ABSTRACT

THESIS :	Centrifugal Modeling of Leaking Underground Storage Tanks Master of Science in Environmental Engineering
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There are between 2 to 3.5 million Underground Storage Tanks (UST) located through this nation. Most of these tanks store gasoline and fuel oil. Many tanks in fact are or will soon be leaking, thus making them one of the major sources of groundwater contamination in USA. The fate and transport of chemicals from the leaking USTs is estimated from mathematical models. A major difficulty in these modeling efforts has been a crucial lack of information concerning the constitutive relationships governing multiphase contaminant movement.

Centrifugal modeling is an alternative to mathematical modeling, which may give an insight to the problem as well as provide information for mathematical models. The centrifuge creates an actual three-dimensional profile and accelerates the transport processes such that a field event which lasts decades may be simulated within hours or days in centrifuge.

In this thesis, the centrifugal modeling technique is used to investigate study the movement of contaminants and the distribution of gasoline within the vadose zone and from vadose zone to the groundwater.

CENTRIFUGAL MODELING
OF
LEAKING UNDERGROUND STORAGE TANKS

by

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CHAPTER I INTRODUCTION

1.1 The Nature of Subsurface Pollution

The Problems of Subsurface Pollution

Since World War II, the production and disposal of hazardous waste has risen tremendously. But not until the publication of "Silent Spring" did people try to learn about the health effects occurring from hazardous wastes, and how to eliminate consequences. Among the hazardous wastes, subsurface pollution is less known than surface pollution like surface water, or solid waste contamination. Even the most knowledgeable scientists struggle with the inevitable difficulties of collecting data by expensive, remote methods which disturb and modify the samples which the data are collected. The problem resulting from subsurface pollution is far more serious than most people can imagine. For instance, even a small concentration of gasoline could render groundwater unfit for domestic use and the gasoline vapors that are released can migrate into basement, sewers, and tunnels causing fire or explosions.

The main source of complexity in studying subsurface pollution is the heterogeneous nature of the subsurface system. The soil and rock which make up the system are commonly nonuniform. Sand has a high permeability and permits easier passage of large volumes of water, while irregular layers or lenses of clay retard it. Furthermore, many other parameters could contribute to the complex dynamic system of subsurface pollution: e.g. occurring of impervious bedrock, fractured clay beds or rocks, holes left by the decay of plants, stream-groundwater interaction, contaminant exchanges by adjacent layers of different permeabilities, adsorption by the soils, degradation by microorganisms, etc. Figure 1 illustrates the complexity of the subsurface systems. Sometimes, it is impossible to determine even the source of pollution unless the historical records and the modern detection technology are available.

The Sources of Subsurface Pollution

The contaminants in groundwater and soil could be from three categories: 1) direct disposal of hazardous materials , 2) indirect disposal of hazardous materials, 3) non-point sources. (see Figure 2). Examples of first category are a) land fills, b) surface impoundment, c) waste piles, d) land treatment, e) underground injection, and f) illegal dumping. In 1984, there were 93,500 landfills and 181,973 surface impoundments in the United States. (EPA, 1984(a), 1984(b)) Of these surface impoundments, nearly 50 percent are over thin permeable soils and about 78 percent are unlined.

Examples of the second category are a) leaks and spills from storage facilities, b) leaks and spills from transport facilities, c) leaks from treatment facilities, and d) leaks and spills from process facilities. The underground storage tanks (USTs) which is the main issue of this study will be discused in section 1-2. The potential leachate generation from the above major surface contamination sources are shown on Table 1.

Examples of non-point sources are a) urban infiltration, b) agricultural operations, c) mining activities, d) oil and gas exploration. Contamination from non-point source is often undeclared in general reports.

1.2 The Leaking Underground Storage Tanks

The Regulations for Leaking USTs

The definition of USTs, regulated by Resource Conservation and Recovery Act (RCRA), is the tanks, including underground pipes connected to the USTs, with 10 percent or more of its volume underground. USTs are used to store CERCLA-regulated (Comprehensive Environmental Responsibility, Compensation and Liability Act, 1980) hazardous chemicals, process chemicals, dilute waste, and petroleum products.

Of the several million of underground storage tanks (USTs) in USA, nearly 25 percent are now leaking. (various estimations for USTs numbers are shown on Table 2). Many more are going to leak in the future, due to: lack of corrosive protection, faulty installation, spills or overfills, and piping failures. Most of the USTs contain gasoline or hazardous chemicals and it makes the leakage a serious problems of the pollution of the soil and grou pollution of the soil and groundwater. It (EPA, 1988). As many as half of these USTs contains gasoline, which consists of lead benzene, toluene, ethylene dibromide and ethylene dichloride.

Most USTs are fabricated with carbon steel and are protected from corrosion with typical design lives between 15 and 20 years. It is estimated that one million USTs are more than 16 years old, of which as many as 700,000 may be leaking now. Newer USTs are designed to resist corrosion and have double liners. Recent regulation has mandated industry and business to comply with regulations regarding detection, prevention and leak control for USTs. (Cheremisinoff et al., 1990).

In 1984, Congress added the Subtitle I to the RCRA to regulate the leaking USTs. All the existing USTs, installed before and after December 1988, should meet the requirements of correct installation, spill and overspill prevention, corrosive protection and detection. (EPA, 1988).

On May 2, 1990, EPA also published amendments in Federal Register affecting the financial responsibility requirement deadlines for USTs, 40 CFR 280.91(c). Under the modification, all petroleum marketing firms owning 13 to 99 USTs at more than one facility will be required to comply with the requirements of 40 CFR part 280, sub-part H -- Financial Responsibility -- as of April 26, 1990. The amendments provide additional time to develop financial assurance mechanisms (especially, state assurance funds) to enable this group to comply.

Public Response

In responding to escalating pollution caused from USTs, most of the states, such as New Jersey, California, and Massachusetts, have proposed very stringent regulation on leaking USTs and the soils contaminated from these USTs. Proportionally, the cost of disposal has increased tremendously and has reached as high as $100 for the disposal of one ton of contaminated soil. In order to reduce the cost and to find the best option for clean-up, US EPA risk reduction engineering lab, Edison, NJ, produced a comprehensive document to direct the users to select the most appropriate corrective action technology (Rosenberg et al., 1988). However, this document does not discuss the extent of contamination or the levels of contamination for the site contaminated by USTs. Therefore, there is a need to establish comprehensive data base for all possible sites contaminated by USTs and to provide the best available correction technology to decontaminate the sites. The best available corrective action technologies are soil venting, soil flushing, soil cover, soil barrier, biodegradation and disposal. Although soil contamination from USTs is an environmental-geotechnical problem, except for a few geotechnical firms, most of the geotechnical professionals shy away from dealing with site contaminated by USTs.

The Difficulties in Investigating Leaking USTs

The research on the behavior of pollutants leaked from USTs becomes very complicated due to the heterogeneous nature of subsurface systems. When dealing with this problem, one should consider the following variables: (1) for the environmental aspect, the variables that should be considered are the amount of leak, duration of leak and the types of oils, including their transformation, degradation and volatilization; (2) for hydrological aspect, the variables that should be included are the amounts of surface runoff and duration and intensity of rainfall; (3) for geotechnical concern, soil types (which contribute to hydraulic conductivity, porosity and adsorption capacity), thickness of soil layers, and inclination of soil layers should be considered as variables; (4) for hydrogeological aspect, the appropriate variables are the water table depth, and the rate and the direction of groundwater movements. In order to establish a data base for the regulators to define the clean-up standards, the researchers must find a feasible way to consider and simplify the complexity of subsurface systems with all the above variables.

1.3 The Scope of This Study

The goal of this study is to investigate the movement of pollutants using the centrifuge: 1) from USTs to the vadose zone to the groundwater ; 2) the distribution of gasoline within the vadose zone; 3) the migration of gasoline leachate to the groundwater; and 4) The impact of residually contaminated soils on the quality of groundwater resource.

CHAPTER II HISTORICAL REVIEW

2.1 Problems and Solutions

There are different approaches to deal with the problems of leaking USTs. Figure 3 shows the logical approaches that may be taken to clean-up for the leaking USTs. Through one or more of the methods, a data base should be established to find the extent of the contamination and the most appropriate decontamination options for each site.

2.2 Field Monitoring and Gathering of Field Data

The first approach is to gather the field data through field monitoring. Though this approach is the best it may not be feasible because of the cost of drilling. Time poses a great obstacle to this method as it is not possible to get data from the past or future. Also, obtaining the undisturbed samples may be a difficult task. Due to loss of volatiles, the analysis of samples may not be reliable if the samples are to be transported to a laboratory. One may also disturb the transport process due to field monitoring to contaminate the region where otherwise would not have being contaminated due to the fate and transport of contaminants.

2.3 Mathematical Modeling

The mathematical models of the fate and transport of pollutants in a multi-media environment are attractive because they offer a relatively rapid and inexpensive way to access the existing or potential environmental hazards during the early stages of chemical production or process developments. But existing multimedia / multiphase models have an inherent deficiency to describe intermediary transport across the soil air and unsaturated / saturated soils zones. Furthermore, pollutant transport in the soil matrix constitutes an unusual local multimedia transport problem. Accurate mathematical model, thus, is hard to develop (Cohen, 1986). For example, PESTAN model (developed by EPA, R.S. Kerr Environmental. Research Lab, Ada, Oklahoma), considers 1st order decay, dispersion, linear sorption. However, relative timing, intensities of water, chemical application rate, and spatial variability will cause underestimation of the model predictions (Donigian and Rao, 1986). SESOIL (Seasonal Soil Compartment Model) [developed by EPA office of Toxic Substances] had integrated hydrologic cycle, sediment cycle, and pollutant cycle. But it needed many input data and it confronted with the difficulties arising from the differences in time scales (Donigian & Rao, 1986).

2.4 Physical Modeling

Physical modeling, on the other hand, may give an insight to the problem and may provide data for the validation of mathematical model. The physical modeling for the movement of fluids immiscible with water had been attempted by numerous researchers (Schwille, 1967, 1981; Mckee, et al., 1972; Payatakes, 1982; Hoag et al., 1986; Ambrose et al., 1988; Farr et al., 1990). Because of the obvious difficulty in gathering long term field data or the numerous input data for mathematical modeling, the researchers turned to the third alternative namely, the physical modeling. Most of the large scale physical models consisted of long one-dimensional columns or big soil box experiments (Parmer, 1989). The soil box experiments yield interesting and useful data for problem caused by leaking USTs. However, the soil box experiment takes considerable time and it is very expensive to construct. It is also difficult to control and achieve constant soil profile due to the size of the box. In this study, the centrifuge modeling which is an improvement upon the physical modeling technique is used to investigate the fate and migration of subsurface pollutants.

CHAPTER III METHODOLOGY

3.1 The Application of Centrifugal Modeling to Simulate Subsurface Pollutant Transport

The centrifuge modeling technique is an improvement upon the physical modeling technique in which the size is made smaller and the time is accelerated through scaling laws. (Arulanandan et al., 1988; Celorie et al., 1989). Due to gravitational field of Ng generated by running the centrifuge, the scale of the sample size, force, and mass can be N^{-1}, N^{-2}, and N^{-3} of that in prototype respectively. The stress which produce varying porosities is as same as it in prototype. In addition, the centrifuge can simulate the in-situ capillary fringe, the water table, the moving of groundwater, any capillary trapping and most of the other features encountered in the field. (Arulanandan et al., 1988). The summary of scaling laws of centrifugal modeling is in Table 3.

The fate and transport of contaminant through soils is due to the physical processes of advection and dispersion, chemical process of adsorption, and the biological process of degradation (Freeze and Cherry, 1979) If the interstitial flow velocity, which is average Darcy velocity over porosity, is made N times of that in prototype, the time for physical processes of advection and diffusion will have the same scale, i.e. N^{-2} of prototype time (Arulanandan et al., 1988). If the centrifugal testing is limited to fine-grained soils (less than 0.015 cm), the interstitial flow velocity may be small enough to make Renold number (Re) less than one ensuring laminar flow regime in the models. Under such conditions model will have the same scaling law for time for problems of advection-dispersion. Since the same soil is used in the model, the chemical process of adsorption can be viewed as a retardation of the physical processes of advection and dispersion hence it will follow the same scaling law, i.e. N^{-2} of the prototype time. (Arulanandan et al., 1988; Celorie et al., 1989). Due to the different time scale the biological process of degradation cannot be modeled in the centrifuge.

3.2 Centrifugal Modeling of the Unsaturated Zone

Studies on unsaturated or multiphase flow using centrifuge technique had been done by several researchers (Alemi, et al., 1976; Hagoort, 1980; van Spronsen, 1982; Nimmo, et al., 1987; Nimmo, 1990). Kool et al.(1987) used the centrifuge to measure transient flow quantities. Through this inverse approach, the flow character can be still identified.

The fate and transport of the contaminants through unsaturated soils can be modeled in the centrifuge provided the following conditions are satisfied: 1) Richard's equation is valid, 2) the capillary fringe is simulated, and 3) suction pressures are simulated.

The unsaturated flow is governed by Richard's equation which incorporates Darcy's Law and the condition of continuity. Nimmo et al. (1987) proved that the Darcy's Law is still valid in centrifuge for steady state flow, and Nimmo (1990) stated that "there is no strong evidence of the violation of unsaturated flow theory in centrifuge", i.e. the Richard's equation is valid even for value of permeability as low as 10^{-10} m/s and under relative dry conditions up to 1/3 of the saturation water content. Table 4 shows the Richard's equation in 1-D form, centrifugal form, and finite difference form.

The capillary rise, H_c, in a unsaturated soil is given by:

$$H_c = 2*T/(r_c * g * d_w) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

where T = surface tension of pore fluid, r_c = radius of the capillary tube, g = acceleration due to gravity, d_w = density of water. Consider two situations: a field situation with a capillary height h_c, and a centrifugal model of size 1/N of the field made of the same soil. Since the same soil is used in centrifugal model under gravitational field of Ng, the capillary head will be reduced to h_c/N according to equation (1). If the scaling laws are applied for the length measurements, the model capillary fringe will coincide with that of the prototype.

The centrifuge can simulate the total potential gradient (P_{total}) that governs the flow of groundwater in saturated and unsaturated zones. Near or below water table, the gravitational potential (P_g) will dominate in P_{total}. At a significant distance from water table, evaporation or infiltration may cause spatially abrupt and relatively rapid change in P_{total} resulting in high suction gradients. At a global scale the net flow through soil layers may be influenced by gravity, while in local scale the flow is caused by suction gradient. (Arulanandan et al., 1988)

The acceptable centrifugal field is up to 200 g for simulating unsaturated flow as shown by Nimmo (1990). For all the tests cited in this paper, the conditions for centrifugal testing are controlled within the above limits (i.e. grain size, total leakage calculation, etc.) in this section to ensure the validity of unsaturated flow modeling.

CHAPTER IV EXPERIMENT PROCEDURE

The flow chart for the experiment described in this study is shown in Figure 5. The four main items are: 1) sample preparation including Gas Chromatography (GC) calibration, 2) centrifugal testing, 3) GC analysis of soil samples, and 4) the interpolation and contouring of data. Six test results are reported in this thesis. The plan area for each site was 26 m * 25 m and the gasoline tank was installed at the middle of the site. The bottom of the tank was at 2.0 m below the ground surface and the top was flush with ground surface. The tank was a cylindrical drum of 6.4 m inside diameter and 2.0 m height. A small hole was drilled at the center of the tank to simulate a leaking UST. The symmetry of the site was taken advantage in the interpolation of the data when generating the equi-concentration lines. Figure 6 shows the layout of the centrifugal sample.

The centrifuge at the New Jersey Institute of Technology is an Genisco Model 1068 with 5g-ton capacity. It is equipped with twenty-four slip-rings and two hydraulic glands. The maximum speed it can read is 450 RPM. The radius of the centrifuge is 0.65 m. The arm of the centrifuge is connected to swing-up buckets of internal dimension 32.0 * 25.4 * 20.3 cm. A special container was designed to maintain the inclined groundwater table. Therefore, the maximum size of the sample is 23 * 22 * 19 cm. A video camera and a preamplifier are mounted inside the centrifuge. A sixteen bits high-speed data acquisition card in a PC-XT is used to acquire the data from the centrifuge.

4.1 The Setup of Experiment

There are four steps for this section, i.e.,(1) gasoline mixing, (2) GC calibration of gasoline mixture, (3) soil mixing and soil compaction.

Gasoline Mixing

In order to get the standard reference mixture for gasoline, one should consider the representiveness of the selected standard (i.e. averaging the differences between the commercial gasoline) and the stability of standard (such as the volatilization and chemical/microbiological transformation). There were several attempts to find a suitable and a representative mixture of gasoline (Snell et al., 1971; Patton and Stevens, 1984; Laseman, 1986; Baehr, 1987; Stetzenbach, 1987; Lyman, 1988; Sutton, 1988; Kreamer, 1990). Between meeting the representativeness and practicability to make the mixture, the author traded it off to get the surrogate gasoline so that it can reach the requirements of vapor density, air diffusion coefficient, and basic chemical constituency. Besides, the surrogate gasoline can also be reproducible easily in the laboratory. The gasoline vapor consists of 3/4 butane and 1/4 pentane. The author selected pentane as a component of gasoline mixture as the butane was difficult to be detected and handled. hexane was also selected from the aliphatic group. From the aromatic group, toluene, o-xylene, and benzene were selected. Instead to meet the EPA regulation that the benzene should be below 2%, the author chose the 6.2% which would produce conservative predictions. The final proportion of the above components is shown in Table 5. The thermodynamic parameters of the components of gasoline are shown in Table 6.

GC Calibration

For GC calibration of the gasoline mixture, three levels of standards, i.e. 400 , 800 , and 1600 ppm, for each chemical, and four levels of standards, i.e. 62.5, 250, 1000, 4000, and 16000 ppm, for the gasoline mixture were used. Those GC calibration were performed using the Perkin-Elmer Model 8500 Gas Chromatographer equipped with Flame Ionization Detector (FID). A Borosilicate Glass capillary column with a SPB1 Stationary Phase, a 0.75 millimeter inside diameter, a length of 60 meters, and a film thickness of 1.0 micrometers, was used. Carbon disulfide was used as a solvent for preparing the standards and for extracting gasoline from soil samples. The oven was programmed at 40 , 50, 80 °C for 1 minute each, and 180 °C for the rest 8 minutes to obtain the highest resolution for the solvent and the five components of the synthetic gasoline. The above temperatures were selected based on the ranges of boiling point for each component and separation of the retention peaks of pentane and carbon disulfide. Each time 0.4 microliter solution was injected into the GC. Each calibration was based on the average of two injections. If there was a considerable deviation, the tests were repeated. The calibration curves for both individual components and for total gasoline mixture are shown on Figure 7 and Figure 8, on which the GC analysis of soil samples are based.

Soil Mixing and Soil Compaction

First, the site was prepared by adding water and maintaining the water table at the desired level and the sand was dry pluviated to obtain a final height of 2.5 cm. The top of the sand layer was leveled by scraping the excess sand. Before placing the soil layer the air dried soil was mixed with desired amount of water (35% by weight for kaoline, 5% by weight for sandy soil, and 15% New Brunswick Soil). The engineering properties of soils tested were shown in Table 7. The clay soil in test 3 and test 8 was compacted to a final unit weight of 13.81 KN/m^2 (88 pcf) in four equal layers. Care was taken to make the clay homogeneous in the transverse direction. The sandy soil in test 4 and test 5 was compacted to a final unit weight of 12.36 KN/m^2 (79 pcf) in four equal layers. The New Brunswick Soil in test 10 and test 12 was compacted to a final unit weight of 15.2 KN/m^2 (97 pcf) in four equal layers. Once the site was prepared the empty gasoline tank was installed after excavating the soil. Then, the site was tested in the centrifuge.

4.2 Centrifugal Testing

The prepared site was installed in the centrifuge and then spun for twenty four hours (real time) so the sample can get further compacted, due to the gravity, and come to equilibrium. At this stage a desired amount of gasoline was added and the clock was started for the gasoline leakage. At the end of the test the centrifuge was stopped and the five excavations were made and samples were extracted as described earlier.

4.3 GC Analysis of Soil Samples

At the end of centrifugal testing, twenty five soil samples were taken from point A, B, C, D , and E in test 3 test 4, and test 5 ; the same number of samples were taken from the point A, C, F, G, and H in test 8, test 10, and test 12. A is under the center of gasoline tank. B, C, D, E, F, G, and H are from the center of gasoline tank in distances of 2, 3, 4, 5, 6, 9, and 12 cm respectively. Five samples were taken from each boring location: 1) from the top of the soil layer; 2) at a depth of 2.5 cm or (2.9 m on prototype); 3) at a depth of 5.0 cm (5.7 m on prototype); 4) at a depth of 7.5 cm (8.6 m on prototype); 5) at the bottom of the soil layer. From each location roughly 0.8 g of wet soil was taken and put in a 5 ml vial. Three milliliter of carbon disulfide was injected to all the soil samples obtained from test 3, and test 8. One milliliter of carbon disulfide was injected into all the soil samples obtained from test 4, test 5, test 10, and test 12.. A larger quantity of the solvent was required for clay type soils. The vial was shaken well to dissolve all the gasoline from the solid, liquid and air phases in carbon disulfide. A 0.4 microliter of solution extracted from each vial was injected into the GC to detect the chemicals and to determine their concentrations.

4.4 Data Interpolation and Contours Generation

In order to generate the contours of concentration, interpolation of the data was made at the symmetrical points corresponding to the data points on the other side of Y-axis, i.e. all the original points except those with zero X-axis were duplicated and change the sign of the X-axis to be negative. Thus, total number of data points would be 45 including 20 points obtained due to symmetry.

All the above data points served as input data to the computer program, SURFER Access System Version 3.00, which generated the contours of concentration. During contours generation, the quadrant search was used to determine which data points to be applied to calculate the current grid point in order to improve the accuracy of the grid for linearly oriented data. The Kriging method, a geo-statistical techniques, was used to calculate the auto-correlation between data points to produce a minimum-variance unbiased estimates.

CHAPTER V EXPERIMENT RESULTS AND DISCUSSION

The parameters for each test are shown on Table 8. These parameters include the types of soil tested, the density of soil, the size of soil particles, the duration simulated, the simulated total leakage of gasoline, and the water level for each sample. The speed of centrifuge was kept constant at 400 RPM for each test to generate the gravitational field of 104 g, the average gravity between values at both ends of sample, i.e. 95 g and 114 g. The following are the results for test 3, 4, 5, 8, 10, and 12.

5.1 Results for Clayey Soil ——— Test 3 and Test 8

For test 3 and test 8, the results added a new dimension to the data base on USTs. The spread of total gasoline concentration in ppm after 30 years of continuous leakage is shown in Figure 8 and Figure 9 for test 3 and test 8. Most of the gasoline appears to be transported by the physical process of diffusion. It appears that there is no hydrodynamic dispersion and there is considerable spread in the lateral directions. The spread in the lateral directions may be due to the anisotropy of the clay layer resulting from lower tortuosity of the path in the lateral direction as a result from manual and centrifugal compaction of the clay. Even though the same volume of gasoline was stored in the tank of test 3 as in the other tests, nearly 1/3 of it did not leak. This is another interesting observation that the amount of leakage appears to be governed by the type of soil underneath a UST and not necessarily the size of the hole.

The spread of individual components of gasoline in ppm after 30 years of continuous leakage is shown in Figure 8A, 8B, 8C, (for test 3) 9A, 9B, and 9C (for test 8). The above figures appear to provide many interesting observation on spread of gasoline in clay type soils. Though both the starting concentrations of Pentane and Hexane were very high (28.5 % each), they almost totally evaporate from the clayey soil at the end of the tests. Benzene, on the other hand, had the lowest concentration in the UST (6.2%), but it appeared to spread faster and yielded the highest concentration levels. The above results verified the predictions of Baehr (1987): the aliphatic group with lower molecular weights (e.g. pentane, hexane, etc) will find their way into the atmosphere, while the aromatic group (e.g. benzene, toluene, o-xylene, etc.) will migrate downwards and spread away from the source. The solubility and other physical/chemical properties of gasoline and the components of gasoline are given in Table 6. Table 6, Figures 8A through 8C, as well as Figure 9A through Figure 9C clearly show that the spread of gasoline in nearly saturated clays is limited by the solubility of the individual components and also that the spread of each component depends on the molecular diffusion of the species and the anisotropy of the medium.

5.2 Results for Sandy Soil ——— Test 4 and Test 5

The contours of total gasoline concentration in ppm for test 4 and test 5 at the end of the simulation are shown in Figure 10 and Figure 11. The contour plot for the test 4 is similar to the plots obtained from field monitoring studies, mathematical model predictions and physical model results (Schwille, 1967; EPA, 1987; Levy et al., 1990). The above researchers showed that the influence of gravity would dominate over that of capillary forces in the unsaturated zone and that the gasoline will move downwards and accumulate when reaching the capillary fringe and water table.

For the test 4, the leaked gasoline moved as a front and accumulated as a pool of free product above the capillary fringe during final 28.5 years. Most of the gasoline had moved due to the physical process of advection. The advection dominated over the diffusion and dispersion even when there was no infiltration of rainwater.

The contour plot of test 5 is quite similar to that of test 4 except there is additional information on the gasoline concentrations below the water table. The gasoline accumulated just above the capillary fringe in this test. There was considerable amount of dissolved gasoline in the soil below the water table. But the maximum amount dissolved was less than 150 ppm. The movement of gasoline below the water table appears to be dominated by the physical process of diffusion.

5.3 Results for New Brunswick Soil ——— Test 10 and Test 12

The patterns of concentration contours for Test 10 and Test 12 (Figure 12, Figure 12A through Figure 12C, Figure 13, and Figure 13A through 13C) show again the influence of diffusion on subsurface gasoline transport. Checking the difference in migration patterns of this set (test 10 and test 12) with the former set (test 4 and test 5), it clearly demonstrates that diffusion will dominate advection in finer soil since the capillary force is relatively higher than that in sandy soil.

The fact that the concentration of pollutant propagates with time was also illustrated by comparing the results of test 10 with these of test 12. The above is a very evident truth, but the patterns give very interesting information of selective transport in vadose zone. In 0.8 year simulation of test 10, the toluene dominates the other component; while o-xylene shows the highest concentration in 24 year simulation of Test 12. The reasons could be the influence of initial proportion of each chemical (30.7% for toluene, 6.1% for o-xylene) and the interactions of their thermodynamic properties (e.g. o-xylene has higher molecular weight, toluene has higher aqueous solubility, etc.).

5.4 Discussion and Suggestion for Further Research

The transports of volatile organics were simulated in different type of soils, with different duration and different leakage, using centrifuge modeling. For all the test results, pentane and hexane are absent due to high vapor pressures and low molecular weights. benzene, toluene, and o-xylene, on the other hand, show a competitive migration in the unsaturated zone. These migration patterns provide an insight to subsurface pollution study.

However, the parameters of test condition should be properly controlled for further research. The temperature during testing should be monitored carefully as the volatiles are sensitive to the temperature changes. The calibration for differential extraction of organic form each type of soil should be done and the results should be compared with these obtained by EPA methods. The blank group or the dummy samples should be controlled with the same condition as those in the tested sample to correct the falling head leakage in warm environment. The field data are also needed to verified the centrifugal modeling and to provide the limitation of the testing parameters. The test with infiltration will be very interesting because it may re-mobilize the deposit of adsorbed contaminant and help to access the impact of residual contaminated soil.

More efforts are also needed to convince the researchers who do not know or doubt the validity and ability of centrifugal models to simulate subsurface pollutant transport. More papers and experiments should be made available to explain and solve the following problems: 1) Since centrifuge force is r-dependent (r is the length from center of centrifuge to the sample), it is spatially non-uniformed. The maximum size of sample should be checked by further experiments. 2) The great body force generated by centrifugal acceleration will dominate all other forces (e.g. suction gradient), change the shape of air-water interface, and alter the micro-scale distribution of water. More validation test should be demonstrated to extend the application of centrifugal modeling to pollutant transport in porous media.

CHAPTER VI SUMMARY AND CONCLUSION

Subsurface pollution caused by leaking underground storage tanks was studied in this paper. Three approaches to investigate the transport phenomena were examined, i.e. the gathering of field data, mathematical models, and physical models. Centrifugal modeling, a improved physical modelling technique, appeared to preferable over the other methods.

Several sites with leaking underground storage tanks were simulated in the centrifuge to find the movement of gasoline in the vadose zone. The synthetic gasoline was calibrated in a GC against different percentages of carbon disulfide to estimate the concentration of total gasoline and the concentration of individual components.

The test results on clayey type soils provided the following conclusions. 1) the rate of leakage depends on the type of soil underneath a USTs; 2) the movement of gasoline is governed by the physical process of diffusion; 3) the spread of gasoline strongly depends on the anisotropy of the soil; 4) the concentration profile depends on the solubility of each of the component and their initial concentrations

The tests results on sand type soils show that gasoline moves due to the physical process of advection and is retained as a pool of free product at the water table. The test results on sandy soil are similar to the results of tests reported based on mathematical models, conventional physical models and actual field data. Therefore, the centrifuge may be used as a research tool to generate data for sites contaminated by USTs.

The test results on New Brunswick Soil show the observation of selective transport of subsurface pollutants. They also demonstrate the propagation of chemical concentration with time in the unsaturated zone. The above phenomena could be resulted from the difference in initial concentrations and the interactions between the thermodynamic properties of chemicals.

The above migration patterns generated by centrifugal modelling provide an insight to the subsurface pollution study. However, the parameters of test condition should be properly controlled to bring about consistent and reproducible results. More efforts are also needed to extend the testing limits of centrifugal modeling.

TABLE 1 Potential Leachate Generation from Major Subsurface Pollution Sources

[Devinny, et al., 1990]

SOURCES	POLLUTED GROUNDWATER (BILLION GALLONS/YEAR)
Landfills and dumps	450
Surface impoundments	300
Septic tanks	1,300
Leaking USTs	2,000*
Others	N/A

* The leakage of USTs is 0.5 billion gallons / year. It needs 4,000 gallons of groundwater to dilute 1 gallon of pure chemical to the concentration of 1,000 ppm.

TABLE 2 Estimations for Number of USTs in USA (1983 -1990)

Estimations for the Number of Underground Stroage Tanks [millions]	Referces
1.4 (Gasoline)	Water Information Center, Inc. (1983)
1.5 - 2.0	Aspen System Corp. (1984), Tejeda (1984) Lehman (1984)
> 3.5	Dowd (1984), Jafek (1985)
4.0 (Patroleum) 0.2- 0.4 (Chemicals)	Dwyer (1985)
2.345	Hansen (1985)
2.5 - 5.0	Cheremisinoff (1990)

TABLE 3 Scaling Laws for Centrifuge Modeling

Quantity		Prototype	Centrifuge Model [n g's]
Linear Dimension		1	1/n
Area		1	$1/n^2$
Volume		1	$1/n^3$
Time	Dynamic Events	1	1/n
	Hydrodynamic Events	1	$1/n^2$
	Viscous Flow	1	1
Velocity (Distance/Time)		1	1
Acceleration (Distance/Time^2)		1	n
Mass		1	$1/n^3$
Force		1	$1/n^2$
Energy		1	$1/n^3$
Stress (Force/Area)		1	1
Strain (Displacement/Unit Length)		1	1
Density		1	1
Frequency		1	n

TABLE 4 Richard's Equations in Different Forms

Forms	Richard's Equation	Remarks
1-D Form	$$\frac{\partial\theta}{\partial\psi}\frac{\partial\psi}{\partial t} = \frac{\partial}{\partial z}\left(k\frac{\partial\psi}{\partial z}\right) + \rho g\frac{\partial k}{\partial z}$$	*Incorporating Darcy's law and condition of continuity
Centrifugal Form	$$\frac{\partial\theta}{\partial\psi}\frac{\partial\psi}{\partial t} = \frac{\partial}{\partial z}\left(k\frac{\partial\psi}{\partial z}\right) - \rho\omega^2\frac{\partial}{\partial r}(rk)$$	*for incompressible medium *a special case of finite difference form
Finite Difference Form	$$C_j^n\left[\frac{\psi_j^{n+1} - \psi_j^n}{\Delta t}\right] = \frac{1}{(\Delta r)^2}\cdot$$ $$\left[k_{j+1}^n\psi_{j+1}^{n+1} - (k_j + k_{j+1}^n)\psi_j^{n+1} + k_j^n\psi_{j-1}^{n+1}\right]$$ $$-\frac{1}{\Delta r}(k_{j+1}^n - K_j^n)\rho\omega^2 r_j - k_j^n\rho\omega^2$$	*Using implicit method (Bear et al., 1984)
Notations	θ : volumetric water content ψ : metric potential t : time z : vertical space coordinate k : hydraulic conductivity as function of θ or ψ ρ : density of water	ω : angular speed of rotation r : distance from the center of rotation C : specific water capacity of medium n : time indice j : space indice

TABLE 5 Components of Synthetic Gasoline

Component	Percentage by Weight
Pentane	28.5
Hexane	28.5
Benzene	6.2
Toluene	30.7
O-Xylene	6.1

TABLE 6 Thermodynamic Parameters for Gasoline and Its Components

[after MacKay et al., 1981 and Bruell et al., 1984]

Chemicals	Molecular Weight	Saturated Vapor Concentration (mg/l)	Aqueous Solubility at 20°C (mg/l)
Pentane	72	N/A	N/A
N-Hexane	86	630	10
Benzene	78	300	1780
Toluene	92	130	515
O-Xylene	106	30	152
Gasoline	100	411	156

TABLE 7 Geotechnical Properties of Soils Tested

Soil Type	Kaolin	New Brunswick Soil	Sandy Soil
Liquid Limits (%)	48	37	N/A
Plastic Limits (%)	36	34	N/A
Surface Area (m^2/g)	55	98	N/A
Caly Fraction (%)	85	24	0.0
D_{10} (mm)	N/A	N/A	0.117
C_u	N/A	N/A	3.4
C_c	N/A	N/A	1.6
Max. Dry Unit Wt. (KN/m^3)	14.1	16.6	13.0
Optimun Water Content (%)	30	19.5	10

TABLE 8 Parameters of Test Condition

Condition	Test 3	Test 8	Test 4	Test 5	Test 10	Test 12
Type of Soil	Klayey Soil	Klayey Soil	Sandy Soil	Sandy Soil	NBS**	NBS**
Density of Soil [pcf]	88	88	79	79	97	97
Grain Size [passing #]	200	200	20	20	40	40
Duration Simulated [yr]	30	30	28.7	30	0.8	24
Total Leak [ml]	22.0	16.5	40.0	16.0	9.0	17.5
Water Level* [cm]	0.2	2.0	2.0	3.5	1.5	0.2

* Water level was measured from the top of aquifer ** New Brunswick Soil

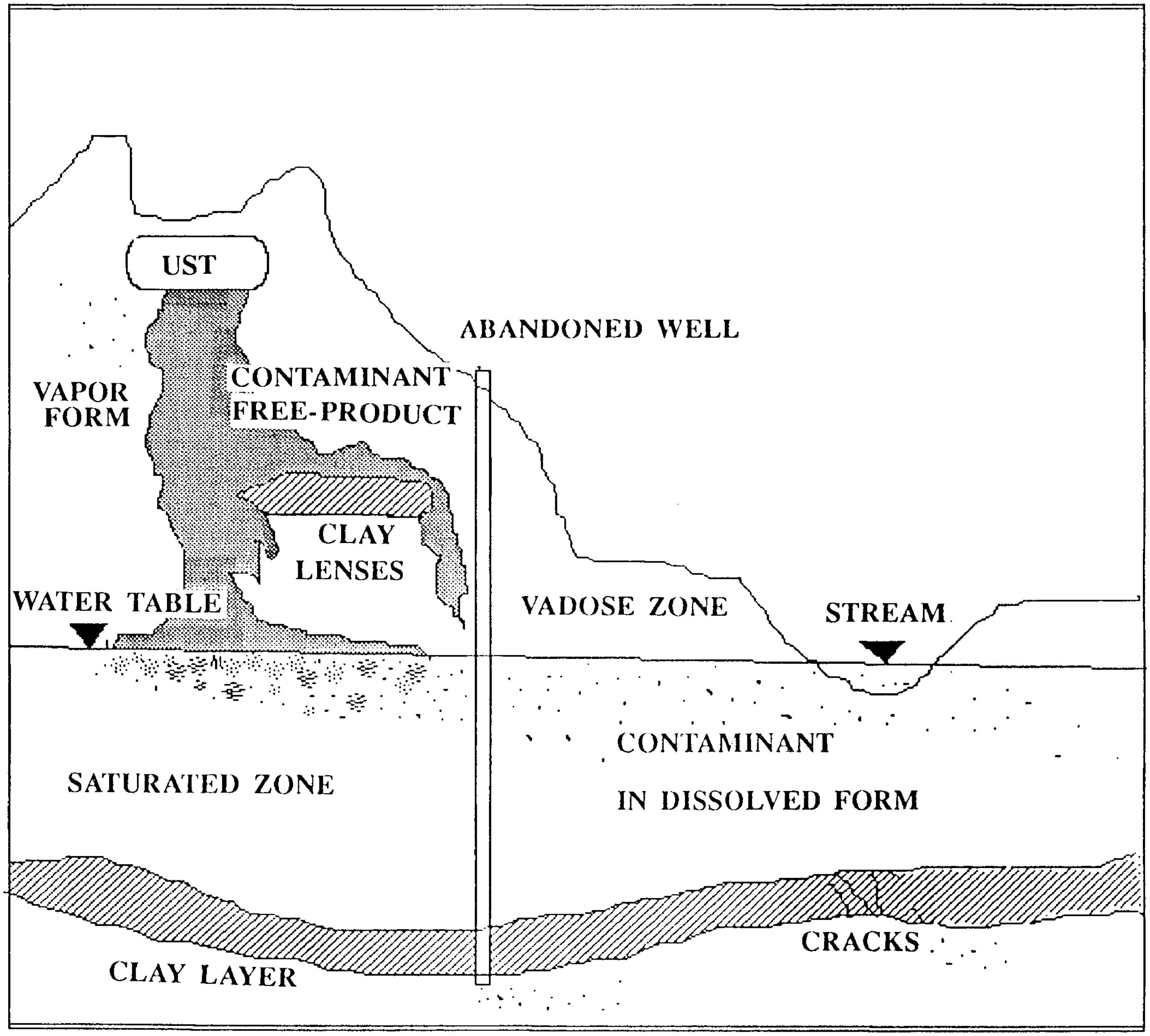


FIGURE 1 Dynamic Subsurface System

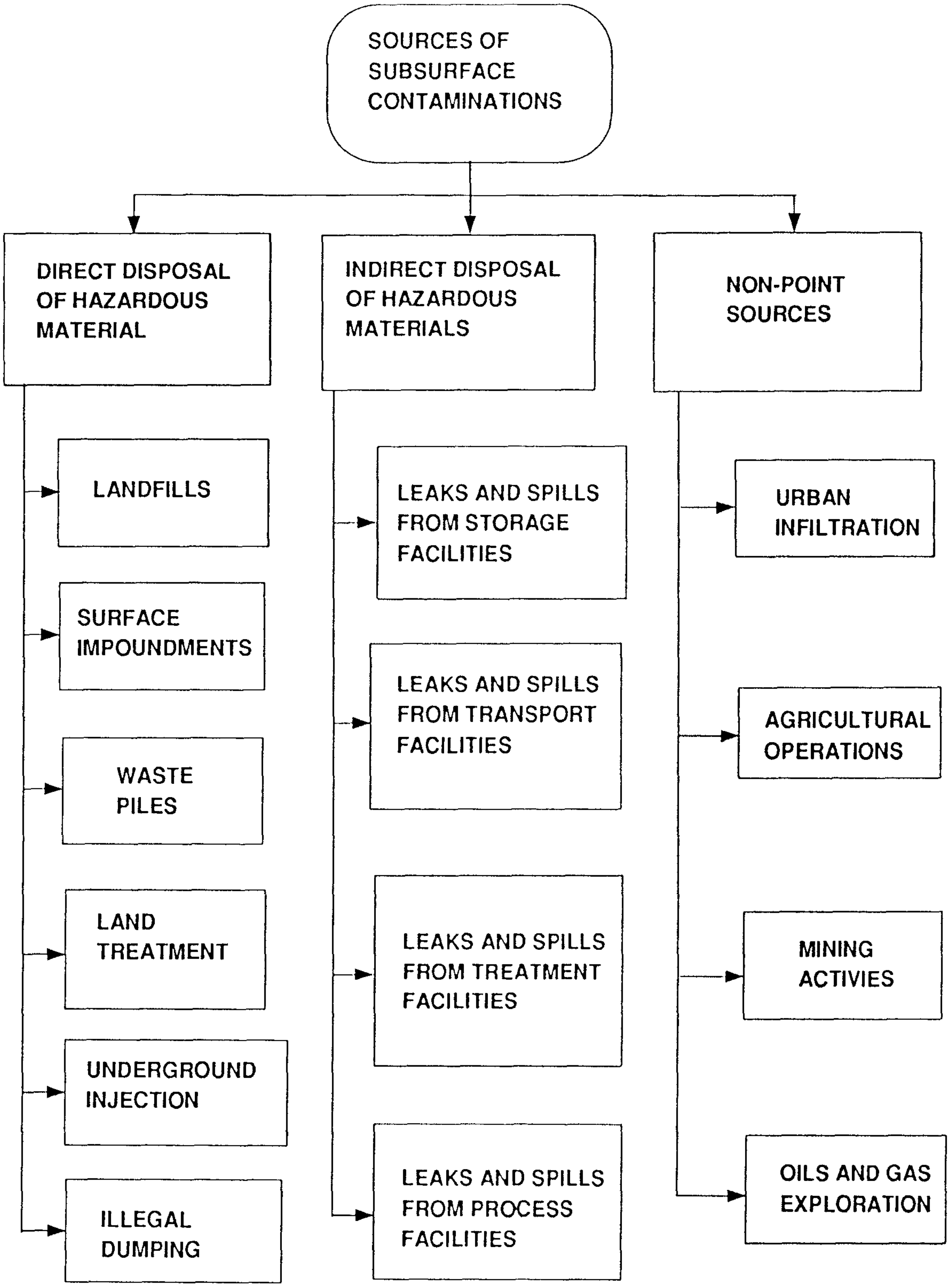


FIGURE 2 Sources of Sub-subsurface Contaminations

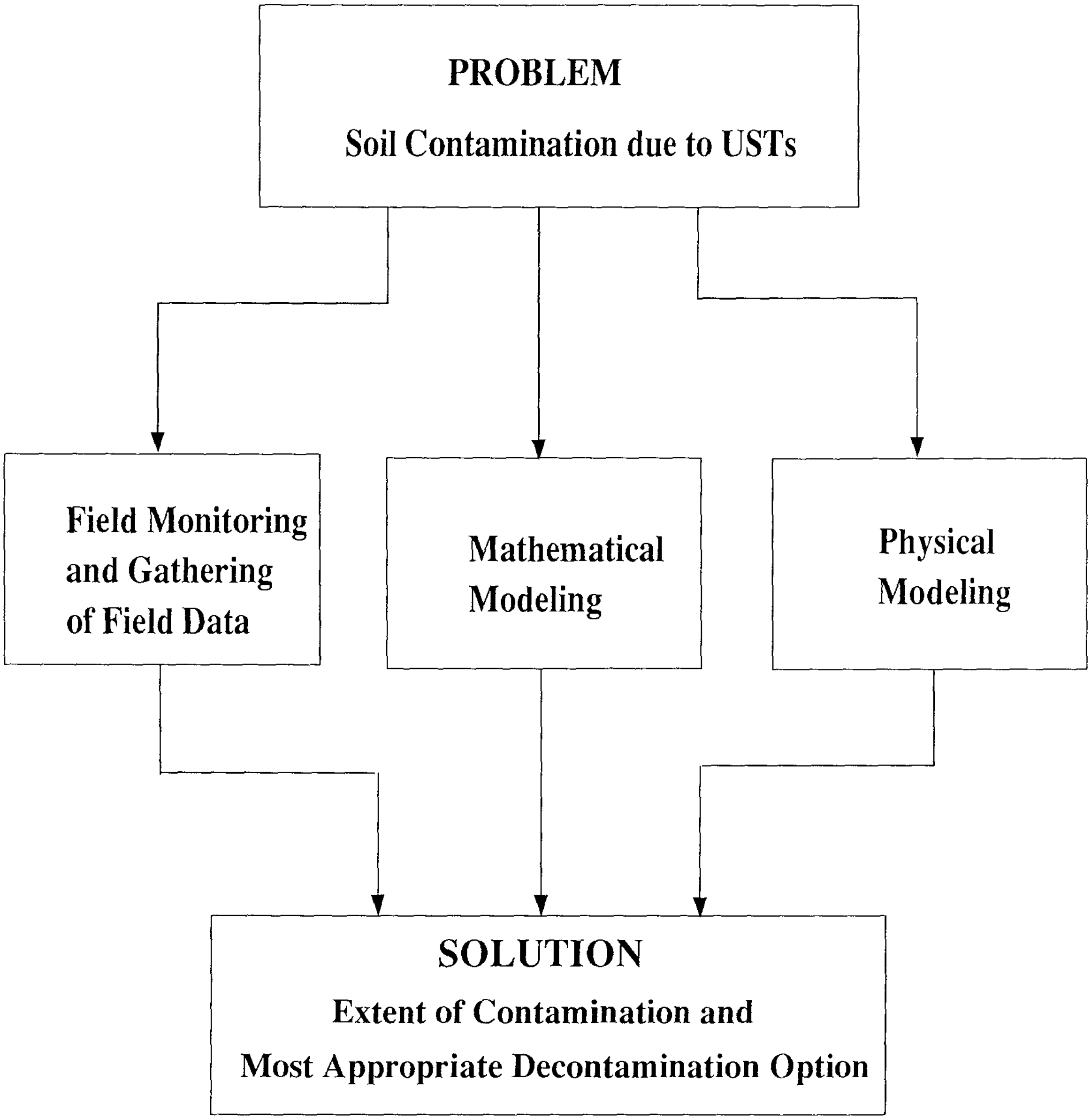


FIGURE 3 Logical Approaches to Clean-up for USTs

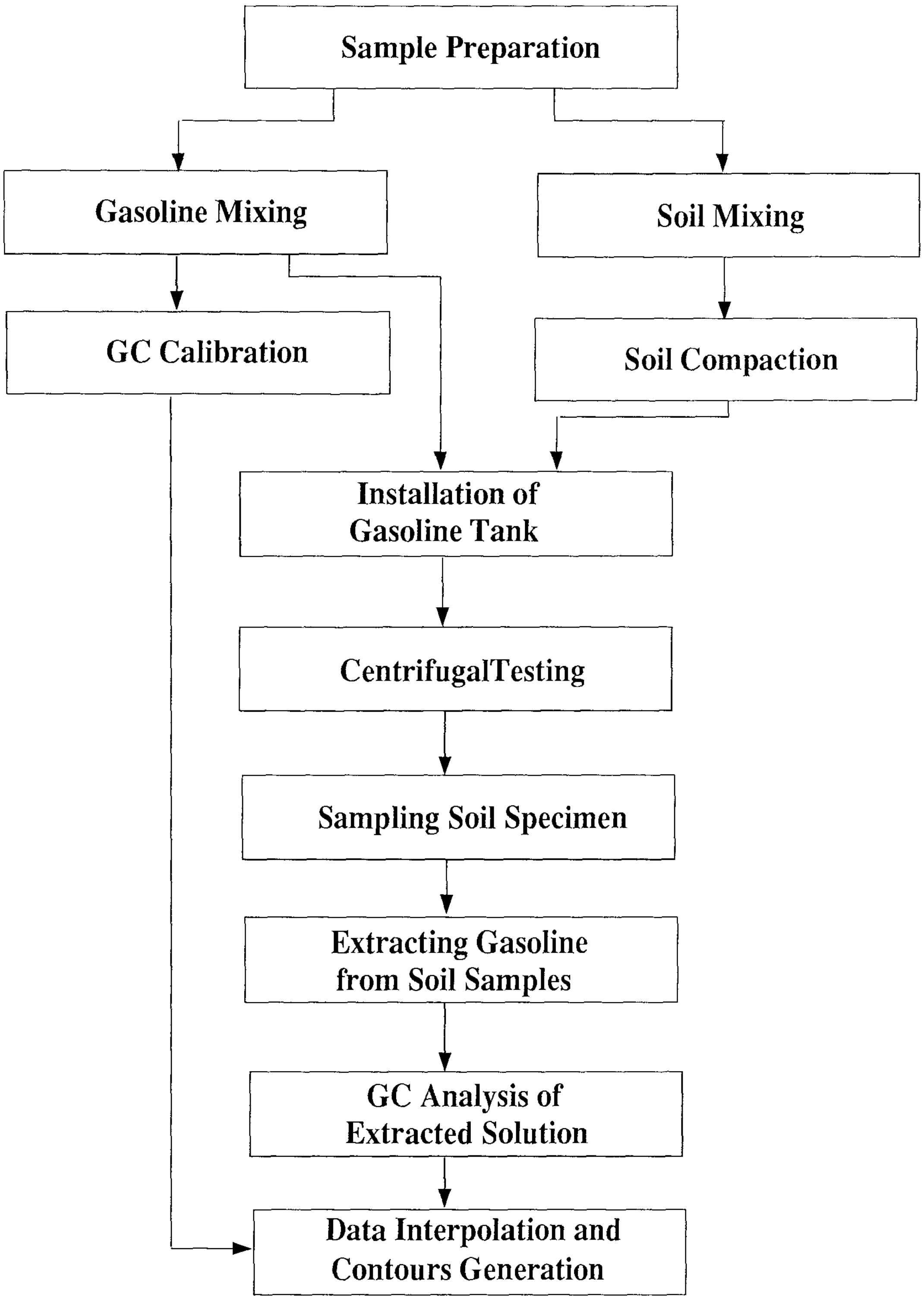


FIGURE 4 Flow Chart of Centrifugal Modeling

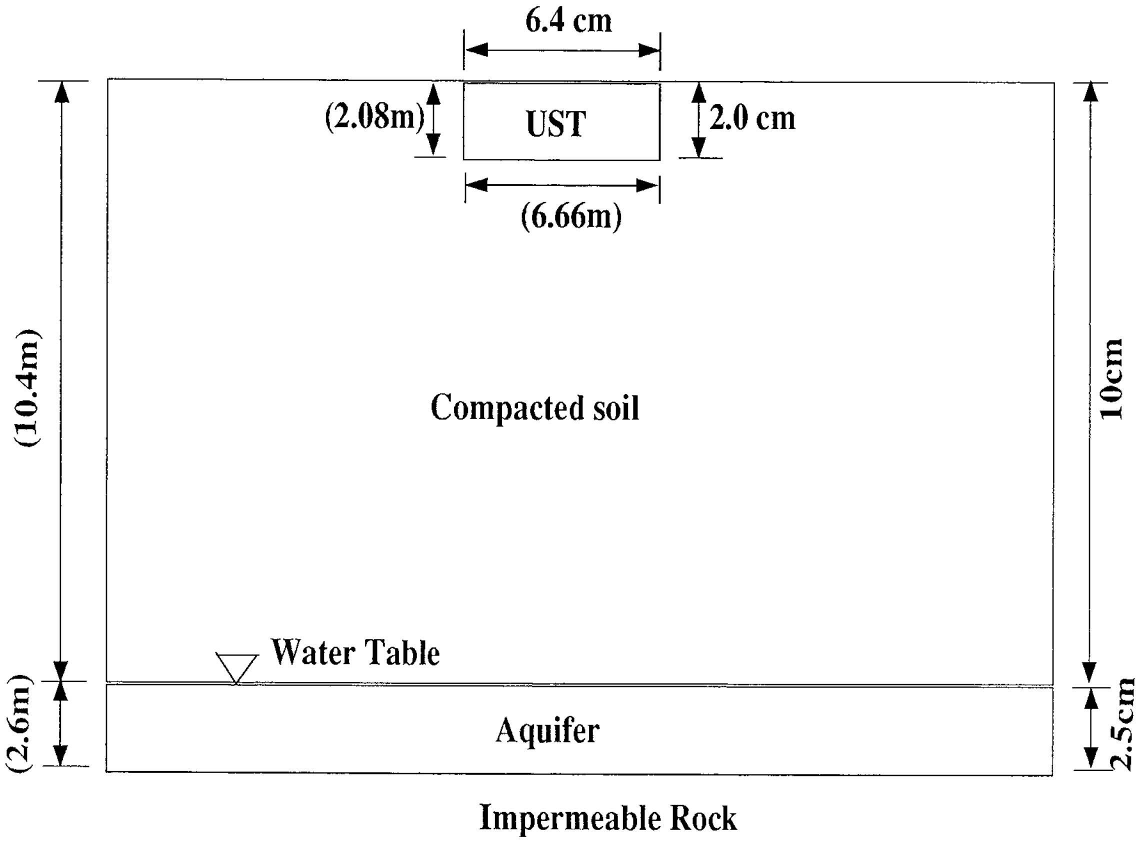


FIGURE 5 Layout of Centrifugal Sample

(scales in brackets are the values simulated)

FIGURE 6 Calibration Curve for Components of Gasoline

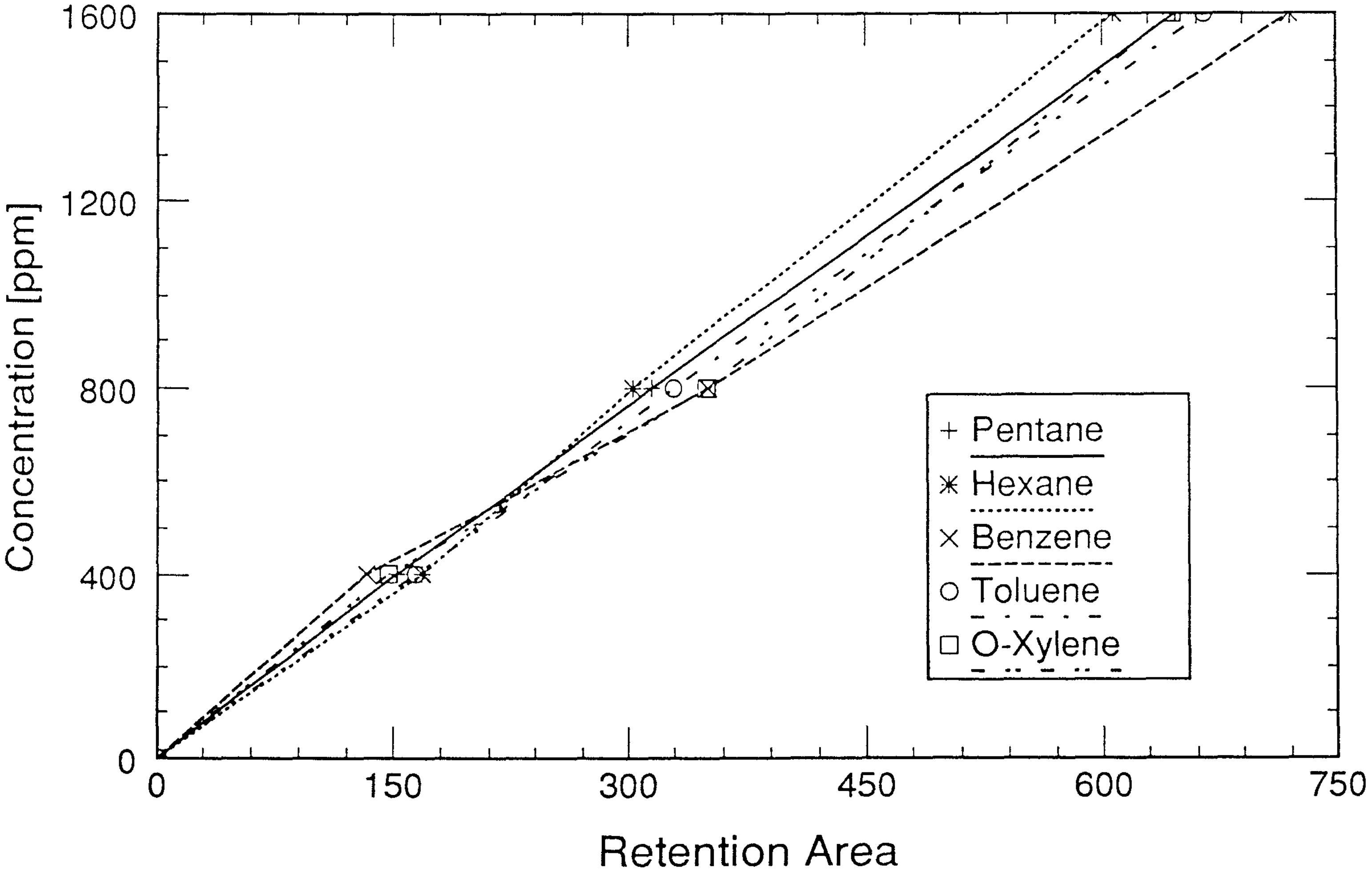
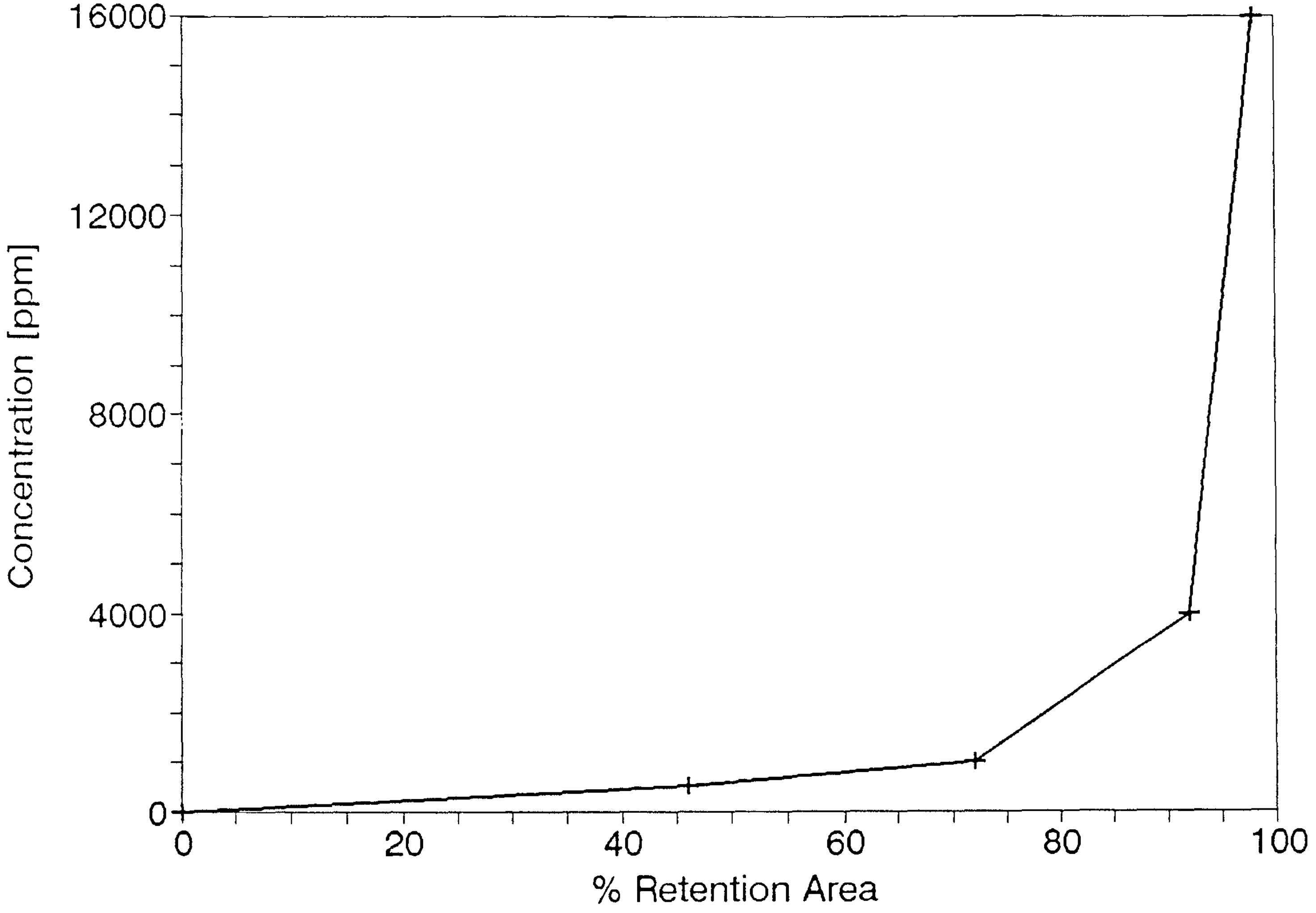


FIGURE 7 Calibration Curve for Sythetic Gasoline



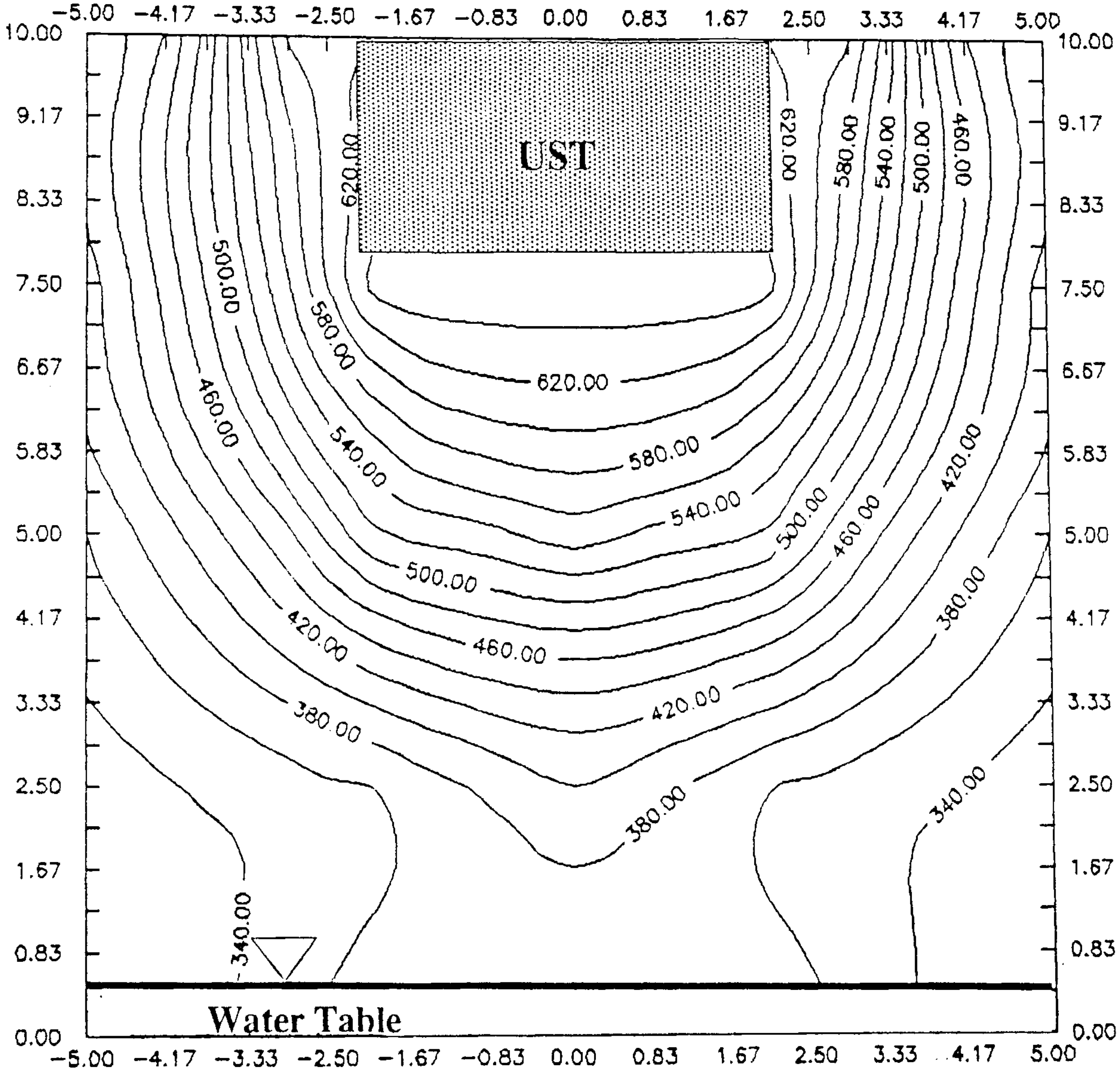


FIGURE 8 Gasoline Concentration Profile for Test 3

[contour lines in ppm, sample layout in cm]

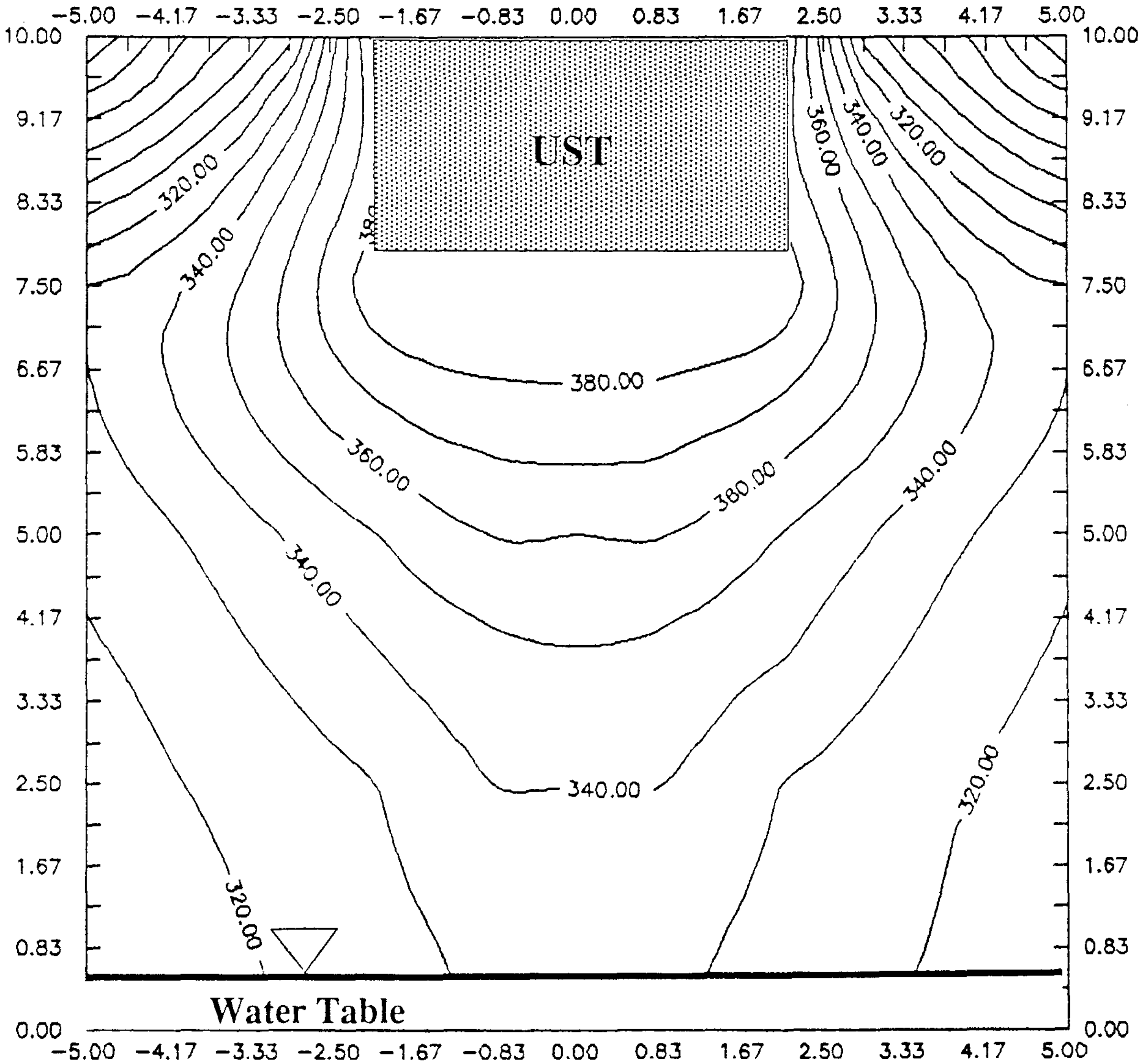


FIGURE 8A Benzene Concentration Profile for Test 3

[contour lines in ppm, sample layout in cm]

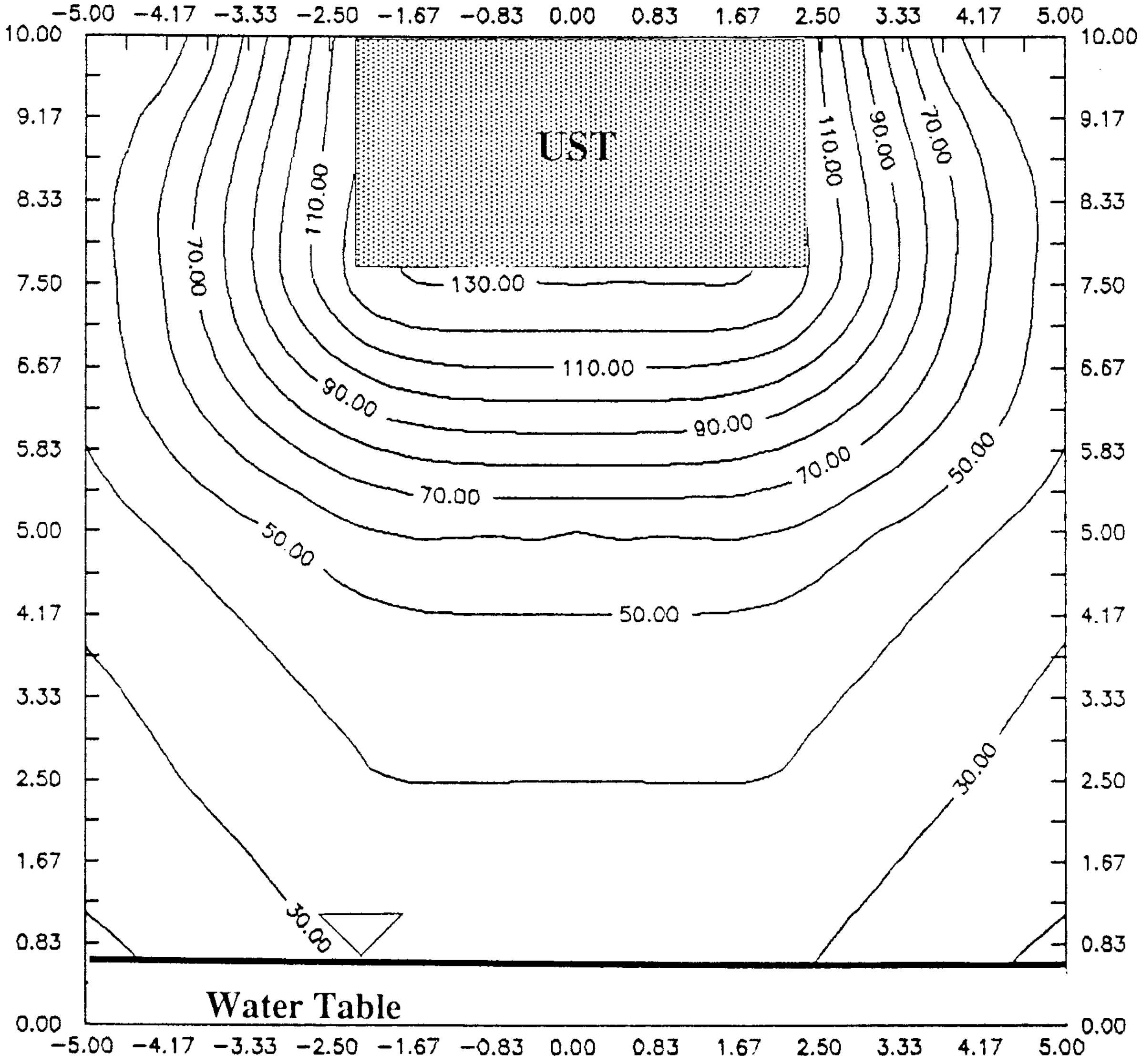


FIGURE 8B Toluene Concentration Profile for Test 3

[contour lines in ppm, sample layout in cm]

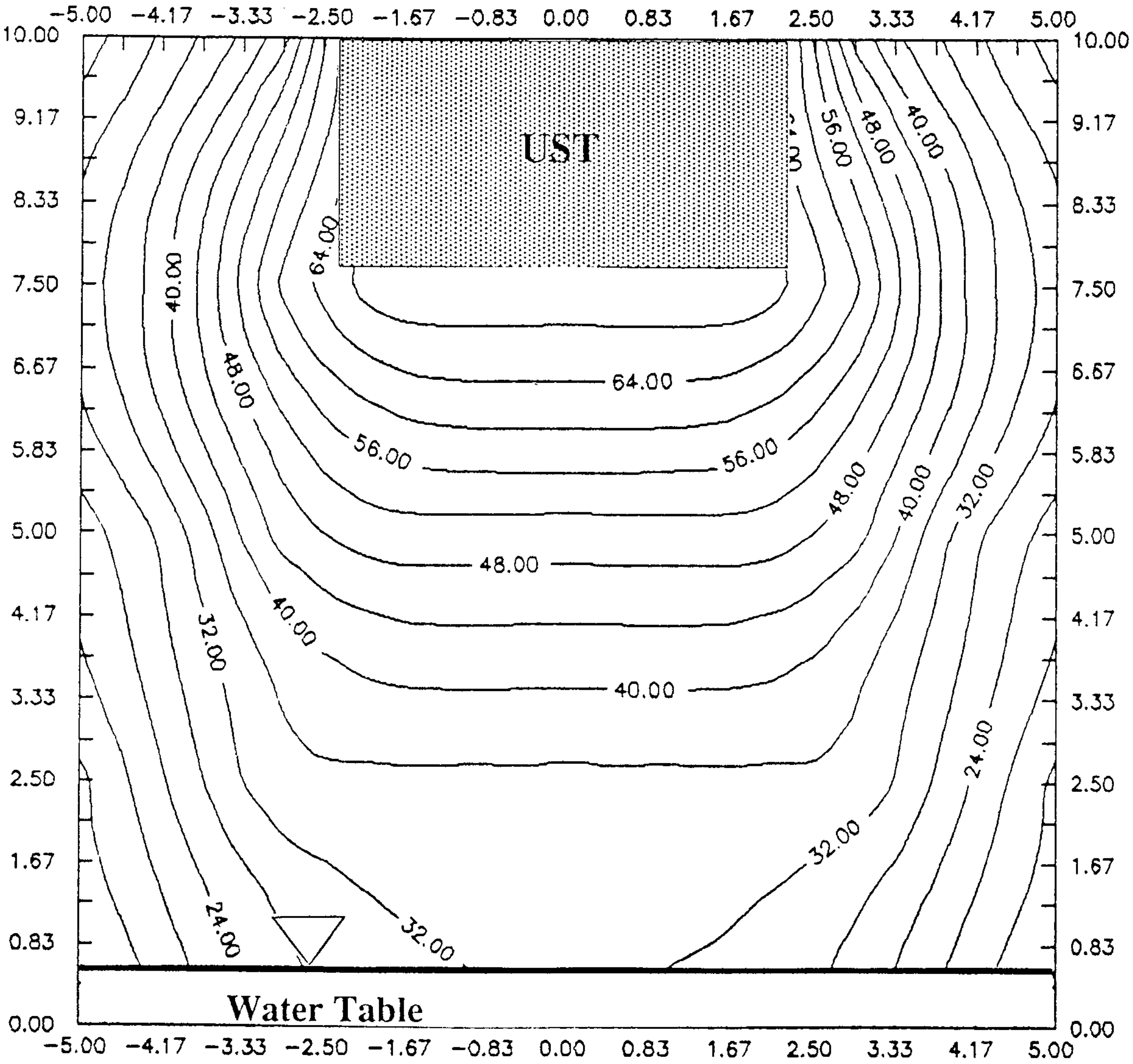


FIGURE 8C O-Xylene Concentration Profile for Test 3

[contour lines in ppm, sample layout in cm]

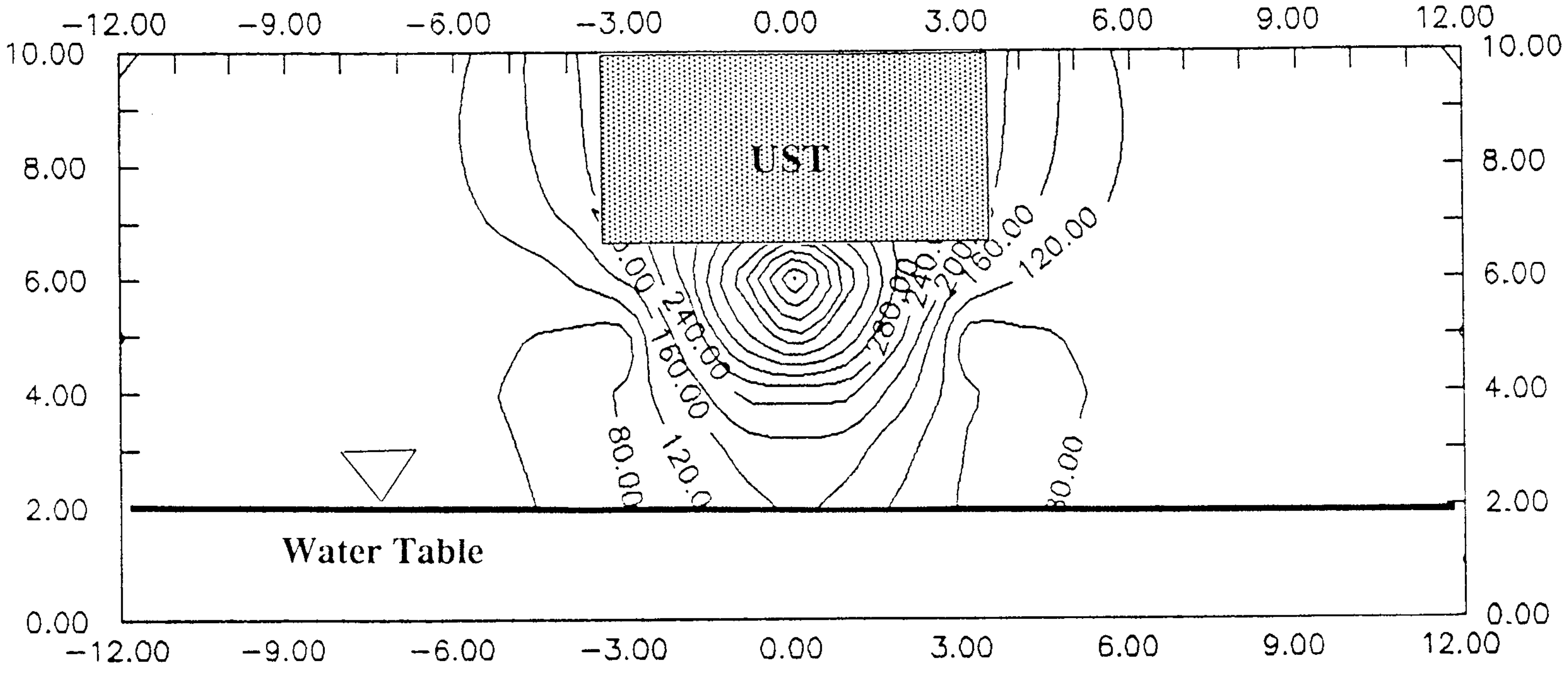


FIGURE 9 Gasoline Concentration Profile for Test 8

[contour lines in ppm, sample layout in cm]

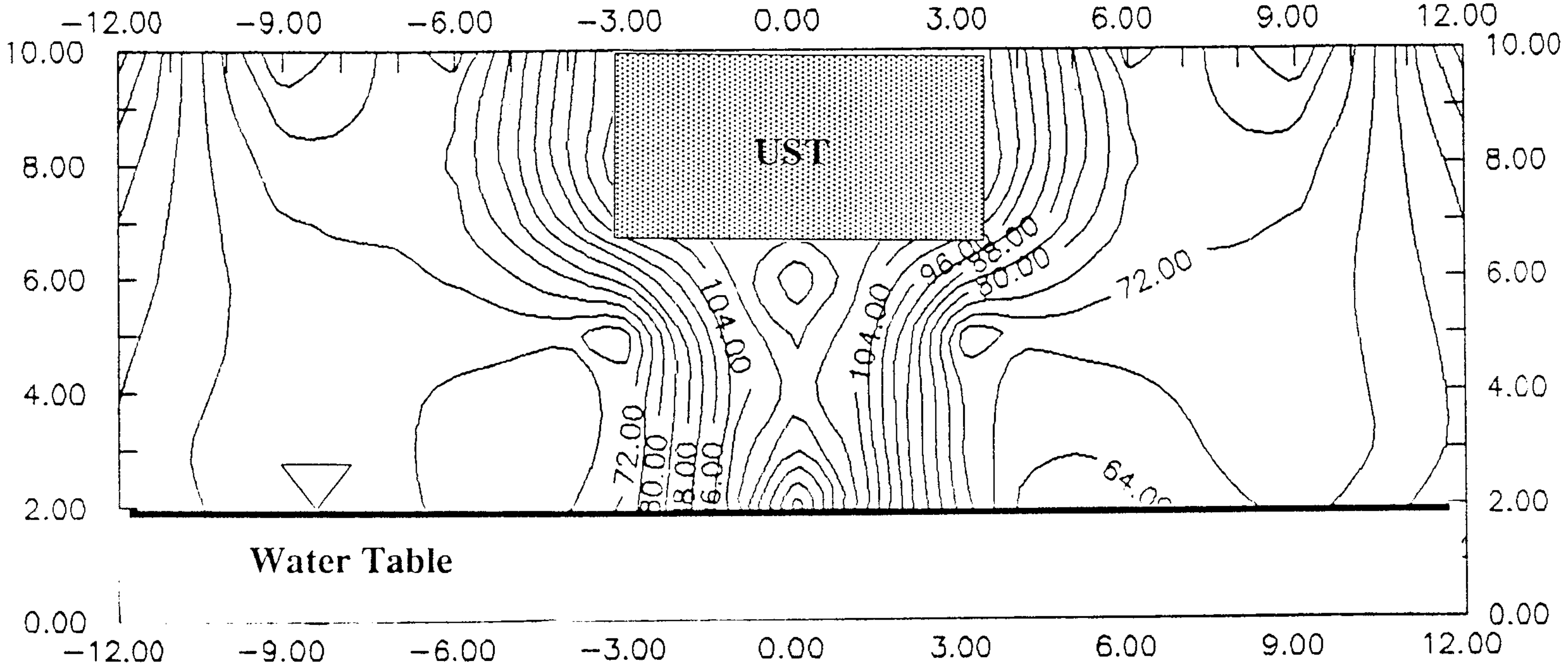


FIGURE 9A Benzene Concentration Profile for Test 8

[contour lines in ppm, sample layout in cm]

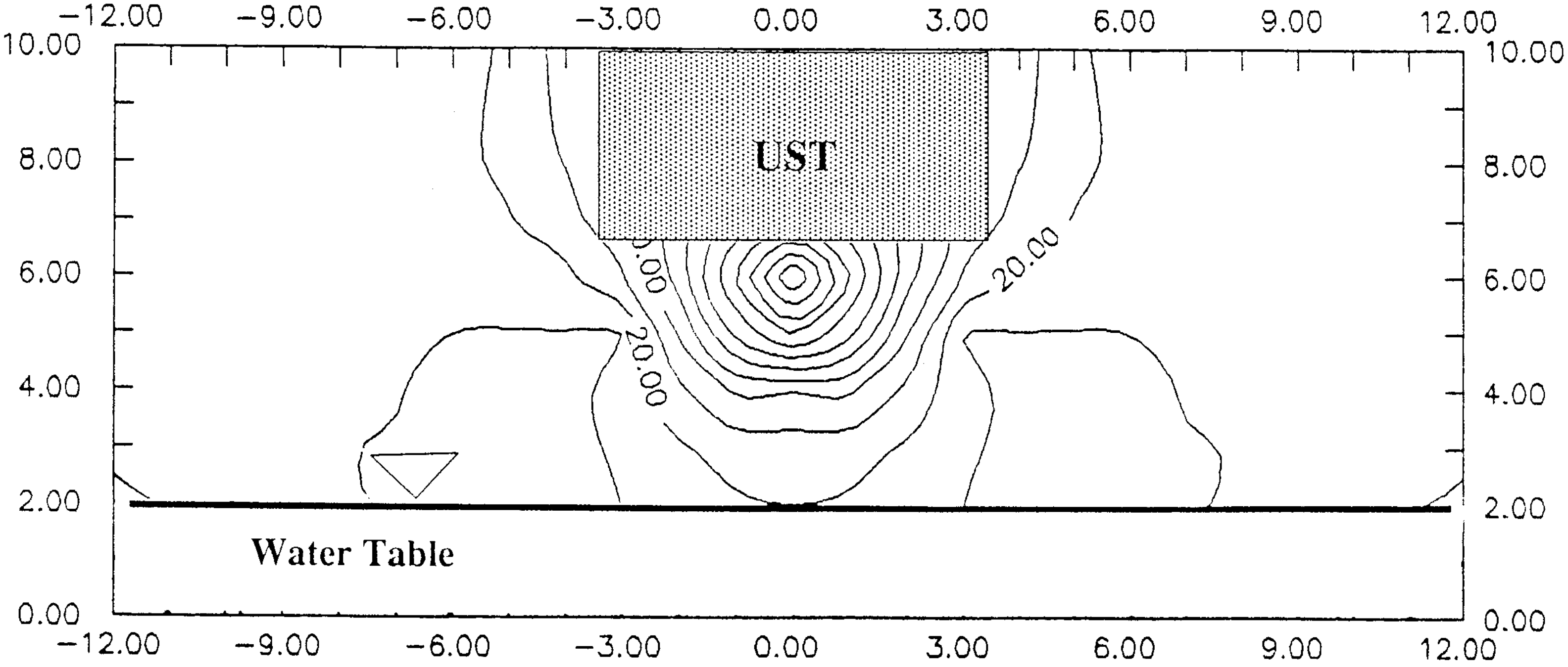


FIGURE 9B Toluene Concentration Profile for Test 8

[contour lines in ppm, sample layout in cm]

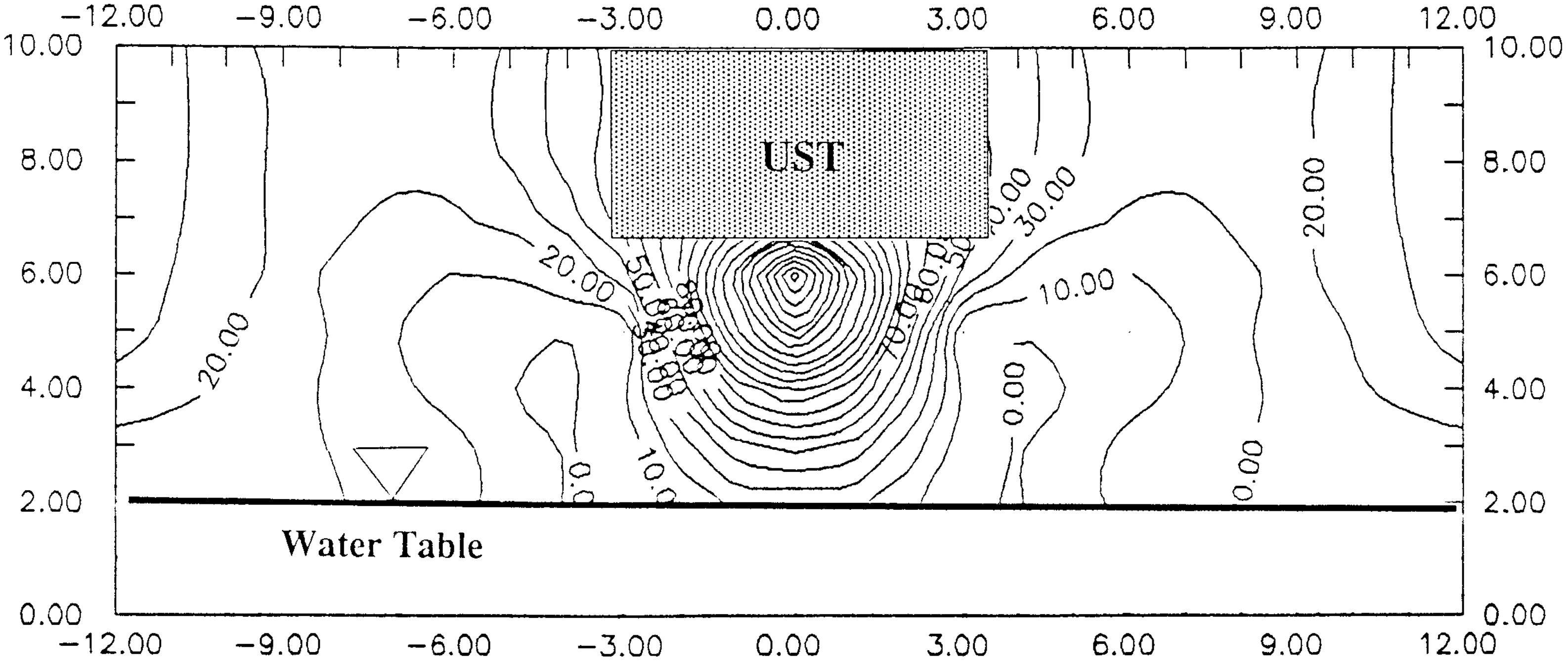


FIGURE 9C O-Xylene Concentration Profile for Test 8

[contour lines in ppm, sample layout in cm]

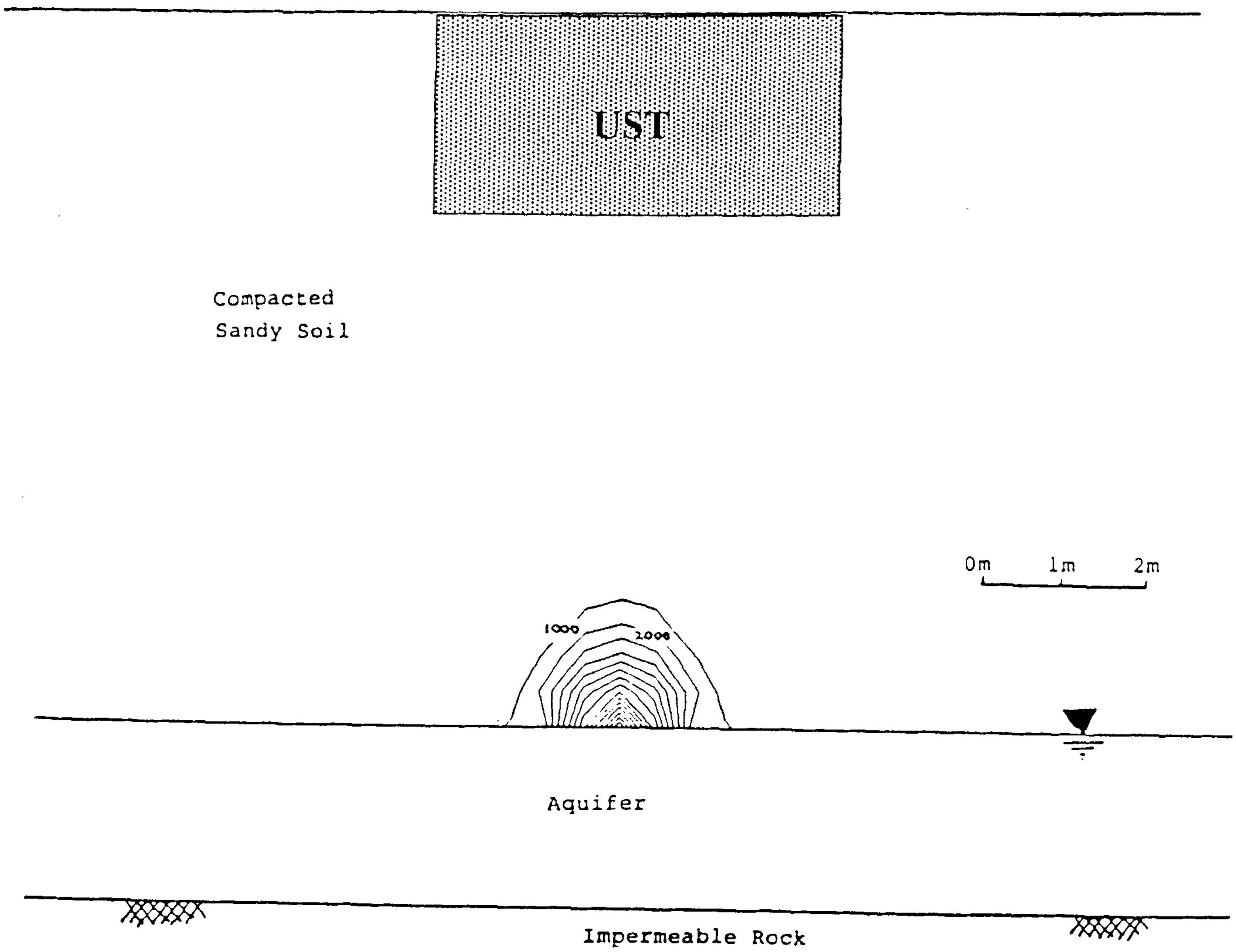


FIGURE 10 Gasoline Concentration Profile for Test 4

[contour lines in ppm, sample layout in cm]

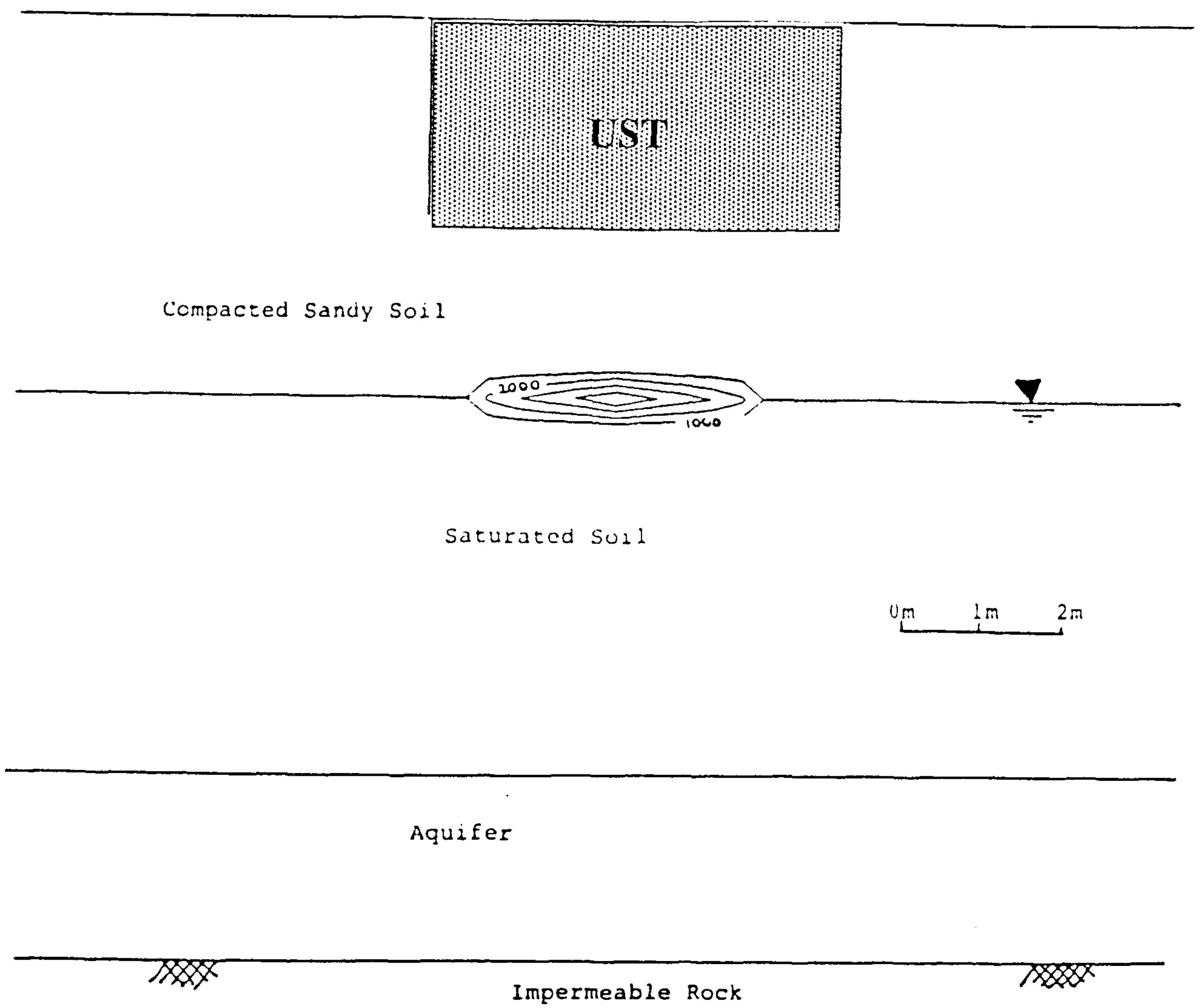


FIGURE 11 Gasoline Concentration Profile for Test 5

[contour lines in ppm, sample layout in cm]

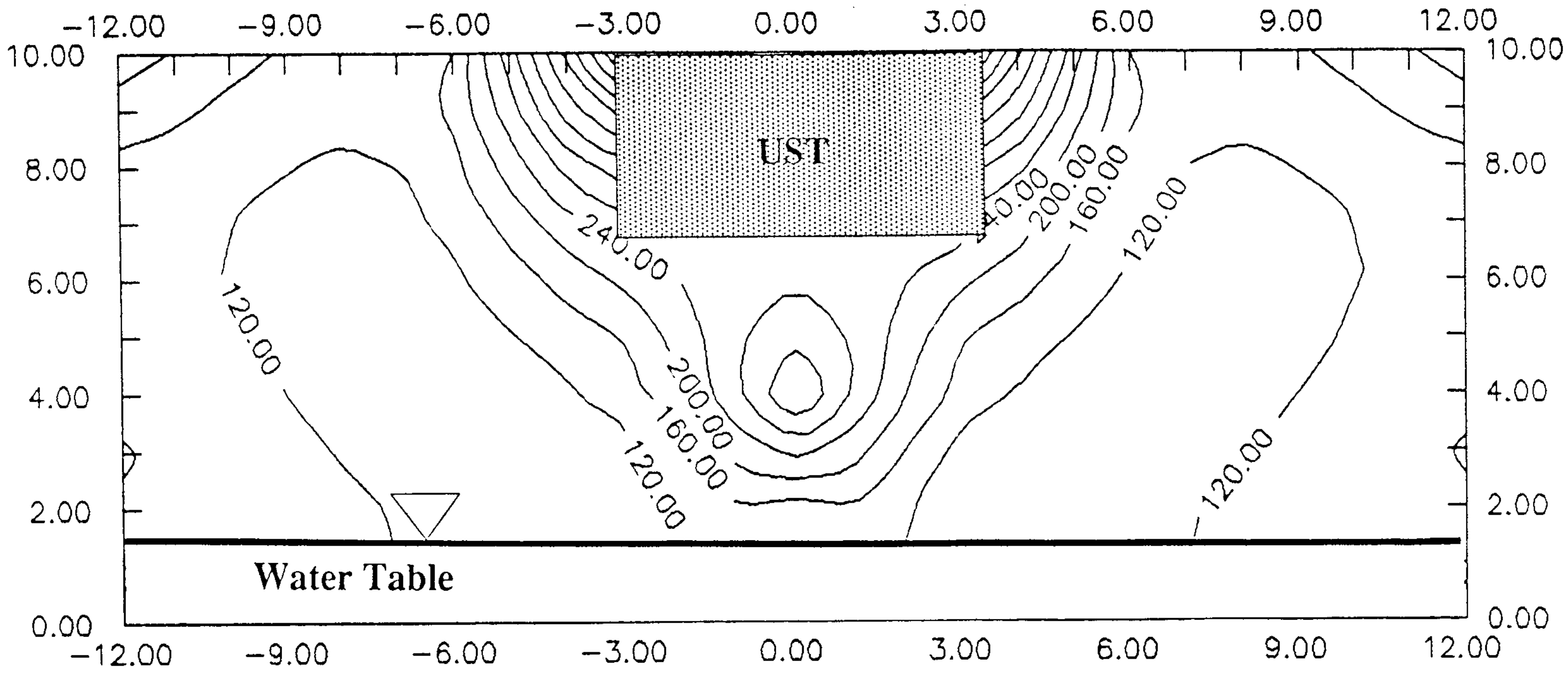


FIGURE 12 Gasoline Concentration Profile for Test 10

[contour lines in ppm, sample layout in cm]

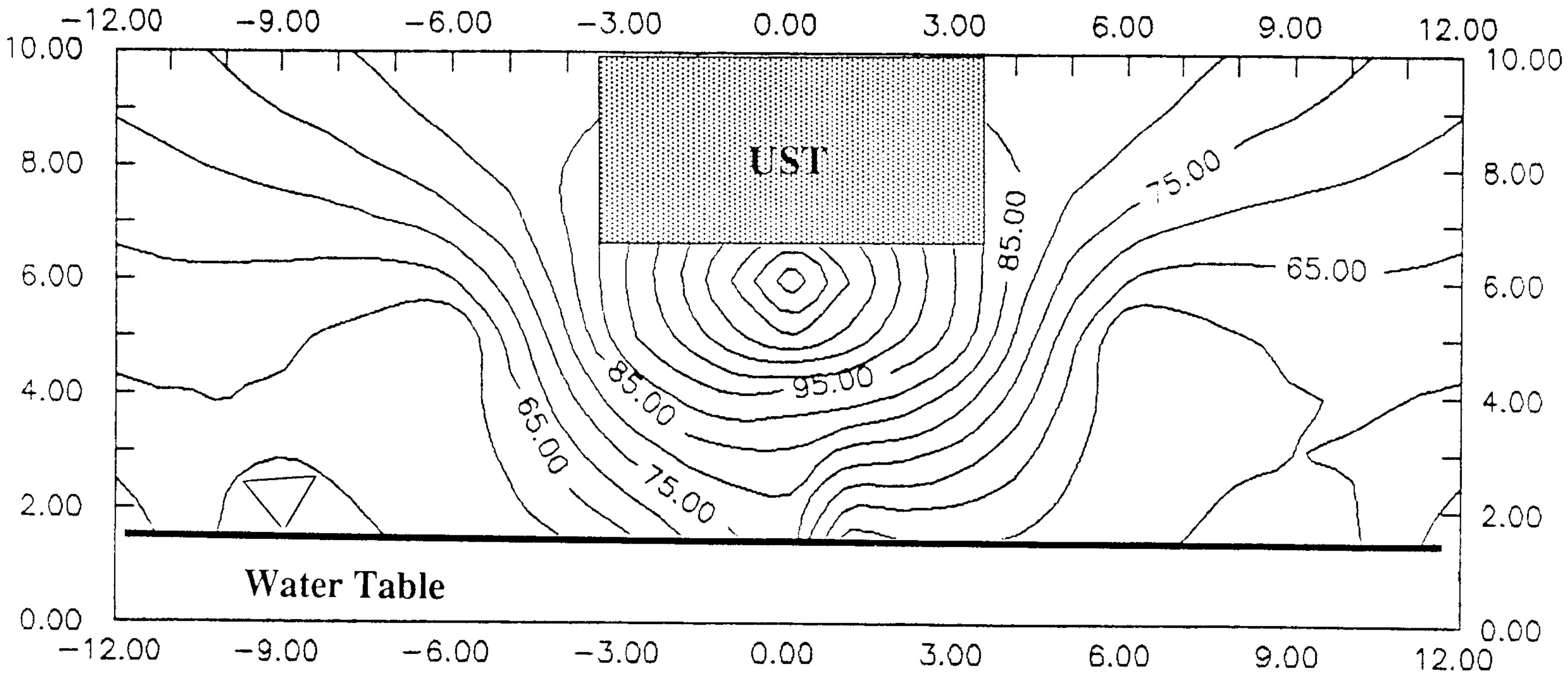


FIGURE 12A Benzene Concentration Profile for Test 10

[contour lines in ppm, sample layout in cm]

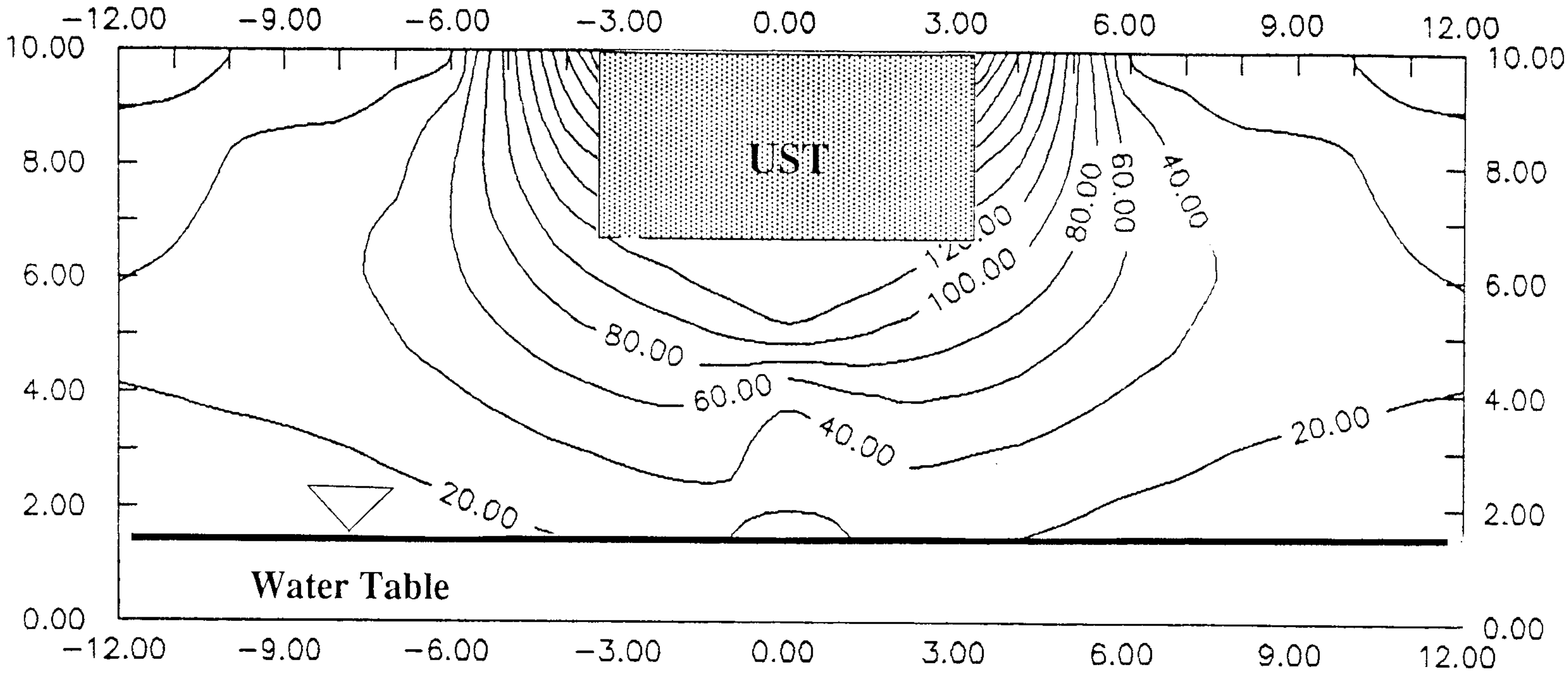


FIGURE 12B Toluene Concentration Profile for Test 10

[contour lines in ppm, sample layout in cm]

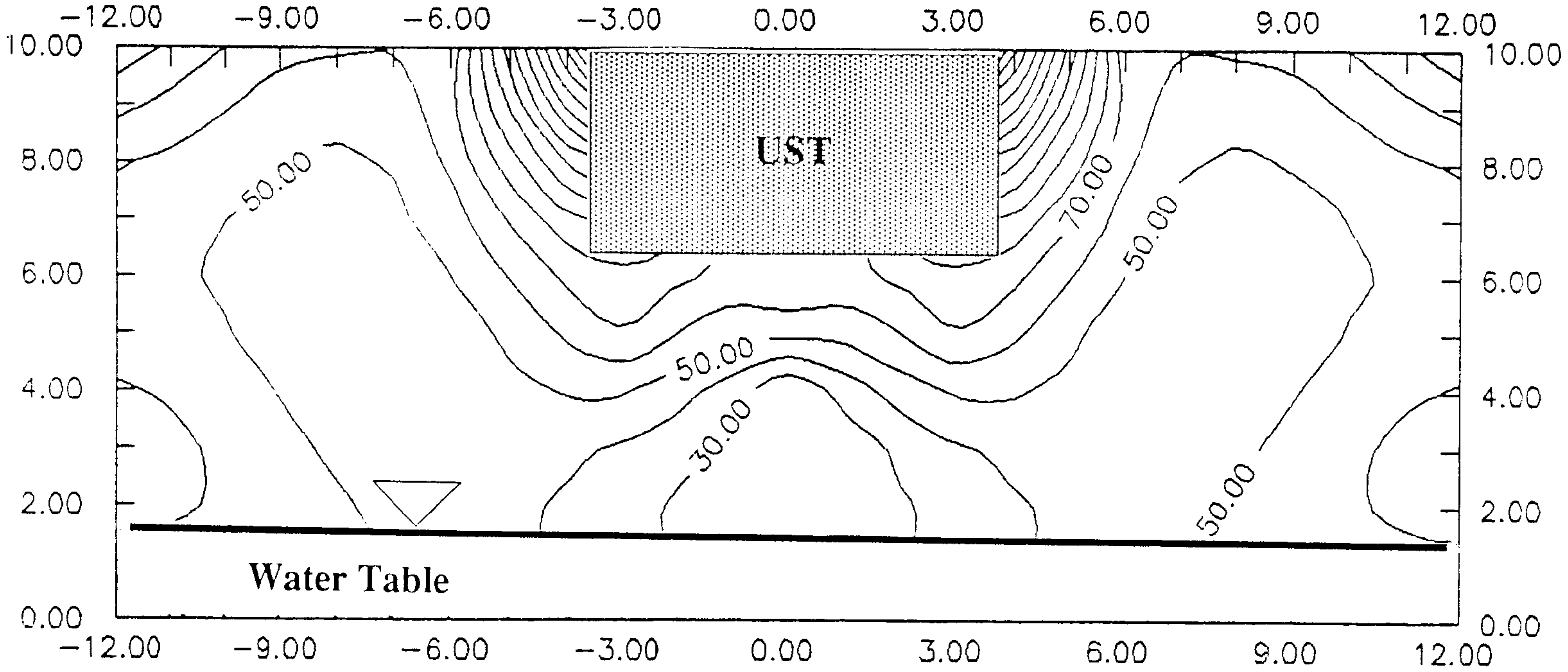


FIGURE 12C O-Xylene Concentration Profile for Test 10

[contour lines in ppm, sample layout in cm]

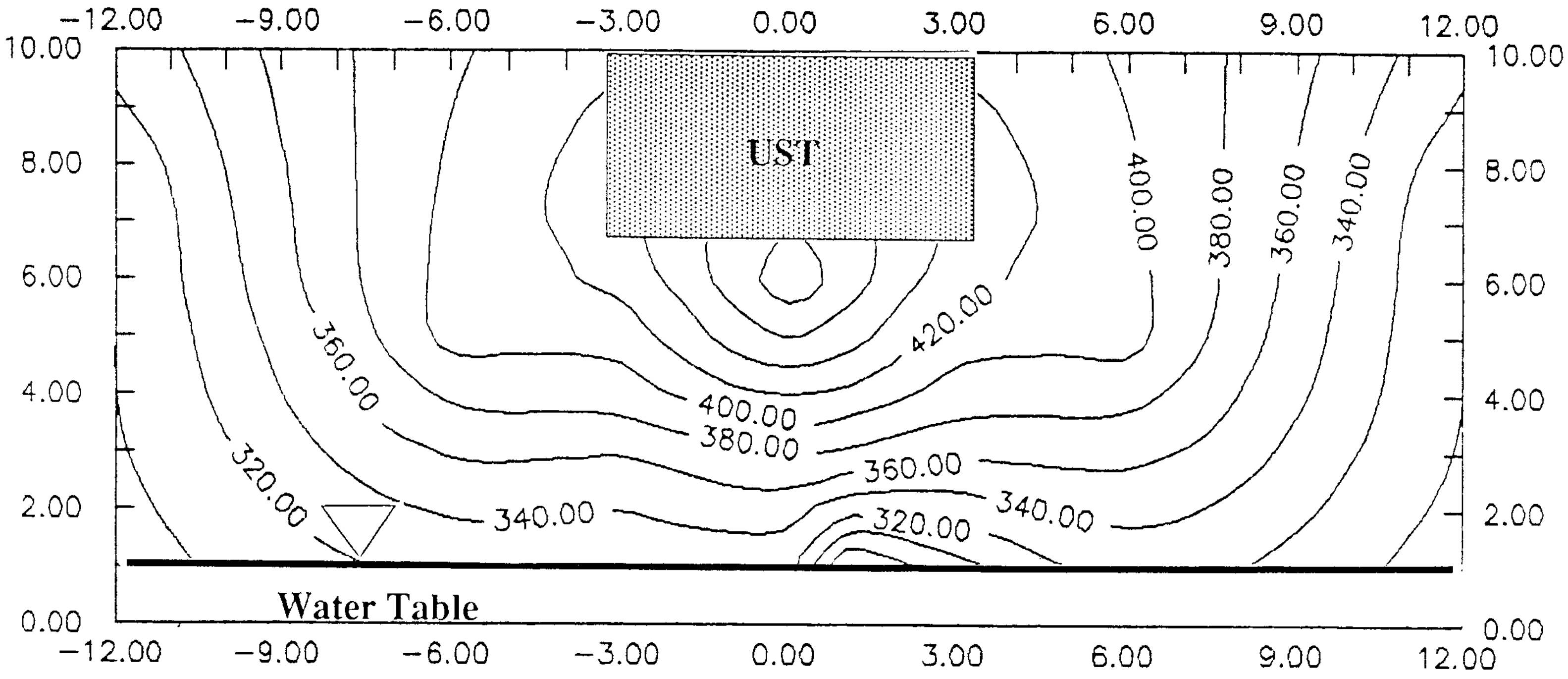


FIGURE 13 Gasoline Concentration Profile for Test 12

[contour lines in ppm, sample layout in cm]

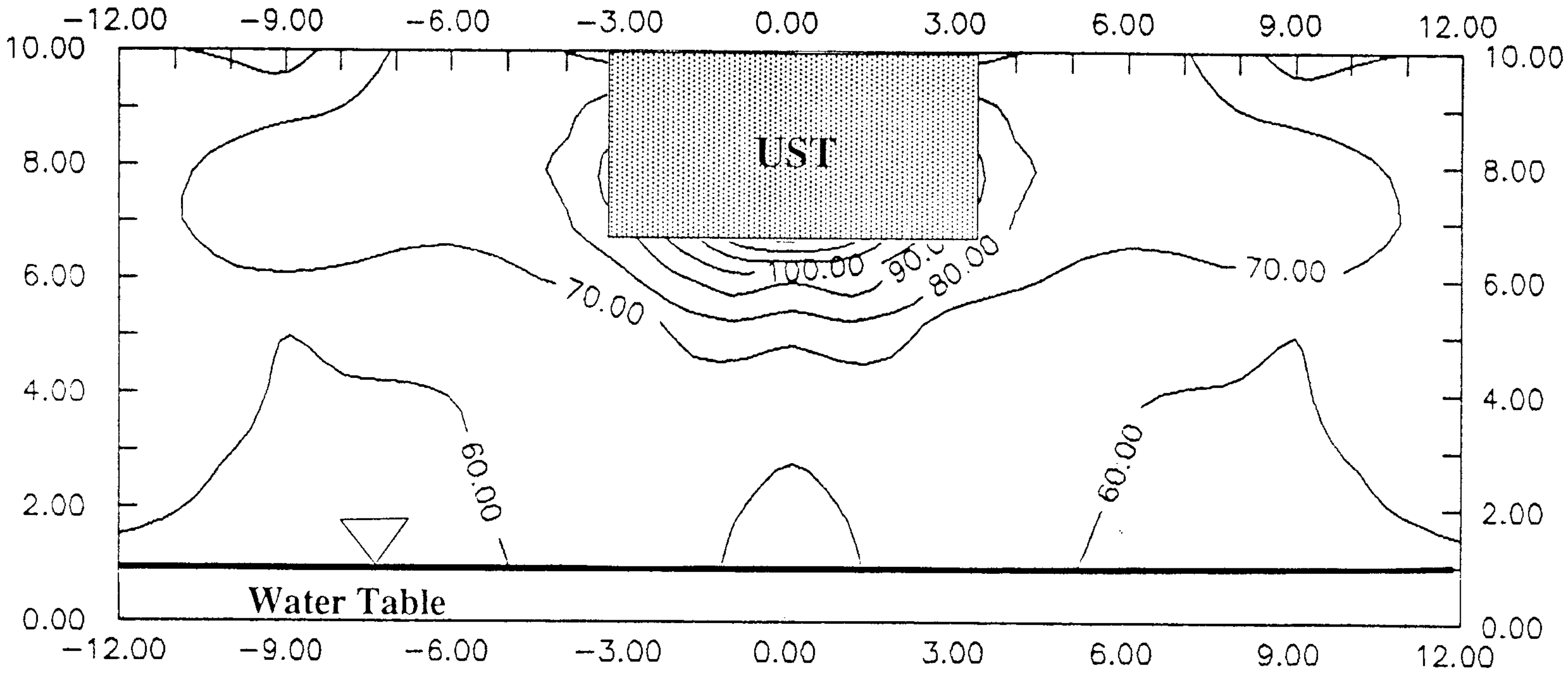


FIGURE 13A Benzene Concentration Profile for Test 12

[contour lines in ppm, sample layout in cm]

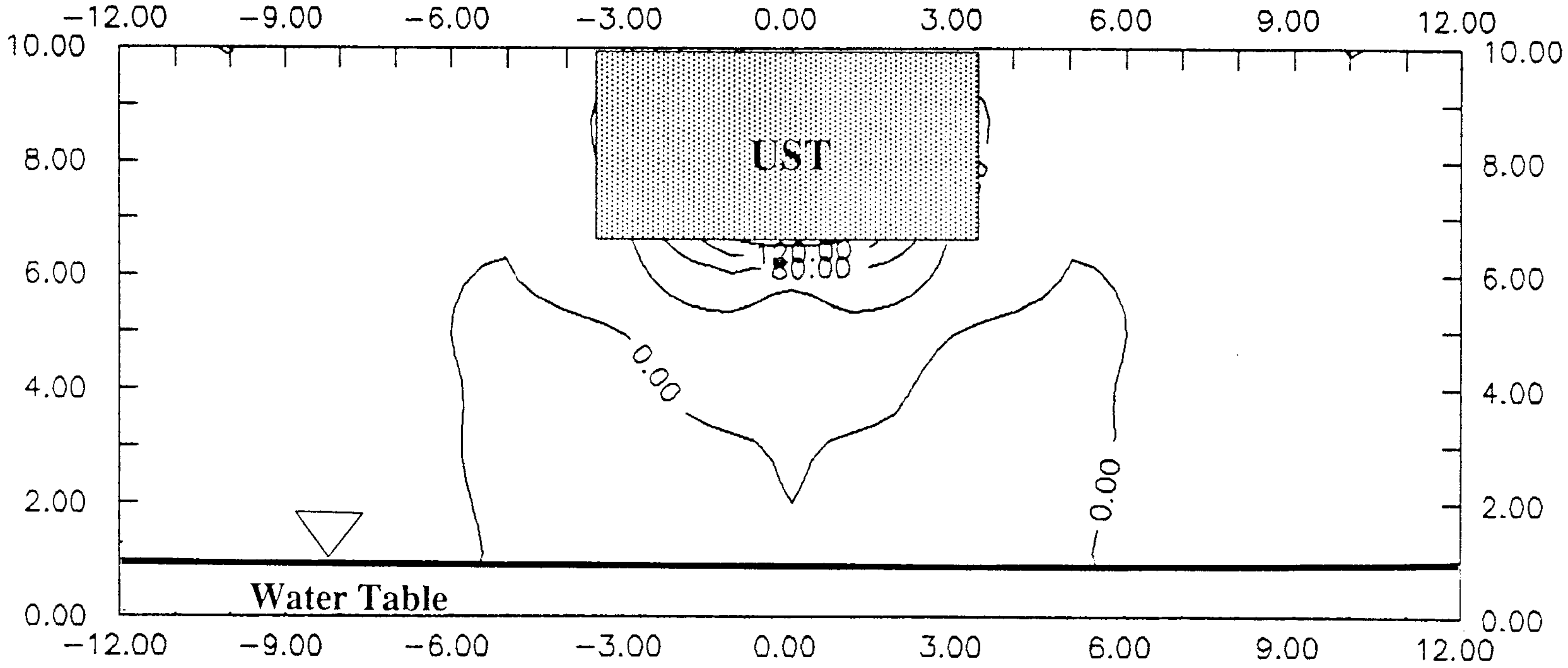


FIGURE 13B Toluene Concentration Profile for Test 12

[contour lines in ppm, sample layout in cm]

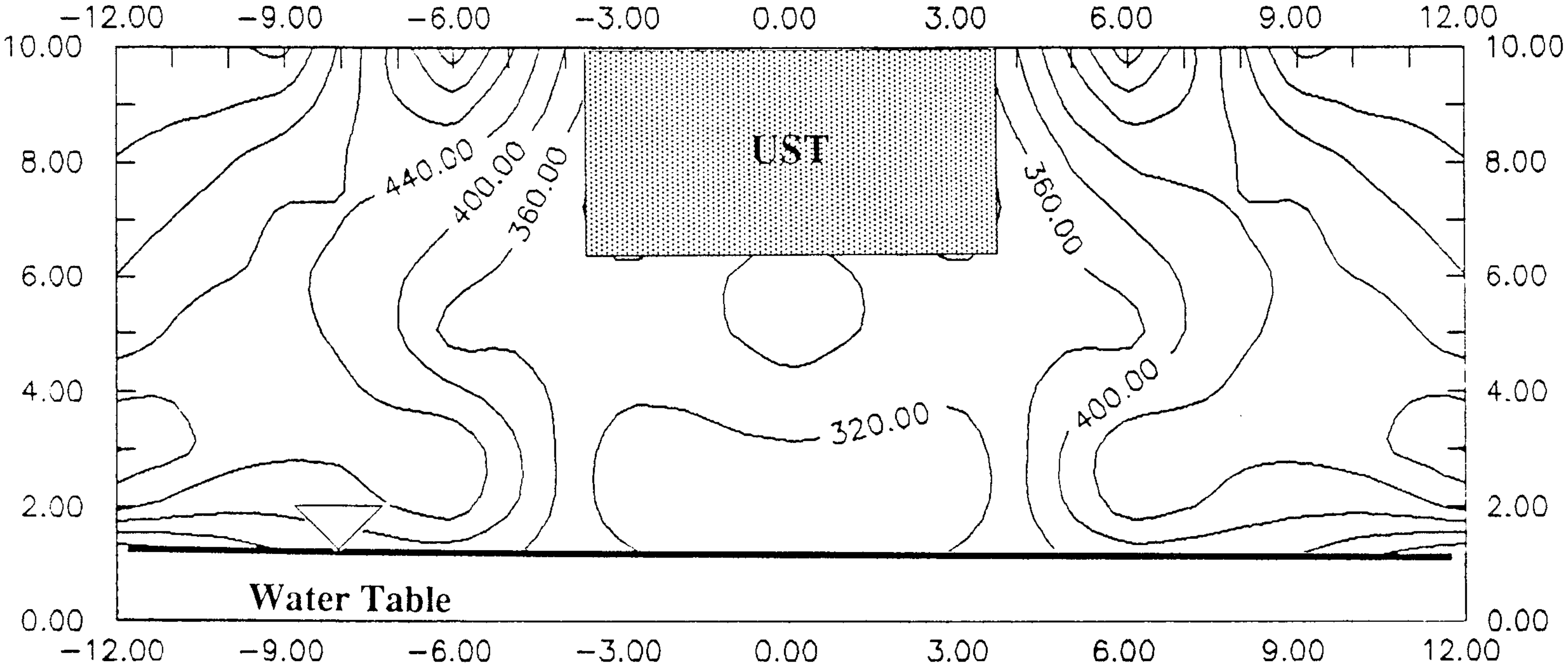


FIGURE 13C O-Xylene Concentration Profile for Test 12

[contour lines in ppm, sample layout in cm]

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