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**The 216-Z-8 French Drain  
Characterization Study**

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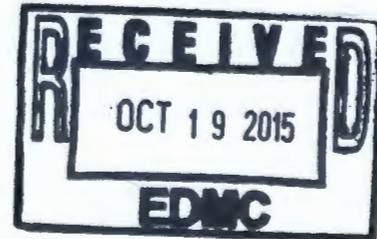
September 1984

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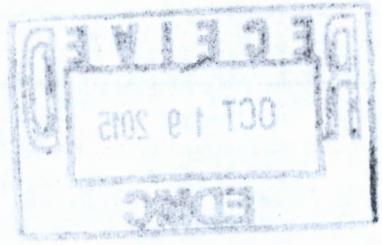
**MASTER**

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DEDICATION

This document is dedicated to M. C. "Maryjane" Marratt. Maryjane was responsible for the planning and execution of most of the field work involved with the characterization of the 216-Z-8 French drain site. Due to a serious, chronic illness, she was forced to withdraw before completion of the project. We regret that we had to lose her expertise and company.



ABSTRACT

Past waste disposal practices at the U.S. Department of Energy's Hanford Site included the disposal of liquid wastes containing low concentrations of plutonium. This 216-Z-8 French drain study is one of a series of studies examining these historical transuranic waste facilities no longer in use at the Hanford Site. The 216-Z-8 site was chosen for study because its design is typical of a number of retired liquid disposal sites. The information obtained from this study will aid the development of technical options for the permanent disposition of this type of facility.

The 216-Z-8 French drain underground disposal system consisted of a large settling tank that overflowed into a French drain. The French drain consisted of two large-diameter, gravel filled, vitrified clay pipes placed on end, end-to-end, over a gravel-filled excavation. The top of the drain was sealed with concrete to prevent the upward flow of waste solution. The waste solution discharged to the 216-Z-8 waste disposal system was a neutralized, transuranic recovery process, filter cake, backflush slurry.

The primary objective of this study was to determine the distribution of plutonium and americium beneath the French drain. Transuranic activity under the French drain did not exceed 5 nCi/g in the soil samples obtained from a well within 1 m of the drain structure. Conservative estimates indicated that 4 to 5 m<sup>3</sup> of radioactive contaminated sediments, greater than or equal to 10 nCi/g may lie directly under the 216-Z-8 French drain.

The secondary objective of the study was to evaluate the possibility of a leak in the settling tank. Results from the analysis of soil samples from wells drilled around the settling tank indicated the presence of low-level transuranic contamination (on the order of 0.001 nCi/g) in the soil surrounding the tank. However, the distribution of the contamination does not support a leak as a plausible mechanism to account for the observed activity surrounding the tank. The bulk of the plutonium was confirmed to be in the sludge that remained in the tank; thus, no significant environmental impact would be expected even if there has been a leak.

## EXECUTIVE SUMMARY

The 216-Z-8 French drain underground disposal system that consisted of a 58,500-L capacity settling tank and French drain received neutralized, filter cake backflush slurry and a rinse water from a plutonium reduction slag- and crucible-dissolution process conducted in the 234-5 facility. The 216-Z-8 system received waste from 1955 until 1962. Waste discharge records, settling tank liquid analysis in 1974, and a copper foil neutron activation analysis over the residual tank content indicated that the tank received about 1 kg of plutonium. The inventory of plutonium in the solution overflowing from the tank to the French drain was reported as 48 g.

In 1974, the settling tank was observed to contain approximately 27,500 L less solution than measurements made in 1958 and 1959 indicated. The remaining contents of the tank were estimated to be 29,000 L of solution and 2,000 L of sludge. The solution was pumped from the settling tank and transferred to a high-level waste tank in 1974. Photographs taken inside the settling tank after the pumping operation indicate that approximately 4,000 L of material remained on the bottom of the tank. Most of the material appears to be solids.

The primary objective of this study was to determine the distribution of plutonium and americium beneath the French drain. To determine the radionuclide distribution, a single well was drilled south and adjacent to the 216-Z-8 French drain. Selected sediment samples collected during the drilling were analyzed for the transuranic (TRU) radionuclides  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Am}$ . Plutonium and americium activity attributed to the waste discharged to the French drain was encountered in a zone extending approximately 5 m from the bottom of the French drain. All measured levels of activity were less than 5 nCi/g of sediment for  $^{239}\text{Pu}$  and less than 0.5 nCi/g of sediment for  $^{241}\text{Am}$ . Considering the initial waste TRU activity level and the activity levels in the sediment samples, estimates indicate that a 1-m-deep zone of greater than 10 nCi/g activity may exist directly below the 216-Z-8 French drain excavation. Conservatively, a 4- to 5-m volume of sediment contaminated to greater than 10 nCi/g of sediment may lie directly below the 216-Z-8 French drain.

The secondary objective of the study was to evaluate the possibility of a leak in the settling tank that would account for the discrepancy of 27,500 L of solution between 1962 and 1974. To evaluate the possibility of the tank leak, four shallow wells were drilled to a depth of 7.6 m near the tank, and selected sediment samples were analyzed for  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Am}$ . The locations of the wells around the tank were selected so that any point on the tank's surface was within approximately 4.5 m of a well. At least one well would be expected to penetrate the volume of sediment contaminated by a 27,500-L leak. Based on the distribution of TRU activity observed beneath the French drain, and depending upon the actual distance for any possible breach in the tank,  $^{239}\text{Pu}$  activity of 1 pCi/g or greater would be anticipated in a well penetrating the contaminated volume. The actual pattern of 1 to 5 pCi/g  $^{239}\text{Pu}$  observed in all four wells was inconsistent with what would be expected a tank leak. All four wells were

not expected to penetrate the plume of 27,500 L, and TRU activity was expected to decrease rapidly with distance from a breach in the tank wall. It was suspected that a systematic error was made in the 1974 measurement of the liquid level of the settling tank that may have resulted in an incorrect calculation of the liquid volume. The liquid volume was never actually measured at that time.

Because the 216-Z-8 facility discharged liquid waste solution to the ground through the French drain, the additional effect of a leak of the same solution from the tank is of little practical consequence. The bulk of the plutonium discharged to the facility was confirmed to remain in the sludge that remained; thus, the environmental impact of a leak if it did occur should be minimal. At this time, no additional work is proposed. If action requiring excavation to or of the tank is indicated, additional drilling prior to excavation should be included to sample sediment around the tank.

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## INTRODUCTION

The U.S. Department of Energy (DOE) Hanford Site has served primarily as a plutonium production facility since its inception in 1943. The Site is located in south-central Washington (Fig. 1). The chemical separation facilities for the purification of plutonium from nuclear reactor fuel rods are located in the central portion of the Site in two exclusion areas known as the 200 East and 200 West Chemical Separations Areas (see Fig. 1).

Before 1973, plutonium-contaminated liquid waste was disposed of directly to the ground via various subsurface structures. This is no longer practiced at the Hanford Site. One type of subsurface disposal system, known as a French drain, consists of a buried, rock-filled encasement with an open bottom through which liquid waste percolates into vadose zone sediments.

The 216-Z-8 French drain disposal system is located in the 200 West Area (see Fig. 1). Liquid wastes discharged to the French drain were neutralized and diluted filter cake backflush slurries from a slag and crucible dissolution process located in the 234-5Z Building in the Z Plant complex (Fig. 1 and 2). The wastes were discharged in relatively small batches and were routed through a large settling tank called the "silica storage tank." Overflow from the tank went to the French drain (Fig. 2 and 3).

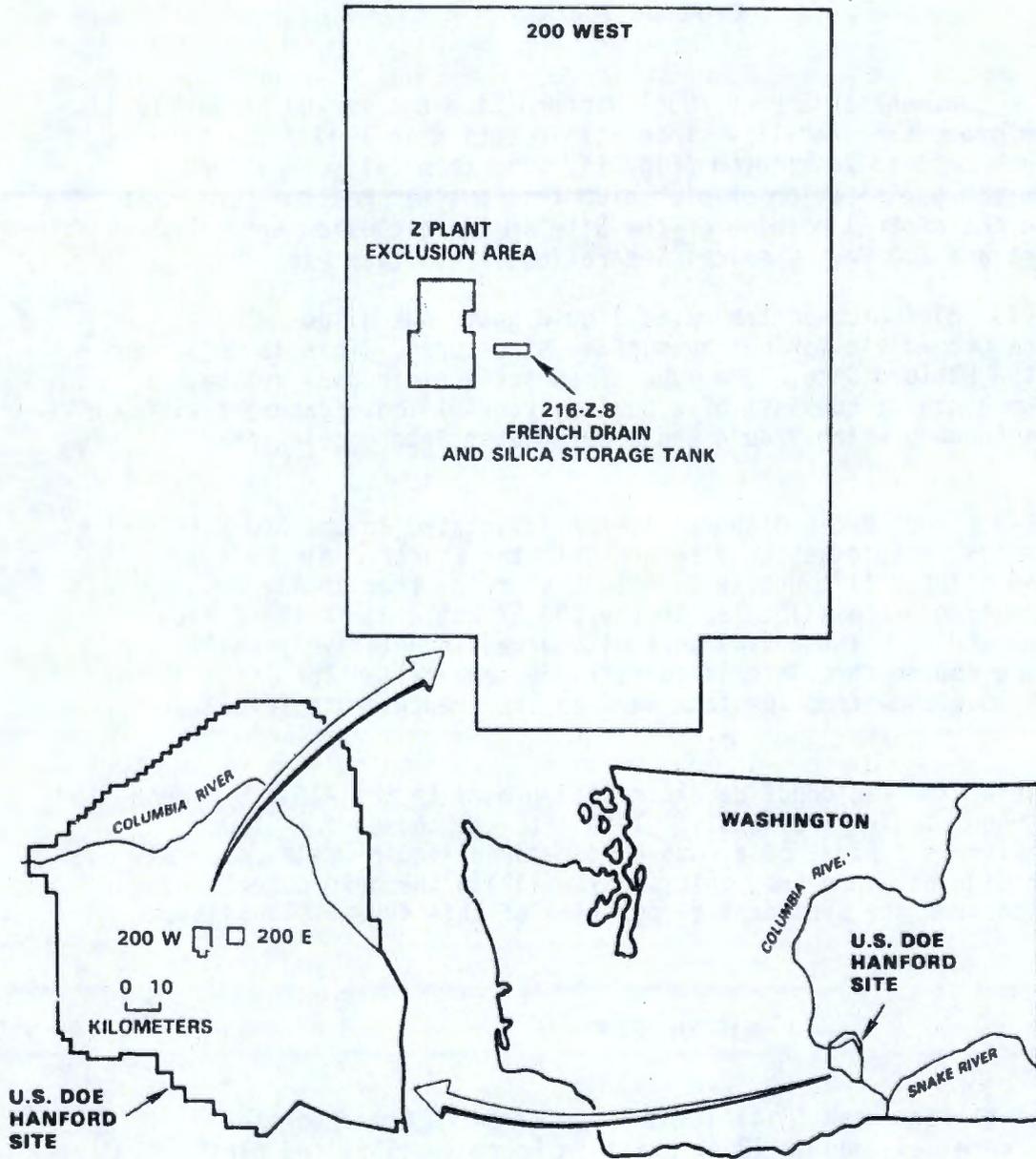
The status of the radionuclide distribution beneath the 216-Z-8 French drain has not been studied previously. This site was chosen for study because its design is typical of a number of retired liquid waste disposal sites. Information obtained from this study will aid the development of technical options for the permanent disposition of this type of facility.

## SITE HISTORY

The silica storage tank (Fig. 3 and 4) and 216-Z-8 French drain (Fig. 3 and 5) were designed in 1954, built in February 1955, and first received waste in July 1955. The 216-Z-8 French drain operated from July 1955 to April 1962 when use of the Recuplex operation was discontinued, and the facility was retired from service.

## CONSTRUCTION DETAILS

The silica storage tank (see Fig. 3 and 4) was constructed of 0.8-cm steel conforming to specifications then applicable for underground oil and gas storage tanks. The tank has a 2.4-m diameter, is 12.2 m long, and has a 58,500-L capacity. The tank has two buried access holes, one 30.5 cm in diameter and another 61 cm in diameter. Two 10-cm-diameter vent risers extend above the ground surface. The outside surface of the tank is painted with a red anticorrosive paint, which is coated with asphaltic pipe enamel.



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FIGURE 1. Map of the Hanford Site.

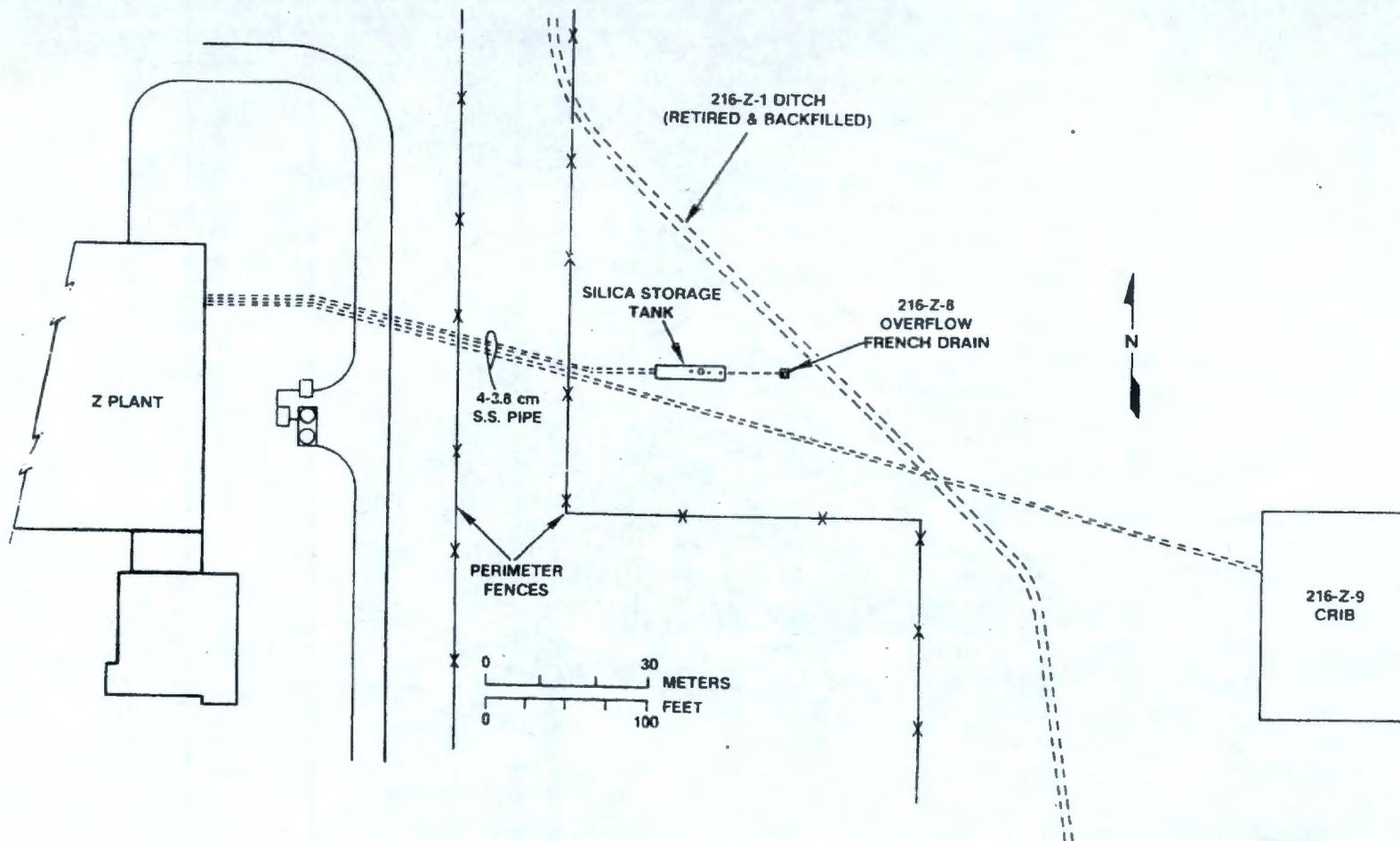


FIGURE 2. The 216-Z Site in Relation to Z Plant.

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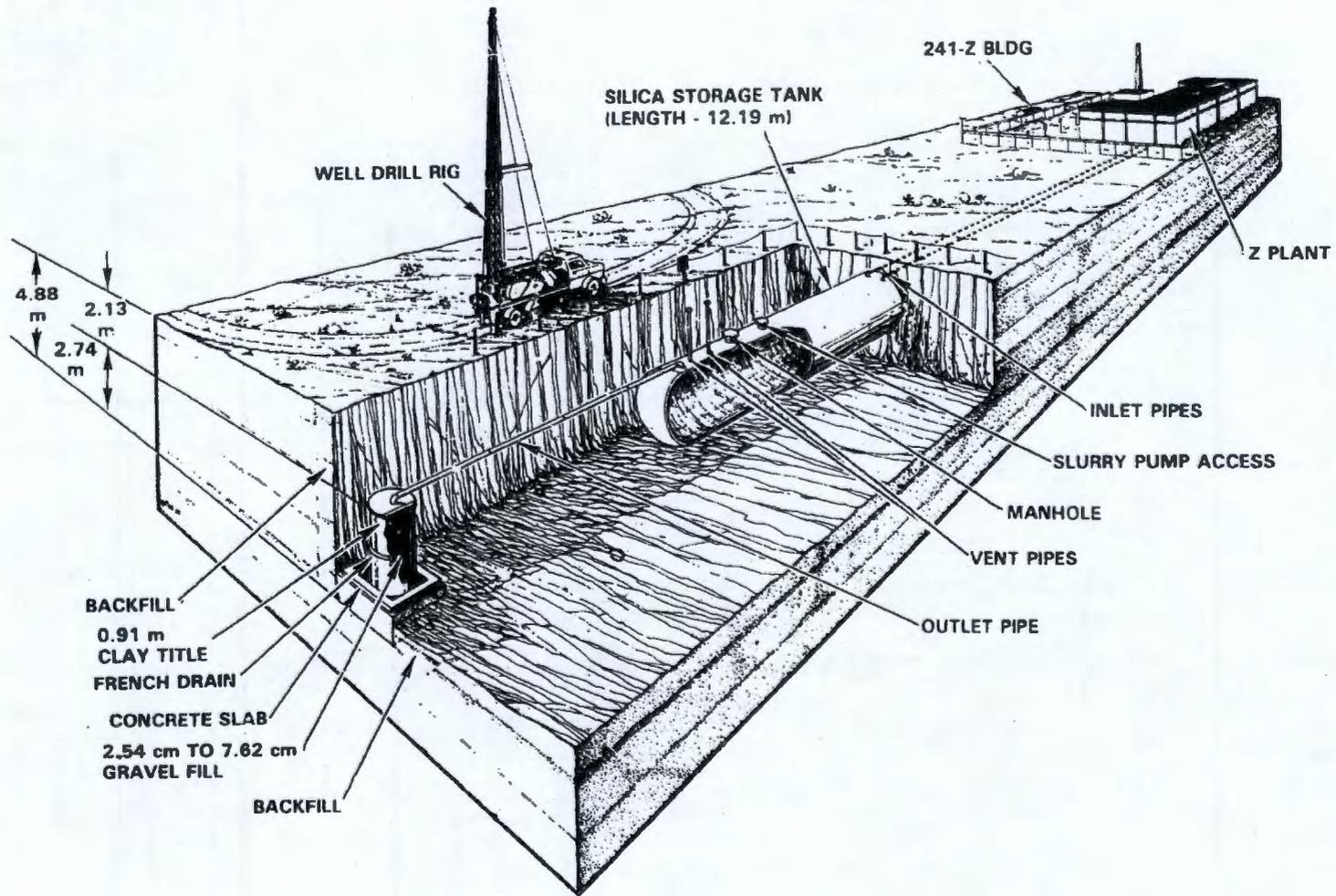


FIGURE 3. The 216-Z-8 French Drain Facility.

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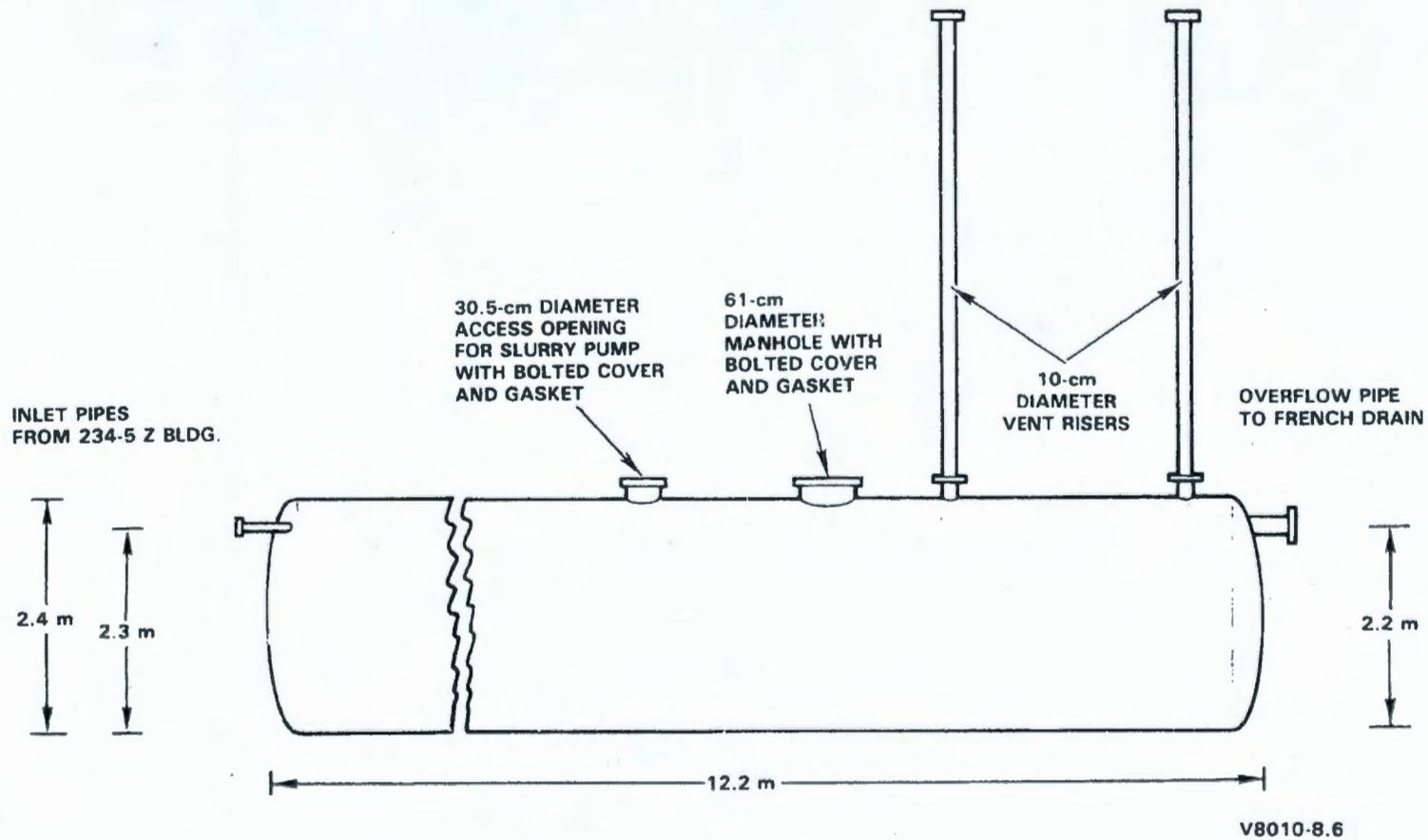


FIGURE 4. Silica Storage Tank Construction Details.

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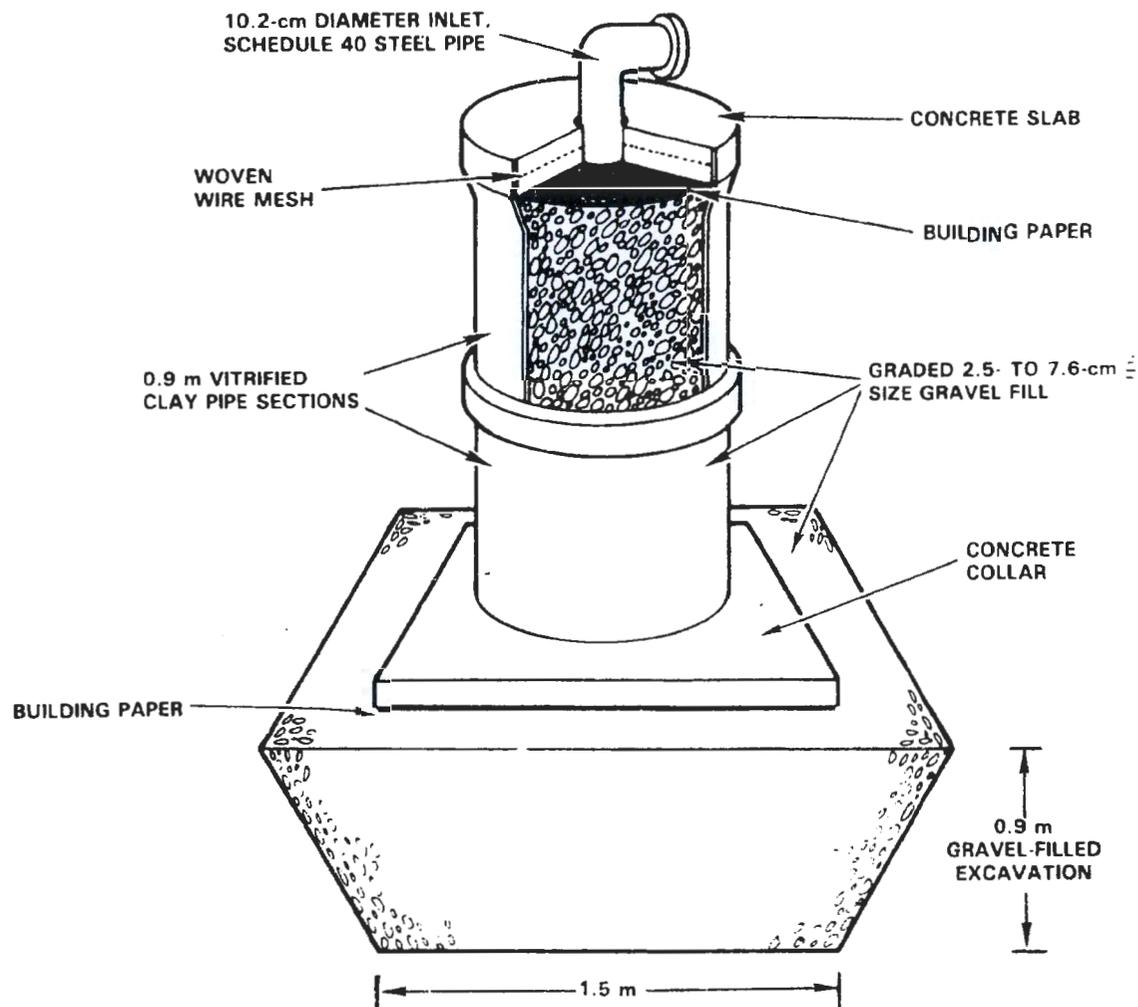


FIGURE 5. Construction Details of the 216-Z-8 French Drain.

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Wastes entered the silica storage tank from the D-10 and D-11 waste collection tanks in the 234-5Z Building of Z Plant via two 3.8-cm-diameter stainless steel lines that enter the tank 15 cm below the top of the tank. The overflow to the French drain, at the opposite (eastern) end of the tank from the inlet pipe, consisted of a single 10.2-cm-diameter, schedule 40 stainless steel pipe located 20 cm from the top of the tank. Waste solution leaving the silica storage tank via the overflow pipe flowed 11 m, at a shallow (~3%) slope, into the French drain structure.

The tank volume was sufficient to permit the main filter cake component, the finely divided diatomaceous earth powder (see following section on Waste Disposal History), to settle in the tank from each batch discharged. It was assumed most of the plutonium would be associated with the particulate material, and very little plutonium would be in the solution that overflowed to the French drain. (The diatomaceous earth was composed primarily of fine-grained silica particles ( $\text{SiO}_2$ ), hence the name silica storage tank.)

The French drain (see Fig. 3 and 5) was constructed by excavating a square area 5.3 m deep with walls at angles so that the bottom dimensions of the hole formed a 1.5- by 1.5-m square. The bottom 0.9-m of the excavation was backfilled with clean, graded gravel. A seal of building paper was laid over this gravel with a 0.9-m-diameter hole to match the two sections of a 0.9-m vitrified clay pipe that were placed on end, end to end, over the hole. A concrete collar was poured around the bottom of the clay pipe, on top of the building paper.

The clay pipe was also filled with gravel and capped with building paper and a wire-mesh-reinforced concrete slab to seal the top of the structure. The overflow pipe from the silica storage tank entered through the center of the French drain's concrete cap. Woven wire mesh was placed at the opening of this pipe into the French drain to ensure a void space at the waste inlet. The entire structure was backfilled, resulting in the top of the structure being 2.5 m below grade. Waste entered the gravel-filled excavation at 4.4 m below grade.

The total volume filled with gravel in the French drain was more than  $4 \text{ m}^3$ . Assuming a net porosity of 30%, more than 1,000 L of solution could be accommodated by the French drain. This was sufficient capacity to permit the waste solution to percolate into the sediments beneath the French drain between batch discharges of the waste and rinse water.

#### WASTE DISPOSAL HISTORY

The 216-Z-8 French drain received a low-level plutonium contaminated waste stream from the 234-5Z Building from 1955-1962. This waste stream was dilute and nearly neutral, had no fission or activation product content, and was relatively low in both disposal rate and total disposal volume.

The nature of this waste stream may be understood better in terms of its position in the plutonium recovery process from which it originated.

Wet chemical methods, such as the Redox or PUREX process in fuel separation facilities in the 200 Areas, separated and purified plutonium from irradiated reactor fuel rods (Liverman, 1975; Cleveland, 1970). The product of the processes was a plutonium nitrate solution, which was transported to Z Plant for preparation of the final plutonium product.

The 234-5Z Building was constructed in 1949 and contained several operations including the plutonium finishing facility and the plutonium scrap recovery facility. In the plutonium finishing facility, plutonium in the nitrate solution was precipitated as an oxalate. The plutonium oxalate precipitate was calcined to produce plutonium oxide. The oxide was converted to plutonium tetrafluoride and then reduced to plutonium metal. Both the oxide and metal were products of the process, depending upon requirements.

The bulk of the waste ultimately discharged to the 216-Z-8 French drain system was derived from the procedure used to reduce the plutonium fluoride and cast plutonium metal. The general process is described in detail by Cleveland (1970). Briefly, the plutonium reduction involved use of a closed pressure vessel. A magnesium oxide crucible was placed in the pressure vessel and the space between the crucible and the inner wall of the pressure vessel was filled with magnesium oxide sand. Calcium fluoride and magnesium oxide sand were placed in the crucible and shaped to make a depression to hold the charge. The charge consisted of a well-mixed blend of plutonium fluoride, finely divided calcium, and some iodine. Air was purged from the pressure vessel and it was sealed securely. The pressure vessel was heated to more than 600°C and the heat of the ensuing exothermic reduction reaction brought the internal temperature of the pressure vessel to about 1600°C.

After reduction and cooling, the crucible was removed and broken to free the plutonium metal "button" weighing approximately 2 kg. The remaining waste material consisted of the magnesium oxide crucible and sand, calcium fluoride, calcium iodide, and approximately 100 g of plutonium. This waste was sealed in metal cans and sent to the plutonium scrap recovery facility (which was called Recuplex during this period) for recovery of the plutonium.

Recuplex was a pilot plant operation that was discontinued in 1962 and replaced by the Plutonium Reclamation Facility (Bruns, 1967). The operations were similar solvent extraction processes. In Recuplex, the cans containing the scrap were fed into special semicontinuous dissolvers that used a concentrated nitric acid/aluminum nitrate [10M HNO<sub>3</sub>, 0.15M Al(NO<sub>3</sub>)<sub>3</sub>] solution to dissolve the can and its contents. Dissolution was incomplete. There was some silica present in the crucible material which, in the presence of aluminum, formed a hydrated, dispersed, and finely divided aluminum-silicon precipitate that interfered in the plutonium solvent extraction step that followed. To remove this material, the process solution was filtered through a sintered aluminum oxide block before the

solvent extraction was attempted. A finely divided, highly inert and pure diatomaceous-earth powder was used to aid filtration. This filter aid was suspended in water and poured over the filter just before the addition of the process solution. The filter aid residue left behind by the draining water presented a uniform, highly porous surface especially suited to the interception of the gelatinous, flocculated, and semicolloidal silica in these process solutions.

The filter aid could be used for several batches before material accumulation plugged the pores and impeded the flow. The filter was then cleaned. Cleaning the filter involved backflushing with approximately 40 L of a 0.3M nitric acid solution. The backflushed solution was collected in the D-10 or D-11 tank. Each tank had a 50-L capacity, and the tanks were connected in parallel. The solution was then neutralized with 2.3 L of 30% sodium hydroxide solution, sampled for accountability, and jetted to the 216-Z-8 French drain disposal system. The batch discharge volume was between 40 and 50 L. Each discharge was followed by a rinse of the system with water. Estimates based on available discharge records indicate that about 200 L of water were used per rinse.

Waste was first discharged to the 216-Z-8 French drain facility in July 1955. Measurements taken of the liquid level in the silica storage tank indicated that a constant level of 2.18 m, the designed overflow level, was attained in October 1957. From October 1957 to April 1962, when the facility was retired, it is estimated that 9,590 L of liquid waste containing an estimated 48.2 g of plutonium overflowed from the silica storage tank to the 216-Z-8 French drain (Hanson et al., 1973). The inventory, reported monthly, was constructed from available discharge records and is given in Appendix A.

#### MONITORING AND PREVIOUS DATA

Monitoring of the 216-Z-8 facility consisted of routine analysis of the alpha activity in the waste solution in the D-10 and D-11 holding tanks before discharge, and nonroutine liquid level measurements and solution analysis in the silica storage tank. The liquid level in the tank was first measured at the designed 2.18 m overflow level in October 1957. Other readings in October 1958 and July 1959 showed that this level was being maintained and the tank was filled to its 58,500-L design capacity and the waste solution then overflowed to the French drain. In October 1958, an analysis of the solution in the silica storage tank reported a plutonium concentration of 5.6 mg/L. The solution sample was reported as being slightly cloudy due to suspended particulate material. The estimate of the plutonium in the waste that overflowed to the French drain from October 1957 to April 1962 is given in Appendix A. In April 1962, the 216-Z-8 facility was taken out of service with the shutdown of the Recuplex facility.

Records indicate that the liquid level in the tank was next measured in April 1974 in preparation for removal of the liquid contents. The 1974 measurement was taken as part of a survey of retired Z Plant tank

facilities. There are no records indicating that a liquid-level measurement was made when the tank was isolated in 1962. The 1974 liquid level was measured at 1.27 m, corresponding to a total volume of 31,000 L. A sequence of weekly measurements showed this new level remained constant. Additional measurements estimated the remaining volume consisted of approximately 29,000 L of solution and 2,000 L of solids (sludge).

Several possible explanations for the apparent 27,500-L discrepancy between the 1958-1959 readings and the 1974 reading were considered during the 1974 investigation. One explanation offered that part of the tank's contents were transferred in 1962 during isolation of the tank for retirement. There are no records documenting any transfer and there would seem to be no reason for transferring approximately half of the solution contents and leaving the remaining half. A second explanation considered was that the liquid evaporated during the 12 yr. Evaporation of this quantity of liquid is not considered feasible from an enclosed tank. The tanks risers were sealed, and the inlet lines were blocked. Only the overflow to the French drain provided a means of removing moisture and there was no mechanism for air movement over the liquid surface. A third explanation could be that there was some systematic error introduced in the depth measurements taken during one measuring period. There are no records validating this possibility. The fourth explanation suggested that the tank had leaked. Although the presence of fluoride in the waste solution suggests the possibility of corrosion of the tank, at the temperature involved, this is not considered probable. The 1974 investigation considered the first or the fourth explanation most likely. The authors of the current study believe the third alternative, a measurement error, should also be considered.

Four samples each of sludge and solution were collected from the silica storage tank during April and May 1974 before removal of the liquid contents of the tank. The solution was found to have a pH of 6. Plutonium concentrations in the solution ranged from 1 to 24 mg/L. In comparison, plutonium concentrations in the sludge ranged from 4 to 755 mg/L, confirming expectations that the plutonium was associated with the solids. No further effort was made to characterize the sludge as a function of location in the settling tank, water content, or chemistry, and the results merely serve to illustrate the heterogeneous nature of the distribution of plutonium within the tank.

Based on the analyses, a conservatively high calculation was made for the tank contents assuming a plutonium concentration of 755 mg/L in the sludge and 6 mg/L in the solution. The calculation indicated a maximum of approximately 1,600 g of plutonium in the tank: 175 g in the solution and 1,400 g in the sludge. Using the same calculations, a maximum of 165 g of plutonium would be in the 27,500 L of solution that presumably leaked from the tank.

In the fall of 1974, the tank was pumped to remove the remaining solution. Because the solution was reported to be too viscous to pump, water was added before pumping. More than 40,000 L of diluted solution were transferred by truck to a high-level waste storage tank.

After the tank was pumped, photographs were taken of the tank's interior by a camera lowered through the pump access manhole. Figure 6 is a view looking toward the outlet end of the tank. The single outlet pipe leading to the French drain is visible. Also visible on the top of the tank are the openings for the larger manhole access and the two risers shown in Figure 4. A conductivity reel used for measuring liquid levels is visible as a line hanging from the foremost riser. The nominal overflow level of the tank is clearly visible as a thick ring of material around the tank. The level mark was probably due to adherence of the fine particulate or colloidal material in the waste at the solution tank wall interface, and not from evaporation of the solution. Several fainter level marks were observed immediately below the overflow level with what appeared to be regular spacings between them (also see Fig. 7). These marks are believed to show temporary liquid levels as batches were added to the tank before the liquid overflowed. When measured in 1974, the level of liquid in the tank was slightly more than halfway up from the bottom of the tank. Several areas of dark coloration appear in the tank that might correspond to this level, but no level marks comparable to the overflow level were observed (see Fig. 6 and 7).

Figure 7 shows a view of the tank looking toward the inlet end. The two inlet pipes can be seen. The level marks for the different solution levels can be seen clearly at the end of the tank. Dark material at the end of the tank is at approximately the correct distance to correspond to the measured 1974 liquid level. A layer of sludge can be observed at the bottom of the tank in this picture. Using the distance from the top of the tank to the bottom of the inlet pipes (approximately 18 cm), the thickness of the sludge immediately below the inlet pipes was estimated as approximately 60 cm.

Figure 8 shows the bottom of the tank toward the outlet end; the conductivity reel seen in Figure 6 is visible. Clearly, a quantity of material remained at the bottom of the tank after the removal of the solution in 1974. The material toward the end of the tank appears to be a solid (sludge). The material visible in the foreground appears to be a liquid. The diluting water added during the pumping operation was reported to have been added using a fire hose. The liquid in the foreground suggests that a depression was made in the sludge layer beneath the manhole. Removal of the solid material under the manhole has apparently resulted in the slumping of the remaining solid material toward the center of the tank as indicated by the presence of sheer planes. These planes indicate that the material was somewhat consolidated. The colloidal claylike precipitate and the diatomaceous filter aid were the major solid components of the waste. This mixture is believed to have gelled in the tank, forming a viscous material with a texture comparable to sticky mud. As indicated, sufficient agitation could resuspend the material and allow it to be pumped.

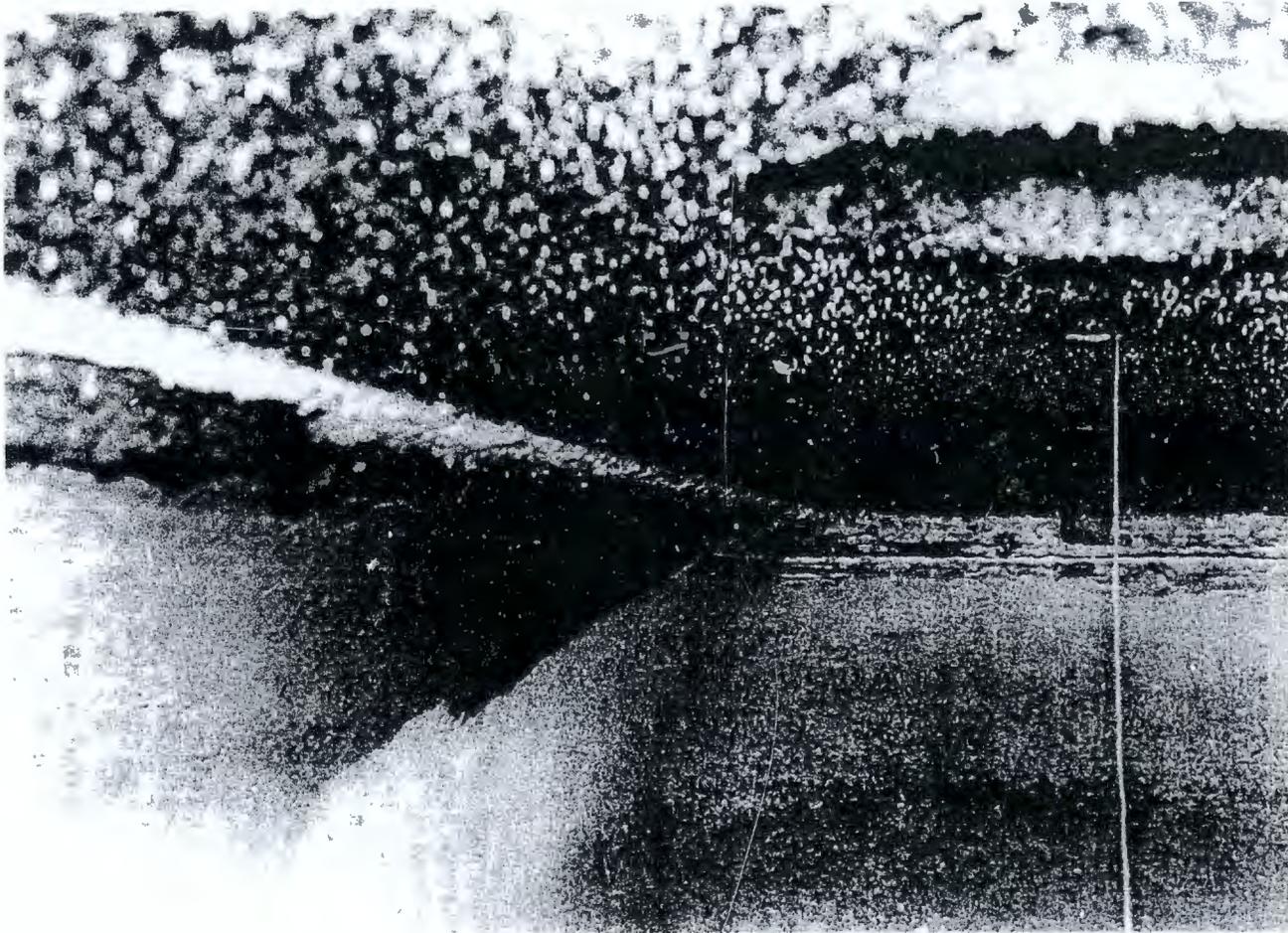


FIGURE 6. View Inside Silica Storage Tank Looking Toward The Outlet End  
(from 1974 study).



FIGURE 7. View Inside Silica Storage Tank Looking Toward the Inlets (from 1974 study).

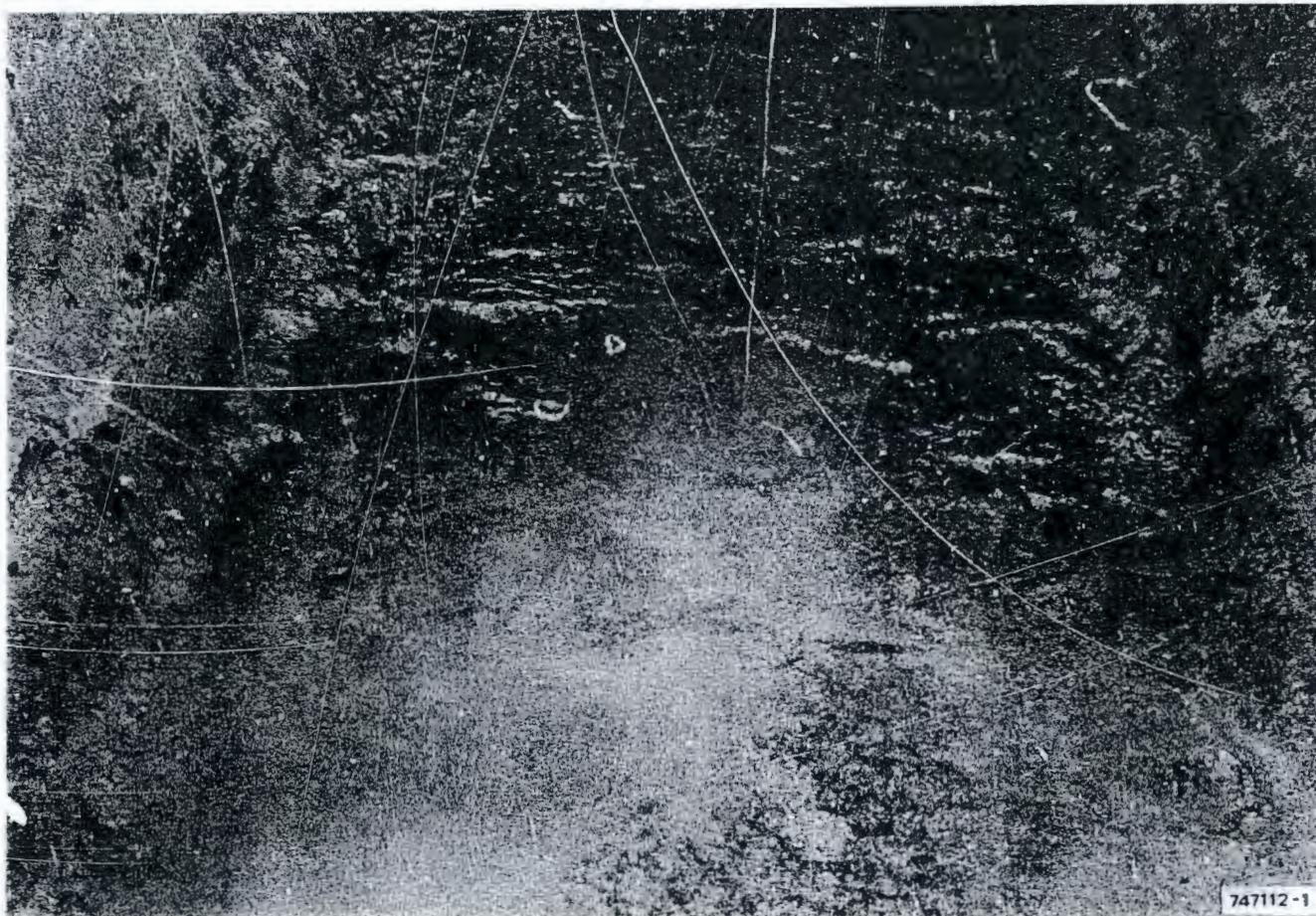


FIGURE 8. View of Sludge at Bottom of the Silica Storage Tank Looking Toward the Outlet End of Tank (from 1974 study).

The thickness of the sludge layer visible in Figure 8 was estimated two ways. Although not visible, the outlet end of the tank was near the upper portion of the photograph. Using the distance from the top of the tank to the overflow level for scale (25 cm) in Figure 6, it was estimated that the layer was 1.6 m wide, corresponding to a thickness of approximately 30 cm for a tank 2.44 m in diameter. Based on the angle at which the tank walls intersected the sludge surface, a comparable thickness was estimated. A uniform layer 30-cm thick in a 2.44-m-diameter tank corresponded to a volume of approximately 4,000 L. This would appear to be a minimum estimate, because the thickness of the sludge was observed to increase toward the inlet end of the tank. The ratio of solution to sludge remaining could not be estimated, but most of the material appeared to be a sludge.

## ENVIRONMENTAL CHARACTERIZATION

Various environmental factors can affect the present distribution of TRU elements beneath the 216-Z-8 French drain. These factors are discussed briefly. A general description of the environment at the Hanford Site is discussed in Liverman (1975). The referenced reports may be consulted for additional information.

## CLIMATE

The Hanford Site is located in south-central Washington State. The climate is semiarid because the Site lies east of the Cascade Mountain Range, which forms a moisture barrier to the general west-to-east weather flow patterns. Average annual precipitation is less than 18 cm. Temperatures are mild, with an annual average temperature of 12°C.

The Hanford Site is subject to occasional high winds. Prevailing winds at the 200 Areas are from the north-northwest through northwest, although the strongest winds are from the southwest. The highest peak wind gust measured was from the southwest at 116 km/h. Winds with peak gusts of 64 km/h or greater have been measured on the average of at least once every month.

## BIOLOGY

Eight major kinds of shrub-steppe communities make up the vegetation mosaic of the Hanford Site. Details of the vegetation and animal population of the Site are available (Liverman, 1975). A sagebrush/cheatgrass community is the dominant vegetative type in the 200 Areas. However, the 216-Z-8 controlled zone and nearby vicinity are man-disturbed areas due to their proximity to the Z Plant perimeter. Vegetation is controlled at Z Plant by herbicide use within the perimeter fence. The lack of native vegetation near the 216-Z-8 site indicates that herbicide has been applied at the site, but the presence of large tumbleweeds (*Salsola kali*) indicates that the application was not recent at the time of this study.

Biological intrusion into buried TRU-contaminated sediments can transport the TRU elements to the surface where resuspension by the wind provides a potential pathway to man. Intrusion can include burrowing by animals and uptake by plant roots. In the absence of complexing agents, however, plutonium and americium are not normally concentrated in plants (Francis, 1973; Price, 1973; Watters, 1978). Because only plutonium was discharged to the site, uptake by plants is not anticipated, but a study of the surface environment at the site has been undertaken to investigate biologic transport (see Acknowledgments).

Animals that burrow into waste sites can bring contaminated sediments to the surface (Liverman, 1975; O'Farrel and Gilbert, 1975). The lack of vegetation cover and present human activity, however, minimize the potential for mammals at the site.

## HYDROLOGY

The movement of ground water at the 216-Z-8 site could transport radionuclides from their present location. A discussion of the hydrology of the site is presented in terms of surface, vadose zone, and ground-water hydrology.

### Surface Hydrology

Low rainfall amounts and intensities, in addition to gently sloping, highly permeable surface soils over most of the Hanford Site, limit water erosion. Most of the 200 West Area landscape lacks evidence of water erosion and, except for localized areas of short steep slopes, surface runoff appears negligible. At present, surface erosion at the 216-Z-8 site appears to be negligible.

### Vadose Zone Hydrology

The rapid infiltration of precipitation and subsequent deeper percolation into the vadose zone create the potential for water to reach wastes beneath the 216-Z-8 French drain. Depth of water penetration is a function of precipitation duration and intensity. Long-term lysimeter experiments have been conducted at Hanford to assess precipitation penetration depths under near-natural conditions (Jones, 1978; Brown and Isaacson, 1977). Moisture was found to penetrate to depths greater than 5 m during the experiments (less than 10 yr), but deeper percolation was negligible.

The highest soil moisture levels and deepest moisture penetrations may be expected in early spring when low evapotranspirative demand has allowed winter snow and rain to accumulate in the soil. During years of extraordinarily high winter precipitation, some ground-water recharge through vadose zone transport may be possible. Normally, the deeply penetrating moisture moves back to the atmosphere in response to evapotranspirative demand. This upward movement of moisture can transport water-soluble radionuclides toward the soil surface (Routson et al., 1981). However, plutonium is not significantly water soluble under environmental conditions. The results of this study indicated that the bulk of the plutonium in the waste stream sorbed onto the sediments within a meter of the point of discharge with no detectable migration beyond a few meters from the bottom of the French drain.

### Ground-Water Hydrology

The water table beneath the 216-Z-8 site is 58.5 m below the ground surface, 145 m above mean sea level. The unconfined aquifer recharges naturally in the Cold Creek and Dry Creek Valley areas west of the 200 Areas. As discussed in the section on Vadose Zone Hydrology, there is

negligible recharge from the 200 West Area surface to the ground water from precipitation. Due to the relatively small volume of aqueous waste discharged at the 216-Z-8 site and the nature of the waste, significant vadose water transport of plutonium to the ground water is unlikely.

## GEOLOGY

The geology of the 200 Separations Area is discussed in detail by Tallman et al. (1979). For the current study, well 299-W15-202 was drilled to a final depth of 53.6 m below the surface to determine sediment distribution beneath the 216-Z-8 site. The results of sediment size (granulometric) analysis were used to construct a lithologic log of the well (Figure 9). The results are reported using an abbreviated sediment size notation. In the notation, the silt and clay size fraction is indicated by "m," the sand size fraction by "S," and gravel by "G." The major component in each sample is indicated by the last letter in the notation, and preceding letters, if present, are modifiers indicating additional components. Thus, "mS" indicates a silty sand, while "Sm" indicates a sandy silt or clay. A letter in parentheses indicates that the fraction is modified by the term "slightly." Thus, "(m)S" indicates a slightly silty sand.

In addition to the major sediment classification, the sand classification is divided into five subdivisions. The subdivisions are indicated by a notation within a bracket that modifies the sand,

where

VC = very coarse  
 C = coarse  
 M = medium  
 F = fine  
 VF = very fine.

Thus, (m)G[F-VF]S would indicate a slightly silty, gravelly, fine to very fine sand. The information for each well was used to determine the sediment stratigraphy for the 216-Z-8 site.

Figure 10 illustrates a simplified stratigraphic cross section of the sediments beneath the 200 West Area near the 216-Z-8 French drain system. The Separations Area is located on a broad bar of sand and gravel, locally called the 200 Areas Plateau, deposited by the spreading flood waters of catastrophic Pleistocene floods. The unconsolidated sands and gravels deposited during the floods are informally identified as the Hanford formation. At the 216-Z-8 site, the Hanford formation is 34 m thick.

A surficial eolian deposit near the 216-Z-8 site was derived from the wind reworking the underlying Hanford Formation sediments. The deposit was removed during excavation of the site and returned as part of the backfill. This deposit is identified by the finer texture of the backfill compared with the underlying sediments of the Hanford Formation.

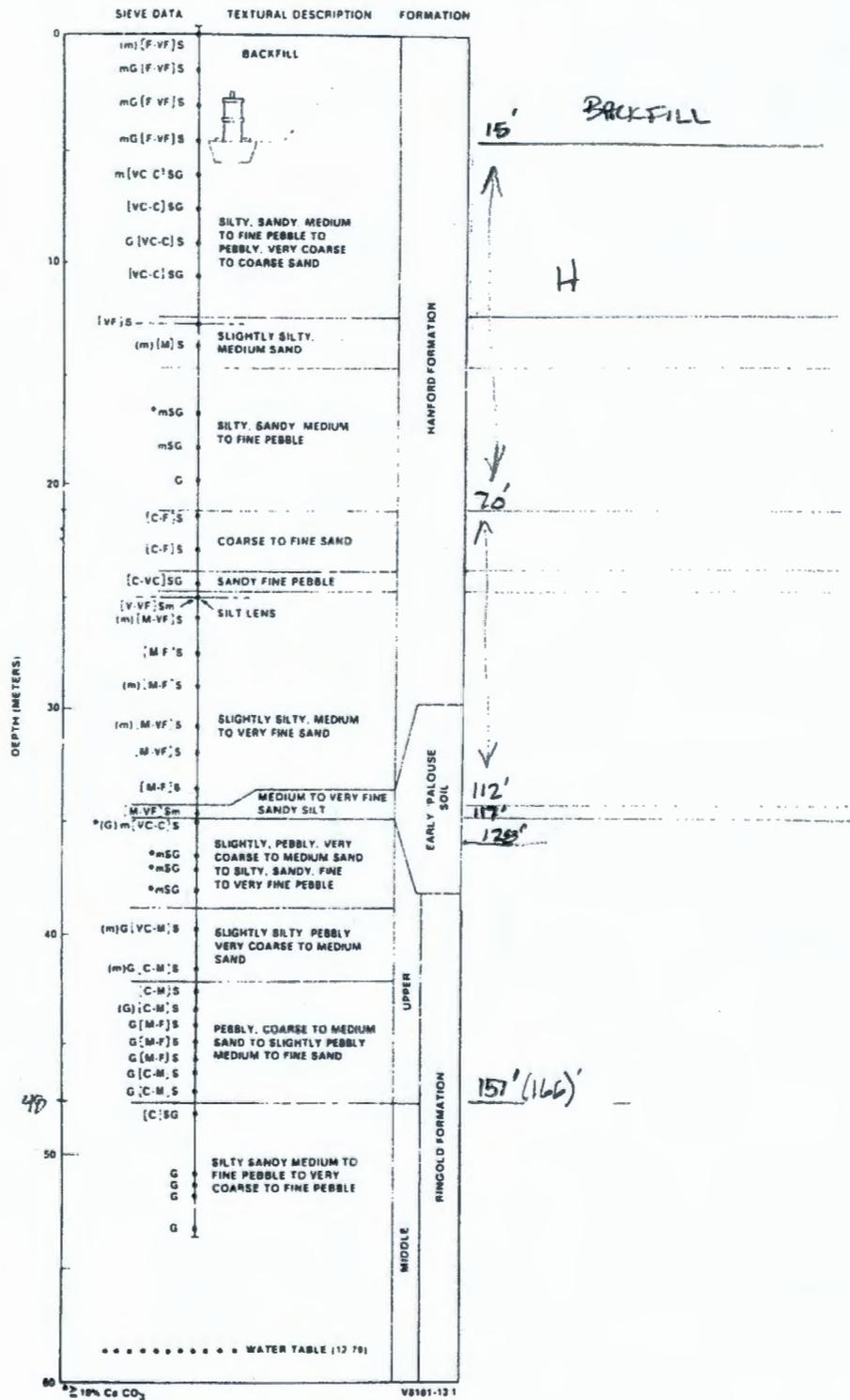


FIGURE 9. Lithologic Log for Well 299-W15-202.

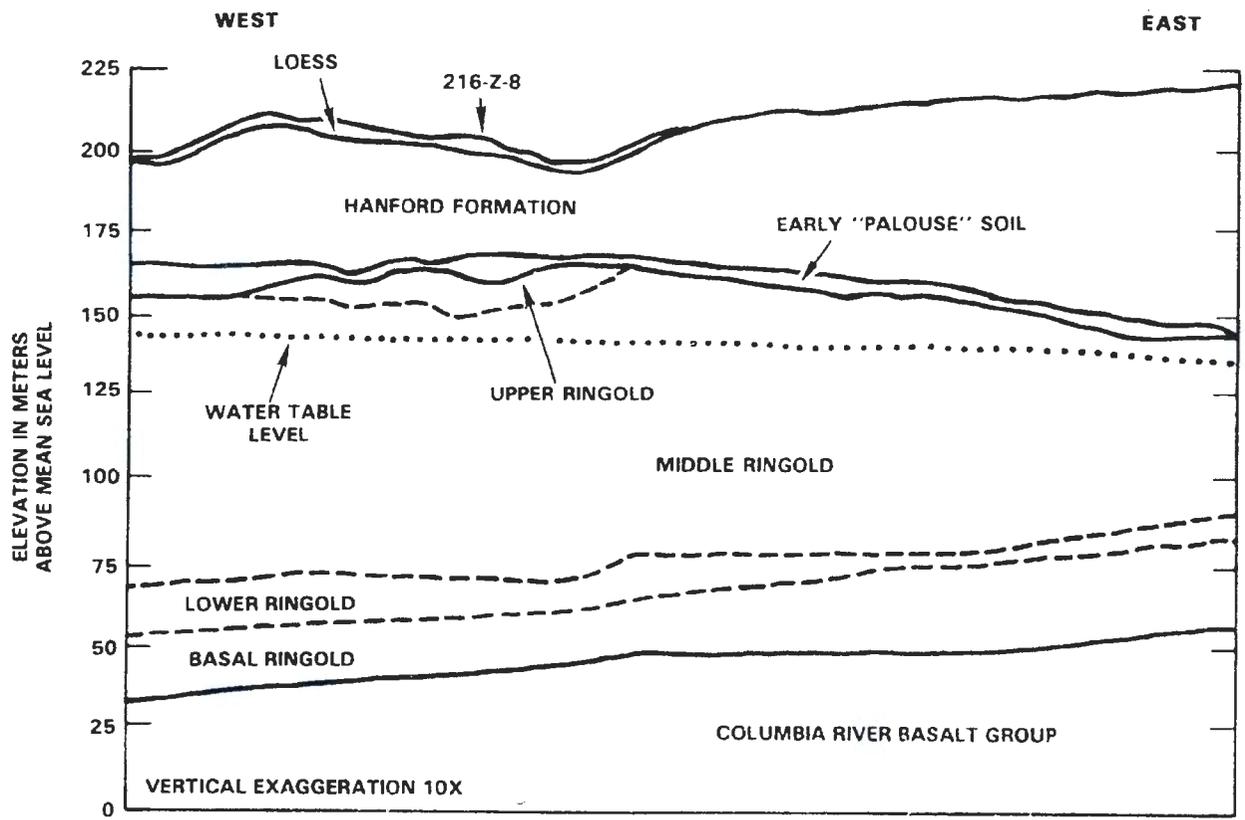


FIGURE 10. Geological Cross Section Through the 200 West Chemical Separations Area (Tallman et al., 1979).

The fine sand and silt below the Hanford Formation is identified as early "Palouse" soil. The soil is so named because of its similarity to the loess soil areas of the same name in eastern Washington. Palouse soil is thought to be the wind-reworked and wind-redeposited fine-grained sand and silt of the underlying Ringold Formation sediments. Beneath the 216-Z-88 site, the surface of the early Palouse soil is at a depth of 34 m and is 1 m thick.

Beneath the early Palouse soil is the Ringold Formation, which has four units: upper, middle, lower, and basal. The upper Ringold sediments are primarily medium-to-fine sands with silt and clay interlayers. The surface of the upper Ringold unit is identified by a well-developed caliche horizon. At the 216-Z-8 site, the surface of the upper Ringold is at a 35-m depth and the unit is 13 m thick. The middle Ringold bed consists of well-rounded pebbles and small cobbles with coarse-to-fine sand and silt filling the interstitial spaces. The single deep characterization well (299-W15-202) was emplaced to a depth of 53.6 m; the middle Ringold was the lowest stratigraphic unit identified. The surface of the middle Ringold occurs at a depth of 48 m below the ground surface. The water table is in the middle Ringold at a depth of 58.5 m below ground surface, 145 m above sea level (Tallman et al., 1979). At the 216-Z-8 site, the middle Ringold's thickness is estimated as 80 m.

The lower Ringold ranges from a silty coarse-to-medium sand to a sandy silt. Beneath the site, the thickness of the lower Ringold is estimated as 13 m. The basal Ringold bed consists of a silty, sandy gravel. Beneath the 216-Z-8 site, the thickness of the unit is estimated as 18 m. The basal Ringold bed overlies the Elephant Mountain Member of the Saddle Mountains Basalt of the Columbia River Basalt Group. The entire basalt group beneath the Hanford Site consists of a 1,500-m-thick sequence of basalt flows and interbedded clastic and volcaniclastic sediments.

## CURRENT STUDY

The primary objective of the 216-Z-8 characterization was to define the plutonium and americium distribution beneath the French drain. This information would provide the input required for engineering evaluation of alternatives for the ultimate disposal of the TRU-contaminated sediments. The information would also provide the input to modeling efforts of similar waste disposal sites. A secondary objective was to evaluate the suspected silica storage tank leak. Because information on the contents of the tank was available, and was of interest as the source of the waste solution, this information is also presented. The general approach was to drill wells near the tank and the French drain to obtain sediment samples. Selected sediment samples were characterized geologically and radiologically.

## WELL DRILLING AND SAMPLING

The wells were drilled using a cable tool drilling rig and a drive barrel sampler (Price et al., 1979). The sampler was surveyed for radioactivity with each sediment sample obtained using portable radiation survey instruments. If no radioactive contamination were detected on the sampler, the sediments in the lower 25 cm of the sampler were emptied into a plastic sample bag and surveyed again. If no contamination were detected on the sediments, samples were collected for storage at approximately 1.5-m increments, or at each observable lithology change. Approximately 1 kg of material was collected at each location. The remaining sediments were then emptied in the same manner, surveyed, and packaged for disposal. Contaminated material was packaged according to applicable requirements for handling and shipping of contaminated TRU material and placed in a separate radioactive material storage. Using the portable radiation survey instruments, only one sediment sample was determined to be contaminated.

## ANALYTICAL TECHNIQUES

Selected sediment samples were analyzed quantitatively for sediment size distribution and plutonium and americium concentrations to determine the geology and radionuclide distribution at the 216-Z-8 site. Portable radiation survey instruments provided only a qualitative measure of contamination and were used only as a guide for sample handling.

Granulometric Analysis

Sediment samples were dry-sieved using nested sieves, which were shaken mechanically by a Rotap shaker (Fecht and Price, 1977). Samples were sieved into seven size fractions, listed in Table 1. Sediment samples were collected and stored in glass jars with a mouth diameter of

approximately 7 cm. The amount of material greater than 7 cm was estimated at the time of the initial sampling and recorded in the well logs. The sediments retained on each sieve were weighed and recorded. Depending upon the weight percent of the various size fractions, the sediment samples were categorized into 1 of 19 sediment classes shown in Figure 11, using the computer program ROC (Fecht and Price, 1977).

TABLE 1. Particle Size Nomenclature.

Particle designation	Particle diameter, mm
Gravel	8-2
Sand	
Very Coarse	2-1
Coarse	1-0.5
Medium	0.5-0.25
Fine	0.25-0.125
Very Fine	0.125-0.0625
Silt (and clay)	<0.0625

NOTE: Nomenclature per Wentworth, 1922.

#### X-Ray and Gamma Ray Energy Analysis

All sediment samples obtained during drilling were nondestructively analyzed (NDA) using a lithium-drifted silicon [Si(Li)] detector system. Approximately 100-g aliquots of each sample were measured. The plutonium X-rays in the 17-keV region and the  $^{241}\text{Am}$  gamma ray at 59.6 keV were counted using appropriate calibration standards. The system had a detection limit for plutonium of 2 nCi/g of sediment and for americium of 0.1 nCi/g of sediment. One sample exceeded the detection limit. This sample was in well 299-W18-202, at a depth of 7.6 m. A plutonium concentration of  $2.5 \pm 2.0$  ( $1\sigma$ ) nCi/g of sediment was reported.

#### Alpha Energy Analysis

Due to the low level of activity found in the samples using the Si(Li) detector, TRU concentrations were determined by alpha energy analysis (AEA) (also called alpha spectrometry). The procedure required chemical digestion of the sediment using nitric-hydrofluoric acids, extraction and purification of the plutonium and americium, and electroplating of the purified elements onto stainless steel planchettes. The alpha particle energy spectrum was then counted using AEA.

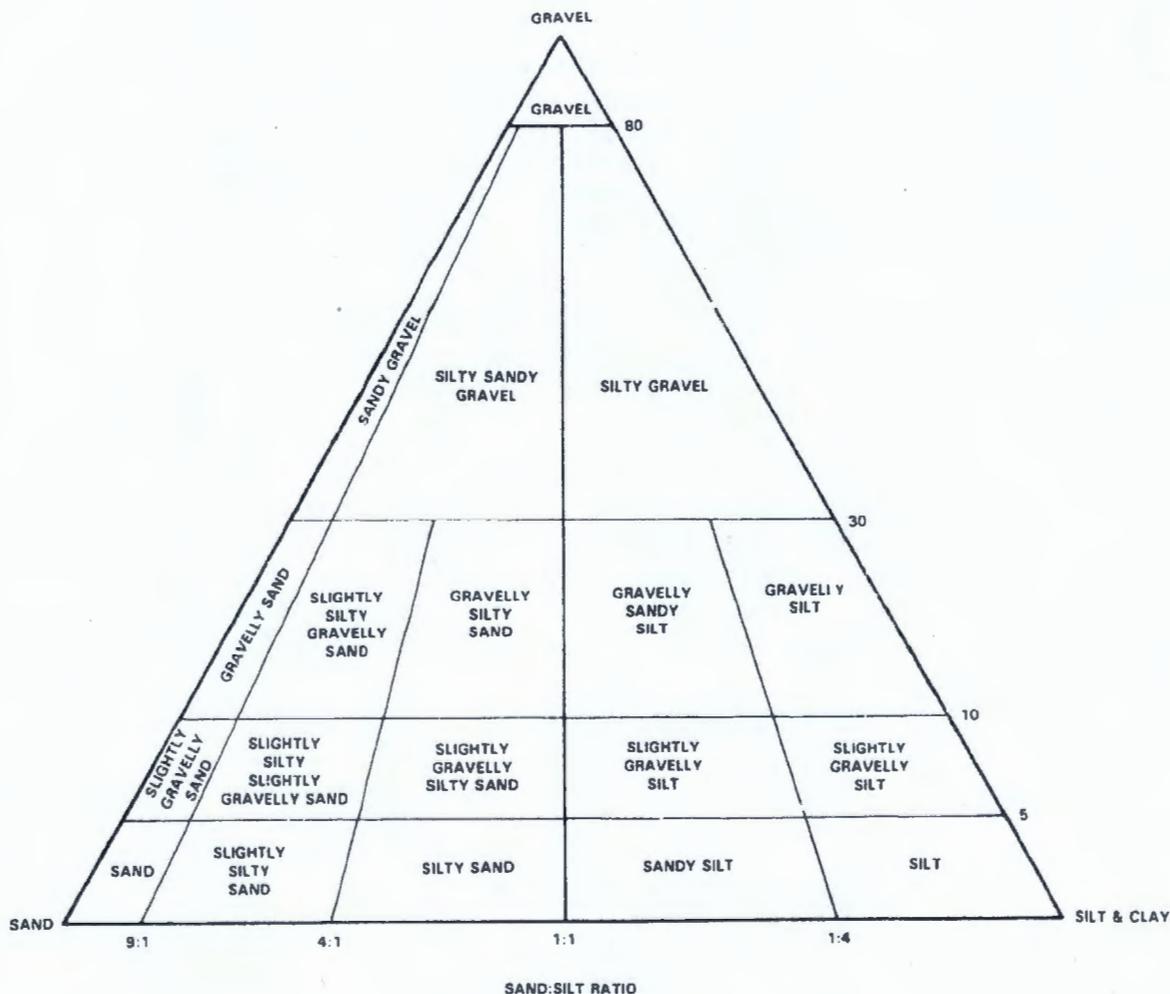


FIGURE 11. Sediment Classification (modified after Folk, 1974).

Normally, low levels of TRU contamination are determined by AEA. The detection limit for the AEA detector was estimated as  $0.02 \pm 0.02$  ( $1\sigma$ ) pCi/analysis, or  $0.002 \pm 0.002$  ( $1\sigma$ ) pCi/g of sediment for a 10-g sample, assuming perfect recovery. Many factors caused the actual limit of detection for AEA to exceed the optimal value. Losses resulting from the digestion, chemical extraction, purification, and counting steps increased the detection limit. For a single analysis, evaluation of the data presented in the "Results" section suggested a detection limit on the order of  $0.005 \pm 0.005$  ( $1\sigma$ ) pCi/g of sediment. However, the uncertainty introduced by the laboratory procedures were generally small compared with the variability of TRU concentration in the original sediment sample and those introduced during sample handling and selection. Based on experience with multiple analyses of 10-g duplicates for a well-characterized sample, a practical detection limit was estimated to be below  $0.1 \pm 0.1$  ( $1\sigma$ ) pCi/g of sediment. Although analytical values are reported for individual analyses

below this limit, the uncertainty was based solely on counting statistics and is not to be taken as an estimate of true sample variability. In this report, for evaluation of the TRU distribution, 0.1 pCi/g of sediment was considered the lower limit of detection for the AEA system. Analyses were made for  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{241}\text{Am}$ . The detector system did not have the resolution to separate the  $^{239}\text{Pu}$  from the  $^{240}\text{Pu}$  alpha energies, and they are reported together as  $^{239}\text{Pu}$ . All of the AEA were performed by Eberline Instrument Corporation\* (Eberline).

Interpretation of the TRU distribution in the sediment depended on the AEA. To supplement Eberline's internal quality control program and to maintain confidence in the reported results, a limited quality assurance program was implemented by Rockwell Hanford Operations (Rockwell) that included the use of double blind standards and duplicate samples. The standard material was provided by the DOE Environmental Measurements Laboratory† (EML). The material was not considered to be a true standard, but to be what is known as a consensus standard or natural matrix standard. The material was a mixture of soil collected at Oak Ridge National Laboratory‡ and the Savannah River Plant§. The material has been analyzed at least three times by 10 laboratories to determine the concentration. The estimated concentration of plutonium in the sediment as reported by EML was  $0.5 \pm 0.05$  pCi  $^{239}\text{Pu}/\text{g}$  and  $0.027 \pm 0.005$  pCi  $^{238}\text{Pu}/\text{g}$ .

Five aliquots of the consensus standard were sent as double blind standards in the batch of sediment samples to be analyzed. The results reported by Eberline (Table 2) agreed with the estimated concentration reported by EML and, when evaluated with the results reported for duplicate sediment samples, provided satisfactory confidence in the interpretation of the TRU distribution.

TABLE 2. Reported Analytical Results for Consensus Standard.

Sample	$^{238}\text{Pu}$ , pCi/g*	$^{239}\text{Pu}$ , pCi/g*	$^{241}\text{Pu}$ , pCi/g*
1	$2.28\text{E}-2 \pm 14.9\%$	$3.53\text{E}-1 \pm 5.2\%$	$2.10\text{E}-1 \pm 9.7\%$
2	$3.74\text{E}-2 \pm 16.6\%$	$4.09\text{E}-1 \pm 7.6\%$	$2.64\text{E}-1 \pm 11.0\%$
3	$2.99\text{E}-2 \pm 17.5\%$	$5.64\text{E}-1 \pm 7.2\%$	$2.36\text{E}-1 \pm 11.1\%$
4	$3.21\text{E}-2 \pm 17.0\%$	$5.37\text{E}-1 \pm 6.5\%$	$2.24\text{E}-1 \pm 6.5\%$
5	$3.35\text{E}-2 \pm 19.8\%$	$4.44\text{E}-1 \pm 7.8\%$	$2.29\text{E}-1 \pm 7.6\%$
Average	$3.11\text{E}-2 \pm 17.4\%$	$4.61\text{E}-1 \pm 19.1\%$	$2.33\text{E}-1 \pm 8.6\%$

\*Uncertainty reported at 1 $\sigma$  as a percentage of the reported value.

\*Eberline Instrument Corporation, Albuquerque, New Mexico.

†DOE Environmental Measurements Laboratory, 376 Hudson Street, New York, New York 10014.

‡Oak Ridge National Laboratory, Oak Ridge, Tennessee.

§Savannah River Plant, Aiken, South Carolina.

Neutron Activation Assay

Several measurements were made in the silica storage tank by a neutron activation technique using copper foils (Brodzinski, 1979). In the procedure, a copper foil was exposed to a neutron flux, which activated the  $^{63}\text{Cu}$  to  $^{64}\text{Cu}$ , by the reaction  $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ . The gamma ray production from the  $^{64}\text{Cu}$  ( $t_{1/2} = 12.7$  h) was measured in the laboratory and used to calculate the neutron flux. Making assumptions about the ratio of the TRU isotopes and the matrix composition, an estimate of TRU concentration was calculated. The technique averaged the neutron flux; hence, averaged the concentration over a large surrounding volume. The technique provided a quick survey procedure for determining the presence of TRU elements.

## RESULTS AND DISCUSSION

The results of the study are presented in three sections:

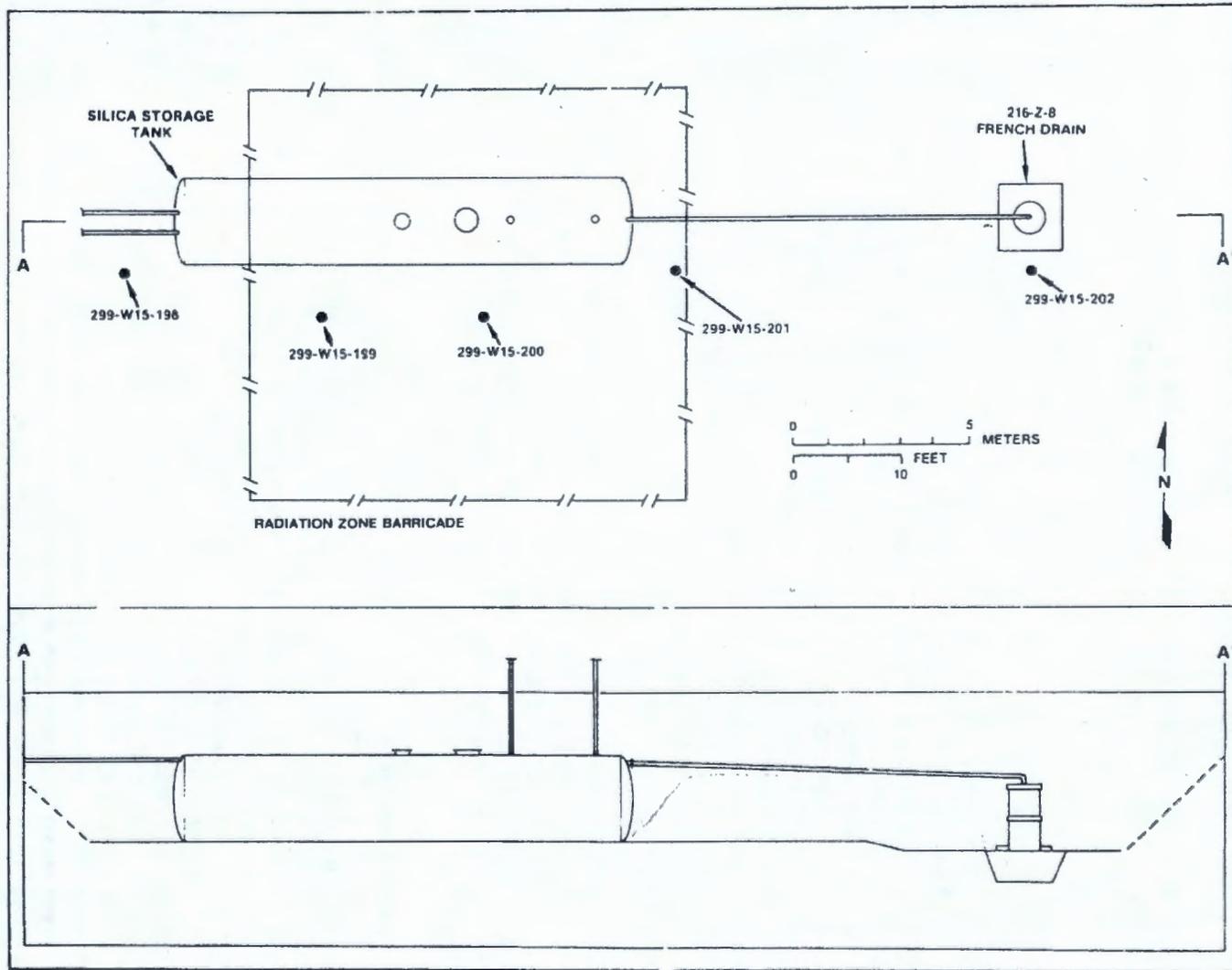
- Delineation of the TRU distribution beneath the French drain
- Evaluation of the suspected silica storage tank leak
- Discussion of the plutonium inventory for the 216-Z-8 French drain system.

## FRENCH DRAIN

A single well (299-W15-202) was drilled less than 1 m south of the French drain. The location of the well in relation to the French drain is shown in Figure 12. The waste plume from the French drain was anticipated to be only a few meters deep due to the basic nature of the waste and the small volume of waste discharged. This well, nevertheless, was drilled to a final depth of 53.6 m to obtain information on sediment distribution in support of general site studies. The results of the sieve analyses of selected sediment samples were used to construct the lithologic log shown in Figure 9. (Notation for granulometric data was discussed in the "Analytical Techniques" section.) The major formations were identified, and separate units based on textural differences were proposed. The formations were discussed in the "Geology" section.

The results of AEA for  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Am}$  content of selected sediment samples from well 299-W15-202 are given in Table 3. Samples were identified by the well number and the depth in meters. Duplicate samples were analyzed at selected depths and were indicated by the A and B designations. Results are reported in picocuries per gram of sediment with the uncertainty of one standard deviation as a percentage of the reported value.

Analytical results were used to construct a cross section through the French drain showing an interpretation of the concentration distribution of  $^{239}\text{Pu}$  and  $^{241}\text{Am}$ . To draw the isopleths, the concentration of the radionuclides was assumed to be symmetrical away from the French drain, with the activity generally decreasing radially away from the bottom of the drain. The estimated distribution of plutonium and americium is shown in Figures 13 and 14, respectively. Isopleths are shown for 0.1, 1, 10, 100, 1,000, and 10,000 pCi/g. The 10,000-pCi Pu/g isopleth (10 nCi/g of sediment) was hypothetical and suggested that there is no more than a 1-m penetration of plutonium contamination levels at or greater than 10,000 pCi/g. For a basic solution, laboratory experiments demonstrated that plutonium was strongly sorbed from solution onto sediment (Rhodes, 1952, 1957; Nishita, 1979), and the highest concentration of plutonium would be expected at the bottom of the French drain at the interface between the gravel backfill and the original sediments. In addition, any particulate material containing plutonium that was not settled in the silica storage tank would be filtered out at the interface, as has been observed in other waste disposal facilities (Price and Ames, 1976).



V8101-13.3

FIGURE 12. Plan View and Cross Section of the 216-Z-8 Site Showing Location of Wells

TABLE 3. Transuranic Activity Results for  
Selected Samples from Well 299-W15-202.  
(Sheet 1 of 2)

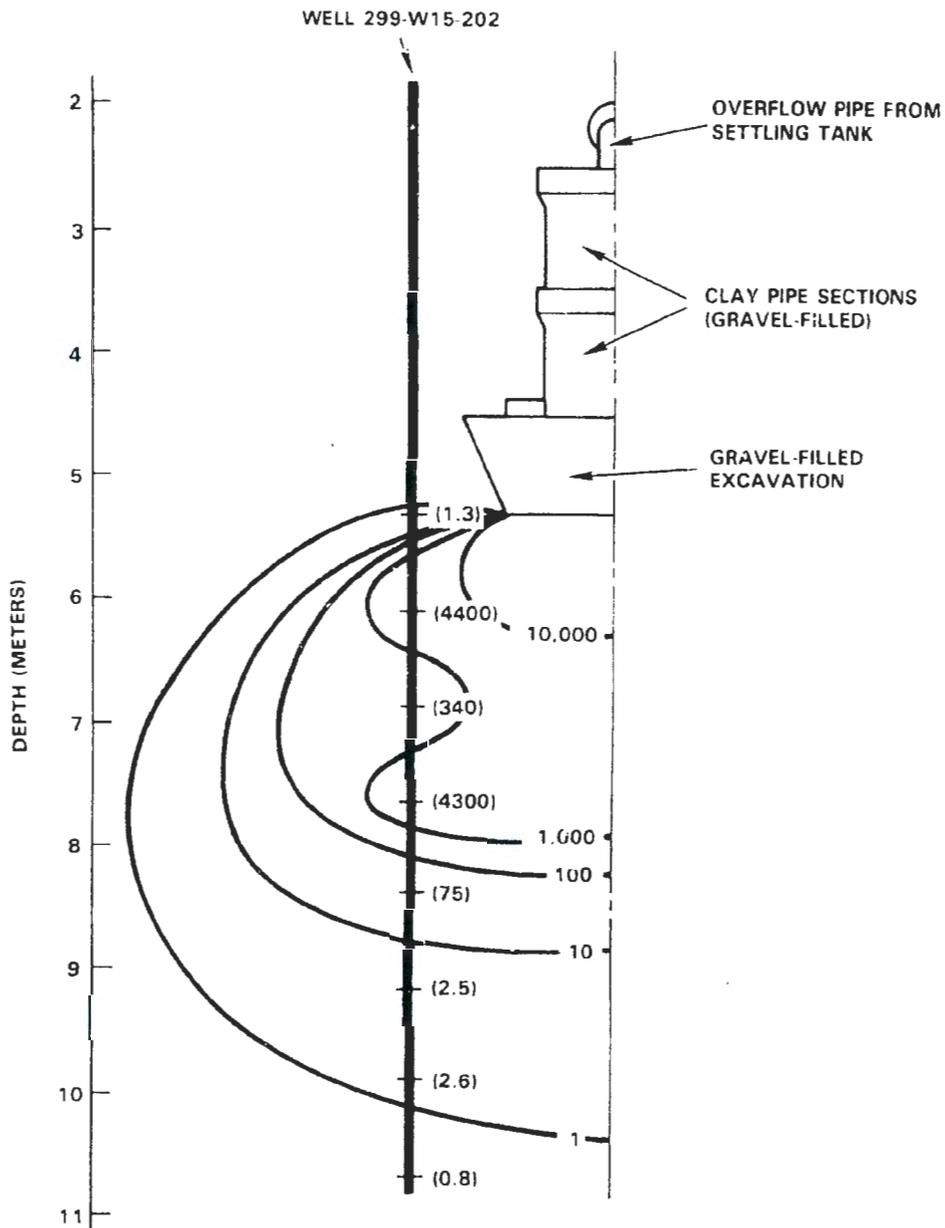
Depth, m	Type of analysis	pCi/g (dry)*
5.3 A	<sup>238</sup> Pu	1.40 E-02 ± 28.9%
	<sup>239</sup> Pu	1.22 E+00 ± 6.8%
	<sup>241</sup> Am	8.09 E-02 ± 22.5%
5.3 B	<sup>238</sup> Pu	1.43 E-02 ± 24.4%
	<sup>239</sup> Pu	1.42 E+00 ± 7.1%
	<sup>241</sup> Am	9.01 E-02 ± 24.6%
6.1	<sup>238</sup> Pu	7.34 E-01 ± 1.9%
	<sup>239</sup> Pu	4.41 E+03 ± 1.6%
	<sup>241</sup> Am	4.57 E+02 ± 1.7%
6.9	<sup>238</sup> Pu	6.03 E+00 ± 3.5%
	<sup>239</sup> Pu	3.41 E+02 ± 1.4%
	<sup>241</sup> Am	4.58 E+01 ± 2.2%
7.6 A	<sup>238</sup> Pu	6.63 E+01 ± 1.5%
	<sup>239</sup> Pu	3.96 E+03 ± 1.2%
	<sup>241</sup> Am	3.68 E+02 ± 1.8%
7.6 B	<sup>238</sup> Pu	7.50 E+01 ± 1.4%
	<sup>239</sup> Pu	4.62 E+03 ± 1.2%
	<sup>241</sup> Am	3.71 E+02 ± 1.6%
8.4 A	<sup>238</sup> Pu	1.66 E+00 ± 6.7%
	<sup>239</sup> Pu	8.76 E+01 ± 1.7%
	<sup>241</sup> Am	9.81 E+00 ± 3.1%

TABLE 3. Transuranic Activity Results for  
Selected Samples from Well 299-W15-202.  
(Sheet 2 of 2)

Depth, m	Type of analysis	pCi/g (dry)*
8.4 P	$^{238}\text{Pu}$	1.08 E+00 $\pm$ 7.3%
	$^{239}\text{Pu}$	6.31 E+01 $\pm$ 1.6%
	$^{241}\text{Am}$	5.57 E+00 $\pm$ 3.3%
9.1	$^{238}\text{Pu}$	2.02 E-01 $\pm$ 27.1%
	$^{239}\text{Pu}$	2.47 E+00 $\pm$ 5.5%
	$^{241}\text{Pu}$	1.81 E-01 $\pm$ 20.4%
9.9	$^{238}\text{Pu}$	2.09 E-02 $\pm$ 120.0%
	$^{239}\text{Pu}$	2.60 E+00 $\pm$ 5.5%
	$^{241}\text{Am}$	1.22 E-01 $\pm$ 31.7%
10.7 A	$^{238}\text{Pu}$	2.29 E-01 $\pm$ 18.6%
	$^{239}\text{Pu}$	9.20 E-01 $\pm$ 8.6%
	$^{241}\text{Am}$	9.51 E-02 $\pm$ 34.0%
10.7 B	$^{238}\text{Pu}$	2.48 E-01 $\pm$ 14.6%
	$^{239}\text{Pu}$	7.18 E-01 $\pm$ 8.9%
	$^{241}\text{Am}$	1.13 E-01 $\pm$ 25.3%

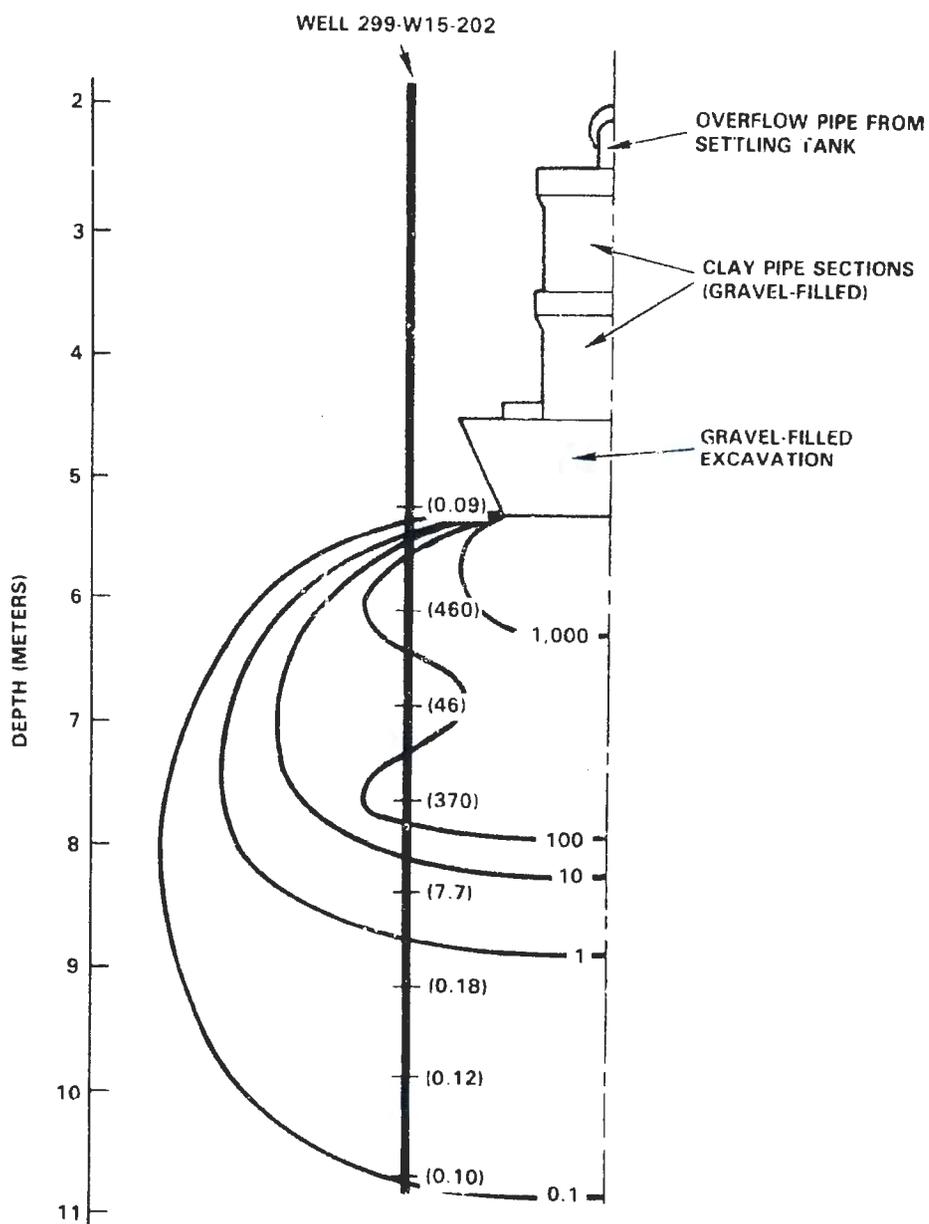
NOTE: A and B indicate pairs of duplicate samples.

\*Uncertainty reported at 1 $\sigma$  as a percentage of the reported value.



V8010-8.7

FIGURE 13. Plutonium-239 Concentrations (in pCi/g) Beneath the 216-Z-8 French Drain. (Numbers in parentheses are reported results from Table 3 for individual sediment samples or average of duplicates. Isopleths shown for 1, 10, 100, 1,000, and 10,000 pCi/g.)



V8010-8.8

FIGURE 14. Americium-241 Concentrations (in pCi/g) Beneath the 216-Z-8 French Drain. (Numbers in parentheses are reported results from Table 3 for individual sediment samples or average of duplicates. Isopleths shown for 0.1, 1, 10, 100, and 1,000 pCi/g.)

The plutonium and americium distributions appeared to be similar; the ratio of  $^{241}\text{Am}$  to  $^{239}\text{Pu}$  was approximately 0.10. The abundance of  $^{241}\text{Pu}$  in freshly processed, weapons grade plutonium was estimated as 0.6 wt% (Emery and Garland, 1974), with no americium present. Americium-241 activity gradually increases from the decay of  $^{241}\text{Pu}$  ( $t_{1/2} = 14.7$  yr). The ratio of  $^{241}\text{Am}/^{239}\text{Pu}$  reaches a maximum 68 yr after production. The ratio reaches a value of 0.1 approximately 20 yr after production, which is consistent with the years of active operation of the 216-Z-8 facility (1955 to 1962). This suggests that the americium present in the sediments beneath the French drain was derived from the in situ decay of  $^{241}\text{Pu}$ , not from a separate source of waste discharge as has been observed at the 216-Z-1A waste disposal facility (Price et al., 1979).

Plutonium activity decreased rapidly with distance from the bottom of the French drain. The background level of plutonium at the 216-Z-8 site was on the order of 1 pCi/g of sediment (see discussion in the following section, "Silica Storage Tank"). If the 1-pCi/g isopleth is used to define the limit of the waste plume beneath the French drain, the plume extends approximately 5 m from the bottom of the gravel-filled excavation. Using the observed  $^{241}\text{Am}/^{239}\text{Pu}$  ratio of 0.10, the extent of the americium contamination is defined by the 0.1 pCi/g of sediment isopleth.

#### SILICA STORAGE TANK

The 1974 project to remove the solution contents of the silica storage tank considered briefly the following four possible explanations for the 27,500-L discrepancy in tank contents (see "Monitoring and Previous Data" section):

1. Part of the tank's contents had been transferred previously
2. The liquid had evaporated
3. A measurement error was made
4. Part of the tank's contents had leaked to the ground.

Available information suggested that explanation 2, that the liquid had evaporated, was not feasible. There are no records to substantiate any of the other explanations. Of the remaining explanations, only the explanation that the tank had leaked can be further evaluated by obtaining new information. If the tank had leaked, plutonium in the waste solution would be sorbed onto the sediment around the tank. If no plutonium contamination is found, then the presumption could be made that the tank did not leak.

To evaluate the possibility of a tank leak, four shallow wells were drilled near the tank to a depth of 7.6 m (Fig. 12). The results of granulometric analysis of selected sediment samples from the four wells, and from the well drilled near the French drain, were used to construct a geologic cross section (Fig. 15). The location of the wells were selected



based on evaluation of the size and distribution of a waste plume from the tank. In an idealized case, 27,500 L (27.5 m<sup>3</sup>) of solution injected at a point source would spread in homogeneous sediments as a roughly spherical volume until it reached the moisture content of the surrounding sediments (~10 vol%). At this point, a volume of approximately 280 m<sup>3</sup> would have been wetted by the waste solution. The radius of this volume would be 4 m. In fact, the sediments are not homogeneous and the idealized case does not consider the volume of sediment displaced by the tank. Experience from the characterization of other liquid waste disposal sites indicates that the waste solution would disperse downward and laterally until it contacted the interface between the backfill and the unexcavated sediments. The waste solution would preferentially spread laterally along this disturbed zone beneath the tank. Thus, the distance the waste solution could be found to have spread from the discharge point would be expected to exceed 4 m. The location of the wells around the tank were selected so that any point on the tank was within 4.5 m of a well. At least one well would be expected to penetrate the volume of sediment wetted by the waste solution.

In considering the level of plutonium activity that might be encountered due to a tank leak, reference is made to the distribution of TRU activity observed beneath the French drain (Fig. 13 and 14). Because the volume of waste (9,590 L to the French drain versus 27,500 L from the tank) and the quantity of plutonium (48 g to the French drain versus a possible 160 g from the tank) are roughly comparable, a crude estimate of the level of activity that could be encountered can be made. At a distance of 4 to 5 m from the bottom of the French drain, plutonium activity is on the order of 1 to 10 pCi/g of sediment. If a breach in the tank wall occurred at the maximum distance from the wells (i.e., 4.5 m to the nearest well), activity on the order of a few to a few tens of picocuries might be anticipated in a well that penetrated the plume. A well located closer to the source of the solution would be expected to have higher activity since the plutonium in the waste solution is observed to sorb quickly onto the sediment.

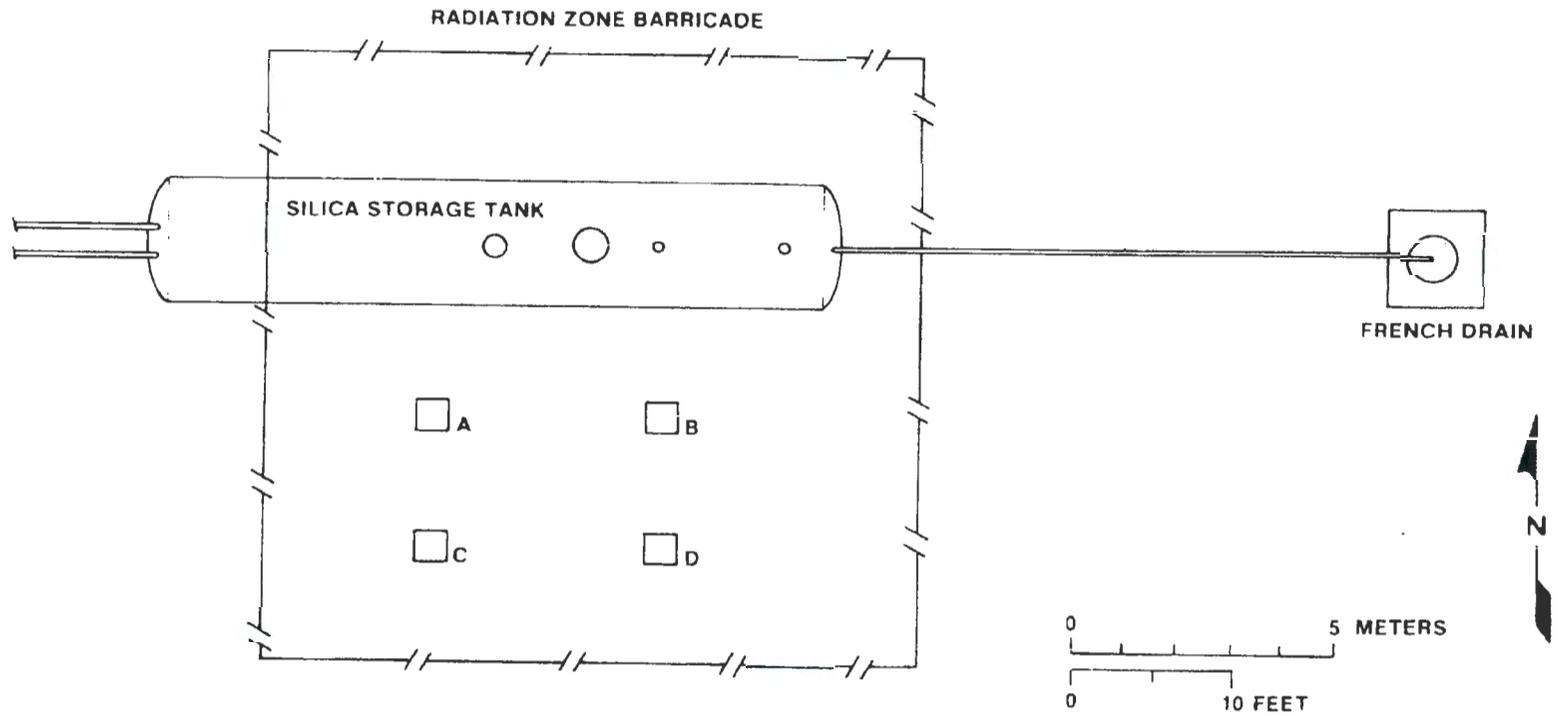
No activity was detected using portable radiation survey instruments in any sample collected in the four wells drilled around the tank. No activity above the limit of detection was found using the Si(Li) detector. Two sediment samples collected from each well at 4.6 and 6.1 m were analyzed for <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>241</sup>Am by AEA. The 4.6-m samples were in the unexcavated sediments just below the backfill-unexcavated sediment interface at 4.3 m. The results are reported in Table 4.

As part of a separate study of the surface environment at the 216-Z-8 site (see Acknowledgments), four core samples were taken south of the silica storage tank at the locations shown in Figure 16. The cores were taken to a depth of 30 cm using a 5.1-cm-diameter split-tube sampler. The cores were subdivided into samples from 0 to 15 cm deep, and 15 to 30 cm deep. Plutonium activity for each of the eight samples was determined by AEA at the Analytical Laboratories of Rockwell. Results are reported in Table 5. The values ranged from approximately 1 pCi/g of sediment to a high of 44 pCi/g of sediment. With the exception of the highest value (44 pCi/g of sediment), the values were comparable to those reported previously for the four wells.

TABLE 4. Transuranic Activity for Selected Samples from Wells 299-W15-198, -199, -200, and -201.

Well	Depth, m	Type of analysis	pCi/g (dry)*
299-W15-198	4.6	$^{238}\text{Pu}$	4.11 E-02 $\pm$ 14.1%
		$^{239}\text{Pu}$	2.14 E+00 $\pm$ 6.5%
		$^{241}\text{Am}$	1.33 E-02 $\pm$ 66.5%
299-W15-198	6.1	$^{238}\text{Pu}$	3.68 E-02 $\pm$ 15.3%
		$^{239}\text{Pu}$	1.34 E+00 $\pm$ 6.9%
		$^{241}\text{Am}$	2.90 E-02 $\pm$ 28.9%
299-W15-199	4.6	$^{238}\text{Pu}$	1.18 E-02 $\pm$ 28.5%
		$^{239}\text{Pu}$	4.12 E-01 $\pm$ 7.7%
		$^{241}\text{Am}$	2.06 E-02 $\pm$ 34.9%
299-W15-199	6.1	$^{238}\text{Pu}$	3.70 E-02 $\pm$ 17.4%
		$^{239}\text{Pu}$	2.12 E+00 $\pm$ 7.2%
		$^{241}\text{Am}$	4.15 E-02 $\pm$ 23.4%
299-W15-200	4.6	$^{238}\text{Pu}$	1.29 E-02 $\pm$ 23.6%
		$^{239}\text{Pu}$	1.94 E+00 $\pm$ 7.8%
		$^{241}\text{Am}$	7.57 E-02 $\pm$ 16.8%
299-W15-200	6.1	$^{238}\text{Pu}$	2.54 E-02 $\pm$ 20.7%
		$^{239}\text{Pu}$	1.01 E+00 $\pm$ 7.1%
		$^{241}\text{Am}$	5.23 E-02 $\pm$ 29.6%
299-W15-201	4.6	$^{238}\text{Pu}$	3.92 E-02 $\pm$ 36.7%
		$^{239}\text{Pu}$	2.08 E+00 $\pm$ 16.9%
		$^{241}\text{Am}$	1.23 E-01 $\pm$ 17.3%
299-W15-201	6.1	$^{238}\text{Pu}$	9.05 E-02 $\pm$ 11.1%
		$^{239}\text{Pu}$	5.31 E+00 $\pm$ 6.8%
		$^{241}\text{Am}$	3.42 E-02 $\pm$ 21.6%

\*Uncertainty reported at 1 $\sigma$  as a percentage of the reported value.



VB101-13.3A

FIGURE 16. Location of Four 30-cm Cores Taken at the 216-Z-8 Site.

TABLE 5. Plutonium Activity  
in Four Cores Near the  
Silica Storage Tank.

Sample location*	Depth, cm	Pu, pCi/g
A	0-15	0.74
	15-30	7.80
B	0-15	1.80
	15-30	0.92
C	0-15	4.70
	15-30	1.10
D	0-15	44.00
	15-30	0.99

\*Sample location shown  
in Figure 15.

Examination of the analytical results reported in Tables 4 and 5 show that with the exception of the 4.6-m-deep sample for well 299-W18-199 (0.4 pCi/g), and the 0- to 15-cm sample from core D (44 pCi/g), the plutonium activity ranges from  $\sim$ 1.0 to 8.0 pCi/g of sediment. This level of activity is what was anticipated around the tank, but the apparent uniformity of activity in the four wells and the four cores is not what would be expected if the tank leaked. For 27,500 L of solution, perhaps two or three, but not four, of the wells drilled near the tank might be expected to intersect the plume. Although a larger volume for the waste plume could be postulated if the leak occurred during active waste disposal operations instead of after, plutonium activity would still be expected to decrease rapidly with distance from the point of release. The plutonium activity in either case would not be expected to be similar in all four wells, based on the proposed model of the transport process.

Other alternate sources for the plutonium activity at the 216-Z-8 site were briefly considered. Migration of a waste solution from another waste disposal facility was discounted because none of the facilities are close enough and have high enough plutonium activity to provide a likely source. Due to processing operations at Z Plant, levels of plutonium activity in surface soil samples above the regional background level of  $0.02 \pm 0.02$  ( $1\sigma$ ) pCi/g of sediment (Houston and Blumer, 1980) are measured downwind of Z Plant. Plutonium activity on the order of 1 pCi/g of sediment is reported by Wheeler and Law (1980). Such a source might explain activity measured in core samples taken from backfill, but would not explain the activity observed in samples from the wells taken from unexcavated, undisturbed sediment.

The alternate sources of plutonium activity considered do not provide an acceptable explanation for the observed distribution activity around the silica storage tank. The simple scenario used to estimate the distribution and concentration of plutonium due to the postulated tank leak does not provide an acceptable explanation either because the scenario does not match the actual observed distribution. Consequently, with the data obtained, the source of plutonium activity cannot be positively identified, and a leak cannot be confirmed. To resolve the question, additional wells would have to be drilled around the tank to define the contamination plume. This additional work was beyond the scope of the present study. Because the 216-Z-8 facility discharged liquid waste solution to the ground through the French drain, the additional effect of a leak of this same solution from the tank would be of little practical consequence. Thus, additional drilling to further define the plutonium distribution is not justified for evaluating a potential tank leak. However, the potentially large quantity of plutonium which remains in the tank needs to be further evaluated and some remedial action will probably be required to stabilize or remove the material. Because the bulk of the small quantity of plutonium released by a leak would still be expected to remain near the tank wall, any action involving excavation near the tank should include additional drilling. This drilling should identify any volume of soil containing elevated levels of plutonium activity required for planning an excavation and would also resolve the questions of whether the tank leaked or not.

To investigate the TRU inventory of the remaining content of the tank, copper foils were lowered on strings down the two vent risers. In each riser, two foils were positioned with one foil just above the sludge layer and the second foil approximately 30 cm above the sludge. The calculated neutron flux measurements for the four foils are given in Table 6. The neutron fluxes are in reasonable agreement, suggesting that the TRU distribution is approximately homogeneous in this portion of the tank.

If the effect of several factors influencing neutron flux were known or could be estimated, the TRU inventory could be estimated from the neutron flux measurements. The first and most important factor is the contribution to the neutron flux from  $\alpha, n$  reactions with light elements, primarily oxygen and fluorine. The contribution to the neutron flux is significantly greater due to the presence of fluorine than oxygen. Because calcium fluoride was produced in the reduction reaction to produce plutonium metal, the presence of significant quantities of fluorine is possible. There were no reported chemical analyses of the sludge from the 1974 study, and such sampling was beyond the scope of the present study. The plutonium content was calculated assuming fluorine concentrations of 0 and 5 wt% in the sludge.

The second factor is the moisture content of the sludge. The addition of water influences oxygen content, but primarily affects the volume of material (called the volume of interrogation) from which the neutron flux is measured. The volume of interrogation for 90% and 99% of the contribution to the measured flux is given in Table 7 for three moisture contents: 16, 42, and 70 wt%.

TABLE 6. Results of Neutron Flux Measurements by Copper Foils in the Silica Storage Tank.

Meters from top of sludge	Neutron flux n/cm <sup>2</sup> /s	Pu concentration, nCi/g					
		5 wt% fluorine <sup>a</sup>			0 wt% fluorine <sup>b</sup>		
		16 wt% water	42 wt% water	70 wt% water	16 wt% water	42 wt% water	70 wt% water
Riser 1							
~0.3	0.6798 ± 5.365%	2,540	4,150	6,070	10,020	16,400	24,600
<0.1	0.6865 ± 5.344%	2,560	4,190	6,130	10,120	16,560	24,200
Riser 2							
~0.4	0.6817 ± 5.355%	2,540	4,160	6,090	10,050	16,440	24,030
<0.1	0.8844 ± 5.321%	3,300	5,400	7,900	13,030	21,330	31,170

<sup>a</sup>Oxygen: fluorine = 19:1<sup>b</sup>Oxygen: fluorine = ∞

TABLE 7. Volume of Interrogation for Copper Foils.

Flux contribution, %	Moisture content*		
	16 wt%	42 wt%	70 wt%
90	820 L (58 cm)	440 L (47 cm)	230 L (38 cm)
99	2150 L (80 cm)	1450 L (70 cm)	830 L (58 cm)

\*Radius of sphere given in parentheses.

A third factor involves the geometry of the TRU distribution around the foil. The foils are calibrated for what is termed a  $4\pi$  model. For this model, the TRU material is assumed to be homogeneously distributed in the matrix and the foil measures the neutron flux at the center of an "infinite" sphere of the matrix. In reality, the sphere is only required to be large enough so that greater than 99% of the neutron flux is measured from within this volume. The sludge layer and the position of the foil above the sludge do not approximate the  $4\pi$  model, but the calibration of the  $4\pi$  model could be used with an appropriate geometry correction. For example, instead of a  $4\pi$  model, a  $2\pi$  model could be used. In the  $2\pi$  model, the foil is located on the flat surface of an infinite hemisphere and the TRU concentration would be double that reported for a  $4\pi$  model. Because the sludge layer is estimated to be too thin to approximate a  $2\pi$  model, the results calculated on the basis of a  $4\pi$  model would have to be multiplied by a factor greater than two to get the correct result. The appropriate geometry correction could possibly be estimated because the approximate dimensions are known.

However, there are sufficient uncertainties in the estimates of the chemical composition and water content of the sludge that the exercise was not considered useful. Thus, the use of the copper foils can only provide an estimate of the TRU activity. If an exact inventory for the tank is later determined to be needed, it would be more appropriate to obtain samples of the sludge for chemical, moisture, and TRU analyses.

The TRU concentration in the sludge was calculated for two different fluoride concentrations (0 and 5 wt%) and three moisture contents (16, 42, and 70 wt%) and assumed a  $4\pi$  geometry with a sediment matrix. The results are reported in Table 6 along with the neutron flux measurements of the four foils.

To estimate the plutonium inventory of the tank, the following assumptions were made.

- A moisture content of 42% is a reasonable approximation.
- The average value for the neutron flux is that represented by the foil at approximately 0.4 m in riser 2.
- The correction for the geometry is a factor greater than 2, but probably less than 5; i.e., the reported concentration in Table 6 must be multiplied by this factor.

With these assumptions, the concentration of plutonium varies from greater than 8,300 nCi/g (5% flourine) to greater than 33,000 nCi/g (0% flourine). Using the previous estimate of 4,000 L of sludge remaining in the tank and a density of  $1.5 \text{ g/cm}^3$ , the plutonium in the tank could range from greater than 650 g to greater than 2,550 g. Due to the many assumptions and uncertainties involved in the calculations, further speculation about the tank contents is not technically useful. Because most of the plutonium was associated with the sludge, the copper foil results generally support the estimated inventory made in 1974 of 1,400 g of plutonium in the sludge. The observations again suggest that the sludge had compacted into a dense (gelled) layer and was not resuspended effectively in the solution during pumping operations.

#### PLUTONIUM INVENTORY FOR THE 216-Z-8 FACILITY

When discussing the waste disposal history, monitoring, and previous data for the 216-Z-8 facility, two estimates were given for the plutonium inventory at the site. The official estimate of plutonium discharged to the 216-Z-8 French drain was reported as 48 g (Hanson et al., 1973; see Appendix A). Measurements taken in 1974, however, made a "worst case" estimate of 1,592 g of plutonium in the silica storage tank. The difference between the two numbers is a result of the different analyses performed on the waste solution before discharge.

As described in the "Waste Disposal History" section, the waste solution discharged to the 216-Z-8 facility consisted of an appreciable quantity of particulate material derived from the backflushing and cleaning

of the filters, and suspended in a large volume of neutralized liquid (supernatant). Because plutonium is very insoluble in a basic or neutral solution, the total amount of plutonium in the waste consisted of the bulk of the plutonium associated with the particulate material and a small portion of plutonium associated with the supernatant. The estimated 48 g of plutonium discharged through the French drain to the ground was presumably derived from analysis of the supernatant. Because the particulate material in the waste would settle in the silica storage tank, the values reported for the French drain probably represent a reasonable estimate of the amount of plutonium discharged through the French drain.

The total amount of plutonium in the waste (particulate and supernatant) was measured, but is not shown in the records of waste discharged to the ground, because the plutonium associated with the particulate material remained in the silica storage tank. The only published reference of the measurements found (Wood, 1958) indicated that from October 1957 to February 1958, 294 g of plutonium were discharged to the 216-Z-8 facility, while the records of waste discharged to the ground indicate a value of approximately 10 g (Appendix A). At the rate of discharge indicated in the 5 mo from October 1957 to February 1958, several kilograms of plutonium could have been discharged to the 216-Z-8 facility in the 81 mo during which waste was discharged. Because use of the facility decreased in the later years, the "worst case" estimate, made in 1974 and supported by measurements made during the current study, of 1.5 kg in the tank is plausible.

## SUMMARY AND CONCLUSIONS

The 216-Z-8 French drain facility consists of a 58,500-L tank, identified as the silica storage tank that overflowed to the 216-Z-8 French drain. Waste liquid was first discharged to the silica storage tank in 1955. The capacity of the tank was reached in 1957 and overflow to the French drain continued until 1962, when the facility was retired.

The waste discharged to the facility was a neutralized filter cake backflush slurry from filtration of a chemical waste stream in the Recuplex operation located in the 234-5Z Building in the Z Plant complex. For this waste type, the bulk of the plutonium was associated with particulate material in the slurry that settled in the silica storage tank. Records suggest that on the order of 1 kg of plutonium could have been discharged to the facility. The bulk of the plutonium remaining in the tank was associated with the layer of sludge. Records of liquid waste discharged to the vadose zone sediments reported that 9,600 L of waste (not including rinse water) containing an estimated 48 grams of plutonium overflowed to the French drain during active operation of the facility.

This study determined the distribution of plutonium and americium beneath the French drain and evaluated the suspected tank leak. For the French drain, results indicated that the plutonium and americium were sorbed onto the sediments immediately beneath the French drain. Estimates indicate that 4 to 5 m<sup>3</sup> of sediment contaminated with greater than 10 nCi/g of TRU activity are located directly beneath the French drain. Transuranic contamination attributable to the waste discharged to the French drain involves a roughly spherical volume extending about 5 m below and laterally from the French drain excavation.

In 1974, as part of a program to reduce the volume of supernatant in tanks at the Hanford Site, the silica storage tank was pumped to remove the solution contents. Measurements taken prior to pumping indicated that a liquid level corresponding to a volume of 31,000 L of solution remained in the tank, which was 27,500 L less than measurements taken in 1958 and 1959. A tank leak was postulated. Samples of solution and particulate material in the tank indicated that the bulk of the plutonium was associated with the particulate material. A conservative estimate of the inventory indicated that approximately 1,400 g of plutonium were associated with the particulate material and 175 g with the solution remaining in the tank. Approximately 165 g of plutonium would have been in the 27,500 L of solution that presumably leaked from the tank. After transfer of the solution in 1974, a camera was lowered into the tank to photograph the interior. The photographs indicate approximately 4,000 L of material, composed mainly of particulate material, remained in the tank.

Based on the observed TRU distribution beneath the French drain, if the silica storage tank leaked, most of the plutonium and americium would be sorbed onto the sediments next to the tank, immediately adjacent to the breach in the tank's wall. Transuranic activity would decrease rapidly with

distance from the tank. Depending upon the actual distance from a postulated breach in the tank to the wells, anticipated levels of activity would have been on the order of 1 pCi/g of sediment or greater for samples collected from a well penetrating the contaminated volume. Levels of TRU activity on the order of 1 to 10 pCi/g were encountered in most of the sediment samples obtained from the four wells drilled around the tank. However, plutonium activity was expected to decrease with distance from a breach in the tank wall and as a result, not be similar in all wells. Alternative sources of the plutonium activity were considered, but do not appear to provide an adequate explanation. With the data obtained, the source of the plutonium activity cannot be positively identified, and a leak cannot be confirmed. Because the 216-Z-8 facility discharged liquid waste solution to the ground through the French drain, the additional effect of a leak of the same solution from the tank is of little practical consequence. The bulk of the plutonium discharged to the facility was confirmed to remain in the sludge that remained in the tank; a fact unaffected by whether or not the supernatant leaked. At this time, no additional work is proposed. However, the potentially large quantity of plutonium remaining in the tank needs to be further evaluated and some remedial action will probably be required to stabilize or remove the material. If an action requiring excavation to or of the tank is indicated, additional drilling prior to excavation should be included to define the suspected contamination plume.

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M. C. Marratt planned this study and conducted the field program. The coauthors, A. E. Van Luik and R. B. Kasper, interpreted the data and wrote the document.

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## APPENDIX

## Monthly Discharge Records to the 216-Z-8 French Drain.

Date	Volume (L)	Pu (g)	Date	Volume (L)	Pu (g)
10/57	300	1.3	1/60	400	1.74
11/57	300	1.3	2/60	300	1.31
12/57	<u>360</u>	<u>1.5</u>	3/60	200	0.872
			4/60	300	1.308
6-month total	960	4.1	5/60	300	1.308
Previous total			6/60	<u>200</u>	<u>0.872</u>
Total to date	960	4.1			
			6-month total	1,700	7.410
7/58 <sup>a</sup>	350	1.52 <sup>b</sup>	Previous total	6,590	32.60
8/58	300	0.13	Total to date	8,290	40.01
9/58	150	0.65			
10/58	250	1.08	7/60	200	0.872
11/58	300	1.30	8/60	100	3.0
12/58	<u>0</u>	<u>0</u>	9/60	0	0.0
			10/60	0	0.0
6-month total	1,350	4.68	11/60	200	0.872
Previous total	3,440	19.9	12/60	<u>0</u>	<u>0.0</u>
Total to date	4,790	24.6			
			6-month total	500	4.744
1/59	0	0.0	Previous total	8,290	40.01
2/59	100	0.440	Total to date	8,790	44.75
3/59	100	0.436			
4/59	200	0.87	1/61	0	0.0
5/59	200	0.87	2/61	500	2.18
6/59	<u>300</u>	<u>1.31</u>	3/61	0	0.0
			4/61	100	0.436
6-month total	900	3.93	5/61	0	0.0
Previous total	4,790	24.6	6/61	<u>0</u>	<u>0.0</u>
Total to date	5,690	28.5			
			6-month total	600	2,616
7/59	100	0.431	Previous total	8,790	44.75
8/59	100	0.650	Total to date	9,390	47.37
9/59	100	0.436			
10/59	100	0.436	7/61		
11/59	200	0.870	8/61		
12/59	<u>300</u>	<u>1.310</u>	9/61	200	0.872
			10/61		
6-month total	900	4.140	11/61		
Previous total	5,960	28.50	12/61		
Total to date	6,590	32.60			
			6-month total	200	0.872
			Previous total	9,390	47.366
			total to date <sup>c</sup>	9,590	48.238

<sup>a</sup>No records for January to June 1958.

<sup>b</sup>This figure calculated from a sample of the settling tank rather than on the amount leaving the building.

<sup>c</sup>No additional discharges from January 1962 to April 1962.

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R. T. Wilde

R. E. Wheeler

D. E. Wood

Basalt Waste Isolation Project Library (2)

Environmental Analysis and Monitoring

Library (5)

Document Control (2)

Publications Services, Station 3,

2750-E, C-135 (2)