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CHEMICAL EFFLUENTS TECHNOLOGY WASTE DISPOSAL INVESTIGATIONS

JULY, AUGUST, SEPTEMBER, 1960

Prepared by members of the

Chemical Effluents Technology Operation

Edited by: W.A. Haney

December 9, 1960

Chemical Research & Development Operation
Hanford Laboratories Operation

Hanford Atomic Products Operation
Richland, Washington

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CHEMICAL EFFLUENTS TECHNOLOGY WASTE DISPOSAL INVESTIGATIONS JULY, AUGUST, SEPTEMBER, 1960

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CHEMICAL EFFLUENTS TECHNOLOGY WASTE DISPOSAL INVESTIGATIONS JULY, AUGUST, SEPTEMBER, 1960

INTRODUCTION

The Chemical Effluents Technology Operation performs research to investigate the chemical and physical aspects of environmental contamination resulting from the disposal of plant effluents or from potential process incidents. This report is primarily concerned with plant assistance research in the field of waste disposal during the quarter July, August, September, 1960.

Ground-water monitoring results utilized in this report were obtained from samples collected routinely by the Environmental Monitoring Operation and analyzed by the Radiological Chemical Analysis Operation.

I. INTERPRETATION OF GROUND-WATER MONITORING DATA (W.A. Haney)

Special Monitoring Well Samples

During this report period large volume samples from six selected wells were obtained and given special analytical processing to further detail the radioisotopes which are included in routine "gross beta" determinations. Samples were analyzed for the predominant long-lived isotopes of interest in separations wastes. The analytical results presented in Table I are in essential agreement with past routine analytical results relating to concentrations and species of radioisotopes of concern.

The quite low Co^{60} concentration noted in well 299E-24-8 confirms the source of the waste at Hot Semi-Works as being the 216-BY scavenged waste cribs. The positive total alpha result noted in the sample

TABLE I

RADIOISOTOPIC ANALYSES OF SPECIAL MONITORING-WELL SAMPLES

(Concentrations in units of μc/cc)

Isotope	Well (1)* 299E-24-8	(2) 699-38-70	(3) 299W-22-19	(4) 299W-22-20	(5) 299W-14-1	(6) 299W-15-3
Total Alpha	$<1.1 \times 10^{-7}$	$<1.5 \times 10^{-7}$	$<7.1 \times 10^{-8}$	$3.4 \times 10^{-7} \pm 2.3 \times 10^{-7}$	$<1.7 \times 10^{-8}$	$<2.7 \times 10^{-8}$
Gross Beta	9.0×10^{-6}	3.3×10^{-7}	$7.2 \times 10^{-6} \pm 2.5 \times 10^{-6}$	1.7×10^{-5}	$1.6 \times 10^{-7} \pm 4.8 \times 10^{-8}$	2.4×10^{-6}
Ru^{106}	8.2×10^{-6}	$5.0 \times 10^{-7} \pm 1.0 \times 10^{-7}$	5.1×10^{-6}	1.5×10^{-5}	$3.6 \times 10^{-7} \pm 2.1 \times 10^{-7}$	2.6×10^{-6}
R.E. + Y	$<2.8 \times 10^{-8}$	$<4.2 \times 10^{-8}$	$<1.4 \times 10^{-8}$	$<2.0 \times 10^{-8}$	$<1.2 \times 10^{-8}$	$<1.8 \times 10^{-8}$
Cs^{137}	$<1.2 \times 10^{-8}$	$<5.3 \times 10^{-9}$	$<1.4 \times 10^{-8}$	$<1.3 \times 10^{-8}$	$<5.7 \times 10^{-9}$	$<8.6 \times 10^{-9}$
Ce^{144}	$<6.3 \times 10^{-9}$	$<3.6 \times 10^{-9}$	$<4.6 \times 10^{-9}$	$<8.9 \times 10^{-9}$	$<8.6 \times 10^{-9}$	$<3.0 \times 10^{-9}$
Pm^{147}	$<4.5 \times 10^{-8}$	$<6.7 \times 10^{-8}$	$<2.2 \times 10^{-8}$	$<3.0 \times 10^{-8}$	$<1.9 \times 10^{-8}$	$<2.9 \times 10^{-8}$
Total Sr	$<2.7 \times 10^{-9}$	$<4.6 \times 10^{-9}$	$<2.6 \times 10^{-9}$	$<4.1 \times 10^{-9}$	$<2.8 \times 10^{-9}$	$<2.6 \times 10^{-9}$
Sb^{125}	$<3.1 \times 10^{-7}$	$<2.5 \times 10^{-7}$	$<2.4 \times 10^{-7}$	$<2.9 \times 10^{-7}$	$<2.4 \times 10^{-7}$	$<2.4 \times 10^{-7}$
Zr^{95}-Nb^{95}	$<1.1 \times 10^{-7}$	$<9.0 \times 10^{-8}$	$<9.0 \times 10^{-8}$	$<9.0 \times 10^{-8}$	$<8.4 \times 10^{-8}$	$<9.0 \times 10^{-8}$
Co^{60}	$5.3 \times 10^{-7} \pm 2.1 \times 10^{-7}$	$<5.3 \times 10^{-8}$	$<1.6 \times 10^{-7}$	$<2.4 \times 10^{-7}$	$<1.6 \times 10^{-7}$	$<3.6 \times 10^{-7}$
Te^{127}	$<2.6 \times 10^{-8}$	$<3.4 \times 10^{-8}$	$<4.2 \times 10^{-8}$	$<2.5 \times 10^{-8}$	$<2.5 \times 10^{-8}$	$<1.6 \times 10^{-8}$

* These numbers refer to the approximate well locations as indicated by the encircled numbers on Figure 1.

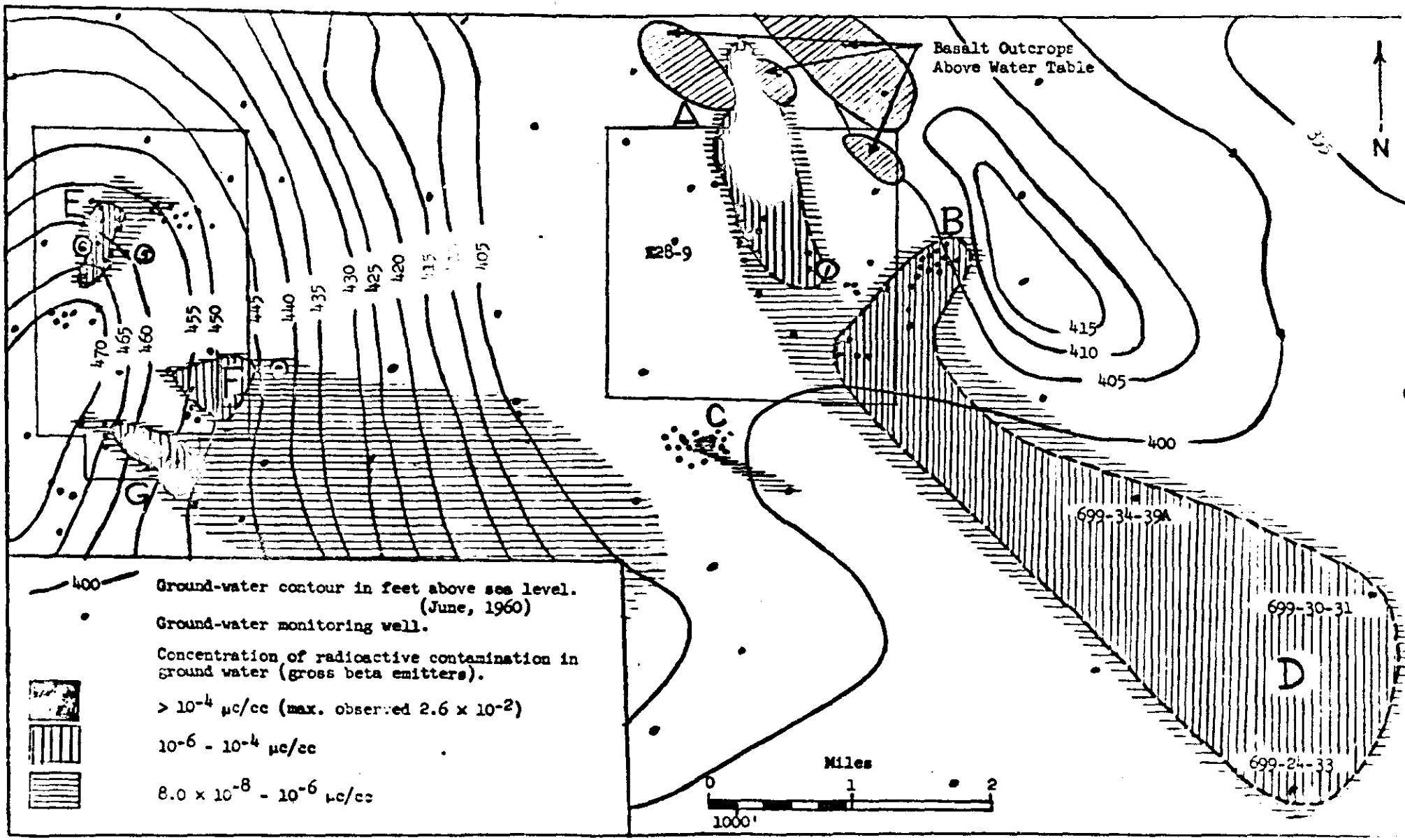

FIGURE 1 -- Map of Separations Area Showing Probable Extent of Ground-Water Contamination, September, 1960

from the 299W-22-20 well monitoring the Redox Laboratory Crib will be investigated further by attempting to determine the isotope(s) contributing this activity.

Special Depth Samples -- 200 East Area

In July, 1960 ground-water samples were obtained at various depths (5 foot intervals) below the ground-water surface from ten wells in 200 East Area. These particular wells were selected because depth samples from the 299E-28-2 well obtained several years ago had shown significant variations in gross beta-emitter concentrations with depth in this area. The source of radioactive contaminants at this location is the 216-BY scavenged waste cribsite where a somewhat unique situation exists in that the surface of the basalt is only a few feet below the local ground-water level. Waste infiltrating to the ground water under the cribs is effectively prevented from further vertical movement by the underlying basalt. Geological data indicate that the surface of the basalt dips to the south to the extent that it is about 130 feet below the regional water table at a distance of 7,500 feet south of the cribs.

All of the wells sampled -- except the well farthest from the disposal site -- showed increasing concentrations of radioisotopes with depth. However, in several instances samples from intermediate depths contained lower concentrations than did the samples from lesser depths. The increasing concentration trend was most evident in wells closest to the disposal site; in general these wells bottomed closer to the basalt than did more distant wells. Nitrate-ion analyses revealed somewhat higher concentrations at depths, but the trend was not nearly so pronounced as it was from radioisotope concentrations. Table II presents analytical data

collected from the depth samples and well location information.

TABLE II

RESULTS OF DEPTH SAMPLES FROM SELECTED WELLS IN 200 EAST AREA

Well No.	Distance And General Direction From BY Cribs	Radioisotope Concentrations (μcβ/cc) Ground-Water Surface Sample	Well Bottom Sample
299E-28-8	1600' S	8×10^{-8}	1.4×10^{-5}
299E-28-2	2500' S	1×10^{-7}	1.3×10^{-4}
299E-28-1	3000' SSE	1.3×10^{-6}	1.4×10^{-5}
299E-28-7	3200' S	5.3×10^{-7}	6.0×10^{-7}
299E-28-3	3400' S	2×10^{-7}	5.6×10^{-7}
299E-28-4	3800' SSE	1.7×10^{-7}	5.5×10^{-7}
299E-28-6	4100' S	4.4×10^{-7}	1.0×10^{-6}
299E-24-8	5600' SE	1.5×10^{-6}	2.0×10^{-5}
299E-23-1	5600' SSE	2.2×10^{-7}	3.8×10^{-7}
299E-24-7	7500' SSE	$<8 \times 10^{-8}$	$<8 \times 10^{-8}$

Other wells in the vicinity of the 200 Areas that have been depth sampled (approximately 20) have shown very little or no variations in radioisotope concentrations with depth. Correlation of the results in Table II with geological, hydrological and waste disposal data will be attempted to determine if this phenomenon is due to aquifer permeability characteristics, waste density effects, or a combination of these considerations. In time, wells to the north of the 216-BY cribsite and additional wells to the south will be depth sampled to define the area over which these anomalous results extend.

200 East Area

Figure 1 is a map of the 200 Areas showing the extent of detectable ground-water contamination, as indicated by analyses of routine samples, during the period July-September, 1960. Only minor changes in the areal

extent of the various contamination zones were evident during this quarter.

(1) 216-BY and 241-B Cribs

Several changes were evident in ground-water contamination patterns at Site A, Figure 1, during this reporting period. Beta-emitter concentrations in well 299E-27-1 just north of the Hot Semi-Works increased five-fold as was noted in Well 299E-24-8 south of the HSW the previous quarter. Future migration of contaminated ground water from this location should be predominantly southward where it will enter the general southeastward path taken by wastes from Purex plant disposals.

The quarterly gross-beta concentration average for Well 299E-28-9 monitoring the abandoned 216-BR-1, 2 and 3 cribs (Figure 1) was below the routine detection limit (8×10^{-8} μcβ/cc) for the first time since use of the crib was discontinued in 1957. The gradual decrease in concentrations of radioisotopes which has been experienced in this well over the past year is probably primarily due to decay of ruthenium-106 and reduced drainage from the vadose zone.

Wells containing the highest concentration of radioisotopes at Site A are those located about 1,000 feet southeast of the 216-BY cribs. Well 299E-33-15 contained the maximum gross beta-emitter concentration this quarter, 1.6×10^{-3} μc/cc. No radiocesium or radiostrontium was detected in wells at Site A during this reporting period; however, several of the wells contain Co^{60} in detectable concentrations. The maximum Co^{60} concentration detected this quarter was 9.0×10^{-5} μc/cc in Well 299E-33-17.

(2) 216-A Cribs and Contaminated Ground Water Southeast of 200 East Area

Radioactive contaminants in the ground water under Purex waste disposal facilities and contaminants which have moved from this area to the southeast are shown as Sites B and D respectively on Figure 1. All of the wells monitoring the 216-A-8 and A-24 tank farm condensates cribs now contain less than 1×10^{-4} μcβ/cc. All but one of these thirteen wells showed a decrease to about one-half the previous quarter's gross-beta concentration. The exception was the 299E-26-5 well monitoring the first section of the active 216-A-24 crib. The two-fold increase in the concentration in this well is probably due to infiltration of most of the waste through the first section of the crib.

A slight decrease in the concentration of radiocontaminants was evident in the three wells which define the contaminated ground-water pattern at Site D. Evidently the activity in these wells peaked last quarter for present ground-water flow and Purex waste disposal conditions. No long-lived radioisotopes were detectable in samples from wells monitoring Purex cribs during this report period.

(3) 216-BC Cribs and Trenches

Only four of the twenty wells monitoring the 216-BC scavenged waste cribs and trenches (Site C, Figure 1) contained concentrations of radioisotopes greater than the routine detection limit (8×10^{-8} μcβ/cc) this quarter. The maximum average concentration was 2.4×10^{-7} μcβ/cc in Well 299E-13-4. No long-lived isotopes have ever been detected in ground water samples from wells at Site C.

200 West Area

Three major areas of ground-water contamination in 200 West Area are shown on Figure 1 as Sites E, F and G. Only minor changes in the areal extent of contaminated ground water under 200 West Area were noted this quarter.

Maximum gross beta-emitter concentrations for the three sites in 200 West Area this quarter, together with maximum concentration averages for the previous quarter are presented in the following table.

TABLE III

MAXIMUM AVERAGE CONCENTRATIONS OF GROSS BETA ACTIVITY IN 200 WEST AREA WELLS

Site	Well Number	July-September, 1960	April-June, 1960
E	299W-15-1	6.8×10^{-6} μc/cc	6.0×10^{-6} μc/cc
F	299W-19-2	1.8×10^{-6} μc/cc	1.8×10^{-6} μc/cc
G	299W-22-14	2.6×10^{-2} μc/cc	2.7×10^{-2} μc/cc

Better definition of the contaminated ground water at Site F was afforded by the installation of three wells to monitor the new 224-U Building process condensate crib. The noticeable decrease in the areal extent of ground-water contamination at Site F is probably due to abandonment of the 216-WR-1, 2 and 3 cribs early this year.

Only one well in 200 West Area contains concentrations of long-lived fission products greater than routine detection limits. Well 299W-22-2 continues to routinely show the present of Sr^{90}; the maximum concentration this quarter was 1.0×10^{-6} μc Sr^{90}/cc which is one-tenth of the previous quarter's maximum. Strontium-90 accounts for about 30% of the gross beta-emitter activity in this well.

II. PLANT WASTE DISPOSAL PRACTICE

Chemical Processing Department

Work on sinking well points near the 106-TY underground storage tank has been abandoned due to the inability to drive the points to the required depth of 60 - 70 feet. Soil conditions are such that maximum depths of penetration on three attempts were 35', 25' and 20'. Even at these relatively shallow depths partial plugging of the pipe resulted from jammed threads on the connecting collars. The possibility of extending the present well drilling contract to provide shallow wells in place of the drive points as a method for determining the contamination status of the soils near the tank bottom is being investigated.

(1) Treatment of Condensate Wastes (J.M. Skarpelos)

The Micro Pilot Plant (MPP) was used to continue to explore the ability of various bed packing materials to remove radioisotopes from Purex tank farm condensate waste. Inorganic ion exchange materials tested during the quarter included clinoptilolite (treated to remove clay from the crude), Decalso (a synthetic gel-type zeolite), and Linde 4-A (a snythetic crystalline-type zeolite). All three materials, even at flow rates of 10 gpm/ft^2, removed Cs^{137} to less than 4×10^{-5} μc/cc, a level sufficiently below maximum permissible concentrations in water* to make them candidates for radioactive waste treatment schemes. The effluent concentration of Sr^{90} varied from $1 - 3 \times 10^{-6}$ μc/cc which is slightly higher than MPC_w.

Other tests evaluated the decontamination ability of synthetic apatite and Canadian apatite in preparation for future tests on the

* NBS Handbook 69.

calcite-phosphate reaction. These materials were not very effective in removing any of the radioisotopes studied.

(2) Waste-Soil Evaluations (A.E. Reisenauer, K.C. Knoll)

Using Redox tank farm condensate as influent solution and soil taken from a well near the 216-SX crib, two soil column tests were run at a flow rate of 0.87 gallons/ft^2/hr. The results of these crib-life evaluations showed no detectable breakthrough of Cs^{137} or Sr^{90} after passage of 100 column volumes. The Redox tank farm condensate crib received 17.7 column volumes of waste through May, 1960 at a flow averaging 2.4 column volumes per year.

The specific retention capacity of Hanford soil for Purex organic waste was determined employing standard centrifuge techniques. A value of 7.8 weight percent was noted for the organic compared to a value of 13.3% for water. The ratio of these measured values agrees reasonably well with that predicted from the surface tensions of the two fluids.

The relative removal of radioisotopes from the solvent by soil was determined by equilibrium experiments using actual samples of organic waste. The radioactive materials in the waste for which soil adsorption was evaluated were Ru, Sr, Cs, I, Co, Zr, Pu, U and rare earths. All of the isotopes were effectively removed by the soil except Ru, I and Co. Since the Co^{60} was present in the waste at a concentration appreciably greater than the present ground-water control limit (5×10^{-5} μc/cc), it was recommended that the Purex 216-A-2 crib not be used beyond its specific retention capacity.

Irradiation Processing Department

(1) NPR Decontamination Wastes (W.N. Koop)

Studies of treatment and disposal methods for wastes resulting from decontamination of the NPR primary coolant loop were continued. One decontamination process involves the stepwise circulation of three cleaning solutions with intermediate water rinses. Most of the radionuclides of concern are effectively scavenged by the precipitates formed upon mixing the spent cleaning solutions from this multi-step process. Laboratory-scale experiments were conducted to determine if natural settling was adequate for separation of the sludge from the supernatant liquid. Scavenging resulting from one month of settling was only slightly less than that noted from centrifuging the waste after a four-day settling period. Results of other tests indicate that scavenging is essentially unaffected when the chemical cleaning waste is diluted with reasonable amounts of water rinses.

A similar treatment method is being sought for wastes resulting from the single-pass decontamination process (a proprietary phosphoric acid cleaner followed by a water rinse) so that both wastes might be processed in the same facility. Since the disposal of soluble phosphates to the river might result in a significant increase in the P^{32} concentration in coolant effluents from downstream reactors, consideration was given to removal of the phosphate ion together with the scavenging of radionuclides by precipitation of insoluble calcium phosphate. This reaction has been used to scavenge radioisotopes from wastes at other nuclear installations.

Efforts to precipitate the phosphate ion with $CaCO_3$ and $Ca(OH)_2$ were unsuccessful presumably because these compounds do not adequately neutralize the acid. Neutralization with NaOH after the addition of $CaCl_2$ resulted in more than 99% of the phosphate being precipitated. Appreciable scavenging of radioisotopes was evident in that less than 1% of the barium, zinc and cobalt remained in the supernatant liquid. However, the sludge volume resulting from this treatment method would be undesirably large, and sizeable quantities of calcium salt and caustic would be required.

Investigations of methods for scavenging the radionuclides without precipitating the phosphate are now underway. If a satisfactory method of this type is developed, it will be necessary to dispose of the supernatant waste either by releasing it locally at a rate slow enough to prevent a significant increase in river phosphate concentration or by releasing the waste at a point downstream of the 100-F Area coolant intake.

(2) Fission Product Release Experiments (R.K. Hilliard, D.L. Reid, and L.F. Coleman)

The study of fission product release from irradiated uranium this quarter emphasized the relationship between release rates during heating and the irradiation level of the uranium specimen. Tests were conducted wherein all experimental conditions were held constant except the irradiation level, which was varied from 2×10^{14} nvt to 2×10^{18} nvt. Little or no effect on the release of most of the fission product isotopes studied has been observed within the range of irradiation levels investigated. For tests at 1[illegible]°C for 24 minutes in air flowing at 2300 cc/minute, the percentage release

of fission product elements were: iodine, 70-90; tellurium, 60-50; xenon, 80-100; cesium, 30-60; strontium, 0.2-0.4; barium, 0.2-0.2; ruthenium 0.5-0.5; and zirconium, 0.05-0.05. Tests on specimens irradiated to 10^{20} nvt, equivalent to 330 MWD/T, will be made to complete this phase of the investigation.

The validity of release calculations was improved this quarter by the installation of equipment capable of dissolving the oxidized residue. Residue dissolution enables a material balance to be made and thus obtain a higher confidence in the fission product release calculations. Previous calculations were based solely on analyses of the volatilized portion and a duplicate specimen.

The new dissolver has provided a means for determining the amount of uranium oxidized during each test. The method entails initial dissolution of the oxide followed by total dissolution of the unoxidized residue. Chemical analysis of the two solutions for uranium content is then performed. Irradiation levels up to 10^{18} nvt had no significant effect on the percent of uranium oxidized under the conditions of these tests.

Past and current work has dealt with determining the fractional release of key fission product isotopes heated under various conditions of temperature, time, atmosphere and irradiation level. Plans were made this quarter to begin a study of the physical and chemical nature of the evolved material. Initial tests will measure the particle size and deposition behavior with various specimen heating conditions. Also tests to determine the distribution of iodine evolved in particulate versus elemental form will be made.

(3) Pilot Scale Study of Reactor Effluent Decontamination Using Aluminum Beds (H.G. Rieck)

A study was initiated to establish on a pilot scale the effectiveness of aluminum shavings beds for removing significant quantities of radioactive constituents in present reactor coolant effluents. The bed is contained in a tank 30 feet long, 3 feet wide and 6 feet deep, with a packed length of 20 feet. A flow rate of 4 linear feet/minute was maintained in the early tests. The pilot scale results were in essential agreement with laboratory scale decontamination achieved. When equilibrium was reached, the bed was removing 68% of the As^{76}, 87% of the Zn^{65}, 57% of the P^{32} and about 25% of the Np^{239}. The significant removal of Np^{239} was greater than in previous laboratory experiments.

Other data obtained in these tests were pressure drop, radiation build-up on the bed and the significance of sludge in operating the bed. Tests employing increased flow rates were started near the end of this reporting period.

III. PARTICLE SAMPLING IN GASEOUS EFFLUENT STREAMS (A.K. Postma, L.C Schwendiman)

In a study supported by the Division of Biology and Medicine, the variables controlling particle deposition and retention in conduits were further studied. The deposition velocity of 30 μ particles (lycopodium spores) in a one-inch pipe was measured for various flow rates.

Results obtained were in reasonable agreement with the correlation developed earlier. Re-entrainment studies with 20 and 30 μ particles established that at velocities lower than 10 feet/second re-entrainment was minimal. At 22 feet/second nearly all the 20 μ particles were re-entrained within 20 minutes.

The velocity required for ren-entrainment of 20 μ particles was significantly higher than that required for 30 μ particles. The results assisted in establishing flow rates below which large particles will be effectively retained in sampling lines. This is of importance in evaluating particle sampling-line losses.

IV. GROUND-WATER HYDROLOGY (D.J. Brown)

A monitoring well, 699-S11-E12, drilled two miles north of the 300 Area penetrated an artesian aquifer in the basalt bedrock. The water in this aquifer was under sufficient pressure to cause it to flow out of the well at the ground surface. The flow rate was estimated to be between two and three gallons per minute. This is the fifth flowing artesian well drilled at the Hanford Works. The other four wells are located on the western edge of the project at the mouth of Cold Creek Canyon. Using the head of water found in wells believed to tap this artesian zone the isopotential pattern for this aquifer may be determined. This pattern, although based on a limited number of observation wells, is believed to indicate the general location of recharge areas. One of these recharge areas appears to be located toward the west, probably in the upland area surrounding Rattlesnake Hills and Yakima Ridge. The isopotential pattern also indicates another area of possible recharge along Wahluke Slope. The broad ground-water mound north of Gable Mountain may be caused in part by leakage from confined aquifers which are recharged from the Wahluke Slope area. Spectrographic analyses of water samples show no significant differences between samples from artesian wells and those water samples taken from the unconfined ground-water body. Thus it might not be possible to identify recharge of the water table aquifer by

leakage from the artesian zone by means of water quality measurements. This recharge could have a very significant effect on the hydrologic flow pattern beneath the Hanford region.

V. WELL DRILLING SUMMARY (D.J. Brown)

Company	Completed Wells	Feet Drilled	Date Completed	Total Feet	To Water	To Basalt
Hatch Drilling Co.	299-W22-22	269	7/16/60	300	Yes	No
Hatch Drilling Co.	199-F8-1	57	7/22/60	57	Yes	No
Hatch Drilling Co.	299-W22-23	310	8/10/60	310	Yes	No
Hatch Drilling Co.	199-D2-5	95	8/11/60	95	Yes	No
Hatch Drilling Co.	199-D5-12	91	8/19/60	91	Yes	No
Hatch Drilling Co.	199-F8-2	55	8/23/60	55	Yes	No
Hatch Drilling Co.	699-15-15	575	8/26/60	722	Yes	Yes
Hatch Drilling Co.	199-H3-1	75	8/30/60	75	Yes	No
Hatch Drilling Co.	299-W22-24	575	9/8/60	575	Yes	Yes
Hatch Drilling Co.	199-B4-4	105	9/13/60	105	Yes	No
Hatch Drilling Co.	699-S11-E12	282	9/14/60	282	Yes	Yes
Hatch Drilling Co.	699-S2-E12	260	9/23/60	260	Yes	Yes

Twelve monitoring and research wells, specified in the drilling contract AT(45-1)-1607, were completed by crews of the Hatch Drilling Company. This work represents 48% of the total drilling designated under this contract. At the drilling rate now maintained by these crews, 36 feet per calendar day, the work on this contract should be completed on schedule. The completion date is now set at January 4, 1961.

The Hatch Drilling Company was low bidder on another drilling contract issued by the AEC. This contract, AT(45-1)-1623, is for the construction of six wells in the 200-East Area. Five of these wells are for monitoring purposes and are to be drilled adjacent to the new 216-A-30 crib located directly east of the 200-East Area. The other well is designed to provide an emergency water supply to the 241-A tank farm condensers.