

# Centrifugal Modeling of Leaking Underground Storage Tanks

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Leaking underground storage tanks (UST) and contaminated soil must be removed to comply with environmental regulations. A major problem associated with removal of contaminated soils is the lack of a proven method to determine the extent of soil contamination. The movement of chemicals and the subsequent fate and transport of chemicals from the leaking UST are often estimated from field explorations coupled with mathematical models. A major difficulty in these modeling efforts has been a lack of information concerning the constitutive relationships governing multi-phase contaminant movement. Centrifugal modeling coupled with field investigation is an alternative way of simulating and estimating chemical movement and transport through soils. In centrifugal modeling an actual three-dimensional profile is created and the centrifuge, because of scaling laws, accelerates the chemical flow and transport processes such that a field event that lasts decades is simulated within hours or days. The centrifugal modeling technique was used to simulate the movement of gasoline through the vadose zone from leaky UST and to study the distribution of various constituents of gasoline. The results show that for granular soils the movement of gasoline was advective and the free product formed a pool on top of the water table directly below the UST. However, for fine grained soils, instead of the pooling of free products, spreading of components of gasoline at different rates was observed to be due to the movement and subsequent transport of gasoline in anisotropic soils.

Underground storage tanks (UST) and contaminated soil are often encountered during new highway construction and highway expansion projects. It is the responsibility of the transportation department that owns the site to remove the leaking UST and contaminated soils to comply with environmental regulations. The ground below the highway and the right of way should be free of soil contamination.

To address soil contamination caused by UST, the U.S. Congress (1) and most states proposed stringent regulations governing leaking UST and soils contaminated by them. To find the best clean-up option, the United States Environmental Protection Agency (USEPA) produced a comprehensive document to direct the users to select the most appropriate corrective action technology (2). However, this document did not address the extent or levels of contamination for sites nor the effectiveness and termination criteria for the corrective action technologies such as soil flushing, soil washing, vacuum extraction, slurry walls, soil cover, and others.

The following factors control the extent of soil contamination. The environmental factors are the volume and duration of leak and types of chemicals leaked including their transformations through degradation and volatilization. The hydrological factors are the amount of precipitation (rain, snow, and ice), evaporation and surface runoff. The geotechnical factors are soil layer thickness and

inclination, anisotropy, soil type, soil properties (such as hydraulic conductivity), adsorption capacity that affects hydrogeological parameters, suction and capillary actions. Hydrogeological factors are the depth of water table, the rate and direction of groundwater movement and all the parameters of the vadose zone.

Field monitoring and gathering of field data, mathematical modeling, and physical modeling are the logical approaches that may be taken to determine the extent of contamination and most appropriate clean-up technology for sites contaminated from leaking UST. A combination of approaches may be used to find the extent of contamination and the most appropriate decontamination option. This will facilitate the decision making or judgment procedure with added confidence and will reduce the cost of the site investigation and subsequent chemical analyses. A detailed description of each approach is given below.

## Field Tests

The first approach is to gather field data through field monitoring. Though this approach is the best, it may not be feasible to create a complete picture because of the high cost of drilling, difficulties of obtaining undisturbed samples, sample contamination, loss of volatiles during transportation to a laboratory, and amount of time needed for such an investigation. However, such data is important in calibrating mathematical and physical models.

## Mathematical Models

Mathematical modeling of flow and fate and transport of contaminants in a multi-phase medium is another popular method. Numerical analyses of multi-phase pollutant migration have been presented by numerous researchers (3,4). The major difficulty in multi-phase modeling efforts has been a substantial lack of information concerning the constitutive relationships governing multi-phase contaminant movement such as the functional relationships between fluid pressures, saturation and permeability of coexisting phases, and separation of components because of different diffusion and adsorption values of components for fluids such as gasoline. Thus, most researchers have simplified the multi-phase problems to three-phase, two-phase, or even single-phase problems (5,6).

## 1-g Physical Models

Physical modeling (1-g), on the other hand, may give an insight to the problem and may provide data for the validation of a mathematical model. The movement of fluids immiscible with water has

been physically modeled by numerous researchers (7). Most of the large scale physical models consisted of long one-dimensional columns or large soil box experiments (8). The soil box experiments yield interesting and useful data, but take considerable time and are expensive to construct.

### Centrifugal Models

The centrifuge modeling technique can be an improvement over the 1-g physical modeling technique because the size is made smaller and the time can be shortened through scaling. Because of a gravitational field of  $Ng$  ( $g$  is the acceleration due to gravity and  $N$  is the factor by which the centrifuge increases the intensity of gravity) generated by the centrifuge, the sample size is made  $1/N$  of that in the prototype. The stress levels (which produce varying porosities) in the centrifugal models match the stress levels of the simulated sites. In addition to the stress levels the centrifuge can also simulate in situ capillary fringe, water tables, moving groundwater, any capillary trappings and many of the other features encountered in the field (9). The scaling laws for centrifuge modeling have been developed (9,10). During the past decade many researchers have used the centrifugal modeling technique to model subsurface flow (9,11).

### Centrifugal Modeling of Vadose Zone

The movement and subsequent transport of contaminants through unsaturated soils can be modeled in the centrifuge with the same scaling laws as that for saturated soils provided that the following are satisfied: (a) Richard's equation is valid, (b) the capillary fringe and capillary forces are simulated, and (c) suction pressures are simulated.

The unsaturated flow is governed by Richard's equation, which incorporates which Darcy's law and the condition of continuity. Darcy's law has been proved to be valid in the centrifuge for steady state flow (12), and it has been stated that "there is no strong evidence of the violation of unsaturated flow theory in the centrifuge," that is, Richard's equation is valid even for permeability values as low as  $10^{-10}$  m/s and under relatively dry conditions (13).

The capillary rise ( $H_c$ ) in an unsaturated soil is given by:

$$H_c = \frac{2 \times T}{r_c \times d_w \times g}$$

where

- $T$  = surface tension of pore fluid,
- $r_c$  = radius of the capillary (or 0.2 times  $D_{10}$  size of the soil), and
- $d_w$  = density of water.

In a centrifugal model of size  $1/N$  of the field model under a gravitational field of  $Ng$ , according to the above equation, the capillary head will be reduced to  $H_c/N$  and hence it is simulated in the centrifuge models.

The total suction pressure that is equal to the sum of the matrix and osmotic suction (14) can also be simulated in a centrifuge. There is a unique water content and thus a degree of saturation versus the total suction relationship for remolded soils (15). The water content or degree of saturation profile for a given site can be simulated in the centrifuge under equilibrium of steady flow condition (13), so the total suction can also be simulated in a centrifuge. The

centrifuge model simulates the total suction pressure and the model is  $N$  times smaller than the prototype, so the suction gradient is  $N$  times higher than that in the prototype. Therefore, hydraulic as well as suction gradients in the model are  $N$  times higher than that in the prototype.

The validity of the centrifugal modeling technique has been verified by: (a) modeling of one-dimensional contaminant transport in saturated soils using the modeling of models technique (9), and (b) comparing the one-dimensional centrifugal simulation of flow and transport of a LNAPL through unsaturated soils with prototype behavior and numerical simulation (16). Thus, the methodology can be applied to real world geo-environmental problems with complicated site conditions. One class of such problems is the simulation of an actual three-dimensional profile of a leaking UST contaminated site in a short duration, that is, to simulate events that occur over several decades in the field in a day or two inside the centrifuge. The goal of this research is to use the centrifugal modeling technique to study the three-dimensional movement of contaminants from leaking UST through the vadose zone to the groundwater table.

## EXPERIMENTAL PROCEDURE

### Soils Tested

The soils used consisted of a kaolin clay and a dredged residue from a gravel that represented a clay and a poorly graded fine sand, respectively. The liquid and plastic limits of kaolin were 48 and 36 percent, respectively. It had a clay fraction of 85 percent and a maximum dry density of  $14.1 \text{ kN/m}^3$  at an optimum water content of 30 percent. The coefficients of uniformity and curvature of fine sand were 3.4 and 1.6 respectively. It had zero clay content, a  $D_{10}$  of 0.117 mm and a maximum dry density of  $14.6 \text{ kN/m}^3$  at an optimum water content of 10 percent. To make the aquifer, Ottawa C 30 sand was used.

### Gasoline Mixture

A commercial gasoline contains numerous organic compounds. Interpretation of data and making logical conclusions will be difficult with the use of commercial gasoline. Hence a synthetic gasoline that was reproducible and representative of the commercial gasoline was used in this research. Commercial gasoline is made of aliphatic, aromatic, and olefin. On average a middle grade gasoline contains 55 percent aliphatic, 42 percent aromatic, and 3 percent olefin. The synthetic gasoline was designed with 43 percent aliphatic and 57 percent of aromatic but olefins were omitted. The aliphatic group consisted of 28.5 percent pentane and 28.5 percent hexane. Butane was omitted completely from the mixture because of difficulties in handling and because of chemical detection, and the potential fire hazard. The aromatic group consisted of 30.7 percent toluene, 6.1 percent *o*-xylene and 6.2 percent benzene. Instead of meeting the USEPA regulation that the benzene be below 2 percent, 6.2 percent benzene was used so that it would result in conservative predictions. The proportions and thermodynamic parameters of the various components of the synthetic gasoline are shown in Table 1. The gas chromatographs of the synthetic and commercial gasoline were compared (17).

TABLE 1 Chemical Parameters of Synthetic Gasoline and Its Components

Chemicals	Percentage by weight	Molecular Weight	Saturated Vapor Concentration at 20° (mg/l)	Aqueous Solubility at 20° C (mg/l)	Henry's Law Constant**	Octanol/water Partition Coefficient at 20° (logP <sub>oct</sub> )
Pentane	28.5	72	1689*	N/A	4299.67	N/A
n-Hexane	28.5	86	630**	10**	5663.95	N/A
Benzene	6.2	78	300**	1780**	18.07	2.13*
Toluene	30.7	92	130**	515**	21.38	2.69*
o-Xylene	6.1	106	30**	152**	21.60	2.77*
Commercial Gasoline	N/A	N/A	411**	156**	N/A	N/A
Synthetic Gasoline	100	N/A	N/A	N/A	N/A	N/A

## Equipment

The centrifuge at the New Jersey Institute of Technology is a 5-g ton machine. It is equipped with 16 slip-rings and two hydraulic glands. It can reach a maximum speed of 450 rpm. The radius of the centrifuge is 0.65 m. The arms of the centrifuge are connected to swing-up buckets of internal dimension 32.0 cm × 25.4 cm × 20.3 cm. A special container was designed to maintain the inclined groundwater table that reduced the maximum size of the sample to 23 cm × 22 cm × 19 cm. This special container with several tubes in the inner walls was able to simulate the inclined groundwater table and the flowing groundwater. A video camera and a pre-amplifier were mounted inside the centrifuge. A 16-bit high-speed data acquisition card in a PC was used to acquire the data from the centrifuge. The speed of the centrifuge during each Test was maintained at 400 rpm. Therefore, the gravitational level for each test varied between 93 at the top of the sample and 114 at the bottom of the sample, times the actual gravity, with an average of 104 g ( $N = 104$ ).

## Dimensions of Simulated Prototype

The plan area of each simulated site was 26 m × 25 m. The gasoline tank was a cylindrical drum of 2.0 m height and 6.4 m inside diameter. A hole was drilled at the center of the tank to simulate a leaking UST. The tank was installed at the middle of the site with its bottom at 2.0 m below the ground surface and top flush with the ground surface. The model dimension may be obtained by using a scale of 1/104. The geometrical symmetry of the site was taken advantage of to interpolate the data.

Three tests are reported in this paper. Test 1 consisted of 10.4 m of compacted kaolin on top of a 2.60 m thick aquifer consisting of Ottawa C30 sand. The water table was located at the top of the aquifer. Test 1 simulated 30 years [model time may be obtained by using a scale factor of  $(1/104)^2$ ] of contamination. The total leakage was 24,747 L [model volume may be obtained by using a factor of  $(1/104)^3$ ]. At the end of the simulation period, five borings extending to the top of the aquifer were made using a miniature boring tool. The borings were located at (a) the center of the tank (Point A); (b) 2.08 m (2 cm on model) north of point A (Point B); (c) 4.16 m east of A (Point C); (d) 6.24 m west of A (Point D); and (e) 8.32 m south of A (Point E). Five soil samples were taken from each bor-

ing, except for Point A, and were located at (a) the top of the soil layer; (b) a depth of 2.6 m (2.5 cm on model); (c) a depth of 5.2 m; (d) a depth of 7.8 m; and (e) the bottom of the soil layer. At Point A only four samples were taken at 2.6 m, 5.2 m, 7.8 m, and 10.4 m depths.

Test 2 consisted of 10.4 m of compacted sandy soil on top of a 2.60 m thick aquifer. The water table was located at the top of the aquifer. Test 2 simulated 28.7 years of gasoline contamination with total leakage of 44,995 L occurring over a period of 12.3 years as determined from the results of preliminary tests. There was no leakage for the rest of the 14.4 years. At the end of the simulation period five borings extending to the depth of the aquifer were made at (a) Point A located at the center of the tank; (b) Point B located 3.12 m (3 cm on model) north of Point A; (c) Point F located 6.24 m east of Point A; and (e) Point G located 9.36 m west of Point A; (e) Point H located 12.48 m south of Point A. Five soil samples were taken from each boring at the depths specified in Test 1.

In Test 3 the soil profile was the same as that in Test 2 except the water table was within the compacted soil layer and was located 5.7 m below the top of the soil layer. This test simulated 29.6 years of gasoline contamination. A total leakage of 17,998 L occurred over the first 6 years with no leakage for the rest of the 23.6 years. At the end of the simulation period five borings were made at the locations described in Test 2.

## Soil and Groundwater Sampling and Analysis

Before describing the procedure for soil and groundwater sampling, and chemical analyses, the field procedure is described. The soil samples from sites contaminated by leaking UST were obtained by boring. The groundwater samples were obtained from wells. In this centrifugal simulation a similar procedure was used to obtain the soil and groundwater samples. A miniature boring tool was fabricated to obtain the soil samples. It had an internal diameter of 0.95 cm and an external diameter of 1.27 cm. A sharp end was obtained by machining as in regular boring devices. The soil and groundwater samples from the field were usually analyzed using EPA Method 5030 (18). It was developed to measure the amount of volatile organic contaminants in the soil or water and was expressed as parts per million or parts per billion of soil or water.

When this research was first initiated, a purge-and-trap device was not available. Hence it was decided to modify the test proce-

sure for analysis. A soil sample containing roughly 0.8 g of wet soil extruding from the boring tool was put into a 5 ml vial with a Teflon septum. Methanol has a similar retention time as pentane and hexane, and carbon disulfide, so a solvent that did not produce a peak was used in this research. A predetermined volume of carbon disulfide was injected into each of the vials. Vials were shaken for 24 hours to extract all the gasoline from the solid, liquid and gaseous phases into the carbon disulfide. A 0.4 gml of solution extracted from each vial was directly injected into the GC equipped with FID for analysis.

### Centrifugal Simulation

The four main items of this phase were: (a) sample preparation, (c) spinning of the centrifuge, (c) soil sampling and GC analysis of soil samples, and (d) interpolation and contouring of data.

#### Sample Preparation

The site was prepared by adding water and maintaining the water table at the desired level. To form the aquifer Ottawa sand was wet pluviated to obtain a final height of 2.6 m (2.5 cm in the model). The top of the sand layer was leveled. For Test-1 air dried kaolin was mixed with 35 percent by weight of water and was compacted to a final unit weight of 13.8 kN/m<sup>3</sup> in four equal layers. Care was taken to make the clay homogeneous in the transverse direction. For Test 2 and Test 3 air dried sandy soil was mixed with 5 percent water by weight and was compacted to a final unit weight of 13.8 kN/m<sup>3</sup> in four equal layers. The empty gasoline tank was installed after excavating the compacted soil.

#### Spinning and Soil Sampling

The container with the prepared site model was installed in the centrifuge and the centrifuge was spun for 20 hours (actual time) for the sample to consolidate and further compact under centrifugal gravity (104 g). After 1 day the centrifuge was stopped. To conform to the design water table, water had to be removed in Test 1 and added in Test 2 and Test 3. The centrifuge was spun again for an additional day for the sample to reach hydrogeological equilibrium (i.e., suction, capillary fringe, and degree of saturation). At the end of 20 hours, if the sample met the desired prototype conditions synthetic gasoline was added to the tank, the centrifuge was accelerated to reach 400 rpm, and the clock was started to account for gasoline leakage. At the end of the simulation time corresponding to the desired prototype, the centrifuge was stopped and borings were made and soil samples were taken at the locations and depths described earlier. A total of 25 soil samples was taken with each sample containing roughly 0.8 g of wet soil, and was placed into 5 ml vials with Teflon septums.

#### GC analysis

Three milliliters of carbon disulfide were injected into each of the vials from Test 1, and 1 ml was injected into each of the soil samples obtained from Tests 2 and Test 3. The clay soil required a larger quantity of the solvent because of large surface area. Vials were

shaken for 24 hours to extract all the gasoline from the solid, liquid and gaseous phases into the carbon disulfide. A 0.4 gml of solution extracted from each vial was injected into the GC for analysis.

#### Data Interpolation and Contouring

To generate the contours of gasoline and chemical fractions (concentrations) interpolation of the data points was made at the symmetrical points. For this purpose the z-axis or vertical axis was located at the center of the tank, which is also the source of leakage. Because of the symmetry about the z-axis the mirror image of a data point was assumed to have the same chemical fractions (concentration) as the data point itself. Thus the 24 real data points yielded a total of 44 data points because the data points along the z-axis yielded points that were coincidental with their respective images. These 44 data points served as input data to generate the contours of chemical fractions (concentrations).

## EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows the contours of total gasoline fraction (concentration) in the soil expressed as ppm for Test 2 at the end of a simulation period of 28.7 years. The leaked gasoline moved as a front and accumulated as a pool of free product above the capillary fringe. In a laboratory physical model test (7) it was reported that in a sand with a permeability of 0.4 cm/s gasoline seeped vertically down and accumulated above the capillary fringe when the water table was stationary, that is, when there was no groundwater flow. Furthermore, a substantially higher fraction (concentration) of gasoline was observed directly below the leakage source. The contours depicting the thickness of the gasoline in the mound showed decreasing thickness with increasing radial distance from the center. Also these contours were symmetrical about the source. The contour plot in Figure 1 corroborates these findings. Gasoline was nonexistent at the source of leakage because it was exhausted at the end of the first 12.3 years. Thus, the hydrocarbon fraction (concentration) decreased from the top of the aquifer toward the UST. The borings did not extend into the aquifer; therefore no information is available for the aquifer. The residual gasoline fractions within the silt layer for all the points except for the points shown in Figure 1 were below 0.01 percent or 100 ppm. The above data suggest that the gasoline infiltrated the soil as a front and became accumulated above the capillary fringe.

The contours for Test 3 are plotted in Figure 2. Note that the contours are quite similar to those in Test 2. Although the total amount of gasoline was about 60 percent less than Test 2, it took 6 years for the leakage to occur. Although there was dissolved gasoline in the water below the water table its maximum gasoline fraction was less than 0.015 percent or 150 ppm. The transport of gasoline below the water table appears to be dominated by the physical process of diffusion.

The contours of total gasoline fraction and its components after 30 years of continuous leakage for Test 1 are shown in Figure 3. Note that the gasoline has moved laterally to a considerable distance and has gone above the location of the leakage source (against gravity) which can occur only because of molecular diffusion. The clay soil is compacted wet of optimum ( $S_r = 60$  percent), and the hydraulic conductivity of the clay was very low ( $k_w = 2 \times 10^{-7}$  cm/sec), so the physical process of advection played a minor role in

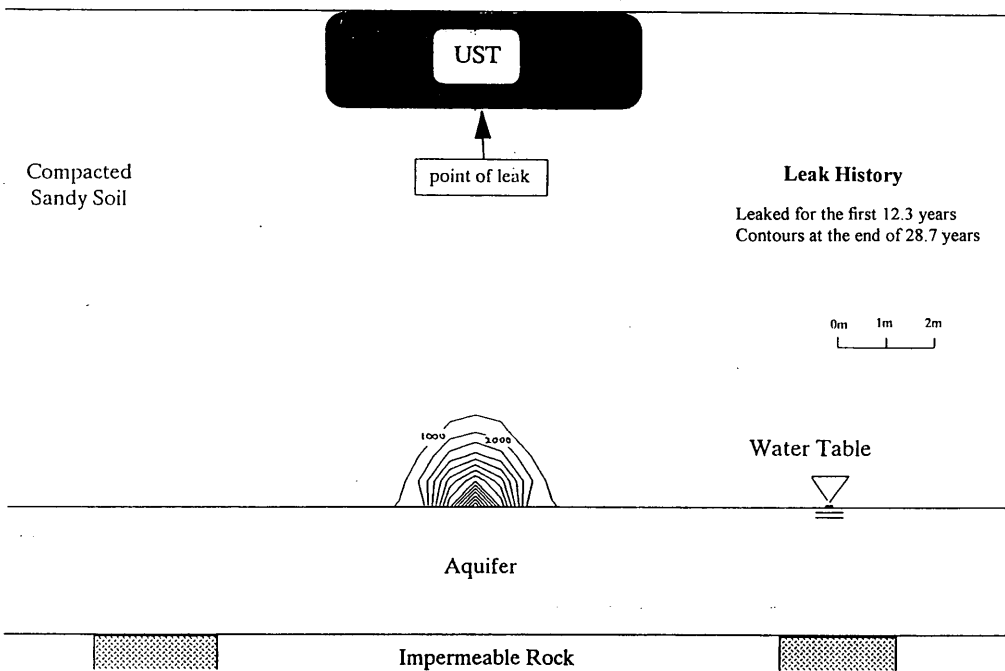


FIGURE 1 Gasoline concentration profile for Test 2 (contour lines in ppm).

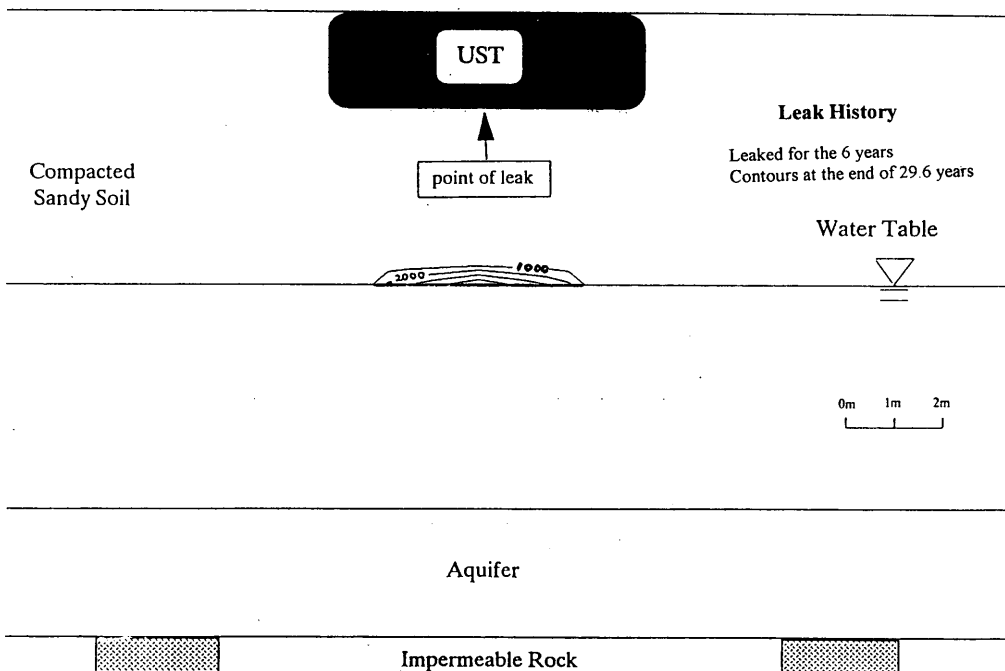
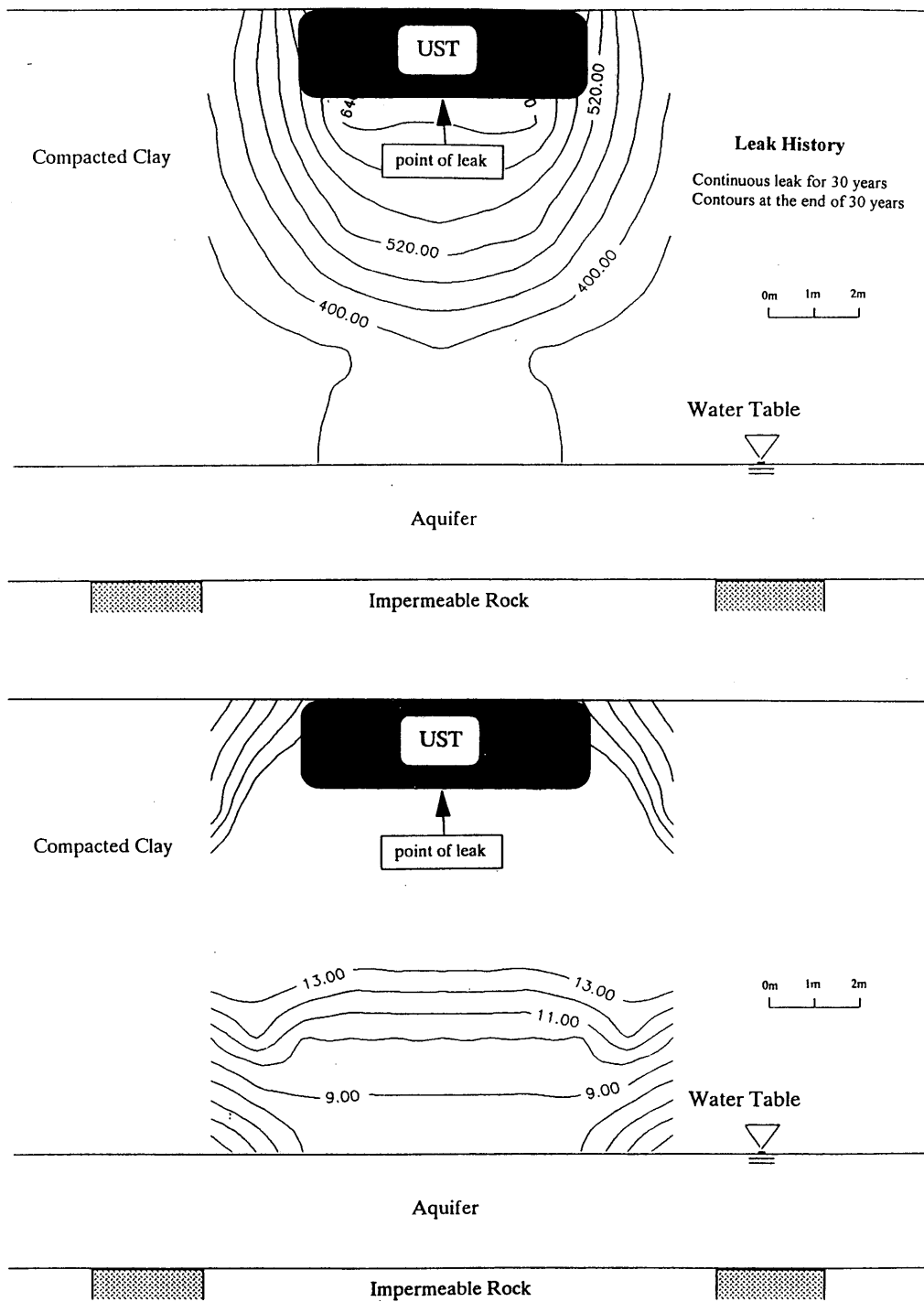
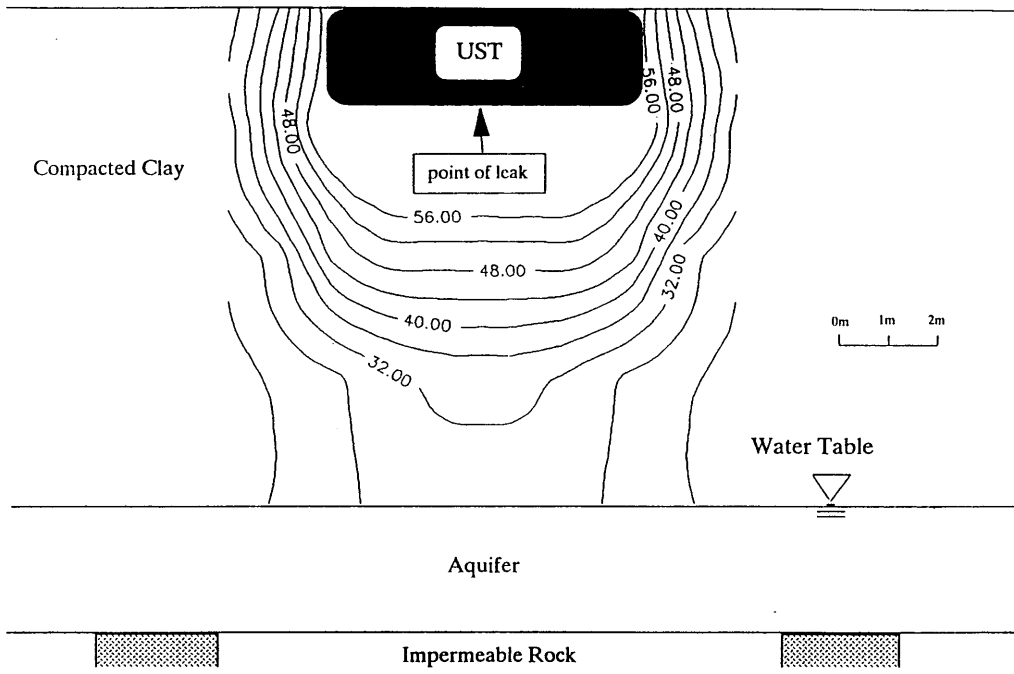


FIGURE 2 Gasoline concentration profile for Test 3 (contour lines in ppm).

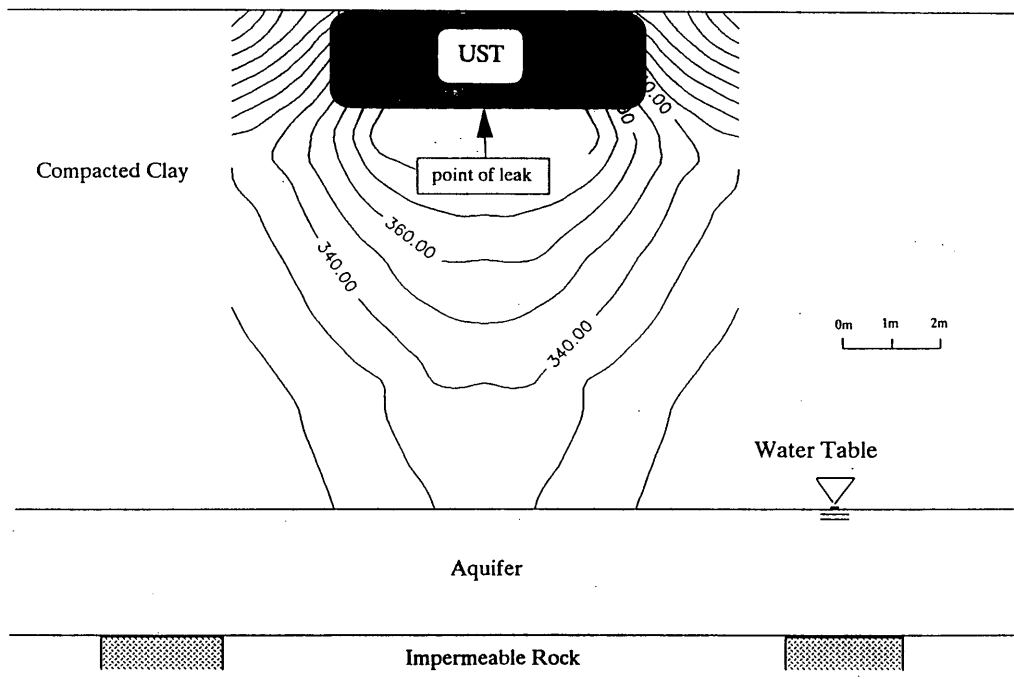


(a)

**FIGURE 3** Gasoline concentration profile for Test 1 (all contour lines in ppm). (a) Pentane concentration profile for Test 1. (b) Hexane concentration profile for Test 1. (c) Benzene concentration profile for Test 1. (d) Toluene concentration profile for Test 1. (e) Xylene concentration profile for Test 1.



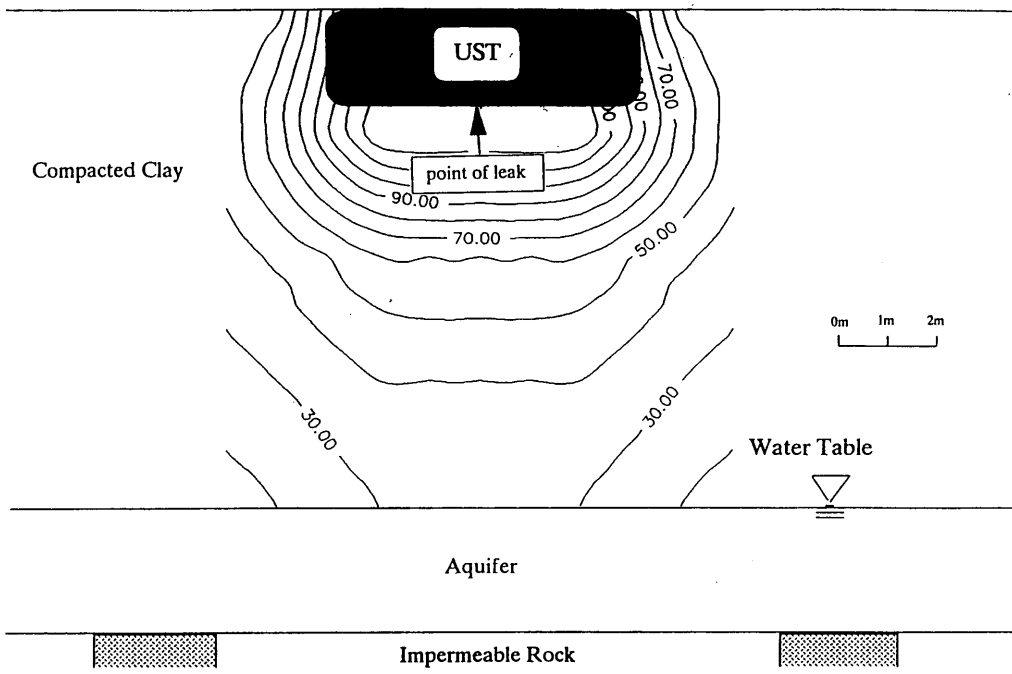
(b)



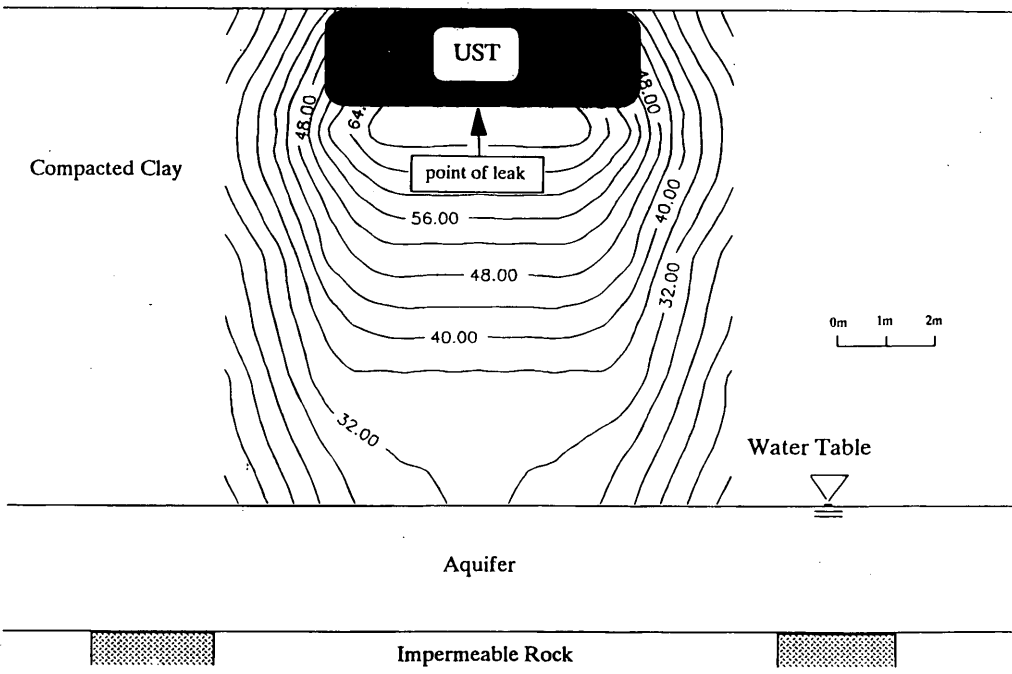
(c)

FIGURE 3 continued

(Continued on next page)



(d)



(e)

FIGURE 3 continued



the movement of contaminants through the clay. The greater spread in lateral directions may be due to the anisotropy of the clay layer resulting from lower tortuosity of the path in the lateral direction as the result of manual and centrifugal compaction of the clay. Even though the same volume of gasoline was stored in this tank as that in Test 2, nearly one-third did not leak. This indicates that the rate of leakage may be governed by both the soil type underneath a UST and the size of the hole.

As seen in Figure 3a, contours for pentane had the lowest level of all of the chemicals present even though its proportion in the tank was among the highest. Its partition coefficient was not available, so no further comments can be made about these contours. It is also believed that the higher vapor pressure resulted in evaporation of most of the pentane leaving only residual values in the soil.

Although the benzene fraction was the second lowest in the synthetic gasoline, as shown in Figure 3c, it yielded the highest levels in the soil. Below the center of the tank at 3 m and 6 m depths the benzene fractions were 360 ppm and 340 ppm, respectively. Even though the amount of toluene in the UST was five times more than benzene (Table 1) it showed the lowest levels of 65 ppm and 39 ppm at the corresponding depths as shown in Figure 3d. Table 1 shows that the water solubility of benzene is more than three times that of toluene. The water content of the soil was about 35 percent, so the higher water solubility of benzene facilitated its movement.

Figures 3a and e show contours for hexane and xylene fractions and both chemicals have essentially identical amounts of about 52 ppm and 34 ppm at 3 m and 6 m depths, respectively. The proportions of free hexane to xylene in the synthetic gasoline were almost the same as that of toluene to benzene but the water solubility of xylene is about 16 times that of hexane (Table 1). Apparently the larger proportion of available hexane has overcome its much lower water solubility.

As seen in Figures 3b and d, hexane showed slightly lower value in the soil than toluene even though both of them comprised about the same fraction in the synthetic gasoline. Note that the water solubility of toluene is 51.5 times greater than that of hexane. The much higher water solubility of toluene did not have as profound an effect on its movement as was observed for benzene. Clearly there is high correlation between water solubilities and the movement of chemicals in soil. Other factors, such as selective transport and saturated vapor pressures, contribute to the movement of chemicals in soils. Therefore, additional experimental investigations are needed to understand mechanisms behind the movement of a group of chemicals through fine grained soils to accurately model their movement and subsequent fate and transport. There was a considerable spread of contaminants in the lateral direction, bulging of contours in the lateral direction, suggesting the contribution of soil anisotropy to movement of chemicals. This is an observation that cannot be simulated in a column test.

The difficulties commonly encountered in obtaining samples during field explorations were also present in this modeling technique. Even when care was taken during soil sampling, there was soil contamination from the boring/sampling device when samples were obtained from different depths. To minimize soil contamination because of sampling, the sampling was started from points with the lowest contamination levels and proceeded to higher levels. Baking the capillary column between GC analyses reduced cross-interference.

## CONCLUSIONS

Three sites with leaking UST were simulated in the centrifuge to determine the movement and subsequent fate and transport of gasoline and its components in the vadose zone. The test results on the sandy or coarse-grained soil showed that gasoline moved because of the physical process of advection and was retained as a pool of free product above the water table. These test results corroborate data reported from laboratory physical models. Therefore it may be concluded that the centrifuge looks feasible to be used as a research/design tool to generate data for unsaturated sites with coarse-grained soils contaminated by leaking UST.

The test results on the clay or fine-grained soil provided the following conclusions:

- The rate of leakage depends on both the type of soil underneath the UST and the amount of corrosion, that is, a UST with a higher corrosion rate may not have larger leakages in fine grained soils.
- The movement of gasoline is not dominated by the physical process of advection as for coarse-grained soils and hence pooled free product was not observed at the water table depth even after 30 years of simulated leakage.
- The movement is not dominated by the physical process of advection, so the contour profile of a given chemical within the soil depends on its water solubility and its fraction in the gasoline.
- There was a considerable spread of contaminants in the lateral direction, an observation that cannot be simulated in a column test.
- The spread of gasoline strongly depends on the anisotropy of the soil through which it travels as indicated by the bulging of contours in the lateral direction.

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