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Tritium Transport Beneath Surface Impoundments at TA-53 Modeling and Analysis in Support of Data Needs

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1.0 - INTRODUCTION

This document examines tritium transport beneath three surface impoundments at the Los Alamos Neutron Science Center (LANSCE), Technical Area 53 (TA-53). LANSCE produces intense sources of pulsed spallation neutrons, which provide the country's scientific community with the capability to perform experiments that support national security and civilian research. LANSCE is comprised of an 800-MeV, proton linear accelerator (linac); a proton storage ring; production targets to the Manuel Lujan Jr. Neutron Scattering Center and the Weapons Neutron Research facility; and a variety of associated experimental areas and spectrometers. Quantities of liquid radioactive waste are generated during normal operation of the facility. One of the primary constituents of the waste is tritium, which quickly forms water in the environment. From 1970-1995, much of this liquid was treated in three surface impoundments, Potential Release Sites (PRS) 53-002(a) and (b) [LANL, 1998, 58841.2; LANL, 1998, 59997.2].   Tritium has migrated into the subsurface at this site and LANL is studying the plume as a component of ongoing RFI and CMS investigations.

We first present site-specific details that are relevant to the conceptual and numerical models of tritium migration through the subsurface. Next we develop the conceptual model of tritium transport that is constrained by both physics and anthropogenic perturbations. Finally, we implement the conceptual model within the framework of a three-dimensional numerical model and explore a variety of possible tritium transport scenarios at the site as a function of both time and space. The results of the numerical analysis are then used to suggest placement of four new boreholes that will provide additional constraints on the extent of the subsurface tritium plume.
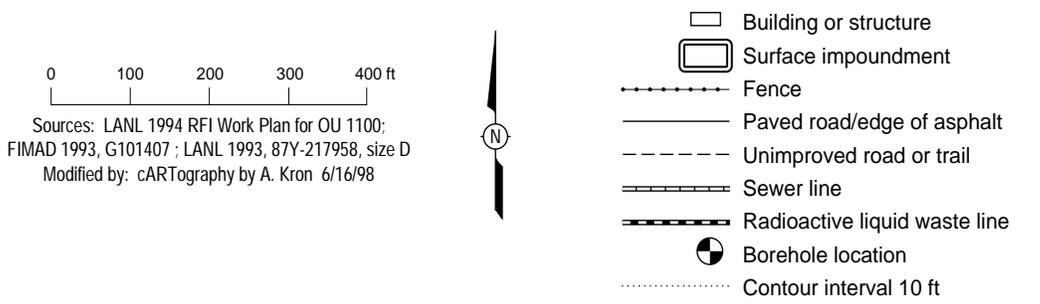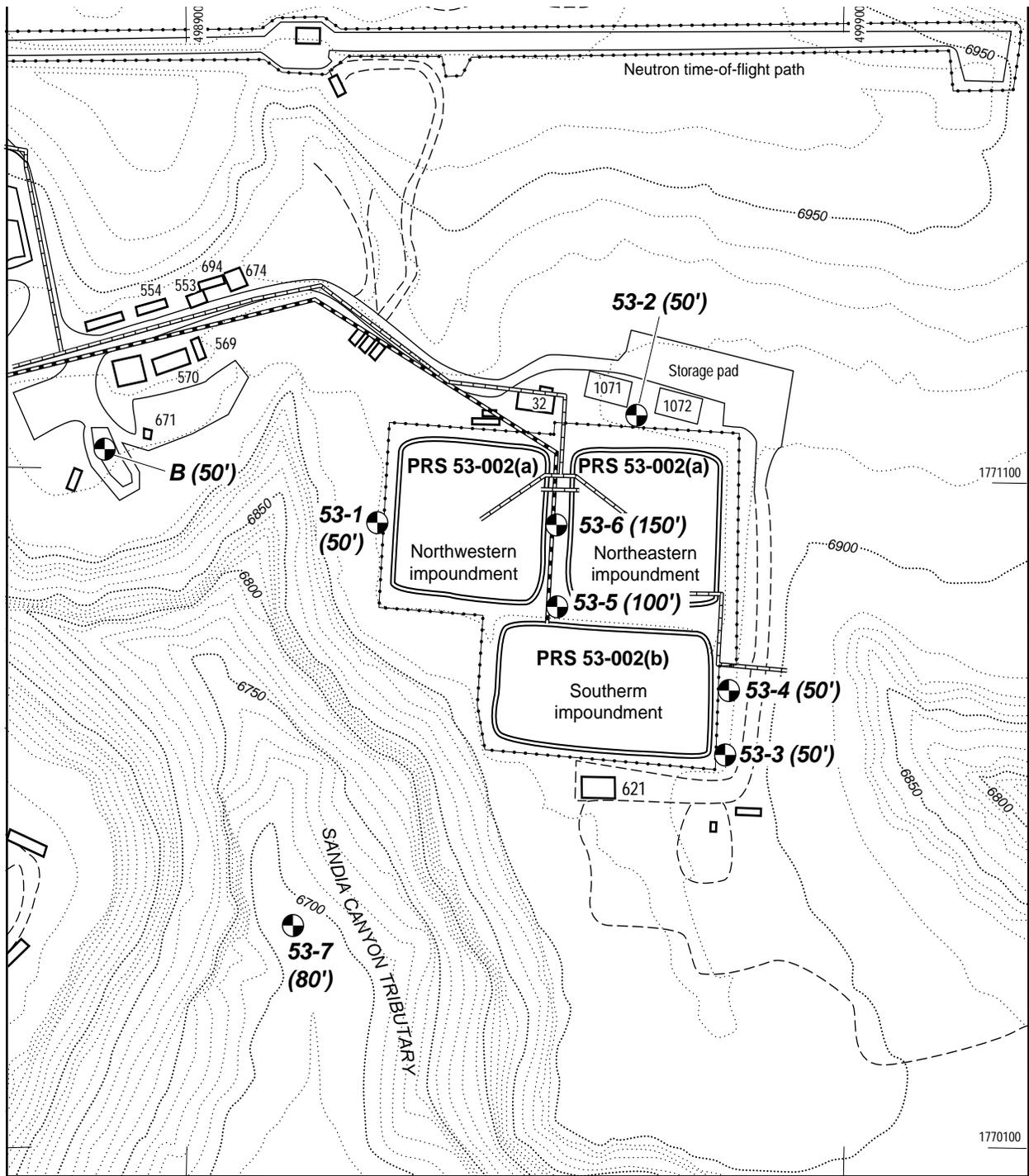
Figure 1   **Approximate locations of boreholes near the TA-53 surface impoundments.**

2.0 -   SITE DESCRIPTION

2.1   Stratigraphy and topography

TA-53 is located on Mesita de Los Alamos, which is bounded to the north by Los Alamos Canyon and to the south by Sandia Canyon (Figure 1). Site-specific geologic stratigraphic and structure maps of the surface impoundments at TA-53 are based on the LANL Site-Wide Geologic Model (Carey et al.1999, 66782); however, the general stratigraphy has been refined from the numerous boreholes around the site (Figure 2). Mesita de Los Alamos is comprised of a series of
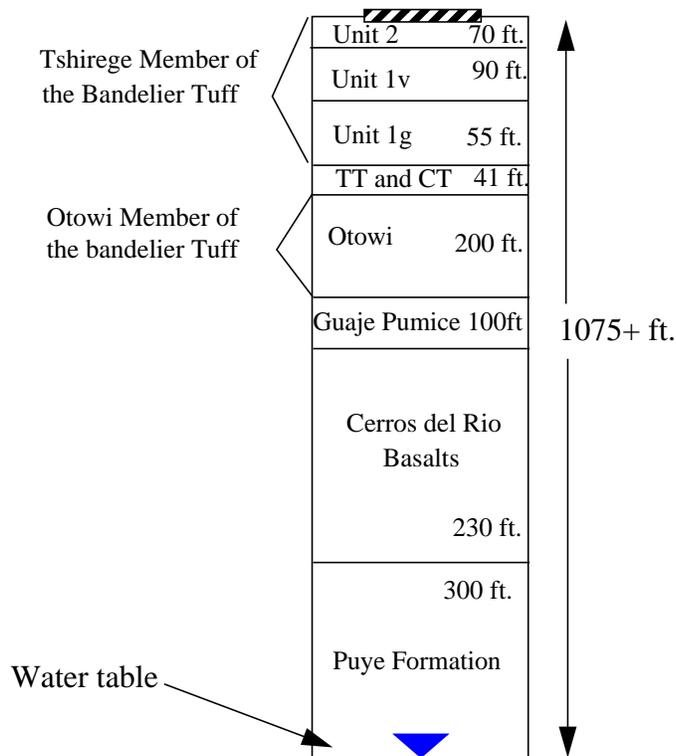
Figure 2   Simplified site stratigraphy.

unconsolidated sediments and volcanic rocks and is typical of a terrain produced by concurrent sedimentation and volcanism. The uppermost rocks that contain the impoundments are within Unit 2 of the Bandelier Tuff, a very porous material containing some fractures in the more welded sections. Between the Tshirege and Otowi members of the Bandelier Tuff are two thin units, the Tsankawi Pumice and the Cerro Toledo rhyolitic sediment. Below the Otowi unit, the Guaje Pumice, Cerros del Rio Basalts, and Puye Formation are less well characterized both with respect to physical parameters and spatial distribution [Broxton and Reneau, 1995; Gardner et al., 1986].

Fortunately, these units lie well below TA-53 and do not appear to play an active role in transport of tritium from the surface impoundments.

2.2   Surface Impoundments

The surface impoundments are located near the eastern boundary of TA-53.   PRSs 53-002(a) and (b) include three surface impoundments (Figure 1). PRS 53-002(a) is comprised of two northern surface impoundments and all associated drainages that were used to treat sanitary and radiological waste from TA-53 facilities. Each northern surface impoundment is 210 ft. in length by 210 ft. in width by 6 ft. deep and has a liquid storage capacity of approximately 1.6 million gal. To minimize the volume of water discharged to the outfall, two Aqua Aerobics^{TM} surface turbine aerators were used in each impoundment for aeration and enhanced evaporation. The northern surface impoundments at PRS 53-002(a) were installed in 1969 and began operating in the early 1970s. The impoundments were constructed by excavating into the tuff, adding a clay liner on top of the tuff, and spraying gunite (concrete) onto the crushed tuff berm. The initial design of the northern surface impoundments included an emergency overflow system that discharged into a south-lying drainage ditch. This overflow system consisted of an overflow pipe connecting the two surface impoundments and a discharge pipe that ran southward through the impoundment berm, terminating immediately after it emerged from the berm. The unlined ditch that accepted effluent discharges from 1970 to 1985 began near borehole 53-5 and headed S.SW. approximately in the direction of borehole 53-4 (Figure 1). An outfall structure with a blank weir plate was located at the junction of the discharge pipes [LANL, 1998, 58841.2; LANL, 1998, 59997.2]. Discharged water may have infiltrated into the host rock exposed within the unlined drainage ditch. Site records indicate that approximately 60 million gallons were discharged from 1970-85, however flow from the outfall regularly reached 1000 m down the small side canyon just to the east of borehole 53-4 [Rogers, 1998, 59169], resulting in an infiltrated volume that is certain to be well below the 60 million gallons of discharged effluent.

PRS 53-002(b), the southern surface impoundment, was constructed and began operating in 1985. It was installed by excavating into tuff, overlain with crushed tuff and sand, and then overlain with a Hypalon TM (rubber polymer) liner. The southern impoundment was constructed to treat both sanitary (1985-1989) and radiological (1985- 1995) waste from TA-53 facilities. In 1992, when Sanitary Wastewater System Consolidation (SWSC) became operational, all sanitary

waste was diverted from the northern surface impoundments [LANL, 1998, 58841.2; LANL, 1998, 59997.2].

Currently (2002), all three surface impoundment have been excavated down to the top of the tuff.

2.3   Tritium data - 1991 Boreholes

During 1991, boreholes were drilled adjacent to the surface impoundments to examine both moisture content and tritium concentration at depth [LANL 1992, 29413)]. Figure 3 shows
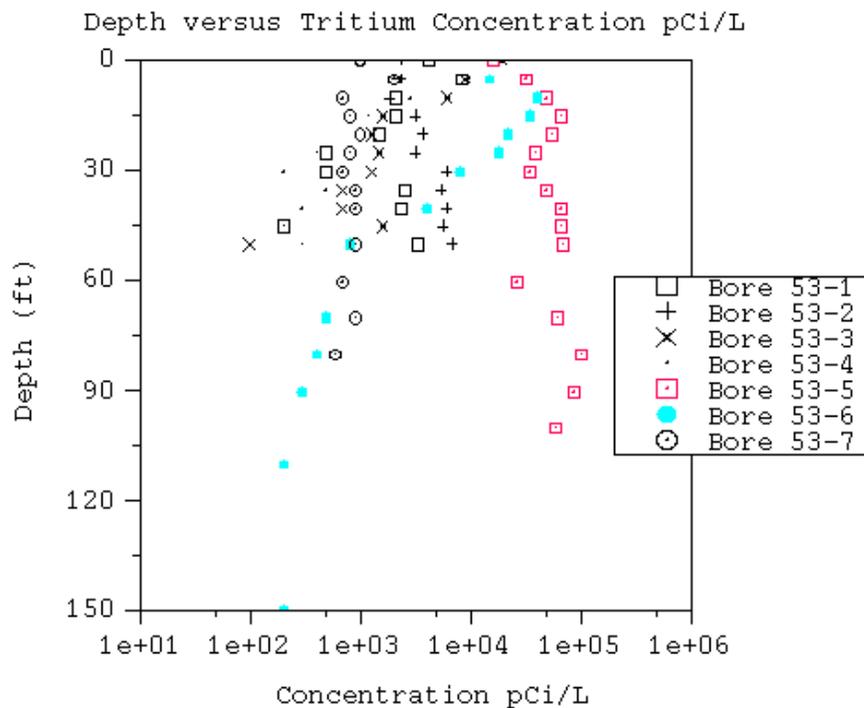
Figure 3 1991 Tritium data for boreholes shown in Figure 1.

the data from this set of boreholes. The only significant concentrations at depth were found in borehole 53-5, near the start of the unlined drainage ditch. We do not believe it is coincidental that the highest subsurface concentrations are very close to being directly beneath what is assumed to be the most likely subsurface tritium release point. However, the high saturations seen only in borehole 53-6 have very low associated tritium concentrations (100 pCi/L). Thus, the 1991 data suggest that the predominant mechanism for tritium transport is via the vapor-phase pathway, while some amount of relatively clean water has leaked into the subsurface near borehole 53-6. A possible source of non-tritiated water near borehole 53-6 is the pipeline that carried sewage to the lagoons.

3.0 - CONCEPTUAL MODEL

Tritium (radioactive hydrogen, half-life = 12.4 years.) exists in the environment as one of the hydrogen atoms attached to a water molecule. Being intimately bound to the water, tritium travels in the vapor phase as water vapor, not as a separate component [Knight 1996, 70152]. Water vapor in equilibrium with tritiated water will have the same concentration of tritium as the source water. However, at 100% humidity and 10 C, water vapor is only about 1% of the partial pressure of the soil atmosphere. Additionally, the density of air is approximately 1000 times lower than the density of liquid water leading to an effective Henry's constant for tritium of about 1x10-5 [(moles/L)air/(moles/L)water]. Within the porous media, equilibrium between water and water vapor is assumed (a well mixed model) because diffusional transport is slow relative to the rate of tritium exchange between pore water and pore gas [Smiles et al. 1995, 70153].   Coupling Henry's Law with equilibrium mixing and radioactive decay results in very different plume behavior than would be generated by compounds with low water solubility and no decay. First, the soil contains water that provides a very large sink for tritium. Second, the equilibrium condition requires that rapid transport in the vapor phase must first fill the available storage in the liquid phase before the plume can advance. Additionally, the short half-life of tritium causes a large reduction in the inventory. For example, if any inventory of tritium were contained for 100 years, then released, the remaining source would be 0.0036 times the initial source.

3.1   HYDROGEOLOGIC DATA

The major rock properties affecting the flow of water beneath the lagoons are the porosity and the permeability of the rock as it becomes wet [Fetter, 1999]. Saturated permeabilities, porosities, and saturations for the tuff units [Rogers and Gallaher, 1995] and the Guaje Pumice were measured on core samples. Estimated values for the porosity and saturation of the Puye Formation are used. No hydrologic property data were available for the basalts at the time this study was performed. Therefore, the porosity of a vesicular basalt located beneath Idaho National Engineering Laboratory is used as an analog [Bishop, 1991]. The saturation of the basalt is based on previous modeling results [Birdsell et al., 1999, 69792].   Fortunately, the model of the contaminant plume is relatively insensitive to the properties of the Puye Formation and Cerros del

Rio Basalts. Table 1 summarizes the hydrologic parameters used for all of the units in the unsaturated-zone flow and transport model.

Table 1: Hydrogeologic parameters used in the numerical model

Unit	Effective porosity	Saturated permeability m^2	Van Genuchten alpha and N (m^{-1}, unitless)
Tsh 2	0.479	7.48e-13	0.66, 2.09
Tsh 1 v	0.528	1.96e-13	0.44, 1.66
Tsh 1 g	0.509	3.68e-13	2.22, 1.59
Tsankawi Pumice	0.473	1.00e-12	1.52, 1.506
Cerro Toledo	0.473	8.82e-13	1.52, 1.506
Otowi Member	0.469	7.25e-13	0.66, 1.71
Guaje Pumice	0.667	1.53e-13	0.08, 4.03
Cerros del Rio basalt (matrix)	0.05	2.96e-13	0.1, 2.0
Puye Formation	0.35	4.73e-12	5, 2.68

3.2   TRANSPORT PROPERTIES

The modeling uses an estimate for the subsurface vapor-phase diffusion coefficient (D*). The estimated D* is based on the work of Vold [1996, 70155] and Vold and Eklund [1996, 70156] and is based on analysis of a large tritium plume at MDA G, located to the east on Mesita Del Buey. Vold determined an effective diffusion coefficient which incorporates near surface processes such as barometric pumping and evaporation of water vapor. The estimated tritium vapor diffusion coefficient (1.e-3 m2/s) is much larger than the best-fit diffusion coefficient (4.e-7 m2/s) for Trichloroethane at MDA L (Stauffer et al., 2002), therefore the model is run with a range of diffusion coefficients to determine which diffusion coefficient best fits the site data.

4.0 - TA-53 TRITIUM TRANSPORT MODEL

A site-specific numerical model that details the geology, waste geometry, and topography was developed to support decisions regarding placement of boreholes to better define nature and extent of tritium contamination below the TA-53 surface impoundments. The numerical site model is based on the conceptual models of unsaturated transport described in various reports [Turin and Rosenburg, 1996, 63559; Stauffer et al. 2000, 69794]. Calculations of mass and energy transport were performed using the LANL developed multiphase porous flow simulator, FEHM [Zyvoloski et al. 1997, 70147]. FEHM has been previously used for modeling tritium transport within the unsaturated zone of the Pajarito Plateau [LANL 2001].

Boreholes drilled in July 2002 will allow the numerical model to be checked and provide additional constraints on the possible behavior of subsurface transport. As new data become available, both the conceptual and numerical site models can be refined to better characterize the site in support of the final closure plan.

4.1   MODEL DOMAIN AND COMPUTATIONAL GRID

Site-specific data and general Pajarito Plateau hydrologic data were gathered and processed for use in the numerical analysis. A numerical grid was designed that captures the land surface to the water table and encompasses a lateral area centered on the lagoons (Figure 4). The grid was generated using GEOMESH grid generation software [Gable et al. 1995], 70148), and incorporates information from the site-wide geologic model [Carey et al.1999, 66782]. Previous tritium modeling at MDA G, TA54 [Vold (1996, 70155) and Vold and Eklund [1996, 70156]] provides an estimate for the mobility of tritium in the near surface. Simulated estimates of tritium migration incorporate the processes of diffusion, dispersion, and advection. Vapor-phase transport is included via an enhanced liquid phase diffusion coefficient [Smiles et al. 1995, 70153]. Simulated tritium concentrations in the subsurface constrained by the data will allow us to best place additional boreholes to better define nature and extent of tritium as well as to benchmark our

conceptual models of the site. The model domain extends vertically from the land surface to below
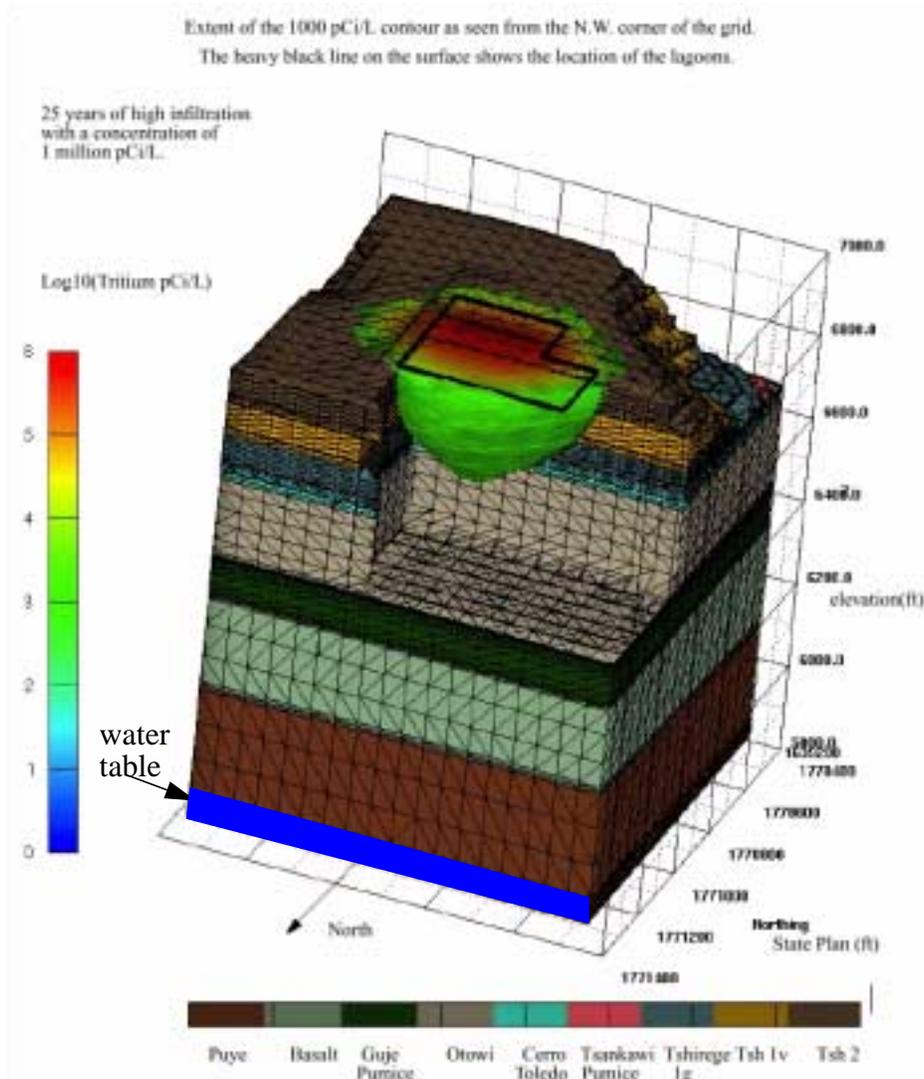

Figure 4   Model stratigraphy below the TA-53 lagoons. Simulation results are for a "Worst Case" scenario.

the water table and delineates a rectangular volume of over 43 million cubic meters. The

stratigraphic configuration used for the model (Figure 4) is derived from the LANL site-wide

geologic model [Carey et al., 1999, 66782.1]. Surfaces and interfaces are loaded into the LaGrit

grid generation software [Gable et al. 1995, 70148], and a computational grid is formulated that

maintains positive definite coupling coefficients at all volume interfaces. Furthermore, the

stratigraphic interfaces are given higher-resolution node spacing to allow for sharp gradients in

material properties. The final grid contains 23,025 nodes and 131,476 tetrahedral volume elements.

4.2   BOUNDARY AND INITIAL CONDITIONS

The bottom boundary of the domain is flat and lies below the water table. The presence of the water table within the model domain allows us to estimate travel times to this important horizon. No flow of water or vapor is permitted across the bottom boundary, and its temperature is held constant at $25^{o}C$, based on well bore measurements [Purtyman, 1995, 45344]. The atmosphere at the top of the model is held at a pressure of 0.078 MPa, and the temperature is fixed to the yearly average of $10^{o}C$ [Bowen, 1992, 12016]. The atmosphere is represented by fixing the concentration at zero in the nodes above the land surface. The vertical side boundaries of the domain are no flow with respect to both mass and heat. Background infiltration is low on the mesa tops of the Pajarito Plateau [Birdsell et al. 1999, 69792], and we initialize the simulations with 1 mm/yr infiltration below the root zone. Before running the contaminant transport simulations, a static air pressure field is established by running the model until pressures and temperatures reach equilibrium. This ensures that the transport simulations are not affected by transient behavior associated with establishing a static air pressure field. The steady-state initial condition has no tritium present and is meant to represent the mesa before release of contaminants.

4.3   Anthropogenic effects

We include several factors that result from the impact of humans on the site. First, the timing of outflow from the North Lagoons (PRS 53-002a) is explicitly simulated, with the initiation of effluent occurring from 1970 to 1985 in the more realistic case and from 1970 to 1995 in the "Worst Case" scenario. The concentrations in the effluent are estimated from site records and average 800,000 pCi/L [LANL, 1998, 58841.2; LANL, 1998, 59997.2]. Additionally, the impact of the unlined drainage ditch is explored by varying the amount of water allowed to infiltrate. Anthropogenic factors not considered in this report include the effects of the geotechnical and clay liners on the ability of tritium to leak and diffuse, and the effects of roads and excavation on the site hydrology.

5.0 - RESULTS

5.1   Worst Case Scenario

The simulation is done in two parts, the first 25 years represents the active leaking period at the site, and the next 75 years of simulation is meant to represent the combined effects of tritium source removal and return to background infiltration rates. The simulation shown in this report uses an infiltration flux of 2m/yr over an area of 10,000 ft2 (929 m2). This infiltration rate was chosen because it is comparable to rates inferred for wet sections of local canyons such as Los Alamos Canyon. Also, this infiltration rate is nearly 10x higher than in-situ estimates (23 cm/yr) based on measured saturations in borehole 53-6 [Rogers and Gallaher, 1995]. At 2 m/yr, the total volume of the simulated infiltration is 12.2 million gallons which is nearly 8 times the volume of an individual northern impoundment and 14% of the total estimated discharge at the site. The simulated source concentration (1 million pCi/L) was chosen to represent a high value from the near surface data set, and is slightly higher than the estimated average effluent concentration [LANL, 1998, 58841.2; LANL, 1998, 59997.2]. At 2 m/yr and 1 million pCi/L, the total injected tritium activity is 48.5 Ci which represents 18% of the total high estimate for tritium discharged from the impoundments.

Figure 5 shows a horizontal slice of the model overlain by an aerial photo of the site. The
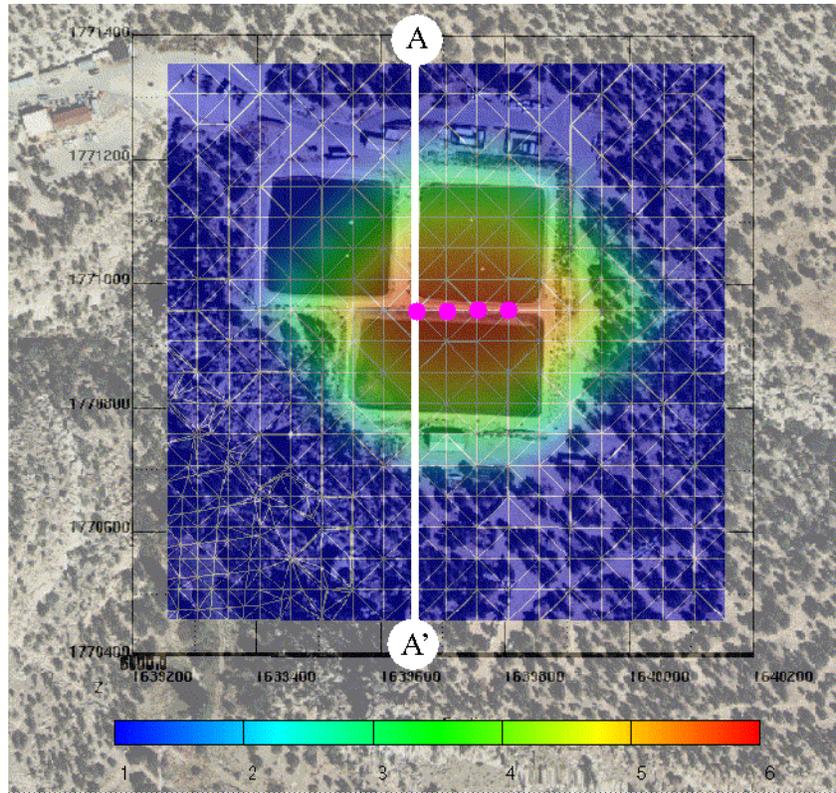

Figure 5   Log10(Concentration pCi/L)

injection nodes are shown as pink spots and were placed near the center of the domain near the

location of the unlined drainage ditch that carried effluent from the northern impoundments LANL,

1998, 58841.2]. By concentrating the injection in a relatively small area, the simulation produces

a deeper plume than applying an equivalent amount of water spread on any larger area.

   Figure 6, a vertical slice of the model along cross-section A-A' (Figure 2), shows that

during the first 25 years of the simulation the leading edge of the tritium plume migrates to a depth

of approximately 370. ft. (6,550 feet elevation) with concentrations of nearly 100,000 pCi/L at 220 ft. depth (6700 ft. elevation).
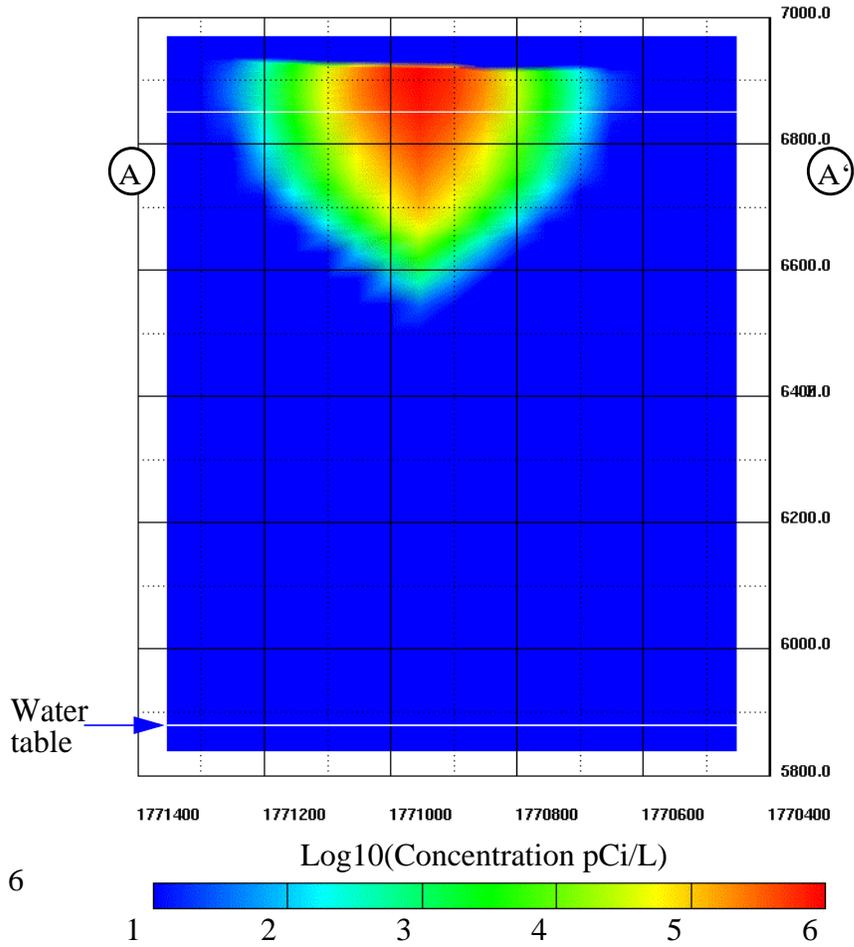

Figure 6

Log10(Concentration pCi/L)

Figure 7 shows the saturation values with depth below the zone of infiltration, generated in

elev(ft) vs saturation
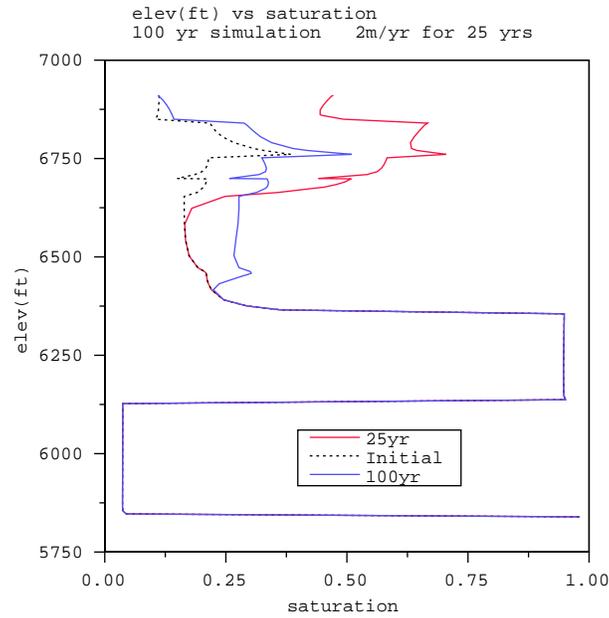100 yr simulation   2m/yr for 25 yrs

Figure 7

the simulation for the years 1970 (initial), 1995, and 2070. Data for borehole 53-5 (Figures 9 and 11) show that the highest subsurface concentrations at the site are not associated with greatly increased saturations. Thus, based on the available data, the "Worst Case" model is clearly overestimating the input of water, and therefore overestimating the total tritium activity in the plume.

In Figure 8 we show that after 75 years, the plume has continued to migrate to a depth of
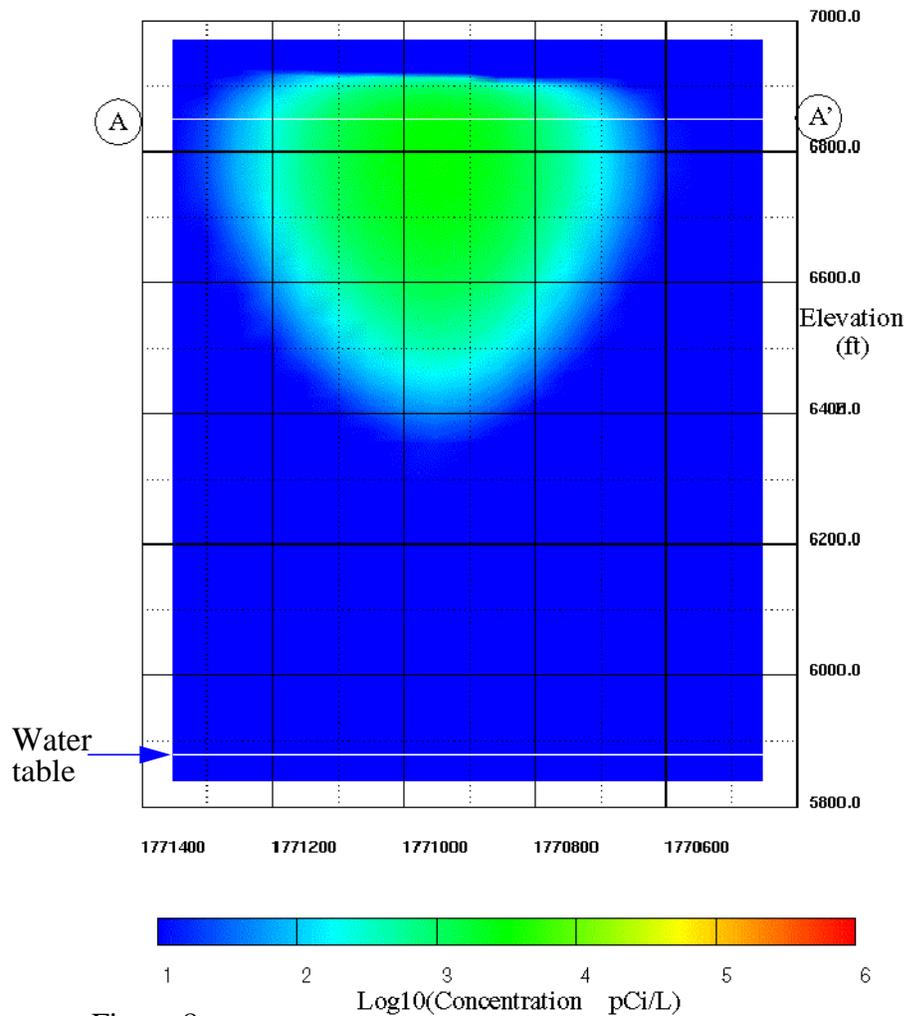

Figure 8

520 ft. (6,400 feet), however with the source removed, the processes of diffusion, dilution, and radioactive decay have reduced maximum concentrations in the plume to 4,000 pCi/L ($Log10(4000) = 3.6$). Thus simulated values for the year 2070 are well below the EPA guideline for tritium in drinking water of 20,000 pCi/L. The uncertain variables (infiltration rate, area, total volume, concentration) used in this simulation are conservative in nature and would represent a worse case scenario. Next, we present simulations that show the relative importance of advection and diffusion on the behavior of the tritium plume.

5.2 Model sensitivity: Advection and Diffusion

The simulations of this section examine the processes of diffusion and advection and their relative importance in the development of the subsurface tritium plume. By varying the amount of water allowed to infiltrate and the effective diffusion coefficient, a range of plume behavior can be

demonstrated. These more realistic (less conservative) simulations covers the years of 1970- 1995, during the first 15 years (1970- 1985) concentrations are fixed in the source region, while during the last 10 years (1985- 1995) concentration at the source is fixed at zero and the plume is allowed to begin relaxing to background values. For this series of simulations, the source concentration is fixed at the estimated average effluent concentration of 800,000 pCi/ [LANL, 1998, 58841.2].

First we present simulations with no infiltration that are designed to show how sensitive the model output is to the specified effective diffusion coefficient. Figure 9 shows several
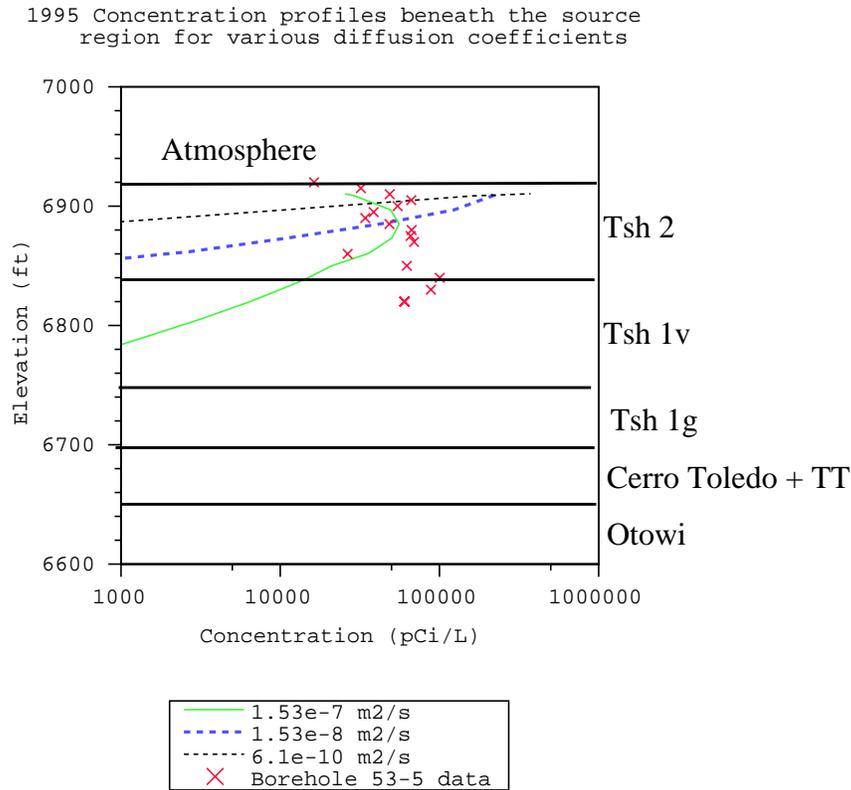

Figure 9

concentration versus depth profiles for different values of the effective diffusion coefficient. In the absence of infiltration, a high diffusion coefficient is needed to push the plume deep enough into the subsurface to approximate the data. However, even the highest diffusion coefficient is not able to match the deepest data. This set of simulations shows that although a higher diffusion coefficient can more quickly spread the source to depth, the high spreading rate leads to much lower peak concentrations in the simulated year 1995, 10 years after the source has been removed.

Figure 10 shows the effect of varying infiltration rate on the 25 year profile of the simulated

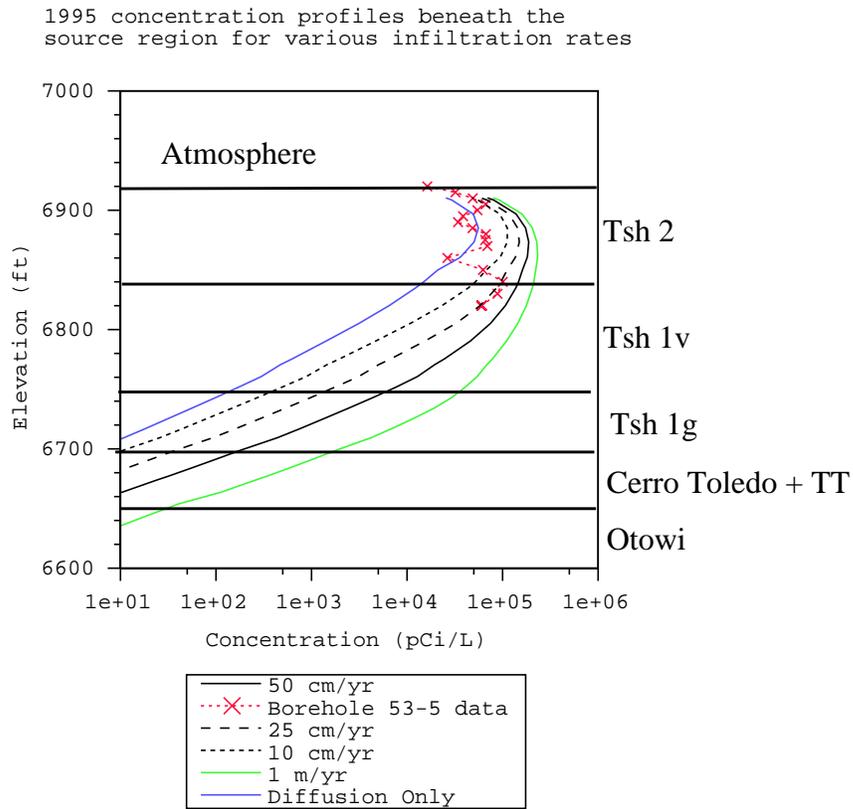1995 concentration profiles beneath the
source region for various infiltration rates

Figure 10

tritium plume. Figure 11 shows the corresponding saturation profiles. Saturations seen in borehole

1995 saturation profiles beneath the source
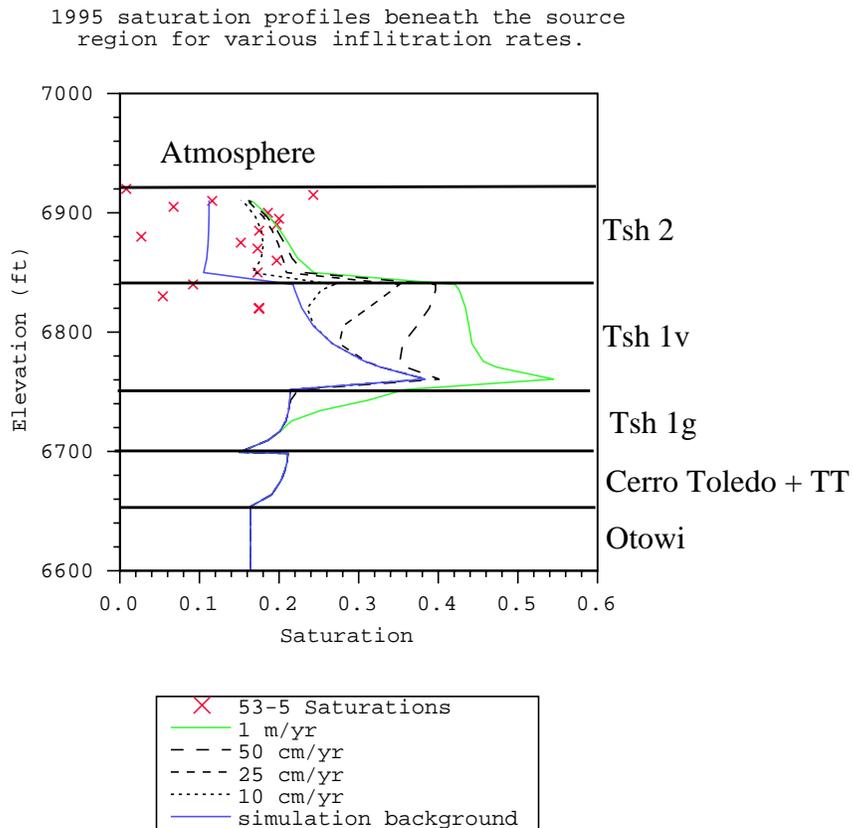region for various inflitration rates.

Figure 11

53-5 fall within the range of the simulated values and could result from heterogeneous material properties and/or heterogeneous flow.

Data from borehole 53-5 and 53-6 (Figure 12) show much different saturation values,

Saturation data from boreholes 53-5 and 53-6
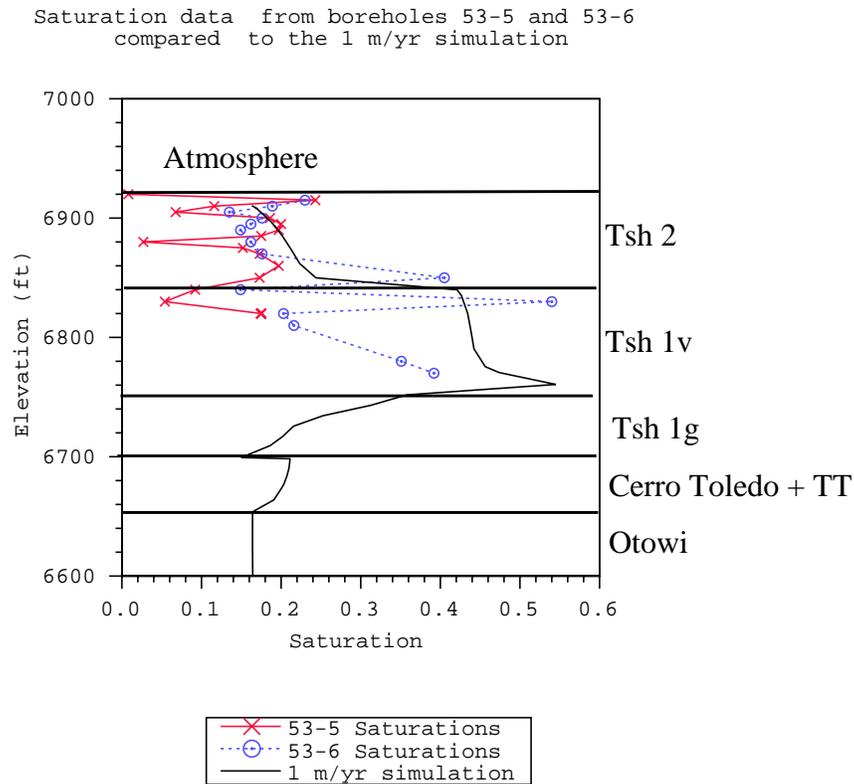compared to the 1 m/yr simulation

Figure 12

suggesting that borehole 53-6 has experienced more infiltration. The saturation profile from the 1 m/yr simulation is presented with the data to show that this amount of water is necessary to create saturations seen in borehole 53-6. The fact that the simulated peak saturation is lower in the profile is related to the timing of the infiltration and/or the location of hydrologic boundaries. A recent site visit revealed water leaking from an undetermined underground source near borehole 53-6 causing a manhole to fill with water and producing a seep in the N.W. lagoon. This observation, coupled with the modeling results (Figure 12) leads us to believe that the water encountered in 53-6 is from a more recent high flux leak. Given the concentrations measured in 53-6, the source of the leak could not have been the radioactive waste line. This leads us to conclude that a sanitary and/or water supply line is the cause of the observed leak and the elevated saturations seen in borehole 53-6. We recommend that the source of this leak be determined and removed, primarily because continued water input to the subsurface will increase transport of waterborne waste constituents from the near-surface, however leaking pipes are also a concern with respect to cost.

5.3   Conclusions

We conclude the TA-53 tritium plume analysis with a discussion of the most beneficial locations for additional boreholes to constrain our knowledge of the extent of the migration. Figure 10 shows that for a wide range of infiltration rates, the vertical extent of the plume (>1000 pCi/L) is unlikely to exceed 6700 ft. Furthermore, the near-surface data (LANL 1998) show that the highest tritium concentrations are found near the intersection of the three lagoons. For these two reasons we have suggested that a borehole be drilled to approximately 6620 ft. (300 ft. deep) at the location marked borehole 53-1-2002 on Figure 13. From our conceptual model of transport in the
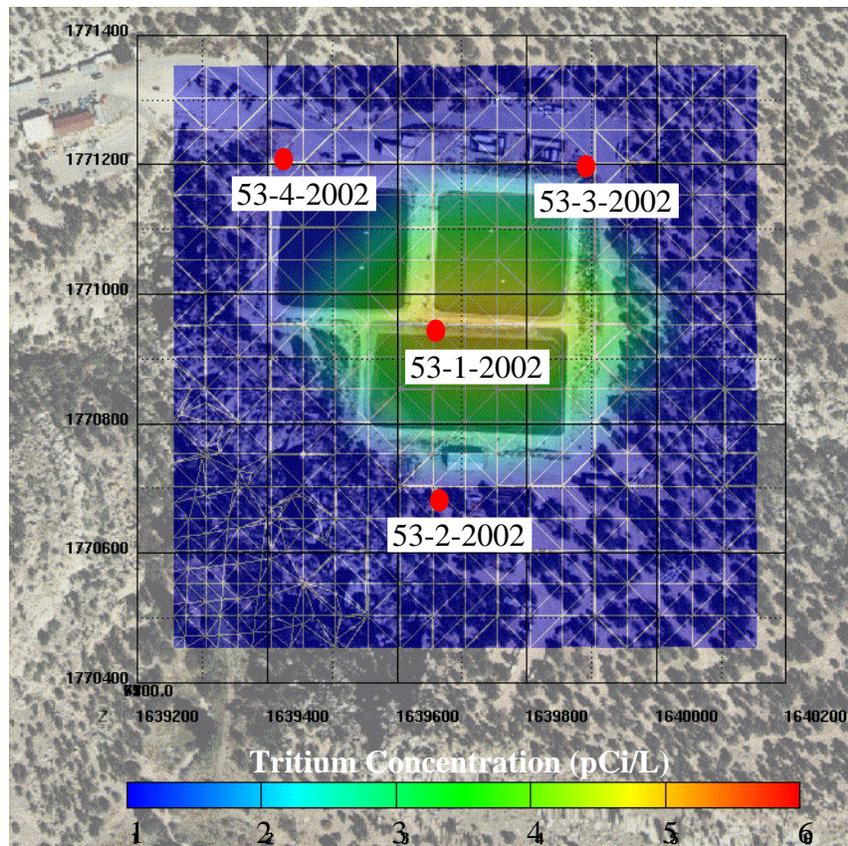
Figure 13 Approximate location of proposed boreholes for July 2002

system, this borehole should capture the bottom of the plume and provide information that was not present in borehole 53-5 due to the limited depth of drilling.

Three additional boreholes are suggested (Figure 13, 53-2-2002 through 53-4-2002) that will provided information to the 200 ft. depth (approximately 6720' elevation) at the estimated lateral boundaries of the plume. These holes are deep enough to intersect the Unit 1v/1g contact

and will be useful in determining if lateral transport has caused higher than expected concentrations at either the Unit 2/1v or 1v/1g boundaries.

5.4   Future work

Tritium samples from the July 2002 boreholes will allow the conceptual and numerical models to be verified. Further simulations which incorporate the new data could be useful in refining our site-wide conceptual model of increased tritium diffusion in the mesa-tops, however a more appropriate way to determine an in-situ tritium diffusion coefficient would be to run a controlled mesa-top release where the important parameters could be well controlled. This style of experiment could use concentrations at or below the 20,000 pCi/L drinking water limit and be done with minimum impact to the environment.

Although the effects of fractures were not examined in this report, the new data from drilling in July 2002 may require the conceptual and numerical models to be revised to include fractures in the appropriate geologic units.

Because of the diffusive nature of tritium and a relatively brief half-life, the current conceptual and numerical models show that there is virtually no future risk associated with the TA-53 tritium plume (Figure 8). Thus, in the real-world setting of limited funding for environmental analysis and remediation, the TA-53 tritium plume should be given a low priority rating when compared to sites such as TA-21, Ten-Site Canyon, TA 54 (Area G or L), or TA-16.

6.0 - ACKNOWLEDGEMENTS

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