

# Demonstration of Performance Modeling of a Low-Level Waste Shallow-Land Burial Site

A Comparison of Predictive Radionuclide Transport Modeling  
Versus Field Observations at the Nitrate Disposal Pit Site,  
Chalk River Nuclear Laboratories

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Prepared by D. E. Robertson, M. P. Bergeron, D. A. Myers, K. H. Abel, C. W. Thomas/PNL  
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## ABSTRACT

Before a license can be obtained to construct a facility for the shallow-land burial of low-level wastes, the U. S. Nuclear Regulatory Commission must be assured that the facility will meet both performance objectives and prescriptive requirements set forth in 10CFR61, "Licensing Requirements for Land Disposal of Radioactive Waste." Subpart D, Section 61.50(a)(2) of 10CFR61 states that a "disposal site shall be capable of being characterized, modeled, analyzed and monitored." In order to test the concept of "site modelability," a 30-year old low-level radioactive waste disposal site at Chalk River Nuclear Laboratories (CRNL), Canada, was used as a field location for evaluating the process of site characterization and the subsequent modeling prediction of radionuclide transport from the site by groundwater. The radionuclide source term was a limestone-lined pit (since covered with soil) which in 1953 to 1954 received approximately 3800 liters of aqueous waste containing 1000 to 1500 curies of aged, mixed fission products, including 700 to 1000 curies of  $^{90}\text{Sr}$  and 200 to 300 curies of  $^{137}\text{Cs}$ . This evaluation was performed by comparing the actual measured radionuclide migration with predicted migration estimated from hydrologic/radionuclide transport models. This comparison has provided valuable insights into the applicability of transport modeling, and in determining what level of effort is needed in site characterization at locations similar to the Nitrate Disposal Pit to provide the desired degree of predictive capabilities.

## SUMMARY

Before a license can be obtained to construct a facility for the shallow-land burial of low-level waste, the U. S. Nuclear Regulatory Commission must be assured that the facility will meet both performance objectives and prescriptive requirements set forth in 10CFR61, "Licensing Requirements for Land Disposal of Radioactive Waste." To test the concept of "site modelability," a 30-year-old low-level radioactive waste disposal site at Chalk River Nuclear Laboratories (CRNL), Canada, was used as a field location for evaluating the process of site characterization and the subsequent modeling predictions of radionuclide transport from the site by groundwater. The radionuclide source term was a limestone-lined pit (since covered with soil) which in 1953 to 1954 received approximately 3800 liters of aqueous waste containing 1000 to 1500 curies of aged, mixed fission products. This evaluation was performed by comparing the actual measured radionuclide migration with predicted migration estimated from hydrologic/radionuclide transport models. This comparison provides insights into the applicability and accuracy of transport modeling and in determining what level of effort is needed in site characterization to provide the desired degree of predictive capabilities.

Extensive field measurements, principally conducted by CRNL and augmented by Pacific Northwest Laboratory (PNL) personnel, provided a well-defined map of the actual radionuclide migration that has occurred during the past 30 years at this site. Strontium-90 is the only long-lived radionuclide which has significantly migrated from the disposal source and presently remains in the soil and groundwater downgradient from the source. The actual areal and vertical distribution of  $^{90}\text{Sr}$  has been well characterized by the field measurements, and actual concentration plumes have been constructed. The  $^{90}\text{Sr}$  has migrated about 350-m downgradient from the disposal pit in a relatively narrow (50 to 100-m wide) plume contained within a shallow aquifer system. Cesium-137, the other long-lived fission product which was relatively abundant in the disposal waste, has not migrated more than a few meters from the disposal pit.

The predictive radionuclide transport modeling was independently conducted in two parts. The first part utilized only a limited amount of existing hydrogeologic data. The initial modeling using the limited data base was meant to simulate the data-gathering process that a prospective licensee would utilize during a low-level waste disposal site characterization effort. For the limited data exercise, 16 borehole/monitoring wells which were part of a much larger CRNL monitoring network for this site were utilized in the site characterization and transport modeling. The second part of the study used the entire, extensive CRNL data base existing for this site, which included applicable data from over 100 monitoring wells containing multi-level piezometers. The purpose of the detailed data exercise was to determine if additional data gathering could substantially improve the accuracy and reliability of the transport modeling.

Because of uncertainties associated with the radionuclide source term, several radionuclide release/migration scenarios were used in modeling the

groundwater transport of radionuclides from the source. The predictive transport modeling for  $^{90}\text{Sr}$  that used a retardation factor of 25 yielded downgradient migration rates that were in good agreement with the actual observed migration. For example, the 100 pCi/l  $^{90}\text{Sr}$ -concentration isopleths downgradient from the disposal pit for the observed and predicted values 30 years after disposal were about 330 m and 320 m, respectively. However, major discrepancies became apparent when the internal distribution of  $^{90}\text{Sr}$  concentrations and the lateral spreading of the plumes were compared. The 1000 pCi/l and 10,000 pCi/l concentration isopleths for the observed and predicted  $^{90}\text{Sr}$  migration downgradient from the disposal pit were separated by about 50 to 70 m. The predicted plumes conservatively estimated spreading of the  $^{90}\text{Sr}$  plume which was approximately triple the maximum width of the observed plume. The predicted spreading is mainly a function of numerical dispersion caused by the coarse grid space currently being used in the model. The amount of spreading predicted by the numerical model could be reduced by refining the finite element mesh in a direction transverse to the principal direction of groundwater movement. The narrow width of the observed plume is mainly a function of the sorting and texture of the surficial sands within which the transport occurs. The rather uniform particle size and fine-grained texture of these sands would have a tendency to minimize the amount of hydrodynamic dispersion that would otherwise occur in a more heterogeneous material.

The detailed data modeling using the entire, extensive CRNL data base did not substantially improve the results of the predictive modeling. However, this similarity in results may have been more a function of the two-dimensional modeling approach or the fact that the bulk of additional well data was closely spaced in and around the observed plume. Thus, for this relatively simple site and modeling approach the minimal site characterization effort (Part 1) involving 16 selected test boreholes/monitoring wells was sufficient to provide a reasonable assessment of the radionuclide transport. A more detailed and costly characterization effort would not have significantly added to the reliability of the predictive modeling using this two-dimensional approach. A more detailed three-dimensional modeling effort would have undoubtedly required a larger array of characterization boreholes.

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## 1.0 INTRODUCTION

Before a license can be obtained to construct a facility for the shallow-land burial of low-level waste, the U. S. Nuclear Regulatory Commission must be assured that the facility will meet both performance objectives and prescriptive requirements set forth in 10CFR61, "Licensing Requirements for Land Disposal of Radioactive Waste." Section 61.5, Subpart D, of 10CFR61 states that any future low-level waste shallow-land burial facility ".... shall be capable of being characterized, modeled, analyzed and monitored..." To assure compliance with these objectives and requirements, applicants will most probably utilize radionuclide transport models to predict the movement of radionuclides in groundwater from a disposal facility.

However, recent studies<sup>(1,2,3,4,5)</sup> at Low-Level Waste (LLW) shallow-land burial facilities and other slightly contaminated sites have shown discrepancies between predicted versus actual radionuclide migration rates in the ambient groundwater. Because of these discrepancies, there is a general understanding within the scientific community that site performance modeling may need further refinement to more accurately predict radionuclide movement from a potential disposal site. Therefore, predictive transport models need to be evaluated under actual field conditions to assess their accuracy and identify the weak links in the modeling process.

One of the ways to test the concept of site "modelability" is to compare predicted radionuclide movement using hydrogeochemical modeling with actual observed radionuclide migration at field sites where radionuclides have been in the ground for many years. Such comparisons will yield insights as to the reliability of models which an applicant might reasonably use to predict LLW disposal site performance at proposed sites.

At the Chalk River Nuclear Laboratories (CRNL), Ontario, Canada, a number of low-level waste shallow-land burial facilities have been in existence for about 25 to 30 years (see Figure 1.1). These sites are proving useful for testing the concept of site "modelability." A cooperative research program was established between PNL and CRNL, and two disposal sites having slightly contaminated groundwater plumes were selected for potential study. This report describes the results of a comparison of predictive radionuclide transport modeling versus field measurements and plume mapping for the Nitrate Disposal Pit site, a low-level disposal facility which received liquid wastes containing approximately 1100 curies of mixed fission products during 1953 to 1954.

### 1.1 PROJECT PLAN AND OBJECTIVES

The Nitrate Disposal Pit site at CRNL was selected as the first field site for testing the concept of site modelability. This site was chosen because of its reasonably well-characterized source term and well-defined <sup>90</sup>Sr plume which has developed in the groundwater over the past 30 years. In addition, this location possesses many of the favorable site characteristics of a Part 61 shallow-land disposal facility in that it is well drained, above the water table, free of active tectonic processes, and capable of being charac-

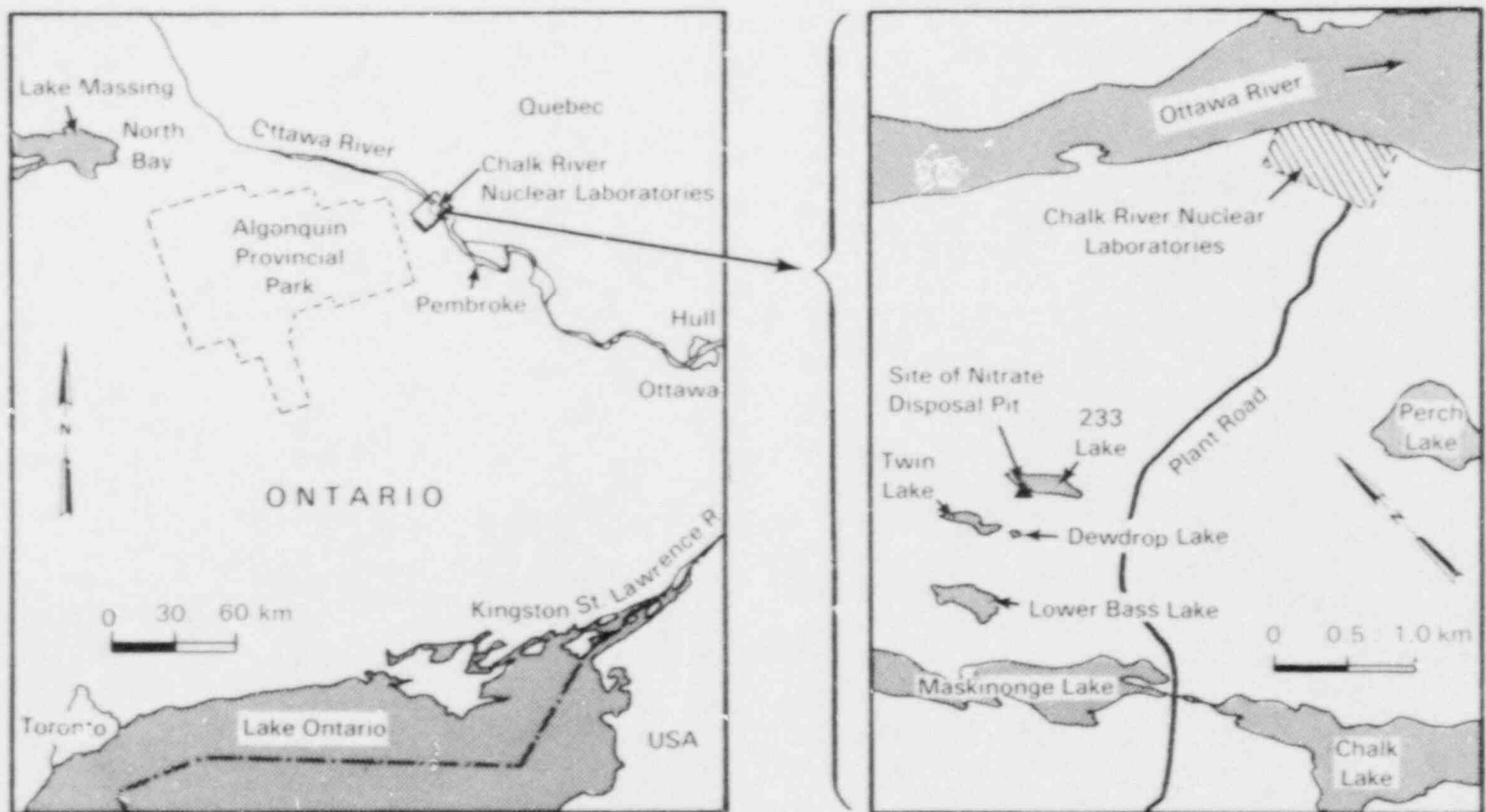


FIGURE 1.1. Location of the Chalk River Nuclear Laboratories and the Nitrate Disposal Pit

terized, modeled, analyzed, and monitored. It should be stressed, however, that this does not assess the overall suitability of this site as a low-level waste shallow-land burial facility, but utilizes it as a favorable field site for testing the modelability concept.

The objectives of this study are to address the regulatory requirement that a site be "modelable," and to use the Nitrate Disposal Pit site as a field test of the reliability of the radionuclide transport modeling.

The project plan is to approach this site as though it were a prospective burial site to be licensed under the requirements of 10CFR61. Under the assumption that this was an undeveloped site and that very little technical information was available from this location, a site characterization plan has been developed which details the geologic/hydrologic/geochemical measurements and analyses which are necessary for predictive modeling of the radionuclide transport in the groundwater from the disposal site (see Appendix A). Using this plan as a guide, a "pre-operational" site performance assessment involving hydrogeochemical modeling has been conducted at the Nitrate Disposal Pit site to predict the temporal movement of radionuclides in the groundwater. The predicted movement is then compared with the actual radionuclide migration which has occurred over the past 30 years to assess the suitability of the modeling. The "modelability" concept (defined in the Site Characterization Plan in Appendix A) for this site is then evaluated.

## 1.2 SCOPE OF STUDY

The Site Characterization Plan (see Appendix A) describes the geologic, hydrologic, and geochemical measurements and information needed to construct a conceptual model for use in performing predictive modeling of the radionuclide transport in the site groundwaters. In the preparation of this site characterization plan it was assumed that there were no existing wells at the site. Only basic data concerning background geological, hydrological, and geochemical parameters were used as input to the preliminary conceptual and/or numerical models of the subject site. The procedure for determining the number and location of test boreholes and data points was one of professional scientific judgment, as opposed to a rigid statistical approach. This flexible process utilizes an iterative methodology in the collection and analysis of characterization data, such that the conceptual model is continually refined and revised as required. This approach is consistent with state-of-the-art efforts to characterize geohydrologic environments. These models would, in turn, be used to guide further development of the characterization plan. A site characterization plan is designed to provide the information necessary to build both a defensible conceptual model as well as reasonable hydrologic flow and transport models of the subject site.

Available hydrological and geochemical data at these sites, previously generated by CRNL investigators, have been utilized to fill the data requirements needed for predicting the movement of selected radionuclides from the site. This site has been extremely well characterized and monitored by the CRNL staff who have installed over 100 monitoring wells and boreholes at this site during the past 30 years. An extensive hydrogeological data base

is available from this monitoring network and both selected information as well as the entire data base has been utilized in this study.

In this project, two levels of numerical modeling were conducted. At the first level of modeling only a portion of the existing extensive CRNL data set, conforming to the well placement selection contained in the site characterization plan, was used to develop a relatively simple groundwater flow and transport model, such as would be done in an actual siting assessment and guidance project. The level of investigation was kept to a minimum, as though licensing were being undertaken in an effective yet minimum cost manner. The numerical model has been assessed to assure that no inconsistencies exist, e.g., the mathematical simulation codes used were commensurate with the assumptions made in the conceptual model and the available data. The transport model was used with a radiologic source term equivalent to actual wastes disposed at the subject site during the early 1950s, and the movement of selected nuclides through the system has been modeled. Simulation has been carried forward and compared with the present day distribution of contaminants as determined by field sampling and mapping. This comparison was done to help assess the usefulness of this initial level of site assessment modeling in: 1) helping to understand the contaminant transport behavior at the site through sensitivity studies, and 2) providing a means to bound the possible consequence estimates.

The second level of modeling has involved a more detailed state-of-the-art effort, carried out using the entire, extensive and available CRNL characterization data set along with the expertise of Atomic Energy of Canada, Ltd. (AECL) personnel. The conceptual model has been adjusted as necessary to take advantage of the 30+ years of experience at the subject site, and appropriate alterations in the modeling technique were made.

Finally, a comparison of the results of both modeling efforts has been made, and those results compared with the field observations of actual migration. This comparison has provided insight into the level of effort necessary to meet the regulatory requirement that a site be "modelable." Additionally, this comparison has served to differentiate the advantages, if any, of a more detailed characterization and assessment activity in the early siting assessment phases.

### 1.3 BACKGROUND INFORMATION

Details of the past low-level waste management activities and of the past and current environmental monitoring and geochemical studies conducted at the Nitrate Disposal Pit site at CRNL are presented in Section 4.1. At this site, mixed fission products were released to an infiltration pit beginning in 1953. Monitoring of contaminant movement in the groundwater began in 1955<sup>(6)</sup> and periodic measurements have continued to map the position of the <sup>90</sup>Sr plume front to the present time. Geochemical studies conducted at or near the site have helped to elucidate the retardation mechanisms afforded by the soil<sup>(7,8,9)</sup>. The extensive array of multi-level monitoring wells at this site has permitted a very detailed mapping of the <sup>90</sup>Sr plume migrating in the groundwater from the disposal pit. Thus, this site affords an excellent opportunity to compare model/code predicted radionuclide transport with actual migration which has occurred during the past 30 years.



## 2.0 HYDROGEOLOGY

### 2.1 GEOLOGIC SETTING

The disposal site is located within the boundaries of the Chalk River Nuclear Laboratories situated in the Province of Ontario about 480 km north of Toronto (Figure 1.1). The site is situated in the Grenville Province of the Canadian Shield. The surficial geology of the region, shown in Figure 2.1, is typified by thin deposits of sand and fluvial gravels that overlie a till or crystalline bedrock with low permeability. Bog deposits are typically found in low-lying areas where surficial sediments are thin. On the average, the sediments are about 5 to 15 m thick. The local bedrock has been scoured by glacial action, resulting in an undulating, yet gently sloping surface that serves to direct and control the movement of groundwater in the surficial sediments.

The disposal site lies near the boundary of the catchment area of Maskinonge Lake that drains into the Ottawa River about 8 km downstream. The site was excavated in a sand dune ridge on a terrace about 50 m above and 1 km east of the lake. The disposal pit is close to three lakes. A moderately sized, shallow lake (Lake 233) is immediately east of the site. Two relatively small lakes, Dewdrop and Twin Lakes, are located about 270 m west of the site. Immediately south of Dewdrop Lake, several small springs issue from surficial sediments into a swampy area. The swamp is drained by nearby streams through a gap bounded by bedrock outcrops to other lakes at lower elevations.

Geologic data from 16 boreholes combined with information on bedrock outcrops were used to map the interface between the surficial sandy sediments and the underlying bedrock and till in the vicinity of the disposal site. The contour map of the bedrock and till surface (Figure 2.2) shows bedrock ridges on each side of a terrace. On the eastern ridge, located along the Lake 233 shoreline, bedrock, which rises to an elevation of about 153 m mean sea level (MSL), is found below about 15 m of sand and silt. In contrast, on the western ridge, the bedrock is thinly covered and crops out at a few locations near Twin and Dewdrop Lakes and immediately west and north of the swampy area. Between the two ridges, ground surface dips gently from east to west, which conceals a depression in the bedrock surface where bedrock elevation decreases to just below 143 m (MSL).

Above the bedrock and till, borehole data indicate that the surficial sandy sediments are predominantly composed of fine-to-very-fine sand. However, these sediments also contain two other distinct facies: sand interstratified with silt found within the shallow fine sand unit, and a fine-to-medium sand generally found just above the bedrock and till surface. The vertical and horizontal continuity of these sediments from Lake 233 to the swampy area southwest of the proposed site is illustrated in the geologic section shown in Figure 2.3.

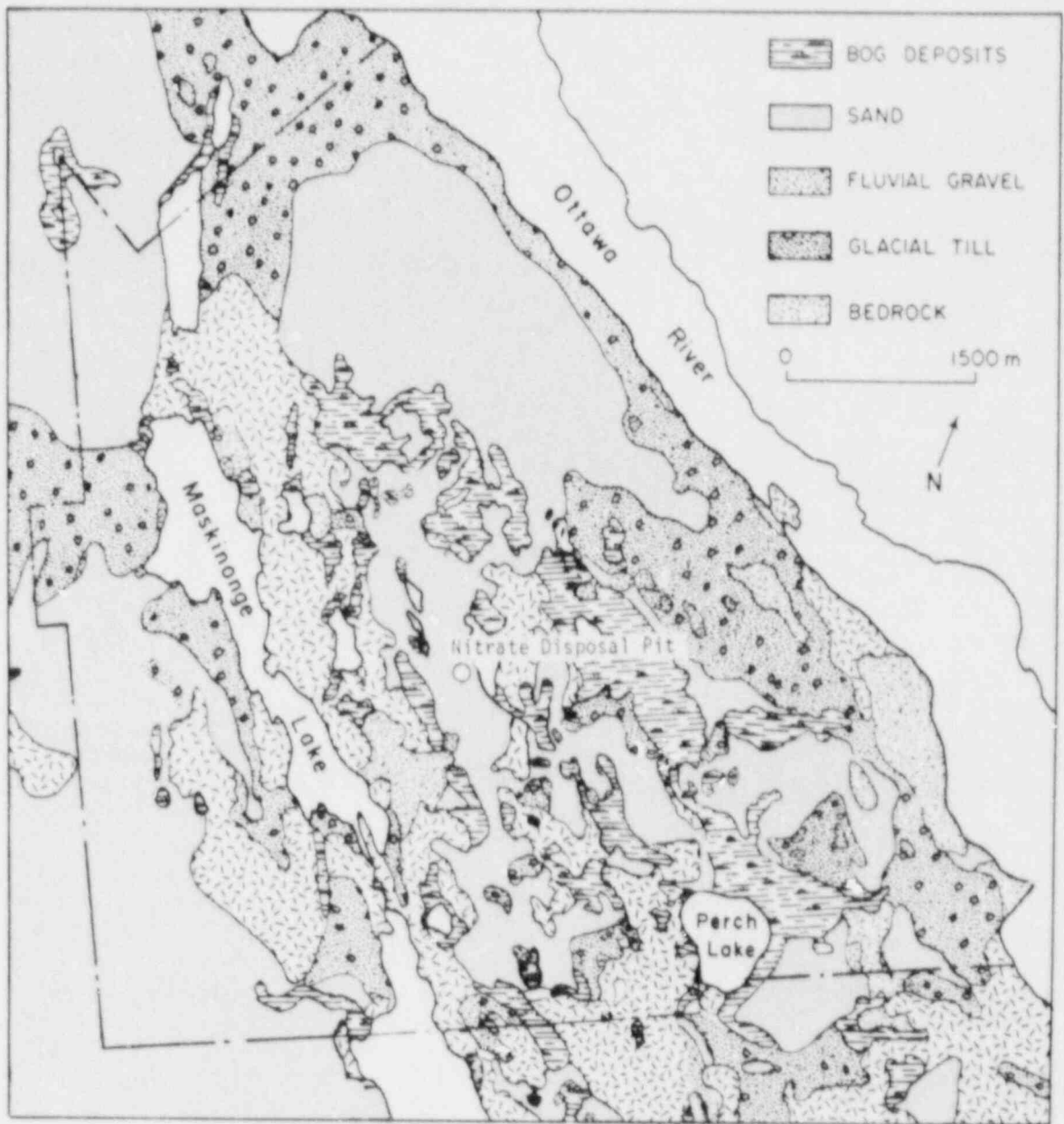


FIGURE 2.1. Surficial Geology of Study Area

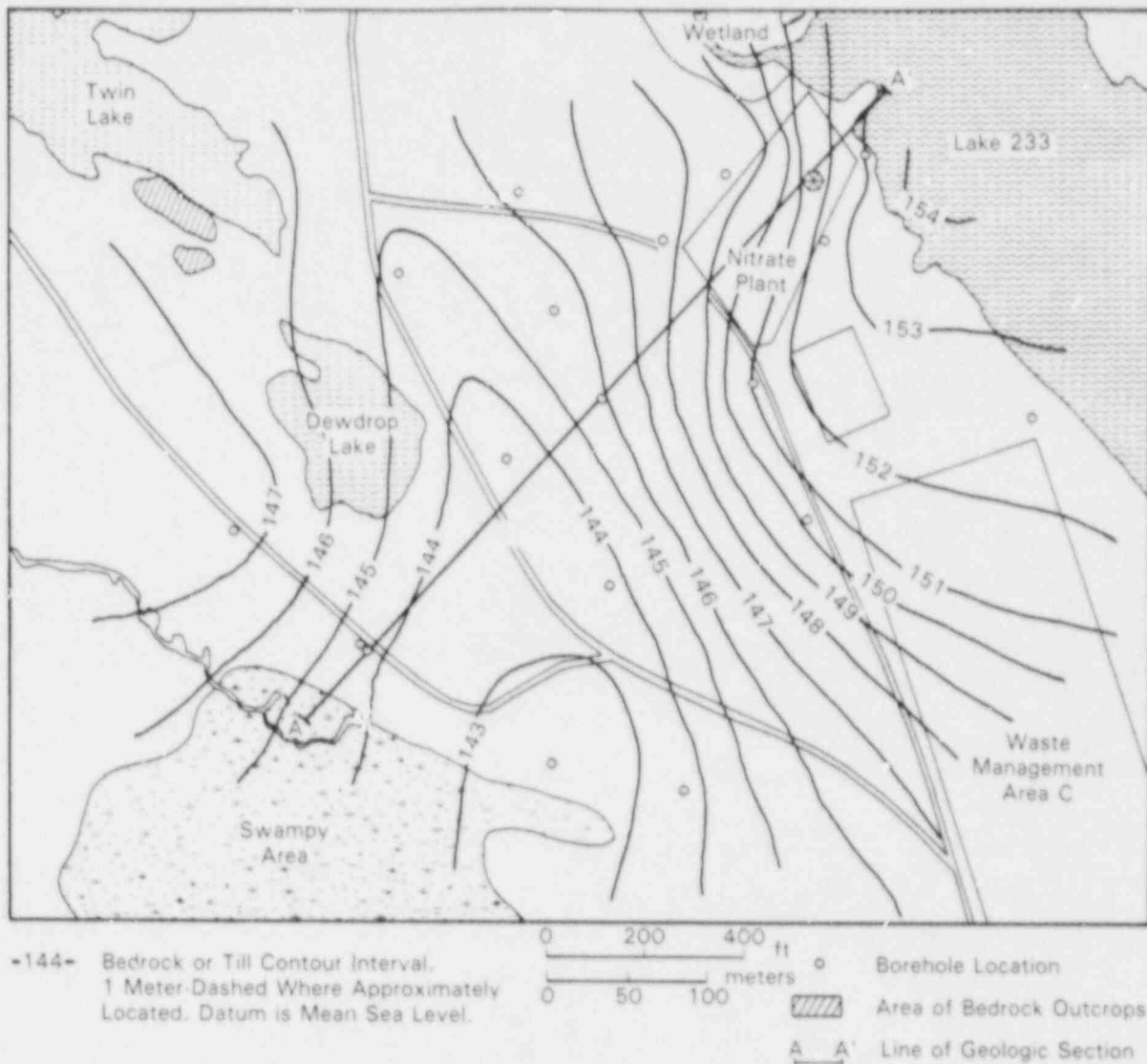


FIGURE 2.2. Map Showing Top Elevation of Bedrock and Till Surface Using the Limited-Data Set Generated by the Site Characterization Plan. See Figure 3.9 for the Maps Generated Through the Use of All Available Data

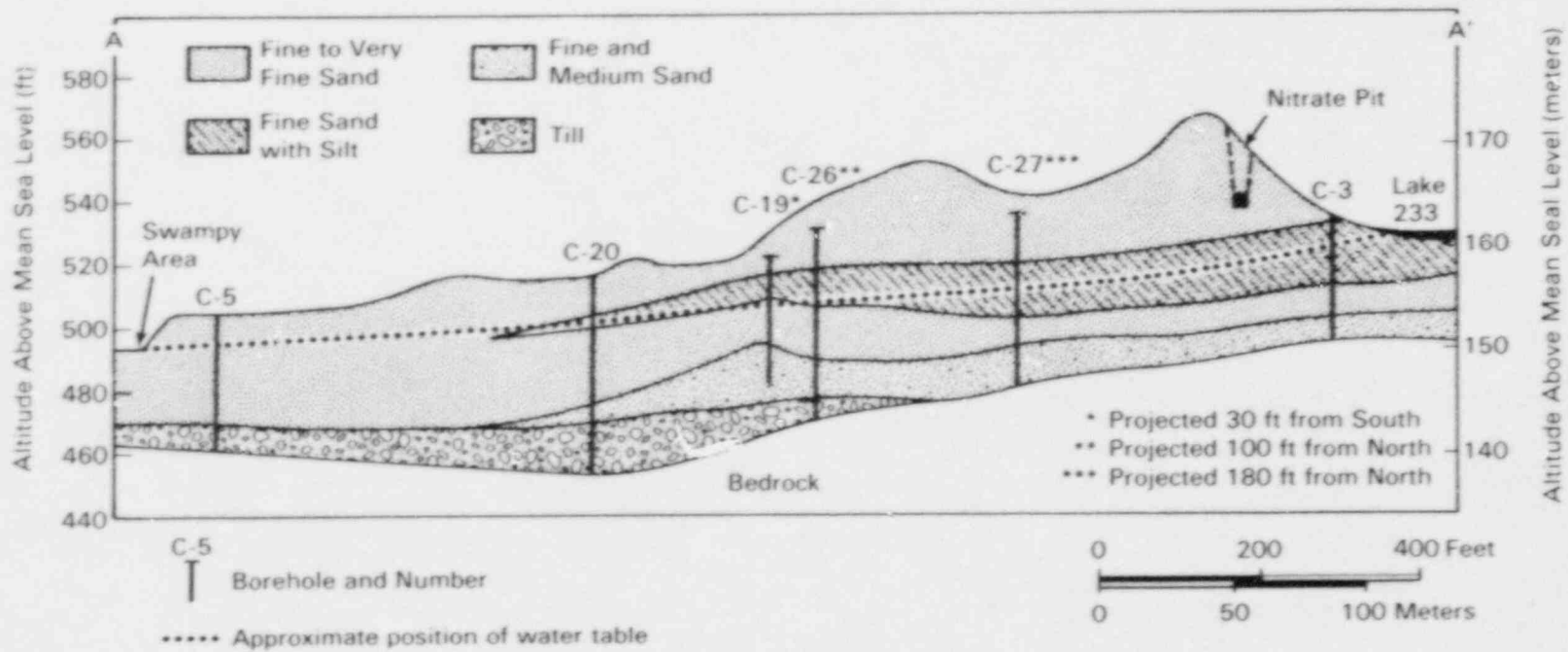


FIGURE 2.3. Geologic Section Showing Major Lithologic Units; Vertical Exaggeration 5:1

## 2.2 GROUNDWATER HYDROLOGY OF SURFICIAL SANDS

The predominant zone of groundwater movement in the vicinity of the disposal site is the surficial sands that overlie the fine-grained till and crystalline bedrock. For this reason, analysis of the groundwater system at the disposal site focused on this unit. Some flow occurs within the till unit, but the clay-rich nature of the till limits the rate and amount of flow. Existing surface-water bodies are generally found in areas where the unconsolidated sediments are thin and bedrock is at or near land surface. This indicates the limited hydraulic conductivity of the underlying bedrock. Although there are visible fractures in the bedrock that could theoretically support fracture flow, the contrast in hydraulic conductivity between the bedrock and overlying sediments is great. Therefore, for the purposes of this study, the bedrock is considered to be impermeable.

### 2.2.1 Areal Extent, Thickness and Composition

The areal extent and saturated thickness of the surficial sands were mapped using geologic data from 16 boreholes and information on bedrock outcrops. The sand was found at all borehole locations. Its saturated thickness, based on water-level measurements made in September 1980, varies from 3 m to just over 10 m in the area of interest (see Figure 2.4). Maximum thicknesses coincide with the areas where the bedrock surface is depressed, about 250 to 300 m west and southwest of the disposal site.

The mineralogy of the surficial sands have been characterized by Pickens et al. as being about 50% feldspar, 30% quartz with minor amounts of sericite, mica, and hornblende. These sediments also contain lesser amounts of garnet, pyroxene, magnetite, and hematite.<sup>(19)</sup>

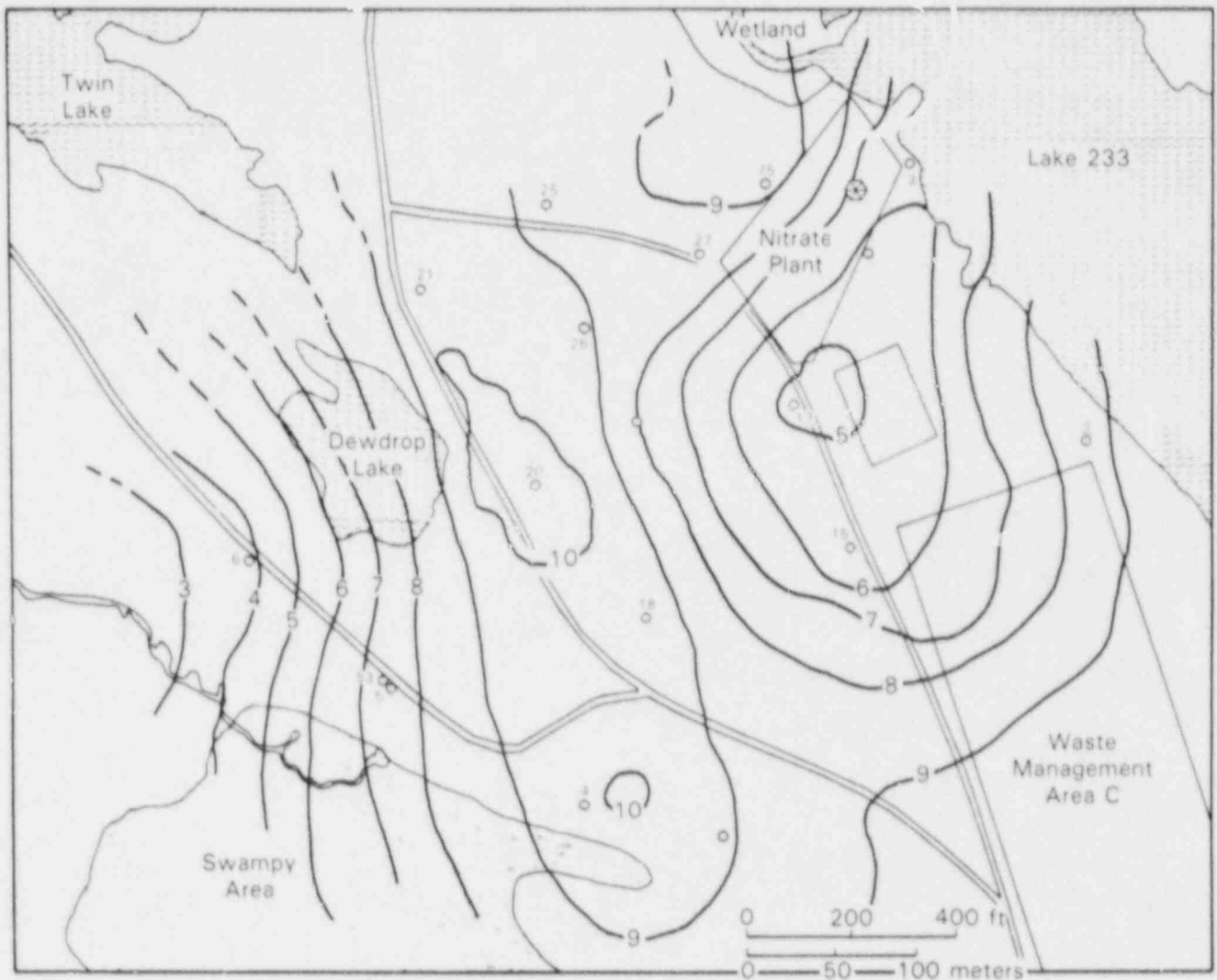
### 2.2.2 Groundwater Flow Patterns

Water-level measurements made in more than 50 piezometers at the 16 borehole locations over the 5-year period 1979 to 1984 were used to construct water-table maps of the surficial sand unit. Figure 2.5 illustrates a typical water-table map, based on measurements made in September 1980. Flow patterns suggested by the map indicate that groundwater in the Lake 233 area and in the vicinity of the site moves laterally toward the southwest before being discharged as seepage to the swampy area lying 460 m southwest of the site. The total drop in water level from the lake to the swamp is about 11 m.

Analysis of water-level data in piezometers at each borehole indicates that, with a few exceptions, water levels measured at the base of surficial sands vary little from those measured near the water table (Figure 2.6). In general, the head differences were less than 0.1 m. However, at some borehole locations, such as borehole C-27, vertical head differences between 0.5 and 1.0 m were observed.

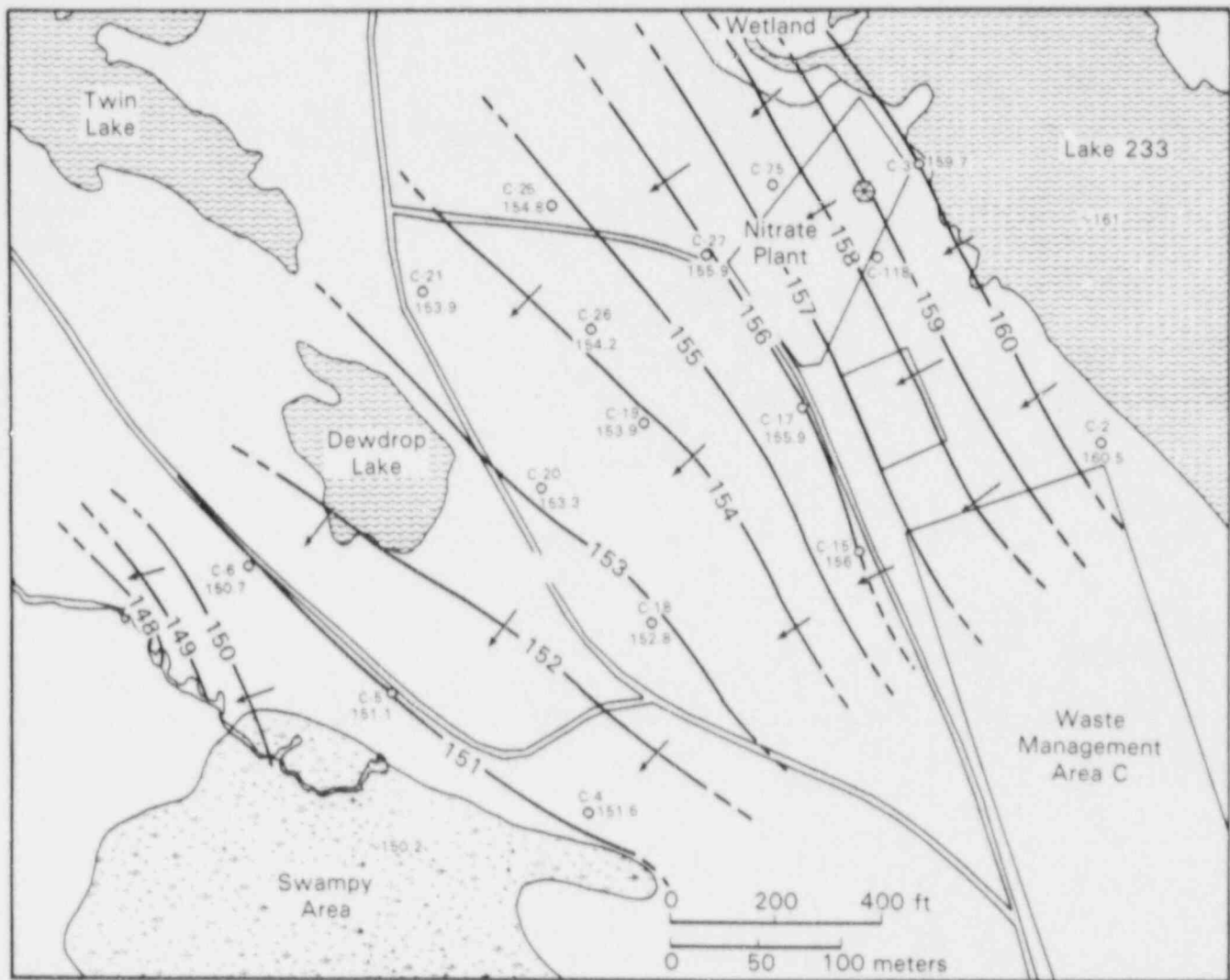
### 2.2.3 Hydraulic Properties

The hydraulic conductivity of the surficial sand aquifer was estimated by the CRNL staff at selected borehole locations with the use of three methods:



- 9- Line of Equal Saturated Thickness  
Interval 1 Meter  
Dashed Where Approximately Located
- 4
- o Borehole and Number

FIGURE 2.4. Saturated Thickness of Surficial Sand Aquifer



-152- Water Table Contour  
Interval 1 Meter  
Datum is Mean Sea Level

C-20  
○ Borehole Number and Water Level  
153.3

← Direction of Groundwater Flow

FIGURE 2.5. Water Table of Surficial Sand Aquifer, September 29, 1980

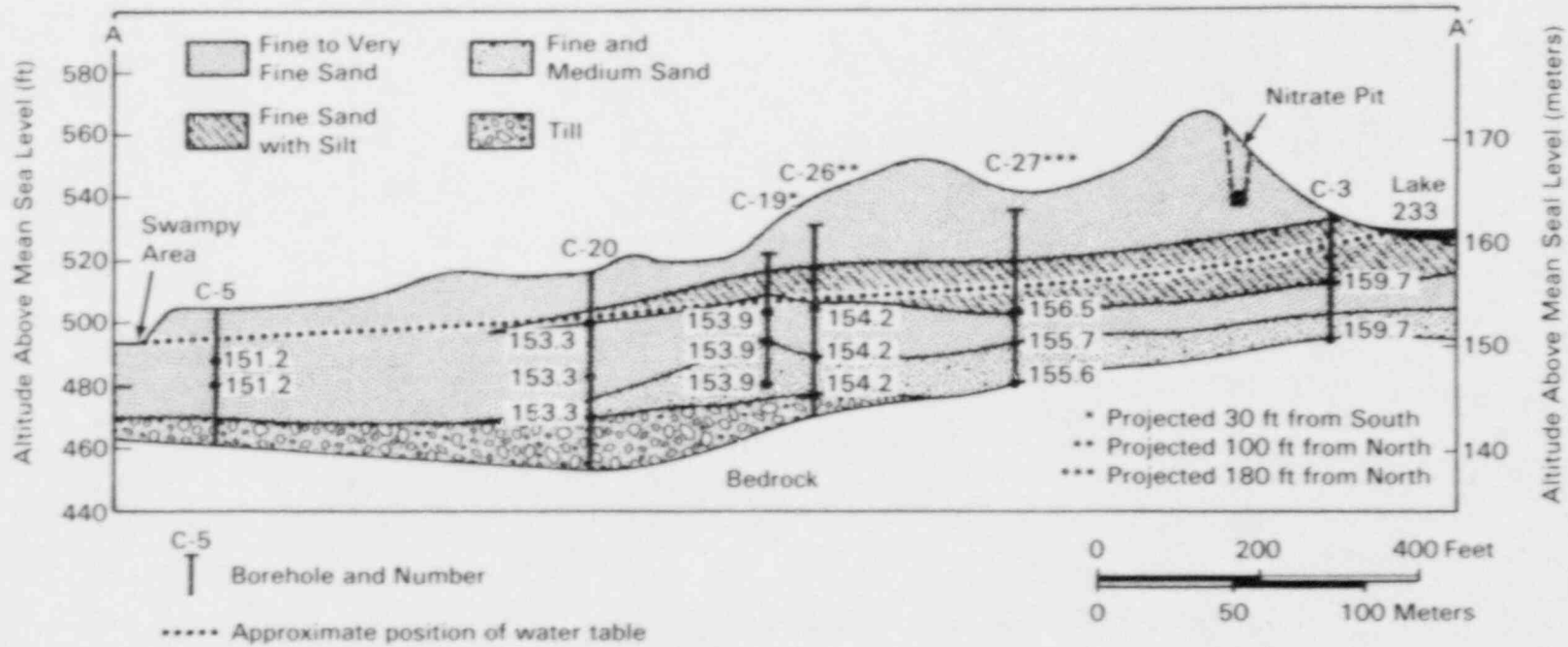


FIGURE 2.6. Geologic Section Showing Major Lithologic Units and Vertical Distribution Head, Vertical Exaggeration 5:1



- empirical measurement of the mean hydraulic conductivity based on grain-size distribution characteristics
- permeameter tests of undisturbed soil cores to yield direct measurements of vertical hydraulic conductivity
- single-well response tests to yield direct measurements of a mean of the vertical and horizontal conductivities.

Individual cores and test intervals were selected to derive hydraulic conductivities that were representative of the three major facies of the sand: the fine-to-very-fine sand, the interstratified sand and silt, and the fine-to-medium sand. The means and ranges of resultant hydraulic conductivities are given by method and hydrostratigraphic unit in Table 2.1.

TABLE 2.1. Summary of Hydraulic Conductivity Values by Hydrostratigraphic Unit and Method, Meters/Day

<u>Hydrostratigraphic Unit</u>	<u>Grain Size Analysis</u>	<u>Permeameter Tests</u>	<u>Single-Well Response Tests</u>
Fine and very fine sand			
Average	5.3	3.1	1.55
Range	4.0 to 7.6	0.4 to 5.3	0.6 to 3.1
Sand interstratified with silt			
Average	-	0.48	-
Fine and medium sand			
Average	9.9	5.9	4.5
Range	6.6 to 17.3	0.95 to 8.6	1.3 to 9.5

The values in Table 2.1 indicate that the hydraulic conductivities derived by the empirical grain-size analyses are consistently higher than those derived from the permeameter and single-well response tests. Results derived from the single-well response tests provided the lowest estimates. In most stratified geologic deposits some degree of anisotropy is present whereby the horizontal conductivity is generally greater than the vertical. Because a single-well response test measures an average of the horizontal and the vertical hydraulic conductivities, values resulting from such analyses would be expected to be greater than those derived from permeameter tests where only the vertical conductivity is measured. This increase does not occur in the data presented in Table 2.1, suggesting that values derived from the single-well response tests should not be considered reliable. The values obtained were in all probability affected by significant frictional losses over the screen interval during the tests.

Hydraulic conductivities derived from the empirical grain-size analyses are probably the most representative estimate of the average conductivity of the

sand. Test results indicate that the fine-to-medium sand is the most permeable unit, with an average of about 10 m/day, and a range between 6.6 m and 17.3 m/day. The next most permeable unit, the fine-to-very-fine sand, averaged 5.3 m/day, and ranged between 4 m and 7.6 m/day. A comparison of the values with those derived from the permeameter test suggests that a small degree of anisotropy may be present, with the horizontal conductivity perhaps being 2 to 3 times higher than the vertical. Although no estimate was made for the sand and silt unit based on the grain-size analysis, the average hydraulic conductivity of this unit was assumed to be about 0.5 m/day, the average value derived from the permeameter tests.

Porosity analyses have been done on some 200 sand samples from the nearby Perch Lake basin<sup>(16)</sup>. Samples of sand with a range similar to the range of grain sizes observed at the Lake 233 area yielded porosity values between 0.33 and 0.43. The median porosity of all the samples was 0.38. These values were considered to be representative of porosities of the sands near the disposal site.

#### 2.2.4 Water-level Fluctuations

Examination of hydrographs of selected wells in the sand aquifer near the proposed site (Figure 2.7) indicates that groundwater levels fluctuate seasonally during each year. These fluctuations reflect changes in storage in response to variations in recharge to and discharge from the aquifer. For the hydrographs and the period of record presented in Figure 2.7, the annual water-level changes varied from about 2 m, as occurred at piezometer III at borehole C-3 (C-3-III) in 1980, to about 0.25 m, as occurred at piezometer II at borehole C-5 (C-5-II) in 1980 and 1983. Water levels generally rise to their highest levels in the spring months, as precipitation, melting snow pack, and associated surface runoff infiltrate a thawing ground surface with little loss to evapotranspiration. During the warmer summer months that follow, precipitation falling on the land surface readily infiltrates the sand sediments, but evapotranspiration can reduce or completely deplete the amount of water actually reaching the water table. As a result, water levels usually decline through the summer and fall. During the winter months, extensive frost in the shallow soils precludes the possibility of rainwater infiltration, and water levels generally reach their lowest level during this period.

Although water levels fluctuate owing to seasonal variations in recharge and discharge to the flow system, they return to approximately the same level each spring. For all wells, annual maximum and minimum water levels stay within a meter of what could be considered an "average" long-term level. For many of the wells, the deviation is less than 0.5 m. The small magnitude of these changes and the lack of evidence of any increase or decrease in water levels, at least in the short term, indicate that the shallow aquifer system near the site is at present in a state of dynamic equilibrium.

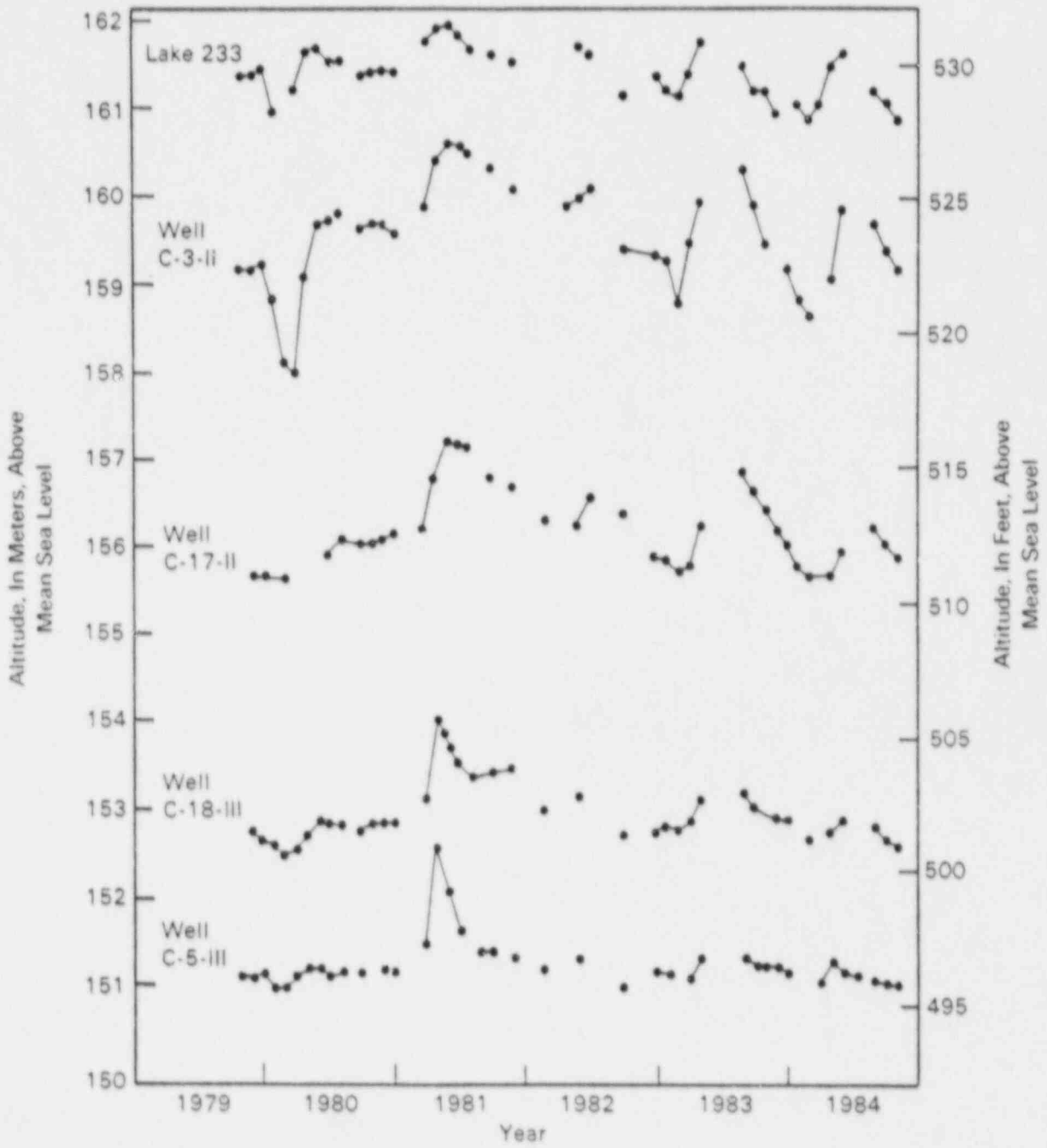


FIGURE 2.7. Hydrographs of Selected Observation Wells, 1979 to 1984

## 3.0 MODELING OF GROUNDWATER FLOW AND TRANSPORT

### 3.1 LIMITED-DATA MODELING

During the first part of this investigation, geologic and hydrologic data from the initial 16 boreholes identified in the site characterization plan were used to develop a groundwater flow and transport model of an area in the vicinity of the Nitrate Disposal Pit site. This model was calibrated with measured water-level data and provided the hydrologic framework for the solute transport model used to predict the movement of radionuclides from the site.

#### 3.1.1 Conceptual Model of Flow and Transport

The direction of groundwater flow indicated by water-table maps suggests that the area of concern for groundwater flow from the Nitrate Pit site is relatively small, extending from about 460 m south-southwest to the swampy area. Most of the groundwater flows through the uppermost sandy sediments overlying the till and bedrock. Flow occurs in the till, but the clay-rich nature of the till inhibits the total amount and rate of flow. Existing lakes and swamps are located in areas where the underlying till and bedrock surface is close to the ground surface or crops out. The occurrence of these surface water bodies is an indication of the limited permeability of the underlying materials. For this study, the modeling analysis focused on groundwater flow and transport in the shallow sands.

#### 3.1.2 Model Selection and Design

##### 3.1.2.1 CFEST Computer Data

The Coupled Fluid, Energy, Solute Transport (CFEST) code developed by Gupta and others<sup>(12)</sup> was selected for use in this investigation. The CFEST code was developed for the Underground Storage Program managed for the Department of Energy by PNL. CFEST is an extension of the Finite-Element Three-Dimensional Groundwater (FE3DGW) code developed by Gupta and others<sup>(13)</sup>.

CFEST is a finite-element code that is capable of simulating groundwater flow and attenuated transport of a decaying radionuclide in a two-dimensional horizontal or vertical plane or in three dimensions. Although geologic data suggest that there may be variations in the surficial sand caused by facies changes, the limited data being used in this investigation did not warrant the use of a detailed three-dimensional analysis. Consequently, the surficial sand was considered a vertically homogeneous and horizontally heterogeneous aquifer and CFEST was applied in a two-dimensional horizontal plane.

##### 3.1.2.2 Finite Element Grid and Boundary Conditions

The model which covers about 0.17 km<sup>2</sup> is based on a bilinear, quadrilateral, finite-element mesh composed of 280 nodes and 247 elements (Figure 3.1)

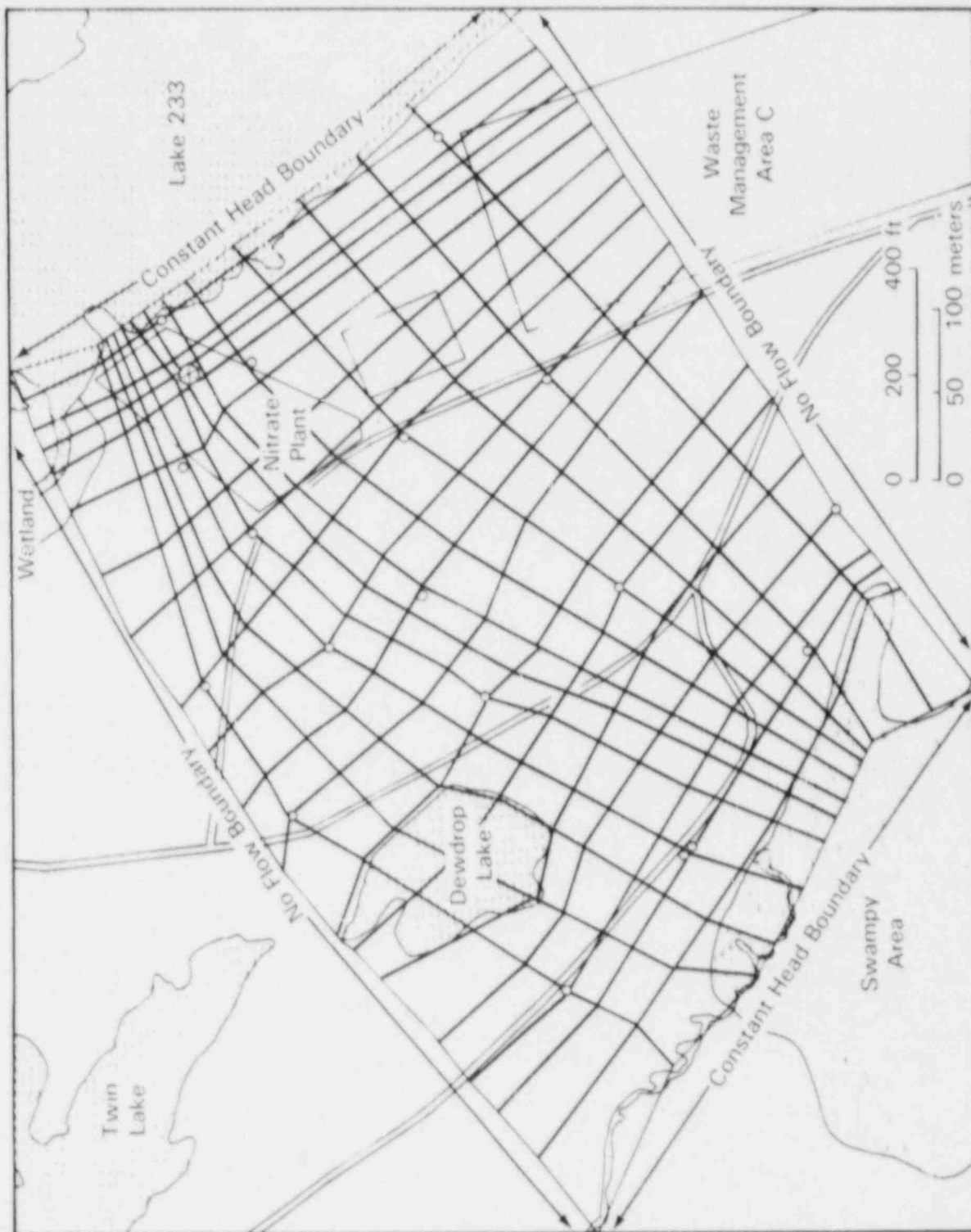


FIGURE 3.1. Finite Element Mesh and Boundary Conditions Used in Groundwater Flow Model

The mesh is designed so that all borehole locations and geographic features pertinent to the simulation approximately coincide with either a node or an element.

Boundary conditions in the model were selected to represent real physical hydrologic boundaries or to approximate as closely as possible artificially imposed boundaries. The location of Lake 233, northeast of the proposed site, represents a relative constant-head boundary for the local ground-water system. In the model, nodes approximating the location of the lake-shore were simulated as constant-head nodes. The heads specified were extrapolated from measured water-table elevations.

Water-level data suggest that the swampy area and nearby streams southwest of the site represent relatively constant-head discharge boundaries for the shallow sand aquifer. In the model, nodes that approximately coincided with the edge of the swamp and channels of nearby streams were simulated as constant heads. No direct measurements of stage in the swamp or streams were taken; therefore, constant heads used in the simulations were estimated from elevations derived from topographic maps.

The water-table map in Figure 2.5 illustrates that water-level contours are roughly parallel to the Lake 233 shoreline to the northeast and to the edge of the swamp and stream channels to the southwest. The model's lateral boundaries on the northwest and southeast were designed to take advantage of this feature and were made perpendicular to the contour lines. Thus, these boundaries represent approximate flow lines and are effective boundaries across which no flow can either enter or leave the model area.

The last boundary considered in the model involves the hydraulic relationship of the surficial sand aquifer and the underlying till and crystalline bedrock. Because the till and bedrock are significantly less permeable than the surficial sand, it was assumed that vertical flow to and from these units was not significant, and the base of the surficial sand was simulated as a no-flow boundary.

#### 3.1.2.3 Model Data Input

Flow in the surficial sand was simulated in the model as a single two-dimensional aquifer layer. Data needed for the simulation included the aquifer top and bottom elevations, a distribution of hydraulic conductivity, and estimates of effective recharge to the aquifer. The aquifer top was approximated from the water-table map illustrated in Figure 2.5. The aquifer bottom was approximated by a contour map of the till and bedrock surface shown in Figure 2.2.

A distribution of hydraulic conductivity for the sand aquifer was developed from the average hydraulic conductivities for each hydrostratigraphic unit. A transmissivity distribution was first developed by multiplying the average permeability of each unit by the saturated thickness of each unit at all of the borehole locations. These values were then interpolated to other locations with the geostatistical technique of kriging to provide an areal transmissivity distribution. This distribution of the transmissivity was divided by the thickness derived from the saturated thickness map (Figure

2.4) to produce an areal distribution of effective hydraulic conductivity. The resulting distribution, which ranged from 5.5 to 9.5 m/day, provided an initial estimate for the model.

Recharge values were obtained by recalculating the water balance presented by Barry.<sup>(14)</sup> In his study of Perch Lake, he presented the annual distribution of recharge. Although the months of heaviest precipitation occur during the warmer periods, evaporation during these months reduces the water actually available for infiltration. Precipitation during the cooler months of the year is predominantly snow, which reduces infiltration during those months. Maximum infiltration occurs during the spring months when accumulated snowfall melt is available for recharge. Approximately 32 to 33 cm of recharge were calculated to occur out of an available 84 cm of precipitation.

### 3.1.3 Model Calibration

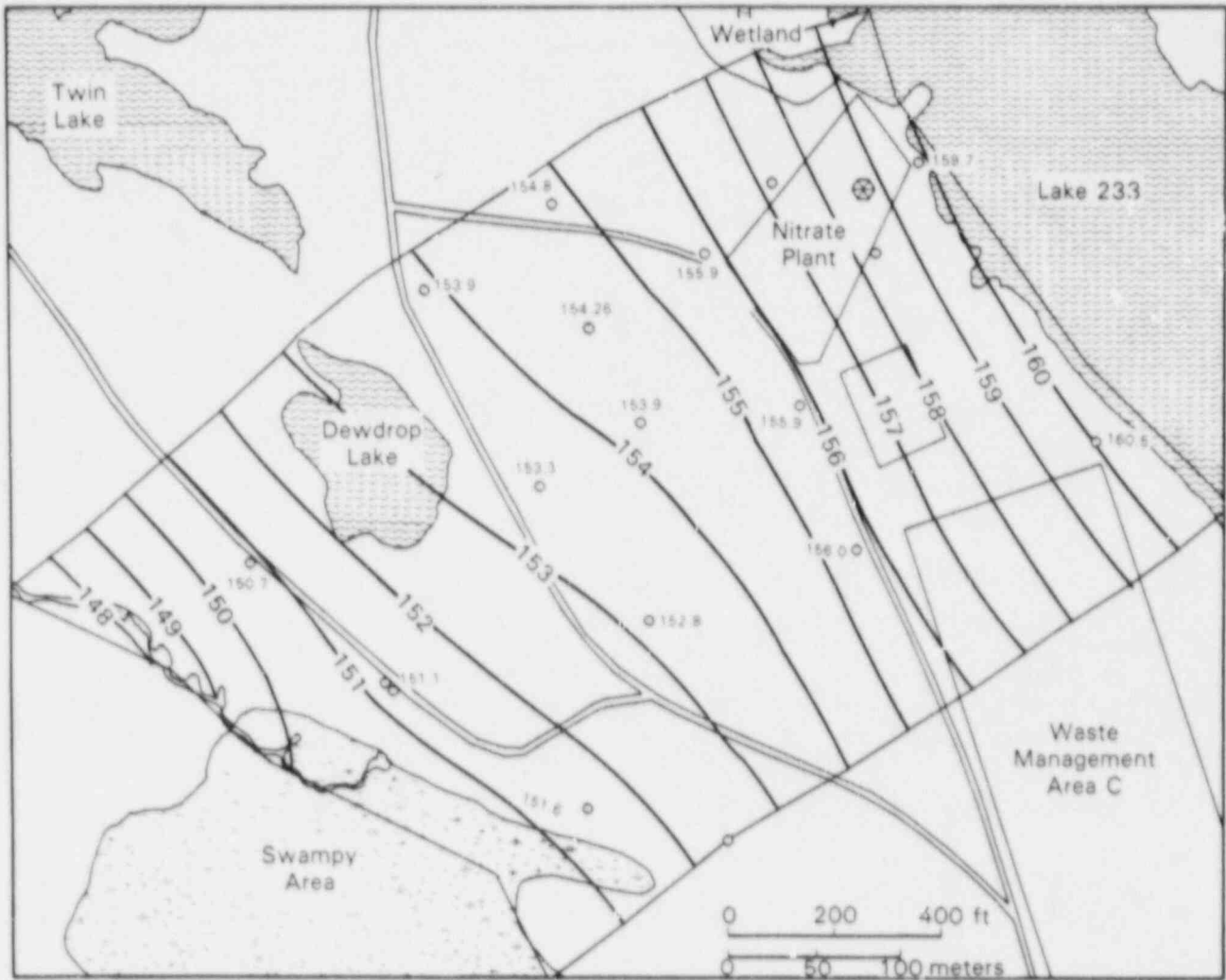
Because water-level fluctuations indicate that the surficial sand aquifer is in dynamic equilibrium, groundwater flow conditions in the aquifer can be simulated adequately with a steady-state model. The hydrographs in Figure 2.7 suggest that conditions observed in the fall of 1980 represent a relatively stable period of water-level changes, and therefore a reasonable period to use for calibration of the model.

The model was applied in steady-state to match predicted water levels to those measured on September 29, 1980, a set of measurements representative of the fall 1980 conditions. During model calibration, input parameters were adjusted within the range of acceptable values until simulated conditions most closely approximated observed conditions. Input variables adjusted during calibration were hydraulic conductivity and recharge.

With the estimated recharge rate of 32.8 cm/yr and some minor adjustments to the initial hydraulic conductivity distribution, the predicted steady-state heads reasonably matched measured heads for the September 1980 conditions. Figure 3.2 illustrates the comparison between simulated and measured head. The mean absolute value of error (observed head minus simulated head) was 0.27 m. Most of the observed heads were matched within 0.45 m and all were matched within 0.7 m.

The distribution of transmissivity shown in Figure 3.3 represents the range of values resulting from model calibration. Final transmissivities were between 40 and 120 m<sup>2</sup>/day, with the maximum values located near the bedrock surface depression west and southwest of the proposed site. Hydraulic conductivity values, derived by dividing the calibrated transmissivity distribution by the saturated thicknesses given in Figure 2.4 ranged between 4.0 and 12.0 m/day. This range is well within that derived from empirical grain-size analyses.

The water budget of the calibrated model, presented in Table 3.1, indicated that infiltration from precipitation, which amounted to  $1.57 \times 10^5$  m<sup>3</sup>/day, accounted for about 36% of the total recharge to the simulated area. The remainder of the recharge,  $2.81 \times 10^5$  m<sup>3</sup>/day, was derived from the Lake 233



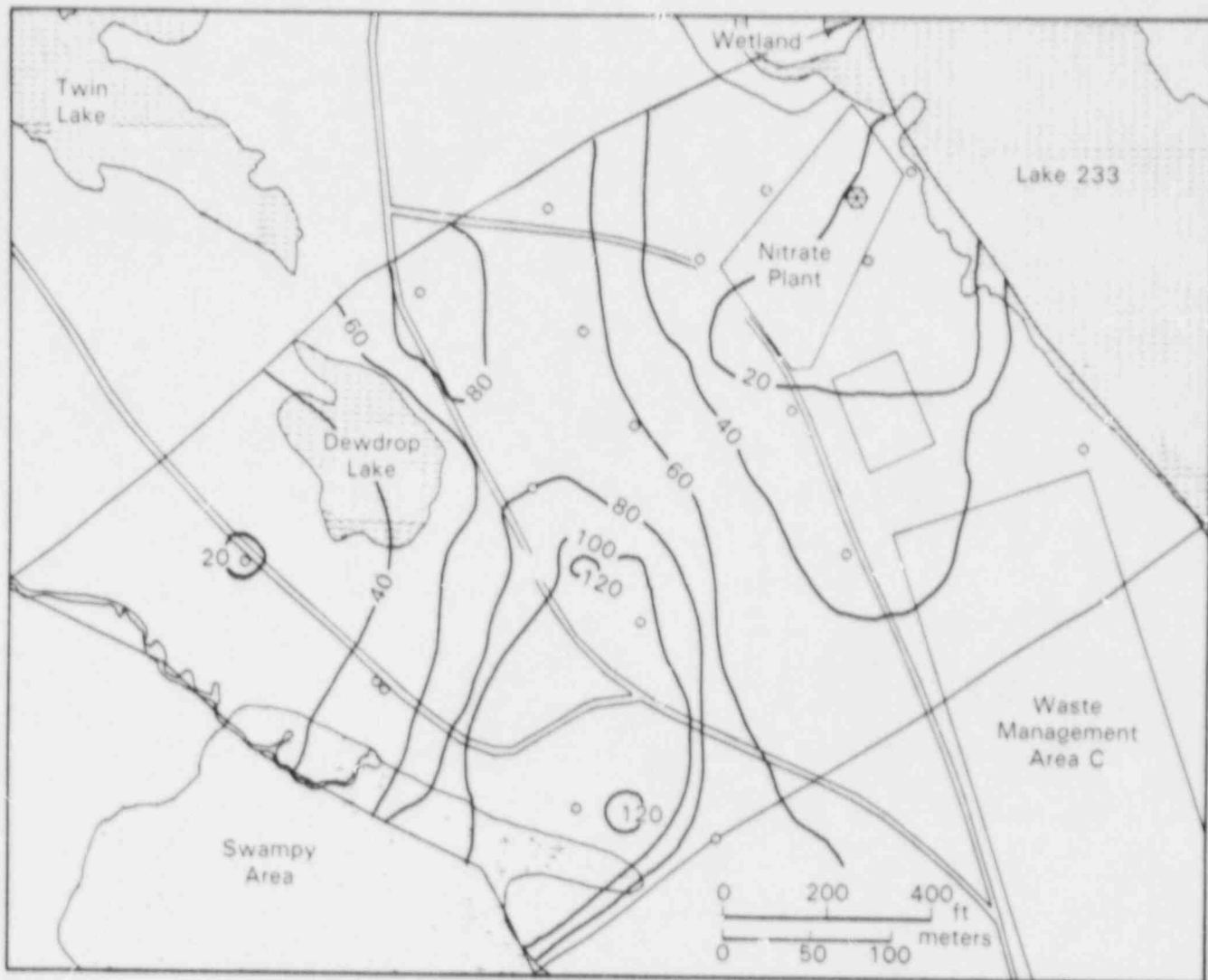
-153- Simulated Water Level Contour  
Interval 1 Meter. Datum  
is Mean Sea Level.

155.9

○ Water Level Measured Sept. 29, 1980

FIGURE 3.2. Comparison of Simulated and Observed Head in the Limited-Data Calibrated Steady-State Groundwater Flow Model of September 1980 Conditions





-100- Line of Equal Transmissivity  
Interval 20  $\text{meter}^2/\text{day}$

C-26  
○ Borehole and Number

**FIGURE 3.3.** Transmissivity Distribution in Calibrated Steady-State Groundwater Flow Model of October 1984 Conditions Using the Limited-Data Set

TABLE 3.1. Water Budget for the Limited-Data Calibrated Steady-state Groundwater Flow Model for September 1980 Conditions (meters/day)

<u>Sources of Recharge</u>	
Infiltration of precipitation	1.570 x 10 <sup>5</sup>
Lake 233 constant-head boundary	<u>2.808 x 10<sup>5</sup></u>
	4.378 x 10 <sup>5</sup>
Swampy area and stream channel constant-head boundary	<u>4.378 x 10<sup>5</sup></u>
Total recharge	4.378 x 10 <sup>5</sup>

boundary. Discharge amounting to  $4.4 \times 10^5 \text{ m}^3/\text{day}$  was simulated at the swampy area and the stream channel boundary along the southwest part of the model.

#### 3.1.4 Simulation of Radionuclide Transport

The calibrated steady-state groundwater flow model provided flow conditions to simulate the transport of selected radionuclides from the disposal pit. In an attempt to minimize numerical dispersion in transport, a submodel of the flow model using a finer finite-element mesh was developed. The position and orientation of the submodel grid containing 777 nodes and 716 elements are shown in Figure 3.4.

##### 3.1.4.1 Source-Term Characterization and Release

From 1953 to 1954, the disposal site received an ammonium nitrate solution from fuel reprocessing operations consisting of about 3800 liters of acid waste containing complexing agents. Waste solutions were drained to a small pit over a period of a year. The pit was lined with limestone to neutralize acidic wastes and to serve as a sorption medium for radionuclides in the effluent solutions. The two primary components in the source material were <sup>90</sup>Sr and <sup>137</sup>Cs with other fission products present at concentrations assumed to be in proportion to their relative fission yields. The total activity from all waste solutions is estimated to be between 1000 to 1500 Ci of mixed fission products, of which 700 to 1000 Ci are <sup>90</sup>Sr and 200 to 300 Ci are <sup>137</sup>Cs.

The principal radionuclides in the source materials for the disposal site are expected to be <sup>90</sup>Sr and <sup>137</sup>Cs. Once introduced into the groundwater system, these radionuclides will migrate in the principal direction of groundwater movement. However, because of their affinity for being adsorbed onto soils, they would be retarded and would migrate at rates much slower than groundwater flow rates. Distribution coefficients ( $K_d$ )

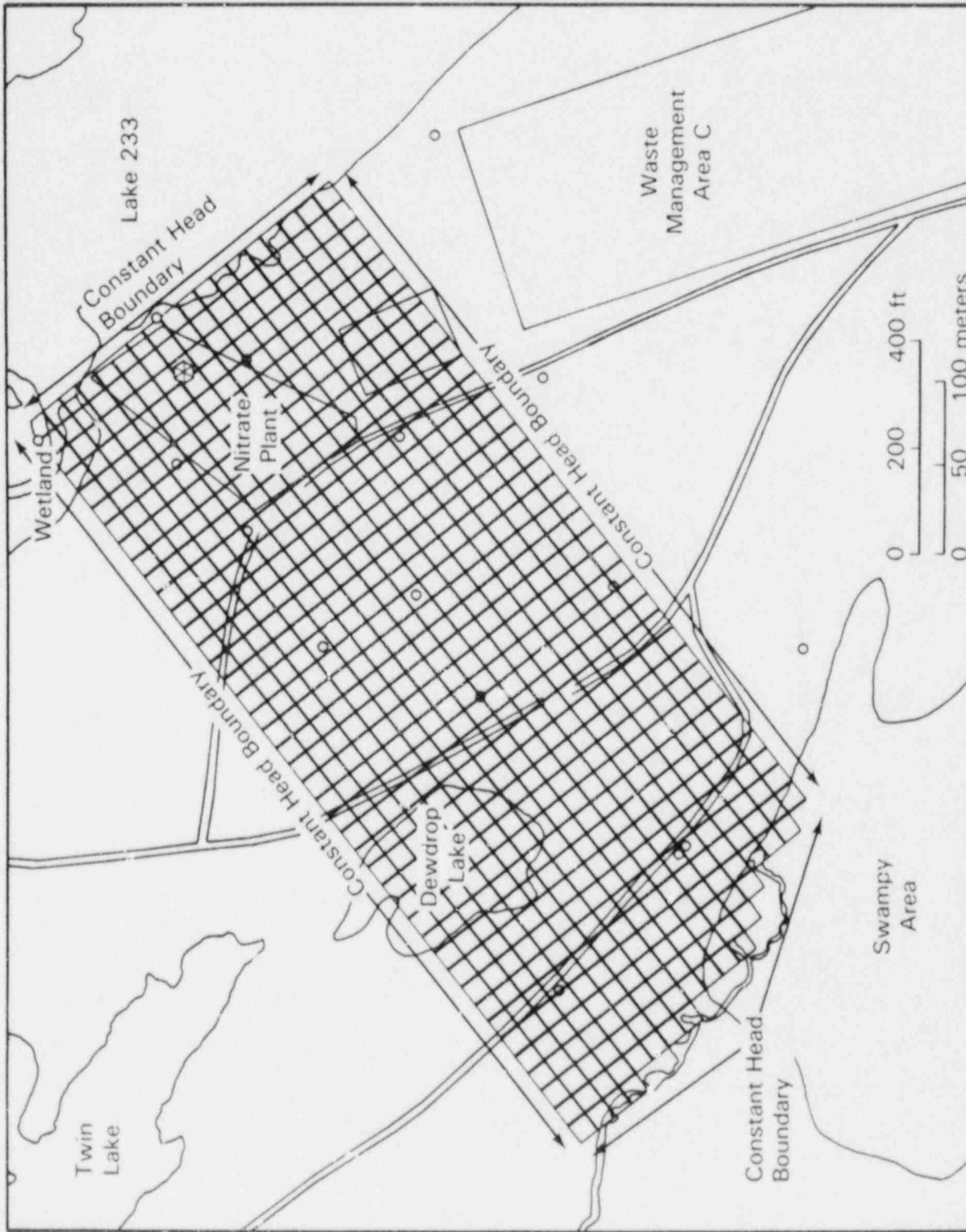


FIGURE 3.4. Refined Finite Element Mesh and Boundary Conditions Used in Groundwater Flow and Transport Simulations

reported by Killey<sup>(7)</sup> and by Jackson, et al.<sup>(8)</sup> from analyses of contaminated soil cores obtained from similar nearby sandy aquifer material generally ranged from 4 to 19 ml/g for <sup>90</sup>Sr and from 50 to 525 ml/g for <sup>137</sup>Cs. The retardation of the contaminant front (R) relative to water movement is described in CFEST by the following equation:

$$R = \frac{v}{v_c} = 1 + \frac{\rho_b}{n} \cdot K_d \quad (3.1)$$

where  $v$  is the average linear velocity of the groundwater,  $v_c$  is the average velocity of the retarded constituent,  $\rho_b$  is the bulk density of the aquifer material, and  $n$  is the aquifer porosity.<sup>(15)</sup> This equation is commonly referred to as the retardation factor. Using a bulk density of 1.7 g/cm<sup>3</sup> reported by Jackson et al.<sup>(3)</sup> and a porosity of 0.38 reported by Parsons<sup>(16)</sup>, these coefficients would translate into retardation factors of 20 to 86 for <sup>90</sup>Sr and 250 to 2540 for <sup>137</sup>Cs. Other radionuclides were present in the waste stream but, because of their short half-lives or their activity levels, they are not expected to be environmentally significant and were not considered in this analysis.

The disposal pit covered an area of about 55 m<sup>2</sup> (7.5 m by 7.5 m) and was constructed so that the base of the pit was within 7 to 8 m of the average water-table elevation. Disposal of 3780 liters of liquid waste occurred over a period of about a year. This rate of disposal represented about 20% of the average annual recharge rate simulated in the calibrated model.

#### 3.1.4.2 Strontium-90 Transport

Release of the <sup>90</sup>Sr inventory from the disposal facility was modeled under three different scenarios. A rapid release of the inventory was modeled by gradually introducing a total of 1000 Ci of <sup>90</sup>Sr into the shallow aquifer over a period of 1 year. This scenario is representative of release conditions that might occur if the limestone lining the disposal pit was ineffective in neutralizing the acidic waste form. The same scenario was simulated with a distribution of retardation factors that varied in space to examine the potential interaction of the acidic liquid waste form with the native groundwater. A third experiment simulated a less rapid release of the same inventory over a period of 30 years. This scenario is representative of release conditions that would occur if the crushed limestone in the pit and underlying soils above the water table were effective in adsorbing the <sup>90</sup>Sr inventory. Each scenario was simulated over a period of 30 years to approximate the historical period since the actual disposal (1953 to 1954). The range of values of pertinent transport parameters evaluated in these simulations was as follows:

Porosity = 0.38  
 Longitudinal dispersivity = 1 to 10 m  
 Transverse dispersivity = 0.01 to 1.0 m  
 Ratio of transverse to  
 longitudinal dispersivity = 0.01 to 0.1  
 Strontium-90 half-life = 28.1 yr  
 Retardation factor = 20 to 100

Because the shallow sediments near the disposal site are, for the most part, fairly well sorted and contain a narrow range of grain sizes, the amount of longitudinal and transverse spreading of the contaminant plume is expected to be small. Thus, we have chosen to present model results derived from simulations that used the lowest values of the longitudinal dispersivity, 1 m, and a small value of transverse dispersivity, 0.1 m. Model results with lower transverse dispersion values were not significantly different than those resulting from the 0.1 m value.

Modeling results of the 1-year release with a constant retardation factor, a longitudinal dispersivity of 1.0 m, a transverse dispersivity of 0.1 m, and retardation factors of 25 and 75 ( $K_d = 5$  and  $15$  ml/g) are presented in Figure 3.5. These retardation factors (25 and 75) are generally representative of the range reported by Jackson, et al.<sup>(8)</sup>. With a retardation factor of 25, the model predicted that groundwater containing 10 pCi/l had migrated a downgradient distance of about 325 m from the disposal site. Concentrations in groundwater directly beneath the disposal pit were about 12,000 pCi/l. Peak concentrations of about 400,000 pCi/l were simulated about 80 m from the site. With a retardation factor of 75, concentrations of about 10 pCi/l were simulated about 180 m downgradient from the pit. Concentrations directly beneath the pit were between 800,000 and 900,000 pCi/l. Peak concentrations of about 1.3 million pCi/l were simulated about 25 m from the disposal pit.

To evaluate the effect of a 1-year release with a variable retardation factor within the plume, we developed a distribution of retardation factors ranging from 2 to 25 (Figure 3.6). The area of lowest retardation was located along a flow path immediately downgradient from the pit. Retardation factors were increased outward in zones to a factor of 25 from the area of lowest retardation.

Results of modeling the 1-year release of the  $^{90}\text{Sr}$  inventory with the variable retardation factors are presented in Figure 3.7. These results are very similar to results of a comparable simulation in Figure 3.5 (top) except that, as expected in an area immediately downgradient from the site,  $^{90}\text{Sr}$  has migrated slightly farther. For this particular set of retardation factors,  $^{90}\text{Sr}$  concentrations of 10 pCi/l were simulated over a downgradient distance of about 160 m after 4 years, 215 m after 10 years, and 365 m after 30 years. Peak concentrations of above 750,000 pCi/l were predicted about 115 m downgradient from the pit.

Results of the 30-year inventory release with constant retardation (25 and 75), a longitudinal dispersivity of 1 m, and a transverse dispersivity of 0.1 m are presented in Figure 3.8. A comparison of these results with results in Figure 3.5 shows that the overall transport of the plume is about the same for all scenarios. However, concentrations within the plume have a distinctly different distribution. While the peak concentration migrates away from the source area in the 1-year release experiments, the peak concentration in modeling the 30-year release generally results in the peak concentration remaining in close to the disposal pit area. The magnitude of peak concentrations during the first 4 to 6 years of the 30-year inventory release simulation were, as expected, generally smaller than peaks predicted for the 1-year release. After 4 years of release and a retardation factor

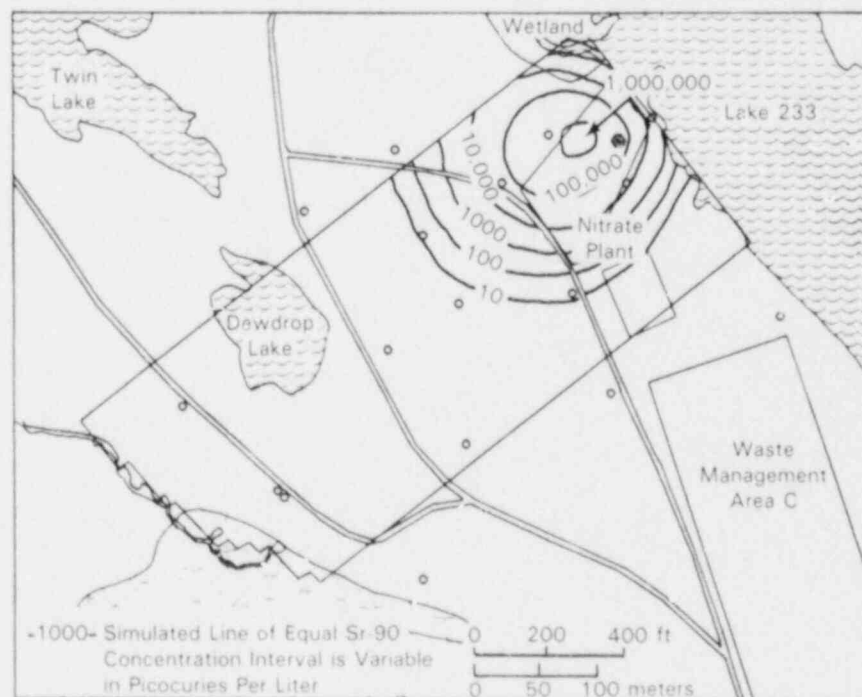
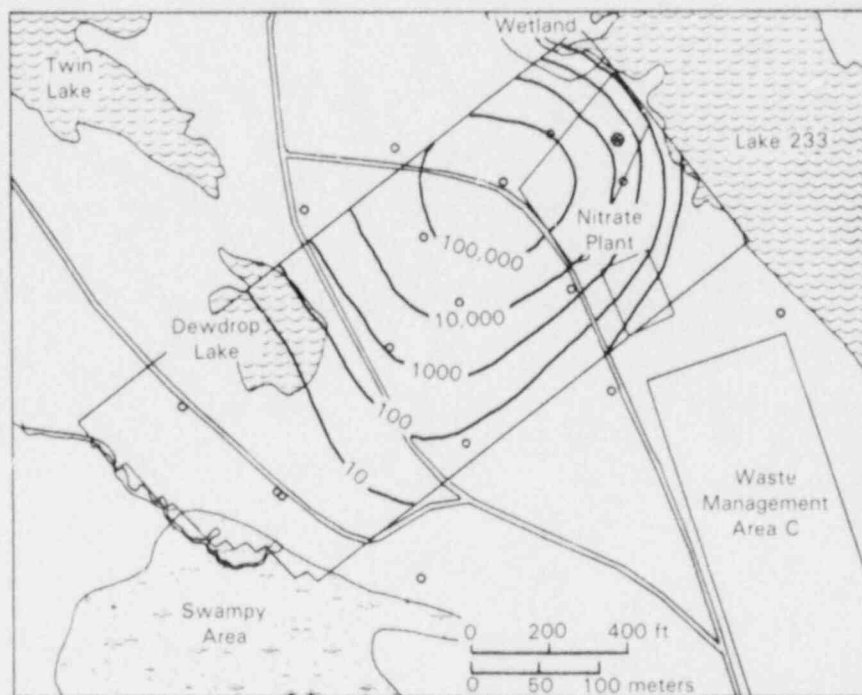


FIGURE 3.5. Strontium-90 Concentrations After 30 Years Resulting from the Limited-Data Simulation of a 1-Year Inventory Release, Using a Longitudinal Dispersivity of 1.0 m, a Transverse Dispersivity of 0.1 m and Retardation Factors of 25 (Top) and 75 (Bottom)

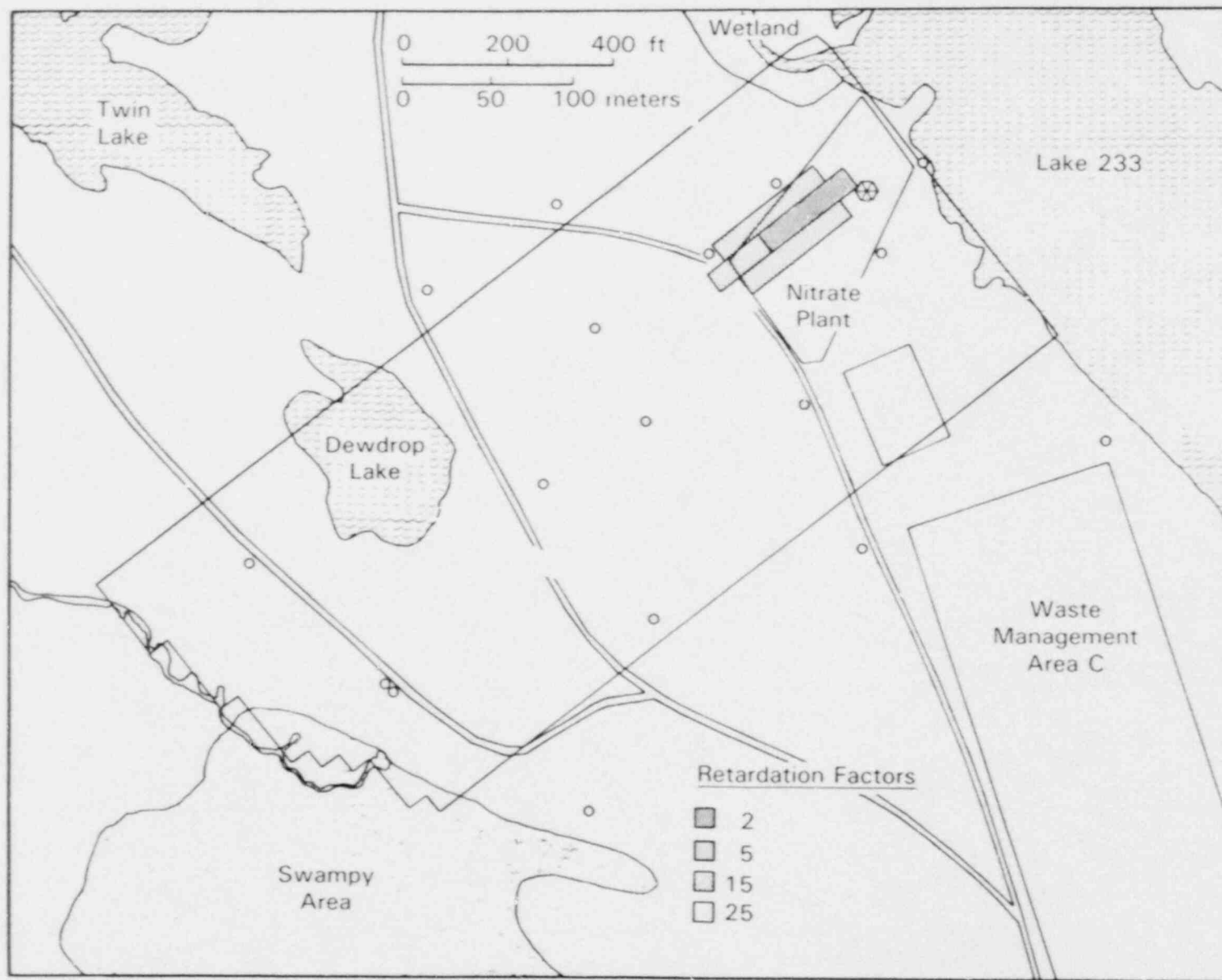
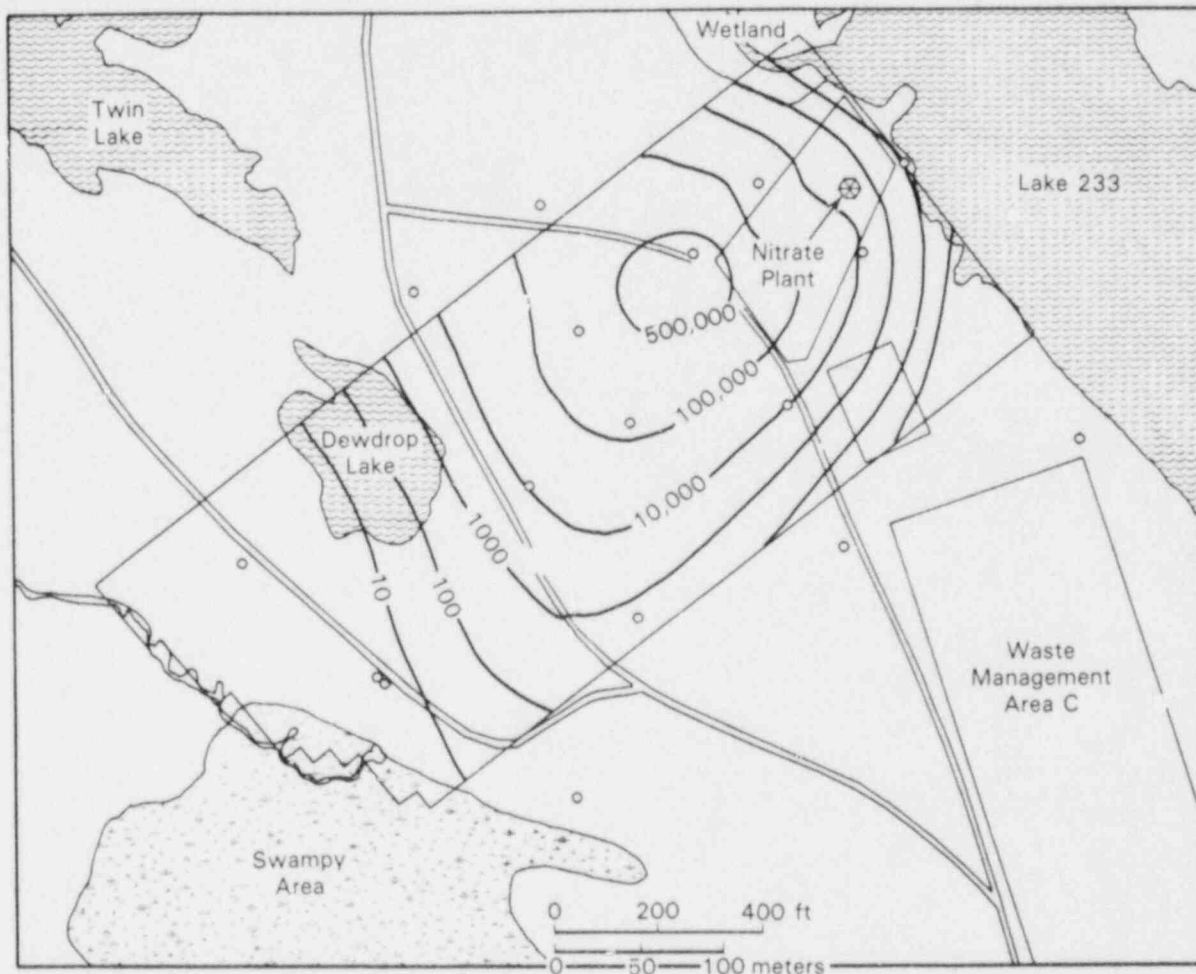


FIGURE 3.6. Areal Distribution of Retardation Factors Used in Simulation of a 1-Year Inventory Release of  $^{90}\text{Sr}$  with Retardation Factors Varying from 2 to 25



-1000- Simulated Line of Equal Sr-90  
Concentration Interval is Variable  
in Picocuries Per Liter

○ Borehole Location

**FIGURE 3.7.** Strontium-90 Concentrations After 30 Years Resulting from the Limited-Data Simulation of a 1-Year Inventory Release Using a Longitudinal Dispersivity of 1.0 m, a Transverse Dispersivity of 0.1 m, and a Variable Distribution of Retardation Factors Ranging from 2 to 25



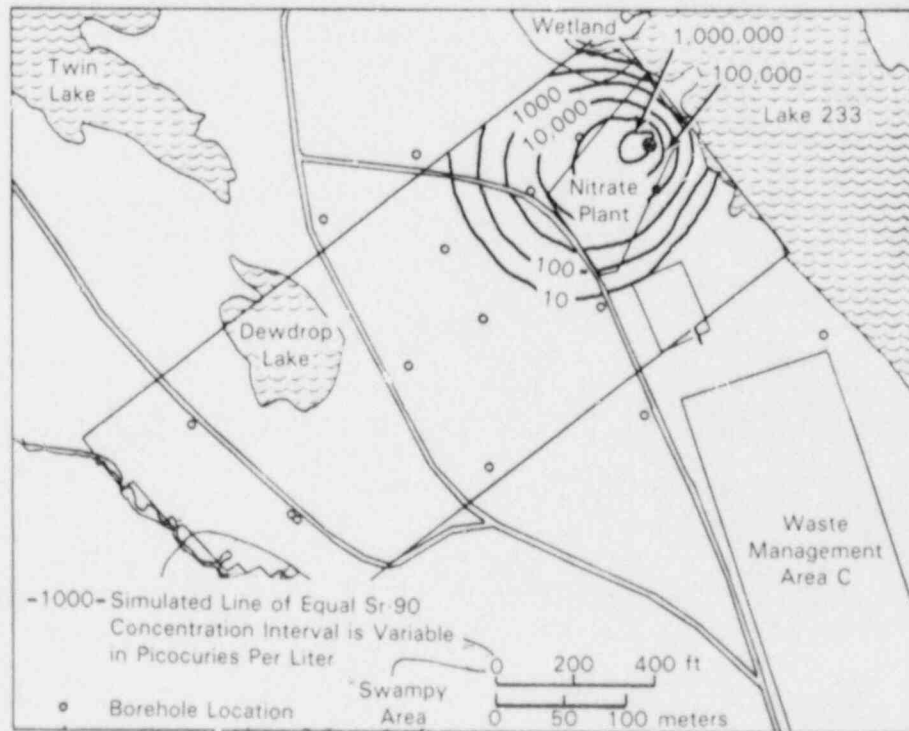
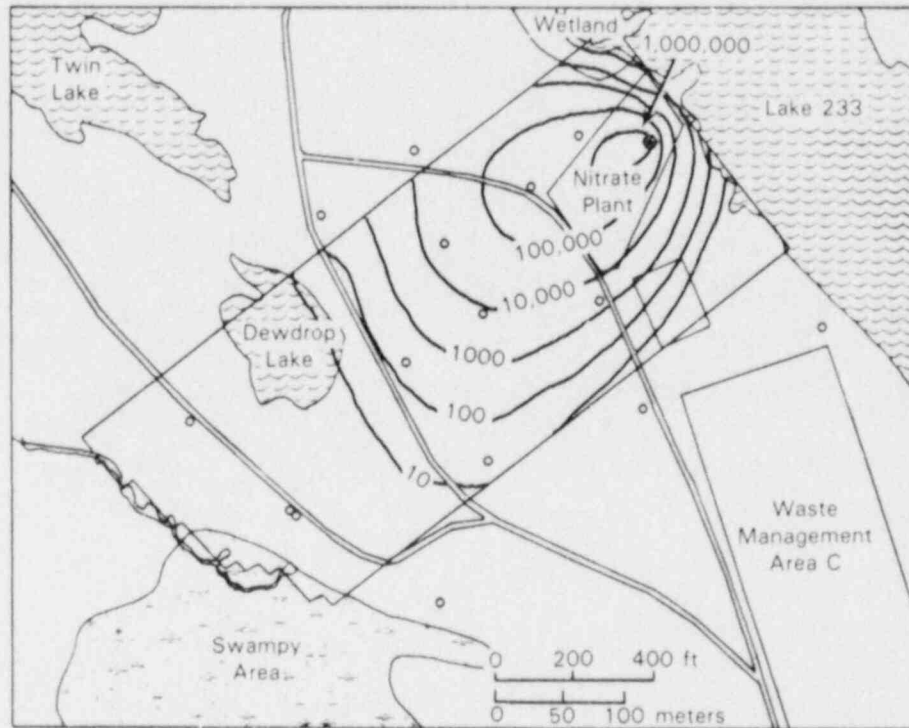


FIGURE 3.8. Strontium-90 Concentration After 30 Years Resulting from Limited-Data Simulation of a 30-Year Release, Using a Longitudinal Dispersivity of 1.0 m, a Transverse Dispersivity of 0.1 m, and Retardation Factors of 25 (Top) and 75 (Bottom)

of 25, the peak concentration under the 30-year release was about 3.6 million pCi/l, a decrease of 2.4 million pCi/l over the peak concentration for the 1-year release. However, during the remaining time, peak concentrations become increasingly larger than those simulated in the 1-year release. At the end of 30 years, peak concentrations for the 30-year release and a retardation factor of 25 was 4.6 million pCi/l, almost a factor of 10 greater than peak concentrations predicted in equivalent 1-year release simulations.

#### 3.1.4.3 Cesium-137 Transport

Migration of  $^{137}\text{Cs}$ , the other principal radionuclide expected in the waste stream, is unlikely to occur over a significant distance from the disposal pit. Because the high affinity of  $^{137}\text{Cs}$  for adsorption onto soils, its migration from the disposal pit bottom to the underlying water table would likely be slow. Estimated travel times in the unsaturated zone, based on a saturated vertical hydraulic conductivity of 3.1 m/day, a porosity of 0.35, a unit gradient, and a retardation factor of 1200 ( $K_d = 268 \text{ ml/g}$ ), suggest that  $^{137}\text{Cs}$  would arrive at the water table an average of 2.5 to 3 years after disposal. The hydraulic conductivity used in this estimate was an average vertical hydraulic conductivity value derived from permeameter tests. If unsaturated permeabilities are considered in these estimates, the estimated arrival time would be much longer. Nevertheless, because  $^{137}\text{Cs}$  is highly retarded, it would not be expected to migrate more than a few meters from the disposal pit once introduced into the shallow aquifer. Simulations made of a release of 200 Ci of  $^{137}\text{Cs}$ , using a retardation factor of 1200, resulted in cesium migration in the groundwater of only a few meters from the disposal pit after 30 years. The peak concentration simulated directly below the pit was about 70,000 pCi/l, and  $^{137}\text{Cs}$  concentrations greater than and equal to 1000 pCi/l were simulated within a few meters of the pit boundaries.

### 3.2 DETAILED DATA MODELING

A second level of modeling was done which made use of the entire extensive characterization data set collected by CRNL personnel over a 30-year period. The conceptual model developed using a limited part of the data was re-evaluated and adjusted as necessary to take advantage of the additional data and the more than 30 years of experience at the subject site.

#### 3.2.1 Effect of Additional Data on Conceptual Model

The additional information from the detailed site characterization data set was examined to reevaluate aspects of the conceptual model that would have some bearing on groundwater flow and radionuclide transport from the prospective site. Specific areas of interest would include the interface between the surficial sands and the underlying bedrock and till, the internal geometry of the facies present in the surficial sands, the hydraulic properties of the sands, and the patterns of groundwater movement inferred from water-level measurements.

### 3.2.1.1 Bedrock-Till Surface

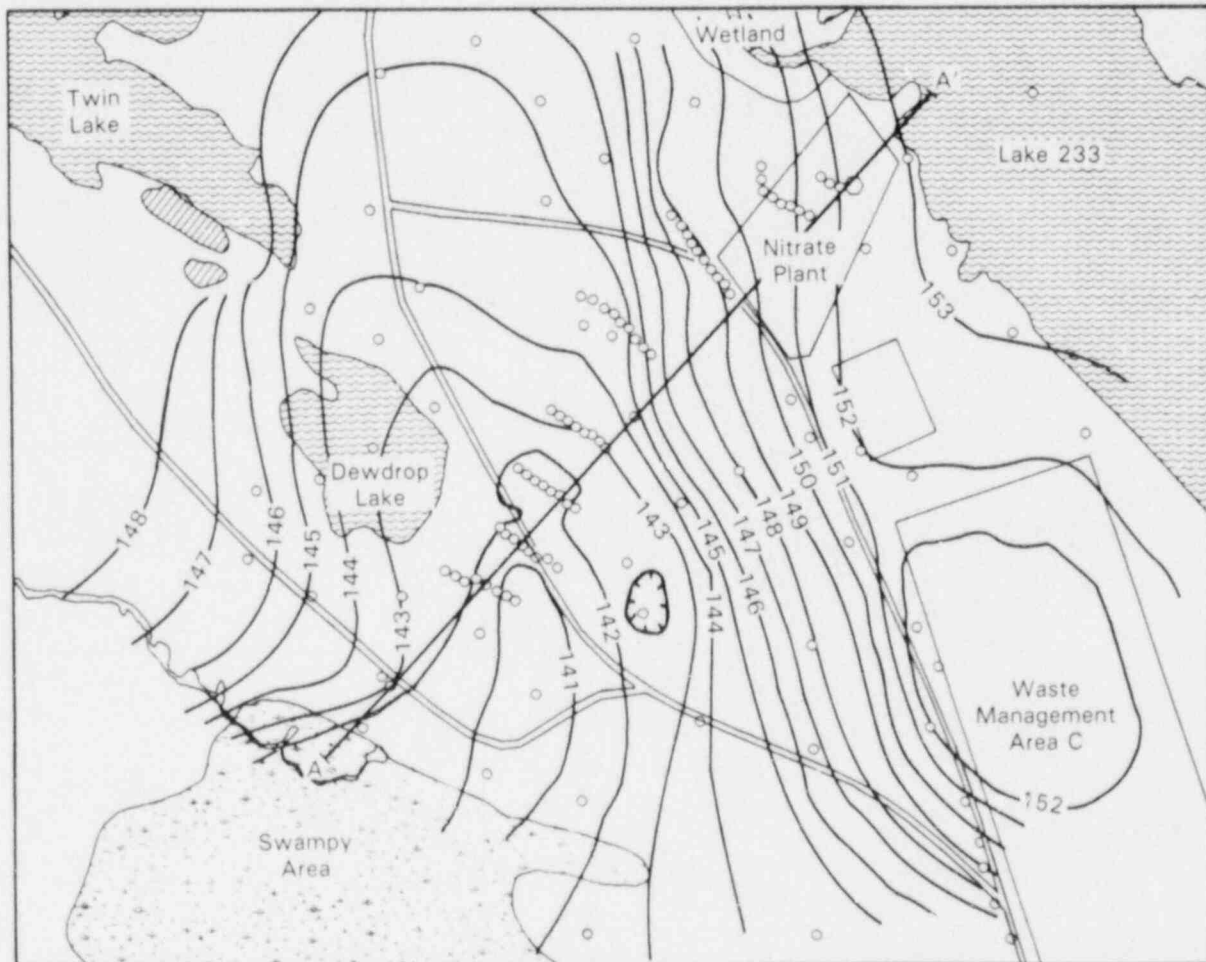
Geologic data from 104 additional boreholes were combined with data from the original 16 boreholes and information on bedrock outcrops used in the limited-data modeling to revise maps of the interface between the surficial sandy sediments and the underlying bedrock and till in the vicinity of the disposal site. The revised contour map of the bedrock and till surface (Figure 3.9) suggests that, although the additional data provides a considerable amount of detail of the till-bedrock interface, the revised surface does not provide any significant changes from the original map (Figure 2.2) which used just 16 boreholes. Both the original and the revised surfaces identified bedrock highs along the Lake 233 shoreline and in the vicinity of Dewdrop and Twin Lakes. Bedrock near Lake 233 was found to rise to an elevation of about 153 m (MSL) below about 15 m of sand and silt. Near Dewdrop and Twin Lakes, the bedrock is thinly covered and crops out at the land surface at a few locations. Between the two ridges, both surfaces provide evidence for a north-south trending depression in the bedrock surface where the bedrock decreases to an elevation of 141.5 m (MSL). However, the revised surface provides additional information that would indicate that the bedrock depression is a little wider and about 2 m deeper than was inferred from the original 16 borehole data set.

### 3.2.1.2 Internal Geometry of Surficial Sand Facies

Above the bedrock and till, the additional borehole data identified the same three major lithologic facies that were detected in the original data set. The three facies included a fine-to-very-fine sand unit, another sand unit interstratified with silt found within the shallow fine sand unit, and a fine-to-medium sand unit generally found just above the bedrock and till surface. The vertical and horizontal continuity of these sediments along a similar section line to that depicted in Figure 2.3, as interpreted by staff of the CRNL<sup>(4)</sup> using the detailed data set, is illustrated in the geologic section shown in Figure 3.10. A comparison of the section developed from the original 16 borehole data (Figure 2.3) with the CRNL section indicates that use of just the 16 boreholes provided sufficient information to approximate the general geometry of the major facies.

### 3.2.1.3 Hydraulic Properties

Hydraulic conductivity estimates by the CRNL staff at selected borehole locations from the additional detailed data were combined with results from the limited-data set to provide new estimates of hydraulic conductivity for the surficial sand aquifer. The combined averages and ranges of hydraulic conductivity estimates that were representative of the three major sand facies, are given in Table 3.2 by analysis method and lithology.



- 144- Bedrock or Till Contour Interval.  
1 Meter Dashed Where Approximately  
Located. Datum is Mean Sea Level.
- 0 200 400 ft
- 0 50 100 meters
- Borehole Location
- ▨ Area of Bedrock Outcrops
- A A' Line of Geologic Section

FIGURE 3.9. Map Showing Top Elevation of Bedrock and Till Surface from the Detailed Data Base

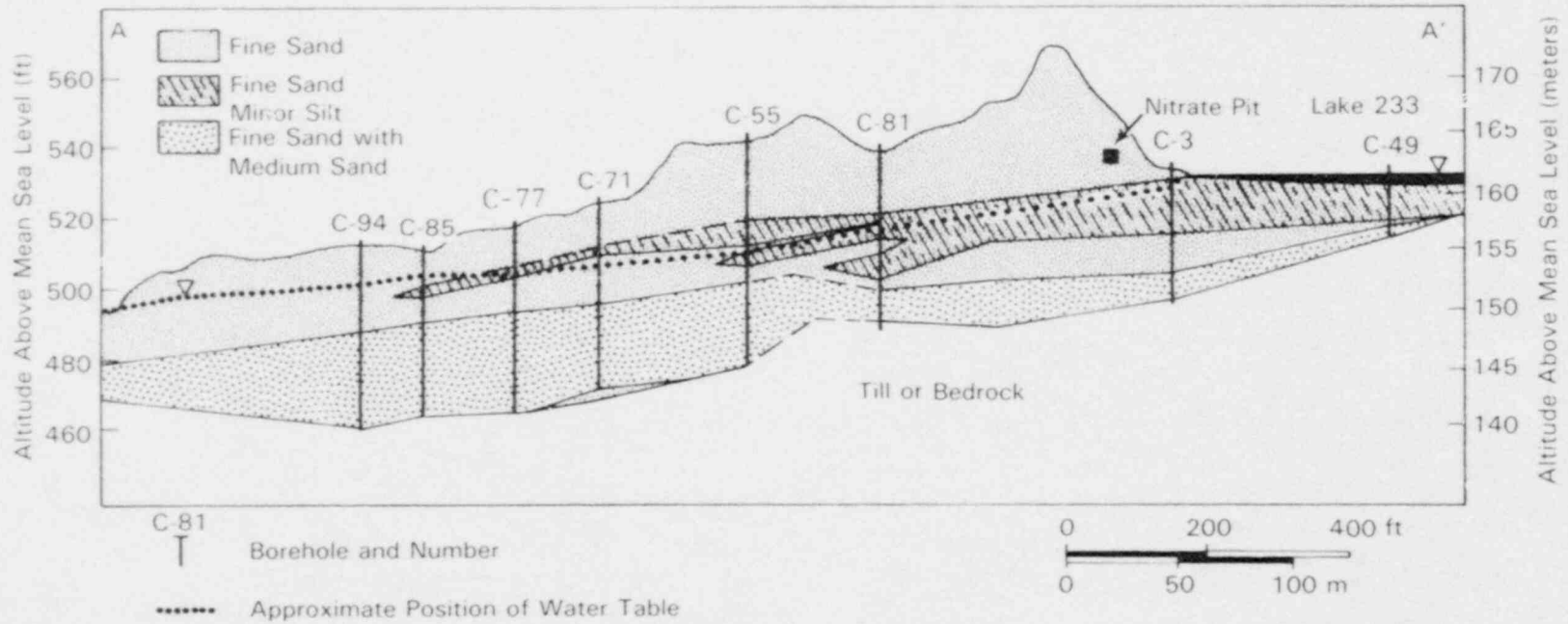


FIGURE 3.10. Geologic Section Showing Major Lithologic Units (Killey 1984), Vertical Exaggeration 5:1

TABLE 3.2. Summary of Hydraulic Conductivity Values by Hydrostratigraphic Unit and Method (meters/day)

<u>Hydrostratigraphic Unit</u>	<u>Grain Size Analysis</u>	<u>Permeameter Tests</u>	<u>Single-Well Response Tests</u>
Fine and very fine sand			
Average	5.4	3.3	1.6
Range	4.0 to 9.5	0.1 to 8.6	0.6 to 3.1
Sand interstratified with silt			
Average	-	0.49	-
Fine and medium sand			
Average	9.5	6.0	5.4
Range	6.2 to 17.3	0.95 to 14.7	1.0 to 9.5

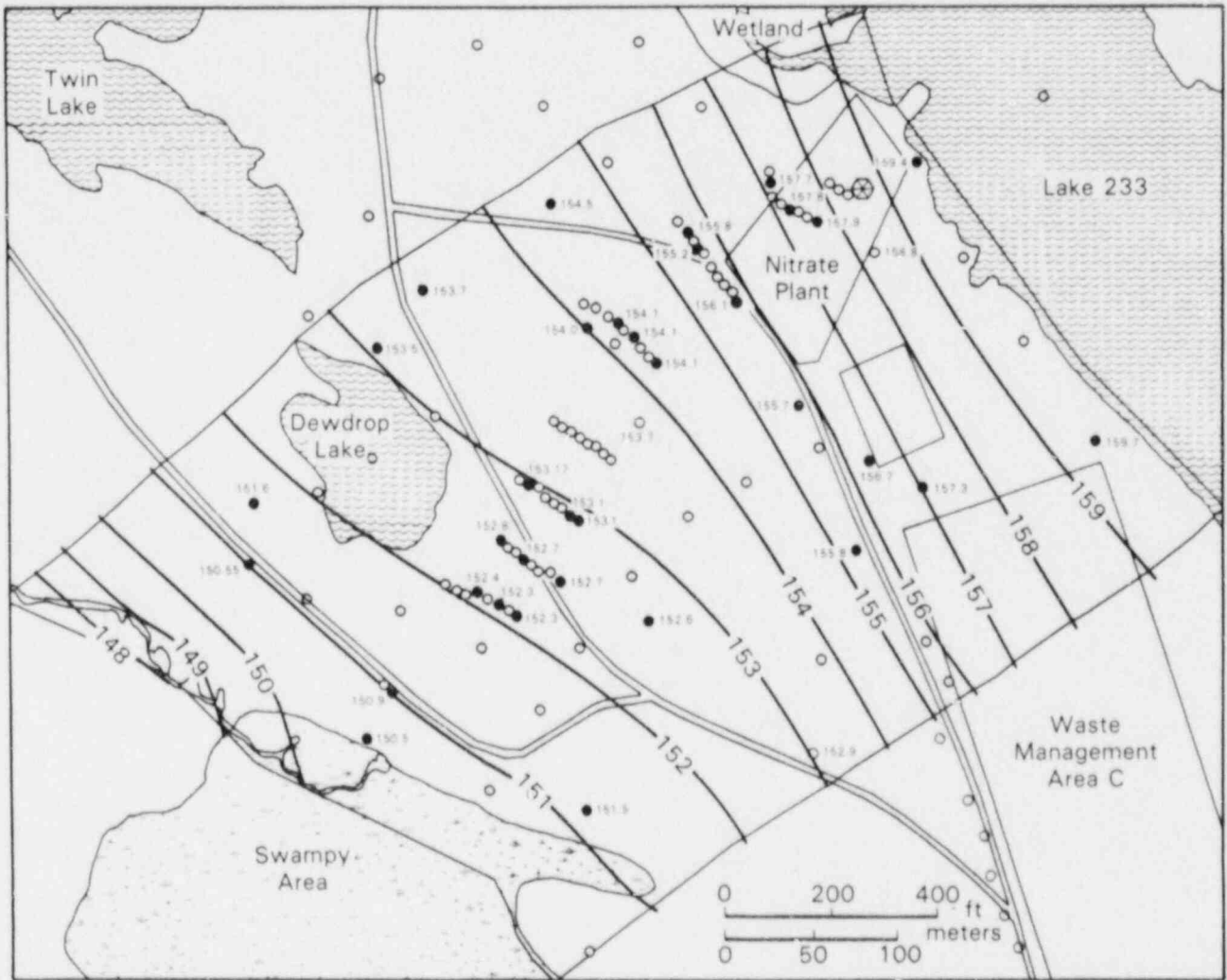
A comparison of these values with those given in Table 2.1 indicates that the additional data did not provide any significant changes in either the means or ranges of hydraulic conductivities from the original values derived in the limited data.

#### 3.2.1.4 Groundwater Movement in Surficial Sands

Water-level measurements made in more than 250 piezometers at a number of the additional 104 borehole locations over the 5-year period 1979 to 1984 were combined with water-level measurements made in the limited data to revise water-table maps of the surficial sand unit. A revised water-table map, based on measurements made in October 1984, is shown in Figure 3.11. Although the additional data provide much more detailed information on water levels along the principal flow downgradient from the disposal to the discharge area southwest of the site, groundwater flow patterns suggested by the map are nearly identical to patterns inferred from maps constructed with the limited-data set. Groundwater in the Lake 233 area and in the vicinity of the site moves laterally toward the southwest before being discharged as seepage to the swampy area lying 460 m southwest of the site. The total drop in water level from the lake to the swamp, 11 m, is about the same as determined earlier.

#### 3.2.1.5 Revised Conceptual Model of Flow and Transport

The conceptual model of flow and transport underwent only minor revisions based on the additional data. Boundary conditions in the model were essentially the same as those used in the simulations based on the limited data. With the limited-data, Lake 233 northeast of the site and the swampy area and stream channel southwest of the site were represented as constant-head boundaries. The lateral boundaries on the northwest and southeast of the model grid approximated stream lines in the water table and, as such, represented no-flow boundaries. The hydraulic relationship of the surficial sand aquifer and the underlying till and crystalline bedrock was represented in the same fashion as done in the limited-data simulations where the base of the surficial sand was simulated as a no-flow boundary.



-153- Simulated Water Level Contour  
Interval 1 Meter  
Datum is Mean Sea Level

159.4

● Water Levels Measured October 1984

FIGURE 3.11. Water Table of Surficial Sand Aquifer, October 1984

Data input for the detailed-data simulations was slightly different than data used in limited-data simulations. Using the limited data, the aquifer top was approximated from the water-table map illustrated in Figure 2.5. The aquifer bottom was approximated by a contour map of the till and bedrock surface developed from the detailed data base shown in Figure 2.2. The initial distribution of hydraulic conductivity was derived by dividing the calibrated distribution of transmissivity developed during the limited-data evaluation shown in Figure 3.3 by the aquifer saturated thickness map shown in Figure 2.4.

### 3.2.2 Model Recalibration

As in the limited-data calibration, the model was calibrated for steady-state conditions. The hydrographs in Figure 2.7 suggest that conditions in the aquifer between 1979 and 1985 have undergone only a few minor perturbations caused by stage fluctuations in Lake 233. For the most part, the aquifer has been in a period of a dynamic equilibrium.

Since many of the wells from the detailed data base had not been installed prior to September 1980, the detailed-data simulations could not use water-level measurements made in September 1980, as a basis for calibration. For detailed-data calibration, the model was used to match predicted water levels to those measured in October 1984, because a complete set of measurements in nearly all available piezometers was made on that date.

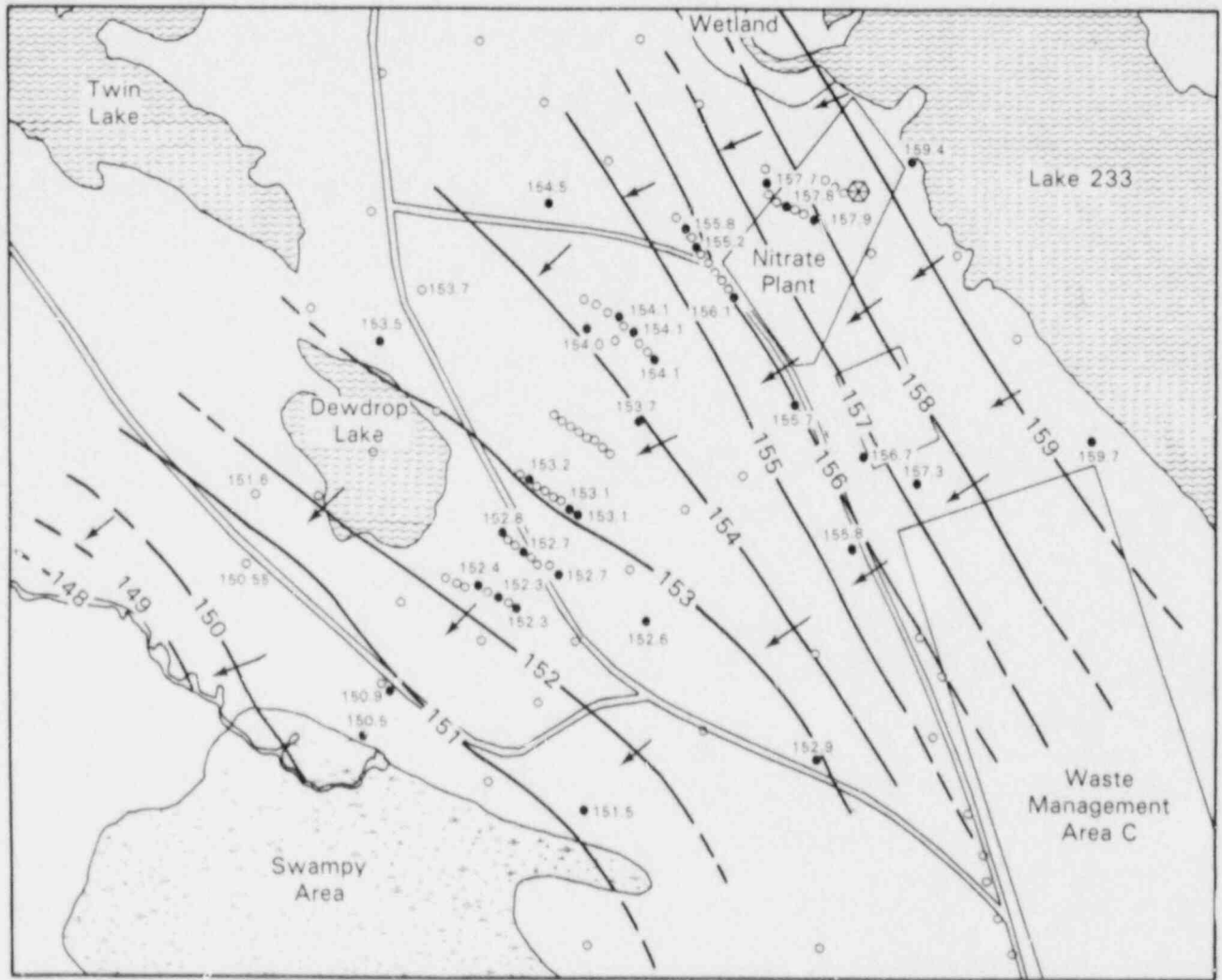
As in the limited-data calibration, input parameters were adjusted within the range of acceptable values until simulated conditions most closely approximated observed conditions. Input variables adjusted during calibration were hydraulic conductivity and recharge.

The calibrated recharge rate of 32.7 cm/yr used in the limited-data modeling and some minor adjustments to the initial hydraulic conductivity distribution provided an acceptable match to observed heads measured in October 1984. Figure 3.12 illustrates the degree of match between simulated and observed head. Most of the observed heads were matched within 0.5 m and all were matched within 0.7 m. This match was similar to the match derived from the limited-data calibration.

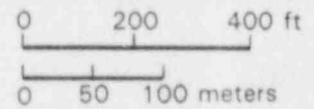
The distribution of transmissivity shown in Figure 3.13 represents the range of values resulting from calibrations of the model based on the detailed data. Final transmissivities were between 20 and 145 m<sup>2</sup>/day, with the maximum values located near the bedrock surface depression west and southwest of the proposed site. Hydraulic conductivity values, derived by dividing the calibrated transmissivity distribution by the saturated thicknesses given in Figure 2.4, were not significantly different than those derived in the initial modeling ranging from 4 and 15 m/day. The increase in transmissivity from the limited-data calibration was mainly a function of the increase in saturated thickness derived from the updated bedrock-till surface.

The water budget of the calibrated model presented in Table 3.3 indicated that infiltration from precipitation, which amounted to 1.569 x 10<sup>2</sup> m<sup>3</sup>/day,



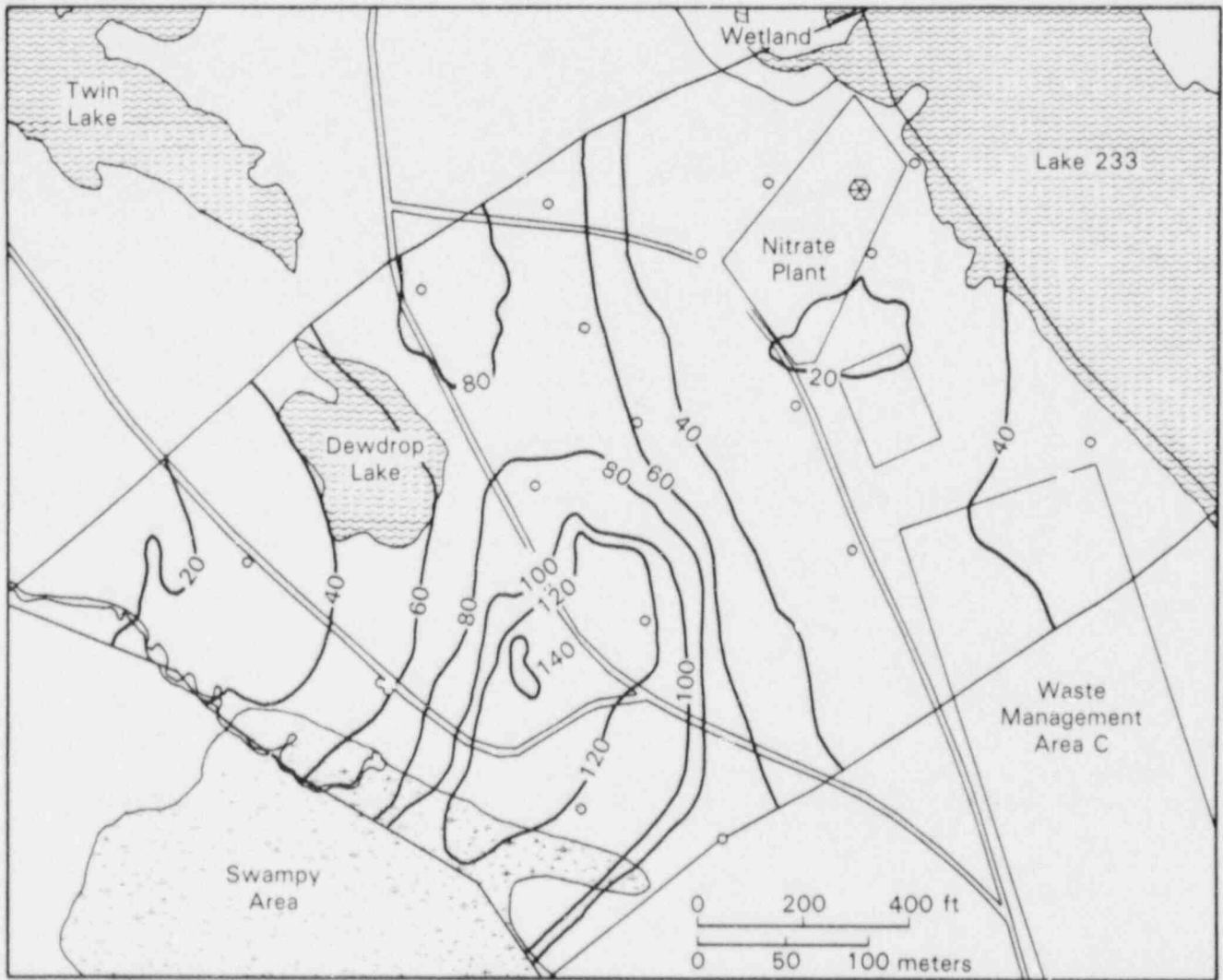


-152- Water Table Contour  
Interval 1 Meter  
Datum is Mean Sea Level



- Borehole and Water Level  
151.5
- ← Direction of Ground-water Flow

FIGURE 3.12. Comparison of Simulated and Observed Head in the Detailed Data Calibrated Steady-State Groundwater Flow Model of October 1984 Conditions



-100- Line of Equal Transmissivity  
Interval 20 meter<sup>2</sup>/day

C-26  
○ Borehole and Number

FIGURE 3.13. Transmissivity Distribution in the Detailed-Data Calibrated Steady-State Groundwater Flow Model of September 1980 Conditions

TABLE 3.3. Water Budget from the Calibrated Steady-State Groundwater Flow Model for October, 1984 Conditions. Values are in cubic meters/day.

Sources of Recharge	
Infiltration of precipitation	1.570 x 10 <sup>5</sup>
Lake 233 constant-head boundary	3.126 x 10 <sup>5</sup>
Total recharge	4.696 x 10 <sup>5</sup>
Sources of Discharge	
Swampy area and stream channel constant-head boundary	4.696 x 10 <sup>5</sup>
Total discharge	4.696 x 10 <sup>5</sup>

accounted for about 36% of the total recharge to the simulated area. The remainder of the recharge,  $2.77 \times 10^5 \text{ m}^3/\text{day}$ , was derived from the Lake 233 boundary. All discharge at the swampy area and the stream channel boundary along the southwest part of the model was simulated. This budget is similar to the budget calculated for limited-data simulations.

### 3.2.3 Radionuclide Transport Modeling

The calibrated steady-state flow model based on the detailed data was used to provide the hydrologic framework for remodeling the limited-data transport simulations made in the early part of this study. This modeling was redone to evaluate the effect of using the detailed site data on initial model results. This modeling used the same grid, boundary conditions, and release scenarios used in the limited-data simulations. The following discussion focuses on results from the simulations of <sup>90</sup>Sr transport because previous results indicated that other radionuclide transport would not be significant.

#### 3.2.3.1 Strontium-90 Transport

The three waste release scenarios simulated with the limited data were reproduced using the detailed-data calibrated steady-state flow model. These scenarios are as follows:

- one-year release of 1000 curies of <sup>90</sup>Sr using a constant retardation factor
- one-year release of 1000 curies of <sup>90</sup>Sr using a retardation factor that varies in space from 2 to 25
- thirty-year release of 1000 curies of <sup>90</sup>Sr using a constant retardation factor.

In general, simulated results of these waste-release scenarios were nearly identical to simulated results of the same scenarios derived from the limited-data model.

Model results from the 1-year release with a constant retardation factor are presented in Figure 3.14. With a retardation factor of 25, the model results at the end of the simulation indicated that groundwater containing 10 pCi/l had migrated downgradient about 335 m from the disposal site. Concentrations in groundwater directly beneath the disposal pit were about 14,800 pCi/l. Peak concentrations of about 415,000 pCi/l were simulated about 68 m downgradient from the site. With a retardation factor of 75, concentrations of about 10 pCi/l were simulated about 180 m downgradient from the pit. Concentrations directly beneath the pit were about 1 million pCi/l. Peak concentrations of about 1.3 million pCi/l were simulated about 50 m downgradient from the disposal pit.

Model results from a 1-year release with a variable retardation factor are presented in Figure 3.15. A comparison of these results with those from the other simulations indicated that, as expected, early migration of  $^{90}\text{Sr}$  was more rapid. For this particular set of retardation factors,  $^{90}\text{Sr}$  concentrations of 10 pCi/l moved downgradient about 160 m after 4 years, 225 m after 10 years, and 380 m after 30 years. Peak concentrations were similar to those predicted for the first scenario but had migrated a significantly longer distance from the disposal pit. After a period of 30 years, concentrations of about 5,000 pCi/l were simulated at the disposal pit area. Peak concentrations of about 680,000 pCi/l were simulated about a distance of 140 m downgradient from the pit.

Model results from a 30-year inventory release are presented in Figure 3.16. A comparison of these results with those in Figure 3.14 shows that the overall transport of the plume is about the same for both scenarios. However, concentrations within the plume have a distinctly different distribution. Unlike the 1-year release where the peak concentration migrates away from the source area, the peak concentration during the 30-year release generally remained in close proximity to the disposal pit area. The magnitude of peak concentrations during the first 4 to 6 years of the simulation were, as expected, generally smaller than peaks predicted for the first year release. After 4 years of release and a retardation factor of 25, the peak concentration from the 30-year release was about 3.6 million pCi/l, a decrease of 2.4 million pCi/l over the peak concentration for the first year release. However, during the remaining time, peak concentrations for the 30-year release become increasingly larger than those simulated in the first year release. At the end of 30 years, peak concentrations for the 30-year release simulation using a retardation factor of 25 were 4.6 million pCi/l, almost a factor of 10 greater than peak concentrations predicted in equivalent first year simulations.

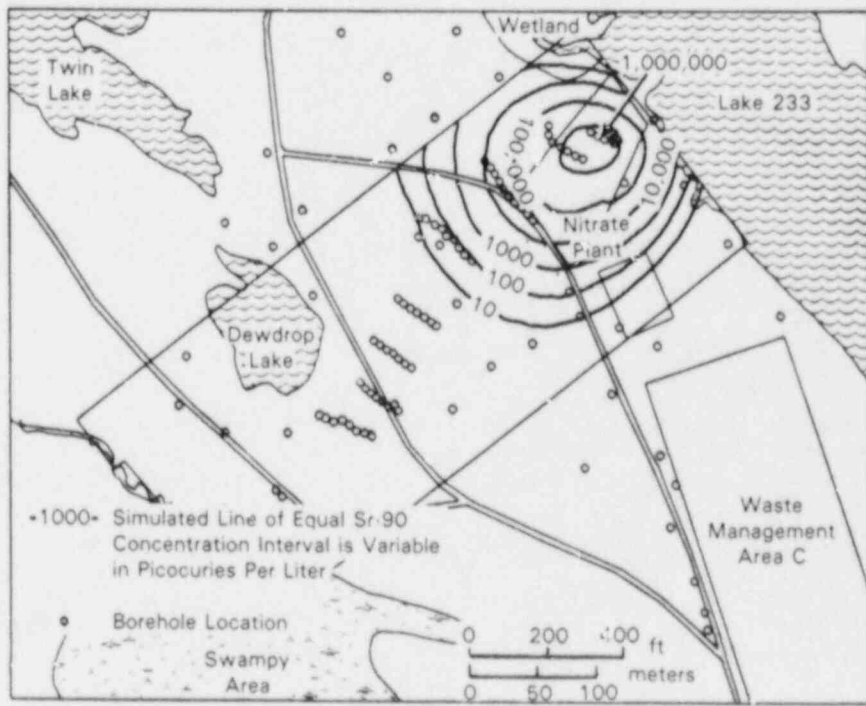
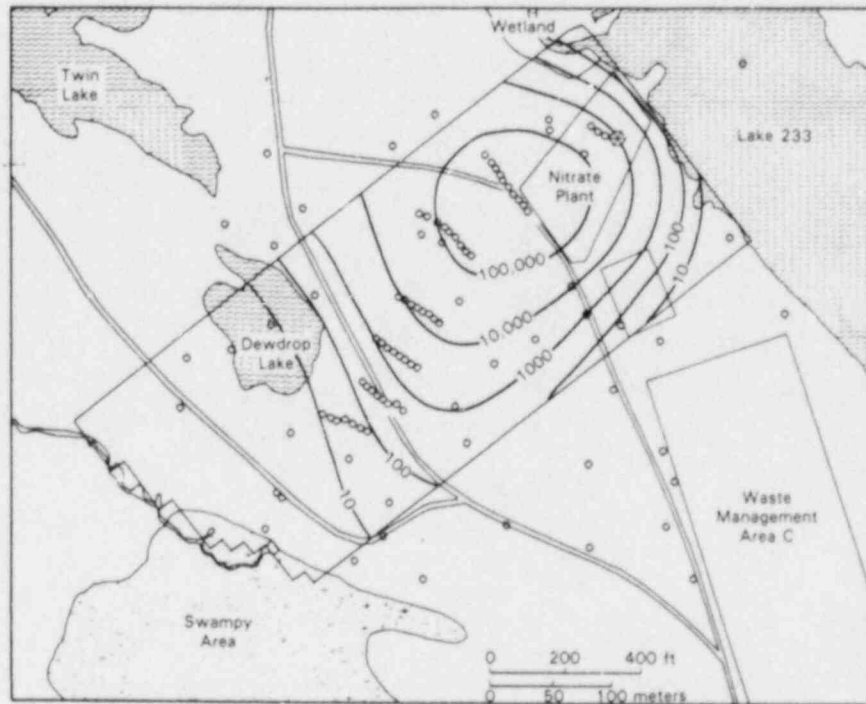
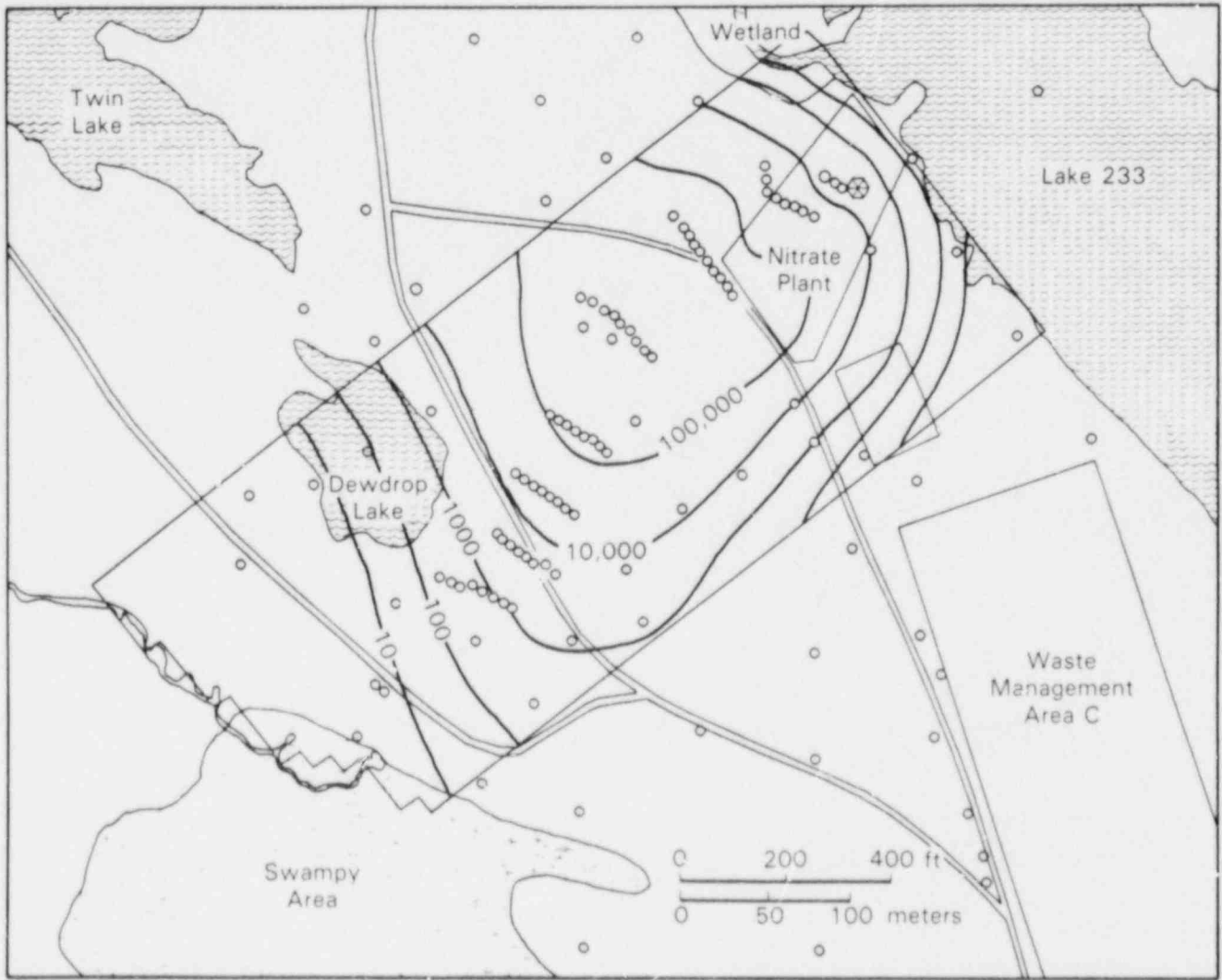


FIGURE 3.14. Strontium-90 Concentrations After 30 Years Resulting from the Detailed-Data Simulation of a 1-Year Inventory Release, Using a Longitudinal Dispersivity of 1.0 m, a Transverse Dispersivity of 0.1 m, and Retardation Factors of 25 (Top) and 75 (Below)



-1000- Simulated Line of Equal Sr-90  
Concentration Interval is Variable  
in Picocuries Per Liter

○ Borehole Location

**FIGURE 3.15.** Strontium-90 Concentrations After 30 Years Resulting from the Detailed-Data Simulation of a 1-Year Inventory Release, Using a Longitudinal Dispersivity of 1.0 m, a Transverse Dispersivity of 0.1 m, and Variable Distribution of Retardation Factors Ranging from 10 to 25

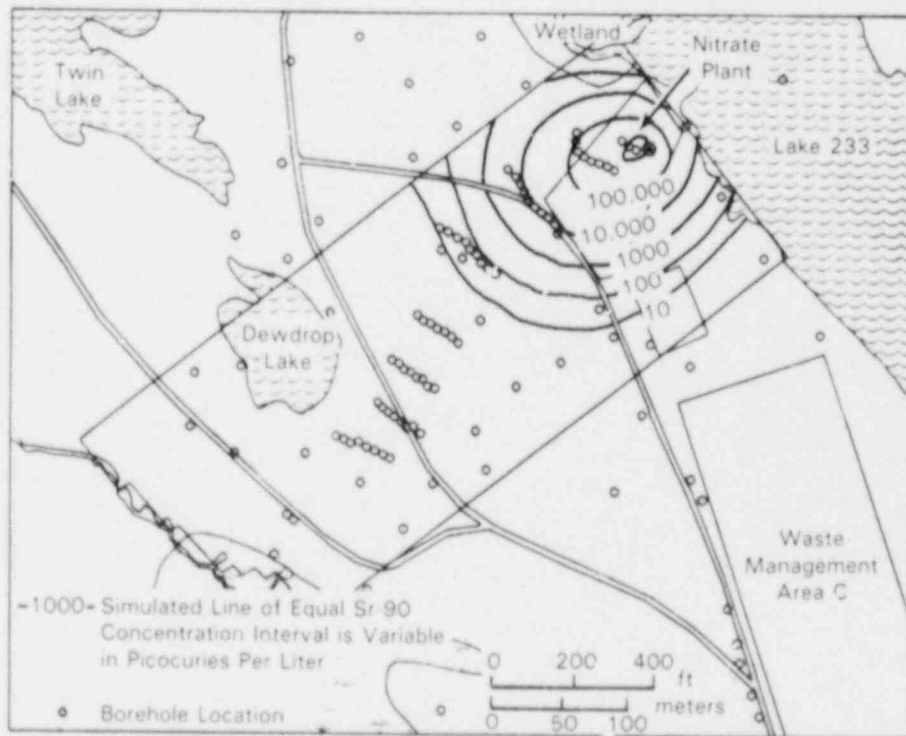
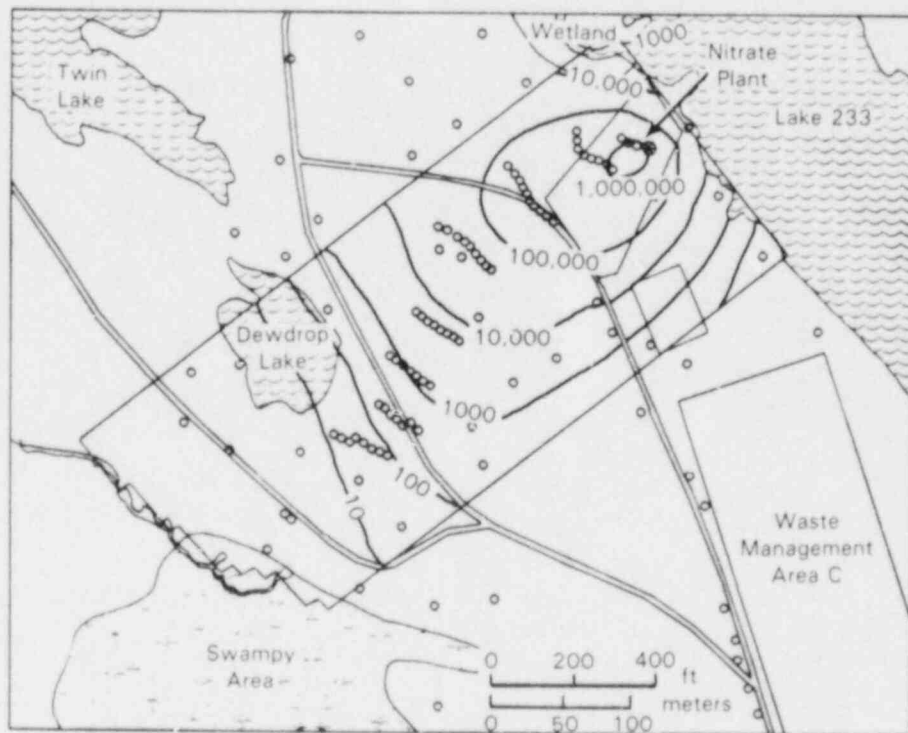


FIGURE 3.16. Strontium-90 Concentrations After 30 Years Resulting from the Detailed-Data Simulation of a 30-Year Release, Using a Longitudinal Dispersivity of 1.0 m, a Transverse Dispersivity of 0.1, and Retardation Factors of 25 (Top) and 75 (Bottom)

#### 4.0 FIELD SAMPLING, MEASUREMENT AND MAPPING OF RADIONUCLIDE MIGRATION

Previous monitoring and hydrogeochemical studies performed by CRNL staff at the Nitrate Disposal Pit Site have defined the early radionuclide migration. In October 1984, a joint PNL/CRNL field sampling and measurement program was conducted at the Nitrate Disposal Pit site to determine the actual radionuclide migration that had occurred during the past 30 years since disposal of radioactive wastes at this site. These measurements, together with extensive groundwater sampling, gross beta, and  $^{90}\text{Sr}$  analyses conducted by R.W.D. Killey of CRNL, have provided the basis for determining the actual dimensions of the migrating radionuclide plume.

#### 4.1 BACKGROUND INFORMATION

Details of the past low-level waste management activities and of the past and current environmental monitoring and geochemical studies conducted at the Nitrate Disposal Pit site at CRNL are presented in this section. At this site, mixed fission products were released to an infiltration pit beginning in 1953. Monitoring of contaminant movement in the groundwater began in 1955<sup>(6)</sup> and periodic measurements have continued to map the position of the  $^{90}\text{Sr}$  plume front to the present time. Geochemical studies conducted at or near the site have helped elucidate the retardation mechanisms afforded by the soil<sup>(7,8,9,10)</sup>. The extensive array of multi-level monitoring at this site has permitted a very detailed mapping of the  $^{90}\text{Sr}$  plume migrating in the groundwater from the disposal pit. Thus, this site affords an excellent opportunity to compare model/code predicted radionuclide transport with actual migration that has occurred during the past 30 years.

During the operation of the Nitrate Decomposition Plant, mixed fission products were released to the infiltration pit beginning in 1953. Among these were  $^{90}\text{Sr}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ , and  $^{144}\text{Ce}$  (see Section 4.2). Monitoring of contaminant movement began in 1955 using geiger counter scans of a series of dry wells, and by 1955 activity had migrated 75 m from the pit.<sup>(6)</sup> By 1957, radiostrontium was detected 150 m from the source, and  $^{106}\text{Ru}$  was detected in the groundwater discharge area. Between 1957 and 1961, the  $^{90}\text{Sr}$  plume front advanced only an additional 50 m, and a detailed map of the radiostrontium plume was constructed based on approximately 600 soil samples collected from 45 boreholes<sup>(6)</sup>. The subsurface inventory calculated from that survey was 800 Ci, in good agreement with the estimated input and implying that all of the  $^{90}\text{Sr}$  had entered the flow system. That survey also showed that 95% of the  $^{90}\text{Sr}$  had migrated less than 160 m; the center of mass of the plume was not advancing as quickly as the leading edge.

The position of the plume front was determined in 1966 and 1971 (W. F. Merritt, unpublished data), and the entire plume was mapped in detail in 1983. The total inventory of  $^{90}\text{Sr}$  calculated from the 1983 survey (and decay-corrected back to 1954) was 680 Ci. The center of mass of the plume in the 1983 data is slightly less than 100 m from the source, although the leading edge has migrated 335 m.



There are several sources of distribution coefficient ( $K_d$ ) data for  $^{90}\text{Sr}$  that can be applied to the Nitrate Plant plume. Conventional laboratory batch testing of sediments collected adjacent to the plume and local groundwater yielded a mean  $K_d$  of  $13.8 \pm 8.4$  ml/g <sup>(7)</sup>. This is similar to other batch measurements of CRNL sands <sup>(8)</sup>. Distribution coefficients can also be calculated for the plume front using the retardation equation. From measured and calculated groundwater velocities and the data on plume front position since 1961, a consistent  $K_d$  of 2 ml/g is obtained. The third source of  $K_d$  information is the samples from the plume itself. Cores obtained during drilling were sectioned and centrifuged to extract pore water. Both sands and waters were counted for  $^{90}\text{Sr}$ , providing in situ values for  $K_d$ . Results of the in situ tests provided a very large range of  $K_d$ 's (1 to 110 ml/g), with a mean value of near 50 ml/g near the center of the mass and decreasing to about 20 ml/g near the front of the plume.

The cause of the discrepancy between the laboratory and in situ  $K_d$ 's is the presence of multiple mechanisms of  $^{90}\text{Sr}$  adsorption. Short-term contact between solutions containing  $^{90}\text{Sr}$  and sediments results in sorption that is truly ion exchange and readily reversible. Longer-term contact results in additional removal of  $^{90}\text{Sr}$  by chemisorption <sup>(7)</sup>. Studies by Melnyk, et al., <sup>(8)</sup> have observed chemisorption of  $^{90}\text{Sr}$  in long-term laboratory experiments. They proposed a first order reaction with a half-time of 2 years when fitting a combined ion exchange/chemisorption model to data from the CRNL glass block experiment <sup>(9)</sup>.

The rapid migration of  $^{90}\text{Sr}$  in the first 2 years after disposal has been attributed to the high ionic strength and calcium concentrations in the aqueous waste following neutralization in the limestone-lined infiltration pit. Chromatographic separation of the calcium and strontium <sup>(10,11)</sup> determined a strontium/calcium selectivity coefficient of 1.3 for Chalk River sands and may be the reason for the decrease in  $^{90}\text{Sr}$  migration for the bulk of the plume. The relatively high velocity of the plume front may be a result of residual enrichment of calcium on the soil exchange sites <sup>(7)</sup>.

Although  $^{137}\text{Cs}$  was a major component of the waste discharged to the infiltration pit, almost all of it has moved less than 15 m during the past 30 years. Sampling in 1984 did detect trace (<10 pCi/l) quantities of  $^{137}\text{Cs}$ , 100 and 400 m from the source, as well as very low levels (<5 pCi/l) of  $^{60}\text{Co}$  and  $^{154}\text{Eu}$ . No  $^{106}\text{Ru}$  or  $^{144}\text{Ce}$  was detected (limit 0.02 pCi/l), both of which would have been reduced to  $1 \text{ E-}9$  of their input inventories by radioactive decay.

Recent field measurements performed by R.W.D. Killey of CRNL have further defined the present  $^{90}\text{Sr}$  migration plume. These measurements have been supplemented by a joint PNL/CRNL field measurement program conducted in October 1984 to further define movement of  $^{90}\text{Sr}$  and other long-lived radionuclides in this slightly contaminated groundwater plume.

## 4.2 RADIONUCLIDE SOURCE TERM

During 1953 and 1954, approximately 3780 liters of radioactive liquid were discharged into the soil at the Nitrate Disposal Pit site in the low-level waste disposal area of the Chalk River Nuclear Laboratories.<sup>(6)</sup> This waste came from a small ammonium nitrate decomposition plant in which solutions of aged waste fission products were treated to remove large quantities of dissolved ammonium nitrate and were then concentrated by evaporation. A small pit (55 m<sup>2</sup>) had been excavated close to the Nitrate Plant and partially filled with crushed limestone to receive the condensate from the decomposer unit. The pit was constructed so that its base was within 7 to 8 m above the average water-table elevation. The limestone lining helped neutralize the acidified waste solutions before they entered the underlying sand layers. After one year's operation the process was halted. By this time the pit had absorbed much more liquid than had been anticipated, owing to spillage and malfunction of the equipment.

During 1954, a number of radioactive spills from the decomposer unit were cleaned by hosing the contaminated area and allowing the washings to drain into the disposal pit. Under these conditions, precise measurements of the waste disposal were not possible although the relative proportions of <sup>90</sup>Sr and <sup>137</sup>Cs were known for the larger spills.

The total activity from all the waste solutions was estimated to be between 1000 and 1500 Ci of mixed fission products, of which 700 to 1000 Ci were <sup>90</sup>Sr and 200 to 300 Ci were <sup>137</sup>Cs. The solutions contained acids and complexing agents, although the concentrations and total quantities of these constituents discharged to the pit have not been documented.

## 4.3 FIELD SAMPLING

The Environmental Research Branch of CRNL has been monitoring the movement of radionuclides in the groundwater near the Nitrate Disposal Pit for many years. A large number of monitoring wells have been installed at this site for this purpose. Recently, CRNL installed additional monitoring wells to complement the existing set, and performed extensive groundwater sampling to characterize the slightly radioactive groundwater plume. The groundwater was analyzed for gross beta activity and some samples for <sup>90</sup>Sr. In October 1984, a joint PNL/CRNL large-volume groundwater sampling program was conducted at this site to measure the 10CFR61 radionuclides. Seven monitoring wells (see Figure 4.1) were sampled by processing 174 to 227 liters of groundwater through Battelle Large-Volume Water Samplers (BLVWS). These samplers have been described in detail elsewhere.<sup>(17)</sup>

The seven wells chosen for large-volume water sampling were located at the approximate center line of the gross beta (essentially due to <sup>90</sup>Sr) plume originating from the Nitrate Disposal Pit. These wells included C-125, C-119, C-27, C-69, C-89, C-54, and C-3 (a well in the opposite direction of the main plume). Groundwater was sampled by connecting to in-place piezometers and pumping the water to the surface with a self-priming, bellows-type

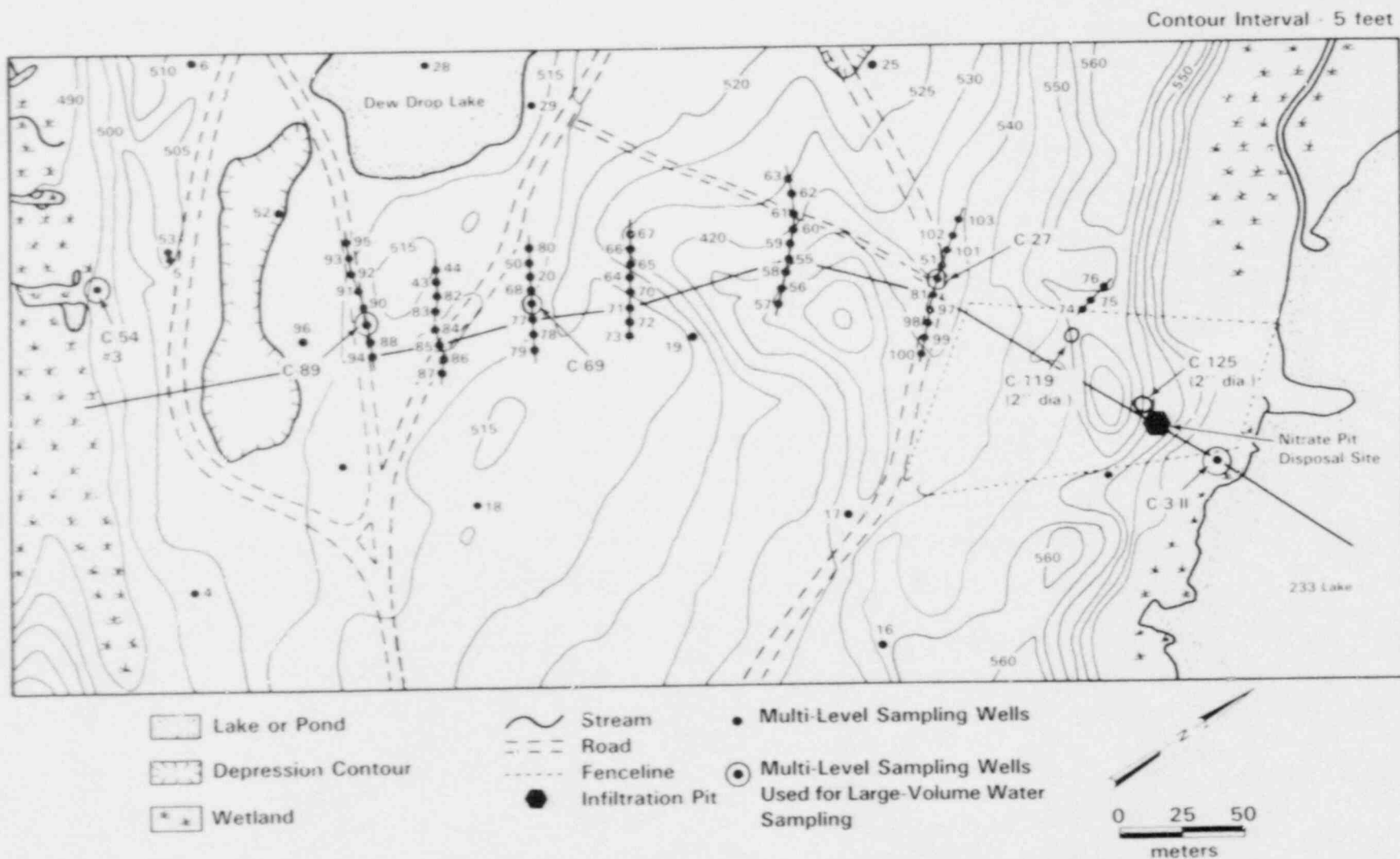


FIGURE 4.1. Large Volume Groundwater Sampling Locations in the Nitrate Pit Disposal Site Plume, October 1984

metering pump (Gorman-Rupp) having wetted parts made of polypropylene (see Figure 4.2). After pumping the piezometers for about 30 minutes to clear each sampling point, the water was then pumped through an acrylic in-line filter assembly loaded with a 30-cm diameter, 0.4  $\mu\text{m}$  Nuclepore polycarbonate filter for removal of particulate radionuclides. The filtered water then passed through a large-volume water sampler designed at Battelle, Pacific Northwest Laboratories to remove cationic, anionic, and uncharged soluble species of the radionuclides from the groundwater. This sampler consisted of three duplicate adsorption beds through which the water flows in the following order: 1) dual 20-cm diameter x 2.5-cm-thick cation resin beds (Dowex 50 x 8,  $\text{H}^+$  form, 200-400 mesh); 2) dual 20-cm diameter x 2.5-cm-thick anion resin beds (Dowex 1 x 8,  $\text{Cl}^-$  form, 200-400 mesh); and 3) dual 20-cm diameter by 0.6-cm-thick beds of activated aluminum oxide. The cation and anion resins quantitatively remove soluble radionuclides in cationic and anionic forms, respectively. The activated aluminum oxide removes radionuclides existing in soluble, nonionic forms. Each bed is constrained by a sharkskin filter and a glass fiber filter. The effluent from each sampler was collected in 250-liter plastic-lined barrels and then pumped through an integrating water-flow meter to measure the total volume processed through the sampler. After the groundwater sample had been pumped through the sampling unit, 10 liters of fresh, filtered water was pumped through the sampler to rinse out the interstitial groundwater from the resin and aluminum oxide beds.

Following the sampling, the samplers were disassembled in the field (Figure 4.3) and the resin and aluminum oxide beds and the prefilters were individually packaged and returned to the laboratory for nondestructive gamma-ray spectrometry, followed by detailed radiochemical analyses for low-energy photon, beta, and alpha-emitting radionuclides of interest on selected samples.

In addition to the radionuclide sampling, various important groundwater constituents and parameters were measured. These measurements are described in some detail in Section 4.5.

#### 4.4 RADIONUCLIDE ANALYSES

The filters, resins, and aluminum oxide were packaged in standard counting geometries and counted on large Ge(Li) gamma-ray spectrometers. The counting intervals ranged from 300 to 1000 minutes. The gamma-ray spectra were analyzed using a computer program that searches for and sums peak area, subtracts background areas, applies decay corrections and volume factors, and calculates an error term based upon background and peak counting rates.

Following the gamma-ray spectrometry, aliquots of the filters, resins, and aluminum oxide were leached with appropriate acid mixtures to remove the radionuclides requiring radiochemical separations. The acid solutions were then subjected to radiochemical procedures for the radionuclides of interest. Strontium-90 was analyzed in all seven samples, and a comprehensive 10CFR61 radionuclide analysis was performed on ion-exchange resin samples from the well (C-119-2) closest to the Nitrate Disposal Pit having the highest gross beta and  $^{90}\text{Sr}$  activity.



FIGURE 4.2. Groundwater Sampling at Chalk River Nuclear Laboratories Using the Battelle Large-Volume Water Sampler

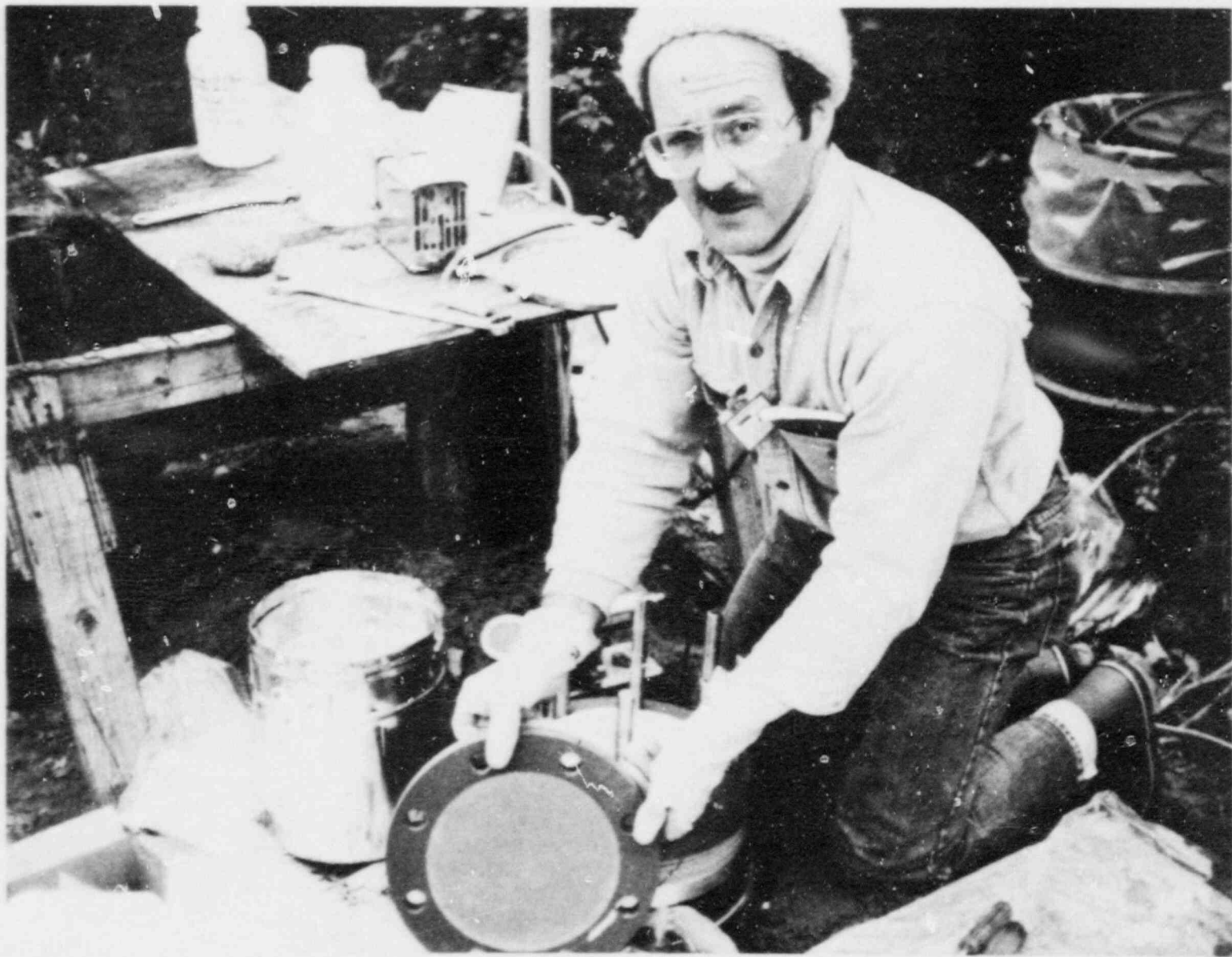


FIGURE 4.3. Disassembly of the Battelle Large-Volume Water Sampler Showing One of the Ion Exchange Resin Beds

The procedures used for the radiochemical analyses are briefly described as follows:

Strontium-90 - Strontium-85 was added as a yield tracer and the radiostrontium was purified by consecutive carbonate and fuming nitric acid precipitations. After an ingrowth period for  $^{90}\text{Y}$ , the yttrium was separated by oxalate precipitation and determined by counting in a thin window beta proportional counter. The  $^{90}\text{Y}$  was confirmed by beta-decay measurements, and the  $^{90}\text{Sr}$  calculated from the  $^{90}\text{Y}$  daughter measurements.

Technetium-99 - Technetium-95 was added as a yield tracer, and the technetium was purified by repeated iron hydroxide scavenging, followed by anion exchange separation. The technetium was then electrodeposited onto a copper disc and  $^{99}\text{Tc}$  measured in a thin window beta proportional counter and confirmed by beta-absorption analysis.

Iodine-129 - Iodine-131 was added as a yield tracer and the radioiodine was purified by anion exchange and solvent extraction with  $\text{CCl}_4$ . The purified iodine was precipitated as cuprous iodide and the  $^{129}\text{I}$  determined by measuring the 29 keV Xe  $K\alpha$  x-rays on a thin window intrinsic germanium detector.

Plutonium-238, 239, 240 - Plutonium-242 was added as a yield tracer, and the plutonium was purified by anion exchange separation and electrodeposited on a stainless steel disc. Plutonium isotopes were determined by counting in silicon surface barrier detectors.

Iron-55 - Iron-59 was added to the filtered and leached samples as a yield tracer and the radioiron was purified by hydroxide precipitation and anion exchange separations. The purified iron was electrodeposited onto a copper disc and  $^{55}\text{Fe}$  determined by measuring the 5.9 keV Mn  $K\alpha$  x-rays using a thin window intrinsic germanium diode.

Nickel-59, 63 - Nickel-65 was added as a yield tracer, and the radionickel purified by hydroxide and dimethylglyoxime precipitation. The purified radionickel was electrodeposited on a stainless steel disc and  $^{63}\text{Ni}$  determined by beta counting on a windowless, anticoincidence shielded beta proportional counter. The  $^{59}\text{Ni}$  was determined by measuring the 6.9 keV Co  $K\alpha$  x-rays using a thin window intrinsic germanium diode.

#### 4.5 CHEMICAL ANALYSIS OF GROUNDWATER

In addition to the radionuclide measurements, groundwater samples were analyzed in the field at the time of sampling for those parameters and constituents requiring immediate measurement. These included temperature, pH, Eh, dissolved oxygen, alkalinity and  $\text{Fe}^{+2}/\text{Fe}^{+3}$ . Also, separate water samples were collected from each site for trace elements, anions, sulfide, and dissolved organic constituents.

Temperature was measured on a flowing water sample with a mercury thermometer. The pH was measured immediately after sampling, using a Ross (Orion Research) combination pH electrode connected to an Orion Model 407A/F Specific Ion Meter. Calibration solutions buffered by pH 7.0 and 4.0 were used to calibrate the pH meter immediately before measuring the pH of the water samples.

The Eh of the water was measured by inserting a standard calomel electrode and a platinum inlay electrode (Corning) into a lucite cell through which the water was allowed to flow at several hundred ml·min<sup>-1</sup>. The electrodes were connected to an Orion Model 407 A/F Specific Ion Meter. Immediately after making the Eh measurement, the electrodes were immersed in Zobell's solution as a check against the platinum-calomel electrode pair.

Dissolved oxygen and alkalinity were measured using a Hach Digital Titrator Kit. Dissolved oxygen was measured by the Winkler method with azide modification and a 200-ml sample size. Titrations were performed with the Hach digital titrator (Hach Company) and a prestandardized solution of phenylarsine oxide. Alkalinity was titrated to pH 4.8 using the digital titrator and prestandardized 0.1600 N H<sub>2</sub>SO<sub>4</sub>.

At each site, a 0.4 µm Nuclepore filtered water sample (2000 ml) for trace element analysis was collected in a carefully acid-cleaned polyethylene bottle, and 20 ml of Ultrex hydrochloric acid (J. T. Baker Chemical Co.) was added as a preservative. This sample was also used to measure the ferrous/ferric iron concentrations in the water within a few days after sampling.

Also at each site, a 250-ml Nuclepore filtered water sample was collected and stored in a pre-cleaned polyethylene bottle for anion analysis by ion chromatography, a 2000-ml water sample (filtered through a silver filter) was collected and stored in a pre-cleaned teflon bottle for analysis of dissolved organic constituents, and a 250-ml Nuclepore filtered water sample was collected in a brown glass bottle and preserved with an alkaline antioxidant for analysis of sulfide. A fresh stock of alkaline antioxidant preservative was prepared by adding 7.2 g ascorbic acid and 18.6 g of disodium dihydrogen EDTA to 100 ml of 12% NaOH. Twenty-five milliliters of this solution were added to 250 ml of each water sample.

#### 4.6 RESULTS OF RADIONUCLIDE AND CHEMICAL ANALYSES OF GROUNDWATER

This section describes the results of the radionuclide analyses of the large volume water sample collected from the Nitrate Disposal Pit plume. It also describes the chemical properties of the groundwater at this site.

##### 4.6.1 Gamma-Emitting Radionuclides

The results of the direct gamma-ray spectrometry of the ion-exchange resins, aluminum oxide and filters are given in Appendix B. The groundwater at this site is practically free of long-lived gamma-emitting radionuclides, which included <sup>54</sup>Mn, <sup>60</sup>Co, <sup>95</sup>Zr-Nb, <sup>103,106</sup>Ru, <sup>134-137</sup>Cs, <sup>125</sup>Sb, <sup>144</sup>Ce, and <sup>152-155</sup>Eu. Well C-3, located between the Nitrate Disposal Pit and Lake 233, was completely devoid of this group of radionuclides. Well C-125, which was drilled closest to the disposal pit, contained only trace concentrations of <sup>60</sup>Co in the groundwater. The highest gross beta and <sup>90</sup>Sr concentrations were found at Well C-119; however, this well contained nondetectable amounts of gamma-emitting radionuclides, except a possible trace of <sup>154</sup>Eu which was very near its detection limit. Well C-27, located some 100 m downgradient from the disposal pit, contained 5 pCi/l of cationic <sup>137</sup>Cs and 0.87 pCi/l of particulate <sup>137</sup>Cs. This well and the adjacent Well C-81 contained some of the highest gross beta and <sup>90</sup>Sr concentrations observed downgradient from



the disposal pit (see Section 4.5.2). The only other well to show traces of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  was C-54-3 which was near the swampy area where the groundwater emerged some 450 m downgradient from the disposal pit.

From the above measurements it is obvious that no significant migration of any long-lived gamma-emitters has occurred in the groundwater at this site. Cesium-137 would be expected to be the most abundant long-lived gamma-emitting fission product present in the disposal pit, but its near absence in the groundwater downgradient from the pit indicates that this radionuclide is firmly attached to the soil immediately under the disposal pit. The observation that  $^{137}\text{Cs}$  has not significantly migrated from its source supports the predictive transport modeling results described in Section 3.

#### 4.6.2 Comprehensive Radionuclide Analyses

The well with the highest gross beta activity (C-119) was selected for comprehensive radiochemical analyses to determine what other long-lived radionuclides may be present in the groundwater at this site. Table 4.1 lists the concentrations of 10CFR61 radionuclides present in cationic and anionic forms in the groundwater of this well. It is obvious that  $^{90}\text{Sr}$  is the overwhelming radionuclide constituent of this groundwater, having a measured concentration of 280,000 pCi/l. Only traces of  $^{59}\text{Fe}$ ,  $^{154}\text{Eu}$  and the plutonium isotopes  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  were the other detectable radionuclides being present near their detection limits. The plutonium isotopic composition indicates its source to be very high burnup reactor fuel or isotopically separated  $^{238}\text{Pu}$ . This plutonium was mainly present in a cationic form and may have been transported to this location as part of a slug of the original low pH aqueous discharges containing complexing agents. The cationic nature of the plutonium at this well was unusual for groundwater at the CRNL LLW management area. Previous studies have indicated that the plutonium migrating from other disposal sites at CRNL is predominantly in an anionic form.<sup>(18)</sup>

Strontium-90 and gross beta measurements were made on all seven well samples used for large-volume water sampling. These data are presented in Table 4.2. The gross beta measurements were determined by evaporating 10 ml of the groundwater samples to about 1 ml and transferring to 2.5-cm diameter stainless steel counting planchets. The samples were evaporated to dryness and counted on a proportional counter. A D/C factor of 2.26 was used for calculating the activity due to  $^{90}\text{Sr} + ^{90}\text{Y}$ . This D/C factor represents an average of the counting efficiencies of the beta energies due to  $^{90}\text{Sr}$  and to  $^{90}\text{Y}$ . The computed disintegration rate due to the  $^{90}\text{Sr} + ^{90}\text{Y}$  was then divided by two to obtain the contribution from only the  $^{90}\text{Sr}$  activity.

As shown in Table 4.2, the gross-beta-to- $^{90}\text{Sr}$ -activity ratio ranged from 0.93 to 1.57. The reason for the ratios which were significantly greater than unity is not known because no other detectable radionuclides were observed in the gamma spectra or from radiochemical separations.

TABLE 4.1. Comprehensive Radiochemical Analyses of Groundwater from Well C-119, Nitrate Disposal Pit Plume

Radionuclide	Concentration (pCi/liter)	
	Cationic	Anionic
<sup>54</sup> Mn	<0.2	<0.04
<sup>55</sup> Fe	1.30 ± 0.76	5.14 ± 0.58
<sup>60</sup> Co	<0.1	<0.03
<sup>59</sup> Ni	<0.6	<0.3
<sup>63</sup> Ni	<0.1	<0.1
<sup>90</sup> Sr	280,000 ± 800	--
<sup>95</sup> Zr	<1.8	<0.2
<sup>95</sup> Nb	<3.7	<0.4
<sup>99</sup> Tc	--	--
<sup>106</sup> Ru	<3.2	<0.3
<sup>125</sup> Sb	<1.1	<0.08
<sup>129</sup> I	<0.9	<0.8
<sup>134</sup> Cs	<0.3	<0.03
<sup>137</sup> Cs	<0.3	<0.03
<sup>144</sup> Ce	<0.5	<0.2
<sup>152</sup> Eu	<1.3	<0.08
<sup>154</sup> Eu	2.0 ± 1.0	<0.1
<sup>155</sup> Eu	<3.4	<0.1
<sup>238</sup> Pu	0.124 ± 0.013	0.019 ± 0.003
<sup>239-240</sup> Pu	0.011 ± 0.005	0.0014 ± 0.0013

Also in Table 4.2 is a comparison of the gross beta measurements made by CRNL with those made by PNL. The sampling dates were not exactly the same, but the samplings were generally made within approximately one year of each other. The only exception was Well C-27 which was sampled by CRNL in January 1980, compared with the October 1984 PNL sampling. Parsons<sup>(16)</sup> has estimated that the equilibrium rate of movement of <sup>90</sup>Sr at this location should be about 0.015 times the velocity of the groundwater (15 cm/day). For the 1700 days elapsed between the January, 1980, CRNL sampling and the October 1984, PNL sampling of Well C-27, the <sup>90</sup>Sr could have moved down-gradient some 3.4 m (11.2 ft). Thus, it is not too surprising that the gross beta values for the two samplings are not in good agreement.

TABLE 4.2. Strontium-90 and Gross Beta Activity of Groundwater Samples from the Nitrate Disposal Pit Plume, October 1984

<u>Sample Location</u>	<u>Sample Date</u>	<u>Volume (liters)</u>	<u>Gross Beta (pCi/l)</u>	<u><sup>90</sup>Sr (pCi/l)</u>	<u>Ratio Gross <sup>90</sup>Sr</u>	<u>CRNL Gross Beta (pCi/l)</u>
C-3-2 (15' depth)	10/17/84	192	610	<14	---	
C-125 (2" dia. well, 40' depth)	10/15/84	174	72,800	46,430	1.57	98,000
C-119 (2" dia. well, 46' depth)	10/13/84	187	406,000	280,000	1.45	675,000
C-27-III (27' depth)	10/16/84	204	34,400	35,900	0.930	594,000
C-69-11 (36' depth)	10/9/85	200	262,000	201,000	1.30	191,000
C-89-11 (36' depth)	10/10/84	215	16,900	15,600	1.08	10,000
C-54-3 (20' depth)	10/10/84	227	<190	<14	---	

\* All of the CRNL gross beta results were for samples collected about one year before the PNL samples, except for Well C-27-III which CRNL sampled 1700 days before the PNL samples.

#### 4.6.3 Chemical Analyses

Table 4.3 shows the chemical analyses of the groundwater sampled in October 1984 by PNL. The measurements were made in the field immediately after sampling or, in the case of  $Fe^{+2}/Fe^{+3}$ , within two days of sampling. In general, most of the groundwater samples were oxidizing and typical of surficial aquifers in this region. The main exception was Well C-69 which had the most reducing Eh, the lowest dissolved oxygen, and the highest alkalinity and  $Fe^{+2}$  concentrations. These conditions may reflect the remnants of a slug of the acidified disposal pit effluent which dissolved  $Ca^{++}$  and  $Fe^{+2}$  from the limestone-lined pit and transported it away at a decelerating rate. This well also contained one of the highest downstream concentrations of  $^{90}Sr$  (see Section 4.7).

TABLE 4.3. Chemical Analysis of Chalk River Groundwater Samples, October, 1984

Sample Location	Sample Date	Sample Volume (liters)	pH	Eh (mV)	Temp. (°C)	Dissolved O <sub>2</sub> (mg/l)	Alkalinity (mg/l as CaCO <sub>3</sub> )	Fe <sup>+2</sup> (µg/l)	Total Fe (µg/l)
C-3-2 (15' depth)	10/17/84	192	4.65	+410	12.0	0.90	7.5	270	330
C-125 (2" dia. well, 40' depth)	10/15/84	174	5.10	+475	12.5	2.74	5.2	7.0	23
C-119 (2" dia. well, 46' depth)	10/13/84	187	5.45	+505	12.0	1.70	5.2	<3	<3
C-27-III (27' depth)	10/16/84	204	6.00	+480	11.0	4.68	10.9	<3	<3
C-69-11 (36' depth)	10/9/84	200	5.30	+330	10.0	0.10	24.0	5000	4600
C-89-11 (36' depth)	10/10/84	215	5.05	+405	10.0	0.57	15.9	410	560
C-54-3 (20' depth)	10/10/84	227	5.30	+460	9.0	3.34	7.7	<3	<3

Two other wells, C-3 and C-89, also tended to exhibit relatively more reducing conditions. Well C-3 was located between the disposal pit and Lake 233 and contained no fission product radionuclides. The chemistry of the groundwater at this site was undoubtedly influenced by recharge from Lake 233.

#### 4.7 MAPPING OF EMPIRICAL DATA

The extensive gross beta measurements made by CRNL, which essentially represent the  $^{90}\text{Sr}$  transport in the downgradient groundwater from the Nitrate Disposal Pit site, were plotted in a two-dimensional plane to define the actual  $^{90}\text{Sr}$  contaminant plume. This CRNL data set, which represents some 100 monitoring wells and over 1000 sample piezometers covering the immediate plume area (see Figure 4.4), has proven extremely useful in defining the areal and vertical dimensions of the plume that has developed over the past 30 years.

The  $^{90}\text{Sr}$  concentrations (represented by the gross beta measurements) have been plotted in a two-dimensional horizontal plane using two different methods. First, the peak  $^{90}\text{Sr}$  concentrations observed over the thickness of the saturated zone at each monitoring well were plotted and contour lines developed to map the plumes of the peak activities. The plume developed from this exercise is shown in Figure 4.5. The principal features of this plume are its relatively narrow width, its converging distribution downstream, and the presence of two "hot spots" at the plume centerline at distances of approximately 330 ft and 740 ft downstream from the disposal pit. The two "hot spots" may be the remnants of slugs of relatively high  $^{90}\text{Sr}$  concentrations released to the groundwater in acidified discharges to the limestone-lined disposal pit. The low initial pH of these discharges and the high concentrations of  $\text{Ca}^{++}$  ion dissolved from the limestone by the acid would minimize the retardation of  $^{90}\text{Sr}$  during its movement through the soil until neutralization and absorption processes reached an equilibrium state. Parsons<sup>(16)</sup> has shown that this has indeed occurred at this site and could explain the reason for the "hot spots" of  $^{90}\text{Sr}$  downstream in the plume.

The second method of plotting the  $^{90}\text{Sr}$  plume was conducted in a manner in which the concentration contours would be more comparable with the predicted radionuclide migration estimated by the predictive modeling processes described in Section 3 of this report. In this mapping, the average  $^{90}\text{Sr}$  concentration throughout the thickness of the saturated zone of the surficial aquifer was determined and plotted in a two-dimensional horizontal plane. The averaging of the  $^{90}\text{Sr}$  concentration was accomplished by computing the total pCi contained in the groundwater in each vertical column of soil and dividing by the total volume of water in the column. A 10 cm x 10 cm surface area was assumed, and this area was multiplied by the depth interval (in cm) sampled by each piezometer in a monitoring well. The resulting volume was multiplied by an average porosity value for the soil (0.35) to give the interstitial volume of the soil which was assumed to be filled with groundwater. The  $^{90}\text{Sr}$  concentration of the sampled groundwater was multiplied by the total water volume contained in each respective sampled section to give the total pCi of  $^{90}\text{Sr}$  in each section of the well. This process was

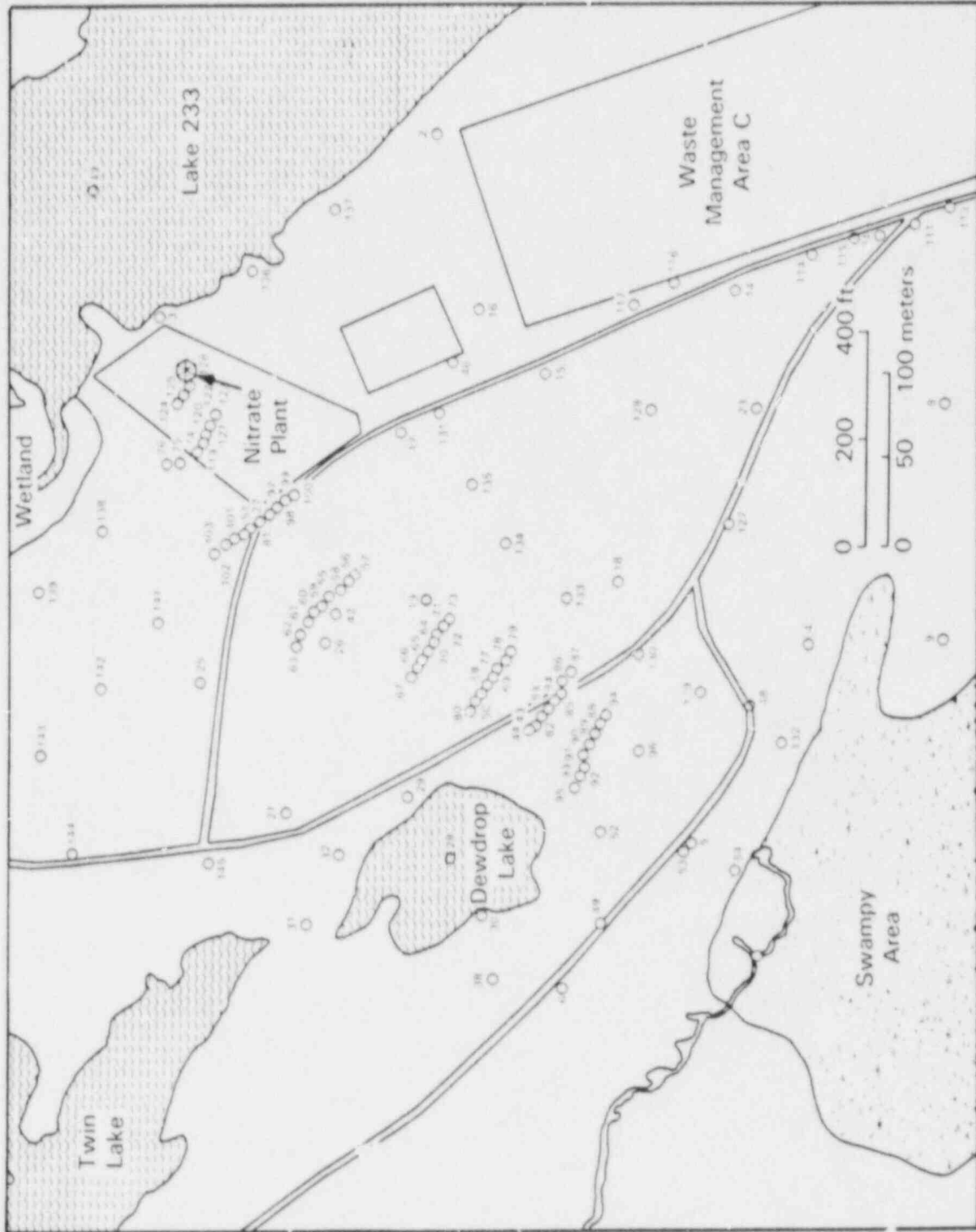


FIGURE 4.4. Locations of Monitoring Wells at the Nitrate Disposal Pit Site

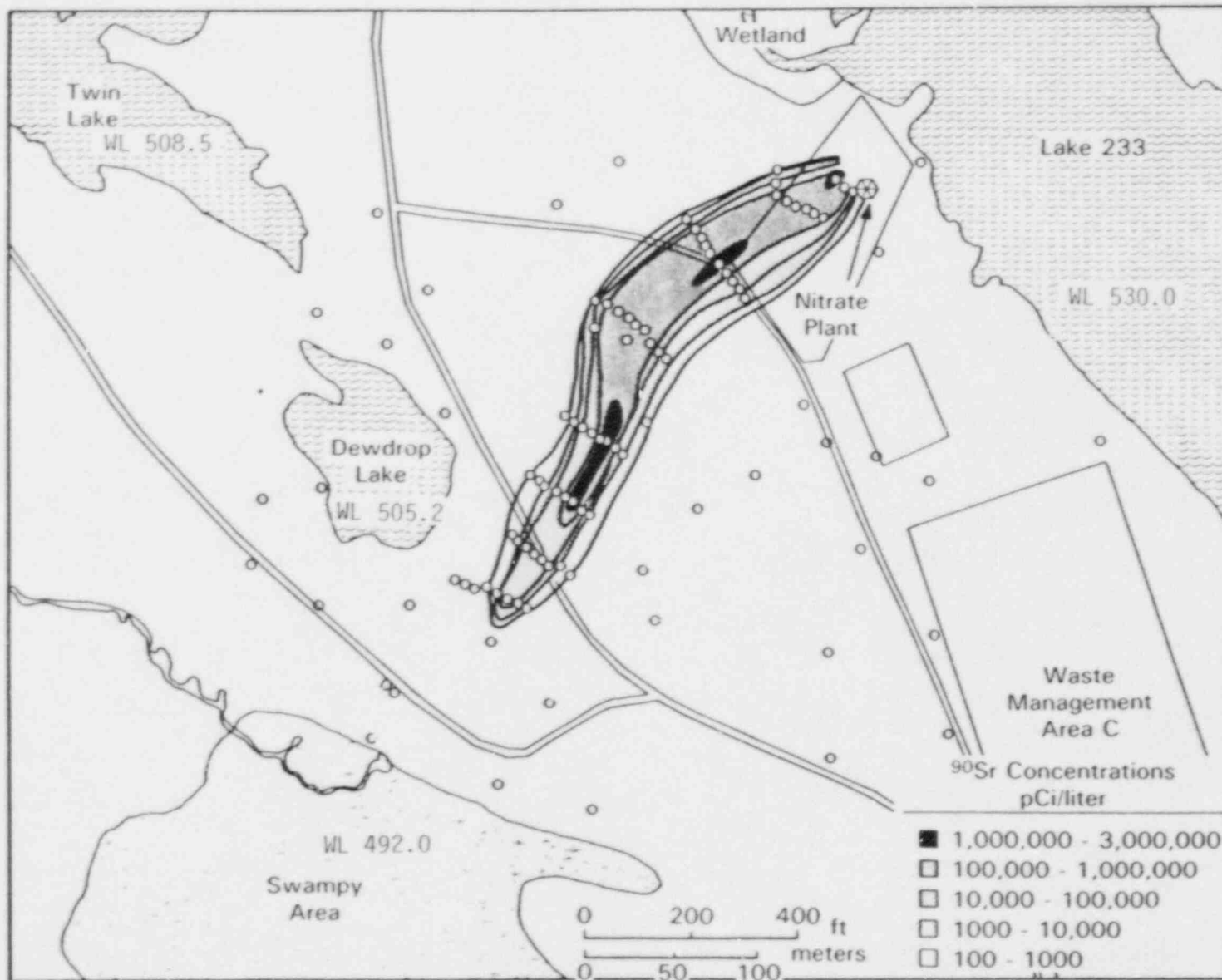


FIGURE 4.5. Observed  $^{90}\text{Sr}$  Plume from the Nitrate Disposal Pit, 1984 (Peak  $^{90}\text{Sr}$  Concentrations in the Saturated Zone at Each Piezometer)

continued through the entire thickness of the saturated zone at each well. Extrapolation was performed for those vertical sections which were not sampled. The total pCi of  $^{90}\text{Sr}$  in the entire vertical section was then summed and divided by the total water volume contained in the entire section to give an average  $^{90}\text{Sr}$  concentration. This provided concentration contours directly comparable with the output of the predictive transport modeling which also produces average  $^{90}\text{Sr}$  concentrations over the surficial aquifer thickness. The  $^{90}\text{Sr}$  plume determined and mapped by this method is shown in Figure 4.6. The general shape of the plume of the average  $^{90}\text{Sr}$  concentrations in the groundwater is very similar to the plume mapped by plotting the peak  $^{90}\text{Sr}$  concentrations at each monitoring well, e.g., each plume is relatively narrow and converges near the downstream end. The averaging of the  $^{90}\text{Sr}$  concentrations, as shown in Figure 4.6, does not show both "hot spots" as seen in Figure 4.5, although the narrow downstream maxima are evident at the transect of wells located distances of 740 and 870 ft downstream from the pit.

The vertical distribution of  $^{90}\text{Sr}$  in a cross-section along the centerline of the plume developed by 1984 is shown in Figure 4.7. The plume map is superimposed over a cross-section of the major lithologic units comprising the deposits in this region. The plume has migrated through the layer of fine to very fine sand. Initially, the upper boundary of the plume closely follows the top of the water table downgradient for about 150 m. Then the plume dips several meters below the water table between about 500 to 1000 m downgradient from the source. The lower boundary of the plume is confined by the bedrock surface over most of the downgradient distance.

The  $^{90}\text{Sr}$  concentrations as a function of depth at each monitoring well along each of the eight transects of wells across the contaminant plume were plotted in cross-section to provide profiles of the plume (see Figures 4.8 to 4.15). A vertical exaggeration of 3.3 to 1 was used to provide more convenient plotting of the data.

Figure 4.8 shows the plume cross-section 50 ft downstream from the pit. This profile is open-ended on each side because it appears that the plume profile at this point was wider than the coverage of the two monitoring wells which were sampled. This profile showed a maximum  $^{90}\text{Sr}$  concentration at a depth of about 42 ft. The  $^{90}\text{Sr}$  cross-section at the 140-foot transect (Figure 4.9) had a width of about 170 ft, with maximum concentrations occurring between depths of 40 to 52 ft. At the 300-foot transect (Figure 4.10), the plume had widened to about 220 ft in Well C-81. Figures 4.11 through 4.15 show the cross-sectional profiles of the  $^{90}\text{Sr}$  as it has moved downstream and converged into a more narrow and dilute plume. The downstream  $^{90}\text{Sr}$  maxima were readily observed at the 740- and 870-foot transects in Wells C-71 and C-77 at depths of 36 and 38 ft, respectively. No detectable  $^{90}\text{Sr}$  could be measured past the 1150-foot transect.



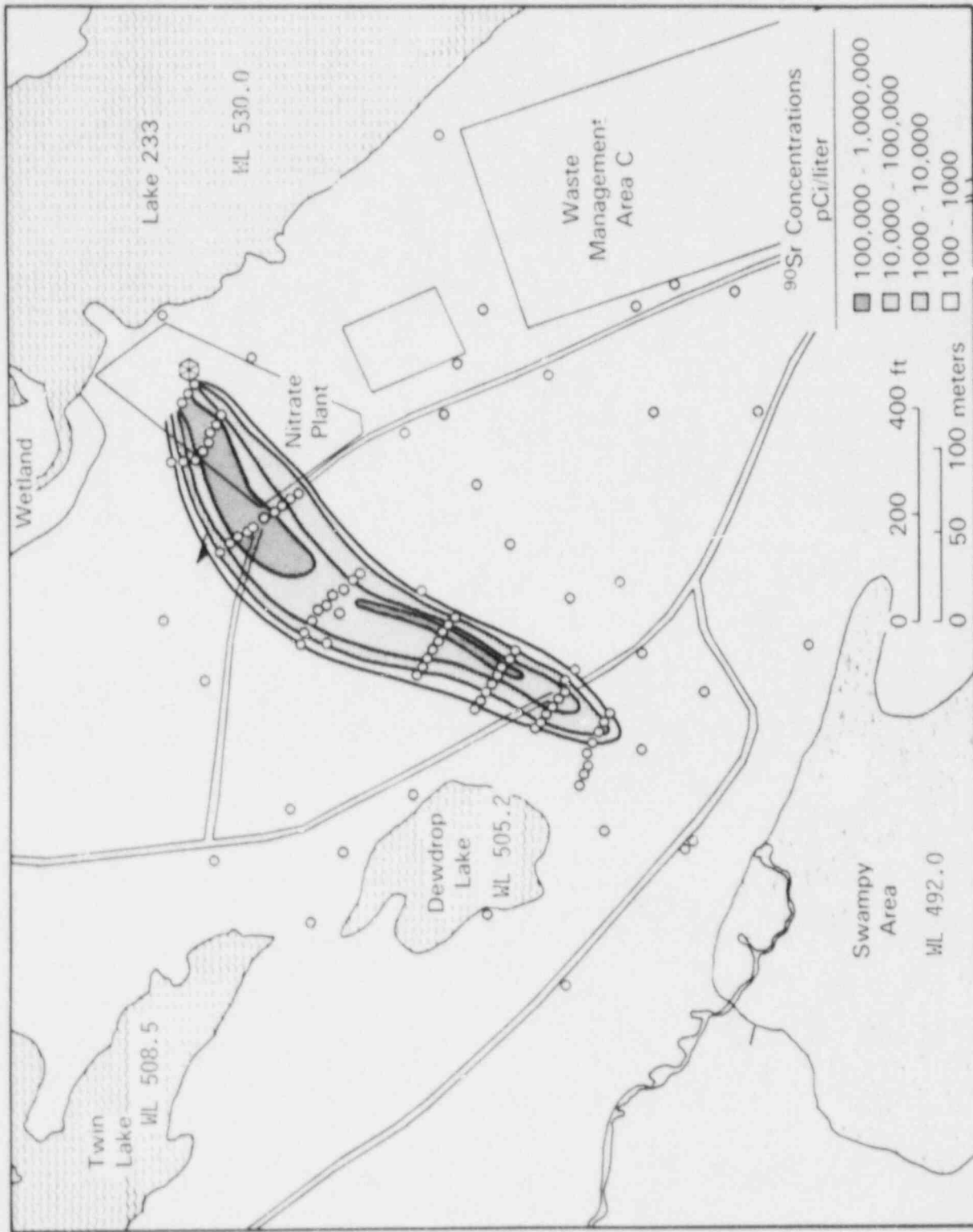


FIGURE 4.6. Strontium-90 Migration in Groundwater from the Nitrate Disposal Pit Site (Concentrations in pCi/liter Averaged Over the Thickness of the Saturated Zone)

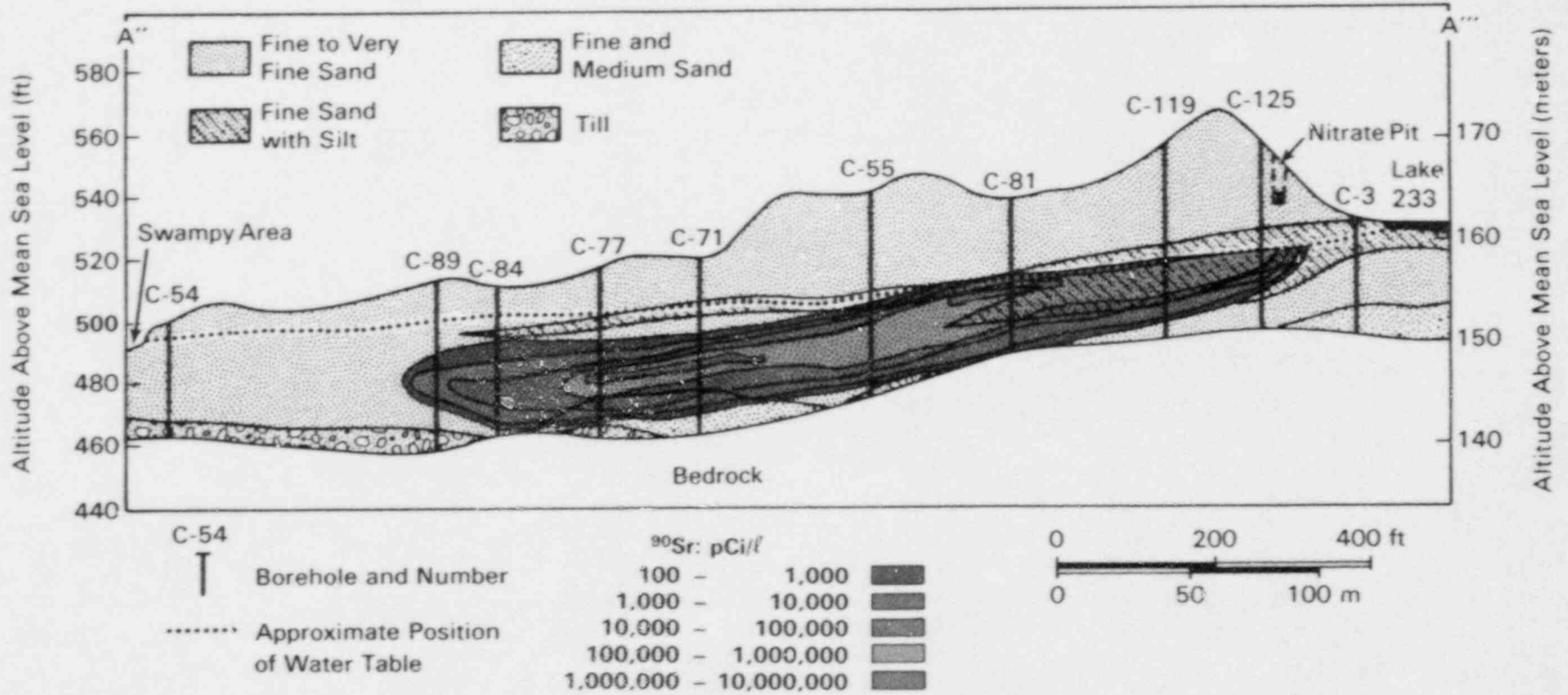


FIGURE 4.7. Vertical Cross Section of the  $^{90}\text{Sr}$  Distribution Along the Centerline of the Plume, 1984

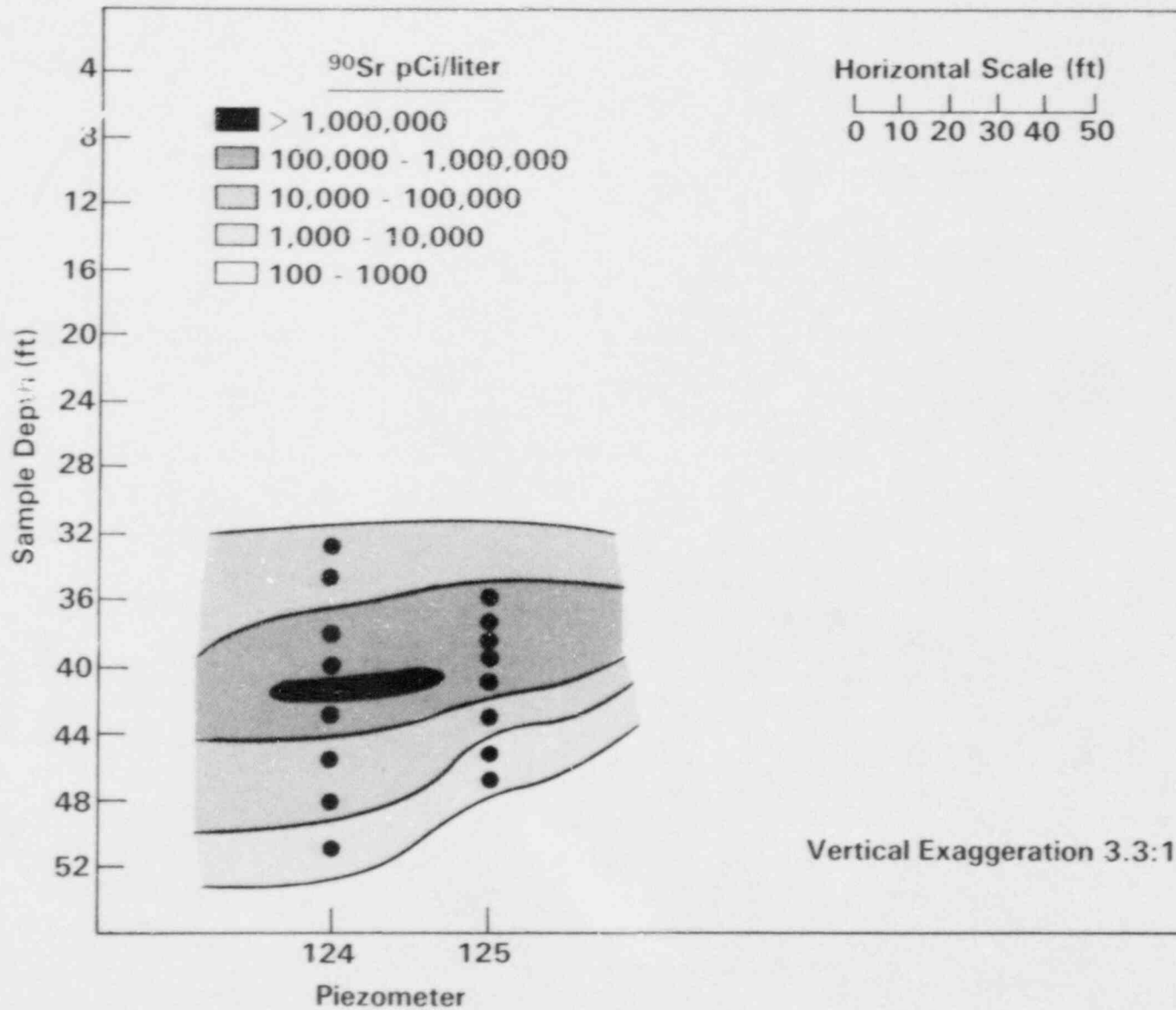


FIGURE 4.8. Nitrate Disposal Pit Groundwater Plume Cross Section, 50-Foot Transect  
<sup>90</sup>Sr Concentration - pCi/liter

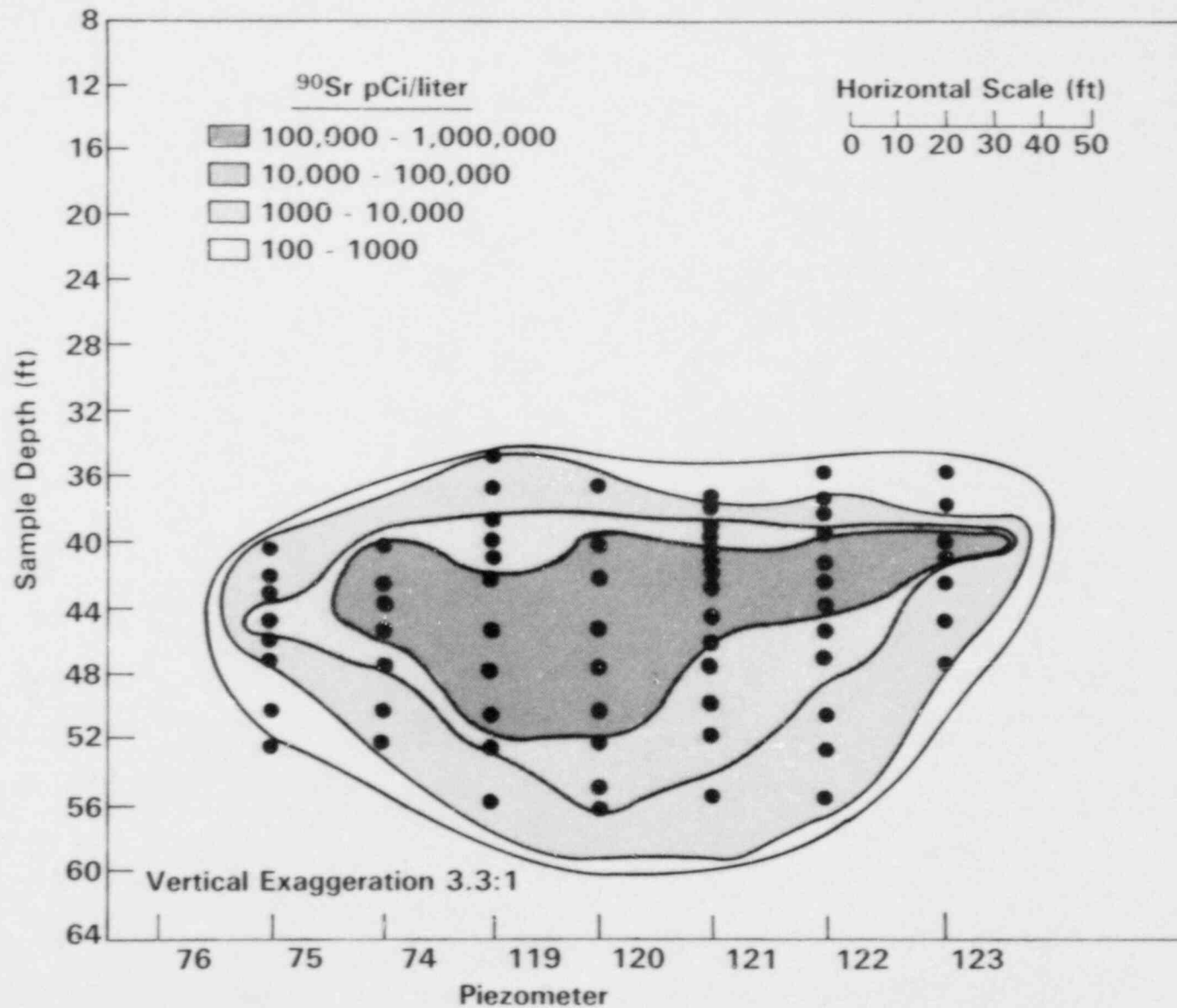


FIGURE 4.9. Nitrate Disposal Pit Groundwater Plume Cross Section - 140-Foot Transect  
 $^{90}\text{Sr}$  Concentrations - pCi/liter

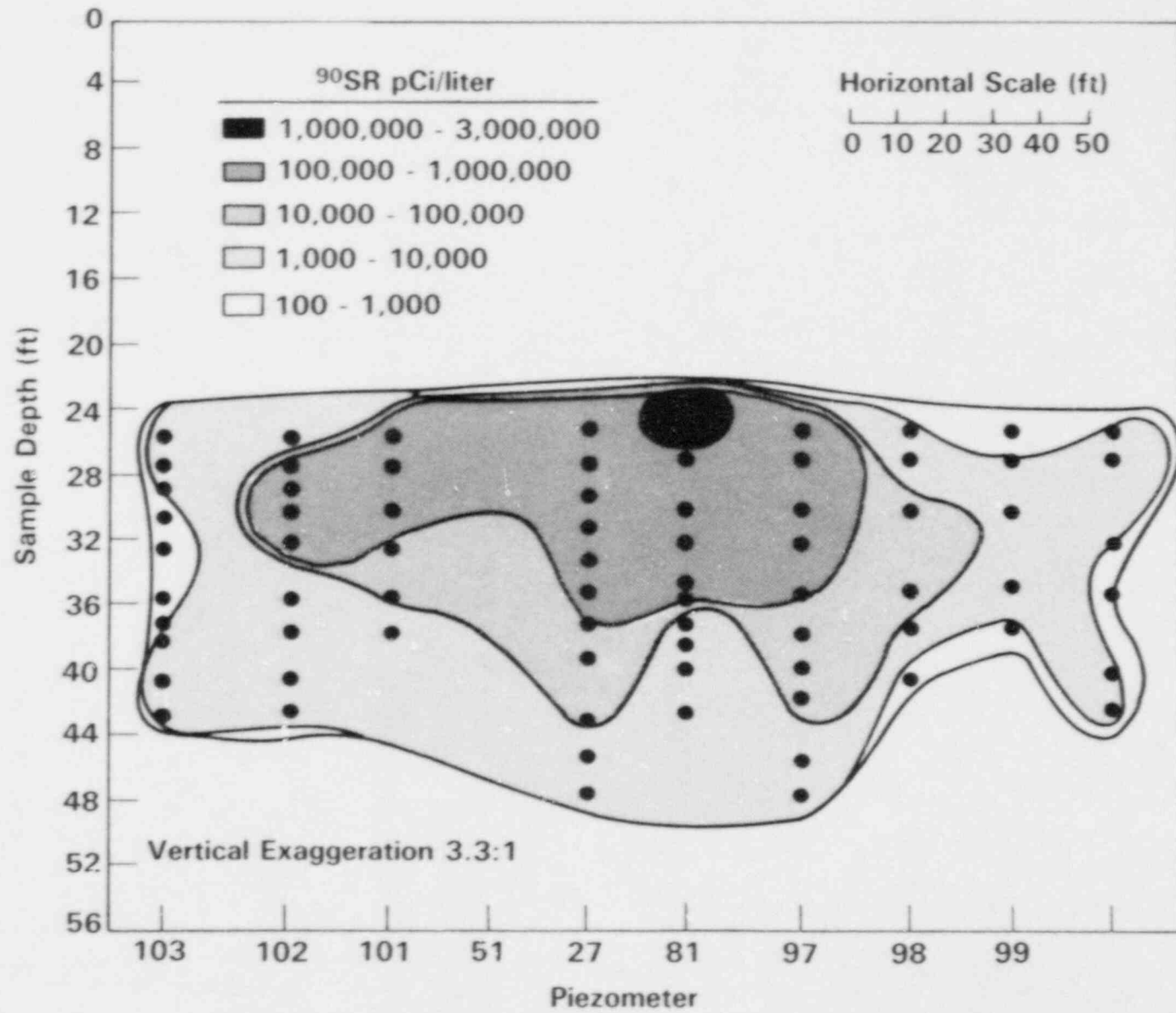


FIGURE 4.10. Nitrate Disposal Pit Groundwater Plume Cross Section - 330-Foot Transect  
<sup>90</sup>Sr Concentrations - pCi/liter

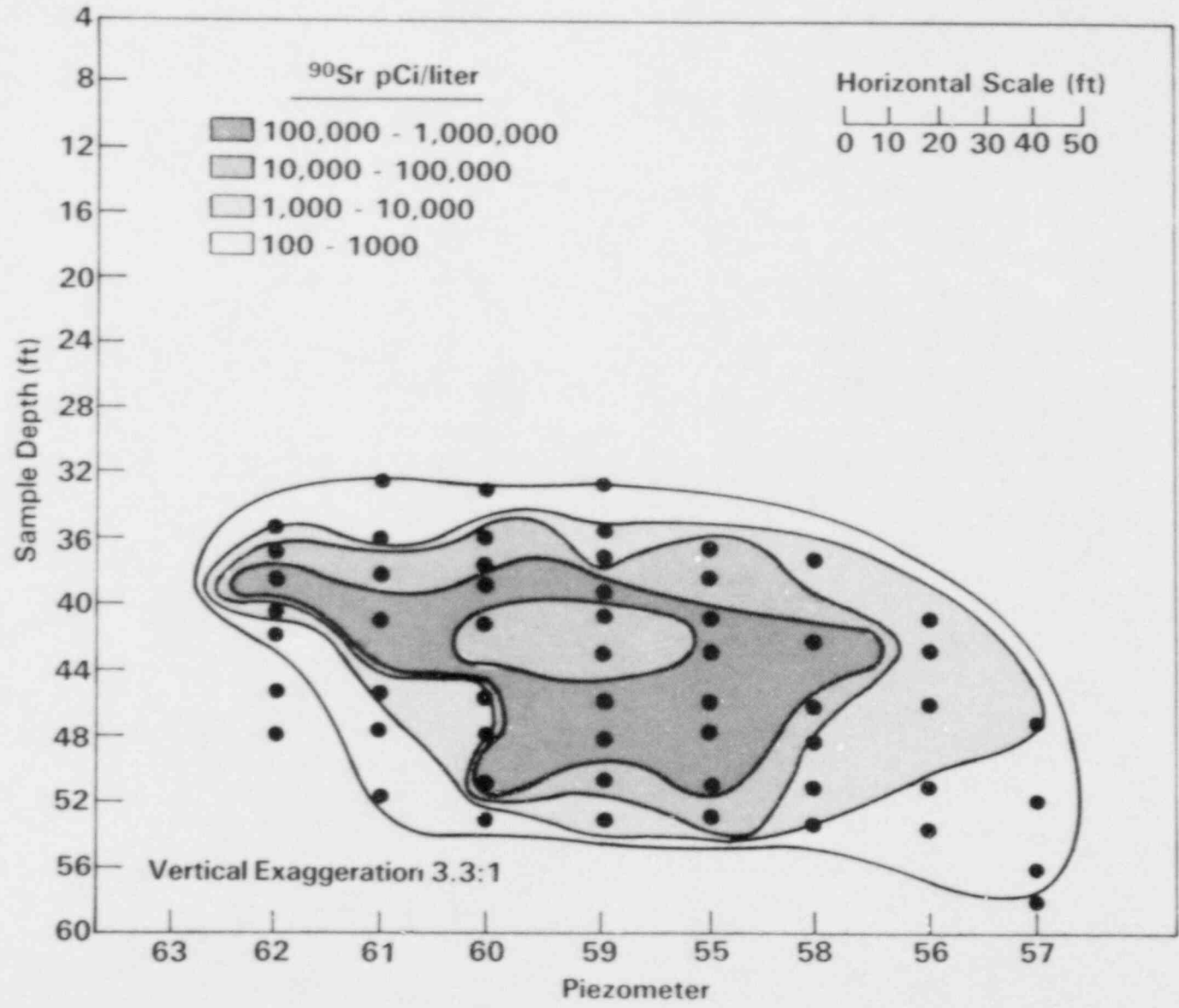


FIGURE 4.11. Nitrate Disposal Pit Groundwater Plume Cross Section - 540-Foot Transect  
<sup>90</sup>Sr pCi/liter

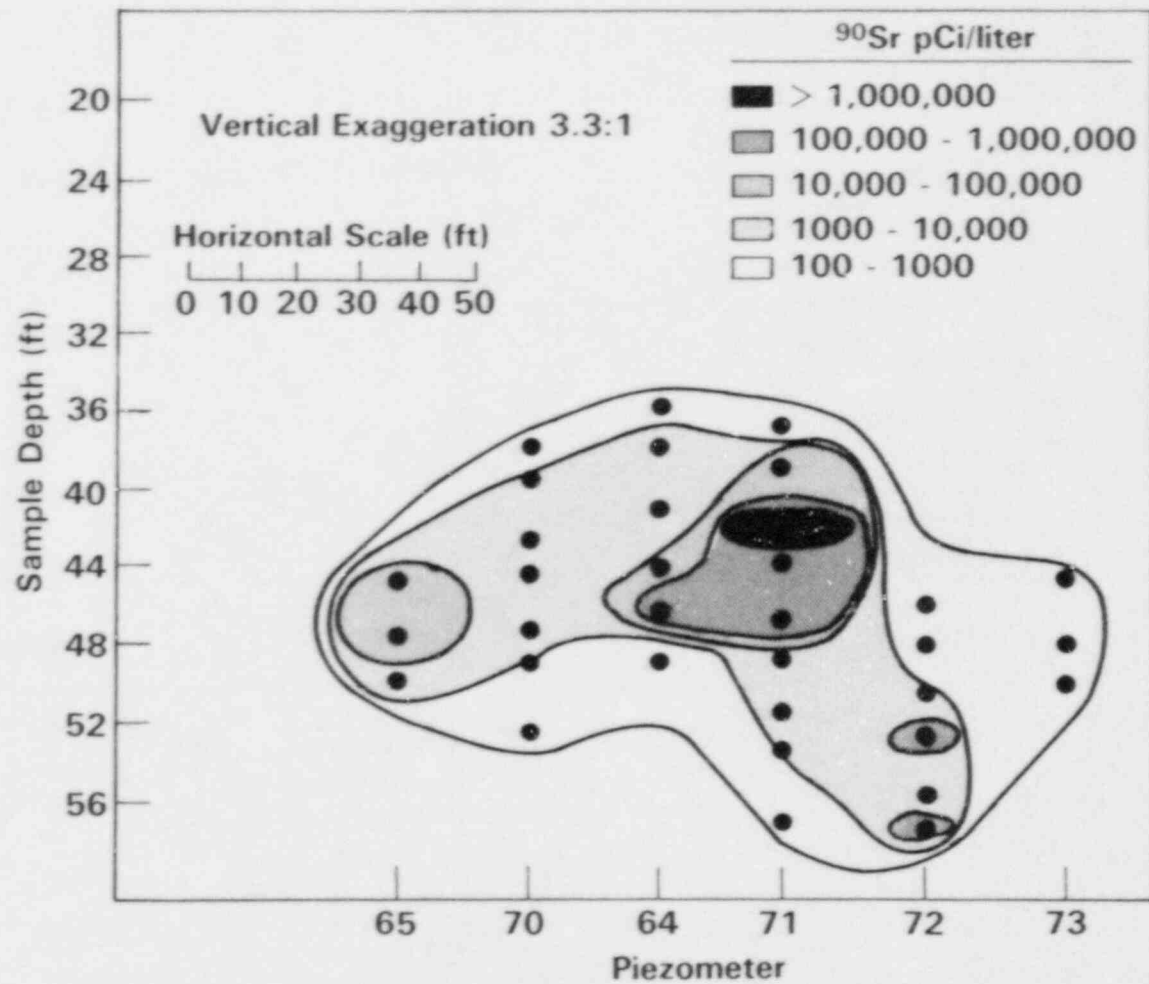


FIGURE 4.12. Nitrate Disposal Pit Ground Water Cross Section - 740-Foot Transect  
<sup>90</sup>Sr Concentration - pCi/liter

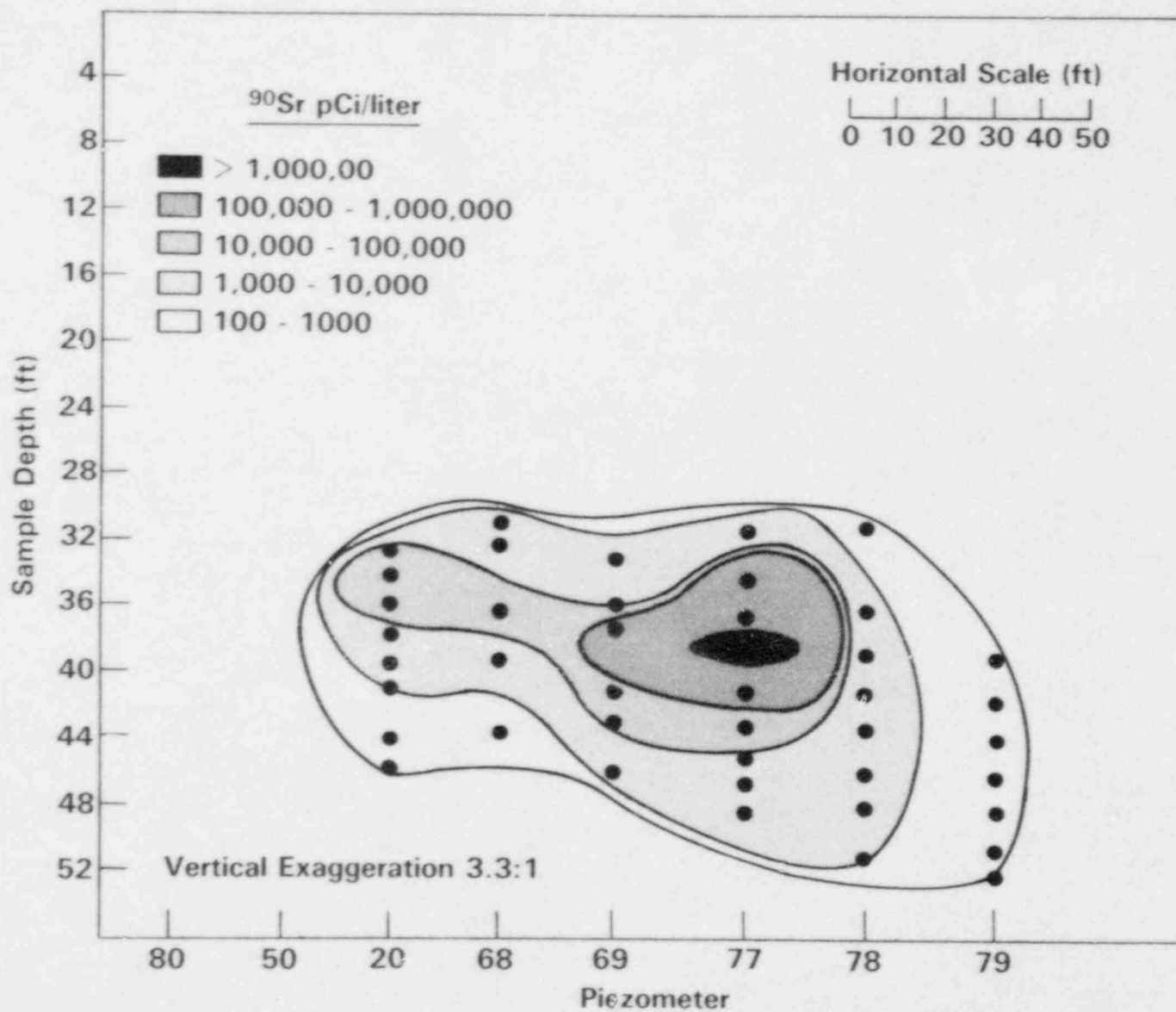


FIGURE 4.13. Nitrate Disposal Pit Groundwater Plume Cross Section - 870-Foot Transect  
<sup>90</sup>Sr Concentration - pCi/liter



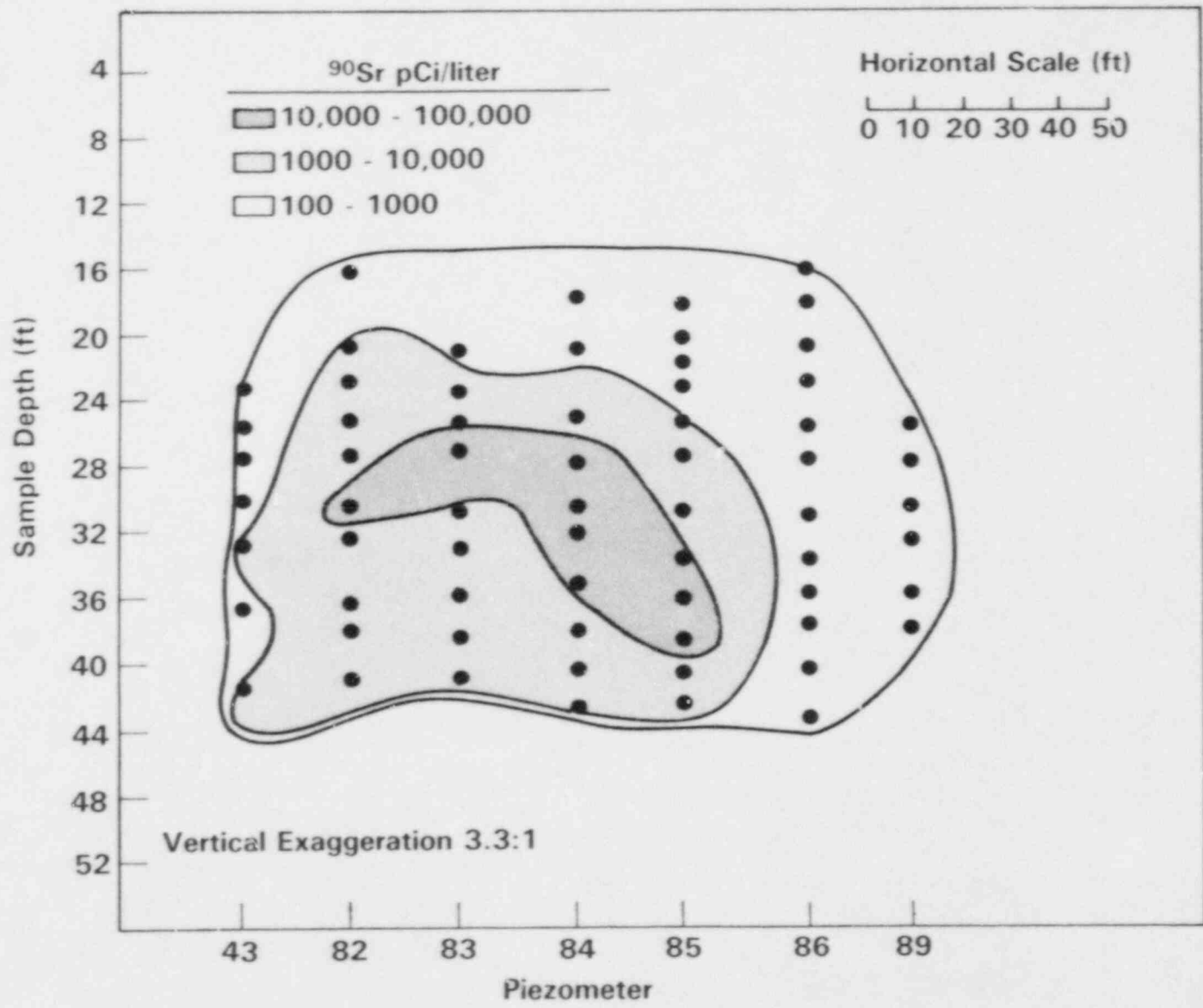


FIGURE 4.14. Nitrate Disposal Pit Groundwater Plume Cross Section - 1020-Foot Transect  
 $^{90}\text{Sr}$  Concentration - pCi/liter

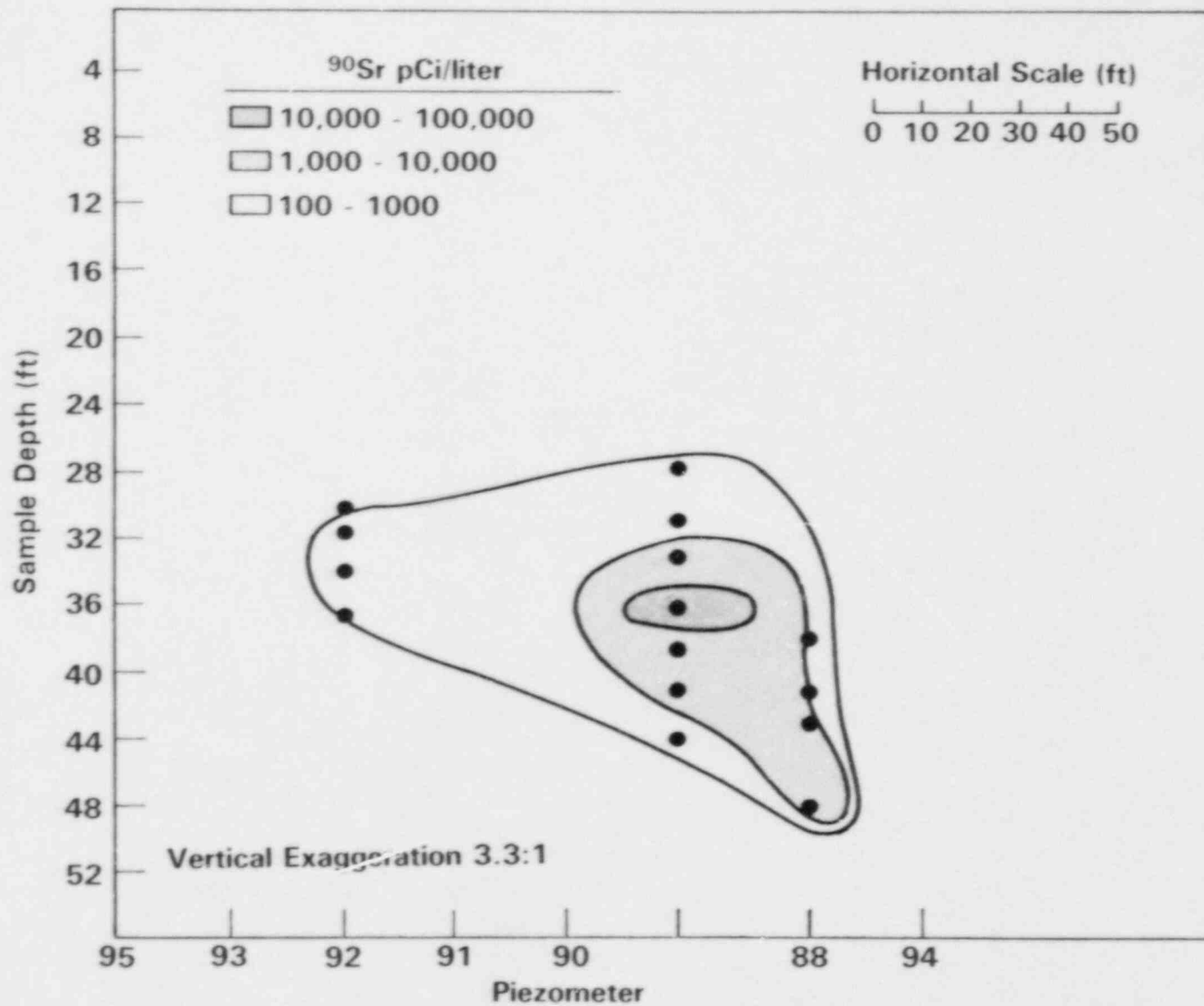


FIGURE 4.15. Nitrate Disposal Pit Groundwater Plume Cross Section - 1150-Foot Transect  
<sup>90</sup>Sr Concentration - pCi/liter

## 5.0 COMPARISON OF PREDICTED AND OBSERVED RADIONUCLIDE MOVEMENT

Field measurements of actual radionuclide movement that has occurred over the past 30 years at this site have indicated that  $^{90}\text{Sr}$  is the only long-lived radionuclide that has significantly migrated from the disposal site. A plume mapped from field measurements (Figures 4.5 and 4.6) indicates that groundwater containing  $^{90}\text{Sr}$  concentrations in excess of 100 pCi/l moved away from the disposal site in a relatively narrow band about 50 to 100 m wide and about 350 m long.

The pattern of  $^{90}\text{Sr}$  concentrations within the plume downgradient from the site indicates that the  $^{90}\text{Sr}$  inventory has not moved at a uniform rate over the 30-year period. High concentrations of  $^{90}\text{Sr}$  at distances between 150 and 250 m from the site suggest that perhaps a part of the inventory entered the groundwater system shortly after disposal and moved rapidly. This early rapid migration of  $^{90}\text{Sr}$  was documented by Parsons.<sup>(6)</sup> These observations are consistent with the 1-year waste release scenarios developed in the present study and has been attributed to the low pH and the high concentration of competing ions in the disposal (ammonium, calcium, etc.) that reduced the retention characteristics of the soil.

The area of high  $^{90}\text{Sr}$  concentration located within 100 m of the disposal site suggests that in time the release of  $^{90}\text{Sr}$  plume from the soils beneath the site became much slower, and, once in the groundwater, the  $^{90}\text{Sr}$  moved at retarded velocities with slight lateral spreading. Indeed, Parsons<sup>(6)</sup> showed that in the first year after disposal the  $^{90}\text{Sr}$  had moved more than 76 m and in the following 5-year period it advanced less than 49 m. A comparison between these values and the corresponding groundwater rates ( $V_g$ ) showed that the mean flow rate for  $^{90}\text{Sr}$  in the first year after disposal was greater than  $0.61 V_g$ , while its mean rate over the following five years had diminished to less than  $0.07 V_g$ .

Strontium-90 transport modeling with a retardation factor of 25 and any one of the waste release scenarios provided plumes that were in close agreement with the measured downgradient movement of  $^{90}\text{Sr}$ . As a comparison, results of the detailed data modeling of the 30-year waste release scenario are given in Figure 5.1. For this case, the 100 pCi/l  $^{90}\text{Sr}$  concentration isopleths for the observed and the predicted values 30 years after disposal were 330 and 230 m downgradient from the disposal pit, respectively. However, major discrepancies become apparent when the internal distribution of concentrations and the lateral spreading of the plumes are compared.

The distribution of  $^{90}\text{Sr}$  concentrations within the predicted plumes deviates from the observed distributions. Unlike the 100 pCi/l  $^{90}\text{Sr}$  concentration downgradient isopleth where a reasonable match was achieved, the 1000 pCi/l and 10,000 pCi/l isopleths for the predicted and observed  $^{90}\text{Sr}$  migration downgradient from the disposal site were separated by about 50 to 70 m. These deviations are primarily because the actual observed distributions are the end result of a combination of the waste release scenarios and migration rates. Early rapid release and migration of the  $^{90}\text{Sr}$  inventory was followed by a slower and perhaps irregular release and transport.

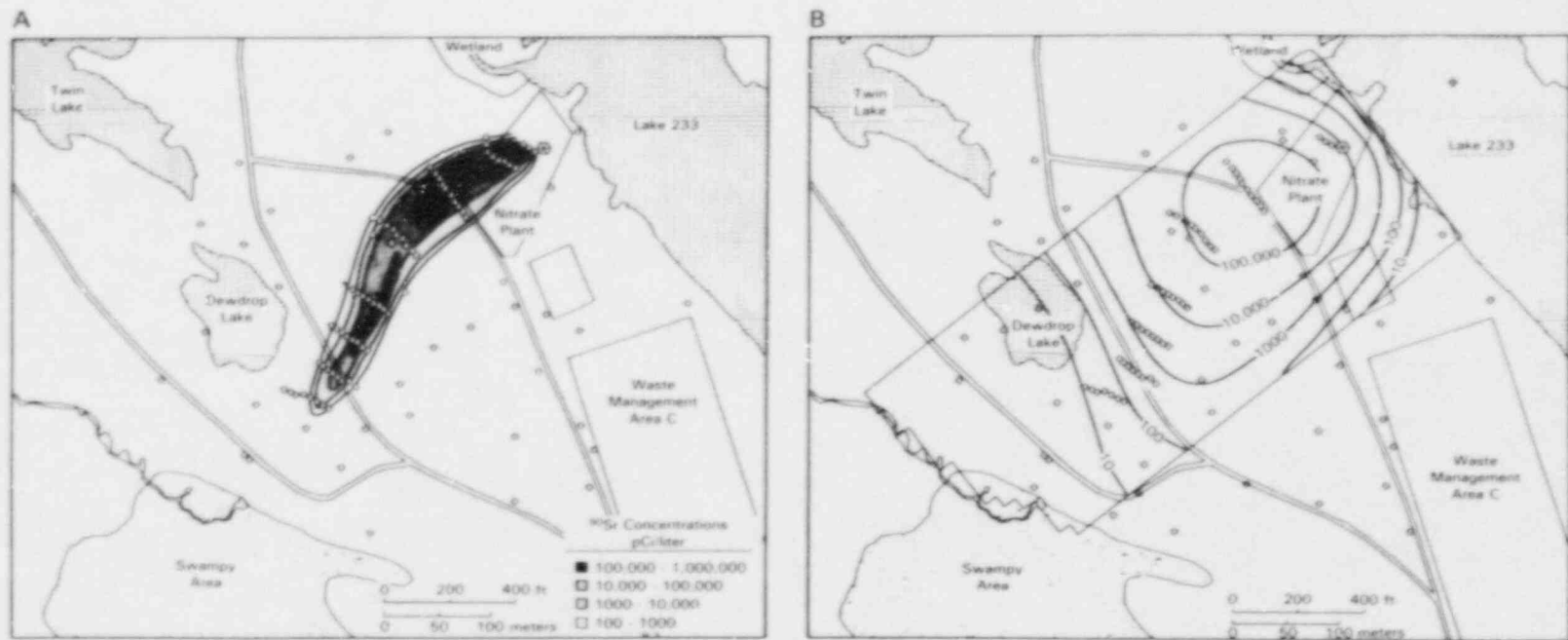


FIGURE 5.1. Comparison of Observed  $^{90}\text{Sr}$  Transport (A) and Simulated Detailed-Data Modeling of  $^{90}\text{Sr}$  Transport (B) After 30 Years

The actual  $^{90}\text{Sr}$  plume was about 3 times narrower at its widest part than the predicted plumes that were in closest agreement, i.e., those using a retardation factor of 25 (see Figure 5.1). The narrow width of the observed plume is probably a function of the rather uniform sorting and fine-grained texture of the surficial sand. These characteristics would have a tendency to minimize hydrodynamic dispersion that might otherwise occur in a more heterogeneous material.

The predicted extent of lateral movement provided a conservative estimate of the spreading and is mainly a function of the numerical dispersion caused by the coarse grid spacing currently being used in the model. This spreading could be reduced by refining the finite element mesh in a direction transverse to the principal direction of groundwater movement. A reduction in transverse spreading would increase contaminant concentrations within the plume. The increased concentrations and an associated increased concentration gradient would result in increased downgradient migration.

For example, if a finite element grid where the grid spacing is decreased by a factor of about 3 in a direction transverse to the principal direction of groundwater movement is used such as is shown in Figure 5.2, results for the same release experiments would be significantly different. The effect of reducing the transverse grid space is demonstrated in the examples of the 1-year inventory release presented in Figure 5.3. With the smaller grid spacing, the simulated plumes are much more narrow and have higher concentrations downgradient than were predicted with the current grid. At the same time, because grid spacing is larger in a downgradient direction, the predicted plumes migrate much further than they do with the current grid. A reduction in grid spacing would continue to reduce the amount of lateral spreading on predicted results until this numerical dispersion becomes less than the amount of dispersion attributable to the simulated longitudinal and transverse dispersivities.

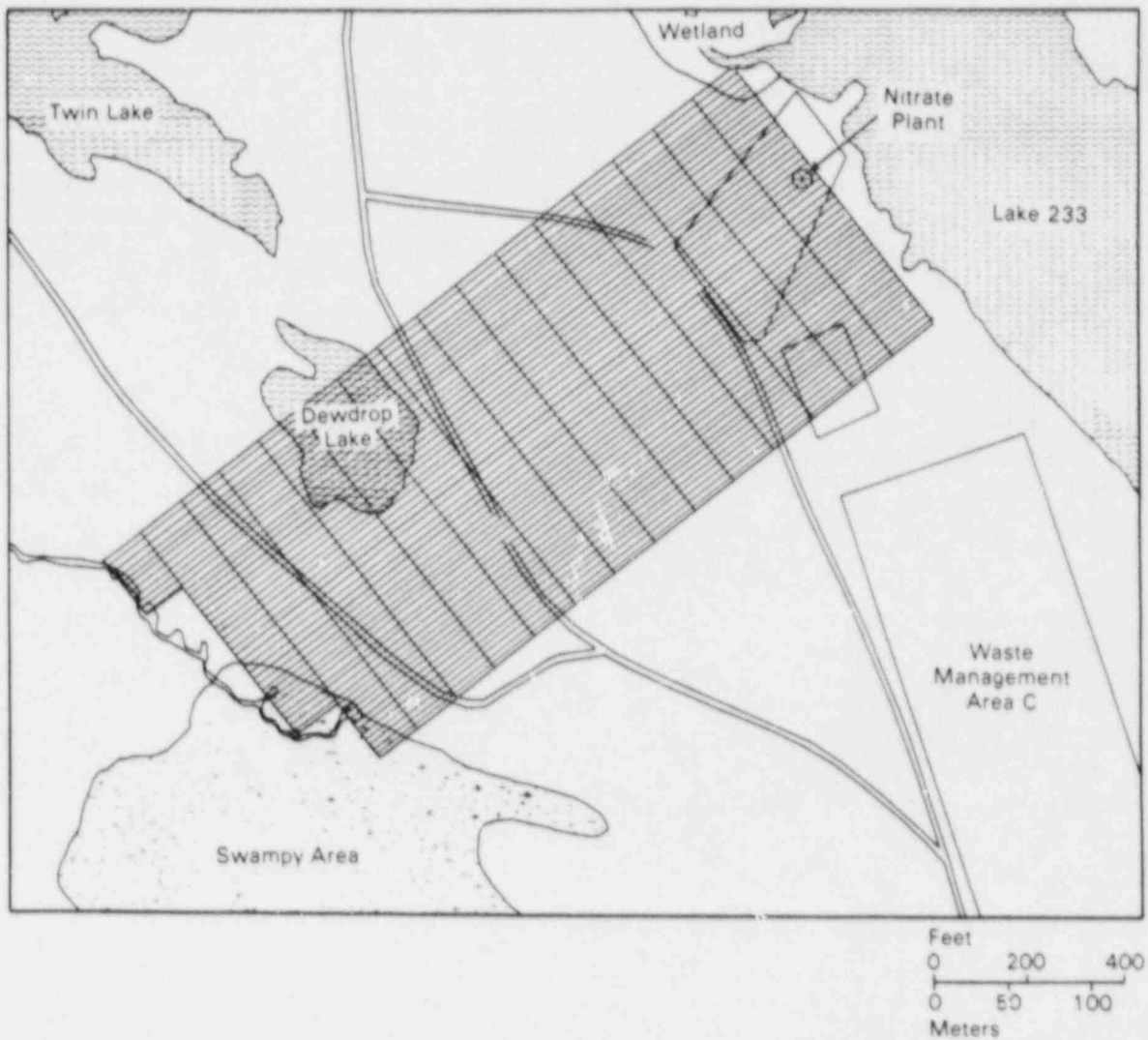


FIGURE 5.2. Revised Finite Element Grid Using a Fine (3 X) Grid Spacing in the Direction Transverse to the Principal Groundwater Flow

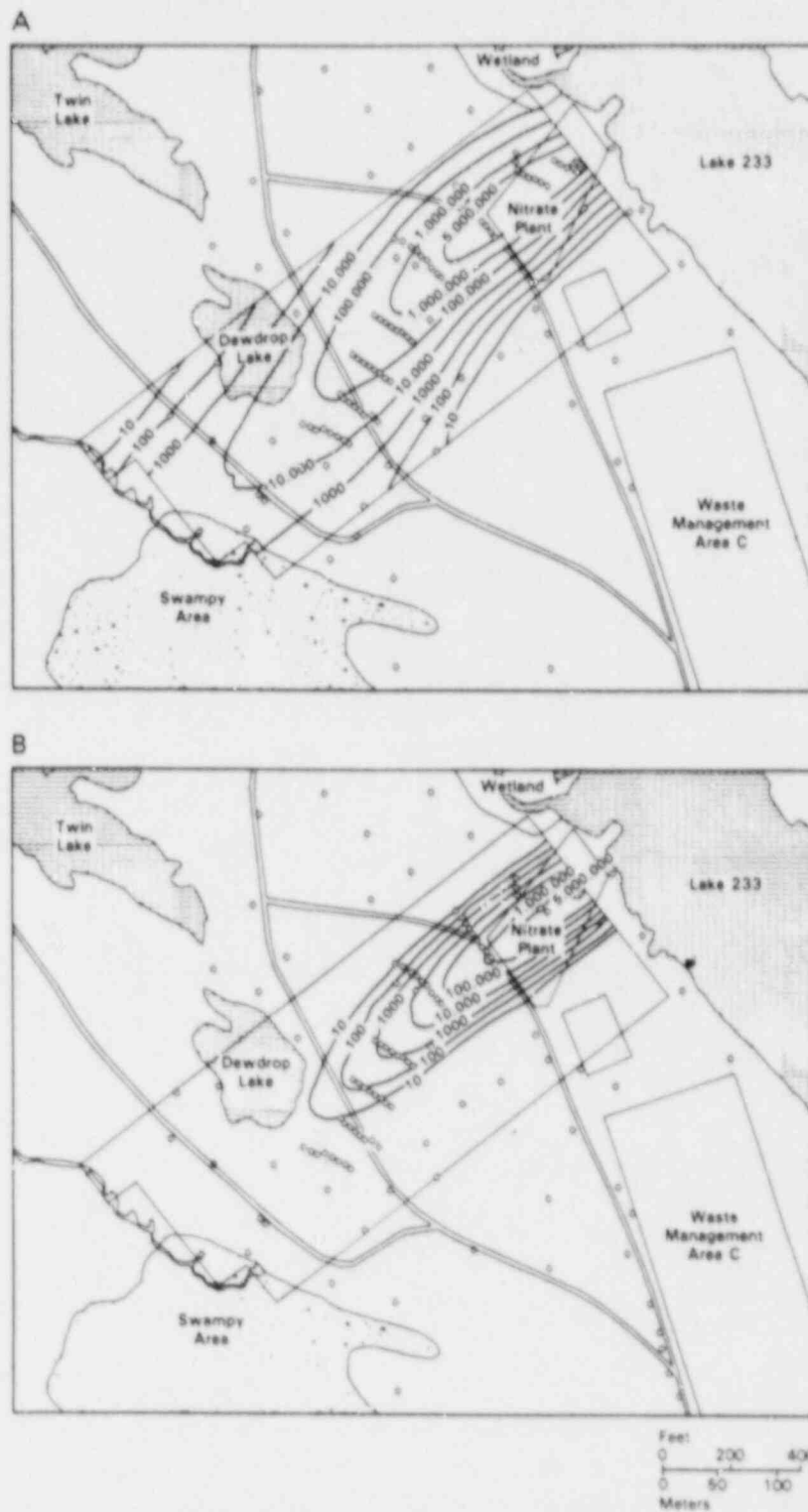


FIGURE 5.3. Strontium-90 Concentrations After 30 Years Resulting from the Detailed-Data Simulation of a 1-Year Inventory Release Using the Revised Finer (3 X) Lateral Finite Element Grid Spacing; Longitudinal Dispersivity of 1.0 m and a Transverse Dispersivity of 0.1 m; Top (A) Simulation Is for a Retardation Factor of 25 and Bottom (B) Simulation Is for a Retardation Factor of 75

## 6.0 CONCLUSIONS

Subpart D, Section 61.50(a)(2) of 10CFR61, "Licensing Requirements for Land Disposal of Radioactive Waste," states that a "disposal site shall be capable of being characterized, modeled, analyzed and monitored." In order to test the concept of "site modelability," a 30-year old low-level radioactive waste disposal site at Chalk River Nuclear Laboratories (CRNL), Canada, was used as a field location for evaluating the process of site characterization and the subsequent modeling predictions of radionuclide transport from the site by groundwater. The radionuclide source term was a limestone-lined pit (since covered with soil) which in 1953 to 1954 received approximately 3800 liters of aqueous waste containing 1000 to 1500 curies of aged, mixed fission products, including 700 to 1000 curies of  $^{90}\text{Sr}$  and 200 to 300 curies of  $^{137}\text{Cs}$ . This evaluation was performed by comparing the actual measured radionuclide migration with predicted migration estimated from hydrologic/radionuclide transport models. This comparison has provided valuable insights into the applicability of transport modeling, and in determining what level of effort is needed in site characterization at locations similar to the Nitrate Disposal Pit to provide the desired degree of predictive capabilities.

The following conclusions have been drawn regarding the process of site characterization and the ability of predictive radionuclide transport modeling to match the observed transport of radionuclides in the site groundwater over the past 30 years:

- The Nitrate Disposal Pit Site at CRNL has provided a very useful field location for testing the concept of "site modelability."
- The site characterization plan initially developed for this exercise proved to be a very useful guide in gathering the necessary data and conducting the subsequent hydrogeologic and transport modeling.
- CRNL and supplementary PNL field studies provided a well-defined map of the  $^{90}\text{Sr}$  plume which has developed over the past 30 years downgradient from the disposal site.
- Because the actual  $^{90}\text{Sr}$  groundwater plume was very narrow, a large number (over 100) of monitoring wells with multi-level piezometers previously installed by CRNL were necessary to adequately define the dimensions of the observed plume. This is consistent with the belief that relatively large numbers of multi-level monitoring wells are needed for operational and post-closure monitoring of low-level waste shallow land burial sites.
- The limited-data predictive transport modeling using the limited-data base generated by the site characterization plan provided a reasonable match with the observed downgradient  $^{90}\text{Sr}$  migration rate. Major discrepancies between the predicted versus observed migration were noted by the higher degree ( $\sqrt{3}$  X) of lateral spreading at the center of mass of the predicted plume, and the different internal distribution of  $^{90}\text{Sr}$  within the predicted plume.



- The detailed-data modeling, using the entire extensive CRNL data base for this site (over 100 monitoring wells), did not significantly alter the results and conclusions of the simulated radionuclide transport provided in the limited-data modeling. However, it should be pointed out that the similarity in results of the two modeling exercises may be more a function of the modeling approach in this investigation than in the level of detail offered by the additional data. For this particular site, a two-dimensional approach was deemed appropriate because the perceived level of heterogeneity did not warrant a full three-dimensional approach. If a fully three-dimensional approach which could account for all the subtle variations of both vertical and lateral heterogeneities offered by the detailed data were considered, slight improvements in the conceptual model of flow and transport would no doubt have slightly increased the accuracy of the model transport results. The similarity in results may also be a function of the well distribution from the detailed data set in that it is closely spaced in and around the plume and may offer little in the way of additional geologic data.
- For this particular site, which is relatively simple geologically and hydrologically, the minimal data set from the 16 test boreholes selected in the site characterization plan was adequate for providing the hydrologic and geologic framework for conducting reasonably accurate predictive radionuclide transport modeling. Fewer boreholes, although adequate for defining the direction of groundwater flow, may not have provided enough detail to adequately define the geometry of the surficial sand unit necessary for performance assessment.
- Model results suggest that a great uncertainty in this investigation was the behavior of the source term. The interaction of the acidic waste, containing complexing agents, with the limestone-lined pit and the subsequent behavior of the effluents on soils in the unsaturated zone, and soil and water in the aquifer system are poorly understood. The inability of the model to properly predict the temporal and spatial variability of adsorption-desorption characteristics of the radionuclides in question resulted in major discrepancies between predicted and observed concentration distributions. These discrepancies indicate the inadequacy of laboratory-derived distribution coefficients in simulating these types of interactions in a model. A better understanding of the waste leachate would have to be acquired with further field and laboratory testing that would evaluate both the temporal and spatial interaction of the waste form with the natural environment.
- Model results also pointed to uncertainties related to the dispersive characteristics of the sediments being modeled. The difficulty of selecting a level of dispersion appropriate for both the types of sediments and the field scale being modeled is a common problem. This understanding could be improved with field (tracer) tests or modeling investigations of contaminant plumes in the same soils or in soils of similar texture and sorting. Laboratory values have been found to be generally inadequate for the scale of field modeling generally used in most studies. A better understanding of the appropriate level of dispersion for a particular field situation will generally lead to a more appropriate model design (i.e., grid discretion) which would help minimize the problems associated with numerical dispersion.

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APPENDIX A

SITE CHARACTERIZATION PLAN  
CHALK RIVER NITRATE DISPOSAL PIT SITE

NRC Fin B 2862

DEMONSTRATION OF PERFORMANCE MODELING  
OF LOW-LEVEL WASTE SHALLOW LAND BURIAL

Task 1

SITE CHARACTERIZATION PLAN  
CHALK RIVER NITRATE DISPOSAL PIT SITE

January 1985

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Pacific Northwest Laboratory  
Richland, Washington

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## PRE FACE

### BACKGROUND INFORMATION

Section 61.5, Subpart D of 10CFR61, "Licensing Requirements for Land Disposal of Radioactive Waste," states that any future low-level waste shallow-land burial facility "shall be capable of being characterized, modeled, analyzed and monitored." Implicit in this requirement is the ability to utilize solute transport modeling to predict the movement of radionuclides in groundwater from a disposal facility. However, recent studies (1,2,3, 4,5) at low-level waste (LLW) shallow-land burial facilities and other slightly contaminated sites have shown discrepancies between predicted versus actual radionuclide migration rates in the ambient groundwater. Because of these discrepancies, there is a general understanding, within the scientific community, that site performance modeling may need further refinement to more accurately predict radionuclide movement from a potential disposal site. Therefore, predictive transport models need to be evaluated under actual field conditions to assess their accuracy and identify the weak links in the modeling process.

One of the ways to test the concept of site "modelability" is to compare predicted radionuclide movement using hydrogeochemical modeling with actual observed radionuclide migration at field sites where radionuclides have been in the ground for many years. Such comparisons will yield insights as to the reliability of models which an applicant might reasonably use to predict LLW disposal site performance at proposed facilities.

At the Chalk River Nuclear Laboratories (CRNL), Ontario, Canada, a number of low-level waste shallow-land burial facilities have been in existence for about 25-30 years. These sites could prove to be useful for testing the concept of site "modelability." Following discussions with CRNL personnel, a cooperative research program was established and two disposal sites having slightly contaminated groundwater plumes were selected for potential study. This exercise addresses the Nitrate Disposal Pit site, a low-level disposal facility which received liquid wastes containing approximately 1100 curies of mixed fission products during 1953-1954.

### PROJECT PLAN AND OBJECTIVES

The plan is to approach this site as though it were a prospective shallow-land burial (SLB) site to be licensed under the requirements of 10CFR61. A "pre-operational" site performance assessment involving hydrogeochemical modeling would first be conducted at the Nitrate Disposal Pit to predict the temporal movement of radionuclides in the groundwater. The predicted movement would then be compared with the actual radionuclide migration which has occurred over the past 30 years to assess the suitability of the modeling. The "modelability" concept (defined below) for this site will then be evaluated.

Under the assumption that very little is previously known about this site, a site characterization plan would first be prepared which describes the geologic/hydrologic/geochemical measurements and information needed to



construct a conceptual model in order to perform predictive modeling of the radionuclide transport in the site groundwaters. In the preparation of this site characterization plan it is assumed that there are few, if any, existing wells at the site. Only basic data concerning background geological, hydrological, and geochemical parameters would be used as input to the preliminary conceptual and/or numerical models of the subject site. The procedure for determining the number and location of data points is one of professional scientific judgment, as opposed to a rigid statistical approach. This flexible process utilizes an iterative methodology in the collection and analysis of characterization data, such that the conceptual model is continually refined and revised as required. This approach is consistent with state-of-the-art efforts to characterize geohydrologic environments. These models would, in turn, be used to guide further development of the characterization plan. The characterization plan is designed to provide the information necessary to build both a defensible conceptual model as well as defensible flow and transport models of the subject site.

After completion of the site characterization plan, available hydrological and geochemical data at these sites, previously generated by CRNL investigators, will be utilized to fill the data requirements of the plan and to predict the movement of selected radionuclides from the site. Finally, the predicted radionuclide migration will be compared with actual migration determined from field sampling and analyses of the slightly contaminated groundwater plumes by CRNL and Pacific Northwest Laboratory (PNL) personnel.

In this project, two levels of numerical modeling are perceived. At the first level of modeling, only a portion of the existing data set, conforming to the well placement selection contained in this site characterization plan, will be used to develop a relatively simple groundwater flow and transport model, such as would be done in an actual siting assessment and guidance project. That portion of the total data set to be used in a typical siting assessment guidance project is described in the accompanying plan. This plan has been designed to provide sufficient data to model the site. The level of investigation is kept to a minimum, as though licensing were being undertaken in an effective yet minimum cost manner. The numerical model will be assessed to assure that no inconsistency exists, e.g., the mathematical simulation codes used are commensurate with the assumptions made in the conceptual model and the available data. The transport model will be exercised using a radiologic source-term equivalent to actual wastes disposed at the subject site during the early 1950s, and the movement of selected nuclides through the system will be modeled. Simulation will then be carried forward and compared with present day distribution of contaminants as determined by field sampling. This comparison is done in order to help assess the usefulness of this level of site assessment modeling in (1) helping to understand the site through sensitivity studies, and (2) providing a means to bound the possible consequence estimates.

The second level of modeling will be a more detailed state-of-the-art effort, carried out using the entire, extensive and available characterization data set along with the expertise of Atomic Energy of Canada, Ltd.

(AECL) personnel (regarding important phenomena which could have been deduced without reliance on observation of the actual waste migration data). The conceptual model will be adjusted as necessary to take advantage of the 30+ years of experience at the subject site, and appropriate alterations in the modeling technique made.

Finally, a comparison of the results of both modeling efforts will be made, and those results compared with the field observations of actual migration. This comparison will provide insight into the level of effort necessary to meet the regulatory requirement that a site be "modelable." Additionally, this comparison will serve to differentiate the advantages, if any, of a more detailed characterization and assessment activity in the early siting assessment phases.

#### PROJECT LIMITATIONS AND PRE-CONDITIONS

Certain limitations are applicable to this task and the final results:

- Only existing data sets and/or knowledge that could be obtained through site characterization efforts based on reasonable time and cost are to be used in gaining the understanding required to develop the conceptual and numerical models of the site.
- Specific data requirements defined by the site characterization plan may not be available from existing sources, thereby necessitating further simplification of the conceptual and numeric models.
- No attempt is or will be made to assess the overall suitability of the study site as a Low-Level Waste Shallow-Land Burial site. The objective is to address the question related to the regulatory requirement that a site be "modelable."

#### CRITERIA FOR EVALUATING THE "MODELABILITY" CONCEPT

Because the purpose of this task is to address "modelability," it is important that some mutually acceptable criteria for judging that concept be developed prior to the actual modeling exercise. This will assure an objective answer to the "modelability" question. Potential criteria for judging successful modeling are as follows:

- The physics of groundwater flow and transport (as currently understood and/or can be deduced from available site data) have not been violated.
- The conceptual model has been developed using a data base sufficiently large such that additional data would have minimal impact on the model.
- The results of the transport analysis are more realistic than a "back-of-the-envelope" calculation and yet conservative, in that they allow the bounds of the expected results to be narrowed through sensitivity studies based on the quality of the characterization data. In this context, conservative means that transport will

occur, yet the concentration of contaminants at the monitoring points, their first arrival, maximum transport rate, and duration will be predicted to be greater than or equal to those observed in the field.

The report which follows constitutes the initial task of this project, the Site Characterization Plan, which a potential applicant might follow for generating the necessary information needed to predict the performance of the site. The results of the predictive modeling and the comparison with actual field observations and measurements will be provided in subsequent reports.

SITE CHARACTERIZATION PLAN  
CHALK RIVER NITRATE DISPOSAL PIT SITE

1.0 INTRODUCTION

Pertinent background information and a description of the design and objectives of the entire project are given in the Preface. This report presents only the site characterization plan which will be used to guide the hydrologic modeling efforts.

The site characterization plan presented herein is developed under Task 1 of NRC Fin B2862, "Demonstration of Performance Modeling of LLW Shallow Land Burial," which is described as follows:

Task 1: Assessment of information needs/site characterization plan

The performing organization is to develop a plan for characterizing the Nitrate Disposal Pit Site at Chalk River as if it were an applicant and the Nitrate Disposal Pit Site was the site to be licensed for LLW SLB disposal. The plan should indicate clearly (and be limited to) the geologic, hydrologic, and geochemical information that would be gathered and the locations on the site where measurements would be taken. The performing organization would employ a statistically based data gathering and analysis plan showing the information needed to characterize a new LLW land disposal site and to predict its performance. Upon completion of this task, the performing organization shall submit, for NRC review, a report describing the characterization plan. The report should show the location of measuring points on maps and it should indicate the measurements/data that would be collected at those points.

This site characterization plan was developed for the Nitrate Disposal Pit Site at Chalk River, although the measurements called for would be identical at the other Chalk River disposal areas. The only thing different would be the measuring points. Any additional modeling of sites at Chalk River, such as the "A" Disposal Site, would apply insights gained at the Nitrate Pit Site. A characterization plan for any additional site would be provided in a future report.

Guidance for determination of those factors necessary to characterize a site for this project were derived from NUREG-0902, Site Suitability, Selection, and Characterization, 1982. The initial steps in site selection have not been addressed, and it has been assumed that the Nitrate Disposal Pit Site has passed all the screening tests necessary prior to detailed site characterization and evaluation.

This characterization plan focuses on the data requirements to initiate and implement predictive transport modeling of selected radionuclides within the hydrogeologic system at the Nitrate Disposal Pit Site and, therefore, is limited to geologic, hydrologic, and geochemical studies necessary to model the site and evaluate the containment and transport of selected radionuclides within the underlying groundwater system. An important aspect of this task is that no new data are to be developed, all data will be derived from existing sources made available through AECL's Chalk River Nuclear Laboratories.

## 2.0 TASK STRUCTURE AND SEQUENCE

Under normal selection and characterization procedures, several steps precede the effort outlined in this plan. A regional assessment of general geologic and hydrologic features would provide initial selection of a number of potential sites. Data collected during the earlier phases of the site selection process include:

1. Regional and local geology, including stratigraphy and structure;
2. Regional and local hydrology, both groundwater and surface water;
3. General meteorology gathered from the closest weather stations.

Preliminary field observations would then be conducted at these initially selected sites to assure that the regional information on surface geology, hydrology, meteorology, and geochemistry was, in fact, as presented in the regional assessment. The more optimal of those sites would then be subjected to more rigorous appraisal, further defining the appropriate data bases. This more rigorous appraisal would provide the starting point for the site specific characterization outlined here. The present study assumes that screening of several potential sites has shown that the Nitrate Disposal Pit Site is worthy of further investigation.

Our approach to site characterization is shown in the flow chart in Figure 1. The overall plan begins with the formation of the regional and local conceptual models using available data, such as topographic maps and logs from any wells previously drilled in the area. Based upon the initial conceptual model and professional judgment, the number and location of the initial field boreholes are determined and the locations prioritized.

The characterization program will then proceed through the drilling and evaluation process until sufficient data are available for initial numerical modeling. At this point the initial numerical model is developed and utilized to guide development and refinement of both the conceptual and numerical models. This process continues until the models, which include the stratigraphy, hydrology, geochemistry, and site structure have been confirmed. This end point is reached when additional data will have minimal impact on the models. Application of rigid statistical analysis at this point in a characterization effort is considered premature.

The characterization program then proceeds into the flow and transport analysis using the finalized models. The flow and transport analysis and its refinement continues until the numerical model reflects the flow and transport expected at the site under the observed field parameters and conditions. It should be noted that the degree of acceptable uncertainty associated with the modeling process is site specific. For simple hydrogeological systems, acceptable uncertainty is expected to be much less than for a more complex system, e.g., a sand-dominated system vs. a karstic limestone. Finally, performance assessment is conducted to predict radionuclide migration and estimate associated uncertainty through sensitivity studies.

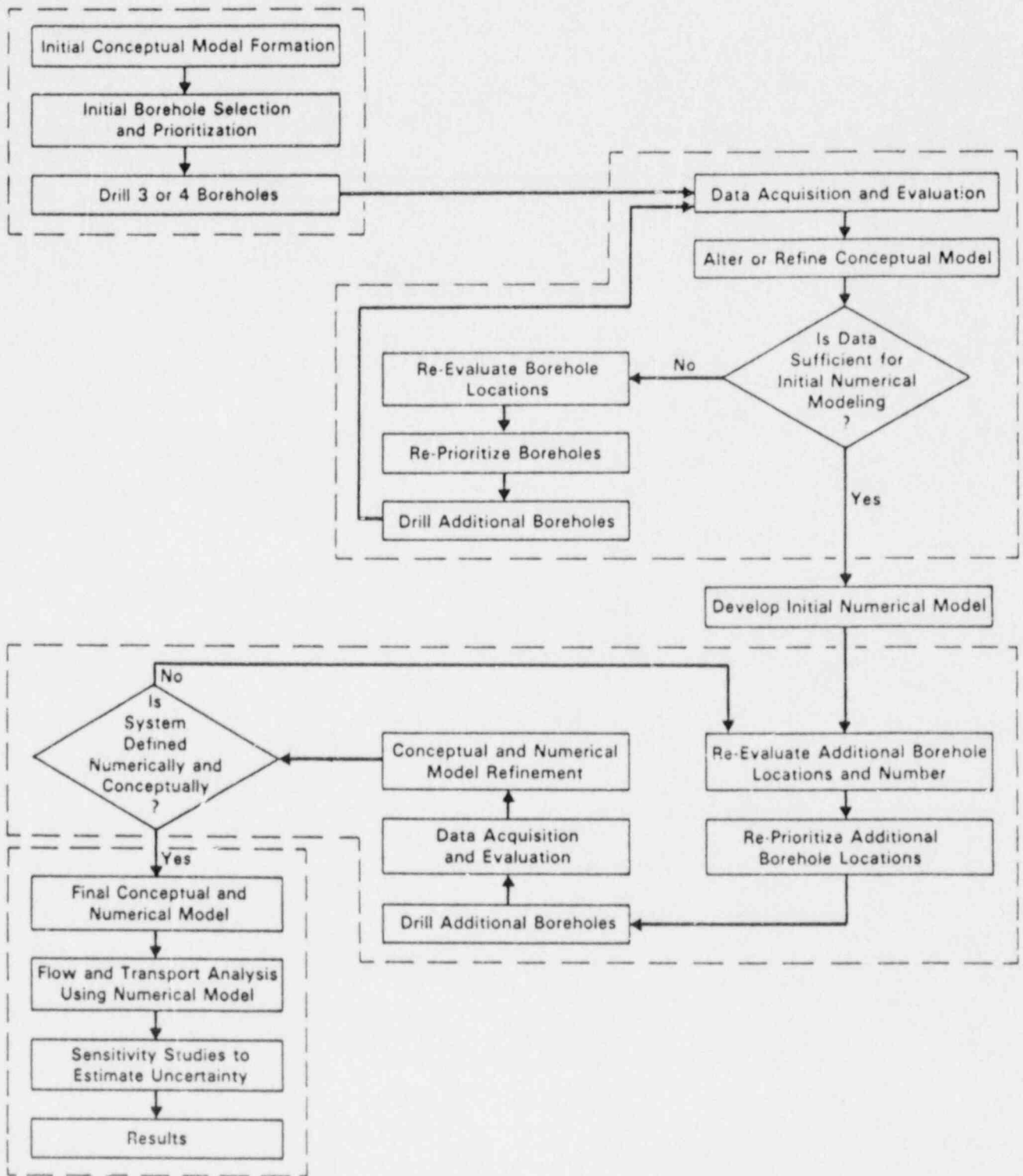


FIGURE A.1. Flow Diagram for the Site Characterization Process

The activities, data requirements, and constraints associated with each of the various steps shown in the project flow chart are described in the following sections, beginning with the formation of the regional and site conceptual models.

## 3.0 INITIAL UNDERSTANDING OF THE SITE

### 3.1 Regional Hydrogeology

The regional hydrogeology is initially evaluated in order to develop a regional conceptual model of the surface water and groundwater regimes and the geology. This information is developed by reviewing available literature, studying topographic maps and aerial photographs, and conducting field observations. The regional assessment permits a better understanding of the large-scale geologic structures and groundwater driving forces which influence the local hydrogeology at selected disposal sites.

#### 3.1.1 Geology

The Chalk River site operated by AECL is located in the Province of Ontario about 300 miles north of Toronto (Figure 2). The general geology is typified by shallow sandy sediments overlying a tight glacial till, which in turn overlies the bedrock. Sediment thicknesses are on the order of a few tens of feet. The bedrock is crystalline and essentially impermeable, supporting a number of small-to-large lakes in the vicinity of the subject site. Although there are visible fractures in the bedrock which could theoretically support fracture flow, the contrast in hydraulic conductivity between the bedrock and overlying sediments is so great that, for the purposes of this project, the bedrock is treated as though it were impermeable. However, the bedrock does play a major indirect role in the transport of groundwater by providing a subsurface topography which ultimately determines the occurrence and thickness of the more recent sediments through which the horizontal groundwater flow occurs. Because of this control, definition of the bedrock surface is essential to the modeling effort.

#### 3.1.2 Hydrology

The hydrology of the region is dominated by numerous small drainage basins which ultimately discharge via small streams to the Ottawa River. These smaller drainage basins often contain moderate-to-small sized surface water bodies which serve to maintain a recharge source for the associated groundwater systems. The local bedrock has been scoured by glacial action, resulting in an undulating, yet gently sloping surface which serves to direct and control the movement of groundwater.

#### 3.1.3 Regional Conceptual Model

The regional conceptual model is one which includes numerous small-to-moderate size drainage basins which ultimately discharge to the regional surface water network. Groundwater flow in these small basins is recharged primarily through precipitation (77 cm/yr), but is maintained through seepage from the numerous lakes. The groundwater flow system is overwhelmingly one of near surface flow, the bedrock being essentially impermeable. Flow in the groundwater system is through fine sandy sediments and to a lesser extent through a till that overlies the bedrock surface. The bedrock surface plays a major role in controlling the direction of groundwater flow;



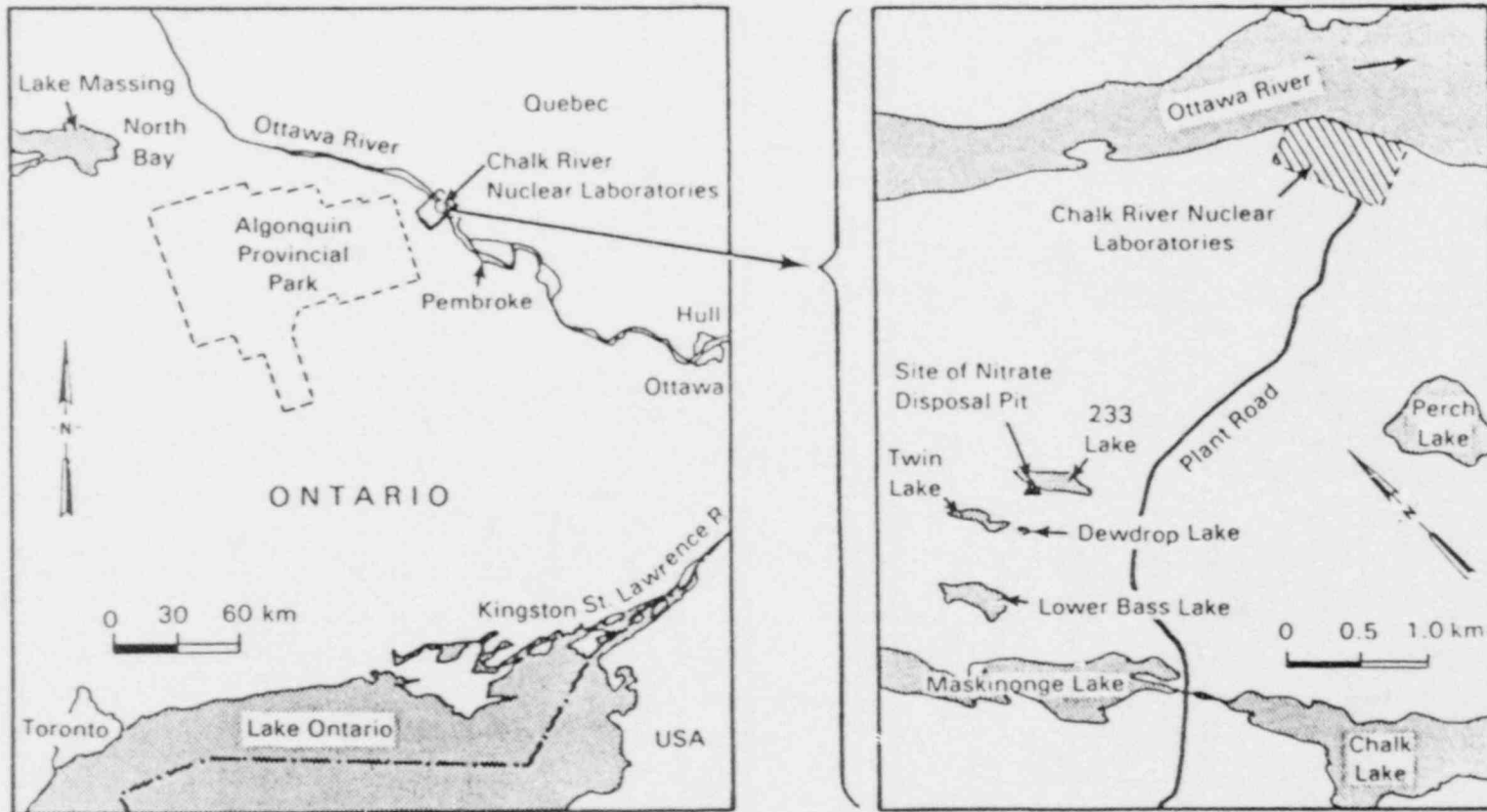


FIGURE A.2. Location of Chalk River Nuclear Laboratories and the Nitrate Disposal Pit Site

springs and swampy areas are prevalent where bedrock crops out or approaches the land surface.

### 3.2 PROPOSED DISPOSAL SITE

The regional assessment of the available hydrogeologic information is followed by an evaluation of the local hydrogeologic conditions at the selected disposal site. The site-specific information is assessed to see how it conforms to the regional conceptual model and to evaluate any localized anomalies or unusual features.

#### 3.2.1 Local Hydrogeology

The hydrogeology of the proposed disposal site conforms with the regional conceptual model. The disposal pit site will lie near the boundary of a catchment area that drains, via Maskinonge Lake, into the Ottawa River about 8 km downstream (see Figure 3). The pit will be excavated in a dune ridge on a terrace approximately 50 m above Maskinonge Lake and approximately 1 km east of the lake. The site is underlain by fine-to-medium grained sands, interbedded with some silty to clayey lenses. These materials overlie a tight clayey till, which in turn overlies the crystalline bedrock. The hydrology of the proposed site is one of predominately groundwater flow. A moderate sized, shallow lake, Lake 233, is immediately east of the proposed site (see Figure 4). To the west, Dewdrop Lake, a small surface water body, is found at an elevation about 8 m lower than Lake 233. Immediately south of Dewdrop Lake several springs issue from the sediments (see Figure 4). The resultant stream flows east out of the small drainage basin through a gap bounded by bedrock outcrops into another unnamed lake. The area in which the springs issue is generally swampy and typical of a groundwater discharge zone in this environment.

#### 3.2.2 Description of Anticipated Wastes and Disposal Pit

The source material for the disposal site will be an ammonium nitrate solution from fuel reprocessing operations, consisting of approximately 1000 gallons of acid waste containing complexing agents. Activities at the disposal site include decomposition of the ammonium nitrate present in the solution and concentration of the fission products by evaporation. The condensate from the evaporation process and any waste solutions will be drained to a small pit lined with limestone. The limestone will serve two purposes; it will tend to neutralize any acidic wastes and it will serve as a sorption media for 90-Sr present in the effluent solutions.

Potential source terms at the site could range from relatively high levels of fission products in the original feed solution, if a spill or overflow should occur during processing, to the relatively low levels of contamination associated with the condensate solution from the evaporation process. The three primary components in the feed solution will be 90-Sr, 88-Y, and 137-Cs, with other fission products present at concentrations which are assumed to be in proportion to their relative fission yields.

It is estimated that some 1100 Ci of fission products will be disposed at the site and subsequently available as a source term for future

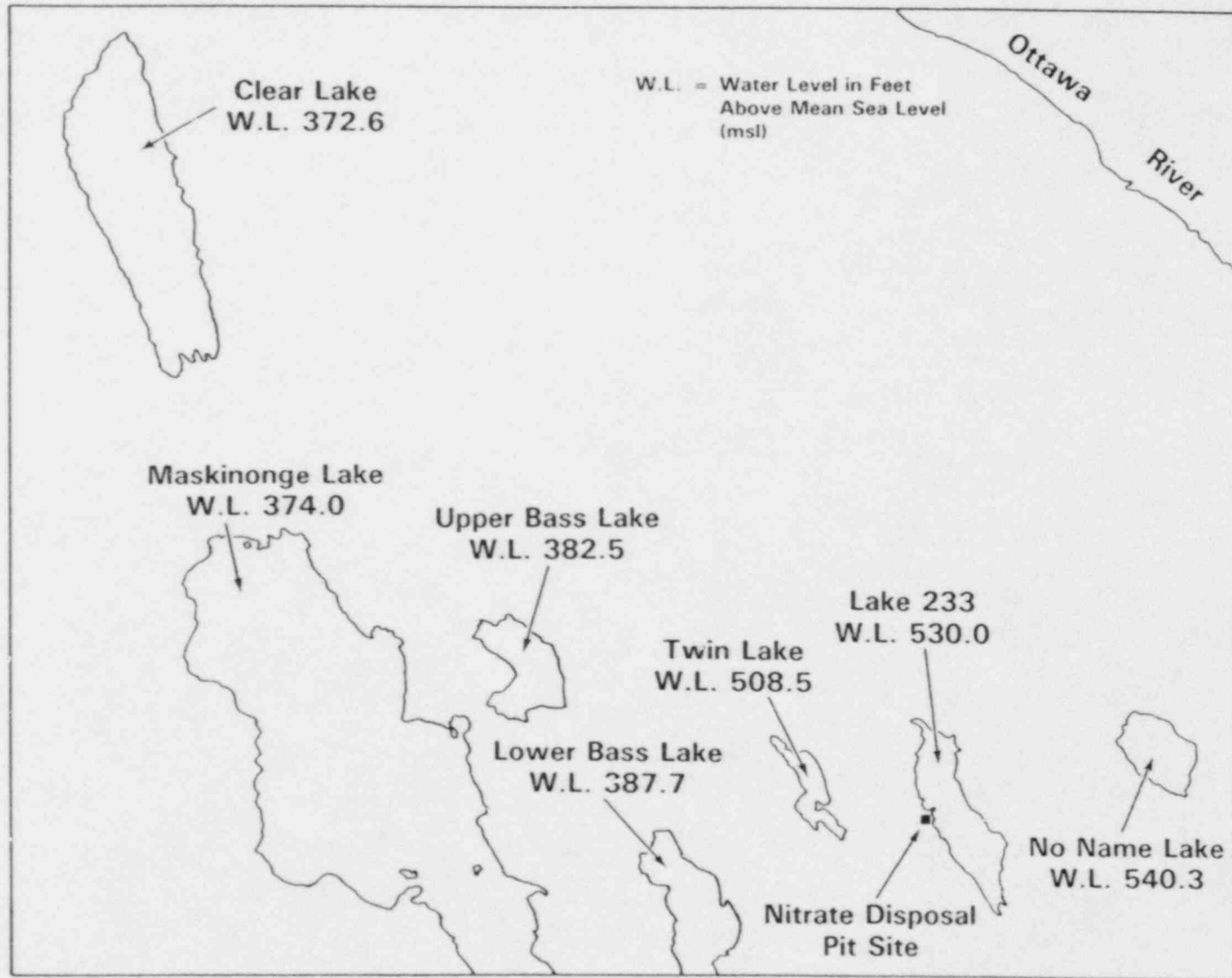
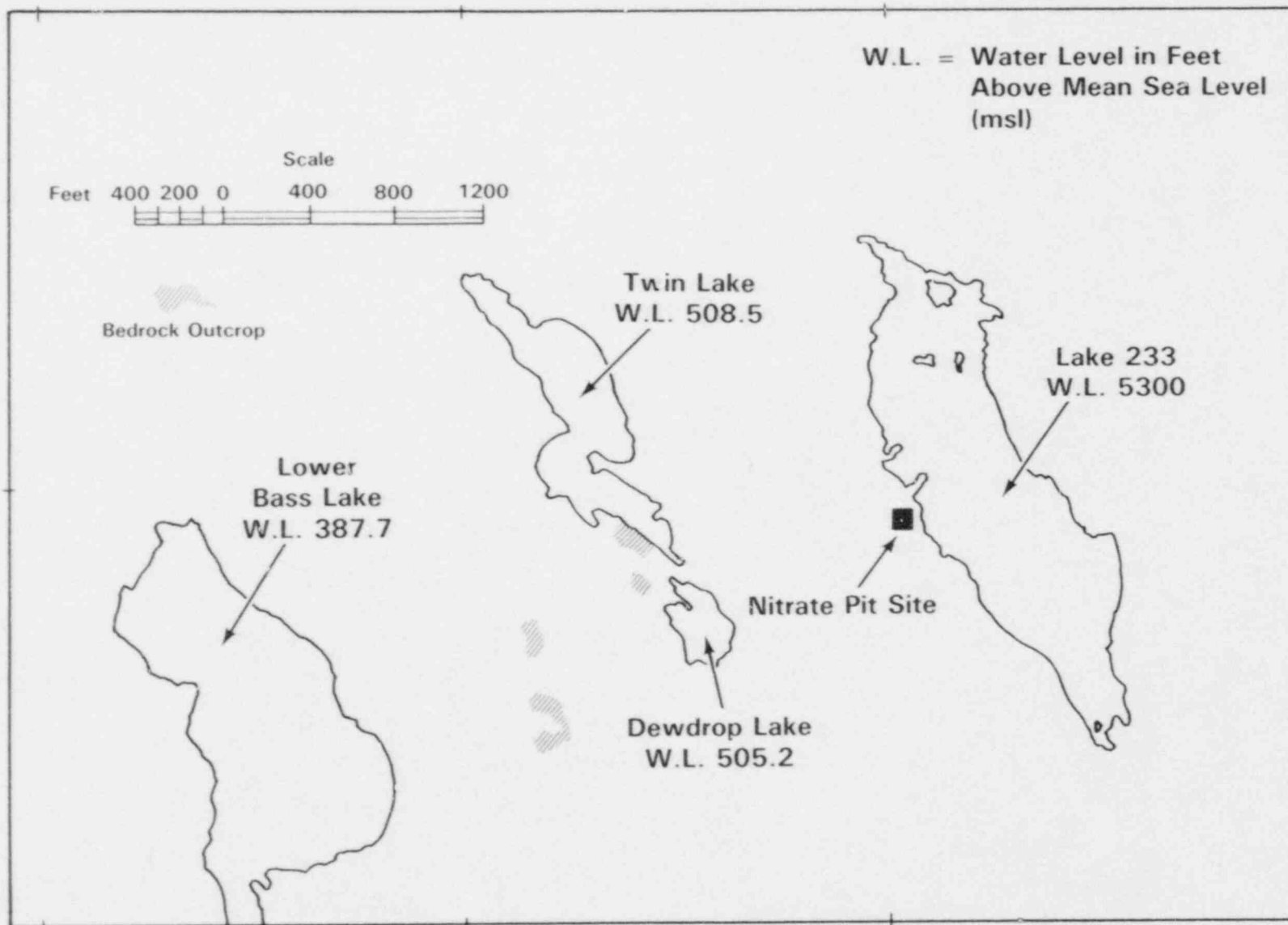


FIGURE A.3. Location and Water Levels of Surface Water Bodies Near the Nitrate Disposal Pit Site



A-16

FIGURE A.4. Regional Topography with Approximate Location of Bedrock Outcrops

migration. The portion disposed at the site will be a small fraction of the radioactive inventory processed at the site.

### 3.2.3 Initial Conceptual Model

The potential area of concern from a groundwater transport standpoint is areally small, extending about 610 m in a west-southwesterly direction (Figure 4). Existing surface water bodies are controlled by the relatively shallow bedrock surface, their existence being an indication of the limited hydraulic conductivity of the underlying rocks. These lakes serve as sources and sinks for the local groundwater systems, providing relatively stable constant head boundaries for the flow system. The majority of the groundwater flow is through the uppermost sediments overlying the till and bedrock. Some flow occurs within the till unit, but the clay-rich nature of the till limits the total amount and rate. Precipitation falling on the land surface during the warmer months of the year readily infiltrates the sandy sediments, with the only losses being to evapotranspiration. During colder periods, extensive and deep frost precludes direct infiltration and runoff occurs during spring melting periods.

The Nitrate Disposal Pit Site, chosen for this modeling exercise, is located on a slight ridge immediately adjacent to Lake 233 (Figure 4). The proximity of the site to this surface water body at first suggests that groundwater flow should be toward the lake, and thus readily accessible to the surface environment. However, it should be noted that the level of Lake 233 is approximately 8 m higher than nearby Dewdrop Lake; also swampy areas with small streams draining them are present near Dewdrop Lake (Figure 4). These additional pieces of information indicate that groundwater flow at the pit site may actually be toward the southwest and away from Lake 233, and that the lake may serve as a relatively constant head recharge boundary for the local groundwater system. For this reason, the majority of the "proposed" monitoring and observation wells should be placed in the southwestern quadrant from the pit site. The initial wells would be placed along the shortest travel path from the Nitrate Disposal Pit Site toward Lake 233 in order to determine if groundwater flow is in this direction.

## 4.0 METHOD OF INVESTIGATION

This section of the report presents the data requirements and field activities necessary for conducting the hydrogeological modeling. The methods proposed for this effort are based on best professional judgment. Rigorous statically based approaches to determine the number and locations of wells were considered. However, it was concluded that the qualitative nature of the decision-making process in early stages of site characterization precludes the use of these approaches. The iterative approach used to develop both the conceptual and numerical models provides flexibility, allowing the optimum use of resources. The application of sensitivity studies to ascertain estimates of uncertainty is reserved for final modeling efforts. The data requirements are listed in Section 4.1 and the means of acquiring these data are discussed in Section 4.2.

### 4.1 DATA REQUIREMENTS FOR SITE CHARACTERIZATION

The data required to characterize the proposed site are centered around those necessary to define and refine the conceptual model as outlined above. These data are to provide the necessary input to numerically model the site for the prediction of radionuclide transport. Because of the limited areal extent of the proposed site, the data gathering activities are concentrated near the actual proposed disposal site, and more disperse in the downgradient direction.

#### 4.1.1 Geology

Site specific geologic sites are centered around the determination of those features which ultimately control the movement of groundwater.

##### 4.1.1.1 Bedrock Surface

Determination of the configuration of the bedrock surface is essential to the understanding of groundwater flow directions and the ultimate controls on the system being modeled.

##### 4.1.1.2 Continuity of Sedimentary Units

In order to successfully model the site numerically, the distribution of the primary geohydrologic units, in this case the glacial sediments overlying the bedrock, must be known. These data provide a means of conceptualizing the three-dimensional distribution of the identified units.

##### 4.1.1.3 Structure

It is essential to recognize and account for any structural effects that may be present in the underlying bedrock. These structural features may control the presence or pathway taken by potential contaminants. These structures may range from joint sets to fault-related features.

## 4.1.2 Hydrology

The data requirements listed here are those which are essential to provide input into the development of the conceptual and numerical models of the site.

### 4.1.2.1 Aquifer Thickness

The saturated thickness of the aquifer being addressed is essential in the determination of hydrologic flow.

### 4.1.2.2 Hydraulic Head

Groundwater hydraulic head measurements, including gradients and directions of flow, are derived from accurate water-level measurements in test wells. Map plots of the groundwater surface will aid in the development of the conceptual model of groundwater flow at the site.

### 4.1.2.3 Water-level Fluctuations

Magnitude and time distributions of water-level fluctuations relate to the changes in storage within the groundwater system and to the overall water-budget of the system.

### 4.1.2.4 Hydraulic Conductivity

This factor is absolutely essential to any hydrologic analysis. It is proposed that multiple aquifer tests be run on several individual well structures to ascertain the statistical variability present in the determination of this parameter. At several locations this parameter should be determined as a function of depth, which will facilitate three-dimensional conceptualization of the flow system. The range of values determined serves to bound the values used in the numerical modeling.

### 4.1.2.5 Storativity

Storativity or specific yield of the sediments is their capacity to hold water, and is a basic parameter essential for modeling of the site.

### 4.1.2.6 Effective Porosity

This parameter, although difficult to obtain, is a controlling factor in the actual rate of groundwater transport and thus the transport of contaminants.

### 4.1.2.7 Dispersion Coefficients

Initial estimates of dispersion coefficients describe the horizontal spreading of a contaminant front as it moves through a groundwater system. Field determinations of this parameter provide the starting point for its inclusion within the modeling effort.

#### 4.1.2.8 Vertical Distribution of Hydraulic Head

Changes in head with depth are indicative of recharge/discharge conditions and thus serve to describe the flow system in the third dimension.

#### 4.1.2.9 Soil Conditions

Partially saturated conditions in the soil column:

- a. Soil moisture versus depth
- b. Hydraulic conductivity versus soil moisture

#### 4.1.3 Geochemistry

Although the distribution coefficient ( $K_d$ ) approach will be utilized in the modeling of the subject site, in order to model the transport of contaminants with any degree of certainty it is essential to establish the initial geochemistry of the system in question. The following information, some of which is only used by project scientists to provide insight into parameter initiation and boundaries, is considered mandatory to perform even a simple analysis of the retardation of radionuclides by the soil.

##### 4.1.3.1 Mineralogical and Chemical Characterization of Rock/Sediments

The mineralogical/chemical nature of the rocks and sediments is a major controlling factor in the distribution of retardation potential within the hydrogeologic system. For transport modeling the relative amounts and distribution of minerals along the flow path should be known. Analysis of cation exchange capacity (CEC), hydrous iron oxide content, and other sediment chemical attributes would be carried out on selected samples representative of the hydrogeologic units. Analyses carried out on several samples will provide some statistical basis regarding the variability of these factors.

##### 4.1.3.2 Distribution Coefficients

This simple approach to the retardation of radiocontaminants is the best available method for an early analysis of contaminant transport. The analyses would be run on several sediment samples obtained from the boreholes, with samples selected so as to represent the hydrogeologic units identified. If there is a wide range in the chemistry of the anticipated waste leachate and/or groundwater, the sorption experiments may also have to be performed on several types of water which simulate the range of chemical composition.

##### 4.1.3.3 Chemical Analysis of Groundwater/Waste Leachate

Chemistry of the native groundwater or soil water, including trace element analysis, would be undertaken. The results of the analyses should show a balance within  $\pm 5\%$  between cation and anion milliequivalents. Laboratory values of the distribution coefficients are accurate only as long as field conditions are reasonably approximated; any changes in the chemistry of the natural system must be known to assess the expected movement of contaminants within the hydrogeologic system.



#### 4.1.3.4 Organic Content of Sediments and Groundwaters

This analysis should be run on several samples to ascertain the distribution of natural and man-made organic material throughout the system. Organic complexation of disposed radionuclides may be controlled by the presence of this matter, thereby greatly affecting the movement of these constituents.

### 4.2 IMPLEMENTATION OF THE PLAN

The exploration boreholes are the basic tools through which the data requirements described above will be fulfilled. The geological, hydrological and geochemical parameters at the site will be defined from analyses of core materials obtained during the drilling process, and also by water sampling and measurements after the boreholes have been drilled. The multiplicity of needs for each borehole (geologic, hydrologic, and geochemical) requires that the locations of the holes be carefully considered. Financial considerations further require that the preliminary number of boreholes be minimized. Hydrogeological considerations dictate both an adequate number of wells and a carefully selected geographic placement pattern, based on an iterative drilling and evaluation program. Our initial decision, after consideration of the above factors, is to drill, sample, and test up to fifteen (15) selected locations on the Nitrate Disposal Pit Site (Figure 5). The Nitrate Pit Site is areally small, being bounded on the west by a lake and to the east by an apparent groundwater discharge area. Therefore, the projected fifteen boreholes should, with their areal coverage, adequately describe the system, if it is as simple as initially conceptualized. The well placement pattern, as shown in Figure 5, and plan are based upon the conceptual model formed from the initial assessment. This site conceptual model is tentative and assumes a relatively simple system at the Nitrate Disposal Pit Site. Should this assumption be disproven during the initial well placements, this pattern and numbers would be revised.

The highest priority boreholes will be drilled initially with data acquisition and interpretation concurrent with the drilling process. The data will be evaluated as the wells are drilled, resulting in site conceptual model refinement. Borehole numbers and locations would be redefined as required, and any additional boreholes prioritized. Then, the next several boreholes in priority will be drilled. The field program will proceed around either loop, as shown in Figure 1, until the initially selected fifteen wells are drilled, or the site has been defined. At that point, the models are reevaluated. If professional judgment indicates that this site is now defined, the program moves forward to the final numerical modeling phase. If, on the other hand, the initially projected fifteen wells indicate a much more complex set of parameters exist at the site than were envisioned, additional boreholes would be required and well emplacement activities would continue until the site conditions are defined or the site shown to be so complex as to be unacceptable for waste disposal.

#### 4.2.1 Well Placement Pattern

It must be stressed again that no new data will be developed during this study; all data to be utilized will be pre-existing and provided by AECL. Specific data desired from these borings include the following:

1. The specific stratigraphic units present, including the fine changes in lithology and fabric which could affect the transport of contaminants;
2. The three-dimensional distribution of the units recognized under 1 above;
3. The saturated thickness of units penetrated. This information is essential to development of a realistic conceptual model and a representative mathematical model;
4. The configuration and integrity of the bedrock surface. This surface is presently thought to control the majority of groundwater flow at the subject site.

The sequentially phased borehole drilling program, as described above, would start with the highest priority locations as follows:

Wells 1,2,3,4      These wells are located in the closest proximity to the disposal site. From the topographic analysis, the groundwater flow at the disposal site would appear to be away from Lake 233 and towards the southwest. However, flow towards Lake 233 cannot be discounted and the initial borehole placements will provide the measurements to elucidate the flow pattern in the immediate vicinity. These wells will provide geologic, hydrologic, and geochemical data on the environment closest to the disposal area. If groundwater gradients are found to be toward Lake 233, the project will be abandoned.

Wells, 5,6,7,8,9      These wells will serve to further define the bedrock surface and the distribution of the underlying sediments. Extensive sampling of these sediments will provide detail on the geochemistry of the units. Hydrologic tests will be run on all of the wells to provide the needed parameters and assess their variability in the lateral direction. Careful attention will be paid to the geohydrological positioning of these wells.

Wells 10,11,12      These wells comprise a second set of relatively close-in wells to further define the variability of geological, hydrological, and geochemical parameters. These wells provide additional data points for that portion of the aquifer system lying about midway along the conceptualized flow path between the suspected recharge zone (Lake 233) and the discharge zone southwest of Dewdrop Lake.

Wells 13,14,15

These wells are located along each of three possible flow paths extending toward the apparent discharge zones. They provide data points for collection of geologic and hydrologic data close to the probable discharge zone. Any changes in the local hydrostratigraphy will be critical in these approximate locations. The greater distance from the disposal area permits their being placed at a greater separation distance from each other. A full complement of hydrologic tests is to be carried out in these wells.

#### 4.2.2 Geologic Data Acquisition

##### 4.2.2.1 Bedrock Surface

Determination of the bedrock surface will be accomplished through the use of boreholes and mapped outcrops. All boreholes "emplaced" (no new or additional field work) during this study will be continued down to the bedrock/sediment contact, and the elevation of that contact recorded. These data will provide a distribution of bedrock elevations that are necessary to plot the surface of that unit and determine the bedrock controls on the shallow groundwater flow system. The use of aerial photography to aid in the mapping of geologic features is precluded due to the classified nature of the area surrounding the site under investigation.

##### 4.2.2.2 Structure and Tectonics

Regional geologic maps and reports will be utilized to ascertain the tectonic mechanisms which have operated in the past or are operating at the present time. These maps and reports will be used to determine the presence of structures such as folds and faults which may control the movement of groundwater in the units. No new or additional geologic field work is planned; only existing published or otherwise available data will be utilized.

##### 4.2.2.3 Hydrostratigraphy

Determination of the detailed hydrostratigraphic sequence of the subject site will be developed from a limited number of data points. This information on near-surface sediments will be derived from the selected boreholes on the site at or near the locations shown on the topographic map, Figure 5. The groundwater flow system in the vicinity of the Nitrate Disposal Pit Site is of limited areal extent as evidenced by the swampy (discharge) area immediately west of the pit. The specific well locations were selected to correspond to the interpreted hydrogeologic flow system. Wells 1-4 are placed to ascertain near-field parameters and the potential gradient from the disposal site toward Lake 233. The remaining wells are placed in concert with the more regional system as indicated by the direction of stream flow and the expression of lake elevations in the direction of Dewdrop Lake (Figures 3 and 4). The boring locations have been selected using the limited available data and professional judgment to give a reasonable picture of the consistency of the sediments and to define, within practical

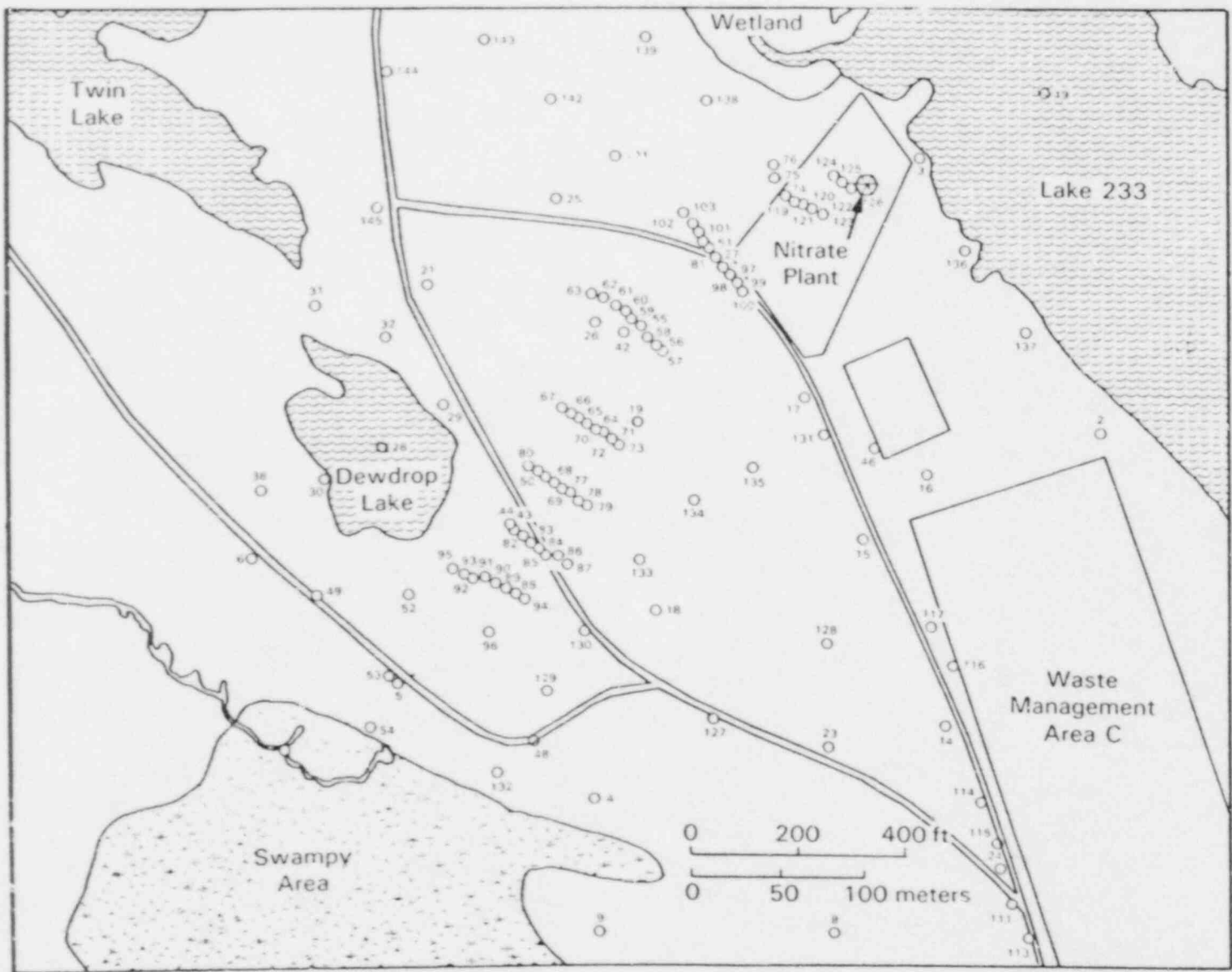


FIGURE A.5. Locations of Monitoring Wells at the Nitrate Disposal Pit Site

limits, the configuration of the bedrock surface. The borings will be completed as observation wells to provide hydrologic data for that phase of site characterization. Attention will be directed at the proposed disposal pit site to determine the specific items enumerated below.

#### 4.2.3 Hydrologic Data Acquisition

The hydrology of the Nitrate Disposal Pit Site will be analyzed based upon data covering meteorologic, climatologic, surface water, and groundwater disciplines.

##### 4.2.3.1 Climate

Essential to the determination of a water budget for the selected site is the determination of the availability of precipitation for run-off, infiltration, and evaporation. The distribution of precipitation throughout the year determines the amount of water potentially available for the three paths noted above. Records for the Chalk River Site will be analyzed to determine the average amount of water that is available to recharge the groundwater system or leach radionuclides from wastes deposited at the proposed site. Precipitation, temperature, latitude, and vegetation type and cover will be used to approximate the annual evapotranspiration from the proposed site.

##### 4.2.3.2 Surface Water

The region surrounding the proposed site has numerous small streams and small-to-large lakes. Available records will be analyzed to estimate the amount of run-off that annually occurs at the site. If available, the amount of water held in the lakes will be determined from lake surface elevation records. These data are critical to the modeling effort as preliminary information indicates that the lakes serve as recharge and discharge boundaries. Flood potentials, where they affect groundwater recharge, will be assessed. Surface-water impact, other than that necessary for development of the groundwater model, will not be assessed.

##### 4.2.3.3 Groundwater

The data requirements listed in Section 4.1.2 will be obtained from the wells or boreholes shown in Figure 5. As they are analyzed, a comparison between the data will be made to ascertain the continuity of units identified as hydrogeologically equivalent. Additional wells may be required if the identified units prove to be of limited extent. Additional wells will be located based upon a combination of professional judgment and the use of geostatistical tools such as kriging. Identification of boundary conditions will be made through analysis of the above data.

#### 4.2.4 Geochemical Data Acquisition

This section describes the methods for acquiring the geochemical information needed for calculating the retardation of radionuclides by the soil phases.

#### 2.4.1 Mineralogical and Chemical Characterization of Rock/Sediment

Representative samples of the rock/sediment are chosen from borehole cores, surface soils or outcrops after visual inspection of texture and color. Small subsamples of the rock/sediment are prepared for X-ray diffraction and optical microscopy by standard methods described in geologic and soil science texts (6,7,8).

The total chemical analysis of the solid may be determined directly on a crushed and homogenized sample by X-ray fluorescence or neutron activation or the sample can be totally dissolved and analyzed by inductively coupled plasma emission spectroscopy, atomic absorption, or optical emission spectrometry. Details for these techniques may be found in appropriate texts (9,10).

Other specific analyses important to determining contaminant retardation potential include the cation exchange capacity, hydrous iron oxide content, organic matter content, particle size distribution, and surface area of the sediments.

Specific procedures for determining the cation-exchange capacity and organic matter content can be found in References 8 and 9. Specific procedures for determining hydrous iron oxide contents can be found in References 8, 9, 11, and 12.

Specific procedures for determining the particle size distribution and surface area of disaggregated sediments can be found in References 7, 8, 13, 14, and 15.

It is prudent to perform the rock/sediment mineralogical and chemical characterization to gather background information to strengthen the understanding of the adsorption processes that are controlling contaminant retardation. Measurement of distribution coefficients without acquiring this ancillary information limits one's ability to infer the controlling adsorption processes, speculate on the effect of future environmental changes, and to judge whether the samples used and conditions studied in fact are representative, comprehensive enough, and applicable to projecting site performance.

Absorption of contaminants is sensitive to the mineralogy of the rock and sediments. Minerals exhibit valuable selectivity and capacity for adsorption; therefore, it is important to delineate the types and relative quantities of minerals present. Much literature exists that describes the selectivity and capacity of various minerals to adsorption of specific contaminants (16, 17, 18, 19).

Cation exchange, surface area, and particle size of the solid adsorbent are three important parameters that correlate well with adsorption of contaminants, especially alkali and alkaline earth metals such as Cs, Sr, and Ra. The hydrous oxide content correlates well with adsorption of transition metals such as Co, Ni, Zn, and Pb and actinides. The organic matter content is important in the adsorption of transition metals, lanthanides, and actinides.

#### 4.2.4.2 Distribution Coefficients

After the range of mineralogy and chemical composition of the rocks/sediments are determined in 4.2.4.1 for materials at the site along probable pathways, one can evaluate how many rock/sediment types should be used to generate distribution coefficients. The second variable, water type (waste leachate and groundwater), must be bounded. The range of groundwaters is determined as described in 4.2.4.3. The waste leachate must be estimated in the conceptual waste source-term model previously discussed. The variability in groundwater and potential waste leachate at the site allow chemists to choose representative solutions to use in the distribution coefficient determinations. The waste type also identifies which contaminants (e.g., radionuclides) should be studied.

The most economical method of generating a distribution coefficient is the batch or static adsorption test. Briefly, the method consists of placing a known mass of rock/sediment into an inert container (typically polyethylene or teflon) and then contacting the sediment with a known volume of groundwater/waste leachate traced with the contaminants of interest. The system is shaken gently for a period of time (typically days to a few weeks) or until steady-state solution concentrations are obtained for the tracers. The sediments and solution are then separated and either the effluent measured and compared to the original influent or the effluent and solids are counted.

The distribution coefficient, or  $R_d$  value, (often called  $K_d$ ) is calculated either as

$$R_d = \frac{(C_{INF} - C_{EFF}) (V)}{(C_{EFF}) (M)}$$

Where:

- $C_{INF}$  = concentration of tracer in influent (original) solution
- $C_{EFF}$  = concentration of tracer in effluent (final) solution
- $V$  = volume of solution used (usually mls)
- $m$  = mass of sediment/rock used (usually grams)

or as

$$R_d = \frac{C_{solid}}{C_{solution}}$$

Where:

- $C_{solid}$  = concentration of tracer on solid (per gram)
- $C_{solution}$  = concentration of tracer in final solution (per ml)

Suggested methods that describe the details of performing batch experiments can be found in References 20, 21, and 22.

A discussion of strengths and weaknesses of the batch method of determining distribution coefficients is found in Serne and Relyea, 1983 (23.).

Because of possible limitations in the batch method, it is prudent to pot check retardation results obtained by the batch method with flow-through column experiments. Briefly, the method consists of packing unconsolidated rocks/sediment into a glass or plexiglass column with an inlet and outlet port. The groundwater/waste leachate traced with the contaminants of interest is percolated through the column and the breakthrough of tracer is monitored in the effluent. Column experiments are useful to determine whether there are mobile species of a contaminant present among other species more prone to absorbing. The batch method assumes that only one species for each contaminant is present whereas the column method can identify the presence of different species with different mobilities and allows separate retardation factors to be calculated. The batch method produces only one distribution coefficient (an average across all species) for each contaminant.

#### 4.2.4.3 Chemical Analysis of Groundwater/Waste Leachate

The chemical composition of the groundwater and waste leachate also affects the retardation potential of contaminants. Knowledge of the concentration of macro cations (Ca, Mg, Na, and K) allows one to determine the degree of competition for exchange sites. The concentration of inorganic ligands such as  $\text{Cl}^-$ ,  $\text{CO}_3^{2-}$ , and  $\text{SO}_4^{2-}$  and organic ligands allows one to estimate the degree to which metallic contaminants might form mobile soluble complexes that resist retardation. The solution pH and Eh (oxidation-reduction potential) also are important parameters that affect adsorption especially for transition, lanthanide, and actinide metals via hydrolysis and redox reactions (i.e., species and valence state changes).

There are numerous standard water sample collection, sample preservation and sample analyses techniques available (24,25,26,27).

Briefly, the most commonly used procedures are to pump groundwater from wells until effects of the well casing are minimal and then to measure sensitive parameters like pH, Eh, and alkalinity in the field. The water sample is then filtered through 0.2 to 0.45  $\mu\text{m}$  membrane filters and split into subsamples and preserved. Macro cation and trace metal samples are acidified to pH 2 while anion and organic samples are kept cool and in the dark until analyzed. Currently major cations and some trace metals are analyzed by inductively coupled plasma emission spectroscopy; other trace metals are determined by graphite furnace atomic absorption; anions excluding bicarbonate and carbonate are measured by ion chromatography and the carbonates by acid titration. Organic content is determined on a TOC analyzer.

It is customary to add up the milliequivalents of cations and anions for the water analysis and compare them. Differences greater than 5 to 10% suggest that some important constituent was not analyzed or that the analytical quality is deficient. A repeat analysis is recommended until balance is achieved.



## 5.0 PRELIMINARY FLOW AND TRANSPORT ANALYSIS BASED ON SITE CHARACTERIZATION

This portion of the project will be carried out in subsequent tasks, but is briefly mentioned here to indicate how the acquired data are used in transport modeling.

It must be recognized that site characterization is a part of the development of the site conceptual model. The initial conceptualization of the site is subject to continual revision as actual field data are gathered. As the data are gathered, they are entered into the preliminary flow model and the model exercised to ascertain consistency with the conceptual model. Where inconsistencies are found, alterations to either or both models may be made. This process is iterative in nature, the end result being the development of defensible conceptual and numerical models. Actual selection of the numeric codes used to model the site is dependent upon the final site conceptual model.

### 5.1 REFINE CONCEPTUAL MODEL

The conceptual model is the driving force that controls the validity of any numerical modeling. All of the data derived from site characterization is utilized to refine and change, if necessary, the original conceptual model. Because of this, actual field studies may be affected as each new data set is added to the conceptualization of the hydrologic system. Refinement of the conceptual model is a controlling factor in the site characterization tasks.

### 5.2 ADDITIONAL DATA NEEDS

In the course of building and testing both the conceptual and numerical models for the site, it is probable that additional data will be required. These additional data needs are identified through the refinement of the conceptual model and are forwarded to the field site characterization task to be provided. Thus, the initial characterization activities are dynamic in that they are subject to continual change.

## 6.0 PERFORMANCE ASSESSMENT

The final effort in the project is the performance assessment modeling of the subject site, and will be conducted in a succeeding task. In this task, all of the data for the particular level of modeling is in place and the simulation carried out to assess the temporal movement of contaminants from the site. The majority of the effort is included in the preceding tasks; this task merely completes the loop.

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APPENDIX B

CONCENTRATIONS OF LONG-LIVED RADIONUCLIDES IN  
GROUNDWATER AT THE NITRATE DISPOSAL PIT SITE,  
CHALK RIVER NUCLEAR LABORATORIES, OCTOBER 1984

1984 CHALK RIVER

NITRATE PLUME

Well No. C-125 - Concentrations in pCi/liter

	Filters		1st Anion		2nd Anion		1st Cation		2nd Cation		1st Al <sub>2</sub> O <sub>3</sub>		2nd Al <sub>2</sub> O <sub>3</sub>	
Ce-144	<0.1		<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.1	<0.1	<0.2	<0.2
Co-60	0.075 ± 0.016		0.1 ± 0.04	<0.03	<0.03	0.59 ± 0.09	<0.03	<0.03	<0.03	<0.03	<0.01	<0.01	<0.02	<0.02
Cs-134	<0.01		<0.03	<0.03	<0.03	<0.1	<0.03	<0.03	<0.03	<0.03	<0.01	<0.01	<0.02	<0.02
Cs-137	<0.02		<0.03	<0.03	<0.03	<0.1	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.02	<0.02
Eu-152	<0.02		<0.1	<0.09	<0.09	<0.6	<0.09	<0.09	<0.09	<0.09	<0.04	<0.04	<0.05	<0.05
Eu-154	<0.06		<0.2	<0.2	<0.2	<0.5	<0.2	<0.2	<0.2	<0.2	<0.08	<0.08	<0.1	<0.1
Eu-155	<0.02		<0.1	<0.1	<0.1	<2.2	<0.1	<0.1	<0.1	<0.1	<0.05	<0.05	<0.1	<0.1
Mn-54	<0.02		<0.07	<0.04	<0.04	<0.1	<0.04	<0.04	<0.04	<0.04	<0.02	<0.02	<0.02	<0.02
Ru-103	<0.2		<0.8	<0.8	<0.8	<4.4	<0.8	<0.8	<0.8	<0.8	<0.3	<0.3	<0.5	<0.5
Ru-106	<0.1		<0.3	<0.3	<0.3	<1.8	<0.3	<0.3	<0.3	<0.3	<0.1	<0.1	<0.2	<0.2
Sb-125	<0.02		<0.1	<0.09	<0.09	<0.6	<0.09	<0.09	<0.09	<0.09	<0.04	<0.04	<0.05	<0.05
Zr-95	<0.1		<0.3	<0.3	<0.3	<1.4	<0.3	<0.3	<0.3	<0.3	<0.1	<0.1	<0.2	<0.2
Nb-95	<0.4		<1.2	<1.2	<1.2	<4.5	<1.2	<1.2	<1.2	<1.2	<0.5	<0.5	<0.8	<0.8

1994 CHALK RIVER

NITRATE PLUME

Well No. C-54-3 - Concentrations in pCi/liter

	Filters			1st Anion			2nd Anion			1st Cation			2nd Cation			1st Al <sub>2</sub> O <sub>3</sub>		2nd Al <sub>2</sub> O <sub>3</sub>	
Ce-144	<0.1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1	<0.1	<0.2	<0.2	<0.2	
Co-60	0.21 ± 0.01	0.24 ± 0.03	<0.02	<0.02	<0.02	<0.02	2.49 ± 0.05	<0.02	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.02	<0.02	<0.02	
Cs-134	<0.008	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.01	<0.01	<0.01	<0.01	<0.01	
Cs-137	<0.03	<0.02	<0.02	<0.02	<0.02	<0.02	0.068 ± 0.024	<0.02	<0.03	<0.03	<0.03	<0.03	<0.03	<0.01	<0.01	<0.01	<0.01	<0.01	
Eu-152	<0.01	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.03	<0.03	<0.03	<0.03	<0.03	
Eu-154	<0.05	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.11	<0.11	<0.08	<0.08	<0.08	
Eu-155	<0.01	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.08	<0.04	<0.04	<0.05	<0.05	<0.05	
Mn-54	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.04	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.03	<0.03	<0.03	<0.03	<0.03	
Ru-103	<0.1	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	<0.2	<0.2	<0.3	<0.3	<0.3	
Ru-106	<0.1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1	
Sb-125	<0.01	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.03	<0.03	<0.04	<0.04	<0.04	
Zr-95	<0.07	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1	
Nb-95	<0.2	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.6	<0.3	<0.3	<0.3	<0.3	<0.3	



## 1984 CHALK RIVER

## NITRATE PLUME

Well No. C-89-11 - Concentrations in pCi/liter

	Filters		1st Anion		2nd Anion		1st Cation		2nd Cation		1st Al <sub>2</sub> O <sub>3</sub>		2nd Al <sub>2</sub> O <sub>3</sub>	
Ce-144	<0.2		<0.2	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.1	<0.1	<0.1	<0.1
Co-60	<0.01		<0.03	<0.03	<0.03	<0.05	<0.05	<0.04	<0.04	<0.04	<0.01	<0.01	<0.01	<0.01
Cs-134	<0.009		<0.03	<0.03	<0.03	<0.07	<0.07	<0.04	<0.04	<0.04	<0.01	<0.01	<0.02	<0.02
Cs-137	<0.03		<0.03	<0.05	<0.05	<0.07	<0.07	<0.04	<0.04	<0.04	<0.01	<0.01	<0.02	<0.02
Eu-152	<0.02		<0.07	<0.1	<0.1	<0.3	<0.3	<0.1	<0.1	<0.1	<0.03	<0.03	<0.04	<0.04
Eu-154	<0.05		<0.1	<0.2	<0.2	<0.3	<0.3	<0.2	<0.2	<0.2	<0.06	<0.06	<0.08	<0.08
Eu-155	<0.01		<0.09	<0.1	<0.1	<0.8	<0.8	<0.1	<0.1	<0.1	<0.04	<0.04	<0.05	<0.05
Mn-54	<0.01		<0.03	<0.04	<0.04	<0.08	<0.08	<0.1	<0.1	<0.1	<0.01	<0.01	<0.02	<0.02
Ru-103	<0.1		<0.6	<0.8	<0.8	<2.1	<2.1	<1.0	<1.0	<1.0	<0.3	<0.3	<0.4	<0.4
Ru-106	<0.09		<0.3	<0.3	<0.3	<0.9	<0.9	<0.4	<0.4	<0.4	<0.1	<0.1	<0.2	<0.2
Sb-125	<0.02		<0.07	<0.1	<0.1	<0.3	<0.3	<0.1	<0.1	<0.1	<0.04	<0.04	<0.04	<0.04
Zr-95	<0.08		<0.2	<0.3	<0.3	<0.7	<0.7	<0.4	<0.4	<0.4	<0.1	<0.1	<0.1	<0.1
Nb-95	<0.2		<0.9	<1.2	<1.2	<2.3	<2.3	<1.5	<1.5	<1.5	<0.4	<0.4	<0.6	<0.6

1994 CHALK RIVER

NITRATE PLUME

Well No. C-69-II - Concentrations in pCi/liter

	Filters		1st Anion		2nd Anion		1st Cation		2nd Cation		1st Al <sub>2</sub> O <sub>3</sub>		2nd Al <sub>2</sub> O <sub>3</sub>	
Ce-144	<0.1		<0.2	<0.4	<0.3	<0.3	<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Co-60	<0.01		<0.03	<0.06	<0.1	<0.1	<0.03	<0.03	<0.01	<0.01	<0.01	<0.01	<0.02	<0.02
Cs-134	<0.01		<0.03	<0.04	<0.2	<0.2	<0.04	<0.04	<0.01	<0.01	<0.01	<0.01	<0.02	<0.02
Cs-137	<0.02		<0.03	<0.04	<0.2	<0.2	<0.04	<0.05	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Eu-152	<0.02		<0.08	<0.01	<2.2	<2.2	<0.09	<0.09	<0.03	<0.03	<0.03	<0.03	<0.07	<0.07
Eu-154	<0.06		<0.3	<0.2	<0.8	<0.8	<0.2	<0.2	<0.07	<0.07	<0.07	<0.07	<0.09	<0.09
Eu-155	<0.01		<0.1	<0.1	<2.8	<2.8	<0.1	<0.1	<0.04	<0.04	<0.04	<0.04	<0.05	<0.05
Mn-54	<0.01		<0.03	<0.04	<0.2	<0.2	<0.04	<0.04	<0.01	<0.01	<0.01	<0.01	<0.02	<0.02
Ru-103	<0.1		<0.4	<0.7	<4.7	<4.7	<0.6	<0.6	<0.2	<0.2	<0.2	<0.2	<0.3	<0.3
Ru-106	<0.1		<0.3	<0.4	<2.7	<2.7	<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.2	<0.2
Sb-125	<0.02		<0.08	<0.2	<0.9	<0.9	<0.09	<0.09	<0.03	<0.03	<0.03	<0.03	<0.04	<0.04
Zr-95	<0.08		<0.2	<0.3	<1.7	<1.7	<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Nb-95	<0.2		<0.6	<1.0	<4.4	<4.4	<0.8	<0.8	<0.2	<0.2	<0.2	<0.2	<0.4	<0.4

## 1984 CHALK RIVER

## NITRATE PLUME

Well No. C-27-3 - Concentrations in pCi/liter

	Filters		1st Anion		2nd Anion		1st Cation		2nd Cation		1st Al <sub>2</sub> O <sub>3</sub>		2nd Al <sub>2</sub> O <sub>3</sub>	
Ce-144	<0.1		<0.2	<0.3	<0.3	<2.3	<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Co-60	<0.01		<0.03	<0.06	<0.05		<0.03	<0.03	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cs-134	<0.009		<0.03	<0.04	<0.1		<0.03	<0.03	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cs-137	0.87 ± 0.02		<0.03	<0.04	5.01 ± 0.17		<0.04	<0.04	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Eu-152	<0.02		<0.08	<0.1	<0.4		<0.1	<0.1	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Eu-154	<0.05		<0.2	<0.2	<0.3		<0.2	<0.2	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06
Eu-155	<0.003		<0.1	<0.1	<1.1		<0.1	<0.1	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Mn-54	<0.01		<0.04	<0.04	<0.03		<0.04	<0.07	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
Ru-103	<0.07		<0.3	<0.3	<1.1		<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Ru-106	<0.03		<0.3	<0.3	<1.5		<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Sb-125	<0.02		<0.08	<0.2	<0.3		<0.2	<0.1	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Zr-95	<0.05		<0.2	<0.2	<0.5		<0.2	<0.2	<0.07	<0.07	<0.07	<0.07	<0.07	<0.07
Nb-95	<0.09		<0.3	<0.4	<1.0		<0.3	<0.4	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1

1984 CHALK RIVER

NITRATE PLUME

Well No. 119-2" dia. Piezo. - Concentrations in pCi/liter

	Filters			2nd Anion			1st Cation			2nd Cation			1st Al <sub>2</sub> O <sub>3</sub>			2nd Al <sub>2</sub> O <sub>3</sub>		
		1st Anion	2nd Anion	1st Anion	2nd Anion	1st Anion	1st Cation	2nd Cation	3rd Cation	1st Al <sub>2</sub> O <sub>3</sub>	2nd Al <sub>2</sub> O <sub>3</sub>	3rd Al <sub>2</sub> O <sub>3</sub>	1st Al <sub>2</sub> O <sub>3</sub>	2nd Al <sub>2</sub> O <sub>3</sub>	3rd Al <sub>2</sub> O <sub>3</sub>			
Ce-144	<0.2	<0.2	<0.4	<0.4	<0.4	<0.5	<0.4	<0.4	<0.4	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1			
Co-60	<0.03	<0.03	<0.04	<0.04	<0.1	<0.1	<0.05	<0.05	<0.05	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01			
Cs-134	<0.01	<0.03	<0.04	<0.04	<0.3	<0.3	<0.05	<0.05	<0.05	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02			
Cs-137	<0.02	<0.03	<0.04	<0.04	<0.2	<0.2	<0.05	<0.05	<0.05	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02			
Eu-152	<0.03	<0.08	<0.1	<0.1	<1.3	<1.3	<0.2	<0.2	<0.2	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04			
Eu-154	<0.07	<0.1	<0.2	<0.2	2.03 ± 0.98	2.03 ± 0.98	<0.3	<0.3	<0.3	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03			
Eu-155	<0.02	<0.1	<0.2	<0.2	<3.4	<3.4	<0.2	<0.2	<0.2	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05			
Mn-54	<0.1	<0.04	<0.04	<0.04	<0.2	<0.2	<0.6	<0.6	<0.6	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03			
Ru-103	<0.1	<0.3	<0.4	<0.4	<4.0	<4.0	<0.6	<0.6	<0.6	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2			
Ru-106	<0.1	<0.3	<0.4	<0.4	<3.2	<3.2	<0.5	<0.5	<0.5	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1			
Sb-125	<0.03	<0.08	<0.1	<0.1	<1.1	<1.1	<0.1	<0.1	<0.1	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04			
Zr-95	<0.09	<0.2	<0.3	<0.3	<1.8	<1.8	<0.3	<0.3	<0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1			
Nb-95	<0.2	<0.4	<0.6	<0.6	<3.7	<3.7	<0.8	<0.8	<0.8	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2			

## 1984 CHALK RIVER

## NITRATE PLUME

Well No. C-3-2 - Concentrations in pCi/liter

	Filters		1st Anion		2nd Anion		1st Cation		2nd Cation		1st Al <sub>2</sub> O <sub>3</sub>		2nd Al <sub>2</sub> O <sub>3</sub>	
Ce-144	<0.06		<0.3		<0.3		<0.2		<0.3		<0.1		<0.1	
Co-60	<0.01		<0.04		<0.04		<0.03		<0.04		<0.01		<0.01	
Cs-134	<0.009		<0.05		<0.04		<0.03		<0.04		<0.01		<0.02	
Cs-137	<0.06		<0.04		<0.04		<0.2		<0.04		<0.01		<0.02	
Eu-152	<0.01		<0.1		<0.1		<0.08		<0.1		<0.03		<0.05	
Eu-154	<0.05		<0.2		<0.2		<0.1		<0.2		<0.07		<0.09	
Eu-155	<0.01		<0.1		<0.3		<0.2		<0.1		<0.04		<0.06	
Mn-54	<0.1		<0.05		<0.04		<0.06		<0.04		<0.02		<0.02	
Ru-103	<0.07		<0.4		<0.4		<0.3		<0.5		<0.1		<0.2	
Ru-106	<0.07		<0.4		<0.4		<0.3		<0.4		<0.1		<0.1	
Sb-125	<0.01		<0.1		<0.1		<0.08		<0.1		<0.03		<0.05	
Zr-95	<0.05		<0.2		<0.4		<0.2		<0.3		<0.1		<0.1	
Nb-95	<0.1		<0.6		<0.6		<0.4		<0.7		<0.2		<0.3	

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13 ABSTRACT (200 words or less)

Before a license can be obtained to construct a facility for the shallow-land burial of low-level waste, the U. S. Nuclear Regulatory Commission must be assured that the facilities will meet both performance objectives and prescriptive requirements set forth in 10CFR61, "Licensing Requirements for Land Disposal of Radioactive Waste." Subpart D, Section 61.50(a)(2) of 10CFR61 states that a "disposal site shall be capable of being characterized, modeled, analyzed and monitored." In order to test the concept of "site modelability," a 30-year old low-level radioactive waste disposal site at Chalk River Nuclear Laboratories (CRNL), Canada, was used as a field location for evaluating the process of site characterization and the subsequent modeling predictions of radionuclide transport from the site by groundwater. This evaluation was performed by comparing the actual measured radionuclide migration with predicted migration estimated from hydrologic/radionuclide transport models. This comparison has provided valuable insights into the applicability of transport modeling, and in determining what level of effort is needed in site characterization at locations similar to the Nitrate Disposal Pit to provide the desired degree of predictive capabilities.

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