

THE COLUMBIA RIVER
ENVIRONMENTAL MONITORING PROGRAM

by

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THE COLUMBIA RIVER ENVIRONMENTAL MONITORING PROGRAMI. INTRODUCTION

The Environmental Studies and Evaluation Operation is responsible for a program of detecting, measuring, and evaluating the radioactive materials released to the environs by the Hanford plant in waste process effluents. One area of primary interest is the potential effect of these waste effluents on the Columbia River and the resultant radiation dose received by people using the river. Through many and varied samples and measurement devices the radiation dose to individuals is estimated. Radiation dose may be received directly from using the river for a water supply or for recreation such as swimming, boating, and fishing, or indirectly from eating produce irrigated with river water, animals grazing on river irrigated fields, fish from the river, or waterfowl residing on or about the river. More people receive exposure from the use of the river for drinking water than from any other environmental source affected by Hanford effluents. Approximately 25,000 of the 80,000 people living in the region around the plant perimeter use the river directly for their water supply and in the near future Richland, with a population of about 25,000, will also install facilities to use Columbia River water.

The present river monitoring program is designed to measure the probable radiation dose to individuals by sampling at several locations including those points where water is used for consumption by large communities. Analyses are made to determine the isotopes contributing approximately 95 percent of the dose. Similar periodic samplings are

made of fish, waterfowl and crops to determine their potential contribution to the total radiation dose from Hanford-created radioisotopes.

II. DEVELOPMENT OF RIVER MONITORING PROGRAM

In May 1945, soon after start-up of the original three Hanford reactors, sampling of Columbia River water was initiated. The original monitoring program was primarily one of detection of reactor-created radioisotopes rather than dose evaluation. Studies⁽¹⁾ at the time of reactor start-up in 1944 determined the radioactive composition of the cooling water effluent released to the river. At that time the main radioisotopes found in order of increasing half-life were: N-16, O-19, Al-28, Mg-27, Cl-38, Ca-49, Al-41, Mn-56, Si-31, Ca-47, Na-24, Cu-64, Zn-65, P-32, Cr-51, and Fe-59. Since limits on the quantities of radionuclides that could be released to the river were not established at that time, an immersion dose of 100 mrep per 24 hours at point of release was adopted as the limiting value.^(2, 3) Review of dose rates during the first several months of operation showed an average of 2.35 mrep per hour at the basin outlets.

Dose rate measurements were taken along the river as far downstream as Pasco, but a short distance away from the reactor effluent outfalls in the river the radiation level had already diminished to ≤ 0.01 mr/hour. However, the need for a long-range environmental monitoring program to detect and measure the quantity of radioactive materials in the river was recognized at that early time and sampling was started.

Originally, sampling locations were established at 181-B, D, and F, Hanford, 300 Area, and Richland.⁽⁴⁾ Later, samples were also collected from both banks and the middle of the river at Hanford, and upstream

from 100-B Area. In late 1947, sampling was initiated near the vehicle and railroad bridges at Pasco, and occasionally river water samples were collected as far downstream as The Dalles and Portland.⁽⁵⁾ Analyses of samples during these early years were made by evaporating 500 mls (some very early samples used only 100 mls) and counting the residue for total beta and alpha activity with a thin-window counter and a standard alpha counter. The detection limit was quoted as being approximately 5×10^{-5} $\mu\text{c/l}$ beta activity and 2 d/m/l alpha activity.⁽⁴⁾

During the early 1950's, comprehensive surveys⁽⁶⁾ were conducted to determine the channeling of reactor effluents and horizontal and vertical mixing. Both gross beta and temperature measurements were used to define the patterns.

By 1957, river monitoring had developed into a comprehensive program of sampling water at several locations along the river below Priest Rapids, and sampling of foodstuffs potentially affected by the river. Also, the advent of gamma energy spectrometric equipment made it more practical to obtain isotopic analyses of the water samples on a routine basis.

The surveillance program continued to grow into one which supplied a large amount of data with emphasis placed more and more on isotopic analysis. During the past two years emphasis has been placed on streamlining the program to obtain an optimum amount and type of data for efficient evaluation of the contribution of Hanford effluents to the radiation dose received by persons living in the vicinity.

III. PRESENT RIVER WATER MONITORING PROGRAM

The river water monitoring program as presently carried on by the Radiation Protection Operation was initiated January 8, 1962. It may

be divided into several distinct parts: (1) raw river water; (2) measurement of immersion radiation dose; (3) sanitary water (water obtained from river for human consumption); and (4) miscellaneous samples for radionuclide content and chemical toxicity.

1. Raw River Water - There are six locations along the river where raw river water is collected routinely for analysis: Vernita Ferry (upstream from the Hanford Plant), Hanford, 300 Area, Richland, Pasco, and Vancouver. Samples from Vernita Ferry, Hanford, and Richland are collected by dipping water from near the riverbank. At the 300 Area, water is collected at the Automatic Columbia River Monitoring Station (ACRMS); at Pasco the raw river water is collected at the pumping station for the city water supply; at Vancouver the U. S. Public Health Service collects samples from the middle of the river at the Interstate Bridge and ships them to us for analysis. Figure 1 shows the geographical location for these sampling points. Volume and frequency of collection are dependent upon the analyses desired from that particular location. Table I shows the frequency of collection at the six sampling sites and the analyses performed.

Routine sampling of river water is carried out as an audit of the burden of radioactive materials added to the Columbia by the Hanford plant and as a basis for estimating the radiation dose to individuals using the river. The repetitive sampling also provides information on seasonal changes and long range trends attributable to modification in operating procedures. The background information accumulated by

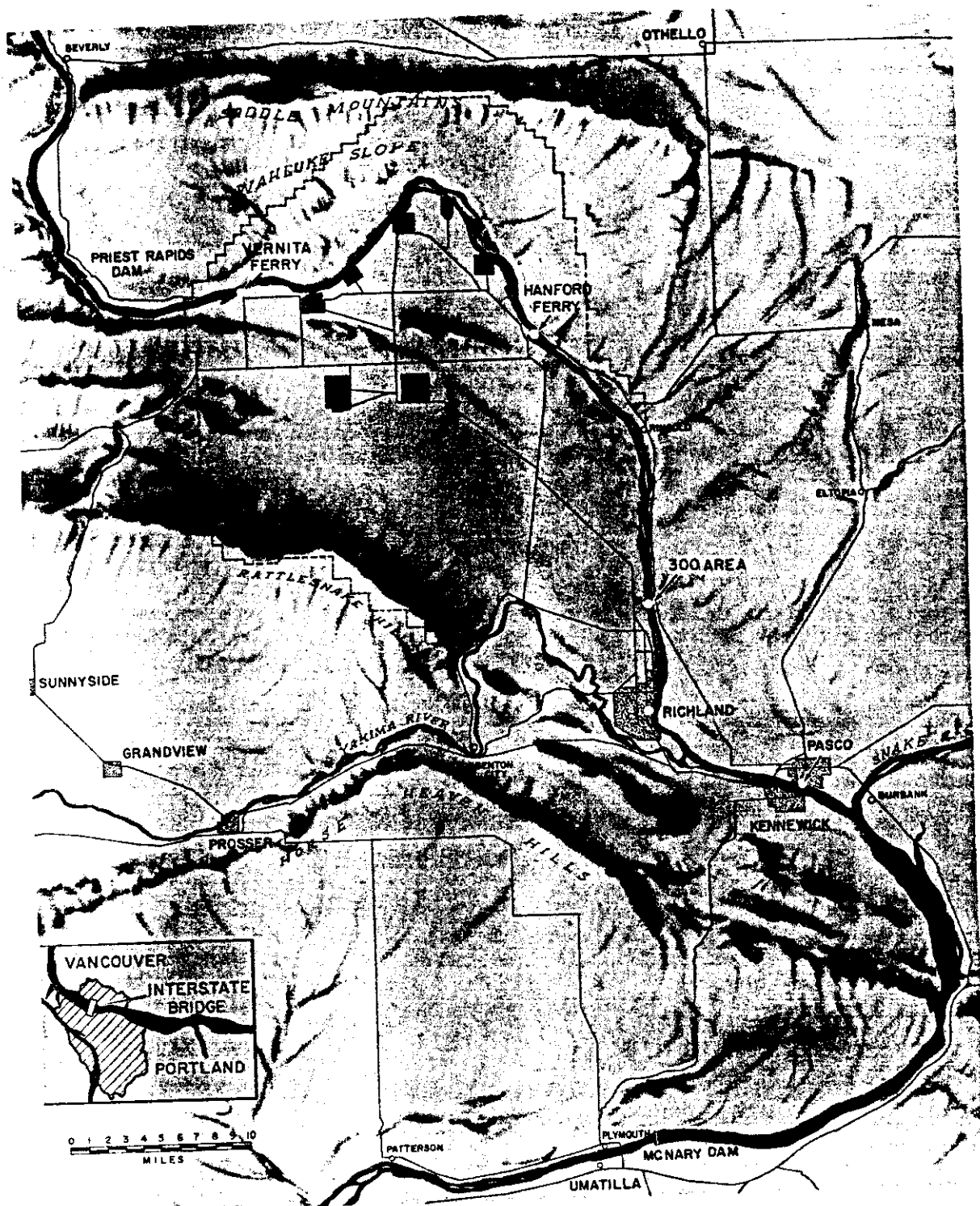


FIGURE 1

Columbia River Water Sampling Locations

TABLE I
TYPE AND FREQUENCY OF ANALYSES - RAW RIVER WATER

Sampling Location		Type of Analyses						
Total Beta	Total Alpha	U	Sr-90	Isotopic Analysis	Integrated Analysis	d/c Factor	Co ⁶⁰ ; Cs ¹³⁷ Ra ²²⁶ , Th ^{nat}	Chemical Analysis
Vernita Ferry	Biwkly	Biwkly	Biwkly	-	-	-	Qtrly	Biwkly
Hanford	Biwkly	Biwkly	-	Biwkly	-	Qtrly	-	Biwkly
ACRMS	Daily	Wkly	-	Wkly	Wkly	Qtrly	-	-
Richland	Wkly	-	-	Qtrly	-	Qtrly	-	-
Pasco	Wkly	-	-	Biwkly	Wkly	Qtrly	-	Wkly
Vancouver	Biwkly	-	-	Biwkly	-	Qtrly	-	-

Isotopic Analysis: As-76, Np-239, REY, Na-24, Cu-64, Ga-72, I-131, P-32, Zn-65, Cr-51, Zn-69, Sc-46, Mn-56, Sr-89 and 90.

Integrated Analysis: P-32, Zn-65, Sr-89 and 90, I-131, Fe-59, Ba-140.

d/c Factor: A weighted factor for converting counts per minute to disintegrations per minute. Determined from concentrations of individual radioisotopes and used for gross activity measurements.

Chemical Analysis: SO_4 , Mg, Fe, PO_4 , Cl, Cu, O_2 , Phth. Alk., M.O. Alk., Hardness, Ca, Solids, Cr^{+3} , and Cr^{+6} .

extensive sampling is also useful in appraising the effects which may result from abnormal operating conditions.

Sampling sites were chosen to provide data at strategic locations. The sample collected at Vernita Ferry provides background information on radionuclides present in the river prior to additions by the Hanford plant. The Hanford townsite is a convenient location for sampling the river just downstream from all of the reactor outfalls and permits detection of some of the short-lived nuclides. Although dispersion of radioactivity is incomplete at Hanford, samples taken at the shoreline are reasonably representative of the average concentration in the river at this location.

The 300 Area sampling location is essentially at the southern boundary of the Hanford Project, and the ACRMS provides continuous monitoring of the river water activity at this point. An emergency alarm is a part of the ACRMS System which would provide an immediate signal in the event radioactivity in the river reached an abnormal level. With such an alarm system several hour's notice is provided for action before any contaminated water reaches the Richland and Pasco pumping stations for domestic use. Isotopic analyses are made most frequently (weekly) on grab samples taken at this point and an integrated sample (composite from a continuous sample stream) provides a complete record of the longer lived nuclides of particular radiological interest.

The Richland sampling location was established to provide information at the site of the future water plant for the City of Richland. Pasco and Kennewick are, at the present time, the closest cities to

the Hanford plant which use Columbia River water for municipal supplies. The radioactive materials present in the water taken into their treatment facilities are, therefore, assayed frequently. The sampling point is the Pasco Pumping Station since this is more convenient than at Kennewick. In addition to grab samples taken weekly, a continuous sample is collected to provide an integrated sample. Vancouver is the furthestest point downstream where it is convenient to obtain samples without complications introduced by tidal action and the intrusion of sea water. Results from samples collected at this point are useful in estimating release of radionuclides to the ocean and loss of radioactivity in the river.

2. Immersion Dose - In addition to analyses of river water for radioactive content, immersion dose is measured directly at six locations in the river. Measurement locations are Vernita Ferry, Hanford Ferry (plant side), 300 Area (near PRTF intake), Richland Marina Dock, Columbia Park Marina Dock and the Pasco Pump House. Five gamma sensitive ionization chambers (Victoreen Model 622) are placed in a sealed plastic bottle and submerged two to five feet in the water. From these measurements the radiation dose to people using the river for recreational purposes such as fishing, swimming or boating may be estimated. These chambers are exchanged weekly and from the average of their readings on a HAPO pencil reader, an estimate of immersion dose is obtained. Figure 2 shows the location of immersion dose measurements.



FIGURE 2

Immersion Dose Measurement Locations

3. Chemical Characteristics - Chemical characteristics of the Columbia River water are determined in samples collected routinely above the plant at Vernita Ferry and below the reactors at Hanford to show the influences, if any, of the plant on these characteristics. Water used for reactor cooling is chemically treated to minimize corrosion and reduce the quantities of radioisotopes formed as it passes through the reactor. Sodium dichromate is added as a corrosion inhibitor. Concentrations of SO_4 , Mg, Fe, PO_4 , Cl, Cu, O_2 , Phenolphthaline Alkalinity, Methyl Orange Alkalinity, Hardness, Ca, Solids, Cr^{+3} , and Cr^{+6} are determined biweekly from one gallon samples collected at the river bank. One liter samples are also collected biweekly at Pasco for analysis of Cr^{+6} and Cr^{+3} . The 190-H Analytical Laboratory, Irradiation Processing Department, performs the analyses.

4. Sanitary Water - Water used for human consumption which originates from the Columbia River is sampled routinely at: 100-F, 200-W, Pasco and Kennewick (See Figure 3). The specific sampling points are 100-F Garage and 108-F Building; 2719-W Building, 200-W Area; Water Plant, Pasco; and Animal Hospital, Avenue "C", Kennewick. 300 Area sanitary water is given the same analyses as other sanitary water samples although it is supplied from the City of Richland which draws water from wells recharged with water from the Yakima River. Plans are under way for the City of Richland, and thus the 300 Area, to obtain its water supply from the Columbia



FIGURE 3

Sanitary Water Sampling Locations

River. Table II shows the frequency of sanitary water samples collected and the analyses performed.

The radioisotopic content of sanitary water is one factor used for estimating the potential radiation exposure of individuals from Hanford effluents released to the river. Results from 100-F samples are used to estimate dose to 100 Area personnel; however, since 100-F is the last production reactor downstream, these results tend to maximize the exposure. The 200-W Area samples provide an estimate of the radiation exposure potential in the 200 Areas. Sampling of Pasco and Kennewick sanitary water is the basis for estimating the potential radiation exposure from drinking water for residents of these cities. Collection of samples at the water treatment plant in Pasco, however, tend to maximize this estimate since the effect of distribution time is eliminated. In Kennewick the samples are collected in a semi-commercial area where the effects of distribution time would tend to provide a more average exposure estimate.

Frequency of sampling at the various locations is established according to need for detecting fluctuations and evaluating their significance. Short term variations in activity measurements occur because of changes in river flow (dilution) and non-uniform releases of radioactive material to the river.

IV. FISH, WATERFOWL, AND FOODSTUFFS SAMPLING PROGRAM

As early as July, 1945, studies were initiated to determine the effects of reactor effluents on fish. (7, 8) The first samples of fish collected from the Columbia River for radioanalysis were obtained near an effluent outfall in the fall of 1945. Low concentrations of

TABLE II

TYPE AND FREQUENCY OF ANALYSES - SANITARY WATERSampling Location

	<u>Type of Analyses</u>			
	<u>Total Beta</u>	<u>Total Alpha</u>	<u>Isotopic Analysis</u>	<u>d/c Factor</u>
100-F Area	4/wk	-	Wkly	Qtrly
200-W Area	Wkly	-	Mo.	Qtrly
300 Area	Wkly	Mo.	-	-
Richland	Qtrly	-	-	-
Pasco	Daily	-	Wkly	Qtrly
Kennewick	Daily	-	Mo.	Qtrly

Isotopic Analysis: As-76, Np-239, RE+Y, Na-24, Cu-64, Ga-72,
I-131, P-32, Zn-65, Cr-51, Zn-69, Sc-46,
Mn-56, Sr-89 and 90.

d/c Factor: A weighted factor for converting counts per minute
to disintegrations per minute. Determined from
concentration of individual radioisotopes and
used for gross activity measurements.

beta emitters were detected with P-32 being the major component. The following year a radiological study⁽⁹⁾ of river organisms confirmed that P-32 was the major radioisotope present. Continuing studies⁽¹⁰⁾ were made of filamentous algae and fish which showed seasonal cycles of radioisotopic concentration and significant differences between species. Studies of the radioisotopic content of the biota of the Columbia River, with emphasis on ecological relationships, have been continued in the Biology Operation through the years with varying intensity.^(11, 12, 13, 14)

Biological studies were also extended to waterfowl residing along the river within the project boundary.⁽¹⁵⁾ Concentration of P-32 and a few other radioisotopes in the flesh of some ducks was found to be similar to that in fish. There are, however, relatively few ducks residing permanently along the river so their population is highly diluted during the hunting season with those migrating south for the winter. This potential source of radiation exposure was recognized soon after start-up of Hanford although sampling of fish and ducks was carried on primarily for biological studies. In 1961, fish and duck sampling became a part of the environmental monitoring program.

1. Fish - Sampling schedules call for the collection of representative samples of several types of fish from five general locations along the river each month (See Figure 4). The four major categories of fish sampled are: (a) whitefish, (b) minnows, (c) spiny-ray, and (d) miscellaneous fish such as steelhead, sturgeon, salmon, and trout. The five sampling locations are Priest Rapids, Hanford, Ringold-Richland,



FIGURE 4

Fish Sampling Areas

Burbank, and McNary Dam. A reasonable effort is made to collect at least five fish of each species for a sample; however, a lesser number will qualify if fishing conditions are poor. Whitefish are almost always caught on hook and line in the vicinity of Priest Rapids, Hanford, and the Ringold-Richland section. Minnows (mostly shiners of the genus Richardsonius but the sample may include young squawfish, chiselmouth, chub and other species) are caught with a small hand seine at Priest Rapids, Hanford, Ringold-Richland, Burbank and behind McNary Dam. Spiny-ray and catfish are gill-netted or caught with hook and line at the same locations. Samples of steelhead, sturgeon, salmon, and trout, etc., are brought in for analysis when caught; however, no effort is made to collect samples of these types of fish routinely. Fish sampling is carried out year around providing conditions permit. The Columbia River is open to fishing all year, but fishermen do not have access to the section of the river within the project boundary.

2. Waterfowl - Waterfowl are collected only during the legal hunting season for this area of Washington State. Collection is made monthly in two general sections of the river, shown in Figure 5; the one designated as "upper river" extends from 100-B to Hanford, and the one designated as "lower river" extends from Hanford to Richland. For sampling purposes, the ducks are grouped into four general classifications; River Ducks, Diving Ducks, Mergansers, and Coots. An attempt



FIGURE 5

Waterfowl Sampling Areas

is made to collect at least three or four but not more than ten from each group each month of the hunting season. Lesser Canadian geese are also collected in the "lower river" region when available. The edible flesh and head of each specimen is taken for radiochemical analysis.

3. Oysters - Several Hanford-produced radioisotopes are sufficiently long lived that they persist to the ocean. Two of the isotopes, Zn-65 and P-32 concentrate in fish and other biota along the Coast of Oregon and Washington. The presence of Zn-65 in shellfish near the mouth of the Columbia River was first noted in 1957.⁽¹⁶⁾ Routine sampling of Willapa Bay oysters was initiated in 1961 with samples obtained from a market in Portland, Oregon. Early in 1962, arrangements were made to have two pounds of oysters shipped from the growing beds at Willapa Bay every two weeks for analysis.

4. Other Foodstuffs - Other foodstuffs produced locally, such as fruit and vegetables, are sampled in season from three areas (Ringold, Riverview, and Finley) along the river where some irrigation is done with river water.

V. ANALYTICAL PROCEDURES

1. Total Beta Analysis - Volumes of 50 or 500 mls per sample are used for river and sanitary water samples depending upon the radioactivity observed in previous samples from that particular location. Analysis of Vancouver and Vernita Ferry river water and Kennewick sanitary water normally

requires 500 mls while most other water samples usually require only 50 mls (see Tables I and II).

Analyses are performed by boiling down the water samples and plating the residue on one-inch stainless steel counting dishes. A gas flow proportional beta counter operated in the Geiger region is used to measure sample activity. Total beta results for water samples are reported in units of microcuries per liter.

The Biology Operation, 100-F Area performs the beta analysis on three of the four sanitary water samples collected from that area per week. The fourth sample is analyzed by the 300 Area Radiological Analysis Group, HLO. Raw counting data from samples analyzed at 100-F Area are forwarded to the HLO laboratory for calculation of results.

Large volume (12 gallons) samples collected weekly or biweekly for isotopic analyses are obtained in four three-gallon containers. Total beta analyses are run in sets of two on samples taken from each container. Any discrepancy in results requires an isotopic analyses performed on samples from each container to determine the cause.

2. Uranium Analysis - All analyses of water for uranium content are made with a fluorophotometer using standard techniques. Usually a 100 milliliter volume is boiled down to ten milliliters from which a 100 lambda aliquot is taken for analysis. Spikes of standard uranium solution are run with each group

of samples. If excess salts are present in the water an alpha extraction is performed prior to fluorophotometric treatment. Results are reported in units of micrograms per liter or microcuries per milliliter.

3. Isotopic Analyses - Large volume (9 and 12-gallon) water samples collected from locations listed in Tables I and II are routinely analyzed for the following radioisotopes: As-76, Np-239, RE+Y, Na-24, Ga-72, I-131, P-32, Zn-65, Sc-46, Mn-56, Sr-89 and Sr-90.

On the day the sample is received in the laboratory, analysis is started immediately for Mn-56 (2.58 hour $T_{1/2}$) using a one liter portion. The Mn, usually present as MnO_4 , is reduced by heating in an acidic solution to MnO_2 , the residue is filtered, washed and counted on a 400 channel analyzer for principal energy of .85 Mev. Analysis for Si-31 is initiated by making a partial distillation in the presence of HCl to remove impurities and is taken down to near dryness. The residue is taken up with HCl and boiled until clear. Si-31, remaining as an insoluble residue, is collected by centrifuging for plating and counting with a beta proportional counter. Decay of the final product is plotted.

A gross rare earth (RE) determination is made by boiling down a two-liter portion to a small volume (~ 100 mls) and making hydroxide and fluoride precipitations. The precipitate is taken up in HCl and re-precipitated with oxalic acid as

the oxalate, ignited to the oxide, plated, and counted in the beta proportional counter. Decay of the final product is plotted. A previously determined disintegration per count factor obtained from previous RE determinations is used to calculate the RE fraction.

Analyses of Cu-64 (12.9 hour $T_{1/2}$) and Na-24 (15 hour $T_{1/2}$) are performed by boiling down a two-gallon sample to which HNO_3 has been added. The residue is plated for counting. Na-24 is counted on a 400 channel analyzer for the 2.75 Mev peak using a 3-inch NaI crystal. Cu-64 is determined by gamma-gamma coincidence counting measurements of the annihilation photons of 0.51 Mev produced by positron emission.

Analyses for Zn-69 (14 hour $T_{1/2}$) and Ga-72 (14.1 hour $T_{1/2}$) are also performed on the day the sample is received. A four-liter portion is obtained and a phosphate precipitate is made which is then dissolved in HCl. Gallium is extracted from the 6N HCl solution and into an organic phase with isopropyl ether. Concentrated HCl is added to the aqueous phase containing Zn and is run through an anion resin column where it stays as a Zn Cl anion. The column is rinsed with 2N HCl and then the Zn is eluted with .05M HCl. A phosphate precipitation is made which is heated to the pyrophosphate. The residue is counted on a 400 channel analyzer. Gallium is back-extracted from the organic phase with water, scavanged with NaOH for Fe and other contaminants, precipitated with NaOH as the hydroxide after proper adjustment of pH, ignited to the oxide, plated, and counted on a 400 channel analyzer.

As-76 (26.5 hour $T_{1/2}$) analysis requires 1000 mls of sample to which 2 mls of As (10 mg/ml) carrier is added. The volume is reduced by heating to approximately 20 mls and then one gram of NH_3I is added to ensure reduction of As^{+5} to As^{+3} . Extraction of As in benzene is accomplished and then it is back-extracted into water. The aqueous phase is divided equally in two 100 ml centrifuge cones to which 30 mls of concentrated HCl and a small amount of $NaHPO_3$ is added. Each cone is heated in a hot water bath until the As precipitates and then the cones are centrifuged for approximately ten minutes. The supernatant is discarded and the precipitates are combined for washing with water and acetone. Discard the supernatant after each wash. The precipitate is finally transferred to a one-inch dish and dried for counting.

Two or three days after receiving samples, counting plates previously used for Na-24 and Cu-64 are again counted with the 400 channel analyzer for Cr-51 (27.8 day $T_{1/2}$) and Np-239 (2.35 day $T_{1/2}$) using a three-inch NaI crystal. Counting results are sent to data processing where the Cr-51 and Np-239 content is calculated after correcting for the complex gamma spectra present. At this time any other isotopes of significance are investigated.

A one-liter portion is used for P-32 determination by boiling down to approximately 100 mls with HNO_3 and filtering the concentrate. A phosphomolybdate precipitation is made

and then the precipitate is dissolved in NH_3OH . The phosphate is re-precipitated as $\text{Mg}(\text{NH}_3)_2\text{PO}_4$, plated and counted on a beta proportional counter. The sample is run on decay for approximately two weeks.

Iodine-131 concentration is determined using a three-liter portion which is first reduced in volume to 200-500 ml by boiling with sodium hydroxide and sodium sulfite to reduce iodine to iodides. The concentrate is then filtered and the iodides contained in the filtrate are oxidized back to the iodine in the presence of NaNO_2 . The iodine is extracted with CCl_4 . Back-extraction into water is made by reducing with sodium sulfite and the iodide is precipitated as AgI . The precipitate is plated, counted on a low background beta proportional counter, and run on decay.

Approximately two weeks after receiving the sample, plates originally used for Na-24 and Cu-64 are used for Zn-65 (245 day $T_{1/2}$) and Sc-46 (84 day $T_{1/2}$) analyses. The Sc-46 determination is made by counting 0.89 and 1.12 Mev photons by coincidence counting on a single channel analyzer using five-inch and three-inch NaI crystals and a Sc-46 reference sample for adjustment. Zn-65 is counted with a five-inch NaI crystal and results from the Sc count are used for correction.

A ten-liter portion, boiled down with HNO_3 to 2-3 liters, is used for Sr-89 and 90 determinations. Precipitations

with NaOH, NaCO_3 , and fuming HNO_3 are made, followed by an Fe scavage and Ba scavage. Strontium precipitated as the carbonate is plated and counted on decay for two weeks. A yttrium separation is then made by dissolving the plated residue in HNO_3 and making three hydroxide precipitations and dissolutions using NaOH, NH_3OH , and HNO_3 . Yttrium is precipitated as the oxalate with oxalic acid and then is ignited to the oxide, plated and counted on decay in a low background proportional counter.

4. d/c Factor - A three-gallon sample is routinely collected at several locations along the river to determine the d/c factor (disintegrations per count) for use with the total beta determinations. All of the isotopes routinely analyzed for, and any additional ones detected on gamma scans, which make up at least 85 to 90 percent of the total count are used to establish the factor. The counts obtained from each isotope are converted to beta counts and are used to determine a gross d/c from a sample of known isotopic content.

5. Radium-226 - Standard techniques of co-precipitation with barium and lead sulfates are used with the addition of EDTA to complex the barium and lead. The radium is re-precipitated with barium, plated, flamed and counted in an alpha scintillation counter.

6. Thorium - Natural - A one liter portion is used for analysis to which Th-234 is added for an internal spike.

A La carrier and concentrated HNO_3 is added prior to

centrifuging in water. Concentrated HF is added and the solution is evaporated with HClO_4 . The precipitate is taken up with HNO_3 and the Th is extracted in TTA and benzene. A small aliquot is taken for counting to check the yield. Back-extraction is made with 2N HNO_3 , evaporated and H_2SO_4 is added. A 100 lambda aliquot is taken in a quartz ampoule for irradiation in a reactor where the Th-232 is transformed to Th-233 and decays to Pa-233. After irradiation, Pa-231 is added to a 25 lambda aliquot as an internal standard and then the Pa-231 and Pa-233 is taken up in di-isobutyl carbonol-benzene (DIBC-B). The aqueous phase is discarded after again adding DIBC-B to clean up any remaining Pa. H_2SO_4 and HF are added to the organic phase and an aliquot is taken for plating and counting. Pa-233 is counted for beta (.26 Mev) and gamma (.31 Mev). Pa-231 is counted for alpha as an internal standard.

7. Analytical Procedures for Foodstuffs - The edible flesh of fish is packed in 9 oz. jars, counted in a nine-inch well crystal and the gamma scans are calculated for all detectable gamma emitters which usually include Zn-65, Sc-46, As-76, Cr-51, Np-239, Cs-137, Co-58, Co-60, Mn-54, and Mn-56. For total beta activity, the flesh is digested in HNO_3 , ashed and counted in a beta proportional counter. Analysis for P-32 is accomplished by digesting the flesh in HNO_3 , filtering the concentrate and then following the previously described procedure used in analysis of water. Sr-89 and 90

analyses are also identical to those outlined for water after completing digestion of the flesh or bony tissue in HNO_3 . In the case of small minnows, the entire fish is used for analysis rather than just the flesh or some other tissue.

Waterfowl collected from the river region are analyzed for total beta activity, P-32, Sr-89 and 90, and gamma emitters. The edible flesh and heads are digested in HNO_3 and analyses are carried out for each radioisotope as previously described for water.

Five-hundred grams of meat from Willapa Bay oysters are packed in a counting jar and counted in a 9-inch well crystal for gamma emitters. Any significant gamma emitter is reported by the laboratory. Another 500 grams is dehydrated, ashed and analyzed specifically for Zn-65, P-32, Sr-89 and 90 by procedures previously outlined.

Samples of fruits and vegetables are given a general gamma scan in a 9-inch well crystal with particular attention being paid to the I-131 content. Analyses are also made specifically for P-32 and Sr-89 and 90 by previously mentioned procedures after digestion in HNO_3 .

VI. DETECTION LEVELS

Listed below are the radioisotopes and their detection limits that are obtained with the previously described analytical procedures:

RADIOISOTOPIC DETECTION LIMITS

<u>Radioisotope</u>	<u>Detection Level</u> <u>uc/ml</u>	<u>Counting Method</u>
As-76	1.5×10^{-6}	Beta Proportional Counter
I-131	1.5×10^{-8}	Beta Proportional Counter and decay
Sc-46	2.4×10^{-8}	Gamma-Gamma Coincidence Counts
Na-24	1.5×10^{-7}	Gamma Spectrometry
Zn-69	6.5×10^{-8}	Gamma Spectrometry (assumes no interference)
Np-239	4.0×10^{-8}	Gamma Spectrometry (assumes no interference)
Cu-64	8.0×10^{-8}	Gamma-Gamma Coincidence Counts
Ga-72	1.4×10^{-7}	Gamma Spectrometry
Cr-51	1.1×10^{-7}	Gamma Spectrometry (assumes no interference)
Th(nat.)	1.4×10^{-9} (approx.)	Gamma Scintillation with Th-234 reference standard and neutron activation
Sr-90, Y-90	2.0×10^{-9}	Low Bkg. Beta Proportional Counter
Sr-89 and 90	1.2×10^{-9}	Low Bkg. Beta Proportional Counter
P-32	6.4×10^{-8}	Beta Proportional Counter
Ca-45	1.8×10^{-8}	Beta Proportional Counter
RE+Y	5.6×10^{-8}	Beta Proportional Counter and decay
Fe-59	1.5×10^{-7}	Beta Proportional Counter and Gamma Spectrometry
Si-31	6.0×10^{-7}	Beta Proportional Counter and decay
Mn-56	5.8×10^{-7}	Gamma Spectrometry
Ba-140	5.7×10^{-9}	Beta Proportional Counter and decay (checked by gamma spectrometry)

There are many things that affect the detection limit for a particular radioisotope. Some of these factors are volume of sample used, chemical yield possible from analytical procedure, sensitivity of counting instruments and amount of interference encountered during gamma spectrometric measurements from other radioisotopes present. Detection levels reported by the analytical laboratory are quoted as having the precision of ± 25 percent at the 68 percent confidence

level. It is very probable that the precision is much better in a majority of cases, however, the varying interference from other radioisotopes and method of determining the detection level does not allow quoting a result more precisely. As-76 is an exception where it is possible to state the detection level as having the precision of ± 10 percent at the 95 percent confidence level. Detection levels are routinely determined from average yields, background count rates and the average d/c factor determined for a particular radioisotope.

VII. EFFLUENT MONITORS AT THE REACTORS

The Irradiation Processing Department maintains a reactor effluent water monitoring program primarily as an operational aid. However, information from their program is useful in predicting the impact of the effluent on the environs particularly when operating irregularities occur.

Monitoring of effluent water as it leaves the reactor is accomplished by directing a small stream of effluent from each reactor crossheader to a flow-through monitoring chamber. The activity from each header can be checked every six minutes, and is used primarily for detection of fuel element ruptures. Back-up instrumentation for rupture detection consists of radiation monitoring devices located at each of the effluent retention basins or on the lines to the basins. The type and location of back-up instrumentation used by each reactor is listed below:

105-B	GM tube on effluent line
105-C	HM chamber at basin inlet
105-D	HM chamber at basin inlet
105-DR	HM chamber at basin inlet
105-H	HM chamber at basin inlet
105-F	HM chamber on effluent line
105-KE	None
105-KW	None

There are GM tube type monitors, connected to count rate meters installed at the outlet of the 105-B and 105-C reactor basins. The remaining retention basins have HM chambers installed at the outlet. Signals from these monitors are recorded continuously at each of the respective reactor buildings where they may be observed at anytime.

Each day, Monday through Friday, one gallon samples of reactor effluent are collected from both the near and far side risers and these samples are analyzed for As-76 and P-32. The samples collected on Monday are also analyzed for total beta activity, Na-24, Mn-56, Np-239, Zn-65, Sc-46, and Cr-51. Once each month samples from both risers are combined and analyzed for Cu-64, I-131, I-133, Ga-72, Zr-95, and Co-60. A monthly composite sample from all reactors is also collected with volume contribution prorated according to flow through the reactor and is analyzed for Sr-89 and 90.

Special analyses are performed whenever a ruptured fuel element is evident as indicated by the routine monitoring program. Effluent from the suspected crossheader is analyzed for Ru-103 and 106, and Ce-141 and 144. Whenever the reactor is purged, analyses for As-76, P-32, Cr-51, Np-239, Zn-65, and Sc-46 are performed.

Samples collected by IPD are analyzed by the Purex Analytical Laboratory in 200-East Area and the results are reported by the Radiological Engineering group, IPD.

VIII. REPORTS ON THE STATUS OF THE COLUMBIA RIVER

Several reports are prepared routinely each year by Environmental Studies and Evaluations Operation that contain extensive information regarding the radiological status of the Columbia River. These reports

are prepared on a monthly, quarterly, and annual basis with generally wide distribution to interested organizations external to the Hanford plant. Monthly reports⁽¹⁷⁾ are titled "Radiological Status of the Hanford Environs"; comprehensive reports titled "Evaluation of Radiological Conditions in the Vicinity of Hanford" are published quarterly⁽¹⁸⁾ and annually.⁽¹⁹⁾

BIBLIOGRAPHY

1. HW-36862, Influence of Hanford Reactors on Domestic Use of the Columbia River, J. W. Healy, R. E. Rostenback, May 25, 1955, (Unclassified).
2. HW-23456, Permissible Limits - Release of Reactor Effluents to the Columbia River, H. M. Parker, May 6, 1952.
3. HW-7-2346, Status of Problems of Measurement of the Activity of Waste Water Returned to the Columbia River, H. M. Parker, September 11, 1945.
4. HW-9871, Radioactive Contamination in the Columbia River, and in the Air and Radiation Levels Measured in the Air at Hanford Works and Vicinity for 1945-46-47 and Early 1948, W. Singlevich, May 24, 1948.
5. HW-7-5042, H. I. Environs Report, Semi-Monthly, Sept. 1946, to May 1947, C. C. Gamertsfelder.
6. HW-32506, Columbia River Survey 1951-1952-1953, J. Honstead, July 21, 1954, (Secret).
7. HW-3-3442, Accumulation of Radioactive Elements in Fish Immersed in Pile Effluent Water, J. W. Healy, 1947, (Secret).
8. HW-7-4759, Some Effects of Pile Area Effluent Water on Young Chinook Salmon and Steelhead Trout, R. F. Foster, 1946, (Confidential).
9. HW-3-5501, Radioactivity in Various Species of Fish From the Columbia and Yakima Rivers, K. E. Herde, 1947, (Secret).
10. HW-11344, A One-Year Study of the Radioactivity in Columbia River Fish, K. E. Herde, 1948, (Secret).
11. HW-36074, Radiological Studies of the Columbia River Through December, 1955, J. J. Davis, D. G. Watson, C. C. Palmiter, 1956.

12. HW-11662, The Accumulation of Radioactivity as Shown by a Limnological Study of the Columbia River in the Vicinity of Hanford Works, R. W. Coopey, November 12, 1948.
13. HW-23093, Accumulation of Radioactivity in Columbia River Fish in the Vicinity of the Hanford Works, P. A. Olson and R. F. Foster, July 1, 1952, (Secret).
14. HW-48523, Concentrations of Radioisotopes in Columbia River Whitefish in the Vicinity of the Hanford Atomic Products Operation, D. G. Watson and J. J. Davis, February 18, 1957, (Secret).
15. HW-59500, Radioactive Contamination in Wildlife, Hanford Biology Research-Annual Report for 1958, pp. 138-143, W. C. Hanson and J. J. Davis, January 5, 1959.
16. Zinc-65 in Marine Organisms Along the Oregon and Washington Coasts, Science, Vol. 133, pp. 1826-1828, June 9, 1961, D. G. Watson, J. J. Davis, and W. C. Hanson.
17. HW-74307-9, Radiological Status of the Hanford Environs for September, 1962, R. F. Foster, October 12, 1962.
18. HW-74398, Evaluation of Radiological Conditions in the Vicinity of Hanford April-June, 1962, Environmental Studies and Evaluation Staff, July 23, 1962.
19. HW-71999, Evaluation of Radiological Conditions in the Vicinity of Hanford for 1961, Environmental Studies and Evaluation Staff, March 1, 1962.