CONF-8205107--2

RADIOLOGICAL SURVEY OF THE LOW-LEVEL RADIOACTIVE WASTE BURIAL SITE AT THE PALOS FOREST PRESERVE, ILLINOIS

by

CONF-8205107--2

DE83 009569

Kimberly A. Hayes
Environmental Monitoring Group
Occupational Health and Safety Division
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439

ABSTRACT

Two landfill sites containing low-level radioactive waste material, Site A and Plot M, are located 14 miles southwest of Chicago, Illinois in the Palos Forest Preserve. Site A is the former location of the Argonne National Laboratory. Buried at Site A in 1956 were the dismantled reactor shells, building walls, and cooling towers from three of the world's first nuclear reactors. Plot M was used from 1943 to 1949 for burial of low-level radioactive wastes derived from Site A operations and from the University of Chicago Metallurgical Laboratory.

Tritiated water was detected in 1973 in some of the Forest Preserve picnic wells located 500-1000 yards north of Plot M. An extensive surveillance program was initiated in 1976 to 1) study the elevated tritium content of some picnic wells and its observed seasonal fluctuations, 2) establish if other radionuclides buried in Plot M or remaining at Site A have migrated, 3) establish the rate of groundwater movement in the glacial till and underlying dolomite aquifer, 4) determine the tritium content of the till and aquifer, and 5) predict future tritium levels in the well water.

Methods used in the survey include radiochemical analyses of well water, surface water, surface soil, soil boring, and vegetation samples from the Forest Preserve. Several test wells were installed in the soil and dolomite bedrock to monitor radioactivity in groundwater, measure water levels, and provide other geohydrological information.

Tritium has migrated from the Plot M burial trenches into the surrounding drift. The tritium plume, the contaminated zone in the drift in which tritium concentrations exceed 10 nanocuries per liter of water (nCi/L), has migrated at least 165 feet horizontally northward and 130 feet vertically downward to the bedrock surface. Small amounts of other radionuclides - uranium, plutonium, and strontium-90 - have been found in boreholes beneath the concrete cap covering Plot M, but not in the subsoil outside of the Plot. The radionuclide concentrations found to date are too low to result in any measureable radiation exposure to the public.

INTRODUCTION

This report describes the ongoing radiological surveillance of the former low-level waste burial site in the Palos Forest Preserve near Chicago, Illinois (Figure 1).

BACKGROUND AND SITE DESCRIPTION

The burial site consisted of two parts (Figure 2): Site A, the former location of Argonne National Laboratory (ANL), the site for the world's first nuclear reactors, Chicago Pile One (CP-1), redesignated CP-2, CP-3, and CP-3', related laboratories, and experimental facilities; and Plot M, a one-acre radioactive waste burial ground used for the disposal of equipment, animal carcasses, clothing, building materials, and other liquid and solid wastes derived from Site A operations and from the University of Chicago Metallurgical Laboratory.

Plot M is rectangular in shape, about 150 feet by 140 feet. It is a flat, open, grassy area on an eastward sloping hill. The site is drained by two intermittent streams which join at a point about 210 feet north and downhill of the Plot. The site is approximately 700 feet above sea level. The Plot M site is underlaid by unconsolidated glacial drift that has an average thickness of 130 feet (Figure 3). Most of the drift can be described as till that is hard, coarse textured, clay and silt rich, and contains discontinuous sand lenses (Olimpio, 1980). The Niagaran dolomite aquifer lies immediately beneath the glacial drift (Willman and others, 1963).

Radioactive material was placed in Plot M from 1944 to 1949, originally in six-foot deep trenches with sufficient soil cover to reduce the radiation to acceptable levels, but by 1948 the waste was packaged in steel bins prior to burial. The steel bins were removed in 1949 and use of the Plot for waste burial was discontinued. In the spring of 1956 an inverted concrete box was constructed to cover the entire plot. The concrete sidewalls were extended eight feet into the ground and a one-foot thick concrete slab was placed over the entire burial area. The concrete was covered with two feet of soil and seeded with grass. Site A was also decommissioned in 1956. The research reactors were disassembled; highly radioactive fuel and other reactor components were removed from the site. The containment shell of CP-3 was filled with concrete and tumbled into a 40-foot deep excavation along with rubble and debris from the dismantling operation. After demolition was completed at Site A, the landfill was filled with dirt, and graded.

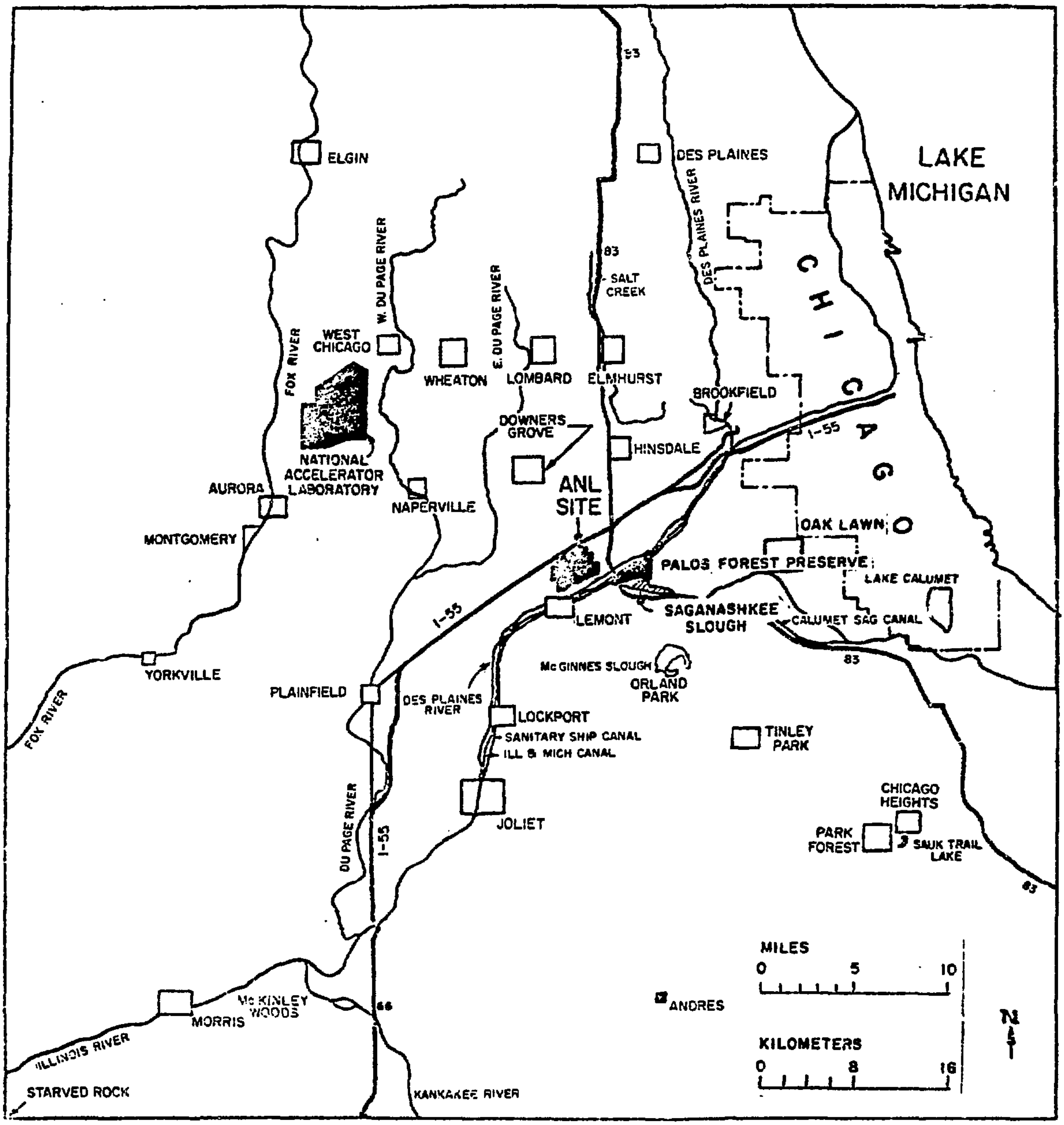

Figure 1. Location of Palos Forest Preserve on Chicago Area Map.

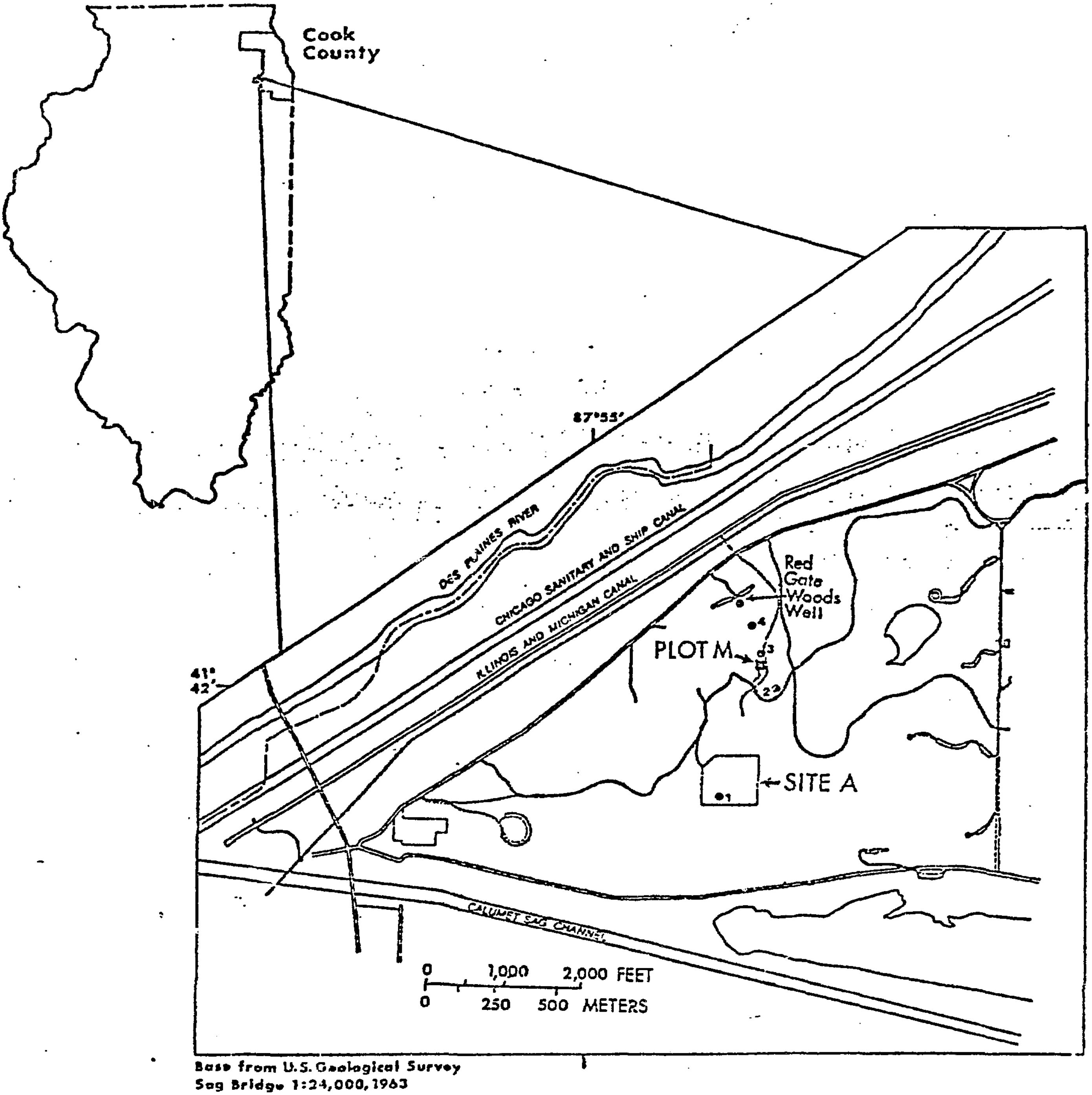

Figure 2. Location of Plot M and Site A, and Dolomite Wells in the Palos Forest Preserve.

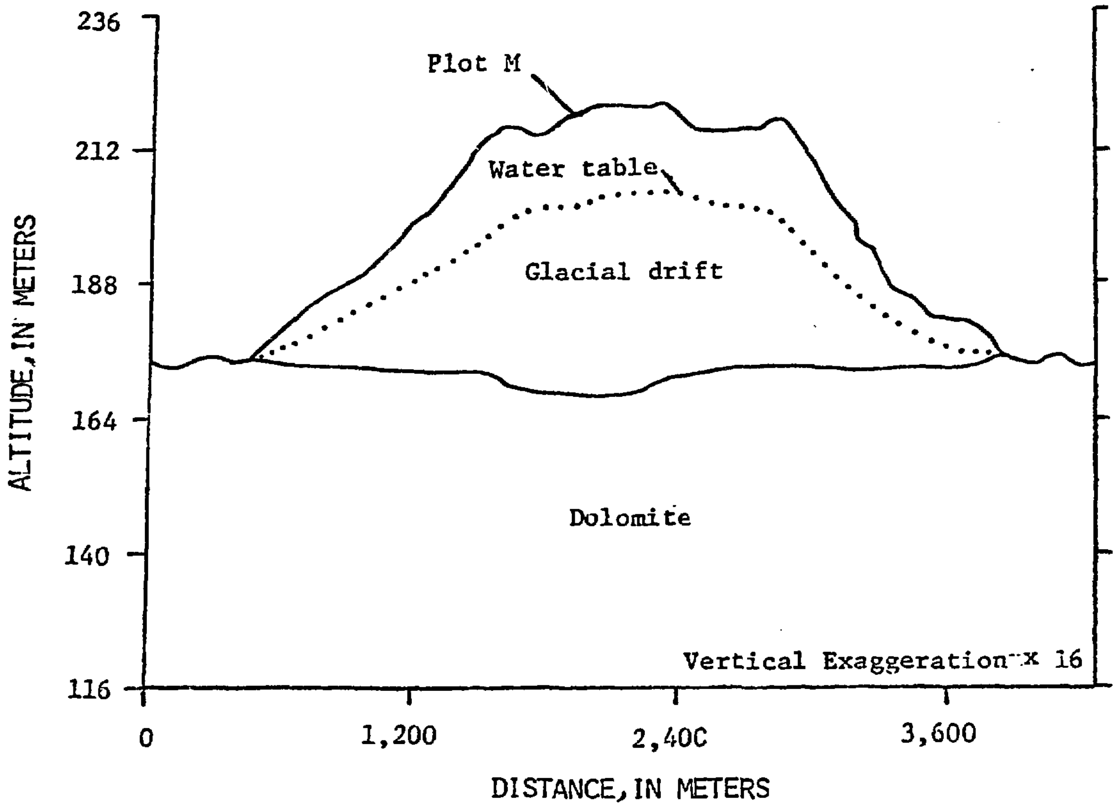

Figure 3. Geologic Cross Section of the Plot M Study Area.

RADIOLOGICAL SURVEY DESCRIPTION

ANL has conducted radiological surveys at both sites since 1952. Radiometric analyses of samples of soil borings, surface water and groundwater show no radionuclides have migrated from Site A. Similar analyses indicate that tritium has migrated from the Plot M site through unconsolidated glacial sediments to the underlying dolomite bedrock (Golchert and Sedlet, 1978).

On February 19, 1976, the Energy Research and Development Administration (ERDA) announced it would conduct an extensive radiological survey in the Palos Forest Preserve, as part of a nationwide ERDA program to resurvey former Manhattan District sites. When the survey was initiated, data had been accumulated from 1973 to 1975 to conclusively show that low levels of tritium were present in two forest preserve hand-pumped wells, approximately 1000 yards north of Plot M.

The Red Gate Woods (RGW) picnic well had the highest activity, with tritiated water concentrations ranging from < 0.2 nCi/l to 14 nCi/l. The concentrations varied seasonally, with a maximum in the winter and a minimum in the summer.

Tritium, a radioactive isotope of hydrogen of mass 3 and with a half-life of 12.4 years, is present in the water as part of the water molecule. "Tritium" and "tritiated water" are used interchangeably in this report. A tritiated water molecule is one in which a tritium atom has replaced one of the two hydrogen atoms normally present. Although the tritium levels detected in the RGW well water were not hazardous, ERDA began the study to determine the extent of movement of radioactive materials from the burial site and any possible future hazard to the health and safety of the public.

Argonne National Laboratory, an ERDA contractor, was asked to carry out the detailed radiological survey and to coordinate related studies. On October 1, 1977, the new U.S. Department of Energy assumed the responsibilities of ERDA, including the surveillance program. The Water Resources Division of the U.S. Geological Survey (initially the Madison, Wisconsin office and now the Champaign, Illinois office) is conducting the groundwater studies.

The survey attempts to answer several questions that arose after the discovery of tritium in the picnic wells in 1973. Assuming that Plot M is the tritium source, how does it migrate from Plot M to the dolomite wells? Does tritium move with groundwater along pathways entirely within the glacial drift, or along pathways through both the drift and underlying dolomite bedrock? What is the rate and extent of tritium migration in the drift and bedrock? Do surface and subsurface conditions at the site promote or retard radionuclide migration? Are any other radioactive materials buried at Plot M or Site A migrating away from their original locations to the surrounding environment? Frequent use of the land and water in the forest preserve around Plot M and Site A emphasizes the need for answers to these questions.

The principal investigative methods used in the survey are periodic radiochemical analyses of water from the existing wells in the area; radiochemical analyses of soil borings, surface soil, and vegetation in the Forest Preserve; installation of several additional wells in the soil and dolomite aquifer to monitor radioactivity in groundwater, measure water levels, and provide other geohydrological information; and determination of the migration potential of selected radionuclides disposed of in the waste burial ground.

TRITIUM MIGRATION IN THE DRIFT

Twenty-nine (29) boreholes have been drilled since 1976 to ascertain the areal and vertical extent of subsurface radionuclide migration in the glacial drift around Plot M. The location of the boreholes are shown in Figure 4. They range in depth from 40 feet to 165 feet from the land surface.

Originally, core samples were analyzed for a variety of radionuclides, and the moisture analyzed for tritium. By 1977, data had been accumulated to show that elevated concentrations of tritiated water existed throughout the Plot M area, and no gamma-ray emitting fission or activation products, uranium, or plutonium were detected in the core samples. In boreholes drilled after 1977, only tritium analyses were conducted to determine the extent of migration.

The tritium plume, defined to be the contaminated zone in the drift in which tritiated water concentration levels exceed 10 nanocuries per liter of water (nCi/l), has migrated horizontally northward at least 165 feet, and 130 feet vertically downward to the dolomite bedrock surface.

The underlying dolomite bedrock is highly permeable, contributing to a significant vertical total head gradient in the saturated zone of the drift. Thus, the primary direction of groundwater flow through the study area is downward. The diameter of the tritium plume decreases with depth and the elongate shape of the plume supports the data which indicate that tritiated water moves primarily downward in the drift - rather than laterally.

The north-south cross section of the study area, E-E' from Figure 4, is displayed in Figure 5. Section E-E' shows the maximum lateral and vertical extent of the tritium plume. This cross section is just east of Plot M.

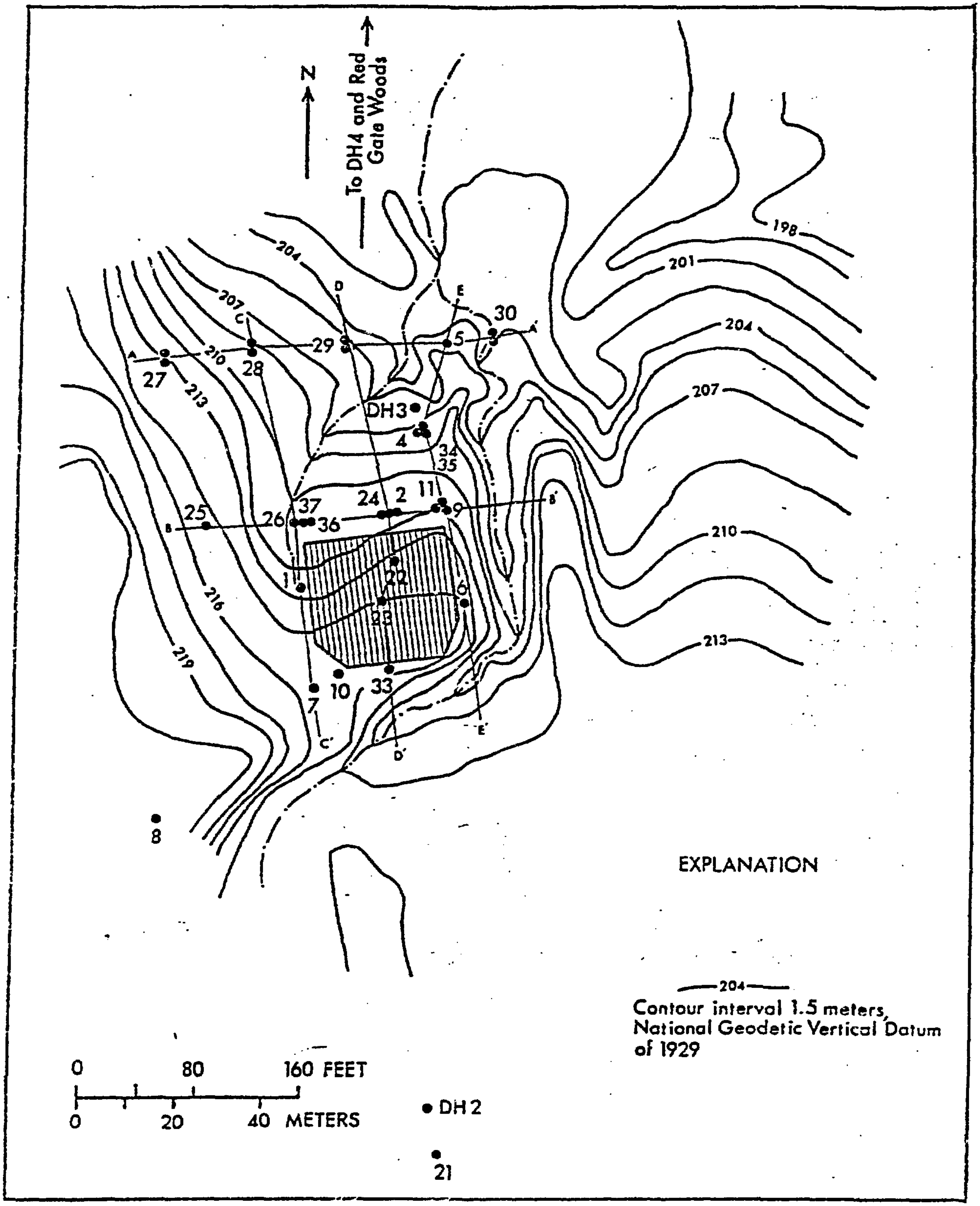

Figure 4. Map of Plot M Burial Site Showing Drainage, Borehole and Well Locations.

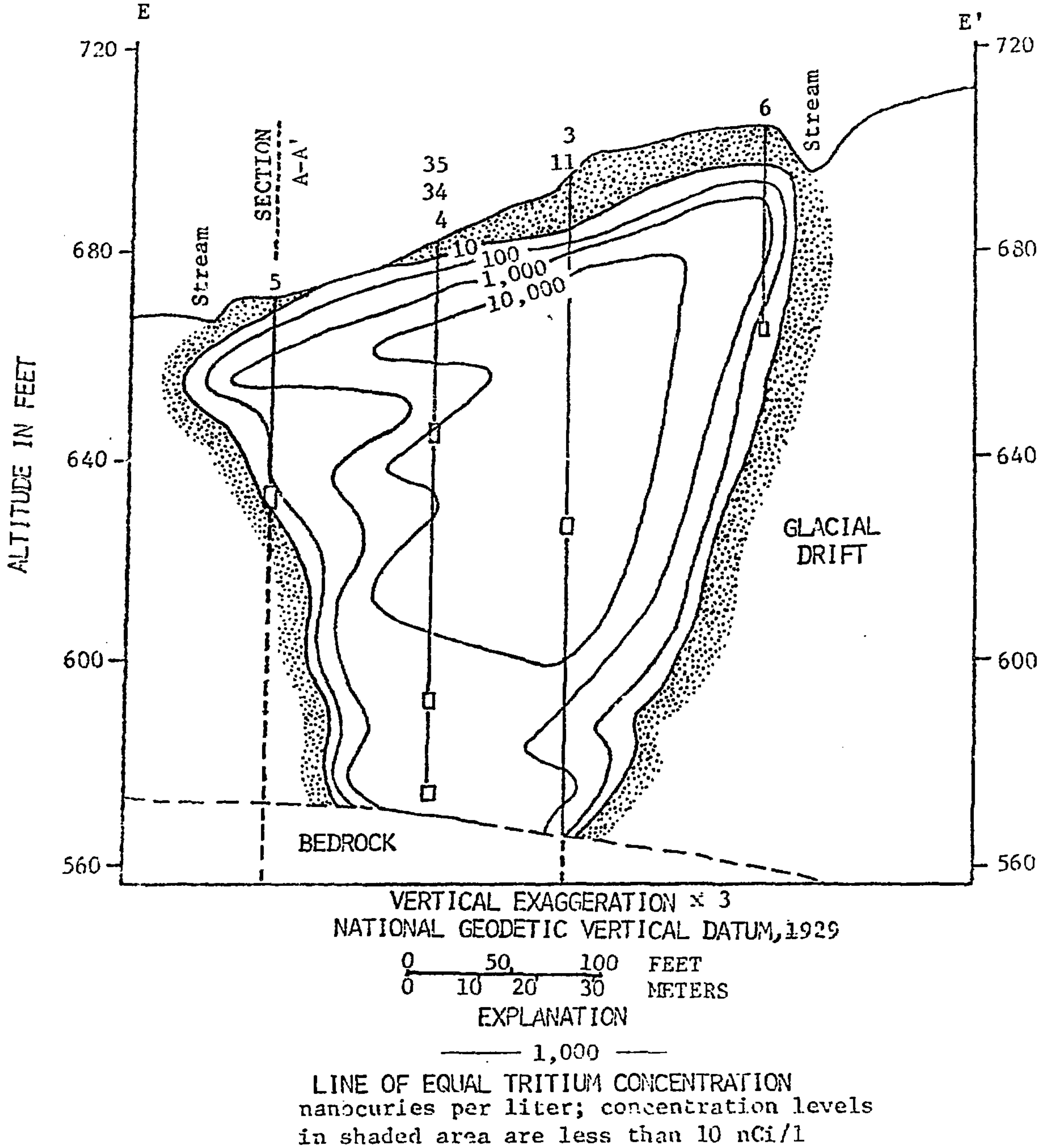

Figure 5. Cross Section E-E'; Maximum Extent of Tritium Plume.

The plume has a "bulls-eye" concentration pattern. Tritium levels are the most concentrated in the center, reaching levels as high as 50,000 nCi/l, and decrease peripherally outward to the 10 nCi/l boundary, which is the leading edge of the plume. The northward extensions in the isocentration lines correspond to the presence of sand layers and the resulting lateral flow of tritiated water. The plume extends laterally from the stream south of the site to the area north of borehole #5 (165 feet north of Plot M), and vertically from 10 feet below the land surface to bedrock.

The leading, northern edge of the tritium plume, cross section A-A' in Figure 4, is depicted in Figure 6. Tritium concentrations are significantly lower at this distance (165 feet) away from Plot M. The plume is considerably thinner (from top to bottom) and does not reach the underlying bedrock aquifer. Highest tritium concentration levels within the cross section, exceeding 1000 nCi/l, are located in a small, thin zone near the surface. This zone underlies numerous sand layers at depths less than 25 feet, near borehole #28 and borehole #29. Tritiated water moves horizontally within these highly hydraulically conductive geologic layers, which occur frequently in the drift around Plot M, to depths up to 65 feet beneath the land surface. Lateral tritium movement usually occurs coincident with the sand lenses within the drift. These lenses allow tritiated groundwater to travel horizontally, whereas the majority of the drift is not very hydraulically conductive due to the high clay and silt content, thus limiting the movement of tritiated water.

Tritiated water in the picnic well in Red Gate Woods has migrated there through the dolomite bedrock, after seepage into the aquifer directly beneath the buried wastes and underlying drift at Plot M. The data clearly show that tritium does not move horizontally through the drift from Plot M to Red Gate Woods.

The estimated total volume of the tritium plume is 8.79 (10^6) ft^3, which is equivalent to a rectangular volume of dimensions 260 ft x 260 ft x 130 ft. If the average total porosity of the drift is estimated to be 30 percent, then the total volume of pore space occupied by tritiated water is 2.64 (10^6) ft^3. Assuming the average concentration of tritium is 10^4 nCi/l the total amount of radioactive tritium in the glacial drift around Plot M is 800 Ci. This estimate is considered reliable to about a factor of 10.

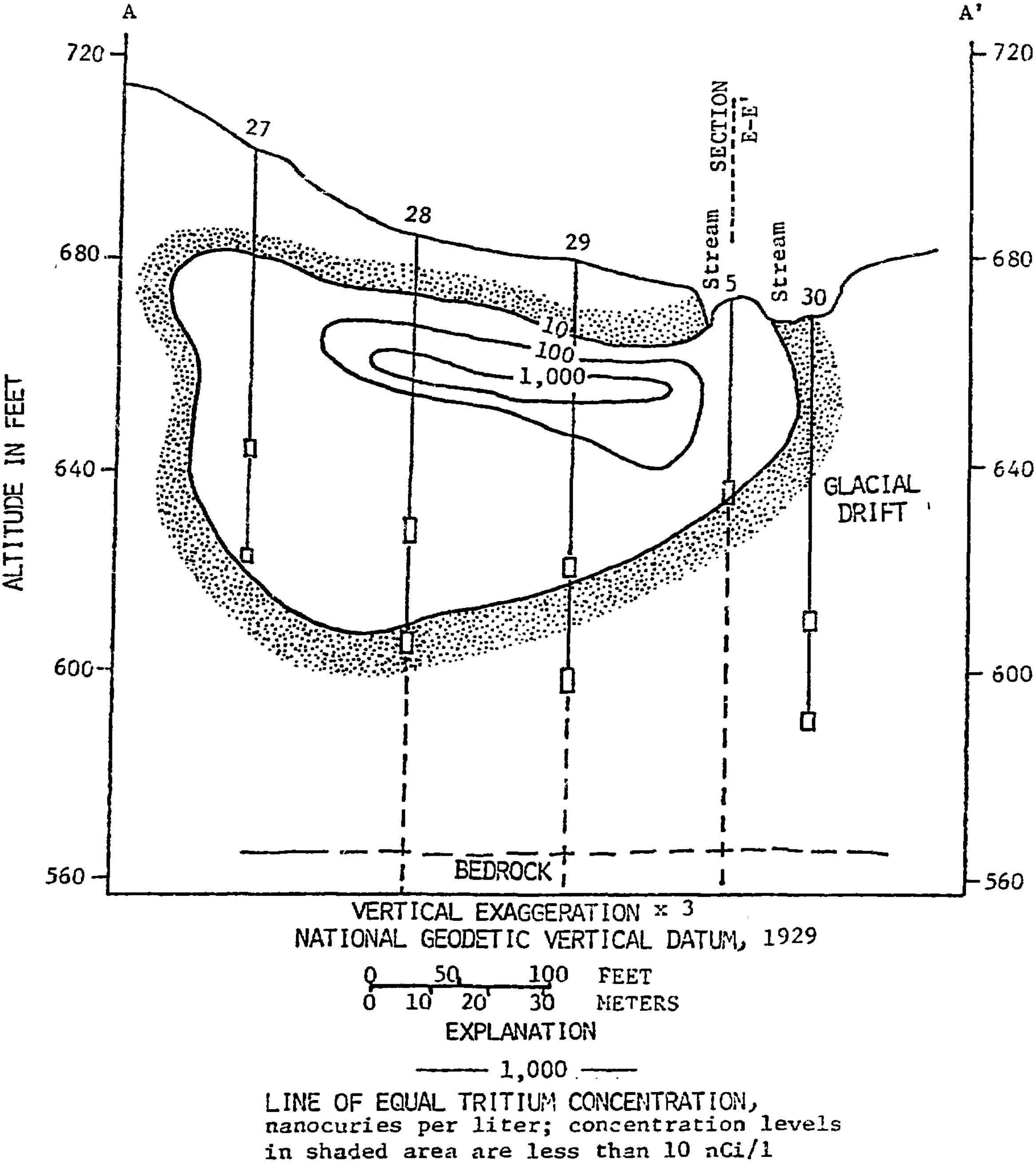

Figure 6. Cross Section A-A'; Leading, Northern Edge of Tritium Plume.

SURFACE WATER STUDIES

Tritiated water measurements were made on samples from 13 locations in the streams draining Plot M (Figure 7). The surface drainage around Plot M is principally by the intermittent stream that travels south to north along the east side of the Plot. A smaller stream drains the west side of the Plot and joins the main stream north of the Plot as shown in Figure 7. The stream continues north until it empties into the Illinois and Michigan Canal about 800 yards north of Plot M. It is only after heavy precipitation, normally in the spring, that the flow reaches the canal. Under lighter rains, the water usually infiltrates into the coarse gravel bed in the vicinity of Red Gate Woods; and during much of the year the entire stream bed is dry.

The general trend of tritium in surface water from 1976 to 1981 was to increase in concentration as water moved from upstream to below the Plot M area, and then to decrease downstream of Plot M as a result of dilution by waters from overland, subsurface, and groundwater flows. All samples were taken from the stream itself, except the sample collected at location #6. This was a low volume seep at the base of the Plot M hill whose water flowed into the intermittent stream bed northeast of the burial area.

Concentrations at locations #3, #4, and #5 are nearly identical, averaging 90 nCi/l, as they all are directly east of the Plot and probably receive the same amount of dilution from water flows in the adjacent land surfaces.

At location #6, the low volume seep at the base of the Plot M hill, tritium concentrations are at least 4 times greater, averaging 810 nCi/l, than those in water from the other sampling locations. The seep is directly north-east of the burial site and does not receive as much dilution as those locations in the stream bed.

Location #9 averages 200 nCi/l of tritiated water which is the greatest of all surface water locations (not including the seep). Sampling location #9 is at the confluence of the streams draining the Plot, which results in higher concentrations due to the joining of flows from both streams. Tritiated water levels decrease north and downstream of location #9.

Other surface water samples in the area were analyzed for tritiated water. Water from the Sanitary and Ship Canal and the Des Plaines River in the vicinity of the Palos Forest Preserve, and Illinois & Michigan Canal water, downstream from the point where the intermittent stream enters, were analyzed for tritium and the concentrations were less than the detection limit of 0.2 nCi/l.

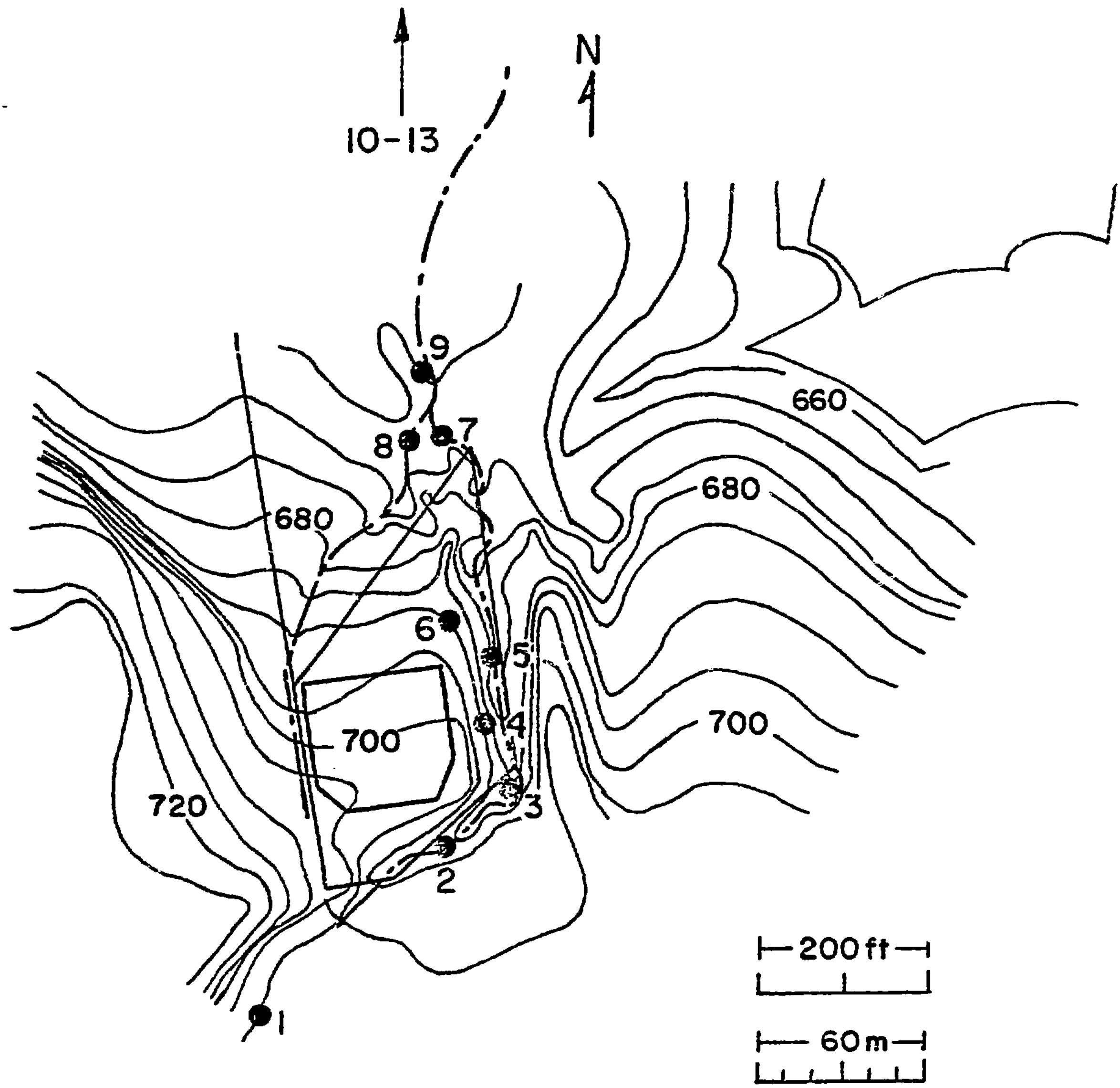

Figure 7. Surface Water Sampling Locations.

In addition to tritiated water, radiochemical analyses were performed on some surface water samples for strontium-90, uranium, plutonium-239, and other transuranic nuclides. Very small amounts of strontium-90 (3 pCi/l) and plutonium-239 (0.003 pCi/l) were detected in the seep, location #6; although the concentrations are in the range found for fallout from nuclear weapons testing.

SURFACE SOIL STUDIES

Surface soil samples were collected near Plot M in the fall of 1979. The samples were collected to a depth of six inches. They were analyzed for strontium-90, cesium-137, radium-226, thorium, uranium, and plutonium, and the soil moisture analyzed for tritium. Strontium-90, cesium-137, and plutonium concentrations were within average fallout levels. The other nuclides, if present, were in concentrations less than detectable.

In the fall of 1976 and in September 1978, surface soil samples were collected at Site A. The cores were analyzed by gamma-ray spectrometry, and the moisture analyzed for tritium. The results show that small amounts of radionuclides from Site A operations persist at various locations in the surface soil south of the buried CP-3 shield, including higher than background concentrations of cesium-137, antimony-125, cobalt-60, and strontium-90. Beyond about 250 feet south of the buried shield, the surface contamination diminishes. The radionuclides present show evidence of activities carried out at the site during research and waste disposal operations and are present at levels which do not present a health hazard.

DISTRIBUTION COEFFICIENTS FOR SELECTED RADIONUCLIDES IN SEDIMENT SAMPLES

Laboratory batch sorption experiments were conducted by Battelle Pacific Northwest Laboratories (PNL) on several soil samples from the study area to analyze the migration potential of tritium, cesium, strontium, uranium, and plutonium (Sherwood and Serne, written communication, 1981).

Batch distribution coefficients (Kd)were calculated for each radionuclide. The Kd is defined as the ratio of the equilibrium radionuclide concentration on the solid phase (microgram per gram), to the equilibrium concentration of the radionuclide in solution (microgram per milliliter). The Kd is dependent upon and therefore reflects - a particular ion, the geohydrologic media, and temperature. If physical and/or chemical parameters are changed, the Kd is likely to vary. A high Kd value results in low radionuclide migration velocities relative to groundwater velocities.

The strontium, cesium, and plutonium Kd values are quite large and suggest that these elements will be rather immobile in the geohydrologic regime. This finding is similar to numerous other observations compiled in a critical review (Ames and Rai, 1978). The amount of these nuclides present at the burial site, in the soluble form capable of interacting with sediments is low (Golchert and Sedlet, 1978). Thus, the potential hazard to the geosphere and biosphere away from the immediate burial site from these nuclides is expected to be extremely small.

The uranium concentrations observed in soil borings collected directly beneath the Plot M cap are in the range of natural uranium,and the Kd value is sufficient to retard migration for long times. Thus, uranium is not expected to create above natural background hazards in the surrounding environs.

The Kd values for tritium were found to be very nearly zero, which means the tritiated water should move essentially with the groundwater, and the rate should be subject to those conditions which generally affect soil water movement: amount of precipitation, hydraulic head, etc. The rate of groundwater movement in the drift has been calculated to be 3.1 feet per year; thus,this is the rate at which the tritium plume travels through the drift.

WELL WATER STUDIES

Five hand-pumped forest preserve picnic wells and four other wells drilled in 1976 to the dolomite aquifer, have been monitored regularly since 1976 for radioactivity. Tritium is the only radionuclide present in higher than background concentrations in the well water. Tritium has travelled vertically into the aquifer beneath Plot M, and laterally through the dolomite to wells as far as 1000 yards north of the Plot.

The greatest concentrations are found at the Red Gate Woods well, (Figure 2) where tritium levels range from < 0.2 nCi/l to 14 nCi/l, and vary seasonally with highest concentrations in the winter and lowest concentrations in the summer. The average tritiated water concentrations are presented in Table 1 for the Red Gate Woods well from 1974 to 1981. Also shown are the annual averages for the well opposite Red Gate Woods, which are much lower than at RGW.

Since the center of the tritium plume in the drift is still about 80 feet above bedrock and is moving deeper into the drift, the more concentrated centers of the plume are reaching the bedrock. This explains why tritium levels in the RGW well have not appreciably declined since 1973, although more than 25 percent of the tritium present in 1973 has decayed (the half life of tritium is 12.4 years). The seasonal fluctuation of tritium concentrations in water at the RGW well, then, are a rough measure of the relative amount of tritium annually entering the dolomite from the drift

TABLE 1

Annual Average - Tritiated Water
(nCi/1)

	Red Gate Woods Well, #5167		Opp. Red Gate Woods Well, #5159	
	Measured	Decay Corrected (to 1974)	Measured	Decay Corrected (to 1974)
1974	7.0	7.0	(4.8)	(4.8)
1975	6.6	7.0	1.7	1.8
1976	6.1	6.8	1.9	2.1
1977	8.2	10.2	2.1	2.5
1978	7.5	9.4	1.3	1.6
1979	7.1	9.4	0.96	1.3
1980	7.0	9.8	1.02	1.4
1981	4.3	6.3	1.06	1.6
		Percent Change		
1974-75	-6	0	-	-
1975-76	-8	-3	+12	+17
1976-77	+34	+50	+11	+19
1977-78	-9	-8	-38	-36
1978-79	-5	0	-26	-19
1979-80	-1	+4	+6	+8
1980-81	-39	-36	+4	+14

below Plot M.

The precise pathway the water follows within the dolomite from beneath Plot M to the Forest Preserve wells is not known. The aquifer is known to be highly fractured and to contain many clay-filled solution cavities (Olimpio, 1982). The position and size of these fractures and cavities may determine the travel route of tritiated water through the dolomite.

The rate of water movement in the aquifer is not known. Although tritium was discovered in the RGW well in 1973, it is not known how long it had been present there.

CONCLUSIONS AND RECOMMENDATIONS

This study concludes that there is no present danger to the health and safety of the public posed by the radioactive materials buried at Site A and Plot M and that none is likely to occur in the future. Removal of these buried materials would not reduce the tritium levels in the groundwater since this radioactive material has moved out of the plot down to deeper levels in excess of 60 feet. There is no evidence that radionuclides other than tritium are leaving the site.

Consequently, the study recommends that the site remain undisturbed and that radiological monitoring be continued and, in some areas, expanded to develop additional information on the groundwater dynamics, radiological condition, and geochemical properties in the area.

ACKNOWLEDGEMENTS

This study is supported by the U.S. Department of Energy and the U.S. Geological Survey. The author thanks J. Sedlet and N. W. Golchert for helpful discussions and criticism of the manuscript.

REFERENCES

1. Ames, L. L. and D. Rai, 1978,"Radionuclide Interactions with Soil and Rock Media,Vol. 1," EPA 520/6-78-007.

2. Golchert, N. W., and Sedlet, J., 1978, "Radiological Survey of Site A and Plot M ", U.S. Department of Energy Report DOE/EV-0005/7, 89 p.

3. Olimpio, J. C., 1980, "Low-Level Radioactive Waste Burial at the Palos Forest Preserve, Illinois, Part I. Preliminary Finite-Difference Models of Steady State Ground-Water Flow ", U.S. Geological Survey Open-File Report 80-775, 40 p.

4. Olimpio, J. C., 1982, "Low-Level Radioactive Waste Burial at the Palos Forest Preserve, Illinois: Part II Geology and Hydrology of the Glacial Drift, as Related to the Migration of Tritium," U.S. Geological Survey Open-File Report 82-78, 52 p.

5. Willman, H. B., Glass, H. D., and Frye, J. C., 1963, "Mineralogy of Glacial Tills and their Weathering Profiles in Illinois. Part 1 - Glacial Tills," Illinois State Geological Survey Circular 347, 55 p.