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THE REMOVAL OF RADIOISOTOPES FROM
THE COLUMBIA RIVER BY NATURAL PROCESSES

by

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ABSTRACT

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Some radioisotopes are introduced into the Columbia River by return of the water used to cool the Hanford reactors. Trace amounts of both fission product and activation product radioisotopes are present and measurable by sensitive radiochemical methods to allow determination of the self-purification processes occurring as the water moves downstream. By comparing the radioisotope input at given times with the amounts present in the river after traveling downstream, a measure of the self-purification rate was obtained. Removal in the first 40 miles downstream from the reactors was about 35 percent for 15 of the most prominent radioisotopes. The removal rate seemed to decrease with subsequent distance traveled downstream for three radioisotopes for which measurements were made. Radiochemical analyses of water, filterable solids and sediment samples taken at downstream locations indicate that incorporation in sediments accounts for most of the removal. Because of the intense scouring action of the Columbia River, sediments are deposited in significant amounts only in slack water sections. The first extensive slack water section downstream from Hanford is the McNary Dam reservoir, Lake Wallula. Radiochemical analyses of some of the long-lived radioisotopes, including Mn⁵⁴, Co⁶⁰, Zn⁶⁵ and Cs¹³⁷ on sectioned sediment core samples indicate a widely varying rate of deposition of radioisotopes. Dating of the various sections of the cores was accomplished using isotope ratio measurements.

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INTRODUCTION

Use of Columbia River water as a coolant for the Hanford reactors and its subsequent return to the river results in the introduction of radioisotopes produced by neutron activation or fission reactions on river salts, water process treatment additives, and reactor corrosion materials. Control and analysis of the amount of radioisotopes released assure that the radiation exposure to humans resulting from downstream use of this water is minimized and within standard control limits. Included among these radioisotopes are some in sufficient concentration and with half-lives long enough to enable their paths through the downstream Columbia River environs to be followed. Significant fractions of these radioisotopes are, or quickly become, associated with particulate material. This may occur by precipitation, adsorption onto particulate materials or adsorption by the biota of the river. Subsequent sedimentation of these materials may result in a self purification of the river water. The studies reported here were designed to measure the magnitude of the self purification by the Columbia River of certain radioisotopes and to study the factors involved in the incorporation of radioisotopes in river sediments.

RIVER DEPLETION

Reactor effluent water outfalls introduce radioisotopes to the Columbia River along a stretch of 14 miles (Figure 1). The first major point of domestic use of the river water is at the Pasco, Washington, Water Plant located 38 miles below the last downstream reactor, a distance which allows a decay of from 12 to 32 hours due to the transit time. Each reactor basin outlet was sampled on a

FIGURE 1

HANFORD AND ENVIRONS

schedule of corresponding times and combined into a composite sample according to each reactor flow rate so that a single sample representing the radioisotope output at a given time was obtained. At the proper time, calculated from river flow time data⁽¹⁾, a corresponding Columbia River water sample was obtained at Pasco, Washington. At this point the specific activity of the water is about $3-15 \times 10^{-6}$ $\mu\text{c/ml}$, depending upon the amount of water in the river at the time. Individual radioisotope concentrations are, in most cases, much lower so that large samples and low level radiochemical techniques are necessary to obtain the accuracy sufficient for these measurements. Wherever possible beta absorption⁽²⁾ and gamma-ray spectrometric⁽³⁾ procedures, which require no chemical separation, were used. The radioisotope content of the composite reactor samples and the river samples were analyzed and the necessary dilution and decay corrections made so that the amounts removed could be obtained directly.

The amounts removed from the Columbia River by natural processes between the reactors and Pasco, Washington, are tabulated in Tables I and II. Table I includes those radioisotopes which comprise 1% or greater of the activity observed at Pasco, Washington. These data are shown in Figure 2 as a bar graph with the width of the bar proportional to an individual radioisotope activity percent compared to the total activity. The average removal is about 30%. Cr^{51} is a K-capture radioisotope emitting only a 0.32 Mev gamma-ray, thus it contributes no beta activity and relatively little to the exposure of persons drinking the water. The average removal of those isotopes which contribute to drinking water exposure is about 40%. As^{76} , with a 39% removal, is the radioisotope which contributes most to the drinking water exposure, largely due to its 2.96 Mev beta radiation.

(1) J.K. Soldat, "Columbia River Flow-Time Calculations", HW-58312, Nov. 21, 1958.

(2) R.W. Perkins, "Determination of P^{32} and As^{76} in Reactor Effluent Water by Beta-Ray Absorption Techniques", HW-50310 (July 2, 1957) Confidential.

(3) R.W. Perkins, "Gamma-Ray Spectrometric Systems of Analysis", Progress in Nuclear Energy Series IX, Vol. I - Analytical Chemistry, 1959, Pergamon Press.

FIGURE 2

NATURAL DEPLETION (EXCLUDING DECAY)

BETWEEN REACTOR OUTFALLS AND PASCO, WASHINGTON

In Table II are tabulated those radioisotopes which individually contribute less than 1% of the radioactivity and for which measurements were made. Co^{60} , Sc^{46} , Ba^{140} and La^{140} had depletion values comparable to the average value for most of the radioisotopes. The Sr^{89} depletion was very low while the rare earths Sm^{153} and Ce^{143} showed the highest depletion values observed.

Recent removal studies have been made more difficult since operation of Priest Rapids Dam just upstream from the Hanford reactors. Normal daily operation varies the river flow past Hanford by factors of 2-4 during most of the year. This adds a large uncertainty to the decay time estimations based on flow rates.

TABLE I

PERCENTAGE DEPLETION OF MAJOR RADIOISOTOPES IN
THE COLUMBIA RIVER FROM THE REACTORS TO PASCO, WASHINGTON

Sample	Radioisotope							
	<u>Cr^{51}</u>	<u>Na^{24}</u>	<u>Np^{239}</u>	<u>Cu^{64}</u>	<u>As^{76}</u>	<u>Ga^{72}</u>	<u>Zn^{65}</u>	<u>P^{32}</u>
11/14/56	21	47	24	56	42		58	51
11/15/56	0	39	17	49	54		19	57
11/16/56	25	54	30	60	32		67	52
3/7/57	10	50	19	60	36		24	0
3/8/57	26	47	46	--	30		21	11
3/9/57	29	61	37	52	45		36	13
1/21/58	8	51	29	59	25	6	28	37
10/9/58	<u>3</u>	<u>49</u>	<u>28</u>	<u>62</u>	<u>50</u>	<u>-</u>	<u>59</u>	<u>36</u>
Average (with standard deviation)	15±11	50±6	29±9	57±5	39±10	6	39±19	32±21

TABLE II

PERCENTAGE DEPLETION OF SOME MINOR RADIOISOTOPES IN THE
COLUMBIA RIVER FROM THE REACTORS TO PASCO, WASHINGTON

<u>Radioisotope</u>	<u>No. of Samples</u>	<u>Average Removal (Percent)</u>
Co ⁶⁰	5	36 ± 34
Sc ⁴⁶	8	43 ± 34
Sr ⁸⁹	1	~0
Ba ¹⁴⁰	4	27 ± 32
Sm ¹⁵³	2	65 ± 5
La ¹⁴⁰	1	34
Ce ¹⁴³	1	65

The radioisotope concentrations of Cr⁵¹, Np²³⁹, P³² and Zn⁶⁵ were measured on samples taken at nine sampling stations on the Columbia River between Arlington, Ore. and Vancouver, Wash., a distance of 132 miles (Arlington is 88 miles downstream from Pasco). These samples were taken over a two-day period although the estimated flow time was 5.5 days (132 hours). Since river flow volume corrections were made and reactor operations during the corresponding period did not vary more than 10%, the measurements are reasonably representative with the largest uncertainty in the flow time estimate. Because of the relatively long half lives of P³², Cr⁵¹ and Zn⁶⁵ the depletion is not greatly dependent on errors in flow time estimate of the order of a day. For example, if the flow time were 6.5 days instead of 5.5 days the depletion of P³² would be 18% instead of 22%. The effect on Cr⁵¹ and Zn⁶⁵ would be even smaller. The value for Np²³⁹ is not very precise but serves to indicate that

the choice of flow time is reasonable. The measured depletion is, in each case, somewhat less than would be expected for the 132 miles traveled if the measured removal rates remained the same downriver as they were from the reactor to Pasco.

TABLE III

DEPLETION OF CERTAIN RADIOISOTOPES FROM THE
COLUMBIA RIVER BETWEEN ARLINGTON AND VANCOUVER, WASHINGTON

<u>Radioisotope</u>	<u>Half Life</u> (Days)	<u>Depletion</u> (%)
Cr ⁵¹	27	21
Np ²³⁹	2.33	< 25
P ³²	14.5	22
Zn ⁶⁵	245	48

All of the river water samples obtained in this study contained particulate suspended material in addition to the dissolved salts. In some cases this suspended material was removed by filtration* in order to provide a measure of the relative amounts of radioisotopes carried by the particulate material and as dissolved salts. The results of these measurements on five river water samples taken at Pasco, Washington, are tabulated in Table IV with the average depletion values for radioisotopes in the river from the reactors to Pasco shown for comparison. Particularly notable are the values for Na²⁴ and As⁷⁶ which show relatively little attached to particulate material but rather large depletion values. Also notable are Zn⁶⁵ and P³² values which show about one-half or more on the particulate material. Although much remains to be explained, association of the radioisotopes with this particulate material

* Millipore HA filter, Millipore Filter Corp., Bedford, Massachusetts.

is probably the main mechanism of loss to the bottom of the river. The particulate material probably consists of bacterial and slowly sedimenting precipitates as well as the usual solid material held in suspension in streams. Various biota of the river can account for a few percent, at most, of the radioactivity in the water. The bulk of the solid material is silt and clay.

TABLE IV
PERCENTAGE RADIOISOTOPES ON FILTERABLE SOLIDS
COMPARED WITH AVERAGE DEPLETION

<u>Sample</u> (Date)	<u>Radioisotope</u>						
	<u>Cr⁵¹</u>	<u>Na²⁴</u>	<u>Np²³⁹</u>	<u>Cu⁶⁴</u>	<u>As⁷⁶</u>	<u>Zn⁶⁵</u>	<u>P³²</u>
1/27/57	3.9		6.5	59		62	86
11/10/58	3.3	0.9	1.4	15	3.6	67	43
10/21/59	3.3	0.6	1.4	15	3.7		
2/16/62	3.3	1.7	8.0	16	4.7	31	
2/17/62	<u>3.4</u>	<u>1.2</u>	<u>7.1</u>	<u>22</u>	<u>6.4</u>	<u>33</u>	
Average (with standard deviation)	3.4±.3	1.1±.3	4.9±.3	25.4±19	4.6±1.3	48.2±18	64.5±30
Depletion	15	50	29	57	39	39	32

RIVER SEDIMENTS

Because of the high velocity of flow, very little permanent deposition of sediment occurs in the Columbia River except in slack water areas. The first extensive slack water section downstream from Hanford is the McNary Dam reservoir, Lake Wallula. This dam is 38 miles downstream from Pasco. The

reservoir was filled in 1954 and studies of the rates of deposition are being made on the sediments laid down since that time. Core samples of the bottom sediments were obtained by means of a modified Phlager design core sampler*. The sediment cores obtained with this device are quickly frozen to eliminate radioisotope migration and cut into 1 to 2 inch sections for radioisotopic analysis. Concentrations of 6 radioisotopes have been made to date on 7 core samples taken just upstream from the dam. The values obtained are tabulated in Table V as curies per square mile of sediment bed, based on the averaged values from the 7 cores. Over two-thirds of the radioactivity is Zn^{65} . Zn^{65} and Cr^{51} represent about 90% of the total radioactivity.

TABLE V
RADIOISOTOPES IN MCNARY RESERVOIR SEDIMENTS

<u>Radioisotope</u>	<u>Half-Life</u>	<u>Curies per Square Mile</u>
Zn^{65}	245 d	230
Cr^{51}	27 d	62
Co^{60}	5.2 y	10
Sc^{46}	85 d	9
Mn^{54}	300 d	6
Cs^{137}	30 y	4
P^{32}	14.5 d	

* Gravity coring tube No. 15M1440, modified Phlager design, G.M. Manufacturing Company, New York, New York, modified to take 2-3 foot cores.

Figure 3 shows the concentrations of Zn^{65} and Co^{60} at various depths in a typical McNary reservoir core sample. Since the half lives of Zn^{65} and Co^{60} are different, (245 d and 5.2 y, respectively) the ratio of their concentrations can be used to determine the age of the various samples provided certain other data are known. These are, the variation with time of the ratio of the Zn^{65} to Co^{60} released from the reactors, and the relative amounts of each radioisotope deposited with time. Data are available to determine the $\text{Zn}^{65}/\text{Co}^{60}$ ratio on water released to the river. No specific data are available to show that the ratio of these two radioisotopes deposited changed with time, and so this ratio was assumed constant. It is hoped that indices to verify the times on some of the cores may be determined by the relationships of core sections to floods, reservoir filling, etc. when further samples are available.

In Figure 4 are plotted the ages of several sediments from McNary Dam reservoir with depth as determined by the $\text{Zn}^{65}/\text{Co}^{60}$ method. The sample taken 3/16/62 shows a relatively uniform rate of deposition of 2.3 inches per year. The 8/10/61 samples show a rate of about 6 inches per year for the last 2 years, then at a rate of about 1.2 inches per year for almost 3 years, preceded by a rapid accumulation of 4 inches of sediment. By studying more cores, using other radioisotope ratios, and sectioning the cores in smaller segments it is hoped that more detailed, definitive determination of the sediment accumulation process may be made.

Zn^{65} AND Co^{60} IN TYPICAL SEDIMENT SAMPLE

FIGURE 4

SEDIMENT AGES AS DETERMINED BY THE Zn^{65}/Co^{60} DATING METHOD