding years, as many as nine reactors were introducing various amounts of radionuclides into the Columbia at one time. With the closure of the K East Reactor in January 1971, direct discharge of once-through cooling water into the Columbia ended. The only radioactivity presently reaching the river is from N Reactor coolant water which is discharged into a seepage trench. From this trench, small amounts of the coolant water reach the river through ground seepage (Robertson et al. 1973).

A large body of literature, abstracted by Becker (1973), has reported the results of radioecological studies of the Columbia River, the most thorough surveys being those of Robeck, Henderson and Palange (1954), Davis, Watson and Palmiter (1956), and Watson et al. (1970). Numerous studies report the accumulation and loss of radionuclides by biota, but the closure of the Hanford reactors provided a unique opportunity to study the pattern of decreasing radionuclide concentrations in the components of the natural river ecosystem over time. In the summer of 1966, a strike closed the Hanford reactors for about 40 days; the results of this shutdown upon radionuclide concentrations in the biota have been published by Watson et al. (1969).

Knowledge of the loss of radionuclides in biota is particularly important today. The number of nuclear power generating plants is increasing and more are projected. The data reported here provide information on the possible effects of the closure of once-through cooling systems on the radionuclide body burdens of the biota, thus ensuring a more complete evaluation of the environmental impact of siting nuclear reactors.

The purpose of this investigation was to determine the decline of the body burdens of radionuclides in the biota of the Columbia River ecosystem after shutdown of the Hanford reactors with once-through cooling systems.



## SAMPLING LOCATIONS

Three sites were sampled from July 1971 through June 1972: 1) immediately below the reactors at White Bluffs (River km 593), 2) in McNary Reservoir (Rkm 473) on the Washington side, and 3) in Bonneville Reservoir (Rkm 245). The Columbia River at White Bluffs is free-flowing, and the littoral and deeper bottom substrates are rounded rubble. The water level fluctuates about 2 m each day because of the operation of Priest Rapids Dam, about 45 km upstream. In backwater areas, the current lessens, and fine silts cover the rubble. Both the McNary and Bonneville stations are typical river-reservoir lentic habitats with little or no distinguishable current. Bottom substrate at McNary is fine silts, while at Bonneville, fine silts and coarse woody debris were present. The decrease in concentrations of radionuclides in biota at White Bluffs and Bonneville, together with the occurrence of most radionuclides in the Columbia River system in sediments behind McNary Dam, dictated eliminating the White Bluffs and Bonneville sites and expanding the sampling effort in McNary from July 1972 through June 1973. Figure 1 shows the sampling locations.

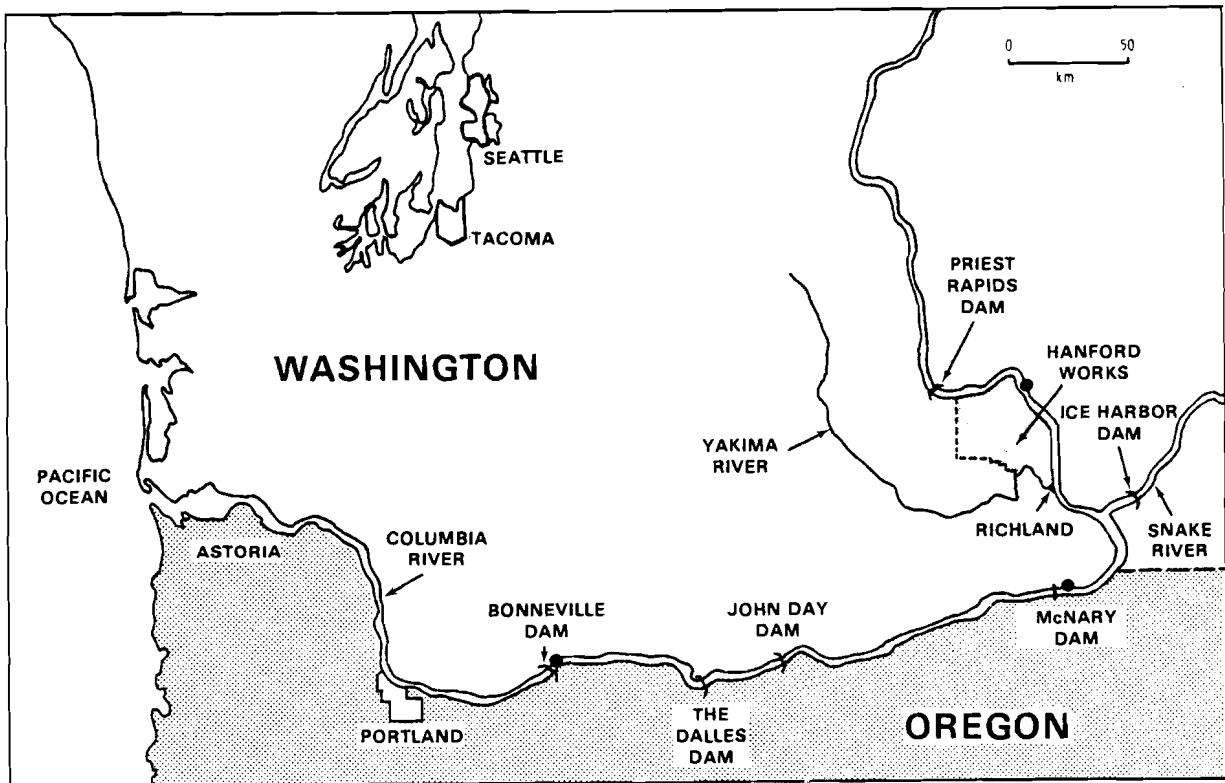


FIGURE 1. Locations (Dots) of Sampling Stations

## METHODS AND MATERIALS

### BIOTA SAMPLED

Initially, we sampled seston, periphyton, a dominant invertebrate species, an herbivorous fish species (sucker, Catostomus macrocheilus) and a carnivorous fish species (squawfish, Ptychocheilus oregonensis) at each site. We believed this selection would give us a representative picture of the various trophic levels and pathways; detailed sampling of all organisms at all sites was beyond the budgetary constraints of the program. This sampling regime was followed as closely as possible during the first year, although some organisms were not available during the entire year, e.g., caddisfly (Hydropsychidae) larvae at White Bluffs. As mentioned above, the sampling effort during the second year was shifted to McNary Reservoir and included additional organisms in the food web as determined from fish stomach content analysis. Table 1 lists the biota and their locations sampled during this study.

TABLE 1. Biota Sampled, Sampling Periods, and Sampling Locations

<u>Locations and Biota Sampled</u>	<u>July 1971</u>	<u>July 1972</u>	<u>July 1973</u>
White Bluffs			
Seston	-----		
Periphyton	-----		
Caddisfly larvae	-----		
Suckers	-----		
Squawfish	-----		
McNary Reservoir			
Seston	-----		
Periphyton	-----		
Chironomids	-----		
Suckers	-----		
Squawfish	-----		
Crayfish		-----	
Forage fish		-----	
Catfish		-----	-----
Mussels			-----
Bonneville Reservoir			
Seston	-----		
Periphyton	-----		
Suckers	-----		
Squawfish	-----		
Sturgeon	-----		

## FIELD COLLECTION

Seston was collected by towing a plankton net (Nitex<sup>®</sup>, 80  $\mu$ m mesh). No attempt was made to separate phytoplankton, zooplankton and detritus. Periphyton was collected mainly from artificial substrates because power production by the dams resulted in water level fluctuations such that accessible natural substrates were not always submerged. Occasional samples were collected from floats, buoys, and other substrates.

Caddisfly larvae at White Bluffs were collected by hand-picking from rocks in riffle areas. Chironomids (Chironomidae) in McNary were initially collected during the first 18 mo from sediments obtained with a 15-cm (6-in) Ekman dredge and washed through screens. Later, scuba divers collected 19-l (5-gal) buckets of sediments which were then washed through screens on shore. This technique was much more efficient and time-saving than using the Ekman dredge. Crayfish (Pacifasticus lenisculatus) and freshwater mussels (Anodonta nuttalliana) were collected individually from the bottom by scuba divers.

Suckers, squawfish, sturgeon (Acipenser transmontanus), and catfish (Ictalurus punctatus) were collected with 46-m long experimental gill nets (mesh size, 2.54 to 7.62 cm). Forage fish, mostly small squawfish, were seined from shallow areas near shore.

## LABORATORY PROCESSING

Samples were prepared for gamma-ray spectrometry by filling a counting container (10 cm in diameter by 2 cm thick) with an homogenized, ashed and weighed aliquot. If sufficient tissue was not present to fill the container, the entire sample was uniformly mixed with agar-agar to make up the difference. The gut contents of suckers, squawfish, sturgeon, and catfish were removed before processing, and only the soft tissues of the mussels were analyzed. Prior to ashing, samples were oven-dried at 60°C and weighed. The number of samples of each type counted varied from one to four, and in some cases for the larger fish, the single analysis was from a composite sample of up to 10 individual fish. Thus, since the majority of measurements presented in the following figures are from sample sizes of one or two, no data are presented on variation.

The samples were counted on an anticoincidence shielded, multidimensional gamma-ray spectrometer; details of the system are described by Wogman, Robertson and Perkins (1967). Data are presented for <sup>46</sup>Sc, <sup>54</sup>Mn, <sup>60</sup>Co, <sup>65</sup>Zn and <sup>137</sup>Cs as pCi/g DW and pCi/g DW  $\pm$  1 standard deviation.

Sampling for this study could not begin until June 1971, about 5 mo after closure of the last once-through cooling reactor. Thus, some unknown change

<sup>®</sup>Use of trade name does not imply Battelle endorsement.

in radionuclide concentration in the biota occurred before our sampling began. This, unfortunately, prevents the calculation of accurate rates of decline or effective half-lives from the time all reactors were operating. However, to provide an indication of the concentrations of these radionuclides in the biota while five reactors were in operation, the data in Table 2 are presented. These data were obtained from organisms collected just downstream from the White Bluffs area and can be compared with values presented in the text and figures. An additional factor to be considered is that between 1967, when the last sampling was done in the studies by Watson et al. (1970), and January 1971, when the last reactor was closed, the four other reactors were shut down sequentially, so that the unknown loss of radioactivity in the organisms in the interim was also probably of a "stair-step" nature. To further aid in assessing this phenomenon, see Watson et al. (1969) for information on immediate loss of radioactivity from Columbia River biota; these data were collected during the 1966-1967 sampling when a work stoppage closed all reactors for about 40 days.

TABLE 2. Radioactivity in Selected Columbia River Biota, 1966 and 1967 (Modified from Watson et al. 1970)

Organism	Number Sampled	pCi/g DW				
		<sup>46</sup> Sc	<sup>54</sup> Mn	<sup>60</sup> Co	<sup>65</sup> Zn	<sup>137</sup> Cs
Seston	45	4630 ± 6060	1030 ± 1270	52 ± 201 <sup>(a)</sup>	10720 ± 8290	23 ± 129 <sup>(a)</sup>
Periphyton	35	4350 ± 4950	1770 ± 1290	240 ± 480 <sup>(a)</sup>	8190 ± 5940	27 ± 53 <sup>(a)</sup>
Caddisfly larvae	39	580 ± 1190 <sup>(b)</sup>	745 ± 568	66 ± 104 <sup>(a)</sup>	3686 ± 2186	0
Suckers	5	0.06 ± 0.13 <sup>(c)</sup>	4.7 ± 1.5	1.4 ± 0.2	249 ± 62	2 ± 0.2
Squawfish	2	0.2 ± 0.3 <sup>(a)</sup>	0.7 ± 0.9 <sup>(a)</sup>	0.5 ± 0.6	141 ± 77	1.2 ± 0.6

(a) 50% or more of values below detection limits  
 (b) 23% of values below detection limits  
 (c) 20% of values below detection limits

## RESULTS AND DISCUSSION

It should be kept in mind that the decreasing concentrations of the radionuclides in the biota described below are a result of three coincident processes: 1) physical decay of the radionuclide, 2) biological turnover of the element by the organisms, and 3) decreasing availability in the food supply (in most cases). In the case of this study, the third item is a function of the first and second. Since the study period included 8.7, 2.5, and 3.0 half-lives for  $^{46}\text{Sc}$ ,  $^{54}\text{Mn}$ , and  $^{65}\text{Zn}$ , respectively, the decline due to physical decay for these isotopes is significant; that for  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  is negligible because of their long half-lives.

### SCANDIUM-46 (HALF-LIFE, 84 DAYS)

Concentrations of  $^{46}\text{Sc}$  in seston decreased at all three study sites to values near or less than 1 pCi/g DW by February and were unmeasurable by late summer, 1972 (Figure 2a). Seston (mostly phytoplankton) and periphyton have rapid reproductive rates and high adsorptive capacities due to large surface-to-volume ratios, thus usually resulting in relatively high concentrations of most radionuclides.

A similar pattern was evident for the periphyton (Figure 2b), except that the decline appears to lag behind that of the seston. This is probably related to the sessile nature of the periphyton as compared to the free-floating seston. Periphyton would retain relatively higher concentrations in the older portion of the community. Absolute concentrations in seston and periphyton were comparable.

Caddisfly larvae were sampled at White Bluffs from September 1971 to February 1972. During this time, they decreased from 3.1 pCi/g DW to below measurable limits. Scandium-46 concentrations in chironomid larvae in McNary Reservoir were measurable only until November 1971 (Figure 2c). During this time, concentrations varied from 5.4 to 34.5 pCi/g DW. Concentrations of  $^{46}\text{Sc}$  in crayfish, mussels, and forage fish in McNary during the second year of the study were unmeasurable. Concentrations of  $^{46}\text{Sc}$  in large fish collected at all sites averaged about 0.03 pCi/g DW, and no sample contained more than 0.6 pCi/g DW.

### MANGANESE-54 (HALF-LIFE, 290 DAYS)

Concentrations of  $^{54}\text{Mn}$  in seston at all sites varied from below measurable levels to slightly over 4 pCi/g DW (Figure 3a). The only exception to this was the April 1972 sample from White Bluffs, which contained 10.4 pCi/g DW. Concentrations of  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ , and  $^{137}\text{Cs}$  were also unusually high on this date, not only in seston, but in other organisms as well. This peak coincides with an increase in runoff and consequent scouring and resuspension of fine radioactively contaminated sediments (Robertson et al. 1973). Resuspension

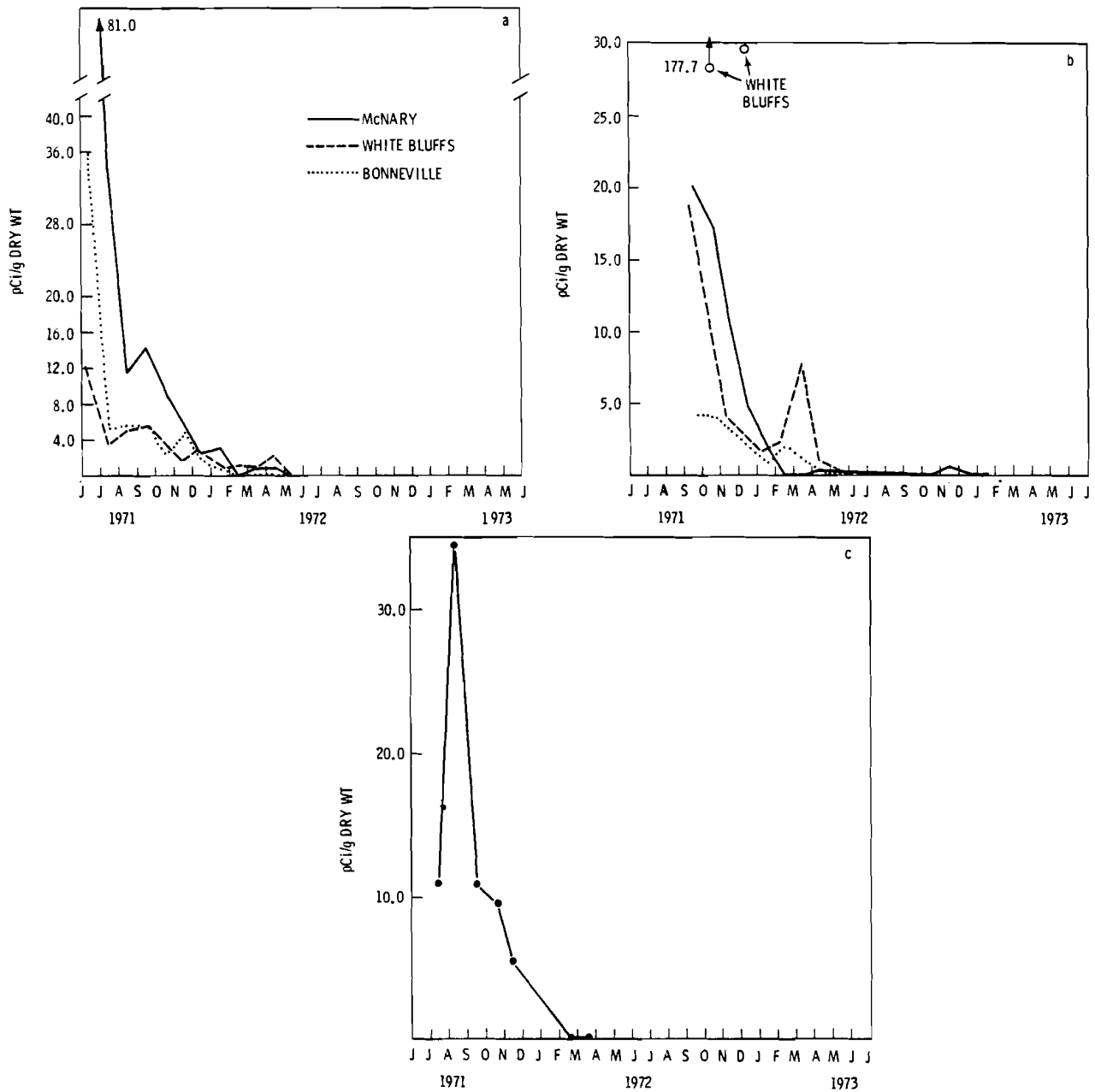
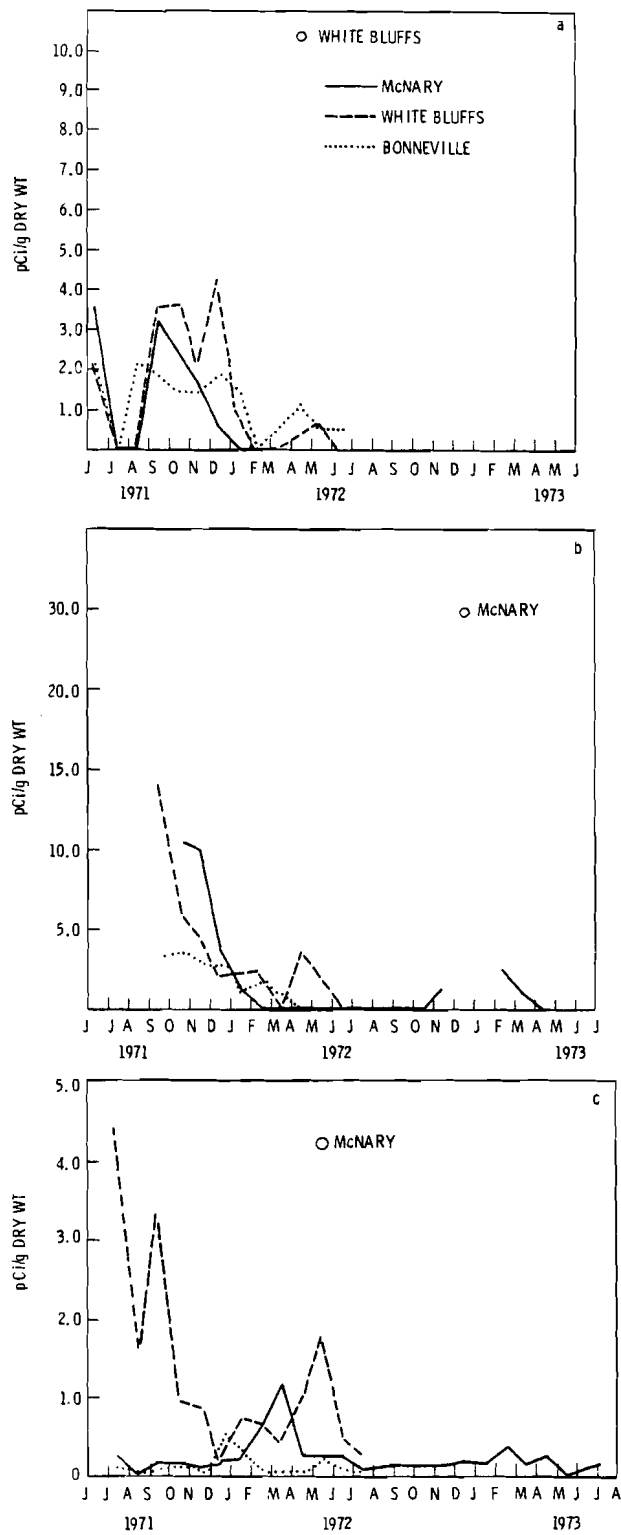


FIGURE 2. Concentrations of  $^{46}\text{Sc}$  in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Chironomids





**FIGURE 3.** Concentrations of  $^{54}\text{Mn}$  in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Suckers

evidently increases the availability of these radionuclides to the food web. Manganese-54 was not measurable in the periphyton after July 1972. Fluctuations reveal a general decrease during the study (Figure 3b).

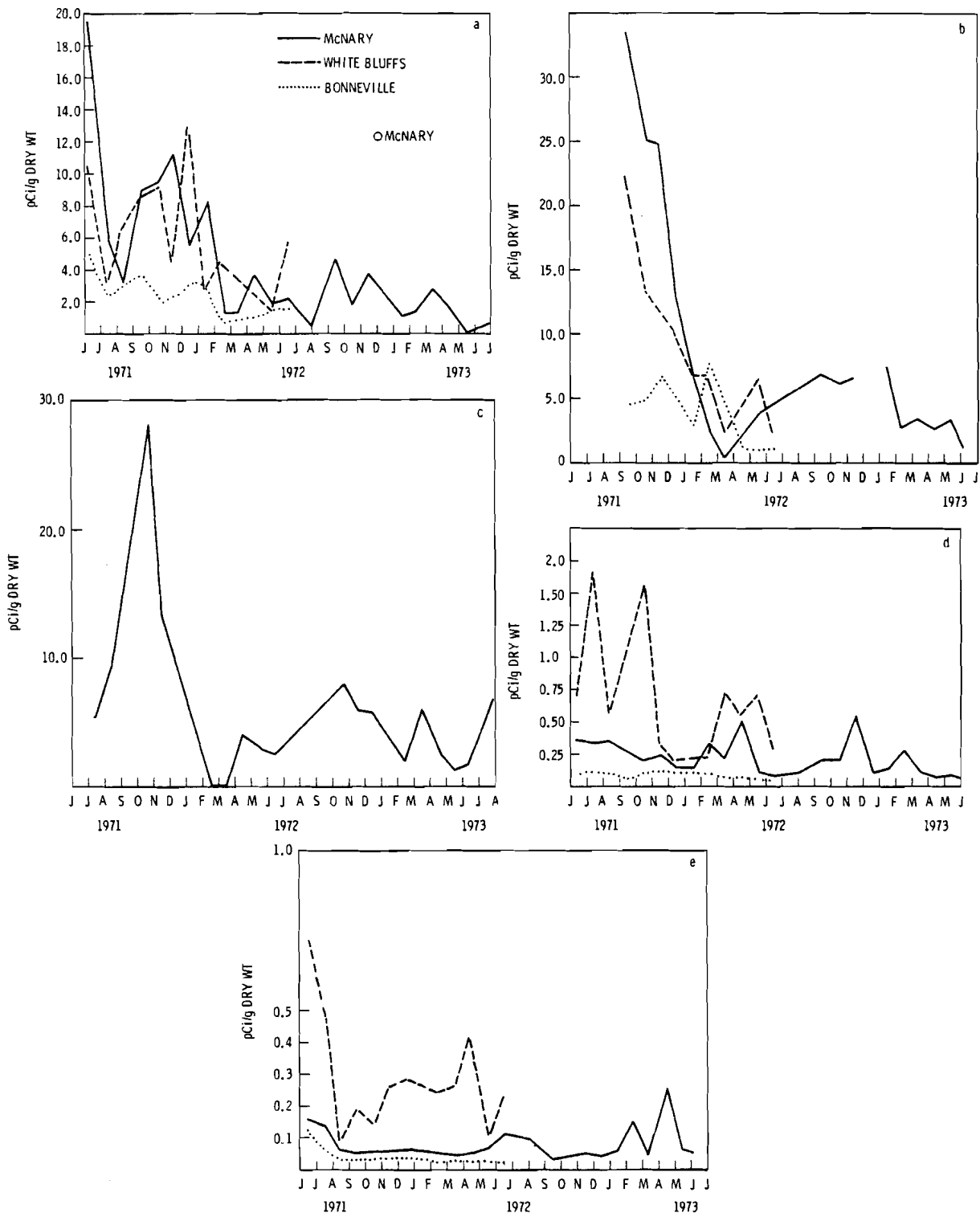
Manganese-54 in caddisfly larvae at White Bluffs decreased from 5 pCi/g DW in September 1972 to unmeasurable levels in February 1973. Manganese-54 in chironomid larvae in McNary was not measurable during the entire study. Mean concentrations of  $^{54}\text{Mn}$  in crayfish were  $0.78 \pm 0.5$  pCi/g DW in McNary between September 1972 and July 1973. The soft parts of the mussels were sampled during the last 4 mo of the study in 1973. Mean concentrations of  $^{54}\text{Mn}$  were  $11.15 \pm 2.1$  pCi/g DW. As mentioned above,  $^{54}\text{Mn}$  was virtually unmeasurable in seston after July 1972; yet this radionuclide was present in highest concentrations of any radionuclide in the mussel flesh. Mussels utilize seston for food and are known concentrators of  $^{54}\text{Mn}$  (Harrison 1969). The relatively high concentrations of  $^{54}\text{Mn}$  are probably related to the long effective half-life ( $T_e$  1/2) of  $^{54}\text{Mn}$  in *Anodonta* of about 1400 days (Harrison 1969). No seasonal trends were apparent in either the crayfish or mussels.

Manganese-54 was not measurable in forage fish in McNary from September 1972 until completion of the study. Concentrations of this radionuclide in suckers at White Bluffs decreased from June 1971 until June 1972 and remained essentially constant at Bonneville and McNary (Figure 3c). Mean concentrations in McNary suckers were  $0.24 \pm 0.2$  pCi/g DW. Manganese-54 concentrations in the carnivorous squawfish were always  $<1$  pCi/g DW and exhibited no overall trends at any study sites; values were usually not measurable during the latter part of the study. Mean concentrations were  $0.28 \pm 0.2$ ,  $0.03 \pm 0.05$  and  $0.01 \pm 0.02$  pCi/g DW at White Bluffs, McNary, and Bonneville, respectively. Concentrations of  $^{54}\text{Mn}$  in sturgeon at Bonneville did not show a noticeable trend; mean value was  $0.04 \pm 0.1$  pCi/g DW. Catfish were collected five times in McNary Reservoir; the mean concentration of  $^{54}\text{Mn}$  was  $0.02 \pm 0.03$  pCi/g DW. Values were not measurable in two of the five samples.

#### COBALT-60 (HALF-LIFE, 5.24 YEARS)

Concentrations of  $^{60}\text{Co}$  in seston at White Bluffs fluctuated during the first year with no appreciable overall decline. This was probably related to the proximity of the N Reactor (15 km upstream), from which  $^{60}\text{Co}$  leaches into the Columbia River through springs draining a disposal trench (D. E. Robertson, personal communication; Cushing and Watson, unpublished data). Mean concentration at White Bluffs was  $6.0 \pm 3.3$  pCi/g DW. An overall decrease from about 20 pCi/g DW to values near unity were measured over the two years in seston in McNary (Figure 4a). Concentrations of  $^{60}\text{Co}$  in seston at Bonneville also decreased during the first year from about 5 to 1 pCi/g DW (Figure 4a).

Cobalt-60 concentrations in periphyton at White Bluffs decreased from 22 to 2 pCi/g DW during the first year of the study (Figure 4b). In McNary, concentrations decreased from 34 to about 3 pCi/g DW during the first year and



**FIGURE 4.** Concentrations of  $^{60}\text{Co}$  in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Chironomids, d) Suckers, e) Squawfish

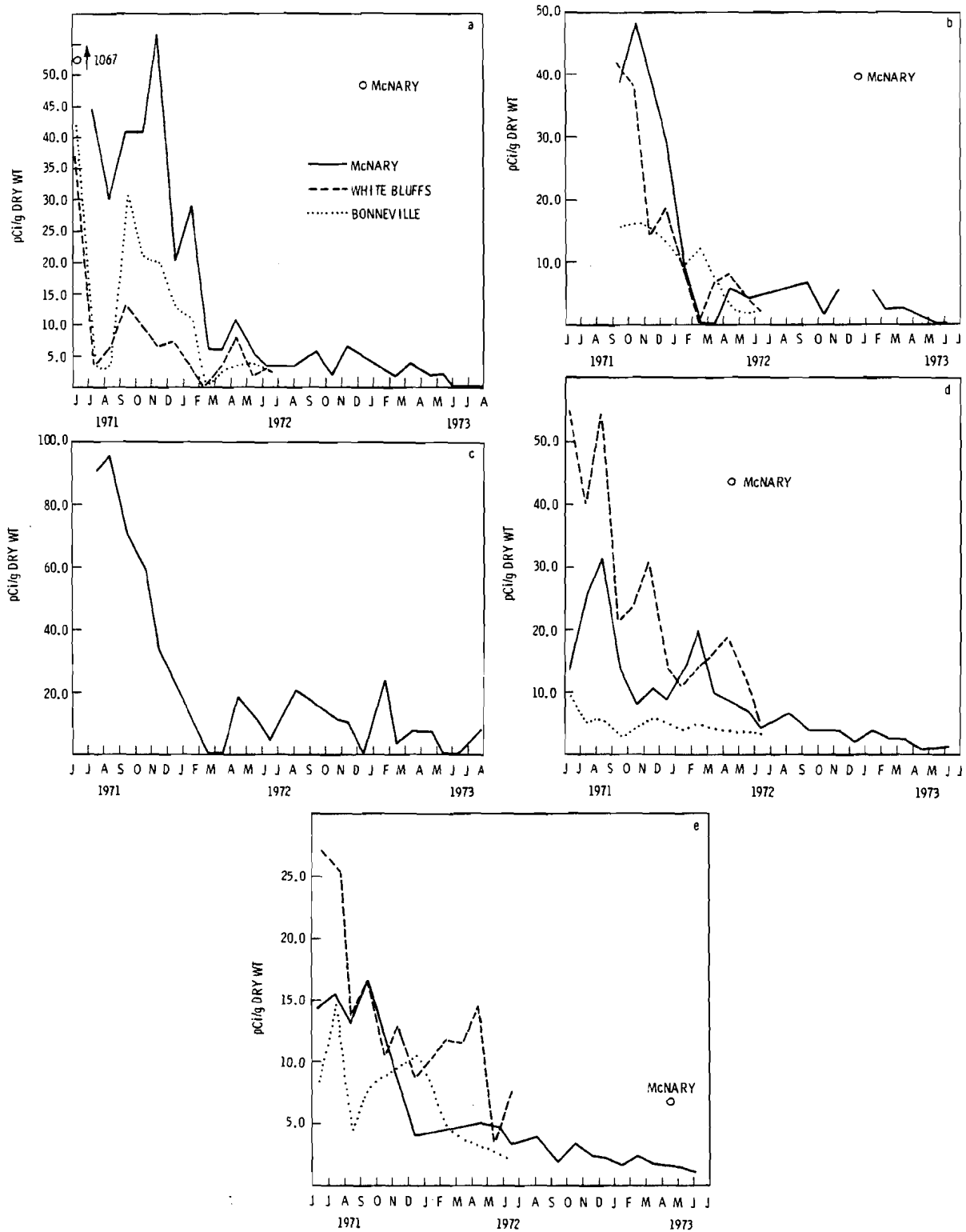
fluctuated between about 1 and 7 pCi/g DW during the second year. Unexplainably high values (ca 30 pCi/g DW) were present in December 1972; these are not shown on Figure 4b and are probably sampling artifacts. At Bonneville, concentrations declined from about 5 to 1 pCi/g DW (Figure 4b).

Caddisfly larvae at White Bluffs showed no appreciable decline of  $^{60}\text{Co}$  during the 6 mo they were available. Mean concentration was  $12.0 \pm 2.5$  pCi/g DW. The pattern of  $^{60}\text{Co}$  concentration in chironomids in McNary was unusual (Figure 4c); concentrations increased from 5.4 to 28.2 pCi/g DW from June to October 1972 and then decreased and remained essentially constant at about 5 pCi/g DW for the duration of the study. Crayfish in McNary contained a mean concentration of  $0.53 \pm 0.2$  pCi/g DW from September 1972 through June 1973; no apparent trends were evident. Freshwater mussels had a mean concentration of  $1.69 \pm 1.5$  pCi/g DW with no apparent trends.

Forage fish collected in McNary from September 1972 through June 1973 contained a mean  $^{60}\text{Co}$  concentration of  $0.2 \pm 0.1$  pCi/g DW and exhibited no discernible trends in concentrations. There is some evidence of a decrease in  $^{60}\text{Co}$  concentration in suckers from White Bluffs (Figure 4d), although the fluctuations at the beginning and end of the first year overlap. Mean concentration was  $0.68 \pm 0.5$  pCi/g DW. Cobalt-60 concentrations in suckers in McNary decreased from about 0.39 to 0.06 pCi/g DW during the two years of study (Figure 4d). No obvious trend in  $^{60}\text{Co}$  concentration in suckers at Bonneville was found (Figure 4d). Cobalt-60 concentrations in squawfish at White Bluffs exhibited alternating increases and decreases (Figure 4e). Cobalt-60 concentrations in squawfish from McNary showed no overall trend; mean concentration was  $0.08 \pm 0.1$  pCi/g DW (Figure 4e). At Bonneville, concentrations in squawfish decreased from 0.12 to 0.03 pCi/g DW in the first 3 mo of study and remained essentially constant for the next 11 mo (Figure 4e). Cobalt-60 in sturgeon was quite uniform; mean concentration was  $0.13 \pm 0.1$  pCi/g DW. Concentrations of  $^{60}\text{Co}$  in catfish from McNary decreased slightly during the last year of study from about 0.13 to 0.05 pCi/g DW.

#### ZINC-65 (HALF-LIFE, 245 DAYS)

In general,  $^{65}\text{Zn}$  was the radionuclide in highest concentration in the biota. Concentrations of  $^{65}\text{Zn}$  in seston at White Bluffs decreased quite rapidly, as would be expected of this transient community (Figure 5a). After the initial decrease from about 38 pCi/g DW to 3 pCi/g DW,  $^{65}\text{Zn}$  levels fluctuated during the following year. The increase of  $^{65}\text{Zn}$  in April 1972 in this and other organisms coincided with the spring runoff, a period in which  $^{65}\text{Zn}$  sorbed to fine sediments is scoured and resuspended in the water column. This could make this radionuclide available for biological uptake. Zinc-65 in McNary seston did not decline as much, initially, as it did at White Bluffs, nor did it decrease as far until about March 1972 (Figure 5a). At Bonneville, seston  $^{65}\text{Zn}$  concentrations fluctuated similarly as at White Bluffs (Figure 5a). At the end of the first year of study,  $^{65}\text{Zn}$  concentrations were about 3 pCi/g DW at all sites. Zinc-65 concentrations in seston at McNary gradually



**FIGURE 5.** Concentrations of  $^{65}\text{Zn}$  in Selected Columbia River Biota; a) Seston, b) Periphyton, c) Chironomids, d) Suckers, e) Squawfish

decreased during the second year of study, with some fluctuations, to essentially unmeasurable levels (Figure 5a). Decreases of  $^{65}\text{Zn}$  in periphyton were similar to decreases in the seston (Figure 5b). The April 1972 pulse was evident at White Bluffs, but it was less evident at McNary and absent at Bonneville. Cushing and Watson (1971) estimated an effective half-life of 15 days for  $^{65}\text{Zn}$  in periphyton in laboratory streams.

The concentration of  $^{65}\text{Zn}$  in caddisfly larvae at White Bluffs decreased from about 11 pCi/g DW in September 1972 to undetectable levels by February 1973. Zinc-65 concentrations in chironomids in McNary decreased rapidly from about 95 pCi/g DW to less than 10 pCi/g DW in 6 mo (Figure 5c). Concentrations fluctuated between undetectable amounts to 24 pCi/g DW from February 1972 to the end of the study. Dean (1974) found that tubificid worms, which ingest sediments as do chironomids, accumulated  $^{65}\text{Zn}$  from the water but not from the sediments. Thus, the decreasing concentrations of  $^{65}\text{Zn}$  in chironomids may be partially related to decreased concentration in the water rather than the sediments. Renfro (1973), on the other hand, found that marine polychaete worms accumulated  $^{65}\text{Zn}$  from radioactively labelled sediments. In the same study, it was demonstrated that considerable  $^{65}\text{Zn}$  was lost from the sediments because of burrowing activities of the polychaetes (Renfro 1973). Although decreasing sediment activity in McNary is related more to resuspension and transport and/or burying by new uncontaminated sediments, loss of  $^{65}\text{Zn}$  due to agitation by chironomid larvae may be a contributing factor.

Crayfish in McNary exhibited an overall decrease from about 1.7 to 0.8 pCi/g DW during the 9 mo they were sampled. Mean concentration was  $1.26 \pm 0.5$  pCi/g DW. Zinc-65 in mussel flesh also decreased during the 4 mo they were collected. Harrison (1969) reported an effective half-life of  $^{65}\text{Zn}$  of about 650 days for the mussel *Anodonta nuttalliana*, and Seymour (1973) reported an effective half-life of 162 days for  $^{65}\text{Zn}$  in a marine mollusc, *Crassostrea gigas*. Mean concentration of  $^{65}\text{Zn}$  in McNary mussels was  $19.5 \pm 2.7$  pCi/g DW.

Concentrations of  $^{65}\text{Zn}$  in forage fish in McNary were essentially constant during the 7 mo they were sampled. The mean concentration was  $2.57 \pm 2.8$  pCi/g DW. Zinc-65 concentrations in suckers at White Bluffs decreased from about 55 to 5 pCi/g DW during the first year of study (Figure 5d). At McNary, concentrations fluctuated during the first year and then gradually decreased during the second year to values of about 1 pCi/g DW (Figure 5d). Lower values were present at Bonneville; they decreased and remained fairly constant at about 3 pCi/g DW by the end of the first year (Figure 5d). Concentrations of  $^{65}\text{Zn}$  in the carnivorous squawfish at White Bluffs fluctuated but showed an overall decrease from about 27 to 5 pCi/g DW during the first year (Figure 5e). At McNary, the rapid decrease during the first year slowed during the second; concentrations at the conclusion of the study were about 1.5 pCi/g DW (Figure 5e). Concentrations also fluctuated at Bonneville, but revealed an overall decline during the first year (Figure 5e). Zinc-65 concentrations in catfish from McNary decreased from 2.8 to 1.3 pCi/g DW from August 1972 to June 1973; the mean concentration was  $2.05 \pm 1.1$  pCi/g DW. At Bonneville,  $^{65}\text{Zn}$  concentration in sturgeon also decreased about threefold from 6 to 2 pCi/g DW during the first year. Mean concentration was  $3.47 \pm$



1.3 pCi/g DW. Jones (1975) calculated ecological half-lives (defined as the time the organism requires to lose 50% of the body burden by both decay and biological turnover) of  $^{65}\text{Zn}$  in various tissues from carp in McNary at the same time our study was in progress. He found no statistical difference among the values for the various tissues and so derived a common loss rate with an ecological half-life of 177 days.

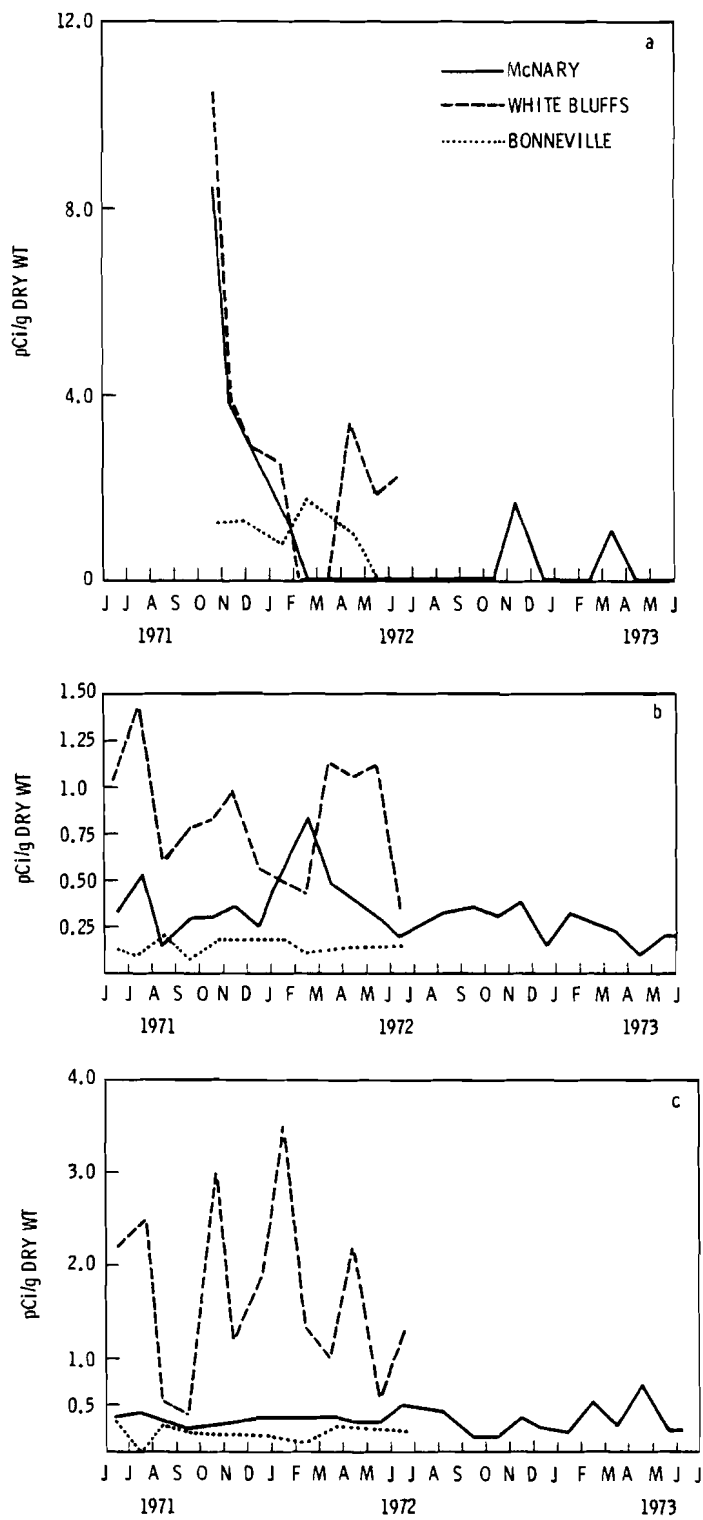
Analyses of water samples showed that about 20% of the  $^{65}\text{Zn}$  was in ionic form when the reactors were operating (Perkins, Nelson and Haushild 1966), but that less than 2% was ionic in 1971 and 1972 (Robertson et al. 1973), a tenfold decrease. With these data in mind, it is of interest to compare ratios of  $^{65}\text{Zn}$  in various organisms and in their principal food during the above periods of time. Suckers graze on periphyton communities; when the reactors were operating, there was approximately 10 times as much  $^{65}\text{Zn}$  in the periphyton as in suckers (Watson et al. 1970). In 1971 and 1972, there was only about two times as much  $^{65}\text{Zn}$  in the periphyton. The same ratios prevailed for the squawfish, which prey on smaller forage fish. The change in ratio approximated that of the changes in ionic  $^{65}\text{Zn}$ . The decrease of  $^{65}\text{Zn}$  by 9 to 10 times in the chironomids closely approximates the tenfold decrease of  $^{65}\text{Zn}$  in the sediments. Since chironomids ingest sediments, the decrease essentially indicates that radionuclide measurements of chironomids are reflective of the gut contents rather than the tissue.

#### CESIUM-137 (HALF-LIFE, 30 YEARS)

Concentrations of  $^{137}\text{Cs}$  in seston at all three sites were too near the limits of detectability to reveal any pattern over the period of study. When measurable, it rarely exceeded 2 pCi/g DW. Figure 6a shows the trends for  $^{137}\text{Cs}$  concentrations in periphyton. After the initial decrease during the first 6 to 9 mo of sampling, measurable concentrations occurred sporadically at the different sites.

Concentrations of  $^{137}\text{Cs}$  in caddisfly larvae at White Bluffs were measurable twice; mean concentration was  $0.89 \pm 0.7$  pCi/g DW. No measurable concentrations of  $^{137}\text{Cs}$  occurred in chironomids, crayfish, or mussels from McNary.

The mean concentration of  $^{137}\text{Cs}$  in forage fish was  $0.11 \pm 0.1$  pCi/g DW; it was unmeasurable in half of the samples. Concentrations of  $^{137}\text{Cs}$  in suckers at White Bluffs fluctuated markedly; the mean concentration was  $0.83 \pm 0.3$  pCi/g DW. No long-term decrease in  $^{137}\text{Cs}$  in suckers from McNary was observed (Figure 6b). The mean concentration was  $0.39 \pm 0.4$  pCi/g DW. At Bonneville,  $^{137}\text{Cs}$  concentrations in suckers were lower and also constant; mean concentration was  $0.15 \pm 0.04$  pCi/g DW. Cesium-137 concentrations in squawfish fluctuated widely at White Bluffs but were relatively constant at McNary and Bonneville (Figure 6c). Mean concentrations were  $1.68 \pm 1.0$ ,  $0.34 \pm 0.1$  and  $0.2 \pm 0.1$  pCi/g DW at White Bluffs, McNary, and Bonneville, respectively. Mean concentrations of  $^{137}\text{Cs}$  in catfish from McNary and sturgeon from Bonneville were  $0.23 \pm 0.1$  and  $0.13 \pm 0.1$  pCi/g DW, respectively; overall values were constant.



**FIGURE 6.** Concentrations of  $^{137}\text{Cs}$  in Selected Columbia River Biota; a) Periphyton, b) Suckers, c) Squawfish

## CONCLUSIONS

The biota in the Columbia River ecosystem below Hanford are being exposed to a changing, and much lower, ambient level of radionuclides in their environment. True effective half-lives of the radionuclides cannot be determined since radionuclides are still available from the sediments, from N Reactor seepage effluents and from the residual radioactivity in the different organisms in the food web. The large sediment-bound radionuclide pool in McNary Reservoir will become less available to biota with time as it is covered with successive layers of sediments carrying only radionuclides related to the N Reactor, a much smaller fraction than originally found.

Nevertheless, these data show that in a river-reservoir complex, the measurable body burden of fission-produced radionuclides decreased to essentially undetectable levels within 18 to 24 mo of cessation of input of once-through cooling water into the river. We further hypothesize, on the basis of the data from the White Bluffs station, that the decline would be more rapid, particularly in the lower trophic levels, in a free-flowing river. Slack water allows the sediments and their associated radionuclide burden to settle and accumulate, thus providing a pool for subsequent resuspension and recycling of radionuclides within the food web.

## REFERENCES

- Becker, C. D. 1973. Aquatic Bioenvironmental Studies in the Columbia River at Hanford, 1945-1971: A Bibliography with Abstracts. BNWL-1734, Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Cushing, C. E., and D. G. Watson. 1971. "Zinc-65 Cycling in a Simple Food Web." In Radionuclides in Ecosystems, Proceedings of Third National Symposium on Radioecology, ed. D. J. Nelson, pp. 318-322. CONF-710501, NTIS, Springfield, Virginia.
- Davis, J. J., D. G. Watson and C. C. Palmiter. 1956. Radiobiological Studies of the Columbia River Through December, 1955. HW-36074, Hanford Atomic Power Operations, Richland, Washington.
- Dean, J. M. 1974. "The Accumulation of  $^{65}\text{Zn}$  and Other Radionuclides by Tubificid Worms." Hydrobiologia 45:33-38.
- Harrison, F. L. 1969. "Accumulation and Distribution of  $^{54}\text{Mn}$  and  $^{65}\text{Zn}$  in Freshwater Clams." In Symposium on Radioecology, Proceedings of Second National Symposium on Radioecology, eds. D. J. Nelson and F. C. Evans, pp. 198-220. CONF-670503, NTIS, Springfield, Virginia.
- Jones, W. M., Jr. 1975. Iron-55 and Zinc-65 in Tissues of Columbia River Carp. Master of Science Thesis, Oregon College of Education, Monmouth, Oregon.
- Perkins, R. W., J. L. Nelson and W. L. Haushild. 1966. Behavior and Transport of Radionuclides in the Columbia River between Hanford and Vancouver, Washington. Limnol. Oceanogr. 11:235-248.
- Renfro, W. C. 1973. "Transfer of  $^{65}\text{Zn}$  from Sediments by Marine Polychaete Worms." Marine Biology 21:305-316.
- Robeck, G. C., C. Henderson and R. C. Palange. 1954. Water Quality Studies on the Columbia River. Special Report, U.S. Public Health Service, R. A. Taft Sanit. Engr. Center, Cincinnati, Ohio.
- Robertson, D. E., W. B. Silker, J. C. Langford, M. R. Petersen and R. W. Perkins. 1973. "Transport and Depletion of Radionuclides in the Columbia River." In Radioactive Contamination of the Marine Environment, International Atomic Energy Agency, Vienna, pp. 141-158.
- Seymour, A. H., and V. A. Nelson. 1973. "Decline of  $^{65}\text{Zn}$  in Marine Mussels Following the Shutdown of Hanford Reactors." In Radioactive Contamination of the Marine Environment, International Atomic Energy Agency, Vienna, pp. 277-286.

- Watson, D. G., C. E. Cushing, C. C. Coutant and W. L. Templeton. 1969. "Effect of Hanford Reactor Shutdown on Columbia River Biota." In Symposium on Radioecology, Proceedings of Second National Symposium on Radioecology, eds. D. J. Nelson and F. C. Evans, pp. 291-299. CONF-670503, NTIS, Springfield, Virginia.
- Watson, D. G., C. E. Cushing, C. C. Coutant and W. L. Templeton. 1970. "Cycling of Radionuclides in Columbia River Biota." In Proceedings of Fourth Annual Conference on Trace Substances in Environmental Health, ed. D. D. Hemphill, pp. 144-157. University of Missouri, Columbia.
- Wogman, N. A., D. E. Robertson and R. W. Perkins. 1967. "A Large Detector, Anticoincidence Shielded Multidimensional Gamma-Ray Spectrometer." Nucl. Instr. and Methods 50:1-10.

DISTRIBUTION

<u>No. of Copies</u>		<u>No of Copies</u>
	<u>OFFSITE</u>	
	A. A. Churm DOE Patent Division 9800 South Cass Avenue Argonne, IL 60439	R. E. Franklin Office of the Assistant Secretary for Environment DOE Washington, DC 20545
	DOE Office of Health and Environmental Research Washington, DC 20545	T. E. Hakonson Environmental Studies Group H-8 Los Alamos Scientific Laboratory Box 1663 Los Alamos, NM 87544
27	Technical Information Center	
	S. I. Auerbach Oak Ridge National Laboratory Oak Ridge Operations Office PO Box X Oak Ridge, TN 37830	D. H. Hamilton Office of Health and Environmental Research DOE Washington, DC 20545
	R. C. Dahlman Office of the Assistant Secretary for Environment DOE Washington, DC 20545	R. A. Lewis Office of the Assistant Secretary for Environment DOE Washington, DC 20545
	J. J. Davis Assistant Director of Research Nuclear Regulatory Commission Washington, DC 20545	Librarian, Building 465 Atomic Energy Research Establishment Harwell, Didcot Oxfordshire OXII ORQ ENGLAND
	J. W. Elwood Environmental Sciences Division Oak Ridge National Laboratory Oak Ridge, TN 37830	Librarian Australian AEC Riverina Laboratory PO Box 226 Deniliquin, New South Wales AUSTRALIA 2710
	W. O. Forster Office of the Assistant Secretary for Environment DOE Washington, DC 20545	



No. of  
Copies

Librarian  
Battelle Memorial Institute  
Columbus Laboratories  
505 King Avenue  
Columbus, OH 53201

Librarian  
Centre d' Etudes  
Nucleaires de Saclay  
PO Box 2, Saclay  
Fig-sur-Yvette (S&O)  
FRANCE

Librarian  
Commonwealth Scientific and  
Industrial Research Organization  
314 Albert Street  
PO Box 89  
East Melbourne, Victoria  
AUSTRALIA 3002

Librarian  
Directorate of Fisheries Research  
Ministry of Agriculture, Fisheries  
and Food Laboratory  
Lowestoft, Suffolk  
ENGLAND NR33 0HT

Librarian  
Lawrence Radiation Laboratory  
University of California  
Technical Information Dept., L-3  
PO Box 808  
Livermore, CA 94550

Librarian  
Los Alamos Scientific Laboratory  
PO Box 1663  
Los Alamos, NM 87544

Librarian  
National Radiological Protection  
Board  
Harwell, Didcot  
Oxfordshire OX11 0RQ  
ENGLAND

No. of  
Copies

Librarian  
Research Library, Reference  
Brookhaven National Laboratory  
Upton, Long Island, NY 11973

Librarian  
World Meteorological Organization  
Geneva  
SWITZERLAND

O. Doyle Markham  
Radiological and Environmental  
Sciences Laboratory  
U.S. DOE  
PO Box 2108  
Idaho Falls, ID 83401

H. M. McCammon  
Office of the Assistant Secretary  
for Environment  
DOE  
Washington, DC 20545

R. E. Nakatani  
Assistant Director  
Fisheries Research Institute  
260 Fisheries Center  
University of Washington  
Seattle, WA 98105

Professor J. M. Neuhold  
Department of Wildlife Resources  
Utah State University  
Logan, UT 84321

I. Ophel  
Atomic Energy of Canada Ltd.  
Chalk River, Ontario  
CANADA

W. S. Osburn, Jr.  
Office of the Assistant Secretary  
for Environment  
DOE  
Washington, DC 20545

No. of  
Copies

No. of  
Copies

<u>No. of Copies</u>		<u>No. of Copies</u>	
	G. Saunders Office of the Assistant Secretary for Environment DOE Washington, DC 20545		<u>ONSITE</u>
	Vincent Schultz Department of Zoology Washington State University Pullman, WA 99163	2	<u>DOE Richland Operations Office</u> M. W. Tiernan M. G. White H. E. Ransom
	Allyn H. Seymour College of Fisheries University of Washington Seattle, WA 98105	2	<u>Rockwell Hanford Operations</u> R. D. Fox P. G. Lorenzini
	Mike Smith, Director Savannah River Ecology Laboratory University of Georgia Savannah River Plant PO Box A Aiken, SC 29801	62	<u>Pacific Northwest Laboratory</u> C. D. Becker L. L. Cadwell J. P. Corley C. E. Cushing (12) D. D. Dauble L. L. Eberhardt R. M. Emery D. H. Fickeisen R. H. Gray J. M. Gurtisen (3) W. C. Hanson J. R. Houston D. N. Neitzel Y. Onishi T. L. Page T. M. Poston K. R. Price W. H. Rickard D. E. Robertson R. G. Schreckhise (4) A. J. Scott (3) J. K. Soldat J. B. States J. A. Strand W. L. Templeton B. E. Vaughan D. G. Watson (4) R. E. Wildung Technical Information (5) Publishing Coordination (2)Le
	J. Swinebroad, Director Office of the Assistant Secretary for Environment DOE Washington, DC 20545		
	A. Walton Director International Atomic Energy Agency International Laboratory Marine Radioactivity Oceanographic Museum Monaco-Ville Principality of Monaco		
	R. L. Watters Nuclear Programs Office of Health and Environmental Research DOE Washington, DC 20545		
	Ward Whicker Colorado State University Fort Collins, CO 80521		