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SUBJECT OR TITLE

Radioactivity of Hanford Reactor Cooling Effluent Water (ROUGH DRAFT)

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FROM J.W. Healy

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Richland, Washington
June 28, 1951 [redacted]

(ROUGH DRAFT)

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RADIOACTIVITY OF HANFORD REACTOR COOLING EFFLUENT WATER

(To be delivered at Chalk River by J. W. Healy, July 9-10, 1951)

The Health Instruments Divisions at Hanford are primarily interested in the reactor effluent water as a potential source of radioactive pollution of the Columbia River. As a result, most of our work has been expended on the measurement of the longer lived activities.

A study of the composition of the water was first made by West when the plant was put into operation. From that time until the end of 1950, no concerted effort was made to obtain a complete picture of the water composition. Recently a series of analyses was started by both the Technical and Health Instrument Divisions to evaluate possible hazards and to estimate the effects of various methods of preliminary treatment of the water.

The reactors at Hanford are located on the Columbia River at distances of 38-53 river miles above the town of Pasco. Figure 1 illustrates the location of the four areas with respect to the river and the towns of Richland, Pasco, and Kennewick.

WATER TREATMENT

The water used for cooling is pretreated by settling, chlorination, and flocculation with ferric sulfate and lime before use. A small amount of sodium dichromate is added as a corrosion inhibitor. An estimate of the mineral impurities is given in the data of Table 2.

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TABLE 2

WATER ANALYSES
PPM

	River Spectrograph	Process Chemical	Process Spectrograph
Ag	0.002	-	0.004
Al	0.2	-	0.3
As	< 0.2	-	< 0.2
Au	< 0.01	-	< 0.01
Ca	> 2	25	> 2
Cr	0.03	0.8	1
Cu	0.07	0.01	0.02
Fe	0.1	0.02	0.5
K	2	-	2
Mg	2	-	2
Mn	0.1	0.005	0.01
Mo	< 0.005	0.1	-
Na	> 2	4-5	> 2
P	< 0.2	0.015	< 0.2
Si	> 2	-	> 2
Sr	0.02	-	0.02
Th	< 0.04	-	< 0.04
U	< 0.2	< 0.002	< 0.2

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These analyses are of a qualitative nature since the spectrograph has not been well calibrated for these samples and the chemical analyses are of a spot nature with only one or two samples for each value. Of particular interest in this table are the low value for manganese and the significant amounts of sodium, calcium, and potassium.

After leaving the reactor, the water is passed through a retention basin designed in two sections so that either may be used independently. Here the water is held for average times of 2.5-4 hours to allow the short lived activities induced in nitrogen, oxygen, aluminum, magnesium, calcium, chlorine, etc. to decay before the water is returned to the river. The actual hold-up time as measured by sampling at the entrance and the exit and comparing these results to a measured decay curve is very close to the theoretical obtained from the volume of the basin and the rate of water flow indicating little channeling.

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II. RADIOCHEMICAL ANALYSES

A complete listing of the major isotopes measured in the water with their relative concentrations is still not available because of the complex nature of the mixture produced and the difficulties in separating traces of one isotope from large quantities of other contaminants. We have identified, however, nearly all of the major contributors and have obtained some data on the relative abundance, although this data has not been obtained for a sufficient length of time to permit evaluation of any possible changes with seasons due to variation in river flow and thus turbidity, mineral content, etc. A summary of the relative activities due to the various isotopes is given in Table 2 arranged according to half-life.

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TABLE 2

EFFLUENT WATER ACTIVITIES
Four Hours Out of Reactor

Isotope	$T_{1/2}$	Radiation Energies Particle Mev	Radiation Energies Photon Mev	*Fraction of Total Percent
$Cl^{34} + Ca^{49}$	34M	5.1,2.4	3.4	3
Mn^{56}	2.59 Hr	0.7,1.05,2.88	0.84,1.81,2.13	31
Na^{24}	14.8 Hr	1.39	1.38,2.75	23
Cu^{14}	12.8 Hr	K,0.57,0.66	1.35	30
As^{76}	26.8 Hr	1.3,2.5,3.0	0.55,1.2,1.8	5
Mo^{99}	67 Hr	0.24,1.3	0.24,0.77,0.82,0.84	0.3
Unidentified	60-70 Hr	-	-	1.3
P^{32}	14 da	1.71	no	0.5
Cr^{51}	26.5 da	K,e^-	0.32	0.4
Fe^{59}	47 da	0.25,0.46	1.10,1.30	0.1
Rare Earths	Mixed	-	-	5.7

*Analyses added to 75%. Above results normalized to 100%.

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All percentages in this work have been based on the activity four hours after leaving the piles since this is an approximation of the holdup time in the retention basin. Relative values at any other time may be obtained by extrapolation with half-life. The Cl^{34} and Ca^{49} values in this table were extrapolated from work done in 1945; all other values have been measured in late 1950 or early 1951 and are representative of low river flows. Since the actual analytical values only added to 75%, the results in the table were normalized to give a total of 100%. This discrepancy in closure is believed to be due to the difficulty in correcting the separated precipitate to the same basis of sample spread, self-absorption, etc. as the original sample, as well as to the possible presence of emitters of half-life less than two hours other than the ones listed.

One isotope not listed in this table is Si^{31} (2.8 hours) which may be present to the extent of 5-10% although the measurements available do not check too well. The material listed as unidentified follows in the analytical procedure at the point where one would expect zirconium, but specific reactions to positively identify it have not yet been carried out. The fraction listed as "rare earths" is that activity that carries through the lanthanum fluoride procedure after removal of interfering ions by an iodate precipitation. The decay curve of this fraction is curved indicating a composite mixture. The activity decreases by a factor of five in the first day, by a factor of 50 in the first ten days and by a factor of 1000 in the first twenty days. The exact isotopes that make up this fraction have not been isolated but are undoubtedly a mixture of short-lived materials produced both by irradiation of traces of rare earths in the water and by fission of the small amount of uranium present in the water and on the outer surfaces of the uranium slugs. The presence of such a process has been indicated by isolating the barium fraction and noting the decay rate. A buildup of activity corresponding closely to that expected from the La^{140} daughter Ba^{140} indicated the presence of fission in such a position that the products are released to the water. A rough estimate of the activity of this fraction at 0.01%

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of the total allowed an estimate of about 0.5 grams of uranium in contact with the cooling system if all of the Ba^{140} produced is dissolved in the water and not adsorbed on surfaces.

From tables 1 and 2 it is apparent that the activities of these fractions are not in the proportion expected from a straight irradiation of the salts in the process water. Such materials as Mn, Ca, Cr, and Fe are higher in proportion than would be expected. This has been shown, in the case of the Mn, to be due to holdup of a portion of the Mn on the walls of the tube through the pile thus resulting in a longer irradiation time. West has estimated that about 2% of the manganese is held up for about 45 minutes to give the increase in activity. The large amounts of Cu, Fe, etc. are undoubtedly due to the same problem of film formation of the colloidal hydroxides with subsequent release to the water. Radiochemical analyses of the film scraped from the inside of the tube have indicated the presence of Mn, Cu, Na, Cr, P, and iron.

These activities form only a small percentage of the total activity as the water leaves the reactor. It has been estimated that the N^{16} (8 sec) contributes about 30,000 times as much activity as the Na^{24} while O^{19} (31 sec) contributes about 100 times and the Al^{28} (2.4 M), from corrosion and the natural aluminum in the water, contribute about 20 times as much as the Na^{24}. These isotopes have practically disappeared by the time the water leaves the retention basin.

Figure 2 illustrates the rate of decay over a period of two months of several samples of this water. The solid line in this figure represents a calculated decay curve from the analyses presented in Table 2, while the points are values measured on a mica window counter. All results have been normalized to the activity after two days for comparison. The check between the two curves indicates that no major fraction has been omitted, particularly from the longer lived isotopes.

Some analyses have been carried out for trace quantities although this program is by no means complete. Our interests in this problem are largely aimed at those isotopes which, although present in small quantities, may present a hazard because of

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their low maximum permissible level in the body. Values for Zr^{95}, Ti^{51}, Ca^{60}, and Ca^{45} are all less than 0.01%, while Zn^{65} is about 0.05% and Sr^{89} is about 0.002%. These values are uncertain at this time because of chemical difficulties involved in the separations and the problem of obtaining a true value of the counting rate from a decay curve of the isolated fraction.

The effluent water is routinely monitored for total radiation rate to a submerged body by plastic ion chambers over the retention pond for the gamma component and by reading a thin sample of the water on a shallow ionization chamber three times a day for the beta component. In addition, periodic samples of the outlet water are taken, evaporated to dryness and measured on mica window counters for total activity. Both of these readings (total dosage rate and activity) have fluctuated with changes in power level of the reactor and possibly with changes in turbidity of the river water.

A comparison of the readings of total dosage and activity should give some clue to the possibility of changing composition of the active components with seasons since the relation between activity and the dosage rate may be expressed by a simple equation of the form:

$$D = KAE$$

Where D is the dosage rate, A is the activity and E is the average disintegration energy of the active components of the water. Thus if there is no change in composition of the water E will remain constant and the ratio of the dosage rate to the activity will be a constant. An examination of the average data for the past few years indicates a fluctuation in this ratio with a maximum energy per disintegration during the months of May to August and a minimum in the winter months. This may be due to an increase in the relative Cu^{64} concentration during the winter since this isotope has an average disintegration energy of 0.22 Mev as compared to 4.8 Mev for Na^{24} and 2.6 Mev for Mn^{56}. Unfortunately these data are not accurate enough to allow exact evaluation of the relative gamma and beta energies and the analytical data are not as yet detailed enough to show actual changes in composition of the water.

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III. SOLIDS ANALYSES

The retention basin provides a settling location for any solid particles that may be in the water from the periodic treatment to remove film from the tubes, and it allows contact between the water and the algae that grows on the sides of the basin. These solid materials tend to adsorb activities from the water and become active. In these solids, the longer lived isotopes are of particular importance because of the greater relative concentration that will occur before their radioactive decay limits the equilibrium value.

Analyses of algae from these basins indicates concentrations of P^{32} up to 18% of the total activity eight hours after removal. The Na^{24} activity is less than 7% while the Cu^{64} is greater than 30%. Other samples 26 days old have P^{32} concentrations up to 25-30%. The total activity of this algae is about 100 times that of the water for the same weights.

Analyses of the activity at the time film is being removed from the tubes indicate that both the particulate and the soluble fractions are relatively increased in Ca^{64} and decreased in Mn^{56} as compared to the normal water. The particulate portion is increased in P^{32}. Total concentrations of activity in the water are increased by factors of about 100 mostly carried by 2-5 PPM of particles about 0.5 micron in diameter.

Both plutonium and polonium have been detected in the algae and in the precipitate at the bottom of the basin although the analytical techniques are not yet sensitive enough to measure them in the water. These materials are believed to originate in trace quantities of U and Bi in the water to the pile.

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IV. RECIRCULATED WATER

During one experiment in the reactor, water was continually circulated through one tube, cooled, and reused in the systems. Samples of this water, after some time of operation, when examined for tritium oxide, gave values of about 0.3 uc/cc. These values did not change over a period of time indicating no further buildup of activity. These results posed three problems: (1) was the activity really due to tritium or was our method of analysis in error? (2) why was there no further increase in activity?, and (3) why should there be this much activity since with the published cross-section of deuterium and the estimated flux, these concentrations should be lower than measured by a factor of 100?

The measurements were made both by the ionization current produced in acetylene and by the counting rate from a hydrogen filled GM counter with good checks between methods. After numerous distillations, burning of hydrogen generated from the sample, electrolysis, etc. it was concluded that the activity must be due to tritium since it followed hydrogen in every case except in electrolysis of a large sample where the gas was depleted in activity by a factor of 5-6 as was expected.

The question of the approach to equilibium was answered reasonably well by examination of the rate of dilution due to water added. Figure 3 presents the measured values for one run showing the buildup of tritium activity approaching an equilibrium value. The solid lines in this diagram represent the expected rate of buildup after taking into account the dilution by water added with the curves normalized to fit in absolute value the rate of increase measured. Two curves are shown, one for a 106 liter system which is the total volume and the other for an 86 liter system which is the volume without a surge tank. The shape of the curves is a reasonably good fit to the experimental data although

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the absolute magnitude of the increase before equilibrium is reached is somewhat different.

The question of the source of this activity is still not settled. The best estimate of the actual rate of production is about 3×10^{-2} uc/cc/day of irradiation. Calculated values from the D_2O would be on the order of 4×10^{-4} uc/cc/day and the water indicates only very low concentrations of lithium.

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V. DILUTION IN RIVER

The water from the 107 basins is released to the Columbia River through a pipe that runs along the bottom of the river bed to the main channel. Measurements are made of the activity of the water and aquatic life at distances up to 100 miles in order to follow any potential hazard from pollution. These measurements are made difficult by the variations in flow rate of the river (20,000-500,000 cubic feet per second) which changes both the flow pattern and the time required to reach a given point and thus the composition of the activity due to decay of the shorter-lived activities. At low water (100,000-200,000 cubic feet/second) the mean velocities range from about two to four miles per hour depending upon the particular location. Thus by the time the water has reached Kennewick or Pasco, the first location at which the water is used for domestic purposes other than irrigation, the total activity has decreased by a factor of three and the composition has changed to give 35% Na^{24}, 40% Cu^{64}, 10% As^{76}, 1.5% P^{32}. etc. Surveys in the Pasco-Kennewick region indicate that the activity is reasonably uniform across the river. At distances closer to the effluent discharge line, the activity appears to follow a relatively narrow channel spreading as it flows down the river. Figures 4, 5, and 6 are drawings made from aerial photographs of the discharge of the ferri-floc from the settling basins in several of the areas. This pattern coorelated very well with activity measurements made at the same time. Of particular interest is the sketch of Figure 5 which illustrates the effect of an island in the river. At low water this island is joined to the shore at the upper end. As the water rises, however, a considerable amount of activity finds its way into the slough behind the island giving the highest shoreline concentrations on the river.

Routine analyses of the water at Pasco and Kennewick indicate that the sum of the individual activities are well below the maximum permissible levels for these isotopes in drinking water. Some concentration of activity is noted on the filters for the domestic water, but this has never reached an alarming level.

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The measurements of aquatic life are of interest because of the concentration of particular isotopes. We have already noted that the algae in the 107 basin concentrates the P^{32} to a large extent. Measurements of fish, plankton, etc. in the river indicate that about 90% of the activity accumulated is P^{32}. This selective absorption has been attributed to the relatively small natural phosphorous content of the river thus making the living organisms compete for the phosphorous present. It is also of interest to note that the uptake is greatest during the months of July to November when the river flow rates are decreasing, but the metabolism rate of the organism is at its peak.

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