Fate and Transport in the Subsurface of Radioactive Waste

by

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DOE Contract No. DE-AC09-96SR18500

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A paper proposed for presentation at the
SCS Advanced Simulation Technologies Conference
Washington D.C.
April 16-20, 2000

and for publication in the proceedings of the meeting

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FATE AND TRANSPORT IN THE SUBSURFACE OF RADIOACTIVE WASTE

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Keywords: Contaminant transport, Radioactive waste, Radionuclides, Vadose zone, Aquifer

ABSTRACT

Disposal of tritium generation wastes in shallow, concrete vaults was modeled to determine aquifer concentrations created by advection and diffusion. A 10,000-year minimum duration was examined, hence material changes in waste containers, vaults and engineered barriers were accommodated in the simulations. Ground-water flow analyses were accomplished in three steady-state stages, representing the intact, cracked, and failed states of the concrete vaults. Radionuclide half-lives and $K_d$ were major inputs to transient transport modeling that was performed to complete the analyses. Contaminant mass fluxes to the water table and concentrations at a hypothetical 100-m down-gradient well from the analyses of two radionuclides were presented displaying distinctively different behaviors.

INTRODUCTION

A linear accelerator option for producing tritium was considered at the Savannah River Site in Aiken, South Carolina (U.S.D.O.E. 1997). Potential waste material would be placed in underground concrete vaults and the interstices would be grouted. Computer simulations were performed to analyze the movement of these buried wastes to a hypothetical well that is 100 meters down-gradient from the vault. Special considerations were made for corrosion of the stainless-steel containers housing the wastes, degradation of the concrete and failure of an engineered infiltration barrier to be placed over the vaults.

Two physical domains were independently modeled, the vadose zone (the unsaturated portion of the subsurface above the water table) and the aquifers below the vadose zone. The model accommodated radioactive decay, production of radioactive progeny, corrosion of waste and waste containers, degradation of concrete and failure of the infiltration barrier.

Contaminant fluxes (curies/year) to the water table produced by vadose zone modeling were input as sources to an aquifer system model. Concentrations (pCi/L) at a 100-m down-gradient location were produced by the aquifer system model. These concentrations were calculated for subsequent comparison with limits to determine which wastes were acceptable based on the specified disposal facility design (not discussed in this paper).

Facility structures that strongly impeded the movement of all radionuclides were the infiltration barrier, waste containers and the vaults. These structures impeded waste movement by restricting water access to the wastes. Factors that strongly affected individual radionuclide movement were the rate of decay and the solid-water partitioning coefficient, $K_d$ (ml/g). Most radionuclides that rapidly decayed never escaped the vault in significant quantities. For a non-infinite source, radionuclides with high $K_d$s were strongly adsorbed to solids, moved slowly and did not attain high concentrations at the 100-m location.

VADOSE ZONE FLOW MODEL

Figure 1 depicts the left half of the vault with the overlaid finite difference mesh that was used for both ground-water flow and contaminant transport through the vadose zone. The right boundary of the modeling domain cuts through the centerline of a cross-section of the vault. This figure shows the waste and the concrete vault covered by clay and surrounded first by sand, then by backfill. The cross-section of the vault shown in Figure 1 is 14.8 m wide by 8.6 m high. The vault is 58 m long. The engineered surface cap was modeled separately and the average infiltration that penetrated the cap was input to the vadose zone model.
The first step in vadose zone modeling was to analyze water flow with PORFLOW©, a finite difference program. The first steady-state flow stage developed for materials in their intact condition produced saturations and Darcy velocities as inputs to the transport analysis.

Boundary conditions at the top of the modeling domain were 4 cm/yr vertically downward fluxes, based on a separate infiltration barrier analysis. Zero flux conditions were prescribed along the sides (symmetry at the right-hand side and far field conditions at the left-hand side). A zero pressure condition was prescribed at the bottom representing the water table.

Selected material properties for the intact vault are shown in Table 1. Concrete has the lowest saturated hydraulic conductivity. Waste was assigned an equivalent saturated hydraulic conductivity due to grout in the voids between waste forms. These two materials in their intact state greatly restrict water flow. The unsaturated flow values are van Genuchten parameters describing the water content — suction curves.

The model described above was analyzed with the PORFLOW© program by executing it in a transient fashion until steady-state conditions were attained. Darcy velocity vectors for the intact stage are plotted in Figure 2. This figure shows that water flow through the vault is very low (the velocity vectors inside the vault were too small to be depicted or seen). Most water from above the vault flows laterally through the overlying sand, then starts moving downward once it passes the edge of the vault.
The vault essentially serves as an umbrella producing a dryer region below it and a wetter region adjacent to it. Time markers indicate that flow reaching the water table near the projected centerline of the vault would arrive in about 300,000 years. Water reaching the water table near the projected edge of the vault that travels through the wetter region would arrive in about 180 years. Water in the far field, only mildly affected by the vault would reach the water table in about 500 years.

The second steady-state flow stage was developed for materials in a degraded condition. During the degraded stage from 576 to 1050 years, cracks are expected to fully penetrate the vault roof and walls. The failed stage occurs at 1050 years when structural collapse of the vaults is expected. Vault degradation and failure and its impacts on waste and the overlying clay were implemented by modifying material properties, assuming that the affected materials were replaced by adjacent material. Table 2 shows the material transformations that were assumed for modeling purposes.

Table 2. Material Transformations Assumed for Degraded and Failed Vaults

<table>
<thead>
<tr>
<th>Description</th>
<th>Degraded Stage</th>
<th>Failed Stage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clay</td>
<td>N/A</td>
<td>To backfill over roof</td>
</tr>
<tr>
<td>Concrete Roof</td>
<td>To clay over roof</td>
<td>To backfill over roof</td>
</tr>
<tr>
<td>Waste</td>
<td>To sand adjacent to walls</td>
<td>To backfill over roof</td>
</tr>
<tr>
<td>Concrete Walls</td>
<td>To sand adjacent to walls</td>
<td>To sand adjacent to walls</td>
</tr>
<tr>
<td>Concrete Floor</td>
<td>N/A</td>
<td>To sand adjacent to floor</td>
</tr>
</tbody>
</table>

The model described above for the degraded stage was analyzed for steady-state conditions. Darcy velocity vectors for the degraded stage are plotted in Figure 3. This figure shows that water flow through the vault is low, but not as low as for the intact stage. Most water from above the vault flows laterally through the overlying sand, then starts moving downward once it passes the edge of the vault.

The degraded vault essentially serves as a smaller, less effective umbrella relative to the intact vault. Time markers indicate that flow reaching the water table near the projected centerline of the vault would arrive in about 23,000 years, vs. 300,000 years for the intact vault. Water reaching the water table near the projected edge of the vault would arrive in about 230 years vs. 180 years for the intact vault. Water in the far field, would reach the water table in about 500 years, the same time as for the intact vault.

The third steady-state flow stage was developed for materials in a failed condition. An upper boundary condition of 40 cm/yr was applied to represent a failed infiltration barrier. The model for the failed stage was analyzed for steady-state conditions. Darcy velocity vectors for the degraded stage are plotted in Figure 4. This figure shows that water flow is primarily downward. The stream trace in the far field reaches the water table in 51 years, a ten-fold decrease in time reflecting the ten-fold increase in the top influx. The sand layer in this stage serves as a minor umbrella, where the flows underneath it are slowed as one moves toward the center of the vault.
Table 3. Distribution Coefficients and Halflives

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Concrete/Waste (ml/g)</th>
<th>Backfill/Sand (ml/g)</th>
<th>Clay (ml/g)</th>
<th>Half-life (ml/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-129</td>
<td>2.0</td>
<td>1.0</td>
<td>1.0</td>
<td>1.57E+07</td>
</tr>
<tr>
<td>Si-32</td>
<td>3.5</td>
<td>35.0</td>
<td>180.0</td>
<td>3.30E+02</td>
</tr>
</tbody>
</table>

Zero mass fluxes through the sides were assigned as boundary conditions. Zero concentrations were assigned at the top and bottom of the model. Initial conditions were zero concentrations.

The vadose zone transport model produced contaminant fluxes to the water table. Results are included and discussed with the aquifer transport results below.

AQUIFER FLOW MODEL

The aquifer flow model was a subset of an extensive regional model consisting of 166,320 elements and encompassing three aquifers. The subset extended from slightly upgradient of the vaults to downgradient seeplines. The submodel consisted of a grid of 50 X-direction cells, 48 Y-direction cells and 20 Z-direction cells. The X- and Y-directions were horizontal. The cell lengths in the X- and Y-directions were a uniform 200-ft width. The Z-length was variable to account for the topography.

Model parameters were copied from the regional model where they had been developed from an extensive data base (Flach and Harris 1997). Seepline fluxes and constant heads from the regional model were applied as boundary conditions and the submodel was executed until steady-state conditions were obtained. Water fluxes and pressures were checked for consistency with the regional model. Water fluxes were input to the aquifer transport model.

AQUIFER TRANSPORT MODEL

The aquifer transport model used the flow model's geometry. Because of limited transport-parameter data, the domain was assumed to consist of sand or clay only. Clay was assigned where the vertical saturated hydraulic
conductivity was less than 1.0E-7 cm/sec. Major input parameters are the decay rates and the $K_d$ values presented earlier in Table 3.

Zero concentrations were assigned as initial conditions and zero contaminant mass fluxes were assigned as boundary conditions at all model edges. Water fluxes from the aquifer flow model were used in the interior of the modeling domain. Contaminant mass fluxes from the vadose zone transport model were input to appropriate aquifer cells as variable sources. The aquifer transport model was executed in a transient manner until the peak concentration at a location 100 meters from the vault was detected.

MODELING RESULTS FOR VADOSE ZONE AND AQUIFER TRANSPORT MODELS

Transport modeling results were based on a unit curie of waste for each radionuclide. Contaminant fluxes at the water table were fractional fluxes (Ci/yr per Ci of inventory) and concentrations at a 100-m well were normalized concentrations (pCi/L per Ci of inventory). Several locations 100-m down-gradient were monitored to ensure that the peak concentration was selected.

Figure 5 shows the fractional fluxes and the normalized concentrations for Si-32. The figure also includes the fractional source release flux, i.e., the rate of flux released from the waste container (Ci/yr per Ci of inventory). The plot of fractional flux from the waste container slopes downward reflecting the relatively fast decay rate (330 year half-life). The relatively low $K_d$ (3.5 ml/g) in concrete by itself would create a relatively fast rise to a peak for the flux to the water table. However, outside the vault, the materials slow the movement where a $K_d$ of 35 ml/g is encountered. That action tends to delay and flatten the peak. The peak flux to the water table occurs at about 2700 years. The peak concentration at a 100-m down-gradient well occurs about 500 years later near 3200 years.

Figure 6 shows the waste release fractional flux, fractional flux to the water table, and the normalized concentrations for I-129. Unlike the Si-32 plot, the I-129 plot of fractional flux from the waste container is flat reflecting the slow decay rate (1.57E7 year half-life). The combination of a low $K_d$ (2.0 ml/g) in concrete and a lower $K_d$ (1.0 ml/g) in the surrounding material creates a fast rise to a peak for the flux to the water table. The peak flux to the water table occurs at about 1051 years. The peak concentration at a 100-m down-gradient well occurs about 11 years later near 1062 years.
I-129 had a peak fractional flux to the water table of 3.9E-4 Ci/yr/Ci versus 8.0E-8 Ci/yr/Ci for Si-32. I-129 had a peak concentration at the 100-m well of 25.0 pCi/L/Ci versus 4.7E-9 pCi/L/Ci for Si-32.

I-129 exhibited a localized dip after its peak. That behavior was likely an artifact of stepping from the degraded steady-state velocity field to the failed steady-state velocity field. When the saturation was instantaneously increased, the mass of contaminant in the water was similarly increased to satisfy the $K_d$ requirement. This discontinuity was exhibited in the form of a short-term peak and dip, typically followed by a gradual increase, often to a higher peak. Smaller simulation time steps near the peak likely would have dampened this effect.

I-129 exhibited the highest and earliest peaks for fluxes to the water table and concentrations at the 100-m well among all the radionuclides modeled. Tritium would have been higher, but it was screened, because the waste container survived for 635 years.

**SUMMARY AND CONCLUSIONS**

Contaminant transport of radioactive waste was modeled from shallow, underground concrete vaults to a hypothetical 100-m down-gradient well. Multiple material changes were simulated along with the radioactive decay process. Plots of fractional fluxes for waste release and masses crossing the water table from the vadose zone were combined with normalized concentrations at a 100-m down-gradient well. Plots for Si-32 and I-129 showed behavior differences reflecting the differences in decay rates and retardation in the materials surrounding the vaults.

**REFERENCES**
