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10 4 Evaluation of Deep Vadose Zone Contaminant Flux into
11 5 Groundwater: Approach and Case Study
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Abstract

For sites with a contaminant source located in the vadose zone, the nature and extent of groundwater contaminant plumes are a function of the contaminant flux from the vadose zone to groundwater. Especially for thick vadose zones, transport may be relatively slow making it difficult to directly measure contaminant flux. An integrated assessment approach, supported by site characterization and monitoring data, is presented to explain current vadose zone contaminant distributions and to estimate future contaminant flux to groundwater in support of remediation decisions. The U.S. Department of Energy Hanford Site (WA, USA) SX Tank Farm was used as a case study because of a large existing contaminant inventory in its deep vadose zone, the presence of a limited-extent groundwater plume, and the relatively large amount of available data for the site. A predictive quantitative analysis was applied to refine a baseline conceptual model through the completion of a series of targeted simulations. The analysis revealed that site recharge is the most important flux-controlling process for future contaminant flux. Tank leak characteristics and subsurface heterogeneities appear to have a limited effect on long-term contaminant flux into groundwater. The occurrence of the current technetium-99 groundwater plume was explained by taking into account a considerable historical water-line leak adjacent to one of the tanks. The analysis further indicates that the vast majority of technetium-99 is expected to migrate into the groundwater during the next century. The approach provides a template for use in evaluating contaminant flux to groundwater using existing site data and has elements that are relevant to other disposal sites with a thick vadose zone.

Keywords: Vadose zone; Numerical modeling; Contaminant transport

1. Introduction

Contamination in the deep vadose zone, defined by Dresel et al. (2011) as “the sediments or rock below the zone of practicable excavation and removal but above the water table” is a significant issue in most regions of the United States, although most of the research focus has recently been on the arid regions regarding contaminant transport (Seaman et al., 2007) or groundwater recharge (Walvoord and Scanlon, 2004; Rimón et al., 2007). A large number of contaminated deep vadose zones are at U.S. Department of Energy (DOE) sites, such as Yucca Mountain (Robinson et al., 2011), Idaho National Engineering and Environmental Laboratory (Nimmo et al., 2004), the Los Alamos National Laboratory (Birdsell et al., 2004) and the Hanford Site (Gee et al., 2007). Although radionuclides are of interest at many deep vadose zone sites, behavior of other contaminants such as metals (Hua et al., 2007), organics (Oostrom et al., 2003; 2007) and excess fertilizer (Onsoy et al., 2005) has been investigated in the unsaturated area below the root zone.

Due to its distance to the surface, a deep vadose zone may pose serious problems for characterization, monitoring, and remediation. Dresel et al. (2011) pointed out that remediation options for the deep vadose zone are often less developed than for the shallow zone or for saturated groundwater and that only a few have been implemented as full remedial actions. They also provided a review of available mass reduction, stabilization, and hydraulic control remedial techniques and identified soil desiccation of sediment layers, in combination with surface barriers, as one of the few potentially effective methods to reduce contaminant flux to groundwater. Recently, Truex et al. (2012) conducted a successful soil desiccation field implementation at the Hanford Site.

1 The nature and extent of groundwater contaminant plumes are often a
2 function of the contaminant flux from the vadose zone to the groundwater.
3 Contaminant movement is typically relatively slow through a deep vadose zone
4 and it is difficult to directly measure contaminant flux in these systems. Instead
5 of directly measuring vadose zone contaminant concentrations and fluxes,
6 theoretical conceptual models are often developed to improve the understanding
7 of deep vadose zone transport (Birdsell et al., 2004; Khaleel et al., 2007; Nimmo et
8 al., 2004; Oostrom et al., 2007; 2013; Robinson, 2011). These conceptual models
9 typically provide characterization information and discussions of plausible
10 subsurface flow and transport processes. Most of these models lack a
11 comprehensive approach to evaluate potentially controlling features and
12 processes, such as recharge, contaminant source characteristics, and lithography,
13 on contaminant flux to groundwater.

14 In this paper, an integrated approach is presented for estimating future
15 contaminant flux to groundwater at deep vadose zone sites. The approach is
16 initiated by gathering available data and information to develop a baseline
17 conceptual site model. An important part of this step is a consideration of the
18 lines of evidence provided by different types of data and any limitations for use
19 of the data. The baseline conceptual model is then refined considering the effects
20 of potential controlling features and processes and relevant boundary conditions
21 in the context of water and contaminant flux through the vadose zone into the
22 groundwater. This refinement takes place based on results from a quantitative
23 analysis using the STOMP flow and transport model (White and Oostrom, 2006)
24 to investigate the role of different driving forces for contaminant flux at the site.

25 The integrated approach was applied to the SX Tank Farm at the U.S.
26 Department of Energy Hanford Site (Fig. 1). As a result of previous tank leaks,
27 the vadose zone beneath this site has become contaminated and some of the

nonsorbing technetium-99 (Tc-99), originating from one of the tanks, has begun to migrate into the groundwater, creating a groundwater plume. However, a large fraction of the contaminant inventory is still present in the unsaturated vadose zone. A predictive analysis, supported by SX Tank Farm site characterization and monitoring data, was applied to estimate future contaminant flux to groundwater in support of remediation decisions for the vadose zone and groundwater. The current work builds on the SX Tank Farm analyses by Zhang et al. (2005) and Khaleel et al. (2007) by considering flow and transport in three dimensions and by comparing predicted trends with field observations. In this paper, an overview is first presented of the available data, followed by the development of a baseline conceptual model. A targeted quantitative analysis is then conducted to determine the flux controlling features and processes. Finally, the analysis is used to explain the existing Tc-99 plume and to refine the baseline conceptual model.

2. Integrated Assessment Method

The first step in the assessment is consideration of available site data and information to develop a baseline conceptual model. After that, potentially controlling features and processes are discussed that need to be considered in an evaluation of contaminant flux to groundwater. The nature and magnitude of how these elements impact contaminant flux are then assessed using a quantitative numerical modeling analysis. Finally, the simulation results are used to refine the conceptual model.

2.1. Site Data Evaluation

A schematic overview summarizing the spectrum of available data for the SX Tank Farm is presented in Fig. 2. Each data type provides input to some element of the baseline conceptual model and subsequent assessment of contaminant flux to groundwater.

2.1.1. Geology and Stratigraphy

Information on the geologic stratigraphy comes primarily from interpretation of 56 shallow and deep borings with geologic contact information. Four principal unconsolidated geologic units underlie the SX Tank Farm and surrounding area, including (in descending order): Holocene surficial deposits (primarily of anthropogenic origin – i.e. backfill), the Hanford Formation, the Cold Creek Unit, and the Ringold Formation. As shown in Table 1, each of these four major units may further be divided into subunits (Serne et al. 2008a,b). Fig. 3 shows the geologic cross-sections using these subunits for the transect AA' in Fig. 1, indicating relative minor changes in layer thickness across the SX Tank Farm. The average thickness of the various layers (Table 1) was compiled by Truex et al. (2015) using information from the available borehole data. Each of these layers can be matched to a soil class from a Hanford Site data base, with laboratory-measured hydraulic properties (Last et al., 2006a). The available borehole data was not considered to be detailed enough to allow for refinement of the stratigraphy depicted in Figure 3.

A mechanism for preferential vertical flow and transport at the Hanford Site is through clastic dikes (Murray et al., 2003). Clastic dikes are (sub)vertical structures that form within sedimentary sequences after deposition and cut across the original layers (Murray et al., 2003). They are common throughout the Hanford Site but a formal analysis has not been conducted for the SX Tank Farm.

1 The clastic dikes have a finer grain size distribution than the surrounding matrix,
2 resulting in a lower permeability but a larger moisture content. This combination
3 might lead to a larger effective vertical permeability under certain recharge or
4 leak conditions suggesting that these features might serve as a conduit for more
5 rapid movement of moisture and mobile contaminants to the water table
6 (Murray et al., 2003).

8 *2.1.2. Hydrology*

9 For the Hanford Site pre-construction period, the recharge rate was assumed
10 to be 3.5 mm/year, based on lysimeter studies, water balance computations, and
11 numerical simulations (Gee et al, 1992; 1994; Last et al., 2006a, Fayer and Keller,
12 2007; Fayer et al., 2010). In this period, vegetation was dominated by shrub-
13 steppe plant communities, composed of summer and winter annual grasses and
14 perennial grasses and shrubs. This desert vegetation is very efficient in utilizing
15 soil water due to its combination of shallow and deep-rooted plants (Gee et al.,
16 1992), resulting in low recharge rates (Gee et al., 1994). After removal of the
17 vegetation during tank construction and replacement of the top soil with sand
18 and gravel, the estimated recharge rate increased to 92 mm/year (Last et al., 2006;
19 Fayer and Keller, 2007). This value, based on primarily lysimeter measurements
20 and chloride mass balance studies, is approximately 55% of the average
21 precipitation of 173 mm/year (Last et al., 2006a). The 92 mm/year estimate is
22 within the range of recharge estimates reported by DePaolo et al. (2004) applying
23 hydrogen isotopes to tank farm subsurface cores. The two recharge rates are part
24 of official Hanford Site data sets (Last et al., 2006a; Fayer et al., 2010) for use in
25 subsurface flow and transport modeling (e.g., Carroll et al. 2012; Oostrom et al.,
26 2013).

1 High volume liquid effluent discharges to nearby ponds and cribs created a
2 groundwater mound beneath the SX Tank Farm, where water levels increased
3 considerably (Hartman, 2000) from its estimated pre-Hanford operations level
4 (Kipp and Mud, 1974). In 1988, production activities on the Hanford Site began
5 to close, resulting in dramatic decreases in wastewater disposal and subsequent
6 gradual decreases in water-table elevation. Local hydrograph data are available
7 to estimate the position of the water table over time. In addition to having a large
8 effect on the SX Tank Farm water table, the high volume discharge facilities also
9 need to be considered regarding potential influences on contaminant flux in the
10 SX Tank Farm vadose zone. Although the discharged volumes from adjacent
11 facilities were large, neutron data at locations just outside the SX Tank Farm
12 footprint showed no water content increases over time, indicating that lateral
13 moisture migration from other sites into the SX Tank Farm subsurface may not
14 have occurred. The groundwater flow direction at the site is shown in Fig. 1, as
15 suggested by Johnson and Chou (2002). Based on an arrival time analysis at
16 groundwater wells, they estimated the Darcy velocity to be approximately 10
17 m/year.

18 Water-line leaks potentially provide other sources of water to drive
19 contamination deeper into the vadose zone and eventually to the groundwater.
20 Typically, evidence of a leak was discovered and the leak was corrected.
21 Anderson (2001) reported at the SX Tank Farm, sustained slow leakage over a
22 long period of time from the indicated location in Fig. 1 (just south of the SX-115
23 Tank) may have occurred. No leak volume estimates exist for the SX Tank Farm
24 water lines, but “the total volume is considered to be substantial” (Anderson,
25 2001) due to the long duration of the leaks. Johnson and Chou (2002) report the
26 development over time of a mature cottonwood tree near the suspected leak,
27 which under normal Hanford Site precipitation conditions could not survive.

2.1.3. Tank Leaks

The SX Tank Farm consists of fifteen 3.8-million liter, high-level waste single-shell underground storage tanks. All of the tanks are believed to have released some liquid to the subsurface (Fig. 1), with the majority coming from the eight tanks indicated in Figure 1. Detailed contaminant release information is documented as part of Hanford Site operations and source-term studies. Shallow vertical boreholes and lateral boreholes beneath some of tanks were logged for gamma radiation to provide evidence for the location and extent of leaks. Neutron logging is also conducted in some vertical boreholes. Although these data have historically provided information about leaks, they are limited in quantifying the extent and exact location of the leak due to spacing of the wells. It is also difficult to track leak movement because of the mixture of gamma-emitting radionuclides and differences in decay rates. Interpretation of neutron logging is also limited due to the construction of some wells.

Best estimate tank leak information on total volume, leak duration, and major radionuclide inventories was compiled by Truex et al. (2015), based primarily on the analysis of leakage events by Johnson and Field (2010), and location/cause analyses conducted by Girardot and Harlow (2014). A summary of this compilation, including computed release rates and concentrations, is provided Table 2. The information shows that four tanks, SX-107, -108, -112, and -115 can be designated as major leakers, while the others may be qualified as minor leakers. The four major leakers account for >90% of the released activity for the nonsorbing contaminant Tc-99.

2.1.4. Vadose Zone and Groundwater Contaminant Plumes

Using Electrical Resistivity Tomography (ERT), Levitt and Henderson provided an estimate of contaminant distribution in three dimensions but the

quality of the results, showing a large co-mingled plume, was negatively affected by the presence of a metallic infrastructure and its use was deemed questionable for tank farms (Johnson and Wellman, 2013). Contaminant distribution information in the vadose zone is available from a series of characterization and direct push boreholes (Serne et al., 2008a,b). Two of the characterization boreholes indicate that the leading edge of mobile tank waste constituents beneath the SX-108 in 2008 was in the CCUz, with the majority of the remaining mass in H2 unit (Serne et al. 2008a). Fig. 4 shows examples of moisture and Tc-99 concentration data from Serne et al. 2008b (borehole 299-W23-234) near tank SX-108, a major leaker. Some mobile constituents such as nitrate and sodium followed a similar pattern to that of Tc-99, while others, such as chromium and iodine-129, appeared to be just slightly retarded, staying within the footprint of the Tc-99 vadose zone plume. Other less mobile contaminants, such as cesium-137 (Cs-137) are even more retarded and are confined well within the Tc-99 plume.

At the case study site, groundwater plumes are expressions of vadose zone contaminant flux. Regional and local groundwater monitoring provide well-documented information about groundwater plumes for Tc-99. This contaminant's arrival before 2000 is observed at W22-46 and W22-50, both located downgradient of the SX-115 tank (Fig. 1). At the monitoring wells downgradient of all other tanks, including the three tanks that are considered to be major leakers (SX-107, SX-108, and SX-112), no significant Tc-99 concentrations have been observed to date.

2.2. Baseline Conceptual Model

The baseline conceptual model and key questions related to controlling elements are shown in Fig. 5. Borehole evidence leads to a conceptual model showing that most of the contaminants originating from the major leakers are located in the lower part of the Hanford Formation and the upper part of the Cold Creek Units of the vadose zone, as illustrated in the Fig. 4 example. An evaluation is needed to determine whether the current subsurface contaminant distribution can be explained using available tank leak and hydrogeologic data, including recharge. Describing the current contaminant distribution is an important technical foundation to support subsequent predictions of future flux to groundwater and assessment of potential flux-mitigating remediation measures.

The influence of the tank leak characteristics (duration, volume, leak location) on transport needs to be considered as controlling features for flux to groundwater. Given the deep vadose zone below the SX tanks, the effects of leak characteristics on vertical migration need to be compared to the influence of additional infiltration due to the increased recharge after 1953.

Vertical heterogeneities, in the form of clastic dikes, have the potential to conduct water and contamination faster than the surrounding matrix (Murray et al., 2003). Although such features are not specifically identified in the tank farm subsurface, the flow and transport effects of potential clastic dikes need to be addressed.

The SX-115 tank leak was of a short duration with a high rate (Table 2). It needs to be determined whether such high intensity leaks can generate the driving force to transport contaminants downwards in a deep vadose zone system. Water-line leaks have occurred at the Hanford Site and a long-duration leak likely occurred just south of the SX-115 tank. Given the location of the

1 water-line leak, an analysis is needed to determine whether a leaking water line
2 could have been responsible for the observed groundwater plume.

3 Recharge may be a key element because it is recognized as a major driving
4 force for deep vadose zone transport (Gee et al., 2007) and the estimated recharge
5 at the case study site has likely increased by a factor of almost 30 (from 3.5 to 92
6 mm/year) compared to pre-Hanford conditions (Fayer and Keller 2007; Fayer et
7 al. 2010) due to construction of the tank farm in 1953. Assessment of the effect of
8 recharge on subsurface transport includes both the large areally extensive
9 recharge (e.g., tank-farm scale and larger) and the impact of localized recharge
10 features such as how the underground tanks impact the spatial distribution of
11 water infiltration.

12 The ERT data (Levitt and Henderson, 2009) suggest the formation of a large
13 co-mingled vadose zone plume. However, the presence of a large plume is
14 uncertain because the ERT-based plume is sensitive to the presence of metallic
15 infrastructure, like the tanks and piping at the site (Johnson and Wellman, 2013),
16 and the use of a limited number of electrodes in the field study by Levitt and
17 Henderson (2009). Data from sparse boreholes do not provide additional
18 evidence for co-mingled plumes. A quantitative approach is needed to
19 determine whether vadose zone plumes from leaks remain separated or if these
20 plumes are expected to consolidate into a broader plume.

21 The baseline conceptual model in Fig. 5 identifies only a single Tc-99
22 groundwater plume, downgradient of the SX-115 (Fig. 1). No additional Tc-99
23 groundwater plumes have yet been established originating from the other seven
24 leaking tanks, making the evaluation of contaminant transport for this particular
25 tank a special case. Given the minor differences in subsurface stratigraphy (Fig.
26 3) at the site, the accelerated migration from the tank to the groundwater below
27 the SX-115 may not be related to the subsurface geology. The available data

1 suggest four possible reasons for the development of the existing Tc-99
2 groundwater plume following accelerated vadose zone transport below the SX-
3 115: 1) lateral water migration from adjacent waste disposal sites, 2) preferential
4 downward flow and transport through clastic dikes, 3) the relatively large
5 estimated leakage rate of this tank (Table 2), and 4) a substantial water-line leak
6 south of the tank (Fig. 1).

7 In principle, it is possible that either one of these four features could have
8 quickly driven contaminants to the groundwater. Reasons 2, 3, and 4 are
9 addressed with the quantitative approach in Section 2.3. The potential effects of
10 lateral migration from adjacent sites were evaluated using additional large-scale
11 flow simulations. It has been well documented that very large water volumes
12 have been disposed in facilities surrounding the SX Tank Farm, notably the U-
13 Pond. It was determined (results not shown) that no reasonable scenario could be
14 devised allowing migration from these sites to the SX Tank Farm subsurface to
15 occur (Truex et al., 2015). The results of the scoping simulations are consistent
16 with borehole neutron data at locations between the tank farm and the high-
17 volume disposal sites.

18 19 **2.3. Quantitative Analysis**

20 A quantitative analysis of controlling vadose zone features and processes was
21 conducted with the water-transport mode of the STOMP simulator (White and
22 Oostrom, 2006). The applicable governing equations are the component mass-
23 conservation equation for water and the solute transport equation, which is
24 solved using a total-variation diminishing scheme. This mode of the simulator
25 has recently been used for a variety of unsaturated flow and transport problems
26 including contaminant transport in perched water (Oostrom et al., 2013), shear-

1 thinning fluid delivery (Oostrom et al., 2014), and vadose zone vapor
2 tomography (Mainhagu et al., 2014).

3 The effects of potentially controlling features and processes on contaminant
4 flux to groundwater were investigated in two three-dimensional (3D) models: A
5 local model of the SX-108 tank subsurface and a large-scale tank farm model
6 including all eight tanks that are confirmed leakers (Table 2). Applying the
7 computationally less demanding single-tank model is useful to determine the
8 controlling features and processes leading to the observation that for all tanks
9 but one, the leaked Tc-99 is still located in the vadose zone. Another justification
10 for this approach is that the borehole data supporting the baseline conceptual
11 model (Fig. 5) suggests that differences in subsurface layering at the SX Tank
12 Farm are relative small. If application of the characteristics of the largest leaker
13 (SX-108; Table 2) leads to a confirmation of the observed vadose zone
14 contamination location, the development of single the Tc-99 groundwater plume,
15 emanating from the SX-115 tank, can then be treated as a special case. The
16 groundwater plume migrates below several other tanks (Fig. 1) and therefore
17 simulations involving the SX-115 need to be conducted with the full model. The
18 large-scale tank model is needed to investigate potential co-mingling of plumes
19 emanating from the various tanks.

20 The single-tank SX-108 model (Fig. 6a) comprises a domain of 61 m in the x
21 direction (west to east), 61 m in the y direction (south to north) and 84 m in the z
22 direction (bottom to top). The 84 m vertical length ranges from 120 m elevation at
23 the bottom of the domain to 204 m elevation at the top surface. The domain was
24 discretized into $51 \times 51 \times 91$ grid cells for a total of 236,691 nodes. The large-scale
25 SX Tank Farm model (Fig. 6b) consists of three submodels representing the SX-
26 115-114-113, the SX-112-111-110, and the SX-109-108-107 west-to-east tank
27 sequences. The SX-112-111-110 and the SX-109-108-107 sequence submodels

1 represent an area of $160 \times 50 \times 84$ m and are discretized using $150 \times 46 \times 91$ grid
2 cells for a total of 627,900 nodes. The SX-115-114-113 sequence submodel
3 represents an area of $160 \times 81 \times 84$ m and is discretized using $150 \times 61 \times 91$ grid
4 cells for a total of 832,650 nodes. This submodel was larger in the y direction to
5 include the area south of the tank farm, ensuring that the well locations W23-15,
6 W22-80, W22-46 and W22-50 (Fig. 1) are part of the domain. In the x direction,
7 the large-scale model extends 30 m beyond the east fence of the tank farm. For
8 both the single-tank SX-108 and large-scale tank farm model, standard
9 refinement procedures were used to obtain the final discretized domains
10 yielding converged mass fluxes across the water table.

11 The two 3D numerical models were configured with eight geologic sediment
12 layers with locations and average thicknesses according to Last et al. (2009) and
13 Truex et al. (2015). The cross-sections of the domains are shown in Fig. 6 and
14 sediment property hydraulic values are listed in Table 1. The water retention
15 values for the van Genuchten (1980) saturation–water pressure constitutive
16 model are obtained from a Hanford Site subsurface data package (Last et al.,
17 2006a) and the water relative permeability is computed using the Mualem (1976)
18 relation. For all layers, similar longitudinal and transverse dispersivity values
19 (0.3 and 0.03 m, respectively) were used as in Zhang et al. (2005) for SX closure
20 assessment numerical simulations using the STOMP simulator.

21 Each simulation consisted of two components: 1) a pre-tank-farm
22 construction simulation with a recharge rate of 3.5 mm/year to obtain steady
23 state initial conditions for the transport simulation, and 2) a post-tank-farm
24 construction transport simulation from 1953 – 4000 with a recharge rate of 92
25 mm/year. For the area outside the tank farm (e.g., the area to the right of the
26 sloped backfill in Fig. 1), the recharge rate was kept at 3.5 mm/year after 1953.
27 The pre-tank-farm construction simulation started with unit hydraulic gradient

conditions from the water table to the surface and was allowed to run for 10,000 years to establish steady state conditions.

In the previous section it was explained that discharges from adjacent sites did not influence water contents below the tanks. For that reason, hydraulic gradient boundary conditions were assumed at the vertical boundaries. Based on local hydrograph data, the water table location was assumed to be at $z = 146$ m from 1953 to 1984, then linearly declining from $z = 146$ to 135 m from 1984 to 2012, and at $z = 135$ m after 2012. The groundwater Darcy flow velocity was assumed to be 10 m/year (Johnson and Chou, 2001) and was imposed through a constant flux (Neumann) boundary condition at the west side of the computational domains, in combination with a constant pressure (Dirichlet) boundary condition at the east side of the domains to indicate the water table position. Tc-99 was discharged into the unsaturated zone of the tank bottom over three periods with rates shown in Table 2. The leachate Tc-99 concentration was assumed to be the best estimate leaked inventory divided by the total leaked volume. The single-tank simulation using the described conditions is referred to as the Base Case. Besides the Base Case simulation, a large number of single-tank sensitivity simulations were conducted testing the influence of leak characteristics (location, duration, and volume), subsurface heterogeneity, hydraulic properties, and recharge. The simulations discussed in this paper are listed in Table 3.

3. Results and Discussion

3.1. Single-Tank Controlling Factors and Processes Simulations

Base Case water saturations and Tc-99 concentrations after the tank leaks (1967) and at 2015 are shown in Figs. 7 and 8, respectively. The Tc-99 concentrations in Fig. 7b show that the contaminant initially migrated primarily with the leaked fluid through advection. As can be seen at the right hand side of the tank in Fig. 7a, the moisture content front resulting from the increased recharge starting in 1953 is located just inside the H1a layer by 1967. In the period after the leaks ceased, the higher water saturations from the leaks rapidly dissipated in all directions due to capillary action and an associated reduction in relative permeability. The additional moisture resulting from the increased recharge imposed in 1953 started to mix with the contaminated water from the leaks shortly after 1968. Although some of the contaminant is transported downward through advection, molecular diffusion in all directions decreased the concentrations in the recharge water as it moved further downwards. Water migrating downward beyond the plume location contained increasingly less contaminant as it moved towards the water table. In the deep vadose zone at the site, the increased moisture content front resulting from the increased recharge imposed in 1953 migrates faster to the water table than the contaminant plume. The groundwater recharge rate changed from 3.5 to 92 mm/year between 2000 and 2007. This means that it took about 54 years for the new steady conditions to be established in the deep vadose zone. As a result, while water from the recharge change in 1953 is predicted to have reached the groundwater at the present time, the center of mass of the Tc-99 contamination is still within the vadose zone.

The delayed response of the contaminant plume compared to recharge is shown in Fig. 8. Predictions for the present time (2015) show that infiltrating water has reached the water table and that steady-state water flow conditions have been reached (Fig. 8a). However, despite the relatively high recharge rate,

the Tc-99 plume lags behind the water front and most of the contaminant is located in the lower part of the H2 layer and the Cold Creek Units. The predicted location is consistent with field observations for the SX Tank Farm (see an example in Fig. 4). Fig. 8b suggests that through 2015, no Tc-99 would have migrated to the water table under the Base Case assumptions (Table 3). The predicted plume in Fig. 8b has become largely asymmetric due to the larger effects of recharge on the part of the plume that is not under the SX-108 tank. In that zone, Tc-99 has migrated downwards faster than in the zone below the tank where the flow rates are smaller. In the latter zone, the plume is obviously more persistent because of the smaller advective transport component. It should be noted that the plume below the SX-108 originated from a leak at the southwest bottom end of the tank and not from a center-tank leak. Over time, contaminant mass has migrated towards the center as a result of primarily molecular diffusion and this mass is less affected by recharge than the outer part of the plume because of the influence of the waste tank on localized water flow conditions.

The predicted contaminant plume in 2100 has decreased in size and the majority of the remaining mass is located directly below the center of the tank and in the Ringold formation above the water table (Fig. 9). The plumes shown in Fig. 8b for 2015 and Fig. 9 for 2100 suggest that the majority of the Tc-99 inventory will migrate across the water table over that period.

Fig. 10 shows that the contaminant discharge behavior into groundwater for the simulations with a constant 92 mm/year surface recharge is quite similar. For all the simulations, except for the center leak simulation, the arrival times are between 2022 and 2027, and the maximum discharges occur between 2065 and 2070. The plot also shows that the vast majority of the released Tc-99 mass would be removed from the vadose zone by the year 2150. The simulation with the center leak shows a considerable delay of about 20 years in the maximum

1 discharge compared with the Base Case, where the leak was at the bottom edge
2 of the tank. The larger the distance of the leak from the tank edge, the less
3 influence recharge has on contaminant migration.

4 The similar results for all the other cases in Fig. 10 demonstrates the
5 important finding that the influence of leak volume, leak duration, heterogeneity,
6 and hydraulic properties on contaminant discharge into the groundwater is
7 limited and that none of these features are therefore controlling contaminant
8 transport at the site. In particular, the results show that for this deep vadose zone
9 configuration, leaked volumes as high as 384 m³ and rates larger than 16 m³/day
10 sustained for 12 days (Table 2), do not lead to enhanced transport towards. It
11 should be noted that for the simulation with the SX-115 leak characteristics, the
12 computed discharge values are smaller because of the smaller inventory
13 associated with this tank. The relative insensitivity of results to heterogeneities
14 includes the simulation with a substantial clastic dike, extending from the leak
15 location to the water table, even showing a slight delay compared to the Base
16 Case. In this simulation, the effective permeability of the surrounding matrix was
17 always larger than in the clastic dike, resulting in slower migration. This result is
18 consistent with the findings of Murray et al. (2003) who showed that for recharge
19 regimes similar to the SX Tank Farm, a clastic dike would not enhance
20 contamination transport in the vadose zone.

21 The insensitivity shown in Fig. 10 to leak characteristics, heterogeneity, and
22 hydraulic properties suggests that recharge likely has a large influence on
23 contaminant transport. A simple calculation considering peak arrival times and
24 average moisture contents illustrates that notion. A distance of approximately 75
25 m from the bottom of the tank to the water table, in combination with a peak
26 arrival time of about 100 years after the leaks, suggests an average contaminant
27 transport velocity of 0.75 m/year. Dividing the recharge rate of 92 mm/year by

the average vertical moisture content at steady-state conditions (0.126) yields a value of 0.73 m/year. Because of the similar numbers, the effect of recharge on contaminant transport was further evaluated by the recharge-related simulations listed in Table 3. The results of this analysis show that using half the recharge (46 mm/year), leads to a peak arrival time at 2162, which is about 200 years after the leaks. This time is about twice as large as the peak arrival time for the 92 mm/year simulation, supporting the notion that recharge drives the contaminant flux into groundwater. Increasing the recharge to the average precipitation (173 mm/year) leads to arrival of contaminants at the water table before 2000 and a peak arrival at 2022. Although this would potentially explain the plume originating from SX-115, it would have also caused groundwater plumes from all other tanks. Because the narrow current plume at the site (Fig. 1) comes from contaminants that leaked from SX-115, the use of a recharge rate larger than 92 mm/year is not realistic. Recognizing that in reality, precipitate occurs in distinct events, simulations have been conducted with six and one daily events per year. Although both of these assumptions are rather extreme, the purpose was to demonstrate that concentrating the average yearly precipitation into a small number of events does not make a difference in contaminant arrival at the water table in a deep vadose zone. Indeed, Fig. 11 shows that the discharge plots are identical to the Base Case. The finding is consistent with the rapid dampening that occurs in the vadose zone after precipitation events, as illustrated in Fig. 12. Already at 10 m below the surface, the average flow rate is, besides some minor fluctuations, similar to the Base Case. The offset in Fig. 12 is due to the way the precipitation is divided over the whole year. For the single event simulation, all recharge is added in the first day of each year. However, for the Base Case, the recharge is constant over the whole year. The results shown in Figs. 11 and 12

1 demonstrate that using a constant recharge rate at the surface boundary is
2 acceptable for this analysis.

3.2. SX Tank Farm Simulations

3 An example of Tc-99 concentrations in a cross-section through the SX-115, SX-
4 114, and SX-113 tanks at 2015 is shown in Fig. 13. The plot shows that, consistent
5 with the results for the single-tank SX-108 Base Case simulation, the majority of
6 the contaminant is in the H2 layer and the upper part of the Cold Creek Units.
7 The plume below SX-113 is migrating slower than for the SX-115 because the
8 contaminant was leaked in 1962 within a relatively small leak volume from the
9 tank bottom center. As a result, the initial plume after the leak was narrow and
10 was mostly protected from downward migration recharge water. Fig. 13
11 indicates that no co-mingling of the individual plumes occurred. The same lack
12 of co-mingling was observed throughout the tank farm.

13 Simulated fluxes to groundwater from all tanks with leak histories (Table 2)
14 are provided in Fig. 14a. The plot shows that for all tanks, most of the released
15 contaminant is predicted to migrate to the groundwater between approximately
16 2020 and 2150, with some tailing afterwards. For the tanks with the largest
17 released activity (SX-107, -108, -112, and -115), the maximum fluxes into
18 groundwater are obtained between 2065 and 2085. Of these four tanks, transport
19 of contaminants emanating from the SX-107 is the slowest because the leak
20 location was at the bottom center. Although the absolute flux into groundwater
21 differs for the four major leakers, the general discharge behavior for these tanks
22 is quite similar, showing that the released activity is also not a controlling factor
23 for contaminant arrival at the water table.

24 Fig. 14b shows that the largest predicted well concentrations are for W22-39
25 and W22-49, which are downgradient of the SX-108 and SX-112 tank, respectively

(Fig. 1). Both these tanks are considered to have been major leakers (Table 2), explaining the relatively high downgradient contaminant concentrations in the future. For both the W22-39 and W22-49 observation wells, the predicted concentration behavior shown in Fig. 14b is consistent with field observations to date, showing no Tc-99 arrival. However, the simulated results for the W22-46 and W22-50 wells do not correspond with measured concentrations. The predicted arrival times for these two wells are considerably later than indicated by field observations (see Fig. 16 for the measured concentrations). Because the Tc-99 arriving at these wells is likely originating from the SX-115, it can be concluded that simulated vadose zone transport is not fast enough below this tank. Of the total travel time from leak location to these observation well, the vast majority is related to migration through the vadose zone because the estimated transport time in groundwater from the projected center of the SX-115 tank to these two wells is less than 4 years.

The simulations for the SX-108 tank have shown that reasonable variations in leak characteristics, heterogeneity, and hydraulic properties do not result in enhanced vadose zone transport that would explain the development of the observed plume downstream of the SX-115 tank. Only considerable increases in recharge rate would accomplish that result but a much larger plume would have been expected under these conditions because the increase would have affected all tanks, not just the SX-115. Based on the results shown in Section 3.1, it can be concluded that for accelerated transport to occur below the SX-115, another controlling feature, very specific for this tank, has to be responsible. It was suggested by, among others, Johnson and Chou (2002) that a substantial water-line leak, located about 15 m south of the SX-115 tank, could have caused an accelerated contaminant migration to the water table and subsequent groundwater transport to the W22-46 and W22-50 observation wells.

1 To investigate this possibility, a series of simulations were conducted to
2 investigate the water-line leak rate and duration that would be necessary to
3 reduce the predicted water table contaminant arrival times to be consistent with
4 the well observations. The results of these simulations (Fig. 15) showed that an
5 order of magnitude leak rate of 1000 L/day over a 40 year period (1961-2001) was
6 needed to reasonably match field data and numerically obtained arrival times.
7 Rates of this magnitude are relatively small and could have persisted for many
8 years before detection in 2001 (Anderson, 2001). Rates larger than 1000 L/day
9 showed converging results (Fig. 15), with a water table arrival time
10 approximately 30 years sooner than for the case without a water line leak.

11 The effect of the water-line leak, using the 1000 L/day rate and a leak duration
12 from 1961-2001, on mass discharge from the SX-115, SX-114, and SX-113 tanks
13 into groundwater and downgradient well concentrations are presented in Fig. 16.
14 Fig. 16a shows that the influence of the water-line leak on contaminant fluxes to
15 groundwater is limited to only the SX-115 tank. The waterline leak also does not
16 affect the groundwater mass flux originating from SX-112 (a major leaker), which
17 is located north of the SX-115. The changes in groundwater flux behavior due to
18 the water-line leak are resulting in earlier contaminant arrival times at the
19 downgradient wells W22-46 and W22-50 (Fig. 16b and 16c). For these wells, the
20 water-line leak caused considerably earlier contaminant arrival times that are
21 more consistent with the field observations. The comparison with the measured
22 concentrations in Fig. 16b (and the zoomed-in version Fig. 16c) shows that this
23 analysis helps in improving the understanding of trends and general behavior.
24 The site data, although fairly detailed for a typical contaminated site, cannot be
25 used to directly match the detailed concentration. The results shown in Fig. 16
26 demonstrate that a long-duration water-line leak, with the characteristics used in

1 this simulation, provide a reasonable explanation for the currently obtained
2 single Tc-99 groundwater plume. Fig. 16 suggests that also for this scenario, the
3 vast majority of the Tc-99 is still located in the vadose zone and that increasing
4 downgradient well concentrations may be expected for at least another 70-80
5 years before peak concentrations are obtained at the individual wells.
6

7 **4. Refined Conceptual Model and Conclusions**

8 This paper provides an analysis of future contaminant flux to groundwater,
9 using the Hanford Site SX Tank Farm as a case study site, integrating available
10 data, knowledge of vadose zone flow and transport processes, and predictive
11 analysis. The approach and results provide a template for use in evaluating
12 contaminant flux to groundwater and the use of site data to support this
13 evaluation. The SX Tank Farm was selected as a case study because of the
14 existing contaminant inventory in the vadose zone, presence of a limited-extent
15 groundwater plume, and the relatively large amount and variety of data
16 available for the site. Although the SX Tank Farm case study is most
17 representative of conditions at the Hanford tank farm sites, the study has
18 elements that are also relevant to other types of deep vadose zone disposal sites.

19 Based on the evaluation of the controlling features and processes for
20 contaminant flux to groundwater, a number of conclusions were reached,
21 providing the technical foundation for supporting estimates of contaminant flux
22 at the site. The major conclusions that are used to arrive at the refined conceptual
23 model shown in Fig. 17 are:

- 24 • The imposed recharge increase in 1953, from an estimated 3.5 mm/yr to about
25 92 mm/yr, resulted in a modified water flux into the groundwater about 50

1 years after the surface change occurred. The current flow conditions in the
2 vadose zone are at steady-state with respect to the increased recharge rate.

- 3 • Unsaturated flow properties and the increased relative importance of
4 molecular contaminant diffusion delay the contaminant flux compared to the
5 water flux. For all tanks except the SX-115, Tc-99 is predicted to arrive at the
6 groundwater shortly after 2020 with peak concentrations occurring at around
7 2070.
- 8 • The influence of leak volume, leak duration, heterogeneity, and hydraulic
9 properties on contaminant discharge into groundwater is limited and none
10 these features are therefore controlling contaminant transport at the site.
- 11 • Preferential flow through clastic dikes does not occur during leakage and
12 under the imposed recharge regimes.
- 13 • While contaminants released from the tanks were initially advectively
14 transported due to the leaks, vadose zone processes quickly dissipated these
15 conditions and the recharge rate became the controlling factor for water flux
16 toward the groundwater. The contaminant plumes from all tanks, except for
17 the SX-115, would be expected to develop based on recharge-driven flux.
- 18 • The Tc-99 plume at the south end of the SX Tank Farms cannot be reasonably
19 explained without imposing a localized water-line leak near the SX-115 tank.
20 In the presence of such a leak, localized flow conditions can be created that
21 intersect vadose zone contamination and accelerate transport towards the
22 groundwater.
- 23 • Because leak volumes are relatively small and recharge controls the water
24 flux through the deep vadose zone, contaminant plumes from individual
25 tanks are not expected to co-mingle significantly in the vadose zone.

26

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References

- Anderson, F. J. 2001. *Engineering Report, Single-Shell Tank Farms, Interim Measures to Limit Infiltration Through the Vadose Zone*. RPP-5002, Rev. 1. CH2M HILL Hanford Group, Inc., Richland, WA.
- Birdsell, K.H., B.D. Newman, D.E. Broxton, and B.A. Robinson. 2004. Conceptual models of vadose zone flow and transport beneath the Pajarito Plateau, Los Alamos, New Mexico. *Vadose Zone J.* 4: 620-636.
- Carroll, K.C, M. Oostrom, M.J. Truex, V.J. Rohay, and M.L. Brusseau. 2012. Assessing performance and closure for soil vapor extraction: Integrating vapor discharge and impact to groundwater quality. *J. Contam. Hydrol.* 128: 71-82.
- Corbin, R.A., B.C. Simpson, M.J. Anderson, W.F. Danielson, III, J.G. Field, T.E. Jones, and C.T. Kincaid. 2005. *Hanford Soil Inventory Model, Rev. 1*. RPP-26744, Rev. 0, CH2MHill Hanford Group, Inc., Richland, Washington.
- DePaolo, D.J., M.E. Conrad, K. Maher, and G.W. Gee. 2004. Evaporation effects on oxygen and hydrogen isotopes in deep vadose zone pore fluids at Hanford, WA. *Vadose Zone J.* 3:220-232.
- Dresel, P.E., D.M. Wellman, K.J. Cantrell, and M.J. Truex. 2011. Review: Technical and policy challenges in deep vadose zone remediation of metals and radionuclides. *Environ. Sci. Technol.* 45:4207–4216, [dx.doi.org/10.1021/es101211t](https://doi.org/10.1021/es101211t).
- Fayer, M.J., A.L. Ward, and V.L. Freedman. 2010. Technical Basis for Evaluating Barriers to Protect Groundwater from Deep Vadose Zone Contamination. PNNL-18661, Pacific Northwest National Laboratory, Richland, Washington.

- 1
2
3
4
5
6 1
7 2 Fayer, M.J., and J.M. Keller. 2007. Recharge Data Package for Hanford Single-
8 3 Shell Tank Waste Management Areas. PNNL-16688, Pacific Northwest National
9 4 Laboratory, Richland, WA.
10 5
11
12 6 Gee, G.W., M. Oostrom, M. D. Freshley, M. L. Rockhold and J. M. Zachara. 2007.
13 7 Hanford Site Vadose-Zone Studies: Overview. Vadose Zone J. 6: 899-905.
14 8
15 9
16 10 Gee, G.W., M.J. Fayer, M.L. Rockhold, and M.D. Campbell. 1992. Variations in
17 11 recharge at the Hanford Site. Northwest Science 66:237-250.
18 12
19 13
20 14 Gee, G.W., P.J. Wierenga, B.J. Andraski, M.H. Young, M.J. Fayer, and M.L.
21 15 Rockhold. 1994. Variations in water balance and recharge potential at three
22 16 western desert sites. Soil Sci. Soc. Am. J. 58:63-72.
23 17
24 18
25 19
26 20 Girardot, C.L. and D. G. Harlow. 2014. Hanford Single-Shell Tank Leak Causes
27 21 and Locations - 241-SX Farm. RPP-RPT-54910, Rev. 0. Washington River
28 22 Protection Solutions, Richland, Washington.
29 23
30 24
31 25
32 26 Hartman, M.J. 2000. Hanford Site Groundwater Monitoring: Setting, Sources,
33 27 and Methods. PNNL-13080, Pacific Northwest National Laboratory, Richland,
34 28 Washington.
35 29
36 30
37 31
38 32 Hua, B., B.L. Deng, E.C. Thornton, J. Yang, and J.E. Amonette. 2007.
39 33 Incorporation of chromate into calcium carbonate structure during
40 34 coprecipitation. Water, Air, Soil Pollution. 179: 381-390.
41 35
42 36
43 37
44 38 Johnson, V.G. and C.J. Chou. 2002. *Groundwater Quality Assessment Report for*
45 39 *Waste Management Area S-SX (April 2000 through December 2001)*. PNNL-13801,
46 40 Pacific Northwest National Laboratory, Richland, Washington.
47 41
48 42
49 43
50 44 Johnson, M.E. and J.G. Field. 2010. *Hanford SX-Farm Leak Assessments Report*.
51 45 RPP-ENV-39658, Rev. 0. Washington River Protection Solutions, Richland,
52 46 Washington.
53 47
54 48
55 49
56 50
57 51
58 52
59 53
60 54
61 55
62 56
63 57
64 58
65 59

- 1 Kipp, K. L. and R. D. Mudd. 1974. *Selected Water Table Contour Maps and Well*
2 *Hydrographs for the Hanford Reservation, 1944-1973*. BNWL-B-360. Battelle Pacific
3 Northwest Laboratories, Richland, Washington.
- 4
- 5 Last, G.V., E.J. Freeman, K.J. Cantrell, M.J. Fayer, G.W. Gee, W.E. Nichols, B.N.
6 Bjornstad and D.G. Horton. 2006a. *Vadose Zone Hydrogeology Data Package for the*
7 *2004 Composite Analysis*. PNNL-14702 Rev. 1, Pacific Northwest National
8 Laboratory, Richland, Washington.
- 9
- 10 Last, G.V., W.E. Nichols and C.T. Kincaid. 2006b. *Geographic and Operational Site*
11 *Parameters List (GOSPL) for Hanford Assessments*. Richland, Washington, PNNL-
12 14725, Pacific Northwest National Laboratory.
- 13
- 14 Last, G.V., P.D. Thorne, J.A. Horner, K.R. Parker, B.N. Bjornstad, D.C. Lanigan
15 and B.A. Williams. 2009. *Hydrogeology of the Hanford Site Central Plateau - A Status*
16 *Report for the 200 West Area*. PNNL – 17913, Rev. 1, Richland, Washington, Pacific
17 Northwest National Laboratory.
- 18
- 19 Levitt, M. and C Henderson. 2009. Surface Geophysical Exploration of the SX
20 Tank Farm at the Hanford Site. RPP-RPT-42513, Rev. 0, Washington River
21 Protection Solutions LLC, Richland, Washington.
- 22
- 23 Mainhagu, J., C. Morrison, M. J. Truex, M. Oostrom and M.L. Brusseau. 2014.
24 Vapor tomography for vadose-zone source characterization: Flow-cell
25 experiments. J. Contam. Hydrology 167: 32–43,
26 <http://dx.doi.org/10.1016/j.jconhyd.2014.07.007>.
- 27
- 28 Mualem, Y. 1976. A new model for predicting the hydraulic conductivity of
29 unsaturated porous media. Water Resources Research, 12:513-522.
- 30
- 31 Murray, C., A. Ward, and J. Wilson. 2003. Influence of classic dikes on vertical
32 migration of contaminants in the vadose zone at Hanford. PNNL-14224. Pacific
33 Northwest National Laboratory, Richland, WA.
- 34
- 35 Nimmo, J.R., Rousseau, J.P., Perkins, K.S., Stollenwerk, K.G., Blunn, P.D.,
36 Bartholomay, R.C., Knobel, L.L., 2004. Hydraulic and geochemical framework of
37 the Idaho National Engineering and Environmental Laboratory vadose zone.
38 Vadose Zone J. 3: 6-34.
- 39

- 1 Onsoy, Y.S., T. Harter, T.R. Ginn, and W.R. Horwath. 2005. Spatial variability
2 and transport of nitrate in a deep alluvial vadose zone. *Vadose Zone J.* 4: 41-51.
- 3
- 4 Oostrom, M. and R.J. Lenhard. 2003. Carbon tetrachloride flow behavior in
5 unsaturated Hanford caliche material: An investigation of residual NAPL.
6 *Vadose Zone J.* 2: 25-33.
- 7
- 8 Oostrom, M., M.L. Rockhold, P.D. Thorne, G.V. Last, M.J. Truex, and V.J. Rohay.
9 2007. Carbon tetrachloride flow and transport in the subsurface of the 216-Z-9
10 trench at the Hanford Site: Multifluid flow modeling and conceptual model
11 update. *Vadose Zone J.* 7: 971-984.
- 12
- 13 Oostrom, M., M.J. Truex, K.C. Carroll, and G.B. Chronister. 2013. Perched-water
14 analysis related to deep vadose zone contaminant transport and impact to
15 groundwater. *J. of Hydrology* 505: 228–239, doi:/10.1016/j.jhydrol.2013.10.001.
- 16
- 17 Oostrom, M. M.J. Truex, V.R. Vermeul, L. Zhong, G.D. Tartakovsky, and T.W.
18 Wietsma. 2014. Remedial amendment delivery near the water table using shear
19 thinning fluids: Experiments and numerical simulations. *Env. Processes*. doi:
20 10.1007/s40710-014-0031-9.
- 21
- 22 Polmann, D.J. 1990. Application of Stochastic Models to Transient Flow and
23 Transport in Heterogeneous Unsaturated Soils. Ph.D. Thesis, Massachusetts
24 Institute of Technology, Cambridge.
- 25
- 26 Rimon, Y., Ofwe, D., Nativ, R. and S. Geyer. 2007. Water percolation through the
27 deep vadose zone and groundwater recharge: Preliminary results based on a
28 new vadose zone monitoring system. *Water Resources Research* 43: W05402.
29 doi: 10.1029/2006WR004855.
- 30
- 31 Robinson, B.A., Houseworth, J.E., Chu, S., 2011. Radionuclide transport in the
32 unsaturated zone at Yucca Mountain, NV. *Vadose Zone J.*
33 doi.10.2136/vzj2011.0133.
- 34
- 35 Seaman, J.C., B.B. Looney, and M.K. Harris. 2007. Research in support of
36 remediation activities at the Savannah River Site. *Vadose Zone J.* 6: 316-326.
- 37
- 38 Serne, R.J., G.V. Last, H.T. Schaef, D.C. Lanigan, C.W. Lindenmeier, C.C.
39 Ainsworth, R.E. Clayton, V.L. Legore, M.J O'Hara, C.F. Brown, R.D. Orr, I.V.
40 Kutnyakov, T.C. Wilson, K.B. Wagon, B.A. Williams, and D.S. Burke. 2008a.

1 *Characterization of Vadose Zone Sediment: Borehole 41-09-39 in the S-SX Waste*
2 *Management Area.* PNNL-13757-3, Pacific Northwest National Laboratory,
3 Richland, WA.

4

5 Serne R.J., G.V. Last, H.T. Schaef, D.C. Lanigan, C.W. Lindenmeier, C.C.
6 Ainsworth, R.E. Clayton, V.L. Legore, M.J. O'Hara, C.F. Brown, R.D. Orr, I.V.
7 Kutnyakov, T.C. Wilson, K.B. Wagnon, B.A. Williams, and D.S. Burke.
8 2008b. *Characterization of Vadose Zone Sediment: Slant Borehole SX-108 in the S-SX*
9 *Waste Management Area.* PNNL 13757-4 Rev. 1, Pacific Northwest National
10 Laboratory, Richland, WA.

11

12 Truex, M.J. and K.C. Carroll. 2013. *Remedy Evaluation Framework for Inorganic,*
13 *Non-Volatile Contaminants in the Vadose Zone.* PNNL-21815, Pacific Northwest
14 National Laboratory, Richland, Washington.

15

16 Truex, M.J., M. Oostrom, G.V. Last, C.E. Strickland, and G.D. Tartakovsky. 2015.
17 *Evaluating Contaminant Flux from the Vadose Zone to the Groundwater in the Hanford*
18 *Central Plateau: SX Tank Farm Case Study.* PNNL-23737, RPT-DVZ-AFRI-026.
19 Pacific Northwest National Laboratory, Richland, Washington.

20

21 Truex, M.J., M. Oostrom, C.E. Strickland, G.B. Chronister, M.W. Benecke, and
22 C.D. Johnson. 2012. Field-scale assessment of desiccation implementation to
23 reduce deep vadose contaminant movement. *Vadose Zone J.*,
24 doi:10.2136/vzj2011.0144.

25

26 Truex, M.J., M. Oostrom, T.W. Wietsma, G.V. Last, and D.C. Lanigan. 2013.
27 *Pore-Water Extraction Scale-Up Study for the SX Tank Farm.* PNNL-21882; RPT-
28 DVZ-AFRI-011, Pacific Northwest National Laboratory, Richland, WA.

29

30 van Genuchten, M.Th. 1980. A closed form equation for predicting the hydraulic
31 conductivity of unsaturated soils. *Soil Science Society of America Journal*
32 44(5):892–898.

33

34 Walvoord, M.A. and B.R. Scanlon. 2004. Hydrologic processes in deep vadose
35 zones in interdrainage arid environments. In: *Groundwater Recharge in a Desert*
36 *Environment: The Southwestern United States.* Water Science and Application
37 Vol. 9. American Geophysical Union, Washington DC.

38

1 White, M.D. and M. Oostrom. 2006. *STOMP – Subsurface Transport Over Multiple*
2 *Phases, Version 4.0 – User’s Guide*. PNNL-15782, Pacific Northwest National
3 Laboratory, Richland, Washington.
4
5 Zachara, J.M., J. Serne, M. Freshley, F. Mann, F. Anderson, M. Wood, T. Jones,
6 and D. Myers. 2007. Geochemical processes controlling migration of tank wastes
7 in Hanford’s vadose zone. *Vadose Zone J.* 6: 985-1003.
8
9 Zhang, Z.F., V.L. Freedman, S.R. Waichler, and S.K. Wurtner. 2005. *2005 Closure*
10 *Assessments for S-SX Tank Farms: Numerical Simulations*. PNNL-15399, Pacific
11 Northwest National Laboratory, Richland, Washington.

Tables

Table 1. Sediment layering and hydraulic properties.

Sediment Layer	Average Layer Thickness (m)	van Genuchten α (1/cm)	van Genuchten n	Residual Saturation ($\text{m}^3_{\text{liquid}}/\text{m}^3_{\text{pore space}}$)	Horizontal K_{sat} (cm/s)	Vertical K_{sat} (cm/s)	Porosity (–)
Backfill	16.0	1.02×10^{-2}	2.177	0.118	7.77×10^{-5}	1.73×10^{-5}	0.323
Hanford H1a	4.1	1.02×10^{-2}	2.177	0.118	7.77×10^{-5}	1.73×10^{-5}	0.323
Hanford H1	4.7	7.90×10^{-3}	2.223	0.133	6.17×10^{-4}	8.87×10^{-5}	0.335
Hanford H2	15.1	1.02×10^{-2}	2.177	0.118	7.77×10^{-5}	1.73×10^{-5}	0.323
Cold Creek Silt	8.7	5.28×10^{-3}	2.249	0.097	9.18×10^{-5}	3.38×10^{-5}	0.420
Cold Creek Carbonate	2.6	1.11×10^{-2}	1.740	0.185	1.39×10^{-3}	5.13×10^{-4}	0.340
Ringold Taylor Flats	3.9	1.37×10^{-2}	1.671	0.120	2.24×10^{-4}	5.01×10^{-5}	0.300
Ringold Wooded Island Unit e	8.1	1.37×10^{-2}	1.671	0.120	2.24×10^{-4}	5.01×10^{-5}	0.300
Clastic Dike	n.a.	5.28×10^{-3}	2.249	0.097	9.18×10^{-5}	3.38×10^{-5}	0.420

Table 2. Estimates of leak times, duration, rates, Tc- 99 inventory, and concentrations (from Johnson and Field, 2010; Girardot and Harlow, 2014; Truex et al., 2015).

Tank	Leak Volume (L) Best/Maximum	Year / Duration (days) Best (Minimum)	Volumetric Rate (L/day)¹ Best/Maximum	Total Tc-99 Inventory (Ci)	Tc-99 Release Concentration (pCi/L)
SX-107	24,200 / 78,100	1964 / 365 (60)	66 / 1,296	4.87	2.01e8
SX-108	1: 9,000 / 9,000 2: 217,000 / 250,000 3: 62,500 / 125,000 Total: 289,500 / 384,000	1964-1965 / 304 (91) 1966-1967 / 365 (61) 1967-1968 / 517 (335)	30 / 99 594 / 4,120 121 / 373	11.70	4.06e7
SX-109	3,800 / 37,900	1965-1966 / 487 (60)	7.8 / 631	0.77	2.03e8
SX-111	11,000 / 11,000	1974 / 27 (27)	407 / 407	0.63	5.73e7
SX-112	102,000 / 167,000	1969 / 16 (16)	6,375 / 10,437	6.45	6.32e7
SX-113	56,800 / 56,800	1962 / 12 (12)	4,733	1.49	2.62e7
SX-114	7,570 / 7,570	1972 / 52 (14)	146 / 542	0.16	2.13e7
SX-115	193,000 / 198,000	1965 / 12 (12)	16,803 / 17,238	4.53	2.35e7

¹The maximum volumetric rate is obtained by dividing the maximum volume by the minimum duration.

²The Tc-99 release concentration is obtained by dividing the inventory by the best volume estimate.

Table 3. Description of single tank simulations to determine flow and transport controlling factors.

Simulation Name	Description
<i>92 mm/year recharge</i>	
-Base Case	Best estimate tank leak information for SX-108 from Table 2 and hydraulic properties from Table 1.
-Center Leak	Leaks originating from tank bottom center.
-Minimum Leak Duration	Minimum leak periods (Table 2).
-Maximum Leak Volume	Maximum leak volumes (Table 2)
-Minimum Leak Duration and Maximum Leak Volume	Minimum leak periods and maximum volumes (Table 2)
-Increased Hydraulic Conductivity	Hydraulic conductivity of all units increased by factor 10.
-Heterogeneous H2	0.5 m silt layers alternating with 0.5 m H2 layers
-Clastic Dike	1 m ² clastic dike (properties in Table 2) extending from leak location at bottom of tank to the water table.
-SX-115 Leak Duration and Volume	Leak volumes and duration for SX-115 tank (Table 2).
<i>recharge sensitivity simulations</i>	
High Recharge	173 mm/year (total precipitation rate)
Low Recharge	46 mm/year (half Base Case)
6 Precipitation Events per Year	15.67 mm/day on first day of fall and winter months
1 Precipitation Event per Year	92 mm/day on January 1

Figures

Figure 1. Location map of the Hanford SX Tank Farm, including observation wells, water-line leak location, and the Tc-99 plume originating from SX-115 tank waste (after Serne et al. 2008c and Johnson and Chou, 2002).

Figure 2. Data availability for evaluating vadose zone contamination and contaminant flux for the SX Tank Farm. An “S” symbol after the data type indicates a single time point is available. A “P” symbol indicates that data are available for periodic or multiple time points.

Figure 3. Hydrogeologic cross-section A-A' as indicated on Fig. 1 (adapted from Serne et al., 2008a,b). The Cold Creek Unit (CCU) consists of the upper silt-dominated layer (dark grey) and the lower cemented carbonate zone (white).

Figure 4. Example of moisture content and Tc-99 distribution for borehole 299-W23-234 (after Serne et al. 2008b). In this figure, the abbreviations PPlz and PPlc are equivalent to CCUz and CCUc, respectively.

Figure 5. Baseline conceptual model, originating from the available data sets shown in Fig. 2, with issues related to controlling features and processes.

(a)

(b)

Figure 6. Sediment zonation for (a) the local SX-108 tank, and (b) the SX Tank Farm numerical model domain. The indicated water table denotes the 1953 location.

Figure 7. (a) Water saturations and (b) dimensionless Tc-99 concentrations for the SX-108 Base Case simulation right after the second leak in 1967.

Figure 8. (a) Water saturations and (b) dimensionless Tc-99 concentrations for the SX-108 Base Case at 2015.

Figure 9. Dimensionless Tc-99 concentrations at 2100 for the SX-108 Base Case.

Figure 10. Tc-99 mass discharge into groundwater for the single tank simulations listed in Table 3.

Figure 11. Effect of recharge on Tc-99 mass discharge into groundwater. The fluxes for the Base Case and the simulations with 6 and 1 precipitation events per year are on top of each other.

Figure 12. Average water flow rates at 5 and 10 m below surface for the Base Case and simulation with a single event per year (92 mm recharge on Jan. 1).

Figure 13. Dimensionless Tc-99 concentration at 2015 for the full SX Tank Farm simulation.

(a)

(b)

Figure 14. (a) Tc-99 mass discharge into groundwater and (b) Tc-99 concentration at downgradient observation wells.

Figure 15. Tc-99 mass discharge into groundwater for various water-line leak rates.

(a)

(b)

(c)

Figure 16. Effect of water-line leak on (a) Tc-99 mass discharge into groundwater (b) simulated and observed concentrations for downgradient wells W22-46 and W22-50, and (c) simulated and observed concentrations for W22-46 and W22-50 from 1990-2015.

Figure 17. Synthesized conceptual model for the SX Tank Farm showing key conclusions related to future contaminant flux conclusions.