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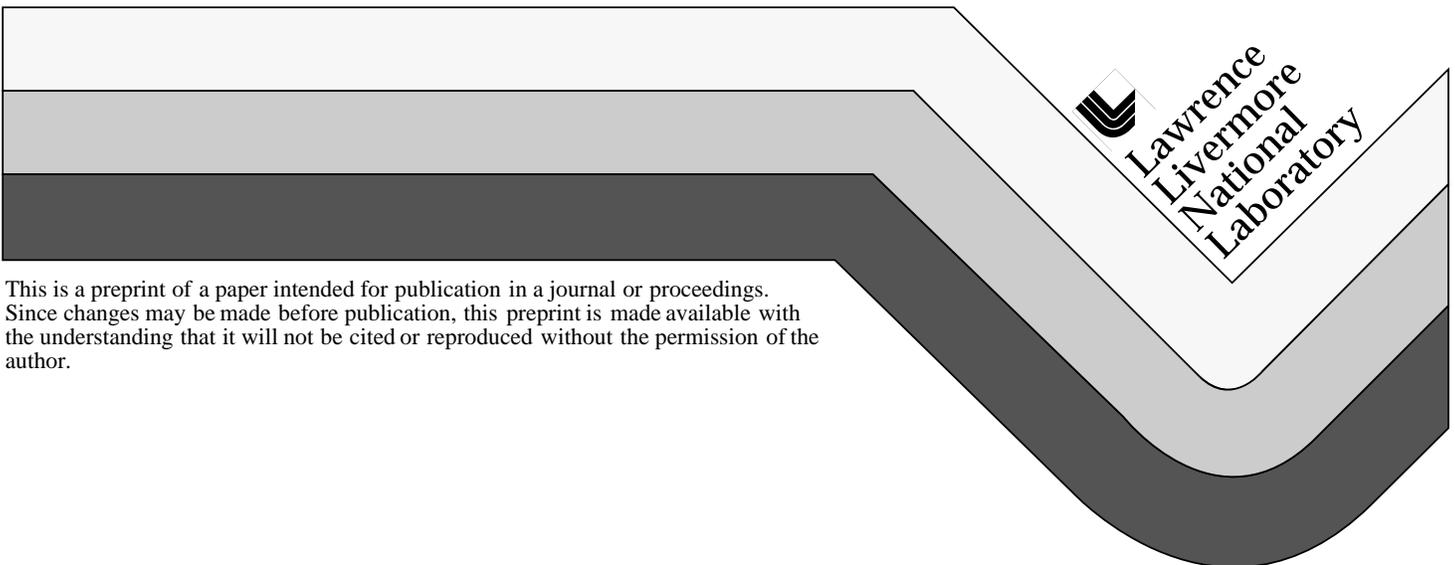
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NATURAL ATTENUATION OF TRITIUM IN VADOSE ZONE MOISTURE AND GROUND WATER AT A LAWRENCE LIVERMORE NATIONAL LABORATORY SITE IN NORTHERN CALIFORNIA, USA

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Abstract

Tritium used in explosives experiments and buried in unlined landfills at a remote Lawrence Livermore National Laboratory (LLNL) site has resulted in three ground water tritium plumes. Using an innovative approach, we determined that despite ground water tritium activities of up to 1.5 million picoCuries per liter (pCi/L) in some locations, natural attenuation processes are significantly limiting the migration of tritium to environmental receptors. We used soil vapor and moisture tritium activity measurements to calculate the source inventory of tritium in the vadose zone. We determined the 12 year annual inventory of tritium in ground water, using objective tritium activity contours and the highly variable saturated thickness of the aquifer. Our analysis indicates that despite seasonal slug releases of tritium, the two plumes emanating from two landfills are stable, with the 1,000 and 20,000 pCi/L contours essentially fixed in space. The third plume emanates continuously from an explosives testing platform; the 1,000 pCi/L contour is translating slightly, but the 20,000 pCi/L contour is retreating upgradient towards the source. Additionally, the long-term trend in total tritium activity for each plume is decreasing. Three processes account for the attenuation of tritium observed: 1) radioactive decay, 2) hydrodynamic dispersion, and 3) dwindling tritium sources. In preparation for the possibility that remediation may be required anyway, we have evaluated innovative remediation technologies for tritium at this site.

Introduction

Analysis of ground water elevation and vadose zone and ground water chemical data collected over several years can provide evidence of the degree to which natural processes are attenuating ground water contamination, and thus limiting the extent to which the contamination can reach environmental receptors. Tritium is an ideal contaminant for such an evaluation due to its perfectly conservative nature in ground water (it actually becomes incorporated into the water molecule) and its short half-life (12.3 years). At the site selected for this study, three overlapping plumes of tritium in ground water emanate from each of two unlined landfills (Pits 3 and 5) and an explosives test platform (the Building 850

firing table) at LLNL Site 300. Site 300 is located in a semi-arid area of rugged topography near San Francisco, California (Figure 1). Ground surface elevations vary



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Figure 1. Location of LLNL Site 300.

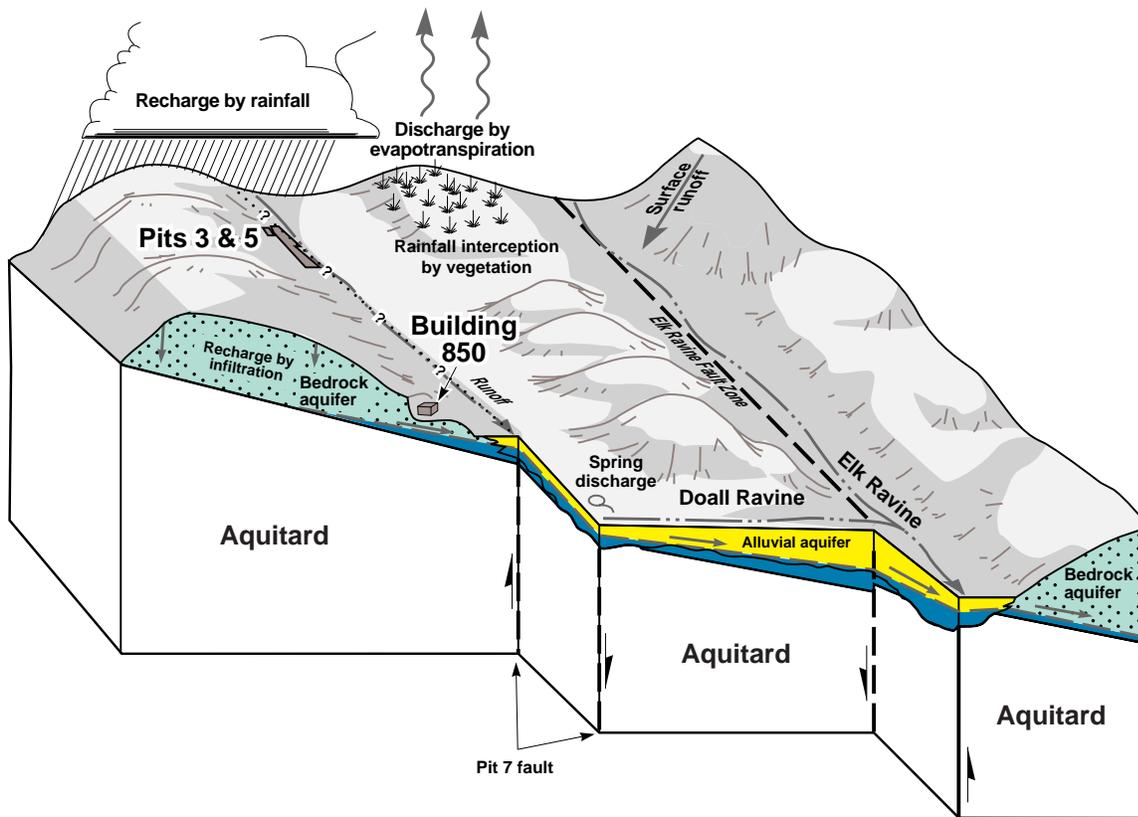
from 150 to 520 meters (m) above sea level. Average annual rainfall is about 26.9 cm, most of which falls between October and March. Site 300 is a Federal Superfund Site, and its environmental cleanup is managed jointly by the U.S. Environmental Protection Agency, the U.S. Department of Energy, and the California State environmental agencies. Over 20,000 Curies (Ci) of tritium were used in explosives experiments conducted at the Building 850 firing table. The resulting debris, consisting primarily of pea gravel contaminated with tritium, uranium, and metals, was disposed in the unlined landfills. Ground water monitoring has been conducted at the site since 1981, and the tritium was first mobilized to ground water after the heavy winter rainfall of 1982-83 (1).

The goals of our ongoing investigations, and of this study in particular, were to determine the inventory and areal extent of tritium in ground water over time, the inventory of tritium in the vadose zone that might still be available for

leaching, and the degree to which the tritium in ground water is migrating away from the sources over time. This evaluation is in preparation for determining what actions are necessary to protect human health and the environment for the long-term.

Contaminant Hydrogeology

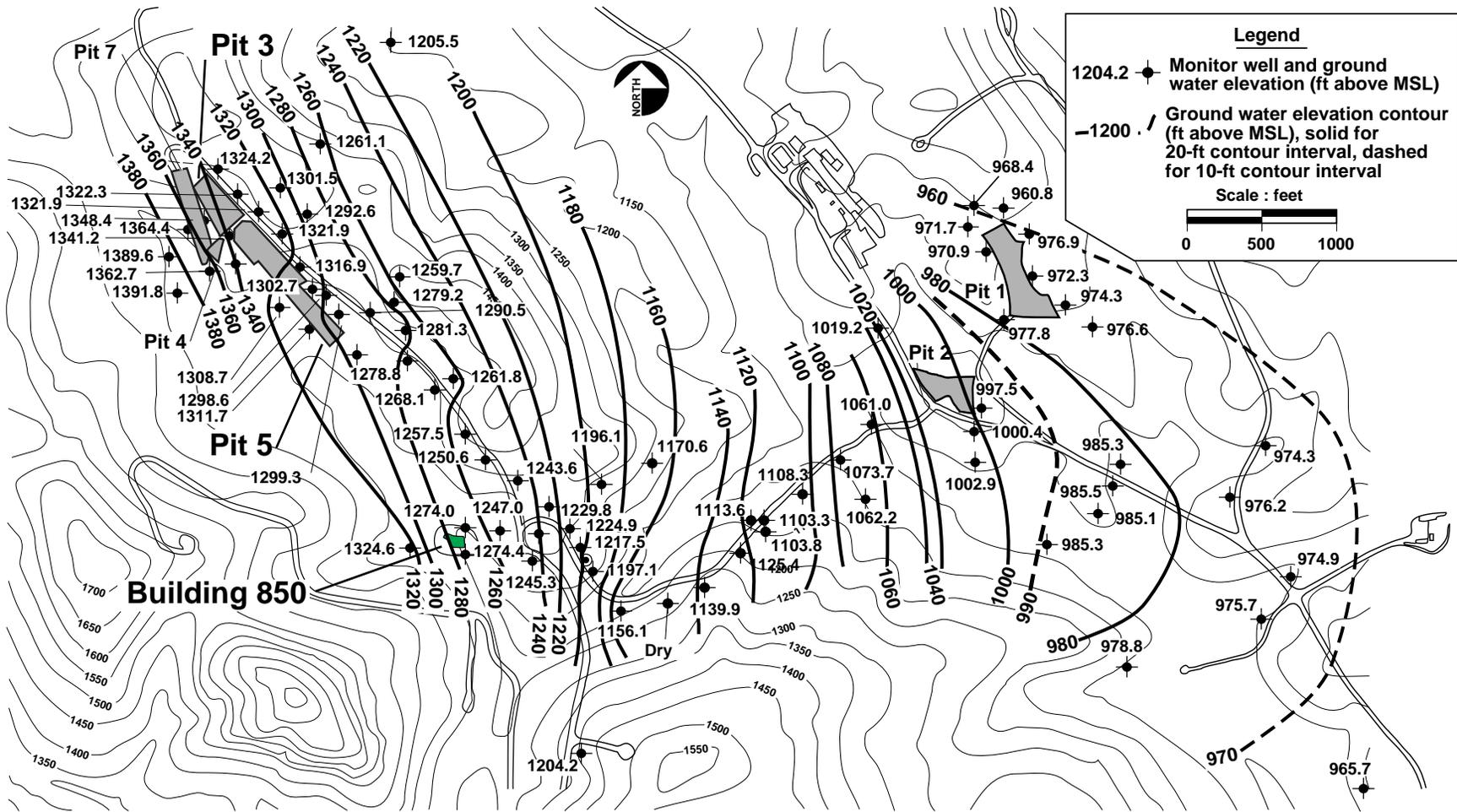
An areally extensive perched water-bearing zone underlies the Building 850/Pits 3 and 5 area (Figure 2). It averages 2 m thick and the water table surface



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Figure 2. Conceptual hydrogeologic model for the Building 850/Pits 3 & 5 area.

averages 7 m below the valley bottom and 11 m below the Building 850 firing table. Ground water flow in the bedrock is east-northeast (at a gradient of about 0.1) and southeast in overlying alluvium (Figure 3). Seepage velocities in the bedrock are



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Figure 3. Potentiometric surface map for the first continuous water-bearing zone beneath the Building 850/Pits 3 and 5 area, October 1996.

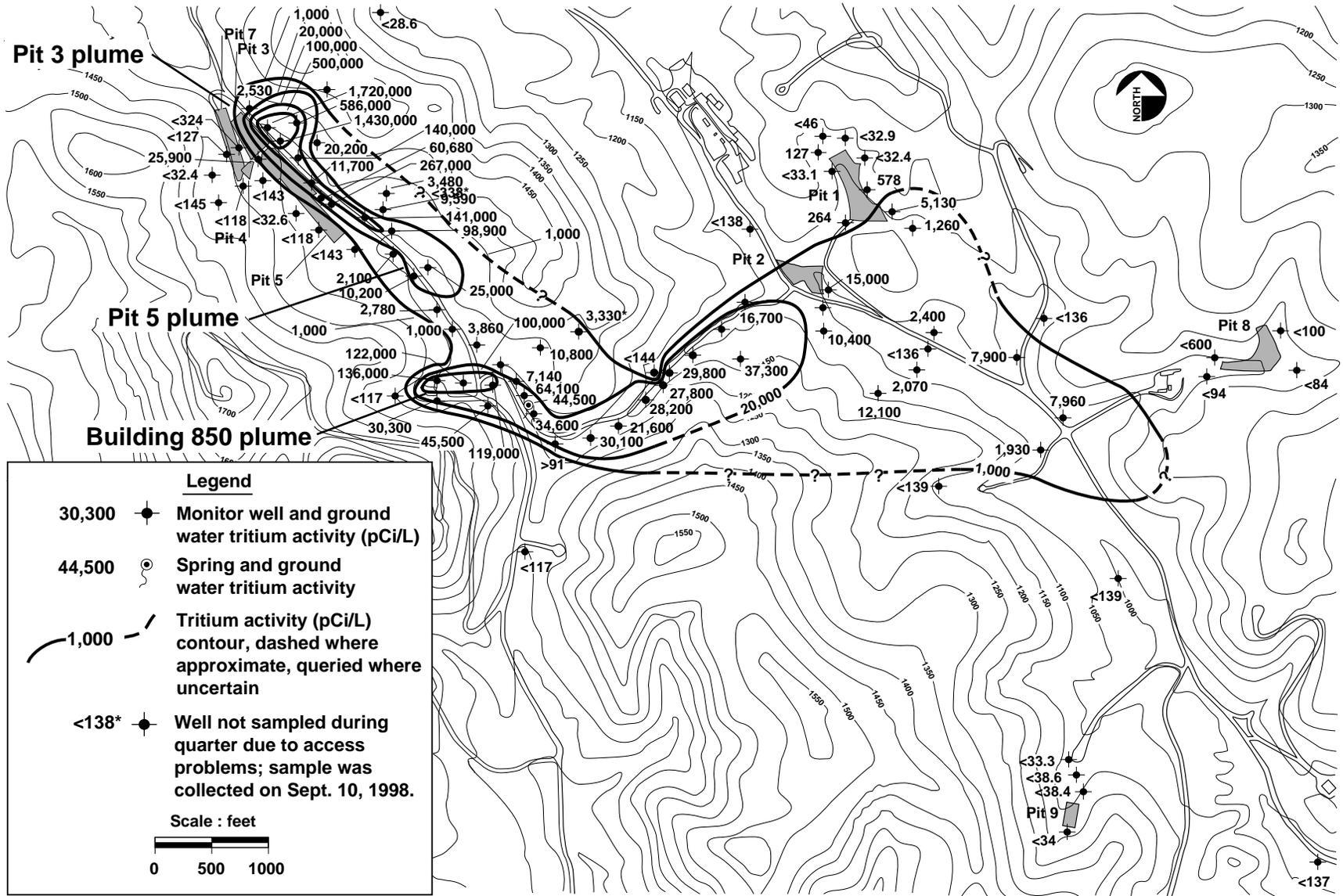
typically about 6-15 m/yr. In the alluvium, seepage velocities are about 40 m/yr. Flow in alluvium occurs only after periods of high rainfall, when the alluvium becomes saturated. During and immediately following winters of high rainfall, the water table may rise by as much as 4 m in a period of several weeks. This water table rise occurs preferentially beneath the landfills. Over the time period from November to February, the water table high propagates downgradient. During the past several winters, lower portions of Pits 3 and 5 have been inundated by the rising water table, resulting in periodic slug releases of tritium to ground water.

At the Building 850 firing table, tritium appears to be released by a more continuous mechanism. There, rainfall recharges the colluvium and flows along the colluvium-bedrock interface, and then laterally at the break in slope into the shallow firing table materials. From the shallow firing table materials, this water percolates to the water table, carrying tritium from the vadose zone 11 m vertically to ground water.

The current maximum tritium activity detected within the Pits 3 and 5 plumes is about 1.72 million pCi/L. The Pits 3 and 5 plumes together are about 1,250 m long and about 300 m wide (Figure 4). The plumes are moving slowly downgradient to the east-northeast in the bedrock and extend 300 m downgradient in that direction. Due to periodic alluvial saturation, the plumes have traveled down the alluvial valley fill and extend 1,825 m to the southeast, where they have merged with the tritium plume emanating from Building 850. The current maximum tritium activity detected in the Building 850 plume is about 136,000 pCi/L. The Building 850 tritium plume extends about 3,040 m from its source at Building 850 and is a maximum of about 1,075 m wide. In addition to seepage velocity constraints on the migration of the plumes, there are several other geologic controls limiting their migration to the Site 300 boundary. The extent of saturation is limited to the east-northeast, which is the direction of ground water flow. The strata containing the tritiated ground water crop out about 2,500 m northeast of the landfills (1,2,3, and 4).

Methods

Several aspects of the hydrologic system required quantification in preparation for assessing changes in tritium distribution in ground water and the vadose zone over time. These included: the degree of ground water inundation of the landfills, residual tritium inventory in the vadose zone and the landfills, and the saturated thickness and volume of the aquifer. To track the likelihood for slug release of tritium to ground water and to relate a particular degree of inundation to later downgradient ground water tritium distribution, we created maps showing the



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Figure 4. Tritium activity contours for ground water in the first-water bearing zone, 2nd quarter 1998, Building 850/Pits 3 and 5 area, LLNL Site 300.

degree of pit inundation by ground water. The height of ground water within the pits was calculated by subtracting the elevation of the contoured bottom of the pits from the elevation of the contoured water-bearing zone. To determine whether there was still significant tritium in the vadose zone and landfills that was still available for leaching, we calculated the total tritium inventory in these compartments. Residual tritium in the soil was calculated using the proprietary software package EarthVision (4). The maximum tritium activity detected in the soil moisture, based on data from geologic logs of boreholes, was contoured logarithmically. We then used EarthVision to calculate the area between contours. A volumetric moisture content of 15% was assumed for the Pits 3 and 5 area, and a moisture content of 10% was assumed for Building 850, based on moisture content. The residual activity in the soil was calculated by multiplying the area between the contours, as calculated by EarthVision, by the vadose zone thickness, the volumetric moisture content, and the geometric mean of the tritium activity for that contour interval. Contributions were summed to yield a total tritium activity in the vadose zone.

To determine the total residual tritium activity in the landfill pits at that time, we hand-contoured soil vapor tritium data collected from the pits on a logarithmic interval. We assumed a volumetric moisture content of 15% and a 3 m thickness for the landfill debris, based on the depth of penetration of the soil vapor drive points. The total residual activity in each of the two landfills was calculated by multiplying the area of the contour interval by 3 m, times the volumetric moisture content, multiplied by the geometric mean of the tritium activity for that contour interval.

To quantify the total tritium inventory in ground water, the aquifer thickness or volume distribution was required to determine the total tritium activity in each contoured ground water tritium region. To calculate the saturated volume of the aquifer, we first determined the areal distribution of the elevation of the bottom of the first water-bearing zone by reviewing geologic logs, determining the depths to very low permeability rocks, and reviewing the completion intervals for monitor wells. The elevations were contoured by EarthVision. The contours for the landfill bottom elevation(s) were then subtracted from the contours of the water table to calculate the volume of saturated aquifer. This volume calculation was generally performed for regions defined by tritium activity contours, thereby calculating an aquifer volume corresponding to a specific range of tritium activities. The aquifer volume was converted to a ground water volume by multiplying by the estimated total porosity (0.20).

To determine the total tritium activity in a ground water plume, the tritium data for a given quarter were contoured using EarthVision. In order to more accurately contour these data, simulated wells were positioned in the model, generally in downgradient locations, for areas where the plume was not well bounded, or to create a boundary between the two release areas (i.e., the area just north of Building 850 where the two plumes are commingled). While the use of

simulated wells created an artificial constraint on the plume, it was necessary to close the contours so that a volume of aquifer could be calculated. Simulated wells were also positioned at two locations downgradient of the Pit 5 portion of the plume, and on strike with a well which monitors known ground water tritium activities. The simulated wells were placed in regions where tritium activities were expected to be at or near background levels. The region with the greatest uncertainty as to the placement of the simulated points is east of the pits. During 1999, we intend to complete one or more new wells in this area to further substantiate our findings. We used the same technique discussed above for calculating the area for each averaged ground water tritium activity. The geometric mean for each interval was multiplied by the volume of ground water for that interval to yield an activity in Ci. The activities for the different intervals were then summed to give a total tritium activity for the plume. Separate calculations were performed for the Pits 3 and 5 plumes and for the Building 850 plume (5).

Ground Water Recharge and Water Table Fluctuations

The distribution and timing of ground water recharge at the site are principally responsible for the observed mobilization of tritium to ground water. Topography has a strong influence on this, because the pits and the Building 850 firing table are both located in the bottom of the valley. The hillslopes west of these facilities slope towards these tritium source areas, in effect amplifying the volume of water available from rainfall falling over a large catchment area, and conveying it preferentially into the landfill and firing table areas. At the Pits 3 and 5 area, most recharge occurs on the upgradient (west) side of the valley and results from shallow saturated flow above the alluvium/bedrock contact. This water appears to percolate to the bedrock aquifer once it reaches the valley bottom (2). The case is similar at Building 850, where this colluvial water laterally enters the shallow firing table materials and then percolates 11 m to ground water.

In the last three water years, slightly below average (1996–97), somewhat above average (1995–96), and twice the long-term average annual rainfall (1997–98), respectively, have occurred in the Site 300 area. The 38-year average annual rainfall for Site 300 is 26.9 cm. Although total precipitation for the 1995–96 rainfall year was only slightly greater than the average rainfall (33.9 cm), 25.8 cm of the rain fell in the three-month period from December 1995 to February 1996. Due to this prolonged period of heavy rainfall, which also followed a year of greater-than-average rainfall (40.6 cm fell during the 1994–95 rainfall year), ground water elevations observed during the spring of 1996 were the highest recorded since 1982–83, the wettest year on record (60.0 cm). Ground water elevations continued to increase even during the slightly below average (26.1 cm) rainfall year of 1996–97 because of very intense rainfall events in December 1996 and January 1997 (two-month total of 17.1 cm). The total rainfall (54.2 cm) for the 1997–98 El Niño water year, however, produced the greatest ground water elevation increases to date, such that the water table is near record high elevations equivalent to those observed when monitoring first began during the last El Niño winter in 1982–83. The near

record high water elevations were the result of several consecutive above-average rainfall years (1994–1998).

At Pits 3 and 5, the relationship between annual winter rainfall (October through March) and net recharge volume for the same time period indicates there is a linear relationship between rainfall and recharge for this small ground water basin. Rainfall in excess of approximately 15.2 cm over the half-year time period appears to be required to produce net recharge. The greatest water table rises continue to occur in the valley bottom and beneath the hills to the west. Wells completed in the hills to the east of the valley exhibit much smaller water table elevation rises. Although the Pits 3 and 5 landfills were initially constructed above the water table, the above average rainfall of 1981–1983 caused the water table to rise locally into previously unsaturated valley alluvium and into Pits 3 and 5, mobilizing tritium into ground water. Subsequent above average rainfall in 1986–1987, 1992–1993, and the three water years discussed here has caused additional mobilization of tritium, although much of this may be from residual tritium in vadose zone soil and rock moisture below and adjacent to the pits.

During the winter storms of 1992–93 and 1994–95, the water table rose preferentially in the vicinity of the southern end of Pit 5 (2). The water table rises generally continued in this pattern in 1995–96 and 1996–97, but changed last year. Based on the distribution of maximum water elevation rises for October 1997 through June 1998, the maximum rise extended further to the north, encompassing all of Pit 5 and the southern portion of Pit 3.

The timing of the recharge followed a pattern similar to that previously observed. Ground water elevations began to rise on the western side of the landfills in November 1997, but did not rise in the area east of the landfills until January 1998. Based on the rapid ground water elevation rises, recharge appears to occur primarily west of the landfills, with the pulse of recharging water propagating downgradient (east-northeast). The recharge pulse migrates from the west side to the east side of the valley over approximately four months. Minimal recharge appears to occur east of the valley (2).

We estimated the bottom elevations of Pits 3 and 5 and compared them to the ground water elevations for several time intervals during the 1998 winter. A very small area in the center of Pit 5 was inundated in January 1998. By mid-February most of the central portion of Pit 5 was inundated. By mid-March a small portion of the southern part of Pit 3 was inundated. During April, May and June, the degree of inundation decreased, and by June it was similar to what it had been in February. In 1998, the south-central portion of Pit 5 was inundated by more than 2.4 m of water for a period of months. Since the bottom elevation of Pit 3 is not well known, the estimated degree of inundation of that pit is less certain.

Fluctuations in ground water elevations in the Building 850 area have typically been less than 1.5 m. However, during the past winter they reached almost

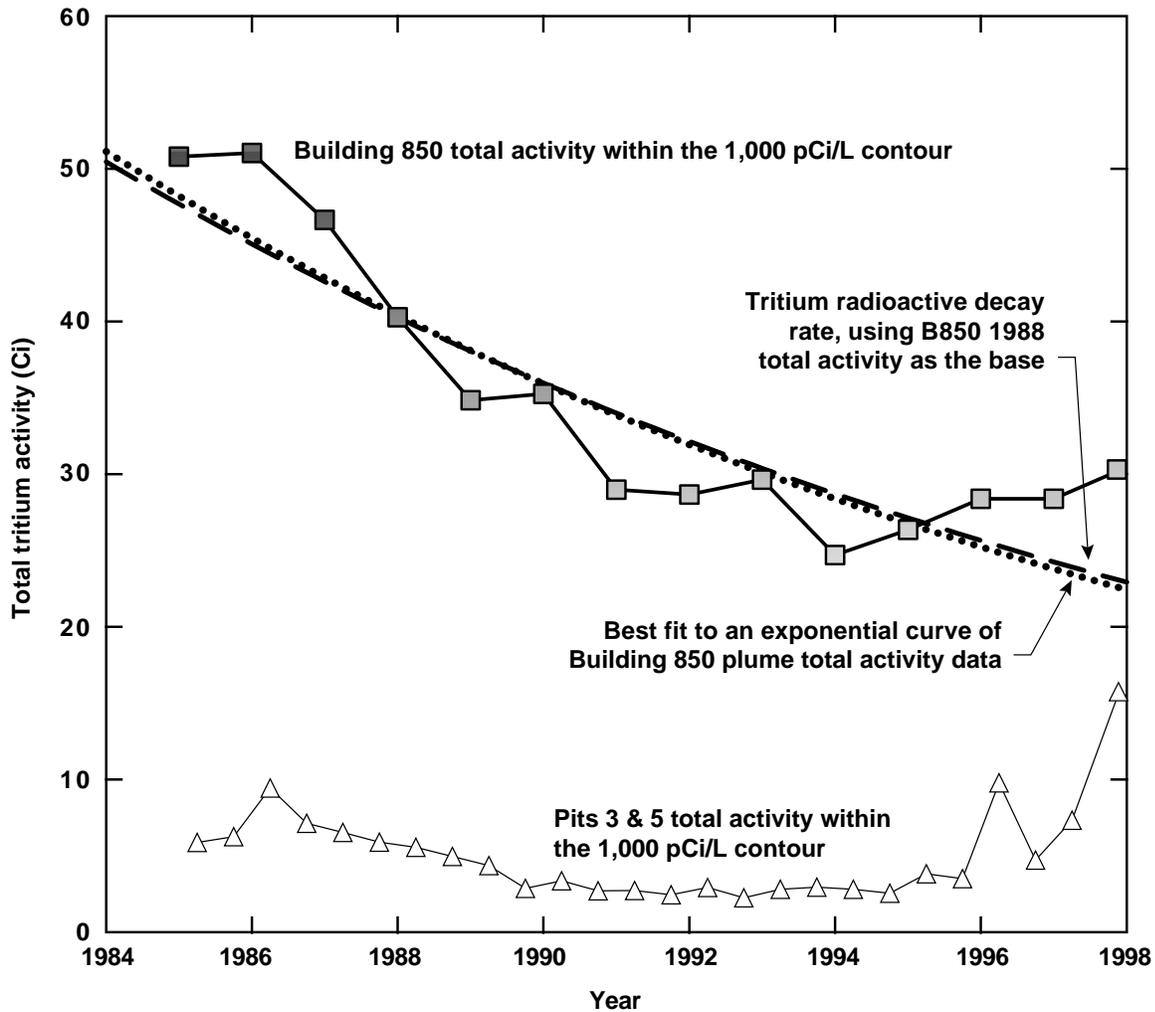
2.1 m at several wells. Ground water elevations in 1998 were among the highest recorded for these bedrock wells near Building 850 during the 14-year period of record. Recharge appears to occur primarily through the hillslope west and above Building 850. Infiltrating rain water may also flow downslope within colluvium and vertically percolate to ground water through the firing table. Recharge in the bedrock to the east appears minimal, based on the smaller and later water elevation rises (5).

Landfill and Vadose Zone Tritium Inventory

To estimate the tritium that could be mobilized from the vadose zone to ground water, an inventory of residual tritium based on analyses of soil core samples was performed. A similar inventory was made for the landfills based on tritium in soil vapor extracted from the landfills. These inventories were calculated by contouring 1985-86 tritium activity data from soil cores and soil vapor analyses over the volumes in the vadose zone. We calculated about 7 Ci remain in Pit 3 and 15 Ci remain in the vadose zone adjacent to the landfill. We calculated that 0.2 Ci of tritium remain in Pit 5 and 2.5 Ci remain in the immediate vadose zone. Much of the tritium has migrated from the pits, particularly from Pit 5, and is now in the vadose zone or the ground water. Within and adjacent to Pit 3, tritium still exists which could become mobilized if water elevations were to rise to levels similar to 1982-83. The residual tritium in soil underlying the Building 850 firing table in 1988 (1.1 Ci) is small compared to the total 1988 tritium activity in ground water (40 Ci). By 1997 we estimated that 99% of the initial (1988) tritium at the firing table source area was removed by migration to ground water or by radioactive decay, i.e., the 1.1 Ci of tritium in the vadose zone in 1988 would have decayed to 0.7 Ci by radioactive decay alone. With a greatly reduced source term and rapidly declining ground water tritium activities, the Building 850 tritium plume is not expected to experience marked increases in activities in the future or pose a health risk at the site boundary. This conclusion is based on a greatly reduced tritium source term, observations of dispersive mixing and radioactive decay, and fate and transport modeling (3).

Total Tritium Activity in Ground Water as a Function of Time

The total tritium in the ground water in the Pits 3 and 5 and Building 850 plumes was calculated semi-annually from June 1985 to June 1998 (Figure 5). At



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Figure 5. Building 850/Pits 3 and 5 total tritium activity in ground water over time.

Pits 3 and 5, the total ground water tritium activity declined sharply through the first few years of monitoring and then stabilized at about 2.5 Ci. After the 1987 to 1991 drought years, the water table rose to heights only previously reached during the major tritium release in 1982–83. Subsequently, the total ground water tritium activity increased again, with the highest total activity to date occurring during the

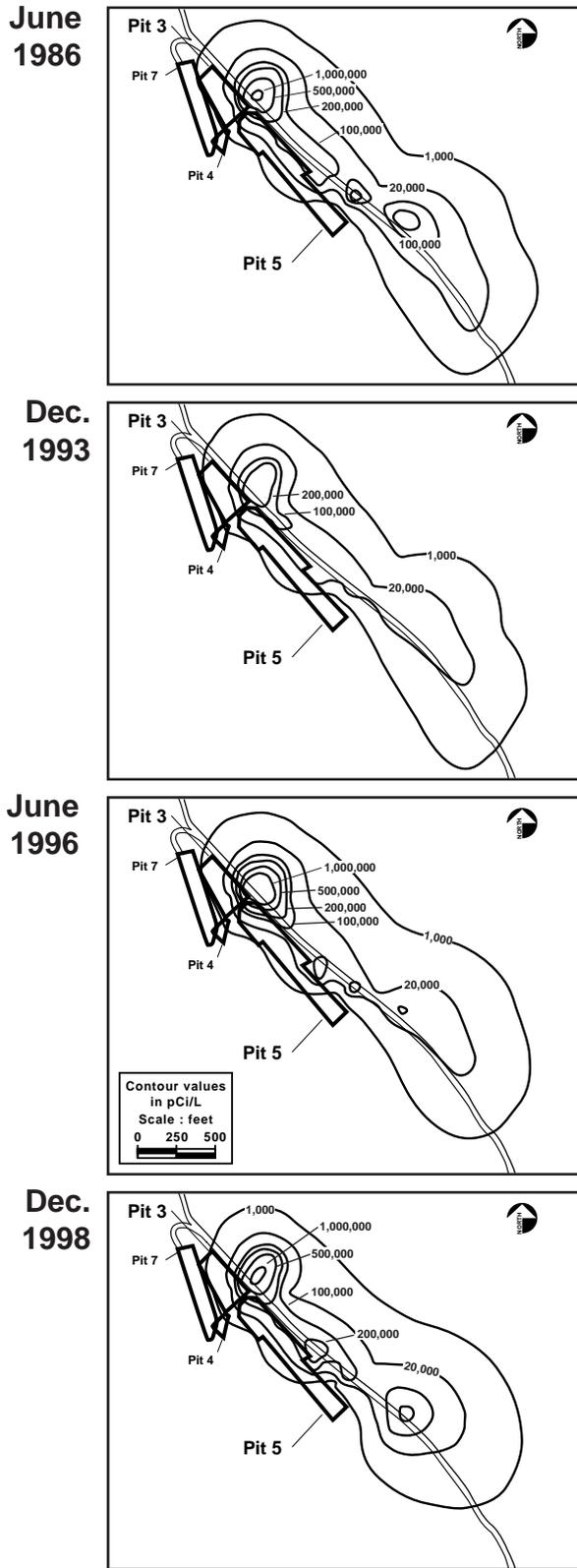
second quarter of 1998 (17.7 Ci). From 1986 through 1990, the slope of the curve plotting tritium activity vs. time has been less than that of the radioactive decay curve for tritium. From 1990 through 1995, the curve of tritium activity vs. time was fairly flat. Since 1995, the ground water tritium activity inventory has been increasing, to the maximum of 17.7 Ci calculated for the second quarter of 1998. The calculated total ground water tritium activity for the last three years shows a great deal of variability, resulting from the fact that two wells downgradient from the pits had highly variable ground water tritium activities. Ground water tritium activities in these wells varied by almost an order of magnitude between quarterly sampling events. This variability is interpreted to be the result of small, localized slug mobilizations of tritium from the vadose zone, rather than a large-scale release. We expect that the tritium activities will decline as they have in the past, as the water table falls over the next few months.

The curve of total ground water tritium activity vs. time for the Building 850 tritium plume indicates that the total tritium activity has decreased significantly from 1985 to 1994. Beginning in 1994, this trend reversed, and by the second quarter of 1998, there were about 30 Ci of tritium in the Building 850 plume. The decrease in total tritium activity from 1985 to 1994 is principally attributable to radioactive decay, indicating that during that time period, there was no significant addition of tritium to the Building 850 plume.

Tritium Plume Size as a Function of Time

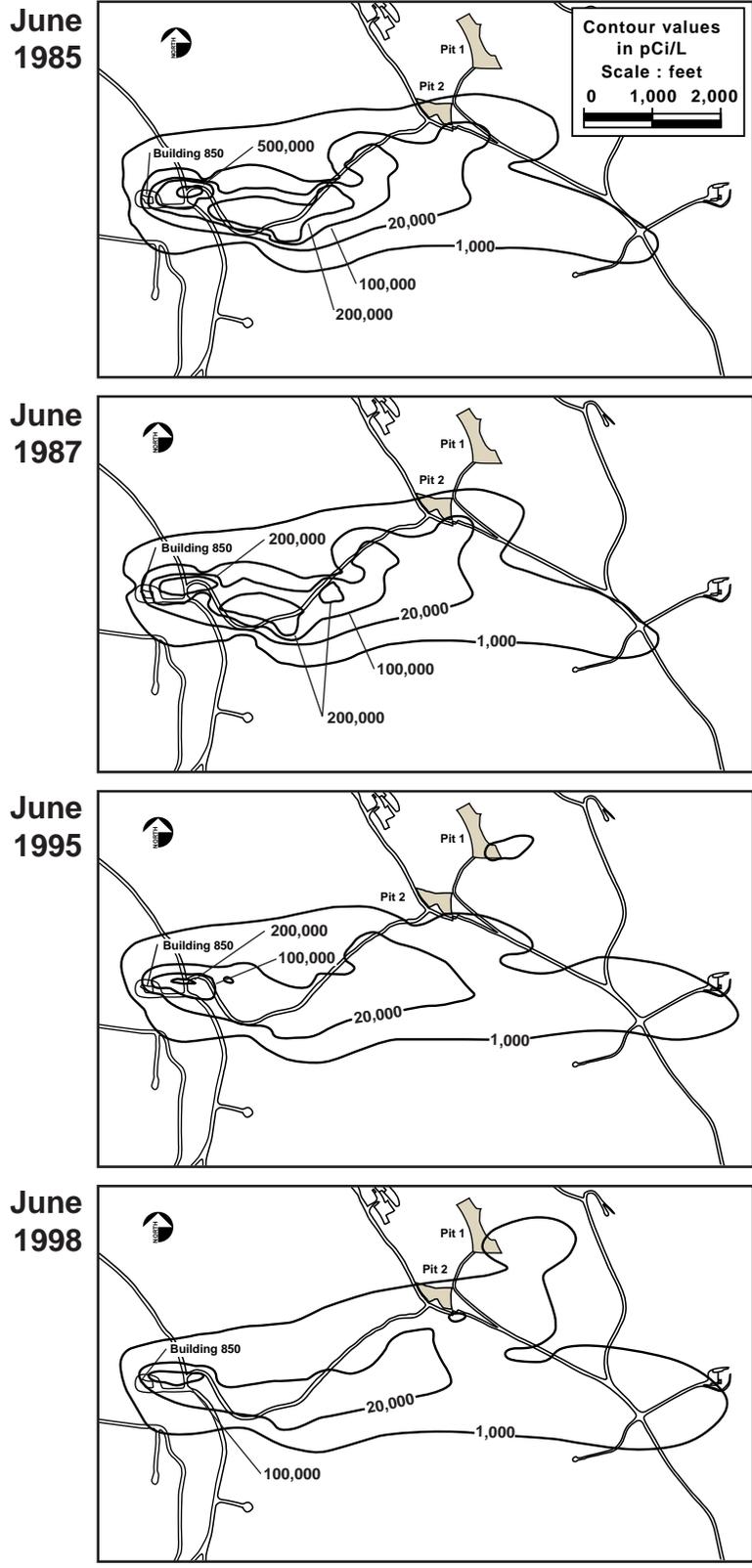
We compared the positions of the 1,000 and 20,000 pCi/L contours over time for each plume to define the migration and extent of the tritium plumes from 1986 to present. For the Pits 3 and 5 tritium plumes, there has been little migration of the 20,000 pCi/L contour over the 13 year period. A comparison of the 1,000 pCi/L ground water tritium activity contours for the Pits 3 and 5 area from 1986 to 1998 was also made (Figure 6). The 1,000 pCi/L activity contour approximately defines the extent of the tritium plume. Comparison indicates little change in the northeasterly extent of the leading edge of the tritium plume, with some increase in length (a maximum of 76 m) to the southeast.

We also compared the 1,000 pCi/L tritium contours over time in the Building 850 area (Figure 7). The extent of the 1,000 pCi/L has remained virtually the same over time except along the distal leading edge of the plume, where there has been an increase of about 300 m in the last few years, reflecting dispersive mixing. As stated previously, the plume extent is increasing only along the distal ends to the east. The length of the plume above the 20,000 pCi/L State and Federal Maximum



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Figure 6. Pits 3 and 5 ground water tritium plumes over time.



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Figure 7. Building 850 ground water tritium plume over time.

Contaminant Level (MCL) for drinking water has decreased from 1985 to 1998 (by about 76 m), reflecting the effects of radioactive decay and dilution by infiltrating surface water. Continued declines in tritium activities should result in further shrinking of the portion of the plume above the MCL. At the same time, the distal portion of the plume, with activities below the MCL, will continue to expand by advection (5).

Conclusions

- Water elevations in the Pits 3 and 5 area are now as high as they were in 1982–83 when ground water monitoring first began, resulting in several feet of landfill pit inundation and consequent mobilization of tritium into ground water (5).
- Tritium within the landfills at Pits 3 and 5 and the adjacent vadose zone could be mobilized by rising ground water, although relatively little tritium appears to exist in the vadose zone beneath the Building 850 firing table (5).
- The horizontal extent of the tritium plumes in the Pits 3 and 5 area has increased very little from 1986–1998, suggesting that natural attenuation by radioactive decay and dispersion is occurring. However, rising annual total tritium activities, and the current 17.7 Ci total ground water activity, the largest estimated compared to the previous high of 10 Ci in 1996, indicate continued re-mobilization of tritium from the pits and/or vadose zone. Additional monitoring will clarify the relationship of the total activity to the actual distribution of tritium in ground water. At present, the tritium plume appears to be naturally attenuating, despite periodic remobilization of tritium to ground water from the sources in the pits (5).
- From 1985–1998, the horizontal extent of the Building 850 ground water tritium plume has increased only along the distal edges. The extent of the 20,000 pCi/L MCL contour has markedly decreased, indicating that natural attenuation is occurring. Until recently, the total tritium activity of the plume had generally decreased at a rate similar to the radioactive decay rate for tritium. During the last few high rainfall years, however, some additional migration through the vadose zone has caused the total ground water tritium activity to increase (5).
- Installation and monitoring of additional wells is necessary to evaluate our conclusions and to monitor the ground water tritium plumes in the long term (5).
- We have not identified any commercially available, nor economically feasible technology for removing tritium from ground water. However, if deemed necessary, several technologies have been proposed for preventing water

from contacting landfill and vadose zone tritium sources. These include interceptor trenches, de-watering wells, and impermeable barriers (4).

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