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A STUDY OF POLLUTANT DISCHARGES
FROM REACTOR OPERATIONS UTILIZING
ULTRACENTRIFUGATION TECHNIQUES

AUTHOR:

J. W. Amburgey, Jr.

UNION CARBIDE CORPORATION
NUCLEAR DIVISION

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A STUDY OF POLLUTANT DISCHARGES FROM REACTOR OPERATIONS
UTILIZING ULTRACENTRIFUGATION TECHNIQUES

J. W. Amburgey, Jr.
Biophysical Limnology Laboratory
Separation Systems Division

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ABSTRACT

Reactor cooling water samples from the Hanford Operation at Richland, Washington, have undergone particulate separations and study utilizing recently developed ultracentrifugation techniques. The suspended particles contained in these samples were separated into classified groups and then subjected to a variety of analytical investigations including elemental and radiochemical analyses. The samples selected for study were taken in sets ranging from one to four samples per set. Six sets or a total of sixteen samples were collected and shipped to the Biophysical Limnology Laboratory at Oak Ridge for study. The analytical results from these investigations are summarized in a series of tables. The content of selected elements and the radioactivity for the more prominent radionuclides are shown as associated with either the particulate or the dissolved fractions contained in the original sample volume.

A few of the radionuclide parent elements such as chromium, sulphur, and zinc were found almost entirely associated with the dissolved materials fraction. These results along with the other data showed that the suspended materials isolated from the various samples of reactor cooling water did not add significantly to the production of radionuclides and the radioactive pollution of the Columbia River at Hanford.

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A STUDY OF POLLUTANT DISCHARGES FROM REACTOR OPERATIONS UTILIZING ULTRACENTRIFUGATION TECHNIQUES

INTRODUCTION

In 1965 a proposal was submitted to the Atomic Energy Commission suggesting that the Technical Division of the Oak Ridge Gaseous Diffusion Plant investigate the potential application of ultracentrifugation techniques to separating particulate materials from natural water and for promoting studies relating to water pollution and control. Similar separating techniques already were being successfully applied to biological systems and the study of cellular components. The Atomic Energy Commission accepted the proposal and provided the necessary funding beginning with the second quarter of fiscal year 1966. One of the first investigations planned under the new program included a study of reactor effluent pollutants discharged into the Columbia River from the Hanford Operations at Richland, Washington.

Representatives from Hanford and Oak Ridge met on two occasions to exchange information and to plan the investigation of reactor cooling water utilizing the latest developments in liquid centrifuge systems and separation techniques. The first meeting was held at Oak Ridge, July 29, 1965. At this meeting, Hanford representatives discussed the production of radionuclides in operating nuclear power reactors and some problems associated with the discharge of such contaminants in reactor effluent cooling water. A second meeting was held at Richland, Washington, July 12, 1967, to review data previously determined and to plan additional experiments.

The scheme established for studying reactor cooling water was divided into two phases: one, the suspended materials contained in samples of cooling water would be separated centrifugally into various particulate fractions; and two, the isolated fractions would undergo selective analytical studies. The intent of the investigation was to obtain reliable analytical data on the waterborne particulate materials and to use this information to help explain the role of these materials in the radioactive contamination of reactor effluent cooling water. The colloidal fractions were of principal concern since very little information had been obtained on these materials and because of previous difficulties in isolating colloidal fractions.

This report presents a summary of all data obtained from the various analytical studies made on both particulate and dissolved fractions centrifugally separated from reactor influent and effluent cooling water samples.

SUMMARY AND CONCLUSIONS

During the first information meeting Hanford representatives described the mechanism of reactor effluent pollution, the problems associated

with such discharges into a natural body of water, and some treatments under consideration. Their presentations can be summarized as follows:

1. The reactor fuel elements tend to accumulate hydrous oxide films that readily sorb certain elements from the cooling water.
2. The sorbed elements are converted into radionuclides after exposure to the high neutron flux field.
3. The radionuclides are discharged in the reactor cooling water effluent stream, apparently resulting from the eventual sluffing-off of the hydrous oxide film.
4. At least four possible corrective approaches have been considered:
 - a. Additional pre-treatment of the inlet cooling water in an effort to reduce the concentration of radionuclide parent elements.
 - b. Treatment of the fuel element cases to inhibit the formation of the film.
 - c. Treatment of the oxide film to reduce its tendency to sorb the parent elements.
 - d. Treatment of the coolant effluent stream to remove the particulate associated radionuclides. Some treatments under consideration included settling basins, filtration, and possibly chemical additives.

The representatives from Hanford were told that the ultracentrifuge could separate and concentrate waterborne suspended materials and thereby permit more reliable analytical studies on individual particulate groups. By using this approach it was thought that a better understanding of the pollution problem could be achieved and hopefully would lead to an economical and practical solution.

A total of sixteen reactor cooling water samples of various sizes were collected and processed between July 1965 and November 1967. The first eight samples, ranging in size from one-half to one liter, were used in scoping-type experiments. The results from these preliminary experiments were extremely helpful in planning full scale fractionations on the larger succeeding samples. The larger sample volumes ranged from eighteen to thirty-eight liters.

The results obtained from the centrifugal fractionations and analytical studies did not reveal appreciable amounts of suspended particulate materials in the cooling water. No significant differences in elemental content were detected between reactor influent and effluent water. Therefore it is concluded that any treatment to be considered probably should involve the hydrous oxide film itself, since it is theorized that the dissolved or very small particulate material is sorbed by the film. Apparently some concentration balance is maintained through continual sorption and resolubilization phenomena taking place within the reactors.

Centrifugation has proven to be an excellent method for fractionating waterborne particulate materials and for promoting studies associated with various aspects of water pollution. It offers very few advantages in the study of dissolved materials. However, the analytical precision associated with the dissolved materials fraction is often enhanced as a result of the removal of the suspended fractions.

These studies utilizing centrifugal techniques have shown that settling basins and ultrafiltration systems, previously being considered, would not have been particularly effective in curbing radionuclide pollution since the radionuclides of principle concern were not found significantly associated with the particulates separated from the effluent cooling water.

Even though material balances were sometimes inconsistent, the revelation of very low particulate content and other analytical data obtained on the various fractions isolated from both influent and effluent cooling water has generated a better understanding of the contamination problems associated with waterborne particulate materials contained in reactor cooling water.

SYSTEMS

Centrifuge systems must meet two basic requirements before satisfactory separation of suspended materials from natural water can be achieved; one, the system must have sufficient capacities to concentrate the suspended materials from large sample volumes (normally up to 50 liters), and two, rotor dimensions and speeds must be sufficient to remove the ultrasmall colloidal particles. The rotor system selected and used for the reactor cooling water fractionations met these requirements. The rotor was approximately 25 cm long, 10 cm in diameter, and designed for a maximum speed of 40,000 rpm. The centrifugal force generated at this speed was $90,000 \times g$ at maximum radius. A Spinco Model L-4 drive system was used to spin the rotor system. Both aluminum and titanium rotors were used in the cooling water studies. The aluminum rotor was used in processing all samples taken prior to July 31, 1967. The titanium one was used on all later samples.

An aluminum rotor core designed for biological separations was modified specifically for separating and concentrating particulate materials contained in large sample volumes of natural water. A schematic diagram of the core and rotor system is shown in figure 1.

FRACTIONATING TECHNIQUES

In the basic separations the suspended particles were removed or concentrated to an approximate volume of 300 ml. The particles were separated into various particulate groups based on a difference in sedimentation rates for the sedimenting particles. The rotor speed and sample flow selected for removing each sample fraction were based on theoretical

Figure 1
SCHEMATIC DRAWING OF CENTRIFUGE CORE,
ROTOR SYSTEM

calculations shown in figure 2 (1) and depended on the particle sizes desired. During each centrifugation cycle, the sample passing through the spinning rotor was divided into two particulate fractions with the larger particles being retained in the centrifuge rotor and the smaller particles being carried out in the effluent stream. In this way the larger particles were concentrated from the original volume to approximately 300 ml. Further concentrations, using other centrifuge systems, were made in some of the tests.

The final effluent volumes containing some ultrasmall particles (estimated to be less than 150 Å in diameter for particles with a density of 2 g cm^{-3}) along with the dissolved materials were concentrated to approximately 300 ml by vacuum evaporation techniques and subjected to the same analytical studies as were the particulate fractions. A schematic of the vacuum evaporation equipment is shown in figure 3.

THEORETICAL CONSIDERATIONS

Figure 2 shows a plot of particle sedimentation coefficient as a function of rotor speed and sample flow. Sedimentation coefficients were approximated from the following equation (1):

$$s = \frac{d^2(\rho_p - \rho_s)}{18\eta}(10^{13})$$

where

s = sedimentation coefficient in Svedberg units,

d = diameter of the sedimenting particle in cm,

ρ_p = density of the sedimenting particle in g cm^{-3} ,

ρ_s = density of the supporting solution in g cm^{-3} , and

η = viscosity of the supporting solution in poise.

A flow rate was selected that would yield a particle fractionation based on its estimated sedimentation coefficient. The sample flow rate requirements were estimated from the following equation (1):

$$Q = \frac{\omega^2 V_s}{\ln R_2/R_1}(3.6)(10^{-13})$$

where

Q is the sample flow in liters per hour,

ω is the angular velocity in radians per second,

Figure 2

RELATIONSHIP BETWEEN SEDIMENTATION COEFFICIENT AND
FLOW RATE AT SEVERAL ROTOR SPEEDS

Figure 3
SCHEMATIC OF VACUUM EVAPORATION SYSTEM

V is the sample volume exchanged in liters,

s is the sedimentation coefficient in Svedberg units,

R_2 is the maximum radius in centimeters, and

R_1 is the minimum radius in centimeters.

EXPERIMENTAL PLAN AND RESULTS

Following the first information meeting, Mr. Robert Geier* and associates left four, one-pint samples of reactor cooling water for our use in some preliminary investigations. The samples represented both influent and effluent water from three different reactor areas. Another set of four, one-liter samples, was received on September 24, 1965. These first eight samples were used in scoping-type experiments. The results from these experiments were used in planning full-scale fractionations on the larger succeeding samples. The following general experimental plan was formulated:

1. Collect minimum sample volumes of nineteen liters.
2. Refrigerate samples (normally less than 5°C) immediately following collection and during transit to suppress biological and chemical activity.
3. Separate and divide the suspended materials into a series of fractions based on a difference in sedimentation rates.
4. Vacuum evaporate the final centrifugal effluent volume and analyze the resulting concentrated dissolved materials fraction.
5. Perform spectrochemical and atomic absorption analyses on each sample fraction.
6. Attempt to identify the larger materials by X-ray diffraction and electron probe techniques.
7. Examine the smaller materials using electron microscopy techniques.
8. Perform radiochemical analyses on all fractions derived from reactor effluent samples.

The analytical data obtained from the larger samples can best be summarized and correlated by combining the results from the different isolated fractions and expressing them all in the same units; preferably in micrograms and picocuries per milliliter in the original sample volume.

* Formerly with General Electric Corporation, currently with Douglas United Nuclear, Inc.

Table I shows miscellaneous data and information for each of the samples fractionated. Tables II, III, and IV show elemental analysis results for the original samples, the particulate fractions, and the dissolved fractions, respectively. Table V shows the percent elemental content associated with the particulate material and based on the total amount found in the two fractions. The elemental data on the original sample aliquots are only used for reference since the precision is often much less than that for the two concentrated fractions.

Table VI shows the radioactivity results associated with the particulate, dissolved, and original sample volumes from the reactor effluent streams. In table VII the results are shown as percent of original count and the amount unaccounted for based on a balance between the two concentrated fractions and the original sample. Some of the losses can be accounted for in that analyses on the original containers for samples HK-6 and -8 indicated that some losses occur as a result of sorbtion effects between the sample and container walls.

In looking at table V we find only iron and phosphorus consistently high in the particulate fractions. Aluminum results are somewhat inconsistent. This may be accounted for in that some of the centrifuge components are made of aluminum and are probable sources of contamination.

Radiochemical analyses (tables VI and VII) revealed that only phosphorus 32 and scandium 46 were significantly associated with the particulate materials. The other radionuclides; chromium 51, antimony 124, strontium 89-90, and zinc 65 were almost wholly associated with the dissolved materials.

The basic elemental and radioactivity data for each coolant sample studied are shown in the appendix. These data and other pertinent information were sent to R. G. Geier following the fractionation and analytical study made on each set of samples. He and his associates have made their own study and interpretations.

REFERENCE

1. Svedberg, T. and Pederson, K. O., The Ultracentrifuge, Oxford, The Clarendon Press, 1940.

ACKNOWLEDGMENTS

The author wishes to acknowledge the assistance of Dr. W. T. Lammers in planning the investigations and making particle counts on the original samples, and the help of the Chemical Development and Chemical Analysis Departments in making special analyses on the various sample fractions.

TABLE I
MISCELLANEOUS SAMPLE INFORMATION

No.	Sample Source	Taken	Received	pH	Conductivity Micromhos	Particle Count per ml			Sample Volume, ml	Total Solids, mg/ml*
						Bacteria	Organic	Inorganic		
H-1	100D Reactor Influent	1/16/67	1/18/67	--	758	2×10^6	1×10^7	9×10^7	19,000	--
H-2	100C Reactor Influent	3/18/67	3/20/67	7.4	140	--	--	--	19,000	0.13
H-3	100C Reactor Effluent	3/18/67	3/20/67	6.7	150	--	--	--	19,000	0.13
H-4	Columbia River	8/28/67	8/30/67	6.7	116	3×10^4	1.5×10^7	1.7×10^8	19,200	0.12
H-5	100K Reactor Influent	8/28/67	8/30/67	6.1	110	< 1	1.5×10^7	9×10^6	19,300	0.12
H-6	100K Reactor Effluent	8/28/67	8/30/67	5.7	164	< 1	2.5×10^6	1.7×10^8	19,000	0.09
H-7	100K Reactor Influent									
	Number 1	11/12/67	11/14/67	6.4	144	10	3×10^7	9×10^7	19,200	0.14
	Number 2	11/12/67	11/14/67	6.6	128	3×10^7	4×10^7	8×10^7	19,200	0.14
H-8	100K Reactor Effluent									
	Number 1	11/12/67	11/14/67	5.4	133	--	None	1.6×10^8	17,000	0.10
	Number 2	11/12/67	11/14/67	5.6	129	--	9×10^8	2.6×10^8	18,000	0.10

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* Solids determined by evaporating sample aliquot to dryness at 100°C.

TABLE II

ELEMENTAL CONCENTRATION IN STARTING SAMPLE, $\mu\text{g/ml}$

No.	Sample Source	Al	Ca*	Cl*	Cr*	Cu*	Fe*	Mg*	Mn	P*	S*	Si	U*	Zn*
H-1	100D Reactor Influent	--	--	--	--	--	--	--	--	--	--	--	--	--
H-2	100C Reactor Influent	0.04	19	1.6	0.02	0.01	0.02	5.0	0.008	0.07	19.0	0.4	0.010	0.30
H-3	100C Reactor Effluent	0.04	19	1.4	0.40	0.02	0.02	5.0	0.008	0.06	16.0	0.4	0.030	0.30
H-4	Columbia River	0.40	17	0.7	< 0.05	< 0.02	< 0.05	3.9	< 0.020	< 0.02	1.0	0.8	< 0.001	0.05 ¹⁰
H-5	100K Reactor Influent	0.20	17	1.5	< 0.05	< 0.02	< 0.05	4.1	< 0.020	0.06	3.0	1.0	< 0.001	< 0.05
H-6	100K Reactor Effluent	0.20	16	1.6	0.30	0.02	< 0.05	4.0	< 0.020	0.05	5.0	0.4	< 0.001	< 0.05
H-7	100K Reactor Influent	0.04	17	1.3	0.01	0.01	0.05	4.6	--	0.02	6.3	0.4	--	0.11
H-8	100K Reactor Effluent	0.03	18	1.1	0.31	0.02	0.01	4.6	0.003	0.01	6.3	0.4	< 0.001	0.20

* Normal precision $\pm 20\%$ of the reported values; all others 1/3 to 3 times this value.

TABLE III

ELEMENTAL CONCENTRATION IN PARTICULATE FRACTION, $\mu\text{g/ml}$

No.	Sample Source	Al	Ca*	Cl*	Cr*	Cu*	Fe*	Mg*	Mn	P*	S*	Si	U*	Zn*
H-1	100D Reactor Influent	0.05	0.31	--	0.002	0.002	0.080	0.110	0.001	0.019	0.390	0.036	--	0.010
H-2	100C Reactor Influent	0.16	0.02	--	0.004	0.045	0.110	0.009	--	1.700	0.350	0.021	0.001	0.063
H-3	100C Reactor Effluent	0.58	0.02	--	0.008	0.344	0.398	0.014	0.003	0.638	0.696	0.006	0.002	0.033
H-4	Columbia River	0.09	0.03	0.010	0.001	0.003	0.042	0.017	0.004	0.003	--	0.296	--	0.003 g
H-5	100K Reactor Influent	0.03	0.08	--	--	0.003	0.030	0.019	--	0.006	0.011	0.051	0.001	0.007
H-6	100K Reactor Effluent	0.05	0.06	0.004	0.001	0.016	0.013	0.015	--	0.004	--	0.020	0.001	0.004
H-7	100K Reactor Influent	0.01	0.14	0.022	--	0.001	0.073	0.037	--	0.003	0.056	0.017	0.000	0.003
H-8	100K Reactor Effluent	0.01	0.17	0.020	0.003	0.002	0.004	0.045	--	0.006	0.070	0.010	0.006	0.003

* Normal precision $\pm 20\%$ of the reported values; all others 1/3 to 3 times this value.

TABLE IV

ELEMENTAL CONCENTRATION IN DISSOLVED FRACTION, $\mu\text{g/ml}$

No.	Sample Source	Al	Ca*	Cl*	Cr*	Cu*	Fe*	Mg*	Mn	P*	S*	Si	U*	Zn*
H-1	100-D Reactor Influent	0.21	10.4	--	0.005	0.021	0.58	2.87	0.029	0.074	6.50	0.79	--	0.204
H-2	100-C Reactor Influent	1.14	11.8	1.60	0.011	2.060	0.44	3.08	0.042	6.550	11.15	1.12	0.006	0.460
H-3	100-C Reactor Effluent	0.13	13.7	1.80	0.250	0.350	0.22	3.37	0.066	2.340	11.37	1.18	0.001	0.360
H-4	Columbia River	0.07	13.4	7.90	0.003	0.007	0.027	3.14	0.004	0.020	2.05	0.82	--	0.026
H-5	100-K Reactor Influent	1.38	13.9	1.42	0.038	0.021	0.139	3.46	0.016	0.060	7.94	1.38	--	0.200
H-6	100-K Reactor Effluent	0.13	8.9	1.36	0.287	0.006	0.008	3.62	0.005	0.011	5.26	0.80	0.005	0.065
H-7	100-K Reactor Influent	0.10	14.3	3.00	0.009	0.008	0.013	3.91	0.001	0.008	7.94	1.25	0.000	0.090
H-8	100-K Reactor Effluent	0.09	14.6	0.92	0.317	0.020	0.008	3.50	0.004	0.014	7.77	1.15	0.000	0.058

* Normal precision $\pm 20\%$ of reported values; all others 1/3 to 3 times this value.

TABLE V

ELEMENTAL DISTRIBUTION - PERCENT ASSOCIATED WITH PARTICULATE FRACTION
 [Particulate (100)/Particulate + Dissolved]

<u>No.</u>	<u>Sample Source</u>	<u>Al</u>	<u>Ca</u>	<u>Cl</u>	<u>Cr</u>	<u>Cu</u>	<u>Fe</u>	<u>Mg</u>	<u>Mn</u>	<u>P</u>	<u>S</u>	<u>Si</u>	<u>U</u>	<u>Zn</u>
H-1	100-D Reactor Influent	19	2.9	--	28.6	9	12	3.7	3	20	6.0	4.4	--	4.7
H-2	100-C Reactor Influent	12	0.2	0.00	26.7	2	20	0.3	0	21	3.0	2.0	14	12.0
H-3	100-C Reactor Effluent	82	0.1	0.00	3.2	49	64	0.4	4	21	5.8	0.5	67	8.4
H-4	Columbia River	56	0.2	1.36	25.0	30	61	0.5	50	13	0.0	26.5	--	10.0
H-5	100-K Reactor Influent	2	0.6	0.00	0.0	13	18	0.5	0	9	0.1	3.6	100	3.2
H-6	100-K Reactor Effluent	28	0.7	0.29	0.3	73	62	0.4	0	27	0.0	20.0	17	5.8
H-7	100-K Reactor Influent	9	1.0	0.73	0.0	11	85	0.9	0	27	0.7	1.3	--	3.2
H-8	100-K Reactor Effluent	10	1.2	2.10	0.1	9	33	1.3	0	30	0.9	0.9	100	4.9

TABLE VI
RADIOACTIVITY ASSOCIATED WITH ORIGINAL SAMPLE AND ISOLATED FRACTIONS
PICOCURIES/ml**

No.	Source	Cr ⁵¹	P ³² *	Sb ¹²⁴	Sc ⁴⁵	Sr*	Zn ⁶⁵
<u>ORIGINAL SAMPLE</u>							
HK-6	100K Effluent	193 ± 14	6.50 ± 0.70	2.82 ± 2.11	3.60 ± 1.10	0.43 ± 0.06	4.80 ± 1.40
HK-8	100K Effluent	240 ± 15	7.40 ± 0.80	3.31 ± 2.13	4.20 ± 1.14	0.04 ± 0.06	5.10 ± 1.40
<u>PARTICULATE FRACTION</u>							
HK-6	100K Effluent	2.88 ± 0.04	0.90 ± 0.05	--	0.32 ± 0.01	--	0.07 ± 0.01
HK-8	100K Effluent	3.30 ± 0.26	2.15 ± 0.11	0.05 ± 0.02	0.52 ± 0.02	--	0.06 ± 0.01
<u>DISSOLVED FRACTION</u>							
HK-6	100K Effluent	178 ± 1.50	3.79 ± 0.20	2.62 ± 0.08	0.30 ± 0.02	0.04 ± 0.01	0.61 ± 0.02
HK-8	100K Effluent	219 ± 8.00	5.08 ± 0.22	1.11 ± 0.12	2.86 ± 0.12	--	4.50 ± 0.17

* Beta activity; all others gamma activity.

** Results corrected to date of sampling.

TABLE VII
 RADIOACTIVITY DISTRIBUTION
 (Percent of Total Activity in Original Sample)

<u>Sample Number and Source</u>		<u>Cr⁵¹</u>	<u>P³²*</u>	<u>Sb¹²⁴</u>	<u>Sc⁴⁶</u>	<u>Sr*</u>	<u>Zn⁶⁵</u>
<u>PARTICULATE FRACTION</u>							
HK-3	100C Reactor Effluent	2.1	14.7	--	8.0	0.4	6.0
HK-6	100K Reactor Effluent	1.5	13.8	--	8.9	--	1.5
HK-8	100K Reactor Effluent	1.4	29.1	1.5	12.4	--	1.2
<u>DISSOLVED FRACTION</u>							
HK-3	100C Reactor Effluent	64.5	9.7	--	23.8	78.8	77.5
HK-6	100K Reactor Effluent	92.2	58.3	--	8.3	--	12.7
HK-8	100K Reactor Effluent	91.3	68.7	33.6	68.2	--	88.3
<u>UNACCOUNTED FOR</u>							
HK-3	100C Reactor Effluent	33.4	75.6	--	68.2	20.8	16.5
HK-6	100K Reactor Effluent	6.3	27.9	--	82.8	--	85.8
HK-8	100K Reactor Effluent	7.3	2.2	64.9	19.4	--	10.5

* Beta activity; all others gamma activity.

APPENDIX

ELEMENTAL AND RADIOACTIVITY DISTRIBUTION DATA

Selected elemental determinations were made on each of the original sample volumes, the different isolated particulate fractions, and the final dissolved materials fraction. In addition, radiochemical analyses were performed on each of the fractions derived from samples of reactor effluent cooling water. These analytical results are shown in tables VIII through XVIII as either total elemental weight or radioactivity where applicable for the different coolant samples and isolated fractions.

TABLE VIII

ELEMENTAL DISTRIBUTION IN REACTOR 100D INFLUENT WATER (H-1)
(Total Weight in μg)

Sample Identification†	Al	Ca*	Cr	Cs*	Cu	Fe	K*	Mg*	Mn	Na	Ni	P*	S*	Si	Sr	Zn*
Particulate Fraction > 0.2 μ	103	410	--	205	6.0	62	62	250	--	--	--	62	410	123	--	45
Particulate Fraction > 0.04 $\leq$ 0.2 μ	140	1750	--	175	9.0	350	140	613	--	--	9.0	18	2975	103	7.0	54
Particulate Fraction > 0.01 $\leq$ 0.04 μ	675	3820	45	450	23	1125	450	1260	23	113	11	292	4050	450	23	101
Dissolved Fraction < 0.01 μ	4000	197000	100	29350	400	11000	11600	54500	550	--	1250	1398	123000	15000	657	3883

† Particle size estimates based on an average particle density of 2 g cm⁻³.

* Precision $\pm$ 20% of reported value; all other determinations 1/3 to 3 times this value.

TABLE IX
ELEMENTAL DISTRIBUTION IN REACTOR 100C INFLUENT WATER (H-2)
(Total Weight in μg)

Sample Identification†	Al	Ca*	Cl*	Cr*	Cu*	Fe*	K*	Mg*	Mn	Na*	Ni	P*	S*	Si	Sr	U*	Zn*
Original Sample	750	361000	--	380	190	380	22800	95000	152	57000	--	1330	361000	7600	--	190	5700
Particulate Fraction > 0.4 μ	920	21	--	11	299	390	45400	69	--	20800	23	16400	440	115	--	25	276
Particulate Fraction > 0.2 $\leq$ 0.4 μ	430	271	--	10	120	163	38200	43	--	17700	22	1720	4500	129	8	2	210
Particulate Fraction > 0.04 $\leq$ 0.2 μ	880	11	--	28	187	990	18920	22	--	9130	22	6960	240	44	--	1	363
Particulate Fraction > 0.01 $\leq$ 0.04 μ	800	32	--	20	240	576	13120	32	--	6288	24	7320	1500	120	--	2	344
Dissolved Fraction < 0.01 μ	21700	224000	30400	217	39150	8350	334000	58500	800	183200	1050	124400	212000	21200	1050	112	8750

† Particle size estimate based on an average particle density of 2 g cm⁻³.

* Precision $\pm$ 20% of reported value; all others 1/3 to 3 times this value.

TABLE X
ELEMENTAL DISTRIBUTION IN REACTOR 100C EFFLUENT WATER (H-3)
(Total Weight in μg)

Sample Identification†	Al	Ca*	Cl*	Cr*	Cu*	Fe*	K*	Mg*	Mn	Na*	Ni	P*	S*	Si	Sr	U*	Zn*
Original Sample	760	361000	26600	7600	380	380	17100	95000	152	57000	--	1140	304000	7600	380	570	5700
Particulate Fraction > 0.4 μ	246	225	--	--	106	74	65600	65	--	30700	--	7380	7200	24	--	9	20
Particulate Fraction > 0.2 $\leq$ 0.4 μ	1050	174	--	10	315	272	24200	88	4	13000	--	2100	2100	10	--	7	42
Particulate Fraction > 0.04 $\leq$ 0.2 μ	6300	4	--	67	4410	4830	21840	65	42	10920	6	1890	2730	42	--	14	399
Particulate Fraction > 0.01 $\leq$ 0.04 μ	3400	3	--	66	1700	2380	25500	57	17	13940	3	748	1200	34	--	6	160
Dissolved Fraction < 0.01 μ	2500	260000	34200	4680	6620	4100	170400	64000	1250	121900	250	44500	216000	22500	750	17	6790

† Particle size estimates based on an average particle density of 2 g cm⁻³.

* Precision $\pm$ 20% of reported value; all others 1/3 to 3 times this value.

TABLE XI
RADIOACTIVITY DISTRIBUTION IN REACTOR 100C EFFLUENT WATER (H-3)

Sample Identification†	(Percent of Total Activity Associated With Each Fraction and Based on an Accumulated Total)					
	Cr ⁵¹	P ³² *	Sb ¹²⁴	Sc ⁴⁶	Sr*	Zn ⁶⁵
Particulate Fraction > 0.4 μ	1.8	25.0	-	15.0	0.2	2.1
Particulate Fraction > 0.2 $\leq$ 0.4 μ	0.2	31.0	-	1.5	0.2	0.9
Particulate Fraction > 0.04 $\leq$ 0.2 μ	0.8	4.1	-	6.2	--	3.0
Particulate Fraction > 0.01 $\leq$ 0.04 μ	0.5	0.8	-	1.9	--	1.1
Dissolved Fraction < 0.01 μ	96.7	39.1	-	75.4	99.6	92.9

† Particle size estimates based on an average particle density of 2 g cm⁻³.

* Beta activity; all others gamma activity.

TABLE XII

ELEMENTAL DISTRIBUTION IN COLUMBIA RIVER WATER (H-4)
(Total Weight in μg)

<u>Sample Identification</u> †	<u>Al</u>	<u>Ca*</u>	<u>Cl*</u>	<u>Cr*</u>	<u>Cu*</u>	<u>Fe*</u>	<u>Mg*</u>	<u>Mn</u>	<u>P*</u>	<u>S*</u>	<u>Si</u>	<u>U*</u>	<u>Zn*</u>
Original Sample	7600	323000	13300	--	--	--	74100	--	--	19000	15200	-	950
Particulate Fraction > 0.4 μ	1400	231	77	14	14	700	245	70	3	--	5110	2	42
Particulate Fraction > 0.01 $\leq$ 0.4 μ	260	286	117	5	52	104	78	4	48	--	520	1	16
Dissolved Fraction < 0.01 μ	1300	254800	150800	52	130	520	59800	78	390	39000	15600	4	468

† Particle size estimate based on an average particle density of 2 g cm^{-3} .

* Precision $\pm 20\%$ of reported value; all others 1/3 to 3 times this value.

TABLE XIII

ELEMENTAL DISTRIBUTION IN REACTOR 100K INFLUENT WATER (H-5)
(Total Weight in μg)

<u>Sample Identification</u> [†]	<u>Al</u>	<u>Ca*</u>	<u>Cl*</u>	<u>Cr*</u>	<u>Cu*</u>	<u>Fe*</u>	<u>Mg*</u>	<u>Mn</u>	<u>P*</u>	<u>S*</u>	<u>Si</u>	<u>U*</u>	<u>Zn*</u>
Original Sample	3280	324700	28650	---	---	--	78310	---	1146	57300	19100	--	--
Particulate Fraction > 0.4 μ	280	1260	--	---	21	399	301	3	70	210	70	2	112
Particulate Fraction > 0.01 $\leq$ 0.4 μ	300	195	--	6	35	165	53	3	48	--	900	8	23
Dissolved Fraction < 0.01 μ	26400	256640	27060	726	396	2640	66000	284	1122	151800	26400	9	3762

[†] Particle size estimate based on an average particle density of 2 g cm⁻³.

* Precision $\pm 20\%$ of reported value; all others 1/3 to 3 times this value.

TABLE XIV

ELEMENTAL DISTRIBUTION IN REACTOR 100K EFFLUENT WATER (H-6)
(Total Weight in μg)

<u>Sample Identification†</u>	<u>Al</u>	<u>Ca*</u>	<u>Cl*</u>	<u>Cr*</u>	<u>Cu*</u>	<u>Fe*</u>	<u>Mg*</u>	<u>Mn</u>	<u>P*</u>	<u>S*</u>	<u>Si</u>	<u>U*</u>	<u>Zn*</u>
Original Sample	3760	300800	30080	5640	376	--	75200	--	940	94000	7520	--	--
Particulate Fraction > 0.4 μ	480	720	80	8	240	120	208	--	53	--	160	9	48
Particulate Fraction > 0.01 $\leq$ 0.4 μ	440	286	--	11	72	121	84	4	26	--	220	7	35
Dissolved Fraction < 0.01 μ	2400	168000	25500	5400	120	150	68100	90	210	99000	15000	102	1230

† Particle size estimate based on an average particle density of 2 g cm⁻³.

* Precision $\pm$ 20% of reported value; all others 1/3 to 3 times this value.

TABLE XV

RADIOACTIVITY DISTRIBUTION IN REACTOR 100K EFFLUENT WATER (H-6)
TOTAL ACTIVITY IN PICOCURIES**

<u>Sample Identification†</u>	<u>Cr⁵¹</u>	<u>P³²*</u>	<u>Sb¹²⁴</u>	<u>Sc⁴⁶</u>	<u>Sr*</u>	<u>Zn⁶⁵</u>
Original Sample	3,620,000 ± 7%	121,500 ± 11%	53,000 ± 75%	67,500 ± 31%	8,010 ± 14%	89,900 ± 28%
Particulate Fraction > 0.4 μ	50,700 ± 1%	14,700 ± 5%	--	5,740 ± 2%	5 ± 37%	1,298 ± 5%
Particulate Fraction > 0.01 ≤ 0.4 μ	3,410 ± 5%	2,180 ± 6%	--	404 ± 5%	1 ± 56%	9 ± 100%
Dissolved Fraction < 0.01 μ	3,340,000 ± 1%	71,300 ± 5%	49,300 ± 3%	5,630 ± 9%	809 ± 9%	11,400 ± 4%

† Particle size estimate based on an average particle density of 2 g cm⁻³.

** Corrected to date of sampling.

* Beta activity; all others gamma activity.

TABLE XVI

ELEMENTAL DISTRIBUTION IN REACTOR 100K INFLUENT WATER (H-7)
(Total Weight in μg)

Sample Identification†	Al	Ca*	Cl*	Cr*	Cu*	Fe*	Mg*	Mn	P*	S*	Si	U*	Zn*
Original Sample	1536	652000	49920	384	384	1920	176640	115	768	241920	15360	-	4224
Particulate Fraction > 0.01	413	5395	819	12	52	2806	1426	7	109	2138	663	8	116
Dissolved Fraction < 0.01 μ	4000	550000	115000	350	300	500	150000	50	300	305000	48000	3	3400

† Particle size estimate based on an average particle density of 2 g cm^{-3} .

* Precision $\pm 20\%$ of reported value; all others 1/3 to 3 times this value.

TABLE XVII

ELEMENTAL DISTRIBUTION IN REACTOR 100K EFFLUENT WATER (H-8)
(Total Weight in μg)

Sample Identification†	Al	Ca*	Cl*	Cr*	Cu*	Fe*	Mg*	Mn	P*	S*	Si	U*	Zn*
Original Sample	1050	630000	38500	10500	700	175	161000	105	350	220500	14400	00	7000
Particulate Fraction > 0.01 μ	385	6034	700	119	53	142	1582	5	225	2450	350	196	105
Dissolved Fraction < 0.01 μ	3060	510000	32300	10880	680	272	122400	136	476	272000	40460	4	2040

35

† Particle size estimate based on an average particle density of 2 g cm^{-3} .

* Precision $\pm 20\%$ of reported value; all others 1/3 to 3 times this value.

TABLE XVIII

RADIOACTIVITY DISTRIBUTION IN REACTOR 100K EFFLUENT WATER (H-8)
TOTAL ACTIVITY IN PICOCURIES**

<u>Sample Identification†</u>	<u>Cr⁵¹</u>	<u>P³²*</u>	<u>Sb¹²⁴</u>	<u>Sc⁴⁶</u>	<u>Sr*</u>	<u>Zn⁶⁵</u>
Original Sample	8,400,000 ± 6%	261,000 ± 11%	116,200 ± 64%	146,800 ± 27%	1320 ± 100%	179,000 ± 27%
Particulate Fraction > 0.01 μ	115,500 ± 8%	75,200 ± 5%	15,900 ± 50%	18,010 ± 5%	21 ± 100%	2,100 ± 24%
Dissolved Fraction < 0.01 μ	7,650,000 ± 3%	178,000 ± 4%	39,000 ± 11%	100,000 ± 4%	5 ± 100%	156,000 ± 4%

† Particle size estimate based on an average particle density of 2 g cm⁻³.

** Corrected to date of sampling.

* Beta activity; all others gamma activity.