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SOIL CONTAMINATION ADJACENT TO TANK 8-F

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Introduction

In 1973, a new series of ground water monitoring wells was installed in the waste farm areas (both F and H Areas) as part of the environmental monitoring program. Two wells in F Area were found to contain low levels of ruthenium activity. A program was undertaken to determine the source of this activity. During this investigation, approximately 1000 cubic feet of soil contaminated with about 5000 Ci of ^{137}Cs was located at the intersection of the fill line encasement and Waste Tank 8.

This cesium resulted from waste which leaked into the soil in 1961 apparently as a result of an overfill of Waste Tank 8. In April 1961, Waste Tank 8 was found to be filled about 5 to 6 inches above the bottom of the fill line. The waste had covered the end of the fill line and about 15,000 gallons of waste leaked through an asbestos-packed sleeve around the fill line into a concrete encasement that provides secondary containment for the fill line entering Tank 8. Most of this leaked waste was collected in an installed catch tank and returned to another waste storage tank. The level of waste in Tank 8 was decreased to normal by transfer of some of its contents to another waste tank. At the time of the leak no waste was thought to have reached the soil surrounding the tank or fill line encasement.

Summary

Investigation of soil contamination found near Tank 8 in F-Area waste tank farm in late 1974 indicates the existence of about 5000 Ci of ^{137}Cs adjacent to the juncture of the fill line encasement and the tank secondary encasement. The location of the contamination and its approximate age as determined by cesium isotope ratio indicate that the release apparently occurred in April 1961 when Tank 8 was overfilled. An analysis of the analytical data accumulated and information on the contents of Tank 8 in 1961 show that about 1500 gallons of high level waste leaked into the soil.

The contaminated soil which resulted from this leak is located about 12 feet below the surface of the soil, and is spread over a horizontal area of 200 to 300 square feet extending 10 to 13 feet from the outer wall of Waste Tank 8. The thickness of the contaminated zone varies from 1 to 14 feet and the lower surface is 14 to 18 feet above the maximum elevation of the water table. This zone of contamination contains an estimated 5000 curies of ^{137}Cs and probably smaller but uncertain amounts of ^{90}Sr and ^{147}Pm, depending on the amount of solids that had settled into the sludge phase in the waste tank and did not leak into the soil. Most of the ^{137}Cs and probably the ^{90}Sr and ^{147}Pm which leaked into the soil are contained in 800 to 1100 cubic feet of soil near the point of leakage. Other radionuclides

contained in this zone are less than a few curies. About 5000 cubic feet of soil containing a small fraction of the released radionuclides surround the central core of 800 to 1100 cubic feet.

Analysis of available information on the leak, current location of the zone of contamination, our knowledge of ground water flow velocities and paths, and the measured ion exchange capabilities of the soil indicate that the measured ^{137}Cs and maximum estimated amounts of ^{90}Sr and $^{238,239}Pu$ which leaked into the soil do not pose any threat to mankind as long as they remain buried at this current location. It should be many years before these radionuclides reach the water table, then several hundred thousand years to reach the nearest stream and ultimately a populated area. In these time periods the ^{137}Cs and ^{90}Sr will decay, leaving harmless solutions of stable barium and zirconium. Plutonium will be diluted by dispersion in ground water to concentrations below the 10 nCi/g limit for unrestricted burial after traveling a few hundred feet from its current location.

Excavation and examination of the Tank 8 spare inlet line encasement, identical in construction to the suspected leak site, showed evidence of very slight leakage of waste into and from this encasement, and several shovelfuls of slightly contaminated soil were removed from beneath the end of the encasement. However, no construction flaws or serious deterioration were observed. Soil core samples along the fill and drain line encasements showed no contamination other than small amounts of ^{106}Ru; further soil exploration is planned in these areas.

The immediate program is to define the location and amounts of other long-lived radionuclides within the contaminated zone near the Tank 8 inlet. Ultimate disposition of this contaminated soil is under study.

Investigation

The arrangement of storage tanks 1 through 8 in the F-Area waste tank farm is shown in figure 1. The concrete encasement between the two rows of tanks encloses the 3"-dia fill lines that distribute the gravity-flow waste from the 221-F Building to the tanks. By installation of jumpers in diversion box 1 (DB-1) the waste can be distributed to any tank. The main encasement slopes toward the south and the fill line encasement to each tank slopes back toward the main encasement. A drain line, inside its own concrete encasement, drains any leakage from the low point of the main encasement to a catch tank for collection and return to a waste tank.

Early in 1974, water samples from two of eight new monitoring wells installed around this group of tanks showed elevated concentrations of ^{106}Ru. A maximum of 1.4 pCi/ml was found in well FTF-7, located between Tanks 3 and 5; a maximum of 66 pCi/ml was found in well FTF-6, between Tanks 5 and 7 near the drain line encasement. These concentrations can be compared to a drinking water limit of 10 pCi/ml for ^{106}Ru.

The immediate supposition was that this contamination originated from the drain line encasement adjacent to well FTF-6, and soil core samples were taken along its entire length. These samples showed no contamination except in the area between Tank 5 and Tank 7, where samples of water from the water table (48 foot depth) indicated a maximum of 34 pCi ^{106}Ru/ml; soil from the 12 to 14 foot depth analyzed a maximum of 0.01 µCi ^{106}Ru/g. A subsequent core sample from the 12 to 14 foot depth adjacent to Tank 8 gave the first indication of ^{137}Cs and ^{90}Sr — 12 µCi ^{137}Cs and 2 µCi ^{90}Sr per gram of soil. Due to the slow movement of these radionuclides in the soil [reference DP-1358], their presence indicated that the original leak site was nearby.

The location of this initial sample of contaminated soil is shown in figure 2, near the juncture of the Tank-8 encasement and the tank fill line encasement. This location and analyses showing a $^{137,134}Cs$ ratio characteristic of waste about 15 years old indicated that the contamination almost certainly originated from the overfill of Tank 8 in April 1961.

The fill line entry into Tank 8 is detailed in figure 3. The tank was filled about 5 to 6 inches above the level of the inlet line in 1961. A recent thorough review of this incident and all available data did not provide sufficient information to quantify movement of waste during the overfill. However, it appears that the tank liquid level was abnormally high for three or four weeks due to a malfunctioning liquid level indicator. During this time about 15,000 gallons of waste flowed through the asbestos-packed joints between the 3" fill line and its 4" sleeve, through the fill line encasement and drain line, into the catch tank. Most of this waste was transferred from the catch tank into Tank 7 after the overfill was discovered. Data from the current investigation indicate that approximately 1500 gallons of waste leaked from the encasement into the ground, probably via the 1/2" molded bituminous filler joint between the tank and fill line encasements. The packed sleeve opening into Tank 8 is peculiar to the design of only the Type I waste tanks (Tanks 1-12). The design of subsequent tanks provided a welded closure rather than packing at this location. A detail of the inlet for the latest (Type III) storage tank is shown in figure 4.

Core samples were taken near other joints in the encasement in an effort to detect other possible zones of leakage from the encasement. Locations sampled are shown on figure 1. Three cores were taken near the intersection of the fill line encasement and the main encasement near Tanks 8, 6, and 4. One was taken at the end of the spare inlet encasement and Tank 8, and another at the intersection of the fill line encasement and Tank 7. No soil contaminated with more ^{137}Cs than background levels was discovered at any of these locations. Only one core sample at the intersection of the fill line encasement and Tank 7, was contaminated with ^{106}Ru activity. Maximum concentration of this soil sample was 0.004 µCi ^{106}Ru/g. These core samples indicate that if any leakage to the soil occurred at these locations it was quite small.

The ground area around the spare inlet encasement (shown 90° from the fill line encasement in figure 1) was excavated. The condition of this similar line-encasement-to-tank-encasement joint was of particular interest since it is identical to the one that leaked. The plan view in figure 5 shows the spare inlet line in its encasement and the section view shows the encasement base pad supported on pilings driven into undisturbed soil. An external inspection showed the waterproofing mastic dry and in good condition. The suspected joint at the tank encasement was also in good condition and there was no evidence of setting, cracking, or leakage. There was a void a few inches deep beneath the entire length of the encasemant base pad, probably from settling of uncompacted backfill. Visual and x-ray inspection of the cap weld on the 3" spare fill line showed it satisfactory with no evidence of leakage. About 15 cubic yards of slightly contaminated soil totaling less than 0.2 Ci of ^{137}Cs were removed from around the encasement. The majority of this contamination was contained in a few shovelfuls of dirt from beneath the end of the encasement (low point) that analyzed 0.006 μCi ^{137}Cs/g soil. This contamination probably resulted from liquid seepage past the grouted end plate of the encasement. A 3"-dia hole was bored through the top of the spare inlet encasement at the location shown on figure 5 to inspect the encasement interior. Figure 6 shows a borescope photograph taken from this location looking toward the tank and shows the concrete encasement of the tank, the 3" spare inlet line and the 4" sleeve. The white asbestos packing is reflected in about 1" of liquid in the line encasement. This liquid, totaling about seven to eight gallons, analyzed less than 1 μCi/ml of ^{137}Cs, a total of less than 0.03 Ci. Such a low concentration would have resulted from leakage of less than 50 ml of high level waste through the packed joint with subsequent dilution by condensation of moisture from the tank in the relatively cool encasement. Drops of condensation were observed on the ceiling and walls of the encasement. This liquid was left in place, and the spare inlet encasement was returned to its original condition.

Defining the extent of soil contamination around the active fill line encasement proceeded in two phases. First, the horizontal extent of contamination and the upper boundary of the contaminated zone were mapped by a series of 30 soil core samples. The cores were terminated at the first indication of radiation on field instruments to avoid radiation exposure to personnel and to avoid a possible avenue for further movement of contamination downward to the water table. This work clearly defined a zone of contamination extending about 12 feet along the tank encasement on each side of the fill line encasement and about 10 to 13 feet out from the side of the tank encasement, with its upper envelope about 12 feet below grade. This area of soil contamination is shown on figure 7, along with the location of the soil cores. The horizontal boundaries were quite distinct with radiation readings inside the contaminated zone ranging from 0.3 to 5 R/hr at the 14 to 18 foot depth and no detectable radiation at a depth of 26 feet immediately outside the zone. Only five of the thirty cores were taken on the southwest side of the inlet line encasement (between Tanks 8 and 7) due to surface interference from a massive berm shielding above ground transfer lines. This berm is scheduled for removal late in 1975 as part of a project to improve secondary containment and leak detection facilities. More core samples on the southwest side will be taken at that time.

The vertical profile of this contaminated zone was obtained by driving nine 2" diameter pipes with sealed ends to depths of 26 feet and taking radiation readings from inside the pipes. Radiation intensities as high as 300 R/hr were recorded toward the center of the previously defined horizontal zone at a depth of about 16 ft. Figure 8 shows the vertical profile of contamination established by four radiation scans along section A-A of figure 7 in relation to the tank, fill line encasement, and the water table.

Analysis of data obtained from the horizontal map and vertical profile of the contamination indicate most of the ^{137}Cs is contained in 800 to 1100 cubic feet of soil near the point of leakage. An additional 5000 cubic feet of surrounding soil contains a small amount of contamination.

Estimate of Contamination

Estimates of the amount of activity contained in the contaminated zone were based on gamma radiation from the soil through the 2" monitoring pipes. This gamma is attributed to the internal conversion of the metastable ^{137}Ba, the daughter of ^{137}Cs. In aged waste this is the predominant gamma activity. Preliminary estimates indicate that about 5000 curies of ^{137}Cs is contained in this zone of contamination. The value is probably good to ±25%.

The radionuclide content of the waste was calculated at the time of the overfill and also that remaining today. These calculations were based on the reactor fuels processed and the measured U and Pu losses while generating the waste stored in Tank 8. Based on the volume of waste in the tank at the time of overfill, the curies/unit volume of waste was calculated assuming the waste to be homogeneous. These results are shown in table 1. Using the measured 5000 curies of ^{137}Cs in the soil and the calculated 3.25 curies/gallon from table 1, it is calculated that 1540 gallons of waste leaked from the fill line encasement into the soil. The assumption of homogeneity (that is, no separation into the sludge) is valid for ^{137}Cs, which does not concentrate in the sludge.

Since ^{137}Cs and ^{137m}Ba were the only radionuclides measured sufficiently to establish the amount in the soil, other radionuclides must be estimated from the ^{137}Cs content. Direct measurements of these other radionuclides through the steel monitoring pipes were not possible due to the softer radiations emitted. Several samples of soil from the upper surface of the contaminated zone were analyzed and found to contain ^{90}Sr and ^{106}Ru in addition to the ^{137}Cs. The $^{90}Sr/^{137}Cs$ ratio varied from 0.17 to 0.00001 at 2 and 10 feet from leak site, respectively. From table 1 the expected ratio would be 0.96 if the ^{90}Sr were released uniformly with the ^{137}Cs and occupied the same position in the soil.

Use of the ratio of each radionuclide listed in table 1 to the listed ^{137}Cs provides a means of estimating the amounts of other radionuclides which might have leaked into the soil. The assumption of homogeneity used in table 1 will result in a maximum release because the radionuclides other than ^{137}Cs do partition and settle with the sludge, depleting the supernate. The amount of radionuclides settling into sludge prior to the leak is not known. The waste was, however, relatively fresh at the time of the leak. The minimum release can be calculated by assuming that all of the sludge had formed and settled and that the supernate (which leaked from the tank) contained no sludge. The former assumption is probably too high and latter assumption too low; however, the two estimates do provide a reasonable range for the radionuclide content of the soil. These are shown in table 2.

Risks Resulting from Radionuclides Released

Most of the ^{137}Cs which leaked with the waste in 1961 and probably most of the other radionuclides are contained in 800 to 1100 cubic feet of soil near the point of leakage. It is our judgement that most of the radionuclides which escaped with this waste are contained in a thin zone of soil pluggage which surrounds the contamination zone. This soil pluggage mechanism has been demonstrated in the laboratory and under field conditions with simulated waste. The pluggage results from dispersion of clay particles of the soil due to monovalent cations of the waste, followed by the filtering action of adjacent soil which removes the dispersed clay, plugging the soil. Laboratory measured permeability indicates a flow reduction of 3700 resulted from this pluggage. (More comprehensive discussion of this soil pluggage mechanism and test data can be found in DP-1358, p C-6.) Several pieces of data collected on the Tank 8 investigation point to pluggage:

- the sharp break from highly contaminated soil to essentially clean soil,
- the additional force required to drive the monitoring pipes through the contaminated zone, and
- the volume of soil containing the radionuclides being essentially that which would be required to contain the 1500 gallons of waste in the free spaces of the soil.

For these radionuclides to migrate into the environment, they must be transported by water. Waste Tank 8 is near the topographic divide between Upper Three Runs and Four Mile Creek, but all meteoric water (rain, snow, etc.) that enters the ground will drain into Four Mile Creek. To spread the contamination, water must soak into the ground, seep through the surface soil, and penetrate the zone of contamination before it can transport the radionuclides from their current location in the unsaturated zone to the saturated zone (water table). Then the ground water must transport the radionuclides until they surface in Four Mile Creek. Ion exchange and cesium fixation will delay the migration by reducing the movement of radionuclides relative to ground water flow. Also, soil pluggage reduces the rate of ground water flow as previously described. Measurements of ion exchange distribution coefficients for the long-lived radionuclides in waste, using SRP ground water and soil, have shown reductions in the movement of ^{90}Sr, ^{137}Cs, and Pu (IV) of 3.6×10^{-4}, 2.9×10^{-4}, and 3.6×10^{-5}, respectively, relative to the rate of ground water flow. Cesium fixation, as the name implies, is a mechanism which selectively removes cesium from the waste and incorporates it into the mica mineral lattice, rendering it essentially unavailable for further movement by ground water. These mechanisms are more completely described in DP-1358.

The area surrounding Waste Tanks 1 through 8 is paved, which should effectively reduce the amount of water that soaks into the soil and moves downward to the saturated zone. The rate of surface water movement in an unpaved area in the burial ground has been measured and found to be about 7 feet/year. If the paving in the vicinity of Tank 8 reduces the downward movement of water by a factor of 10 and the soil is not plugged (contrary to our current understanding), it would require on the order of 25 years for water to percolate the 14 to 18 feet to the water table. The mechanism of ion exchange and Cs fixation would tend to further slow migration of the activity to the water table. As indicated earlier, we feel soil pluggage is probably responsible for the current slow movement of the radionuclides toward the saturated zone. This consideration should extend the estimated time manyfold. (Flow reduction of 3700 times have been measured in laboratory experiments.)

Figure 9 shows the flow path for water in the water table from F and H Areas to the stream. As indicated earlier, Tank 8 is near the water table divide but water from below Tank 8 will flow to Four Mile Creek. From this flow path and knowledge of ground water flow rates, it has been estimated that about 200 years is required for ground water from the vicinity of the waste tanks to reach Four Mile Creek. Time to move to Upper Three Runs Creek would be similar if the water moved in that direction. Soil pluggage, ion exchange, and cesium fixation would slow the movement of the radionuclides beyond this time. Considering soil ion exchange, cesium and strontium would require several hundred thousand years to reach any SRP stream and in that time they will have decayed.

As shown in table 2, between 0.1 and 1.8 curies $^{238,239}Pu$ are estimated to have leaked into the soil. If the release was the maximum amount (1.8 curies) and all of the plutonium was the longer-lived ^{239}Pu, it would take many hundreds of thousands of years for the plutonium to decay. In the meantime, dispersion of this plutonium would occur during its extremely slow migration (Pu moves at 1/8 the rate of Cs) toward the creek, diluting the plutonium. By the time the plutonium has migrated more than a few hundred feet from its current location, dispersion will have diluted its concentration to below the 10 nCi/g limit for unrestricted burial.

Program

The program for the next year will be to:

- Periodically repeat radiation readings in the existing nine 2" pipes to monitor any movement of radioactivity in the soil. This monitoring will provide continuing assurance that the radioactivity remains fixed in the soil above the water table.
- Insert additional monitoring pipes on the southwest side of the inlet encasement between Tanks 7 and 8 (figure 1) later this year to better define the zone of contamination in that area.
- Obtain additional core samples from within or below the zone of contamination and analyze for other radionuclides, particulary ^{90}Sr.
- Insert radiation monitoring pipes in vicinity of drain and fill line encasements and pump ground water wells as part of general monitoring program described in letter, K. W. French to N. Stetson, "Ground Contamination in 200 Area Tank Farms," April 24, 1975.
- Regrade and waterproof the ground surface as need above the contaminated zone to minimize percolation of surface water through the soil, which could cause some downward migration of the contamination. Surface water will be drained away from the area. Measure percolation rate of surface water in the waterproofed area.
- Obtain technical data on cesium and strontium behavior in soil from the Tank-8 area, including leachability, radionuclide distribution coefficients (Kd), etc.
- Complete design of a high-liquid-level alarm system as an independent backup for the improved automatic level-sensing tapes recently installed in all waste tanks.
- Study the disposition of the contaminated soil. Unless our current knowledge regarding the amount, location, and movement of this contamination is greatly altered, excavation will not be recommended.

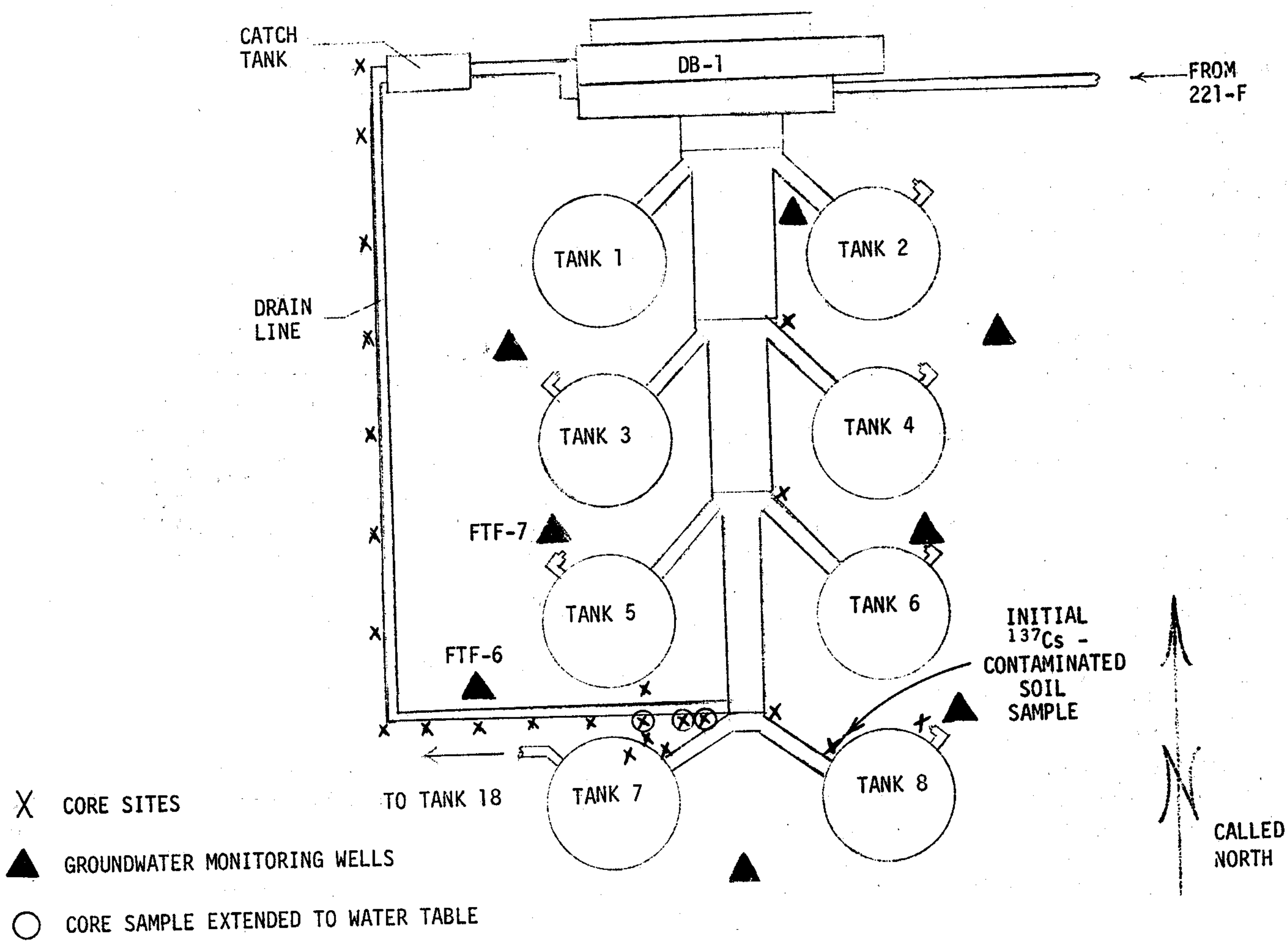

FIGURE 1. 241-F UNDERGROUND ENCASEMENTS

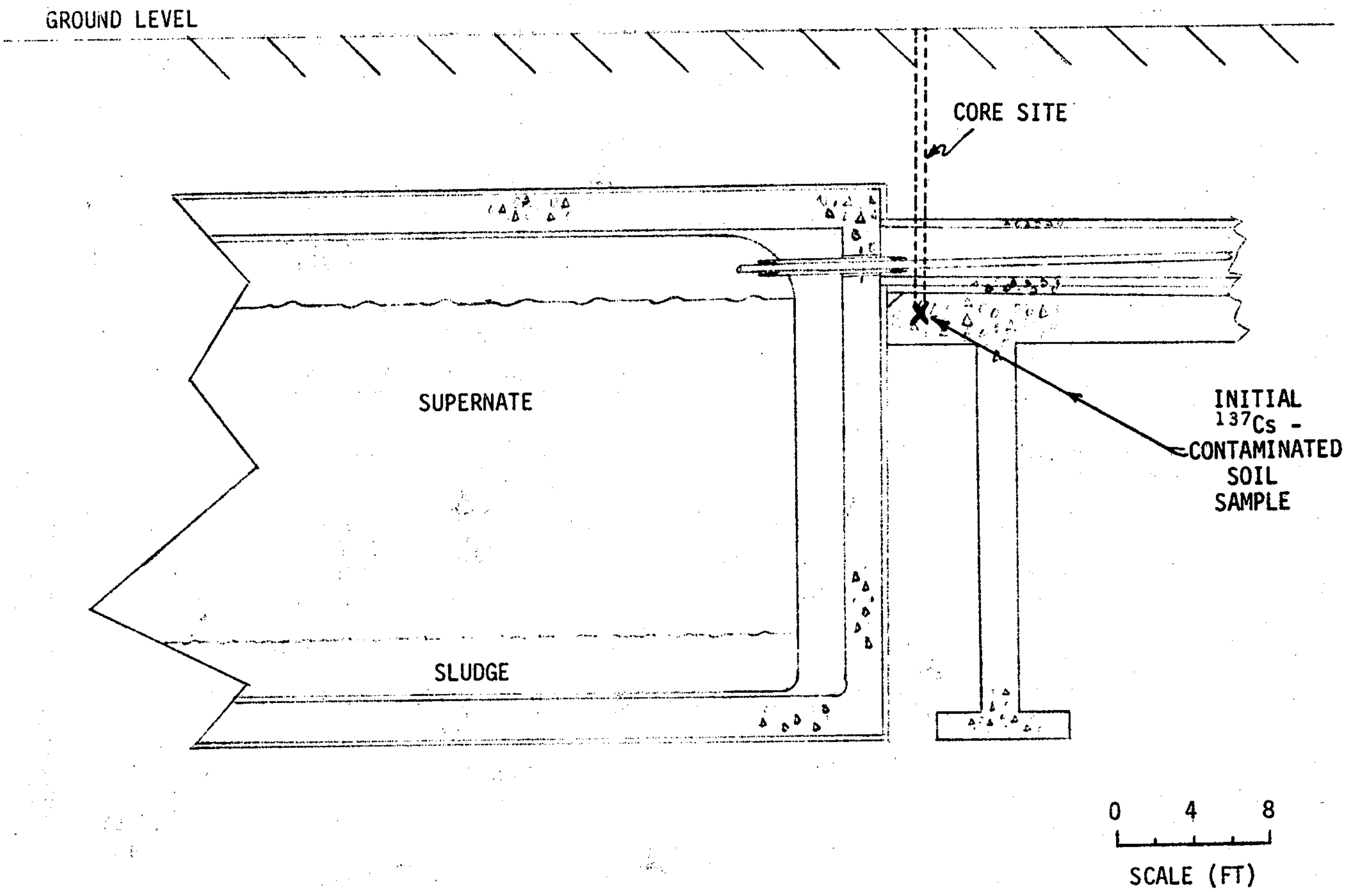

FIGURE 2. WASTE TANK 8, SECTION VIEW

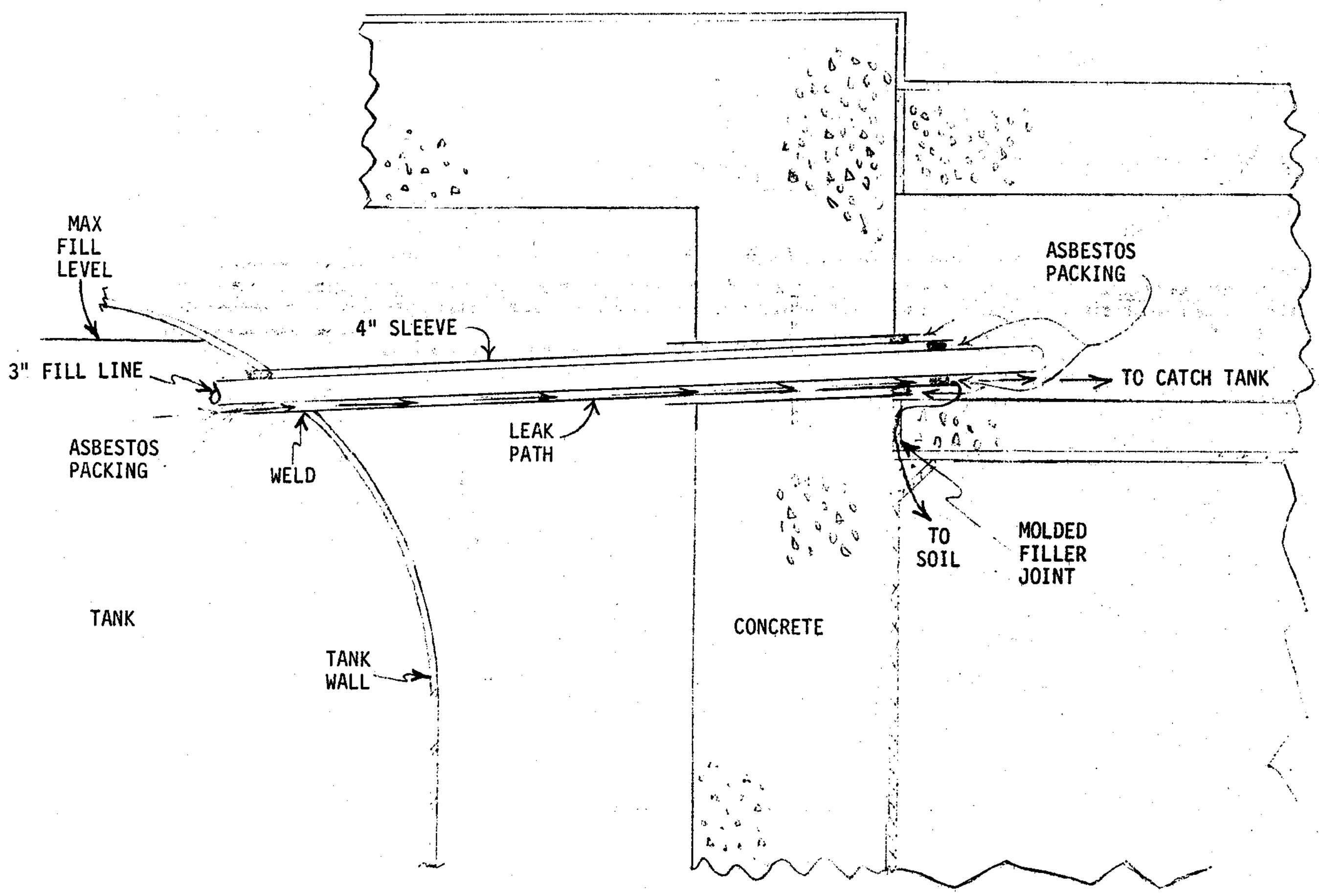

FIGURE 3. TYPE I WASTE TANK INLET

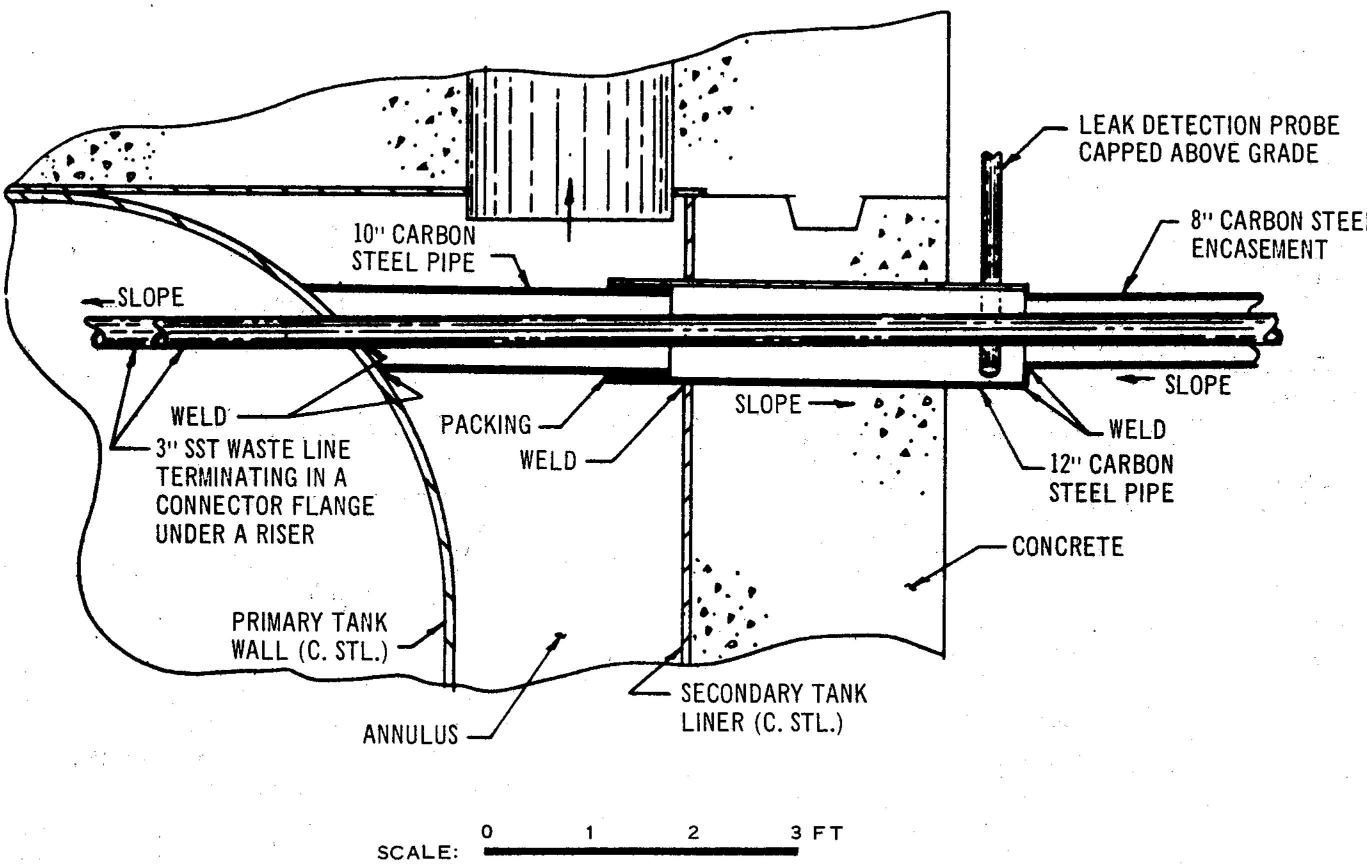

FIGURE 4. TYPE III STORAGE TANK INLET

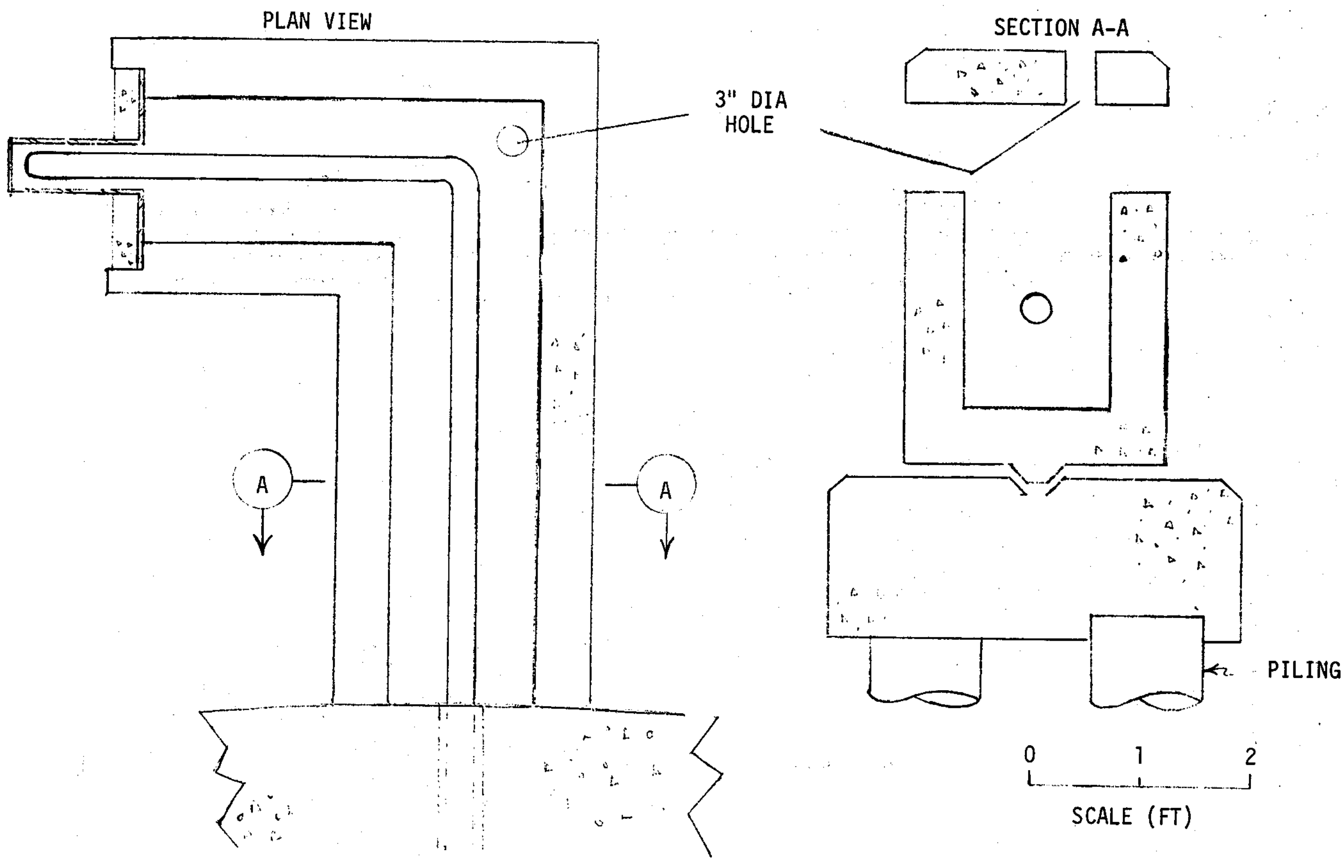

FIGURE 5. TYPE I TANK SPARE INLET LINE

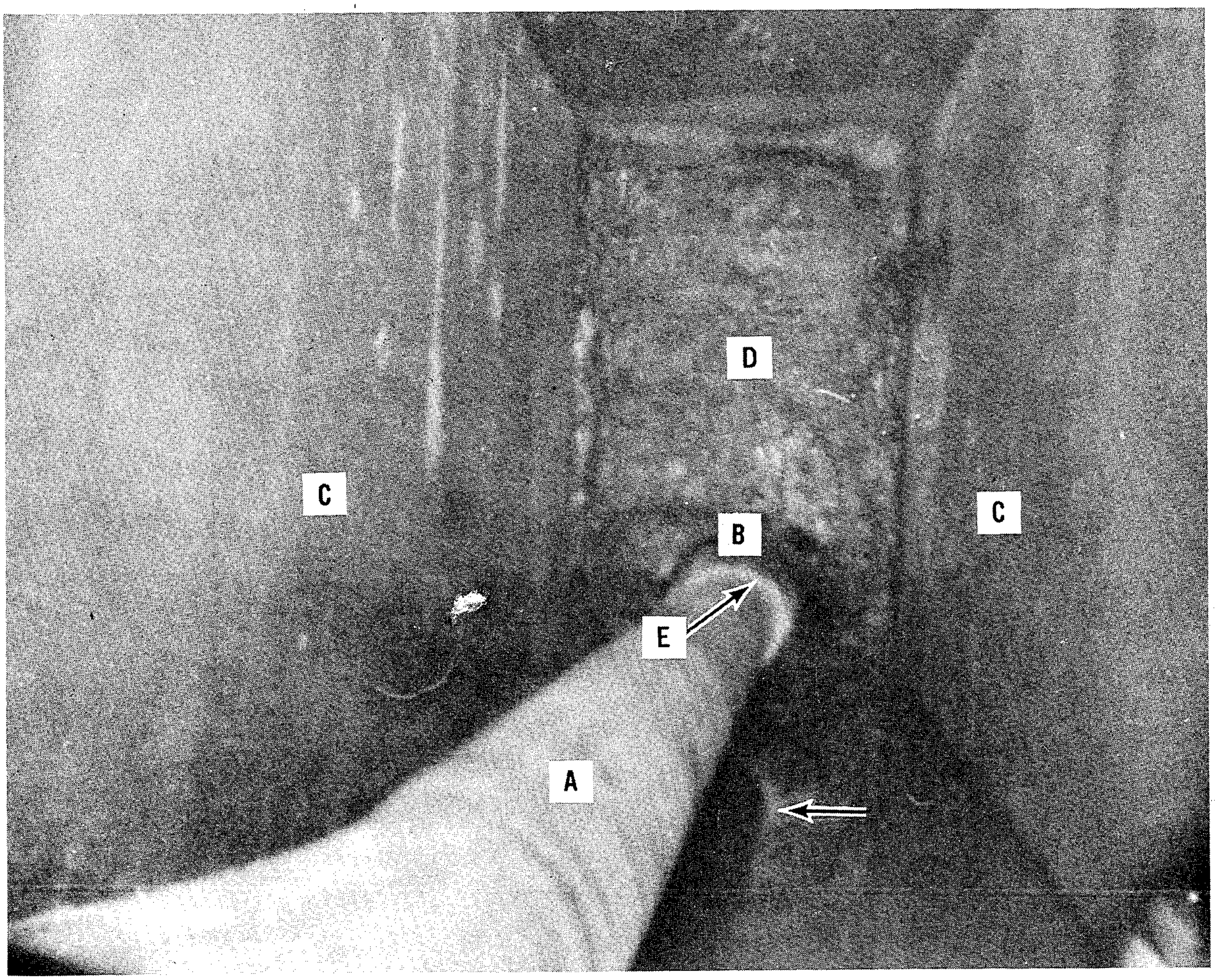

FIGURE 6. INSIDE CONCRETE ENCASEMENT LOOKING TOWARD TANK NO. 8. Note pipe reflection in liquid on bottom (arrow). Photograph reversed 180° because of mirror image. Approx ¼X, DPSPF 18587-7.

A 3-inch process line
B 4-inch sleeve
C Concrete encasement
D Concrete shell on tank
E Asbestos packing

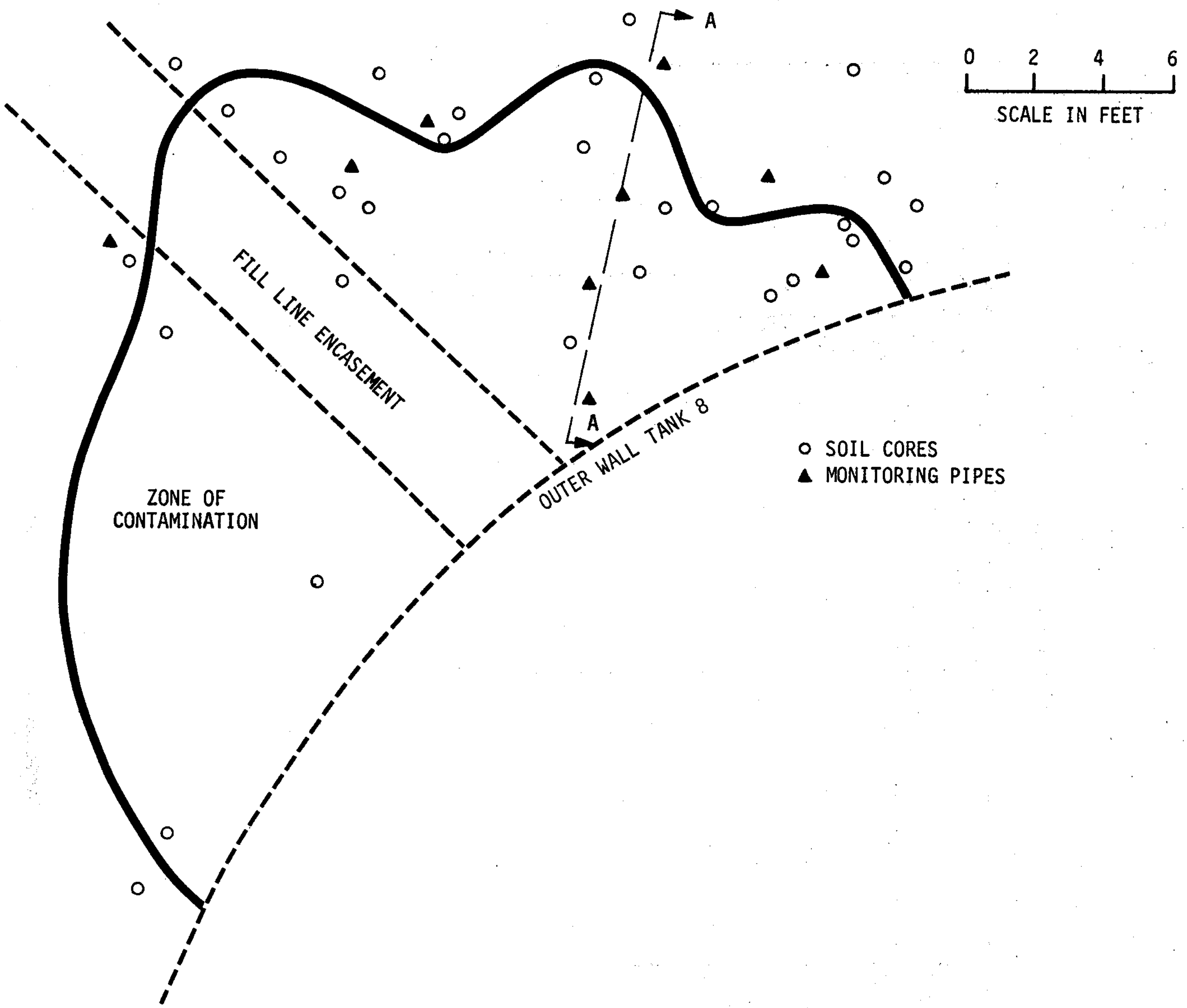

FIGURE 7. ZONE OF CONTAMINATION AROUND TANK 8 FILL LINE

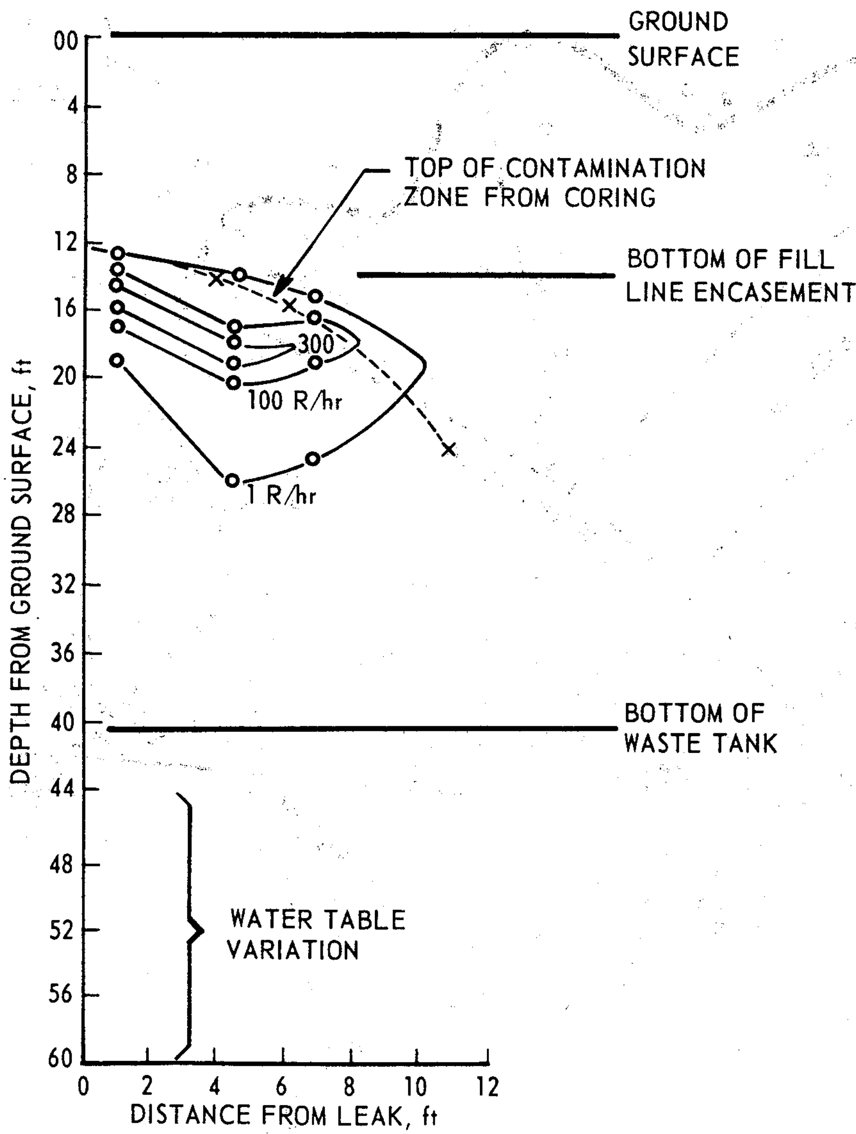

FIGURE 8. VERTICAL DISTRIBUTION OF WASTE
(section A-A, figure 7)

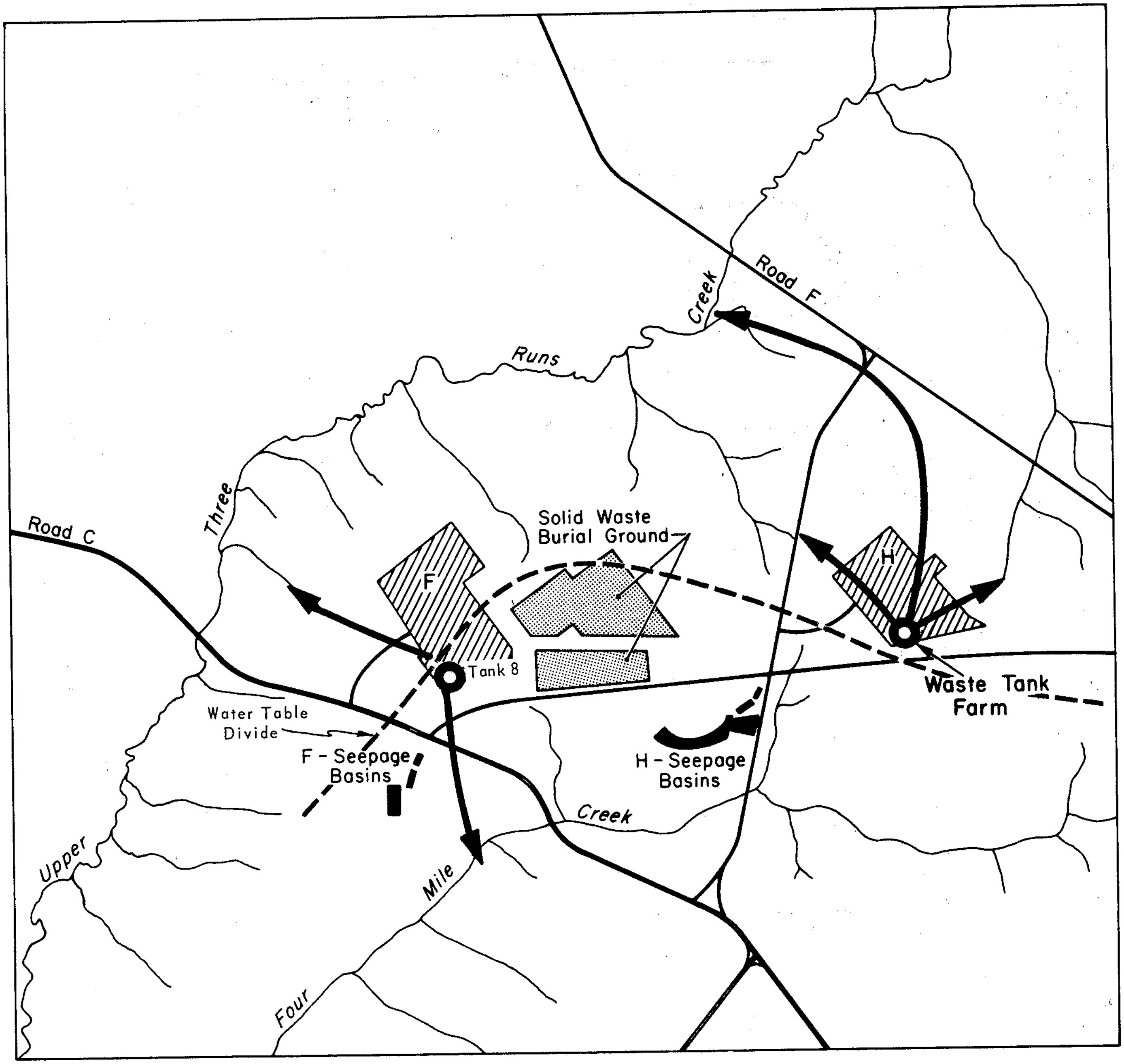

FIGURE 9. FLOW PATH IN WATER TABLE

TABLE 1
TANK 8 CALCULATED WASTE COMPOSITION, Ci/gal

	At Time of Overflow April 1961	Current April 1975
^{137}Cs	4.48	3.25
^{137m}Ba	(4.14 γ)	(3.01 γ)
^{90}Sr	4.36	3.12
^{90}Y	4.36	3.12
^{106}Ru	3.50	2.36×10^{-4}
^{106}Rh	3.50	2.36×10^{-4}
^{144}Ce	53.35	2.08×10^{-4}
^{144}Pr	53.35	2.08×10^{-4}
^{147}Pm	14.07	0.347
$^{238,239}Pu$	1.16×10^{-3}	1.16×10^{-3}
^{89}Sr	1.75	a
^{91}Y	4.30	a
^{95}Zr	7.53	a
^{95}Nb	14.43	a

a Less than 1×10^{-6}.

TABLE 2
ACTIVITY IN THE SOIL, Ci

	At Time of Overflow April 1961		Current April 1975	
	Minimum[a]	Maximum[b]	Minimum[a]	Maximum[b]
^{137}Cs	6,890	6,890	5,000	5,000
^{137m}Ba	-	-	-	-
^{90}Sr	135	6,710	100	4,800
^{90}Y	135	6,710	100	4,800
^{106}Ru	1,080	5,390	0.08	0.36
^{106}Rh	1,080	5,390	0.08	0.36
^{144}Ce	4,110	82,160	0.02	0.32
^{144}Pr	4,110	82,160	0.02	0.32
^{147}Pm	1,080	21,670	25	530
$^{238,239}Pu$	0.1	1.8	0.1	1.8
^{89}Sr	55	2,700	c	c
^{91}Y	130	6,620	c	c
^{95}Zr	2,320	11,600	c	c
^{95}Nb	4,440	22,220	c	c
Total →	25,560	260,220	5,230	15,130
Total with $T_{1/2}$ >10 Yr →	8,240	41,980	5,230	15,130

[a] Assumes radionuclide partitioning between sludge and supernate was complete and only supernate leaked into the soil.
[b] Assumes no radionuclide partitioning and radionuclides uniformly distributed in the waste which leaked into the ground.
[c] Less than 0.01 Ci.