ESTIMATING SURFACE WATER RISK AT OAK RIDGE NATIONAL LABORATORY: EFFECTS OF SITE CONDITIONS ON MODELING RESULTS

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ABSTRACT

Multiple source term and groundwater modeling runs were executed to estimate surface water $^{90}$Sr concentrations resulting from leaching of sludges in five 180,000 gallon Gunite™ tanks at Oak Ridge National Laboratory. Four release scenarios were analyzed: 1) leaching of unstabilized sludge with immediate tank failure; 2) leaching of unstabilized sludge with delayed tank failure due to chemical degradation; 3) leaching of stabilized sludge with immediate tank failure; and 4) leaching of residual contamination out of the shells of empty tanks. Source terms and concentrations of $^{90}$Sr in the stream directly downgradient of the tanks were calculated under these release scenarios. The following conclusions were drawn from the results of the modeling: 1) small changes in soil path length resulted in relatively large changes in the modeled $^{90}$Sr concentrations in the stream; 2) there was a linear relationship between the amount of sludge remaining in a tank and the peak concentration of $^{90}$Sr in the stream; 3) there was a linear relationship between the cumulative $^{90}$Sr release from a tank and the peak concentration of $^{90}$Sr in the stream; 4) sludge stabilization resulted in significantly reduced peak concentrations of $^{90}$Sr in the stream; and 5) although radioactive decay of $^{90}$Sr during the period of tank degradation resulted in incrementally lower peak $^{90}$Sr concentrations in surface water than under the immediate tank failure scenario, these concentrations were equivalent under the two scenarios after about 90 years.

I. INTRODUCTION

At Oak Ridge National Laboratory (ORNL) shallow groundwater in the water table aquifer generally moves along the soil/bedrock interface and discharges to bordering surface water bodies. To estimate surface water $^{90}$Sr concentrations resulting from leaching of sludges in five 180,000 gallon Gunite™ tanks, multiple source term and groundwater modeling runs were performed. The reason for modeling the release of $^{90}$Sr from the tanks was that it is the major potential contributor to human health risk from the tank sludges due to its relatively high inventory in the sludges and its relatively high mobility. Four release scenarios were analyzed: 1) leaching of unstabilized sludge with immediate tank failure; 2) leaching of unstabilized sludge with delayed tank failure due to chemical degradation; 3) leaching of stabilized sludge with immediate tank failure; and 4) leaching of residual contamination out of the shells of empty tanks. Time-dependent source terms for these four scenarios were calculated, based on $^{90}$Sr inventories and tank sludge volumes inferred from sampling results and visual observations of the tank contents, using the Disposal Unit Source Term (DUST) model. The computer code used for the groundwater flow portion of the modeling was FWORK, a 3-dimensional, finite difference groundwater flow and solute transport code developed by GeoTrans, Inc. The contaminant transport was simulated by means of an analytic solution based on one-dimensional transport through a porous medium with linear sorption and radioactive decay. Concentrations of $^{90}$Sr in the stream directly downgradient of the tanks under the various release
scenarios were calculated by diluting the mass flux in groundwater predicted during the transport modeling by the mean flow in the stream.

II. MODEL COVERAGE AND BOUNDARIES

The groundwater model was set up to encompass the main plant area of ORNL, with a grid size of 81 by 66 blocks in the x-y plane and a grid spacing of 15 m (50 ft). There were four layers in the z-direction: layer 1 was approximately 7.6 m (25 ft) thick; layer 2 was about 3 m (10 ft) thick; and layers 3 and 4 were each 7.6 m (25 ft) thick. Layer 1 represented the soil and shallow bedrock and Layers 2, 3, and 4 represented deeper bedrock zones. The aquifer was assumed to be unconfined. Areal recharge was fixed at 0 cm/yr (0 in/year) in areas corresponding to roads, buildings, parking lots, etc., and at 15 cm/yr (6 in/year) in all other areas. Model boundary blocks and the model grid in the area of the Gunite tanks are shown on Figure 1. As seen from this figure, streams, tank dry wells, building sumps, and pipelines were defined as specified head leaky drain boundaries; surface impoundments were defined as constant head boundaries; and the northern edge of the model was defined as a constant head boundary. The model was calibrated to groundwater elevations measured in wells and piezometers and run under steady state conditions.

III. HYDRAULIC PROPERTIES

The bedrock underlying the tanks consists of interbedded limestone and siltstone with minor amounts of shale. The “native” soil (overburden) in the area of the tanks is about 3 m (10 ft) thick and consists primarily of silty clay with minor amounts of gravel backfill material. The east/west (x-direction) hydraulic conductivity value in Layers 1 and 2 of the model was set at 3.5 x 10^{-3} cm/sec (0.1 ft/day), the north/south (y-direction) hydraulic conductivity value was set at 1.8 x 10^{-3} cm/sec (0.05 ft/day), and the vertical (z-direction) hydraulic conductivity value was set at 7.1 x 10^{-4} cm/sec (0.02 ft/day). For Layers 3 and 4, the hydraulic conductivity values were 1.4 x 10^{-4} cm/sec, 3.5 x 10^{-4} cm/sec, and 1.8 x 10^{-4} cm/sec (0.4, 0.1, and 0.05 ft/day) in the x-, y-, and z-directions, respectively. Effective porosity values of 3.5%, 1.0-3.5%, 1.0%, and 1.0% were assigned to Layers 1, 2, 3, and 4, respectively. A mean annual flow in the reach of the stream directly downgradient of ORNL of 170 L/sec (6 ft^3/sec) was used to calculate dilution of contaminants.

IV. MODELING SCENARIO AND METHODOLOGY

Under current conditions, groundwater levels are maintained below the bottom of the tanks by the operation of a collection system consisting of dry wells associated with each tank connected to a process waste pumping station (PS 1) which pumps the water to a process wastewater treatment plant. Under the modeling scenario, PS 1 was assumed to be inoperative, which resulted in groundwater levels around the tanks rising about 1.5 m (5 ft), or about midway up the tanks.

The conceptual model for the movement of ^{90}Sr from the tanks to surface water was as follows:

For a failed or degraded tank, the sludges are fully saturated. ^{90}Sr is leached from unstabilized sludges by groundwater moving laterally through them. For stabilized sludges, the ^{90}Sr diffuses to the surface of the waste form and is washed off by groundwater moving past the surface. For the case in which the tank has been emptied, residual ^{90}Sr contamination in the tank shell diffuses to the inner surface of the shell and is washed off by groundwater moving past the surface. The resulting contaminated groundwater is captured by the dry well associated with the tank and moves through a 20 cm (8 in) vitreous clay east-west header to a 20 cm (8 in) vitreous clay north-south process waste line (see Fig. 2). It then moves through the process waste line to PS 1. Under this scenario, the pumping station is assumed to be inoperative. The contaminated groundwater leaks out of the pumping station manhole, moves through approximately 10 feet of native soil, and leaks into a north-south storm drain line. It then moves in this storm drain line, discharging to the stream via a storm drain outfall (Outfall 302), where dilution occurs in the stream. The total transport path length for this scenario was about 300 m (1,000 ft) from the tanks to the stream.

The following methodology was used for the source modeling using the DUST model:
Figure 1. Model boundary blocks in the ORNL main plant area.
Figure 2. Transport pathway for $^{90}$Sr from the Gunite™ tanks to surface water.
1. Release of $^{90}\text{Sr}$ from unstabilized sludge
The tank shell structure was assumed either to fail immediately (disappear) or degrade over a given period of time at the end of which its failure is total. After tank failure, the sludges were assumed to be completely below the water table as the pumping station (PS1) that depresses the groundwater table below the bottom of the tanks was assumed to be inactive. A surface rinse release with a partitioning mechanism governed by a sludge distribution coefficient, $K_d$, was used to model the transfer of contaminants between the sludge and initially uncontaminated groundwater moving horizontally through the sludge. A mass flux in groundwater was calculated by multiplying the groundwater concentration obtained from the DUST model by the Darcy velocity in the vicinity of the tank calculated using the FTWORK model.

2. Release of $^{90}\text{Sr}$ from stabilized sludge
A diffusion-controlled release mechanism was used to model $^{90}\text{Sr}$ release from a disc-shaped stabilized waste form. The sludge was assumed to be stabilized in a concrete-like material that occupied 50 percent of the tank volume. The waste form was assumed to be entirely below the groundwater table with groundwater moving horizontally past the outer surface of, and not through, the waste form. The $^{90}\text{Sr}$ within the stabilized waste form was assumed to diffuse out of the bulk waste, with the diffusion process being controlled by the effective diffusion coefficient ($D_e$) for the $^{90}\text{Sr}$. The effective $D_e$ accounted for solid-to-liquid partitioning effects and radioactive decay within the waste form. The $^{90}\text{Sr}$ was assumed to be immediately rinsed without partitioning from the waste form into the groundwater, effectively maintaining the $^{90}\text{Sr}$ concentration at the surface at zero. A mass flux in groundwater was calculated by the same method used for the unstabilized sludge modeling runs.

3. Release of $^{90}\text{Sr}$ from the shell of an empty tank
The tank shell $^{90}\text{Sr}$ inventory was estimated by an analytic diffusion solution in which the $^{90}\text{Sr}$ in the sludge pore water diffuses molecularly through the tank shell pore water and is adsorbed onto the tank shell matrix via a solid-to-liquid partition factor for concrete ($K_s$). The sludge $^{90}\text{Sr}$ inventory and the sludge height in the tank were assumed to remain constant over the operating life of the tank. The release of $^{90}\text{Sr}$ from the tank shell was then modeled as a release from a stabilized waste form by a methodology similar to that used for the release modeling described above for the stabilized sludge.

Transport through the soil pathway between PS1 and the storm drain was simulated by means of an analytic solution based on the following equation for one-dimensional contaminant transport through a porous medium with linear sorption and radioactive decay,

$$-v \frac{\partial C}{\partial x} + D \frac{\partial^2 C}{\partial x^2} - \lambda RC = R \frac{\partial C}{\partial t}$$

where: $v =$ seepage velocity;
$D =$ dispersion coefficient;
$C =$ concentration;
$\lambda =$ decay coefficient;
$x =$ distance;
$t =$ time; and
$R = 1 + K_d \rho / \phi$

Solving this equation for concentration, $C$, at time, $t$, and distance, $x$, gives the expression:

$$C_{x,t} = \exp \left[ \frac{\gamma}{2D} \right] \int \frac{x}{\sqrt{4\lambda T}} \exp \left( -\frac{T^2}{4\lambda T} - \frac{T^2}{4D} \right) dT$$

where: $V_{x} = vR$; and
$D_{x} = D/R$.

The initial concentration for a given time step, $C_{x}$, in the above expression was obtained by dividing the mass flux source term from the relevant DUST run time step by the volumetric water flux, calculated by FTWORK, in the drain associated with the tank dry wells. The seepage velocity, $v$, was determined by dividing the Darcy velocity calculated by FTWORK for the area around PS1 by the effective porosity for WAG1 soils of 0.035. The retardation factor, $R$, was calculated using a site-specific $K_s$ for $^{90}\text{Sr}$ of 13.5 ml/g.
porosity, \( \phi \), of 3.5%, and a soil bulk density, \( \rho \), of 1.50 g/cm³. The unretarded dispersion coefficient, \( D \), was 2.56 ft²/yr, which was equal to the seepage velocity (5.11 ft/yr) times a dispersivity of 0.5 ft. The transport distance, \( x \), was set at 3 m (10 ft) for most of the simulations, as this was the assumed amount of “native” soil between PS1 and the storm drain adjacent to PS1. When the above expression was evaluated at a given time step, the resultant concentration, \( C_{\text{result}} \), was multiplied by the volumetric water flux in the storm drain, calculated by FTWORK, to obtain a mass flux. This mass flux was then divided by the flow rate in the stream of 170 L/s (6 ft³/s) to obtain the concentration of the \( {}^{90}\text{Sr} \) in the stream at that time step. A total of 15 modeling runs were performed for the four release scenarios using the \( {}^{90}\text{Sr} \) transport conceptual model outlined above and removing none, 50%, and 90% of the sludge in the tanks. In addition, a sensitivity analysis of the length of the native soil pathway was also performed under the unstabilized sludge in failed tank scenario in Tank W-10 for soil path lengths of 1.5, 3.0, 6.0, and 15 m (5, 10, 20, and 50 ft).

V. RESULTS AND DISCUSSION

The results of the source term and groundwater transport modeling are summarized in Table 1. For tank W-10, peak concentrations resulting from leaching of stabilized sludges were about 10% of peak concentrations resulting from leaching of unstabilized sludges and peak concentrations from a degraded tank were about two-thirds of peak concentrations from a failed tank.

The effects of variations in soil transport path length on \( {}^{90}\text{Sr} \) concentrations in surface water for unstabilized sludge in a failed tank W-10 are summarized in Table 2.

Table 2. Soil transport path length effects on \( {}^{90}\text{Sr} \) concentrations in surface water

<table>
<thead>
<tr>
<th>Soil path length, m (ft)</th>
<th>( {}^{90}\text{Sr} ) conc., pCi/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 (5)</td>
<td>1,160</td>
</tr>
<tr>
<td>3.0 (10)</td>
<td>650</td>
</tr>
<tr>
<td>6.0 (20)</td>
<td>230</td>
</tr>
<tr>
<td>15 (50)</td>
<td>11</td>
</tr>
</tbody>
</table>

Increasing the soil transport path length by a factor of ten decreased the peak \( {}^{90}\text{Sr} \) concentration in the stream by a factor of about one hundred.

Figure 3 is a plot of surface water \( {}^{90}\text{Sr} \) concentration vs. time for the tank degradation and tank failure scenarios for tank W-10. As seen from this plot, although the peak concentration was higher for the tank failure scenario, the concentrations under the two scenarios were essentially equivalent after about 90 years.

VI. CONCLUSIONS

The following conclusions were reached from the results of the source term and groundwater transport modeling:

1) The most critical part of the analysis was the effect of retardation in soil on the peak \( {}^{90}\text{Sr} \) concentrations in surface water. Small changes in soil path length resulted in relatively large changes in the modeled maximum \( {}^{90}\text{Sr} \) concentrations in the stream.
Table 1. Source term and groundwater transport modeling results.

<table>
<thead>
<tr>
<th>Tank</th>
<th>Release scenario</th>
<th>Sludge remaining in tank (%)</th>
<th>UCL_{68} total ⁹⁰Sr sludge inventory (Ci)</th>
<th>Total cumulative release from tank (Ci)</th>
<th>Measured sludge K_d</th>
<th>Literature sludge K_d</th>
<th>Measured UCL,</th>
<th>Literature UCL,</th>
</tr>
</thead>
<tbody>
<tr>
<td>W-6</td>
<td>Sludge in failed tank</td>
<td>100</td>
<td>1,000</td>
<td>27</td>
<td>74</td>
<td>25</td>
<td>68</td>
<td></td>
</tr>
<tr>
<td>W-7</td>
<td>Sludge in failed tank</td>
<td>100</td>
<td>1,200</td>
<td>NC</td>
<td>70</td>
<td>NC</td>
<td>77</td>
<td></td>
</tr>
<tr>
<td>W-8</td>
<td>Sludge in failed tank</td>
<td>100</td>
<td>3,600</td>
<td>NC</td>
<td>250</td>
<td>NC</td>
<td>280</td>
<td></td>
</tr>
<tr>
<td>W-9</td>
<td>Sludge in failed tank</td>
<td>100</td>
<td>780</td>
<td>NC</td>
<td>54</td>
<td>NC</td>
<td>58</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Sludge in failed tank</td>
<td>100</td>
<td>8,500</td>
<td>140</td>
<td>600</td>
<td>150</td>
<td>650</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Sludge in failed tank</td>
<td>50</td>
<td>4,300</td>
<td>67</td>
<td>300</td>
<td>74</td>
<td>330</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Sludge in failed tank</td>
<td>10</td>
<td>850</td>
<td>13</td>
<td>70</td>
<td>15</td>
<td>65</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Sludge in degraded tank</td>
<td>100</td>
<td>8,500</td>
<td>NC</td>
<td>380</td>
<td>NC</td>
<td>410</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Sludge in degraded tank</td>
<td>50</td>
<td>4,300</td>
<td>NC</td>
<td>220</td>
<td>NC</td>
<td>260</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Sludge in degraded tank</td>
<td>10</td>
<td>850</td>
<td>NC</td>
<td>38</td>
<td>NC</td>
<td>43</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Stabilized sludge in failed tank</td>
<td>100</td>
<td>8,500</td>
<td>NC</td>
<td>36</td>
<td>NC</td>
<td>62</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Stabilized sludge in failed tank</td>
<td>50</td>
<td>4,300</td>
<td>NC</td>
<td>18</td>
<td>NC</td>
<td>31</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Stabilized sludge in failed tank</td>
<td>10</td>
<td>850</td>
<td>NC</td>
<td>3.6</td>
<td>NC</td>
<td>6.2</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Tank shell assuming sludge height of 360 cm³</td>
<td>0</td>
<td>85</td>
<td>NC</td>
<td>10.7</td>
<td>NC</td>
<td>3.5</td>
<td></td>
</tr>
<tr>
<td>W-10</td>
<td>Tank shell assuming sludge height of 14 cm³</td>
<td>0</td>
<td>62</td>
<td>NC</td>
<td>7.9</td>
<td>NC</td>
<td>2.2</td>
<td></td>
</tr>
</tbody>
</table>

* UCL_{68} = upper 95th percentile confidence limit for the triangular distribution of sludge inventory.

* The measured sludge K_d value of 65 mL/g was estimated using sludge samples collected from Tank W-10 in 1995.

* The literature sludge K_d value of 13.5 mL/g was the same value used for soils and was a conservatively low estimate.

* Tank full of sludge during its 52-year lifetime.

* Average sludge height during the 52-year lifetime of the tank.
2) There was a linear relationship between the amount of sludge remaining in a tank and the peak concentration of $^{90}$Sr in the stream; 3) There was a linear relationship between the cumulative $^{90}$Sr release from a tank and the peak concentration of $^{90}$Sr in the stream; 4) Sludge stabilization resulted in significantly reduced peak concentrations of $^{90}$Sr in the stream; and 5) Delaying tank failure resulted in lower peak $^{90}$Sr concentrations in surface water. This reduction in peak concentration was attributed to the radioactive decay of $^{90}$Sr during the period in which the tank is chemically degrading and is allowed to remain intact. However, modeled concentrations under the degraded tank scenario were equivalent to those calculated under the failed tank scenario after about 90 years.

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REFERENCES


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