

Effect of Selected Modeling Assumptions on Subsurface Radionuclide Transport Projections for the Potential Environmental Management Disposal Facility at Oak Ridge, Tennessee



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June 28, 2016

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Environmental Sciences Division

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Projections for the Potential Environmental Management Disposal Facility at Oak Ridge,
Tennessee**

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ABSTRACT

The Department of Energy's Office of Environmental Management recently revised a Remedial Investigation/ Feasibility Study (RI/FS) that included an analysis of subsurface radionuclide transport at a potential new Environmental Management Disposal Facility (EMDF) in East Bear Creek Valley near Oak Ridge, Tennessee. The effect of three simplifying assumptions used in the RI/FS analyses are investigated using the same subsurface pathway conceptualization but with more flexible modeling tools. Neglect of vadose zone dispersion was found to be conservative or non-conservative, depending on the retarded travel time and the half-life. For a given equilibrium distribution coefficient, a relatively narrow range of half-life was identified for which neglect of vadose zone transport is non-conservative and radionuclide discharge into surface water is non-negligible. However, there are two additional conservative simplifications in the reference case that compensate for the non-conservative effect of neglecting vadose zone dispersion: the use of a steady infiltration rate and vadose zone velocity, and the way equilibrium sorption is used to represent transport in the fractured material of the saturated aquifer. With more realistic representations of all three processes, the RI/FS reference case was found to either provide a reasonably good approximation to the peak concentration or was significantly conservative (pessimistic) for all parameter combinations considered.

1. INTRODUCTION

The Department of Energy's Office of Environmental Management recently revised a Remedial Investigation/ Feasibility Study (RI/FS) that included an analysis of subsurface radionuclide transport at a potential new Environmental Management Disposal Facility (EMDF) in East Bear Creek Valley near Oak Ridge, Tennessee. This report summarizes a set of supplemental analyses of radionuclide transport that evaluate the effect of several simplifications used in analyses to determine preliminary Waste Acceptance Criteria.

The RI/FS used the PATHRAE-RAD [Merrel et al., 1995] software. PATHRAE-RAD's pathway conceptualization considers one-dimensional vertical transport through the vadose zone beneath the land disposal facility followed by horizontal transport through a groundwater aquifer.

PATHRAE-RAD makes several simplifying assumptions about transport in the subsurface. Most important, it neglects dispersion in the vadose zone. Neglect of dispersion in the vadose zone may cause radionuclide releases to the environment to be underestimated (non-conservative) in some situations. In particular, when the radionuclide half-life is short compared with retarded travel time, dispersion may cause an increase in peak breakthrough because some fraction of the mass may arrive before having time to decay. However, the conditions in which this is expected also correspond to the conditions where the radionuclide discharge is highly attenuated in the subsurface. In addition, there are several other simplifications in PATHRAE that make it necessary to use conservative bounding approximations for some parameter values. For example, matrix diffusion in a fractured rock system is not included. In addition, transient infiltration rates are not represented. The combined effects of those neglected processes are assessed in this report. In particular, the effect of vadose zone dispersion is evaluated, as is the potentially compensating effects of matrix diffusion and explicit representation of transient flow in the vadose zone.

2. METHOD OF ANALYSES

The analyses undertaken here use the same pathway conceptualization as PATHRAE's groundwater transport to surface water pathway. Specifically, radionuclide leaching from the source zone, transport in

the vadose and saturated zones, and dilution by surface water are represented. However, the method of analyses used here is more flexible than PATHRAE and accommodates dispersion in the vadose zone, transient flow velocities, and matrix diffusion effects.

The leach rate from the source zone and surface water dilution is modeled the same as in PATHRAE. Vadose zone transport is represented by solving the advection dispersion equation with equilibrium sorption so that dispersion in the vadose zone and transient infiltration effects can be accommodated. Transport in the saturated aquifer is evaluated with the advection dispersion equation coupled to a matrix diffusion system.

2.1 RADIONUCLIDE SOURCE

The flux of radionuclides from the source zone into the underlying vadose zone is calculated with a simple leaching calculation, identical to PATHRAE. Specifically, the activity flux f_{leach} (Ci/m² - yr) leaving the source zone is

$$A_{sz}f_{leach}(t) = \frac{Q_{sz}(t)}{R_{sz}L_{sz}\phi_{sz}}I(t)$$

where Q_{sz} is the percolation flux through the source zone, and R_{sz} , L_{sz} , and ϕ_{sz} are the retardation factor, thickness, and porosity of the source zone. The repository footprint area is denoted A_{sz} . The time-dependent inventory $I(t)$ is governed by the ordinary differential equation for decay and leaching

$$\frac{dI}{dt} = -\lambda I - \frac{Q_{sz}(t)}{R_{sz}L_{sz}\phi_{sz}}I$$

subject to the initial condition $I(0) = I_0$.

2.2 VADOSE ZONE TRANSPORT

Vadose zone transport is represented by the advection dispersion equation on the one-dimensional pathway from $z = 0$ to $z = \Delta_z$ where z is depth in the vadose zone beneath the landfill liner and Δ_z is the thickness of the vadose zone. The governing equation for radionuclide concentration $C_v(t, z)$ in the vadose zone is

$$R_v \frac{\partial C_v}{\partial t} = -V_v(t) \frac{\partial C_v}{\partial z} + \alpha_v |V_v(t)| \frac{\partial^2 C_v}{\partial z^2} - \lambda R_v C_v$$

with initial condition

$$C_v(0, z) = 0$$

and boundary conditions

$$V_v(t)C_z(t, z) - \alpha_v |V_v(t)| \frac{\partial C_z(t, z)}{\partial z} \Big|_{z=0} = f_{leach}(t)$$

$$C_v(t, 3\Delta_v) = 0$$

Here R_v is the retardation factor, V_v is the vadose zone velocity, α_v is the vadose zone dispersivity, λ is the decay constant, f_{leach} is the rate of leaching from the source zone, and t is time. Molecular diffusion has been neglected in the dispersion term relative to hydrodynamic dispersion.

Note the boundary condition is applied at a distance $3\Delta_z$ as an approximation for a distance boundary, but the flux into the underlying aquifer is evaluated at Δ_z . The flux from the vadose zone into the underlying aquifer is then evaluated as

$$f_{vz}(t) = V_v(t)C_z(t, \Delta_z) - \alpha_z |V_v(t)| \left. \frac{\partial C_z(t, z)}{\partial z} \right|_{z=\Delta_z}$$

2.3 SATURATED ZONE TRANSPORT

Transport in the saturated aquifer is evaluated with the advection dispersion equation coupled to a matrix diffusion system. The governing equations for radionuclide concentration in the fractures $C_f(t, x)$ and matrix $M(t, x)$ as function of time t , travel distance x , and distance y from a fracture are

$$R_f \frac{\partial C_f}{\partial t} = -V_a(t) \frac{\partial C_f}{\partial x} + \alpha_a |V_a(t)| \frac{\partial^2 C_f}{\partial x^2} - \lambda R_f C_f + \frac{D_m}{b} \frac{\partial M}{\partial y} \Bigg|_{y=0}$$

$$R_m \frac{\partial M}{\partial t} = D_m \frac{\partial^2 M}{\partial y^2} - \lambda R_m M$$

with initial conditions $C_f(0, x) = 0$ and $M(0, x, y) = 0$ and boundary conditions

$$C_f(t, x) = M(t, x, 0)$$

$$\left. \frac{\partial M}{\partial y} \right|_{y=B} = 0$$

$$V_a(t)C_f(t, 0) - \alpha_a |V_a(t)| \left. \frac{\partial C_f}{\partial x} \right|_{x=0} = f_{vz}(t)$$

Here R_f is the retardation factor in fractures, R_m is the retardation factor in the matrix, V_a is the groundwater velocity, α_a is the dispersivity, λ is the decay constant, D_m is matrix diffusion coefficient, b is the fracture half-aperture, and B is one-half the fracture spacing (matrix block size).

When the matrix effective diffusion coefficient is set to zero, this system reduces to the equilibrium sorption system.

2.4 SURFACE WATER DILUTION

The concentration in surface water is calculated by executing the model chain starting with a specified unit concentration in the source zone. The result is a normalized discharge in units of yr^{-1} , which is then scaled by the assumed RI/FS initial inventory of $1.68 \cdot 10^6$ Ci and diluted into surface water discharge $7.36 \cdot 10^5$ m^3/yr to obtain a concentration.

2.5 SOLUTION METHOD

The above system of equations describing vadose zone transport was implemented in Mathematica™ (Wolfram Research, 2014) and solved with Mathematica's NDSolve module. In the variants that neglected matrix diffusion in the saturated zone ($D_m = 0$), Mathematica™ was also used to solve for saturated zone transport. When matrix diffusion was represented, the MARFA (Painter et al., 2007) software was used to solve for saturated zone transport.

3. PATHWAY PROPERTIES

Reference case pathway property assumptions are given in Table 1.

Table 1. Pathway properties for the reference case

Parameter	Value
Source zone thickness [m]	16.16
Source zone porosity [-]	0.44
Source zone bulk density [kg/m^3]	1600
Vadose zone thickness [m]	6.7
Vadose zone porosity [-]	0.44
Vadose zone water content [-]	0.38
Vadose zone bulk density [kg/m^3]	1800
Vadose zone velocity [m/yr]	0 <200 years
	0.029 200 – 1000 years
	0.088 >1000 years
Travel distance in aquifer [m]	719 ¹
Aquifer porosity [-]	0.04
Aquifer bulk density [kg/m^3]	1800
Aquifer velocity [m/yr]	21.3
Dispersivity in aquifer [m]	47.6

¹The value is the travel distance from the repository edge to surface water discharge point plus one-half the repository footprint size in the direction of groundwater flow.

4. GENERIC ANALYSES

4.1 EFFECT OF NEGLECTING DISPERSION IN THE VADOSE ZONE

Several sets of simulations were undertaken to identify combinations of half-life and equilibrium distribution coefficient (K_d) for which PATHRAE's neglect of dispersion in the vadose zone is not conservative. When the radionuclide half-life is short compared with retarded travel time, dispersion may cause an increase in peak breakthrough because some fraction of the mass may arrive before having time

to decay. However, the conditions for which this is expected correspond to the conditions where the radionuclide discharge is highly attenuated in the subsurface.

The simulations considered transient infiltration with two dispersivity values in the vadose zone (1 cm and 67 cm). Three values of K_d in the vadose zone were considered. A range of half-lives was used for each value of K_d .

Figures 1 through 4 show results for K_d of 1, 2, 3 and 10 ml/g, respectively. Each of the three figures has two subplots. The top subplot in each shows the ratio of surface water concentration assuming minimal (1 cm) and significant (67 cm) dispersion in the vadose zone on the y-axis. The case with 1-cm dispersion is not expected to be significantly different from the case with no dispersion. The x-axis is half-life. The horizontal line is the value 1. Below that line, neglect of vadose zone dispersivity is conservative, while that assumption is not conservative for points above that line. The lower subplot in each figure is the surface water concentration for the 67 cm dispersivity case. The horizontal line in the lower plot is the value 0.07 pCi/l. For two-thirds of the radionuclides, the surface water concentration corresponding to an excess lifetime cancer risk of 10^{-5} is 0.07 pCi/l or greater (Kenworthy, 2016). Thus, the value 0.07 pCi/l is a convenient threshold for identifying combinations of K_d and half-life that are of no interest due to high amount of decay, even if neglecting dispersion is not conservative.

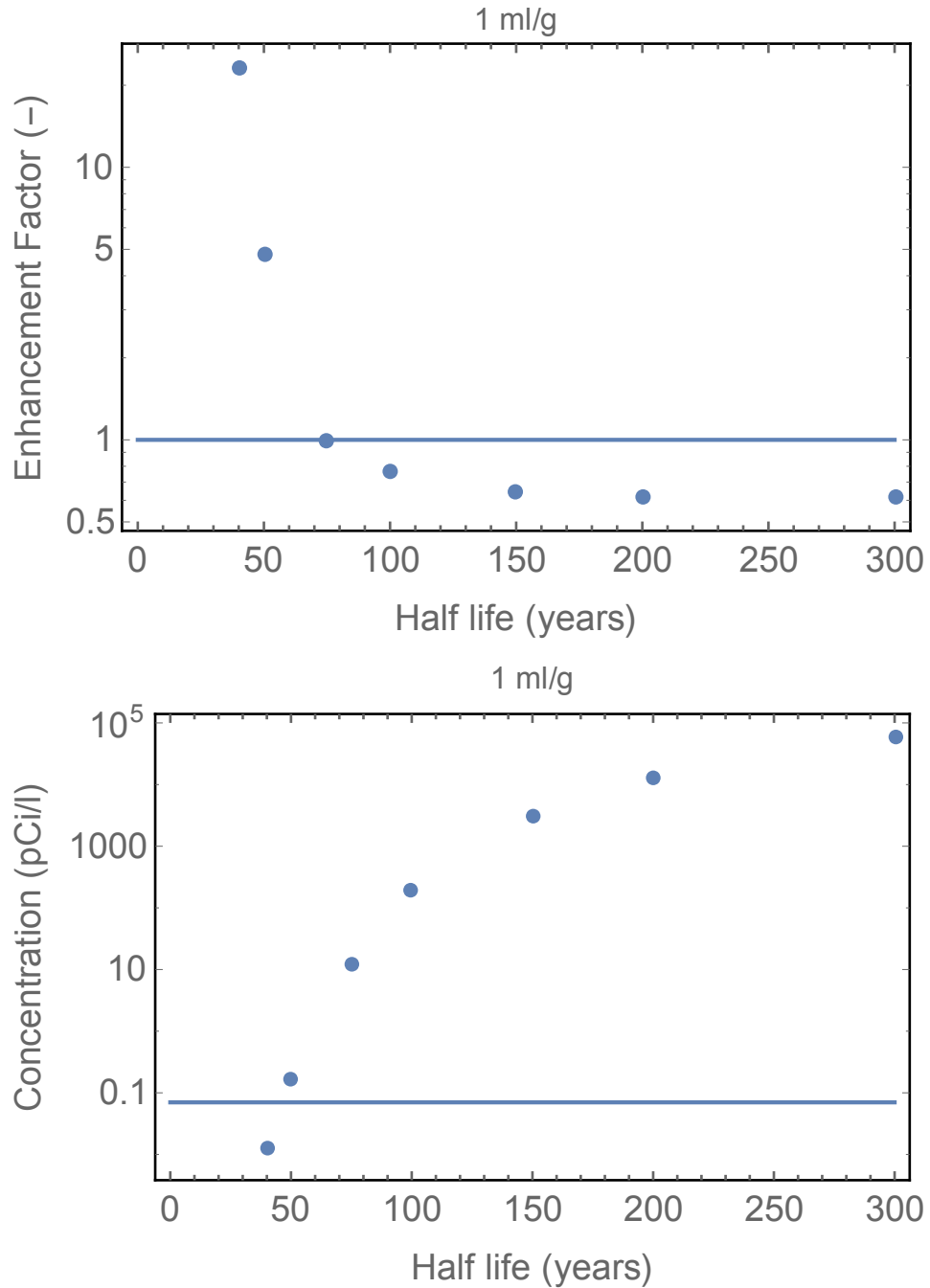


Figure 1. Surface water concentration with dispersion in the vadose zone relative to value calculated with minimal dispersion (upper plot, described as “enhancement factor”) and surface water concentration calculated with dispersion in the vadose zone (lower plot) for a K_d of 1 ml/g in the vadose zone. The range of half lives for which neglect of vadose zone dispersion is non-conservative and resulting surface water concentration is above a risk threshold of 0.07 pCi/l is approximately 50 to 75 years in this case.

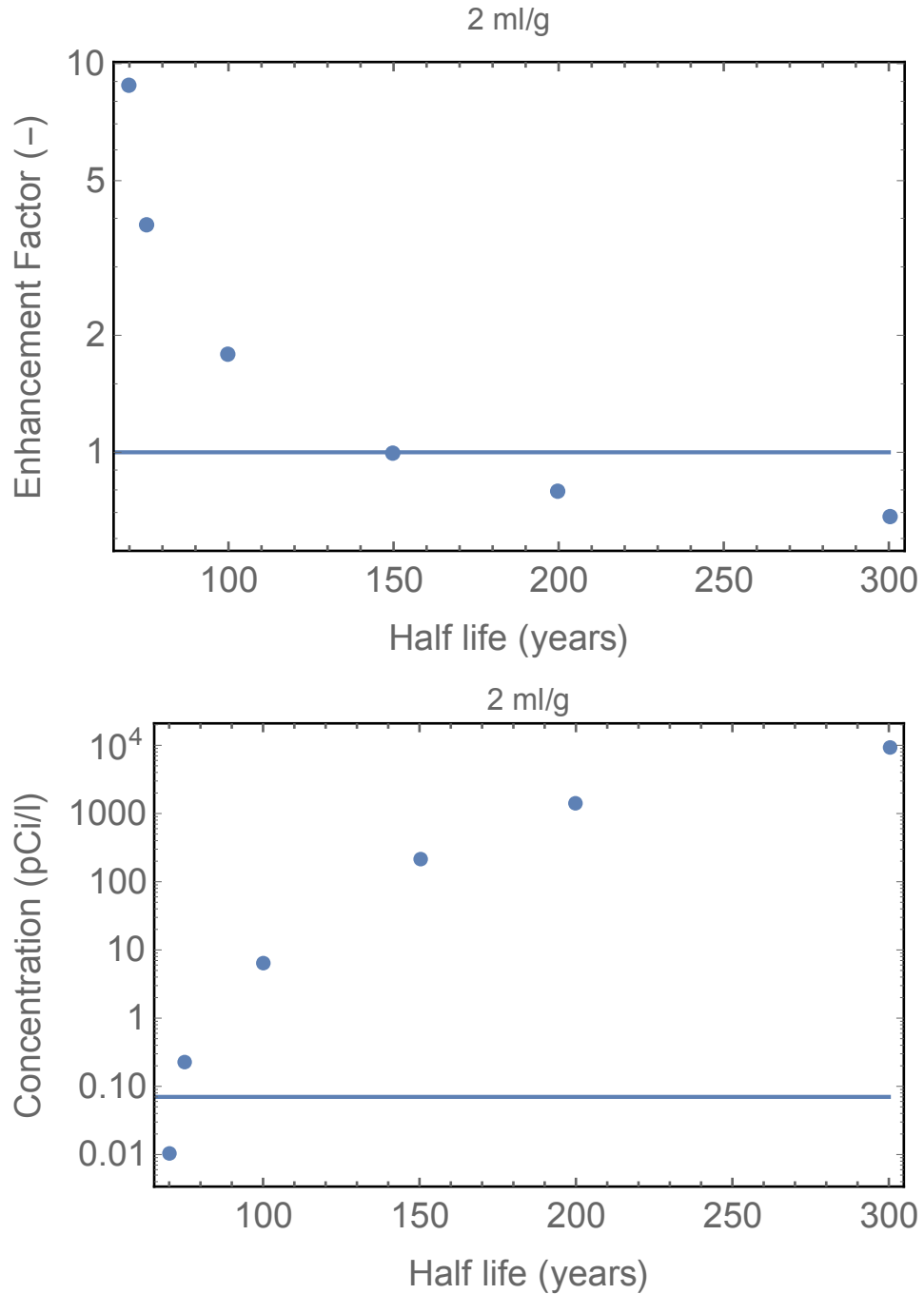


Figure 2. Surface water concentration with dispersion in the vadose zone relative to value calculated with minimal dispersion (upper plot, described as “enhancement factor”) and surface water concentration calculated with dispersion in the vadose zone (lower plot) for a K_d of 2 ml/g in the vadose zone. The range of half lives for which neglect of vadose zone dispersion is non-conservative and resulting surface water concentration is above a risk threshold of 0.07 pCi/l is approximately 75 to 150 years in this case.

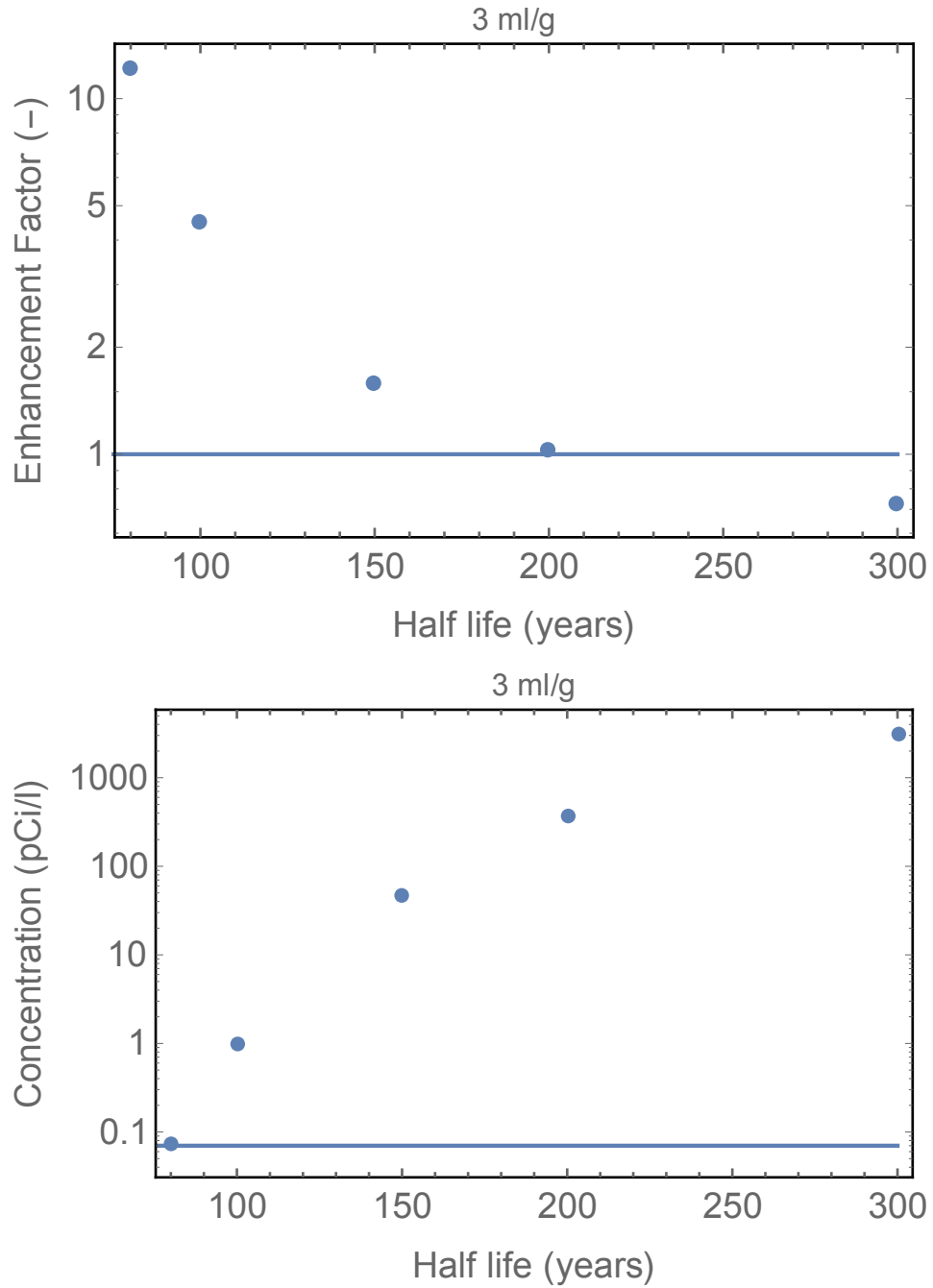


Figure 3. Surface water concentration with dispersion in the vadose zone relative to value calculated with minimal dispersion (upper plot, described as “enhancement factor”) and surface water concentration calculated with dispersion in the vadose zone (lower plot) for a K_d of 3 ml/g in the vadose zone. The range of half lives for which neglect of vadose zone dispersion is non-conservative and resulting surface water concentration is above a risk threshold of 0.07 pCi/l is approximately 80 to 200 years in this case.

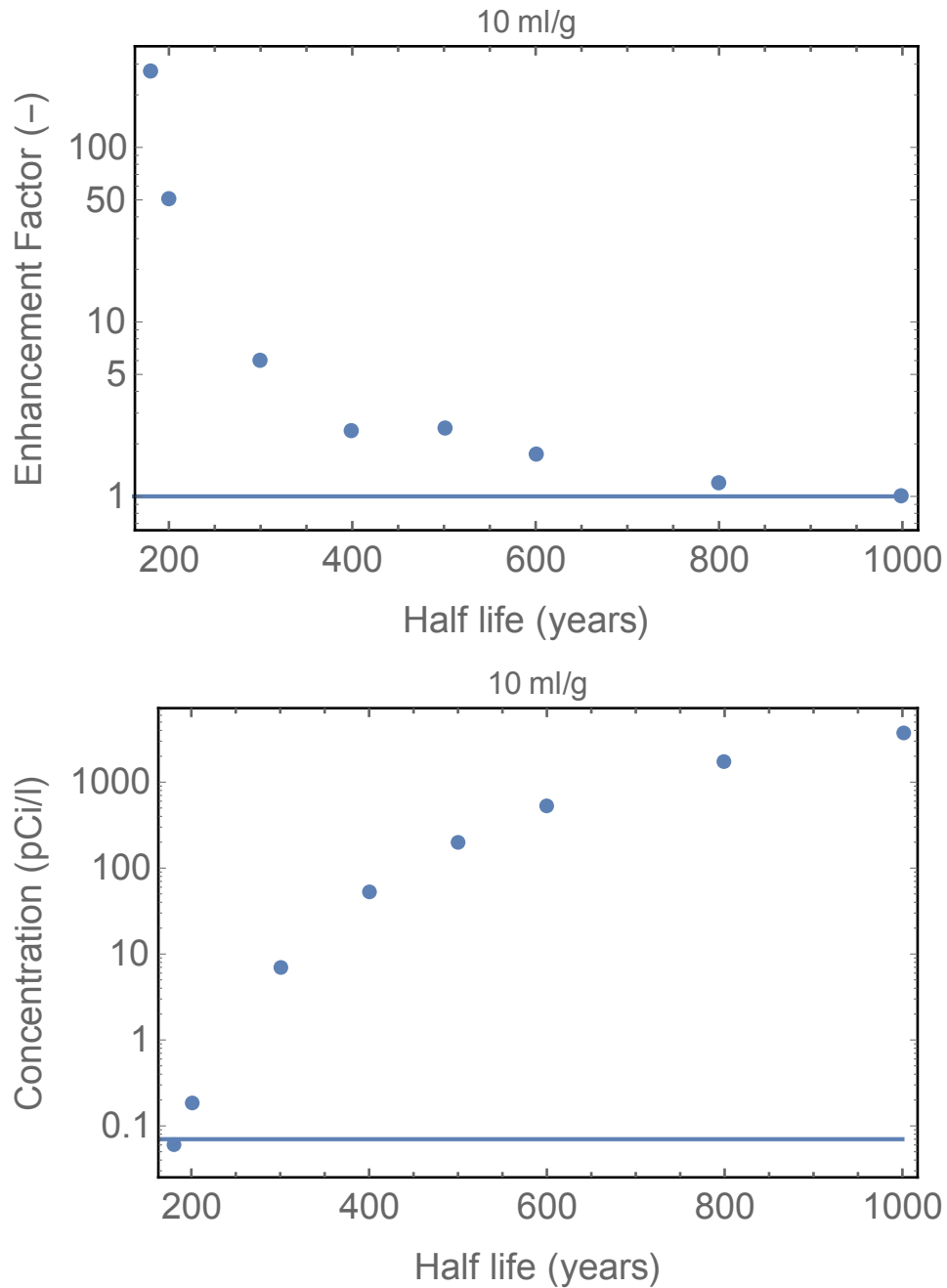


Figure 4. Surface water concentration with dispersion in the vadose zone relative to value calculated with minimal dispersion (upper plot, described as “enhancement factor”) and surface water concentration calculated with dispersion in the vadose zone (lower plot) for a K_d of 3 ml/g in the vadose zone. The range of half lives for which neglect of vadose zone dispersion is non-conservative and resulting surface water concentration is above a risk threshold of 0.07 pCi/l is approximately 200 to 1000 years in this case.

It can be seen from Figures 1 through 4 that for a given K_d value, a relatively narrow range of half-lives exists for which neglect of vadose zone dispersion is negligible and surface water concentration resulting from that radionuclide is non-negligible. The range is approximately 50 to 75 years for K_d of 1 ml/g, 75 to 150 years for K_d of 2 ml/g, 80 to 200 years for K_d of 3 ml/g, and 180 to 1000 years for K_d of 10 ml/g.

4.2 COMPENSATING EFFECT OF TRANSIENT VADOSE ZONE VELOCITY

It is important to note that the results in Figures 1 through 4 were obtained with transient infiltration rates, which affected both the leach rate from the source zone and the vadose zone velocity. Transient velocities are not accommodated by PATHRAE. Thus, in the reference case PATHRAE simulations, the higher vadose zone velocity of 0.088 m/yr was applied to the entire simulation period. The effect of using a steady infiltration rate on calculated surface water concentration is shown in Figure 5 for two combinations of K_d and half-life. The red curve in each plot is the case with minimal dispersion and steady infiltration, similar to PATHRAE reference case. The other two curves in each plot are for the transient infiltration cases with minimal and significant dispersion in the vadose zone. As discussed above, adding dispersion in the vadose zone increases the calculated surface water concentration for some combination of K_d and half-life, which is not conservative from a risk perspective. However, the more realistic representation of transport including both transient infiltration and vadose zone dispersion results in smaller surface water concentrations when the K_d is not large. Stated differently, the conservatism of using a steady infiltration rate is a larger effect than the non-conservative neglect of vadose zone dispersion when K_d is not large. For the $K_d=10$ ml/g case, the non-conservative neglect of vadose zone dispersion is the larger effect. However, the surface water concentration is small in this case and close to the risk threshold of 0.07 pCi/l.

4.3 COMPENSATING EFFECT OF MATRIX DIFFUSION

Matrix diffusion is not represented directly in PATHRAE. Instead, the parameters appearing in the equilibrium sorption model are selected to bound the peak breakthrough from the saturated zone. Here the degree of conservatism inherent in that approach is evaluated. The model chain is similar to the other calculations in this supplemental analysis except that the computer code MARFA is used for the saturated zone so that matrix diffusion effects can be represented.

The saturated zone parameters were modified from the PATHRAE reference case for the MARFA runs. This adjustment is required because of differences in the two model conceptualizations. In particular, the matrix diffusion model in MARFA provides a more realistic representation of transport in the fractured material in the saturated zone, and thus requires fewer conservative bounding-type approximations.

The fracture parameters assumed are based on previous modeling work at the site (McKay et al., 1997). A fracture spacing of 10 cm and a fracture aperture of 0.12 mm were used. This corresponds to a fracture porosity of 0.0012. This value of fracture porosity was then used to scale the saturated zone travel time from the value used in the PATHRAE reference case. The resulting travel time is 0.67 years. In addition, the K_d in the saturated zone was not reduced from the vadose zone value, as opposed to the PATHRAE reference case, which used a saturated zone K_d that is 10% of the vadose zone value.

The basis for these changes from the PATHRAE reference case is that the PATHRAE equilibrium sorption model does not distinguish between primary and secondary porosity and does not represent mass transfer limitations between flow zones (fractures) and sorption sites that are located in the matrix. Thus,

the PATHRAE reference case reduced K_d in the saturated zone compared to the vadose zone and used an effective porosity that is intermediate between the fracture and matrix values. Because MARFA represents primary and secondary porosity directly and has an explicit representation of mass transfer limitations through the matrix diffusion model, those conservative assumptions were not needed.

As for effective matrix diffusivity, McKay et al. (1997) estimate a value of $1.89 \cdot 10^{-2} \text{ m}^2/\text{yr}$. Given the uncertainty in this number a conservative value of $1.89 \cdot 10^{-4} \text{ m}^2/\text{yr}$ is used here.

Surface water concentrations versus time with and without matrix diffusion are shown in Figure 6 for two combinations of K_d and half-life. The red curve in each plot is the case with minimal dispersion in the vadose zone, the solid blue curve is the case with dispersion, and the dashed curve is for the case with dispersion and matrix diffusion. Similar results were obtained for the 3 ml/g and 1 ml/g cases (results not shown). As shown in the previous subsection, dispersion in the vadose zone is non-conservative from a risk perspective for a limited range of half-lives. The results in Figure 6 demonstrate that other conservatisms in the PATHRAE reference case – specifically the way the equilibrium sorption model is parameterized to compensate for lack of a matrix diffusion representation – more than compensates for the non-conservative neglect of dispersion in the vadose zone for all but one parameter combination. In the lower plot of Figure 6, it can be seen that matrix diffusion does not fully compensate for the non-conservative neglect of dispersion. However, the peak concentrations calculated within the PATHRAE-like approximation (red curve) and the more realistic representation (dashed curve) are not significantly different, indicating that PATHRAE provides a good approximation in that case albeit not a strictly conservative one.

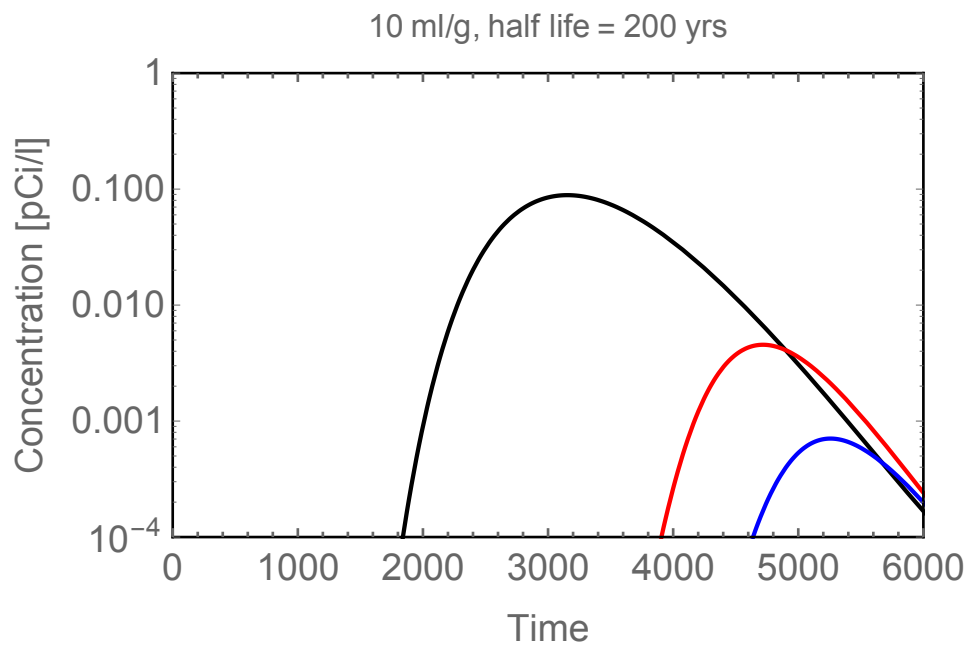
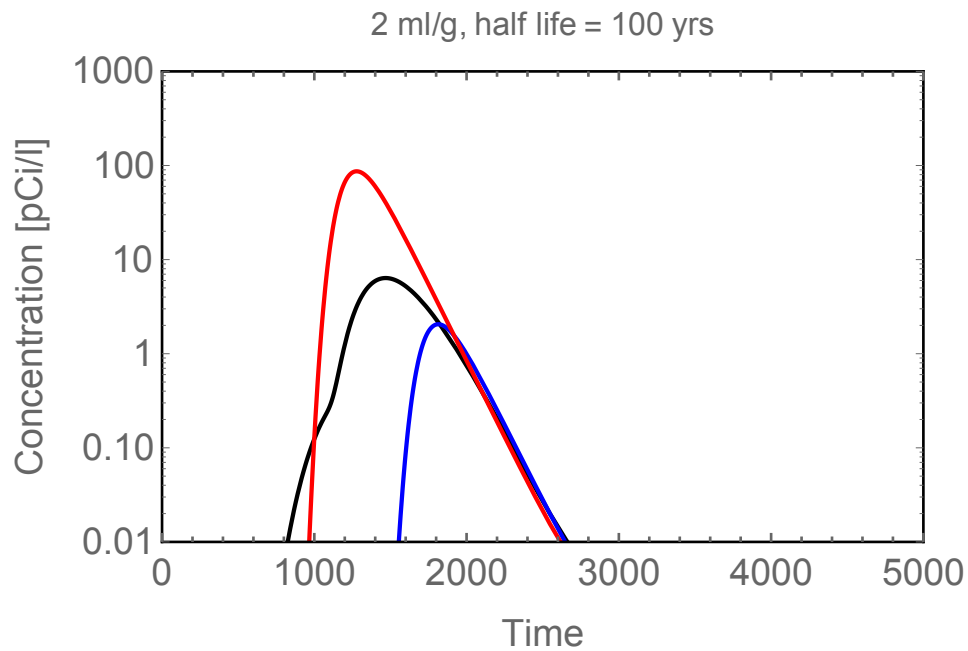


Figure 5. Calculated surface water concentration versus time for two combinations of K_d and half-life. The red curve is the case with minimal dispersion in the vadose zone and steady infiltration (similar to PATHRAE reference case), the blue curve is for the case with minimal dispersion and transient infiltration, and the black curve is the case with dispersion and transient infiltration. Although dispersion increases the calculated surface water concentration in the higher K_d case, the concentration is low and close to the risk threshold of 0.07 pCi/l.

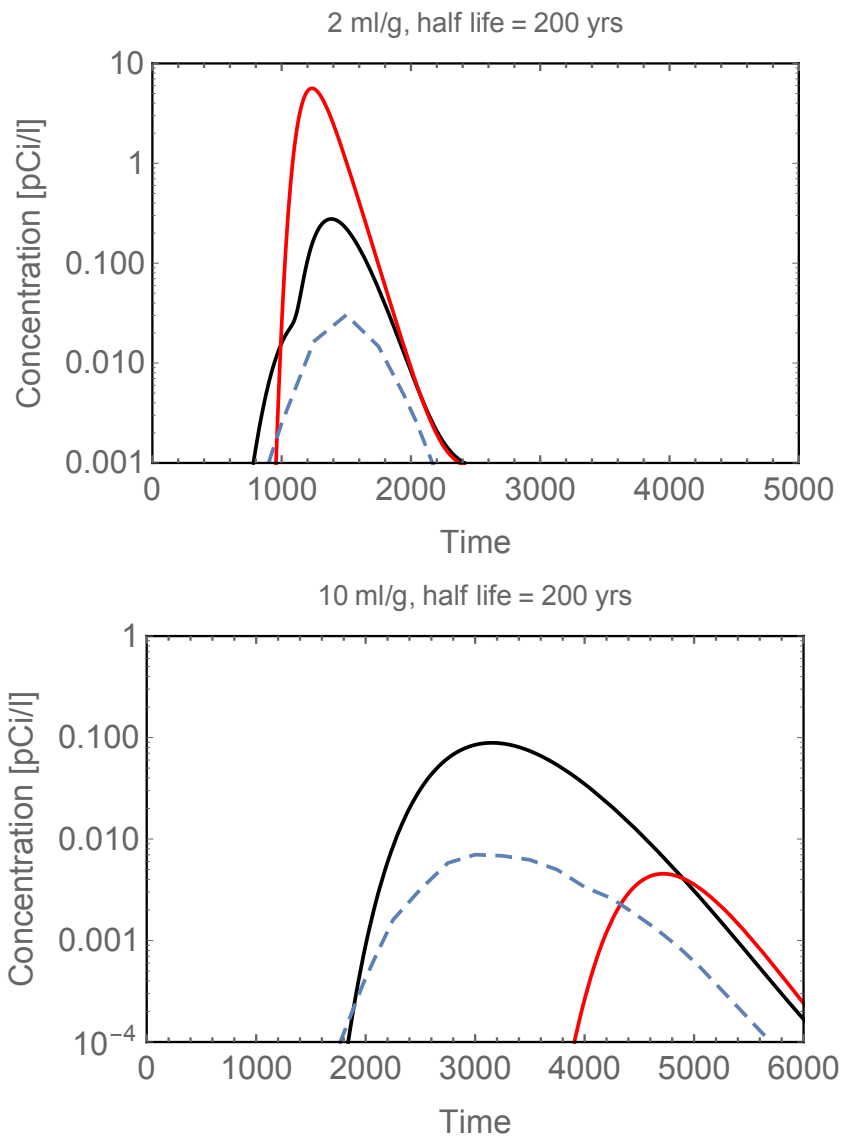


Figure 6. Calculated surface water concentration versus time with and without matrix diffusion. The red curve is the case with minimal dispersion in the vadose zone and steady infiltration (similar to PATHRAE reference case), the black curve is the case with dispersion and transient infiltration, and the dashed curve is for the case with dispersion, matrix diffusion and transient infiltration.

5. SUPPLEMENTAL ANALYSES OF TRANSPORT FOR SELECTED RADIONUCLIDES

We consider steady and transient infiltration with vadose zone dispersivity of 1 cm and 67 cm, for a total of 4 variant simulations. The steady infiltration case with 1 cm dispersivity is similar to PATHRAE, which has steady infiltration and no dispersion in the vadose zone. Peak surface water concentration results for C14, Tc99, and I129 are given in Table 2. Times at which the peaks occur are given in Table 3. For these radionuclides, the effect of vadose zone dispersivity is to reduce the peak concentration and shift the time of peak to later times.

Table 2. Peak concentration for selected radionuclides

	Steady Flow		Transient Flow	
	VZ dispersivity 1 cm	VZ dispersivity 67 cm	VZ dispersivity 1 cm	VZ dispersivity 67 cm
C-14	1.66e6 pCi/l	1.28e6 pCi/l	1.53e6 pCi/l	1.20e6 pCi/l
Tc-99	1.43e6 pCi/l	1.11e6 pCi/l	1.39e6 pCi/l	1.11e6 pCi/l
I-129	6.01e5 pCi/l	4.72e5 pCi/l	6.01e5 pCi/l	4.68e5 pCi/l

Table 3. Time of peak concentration for selected radionuclide

	Steady Flow		Transient Flow	
	VZ dispersivity 1 cm	VZ dispersivity 67 cm	VZ dispersivity 1 cm	VZ dispersivity 67 cm
C-14	Year 928	Year 1037	Year 1463	Year 1581
Tc-99	Year 1162	Year 1327	Year 1709	Year 1871
I-129	Year 2557	Year 2969	Year 3111	Year 3507

Times are in years after closure (failure of liner at year 200).

6. CONCLUSIONS

Three simplifying assumptions in the PATHRAE reference case were relaxed in this analysis, using the same pathway conceptualization but with more flexible modeling tools. Neglect of vadose zone dispersion was found to be conservative or non-conservative, depending on the retarded travel time and the half-life. For a given K_d , a relatively narrow range of half-life was identified for which neglect of vadose zone transport is non-conservative and radionuclide discharge into surface water is non-negligible. That range of half-lives increases with increasing K_d . However, there are two additional conservative simplifications in the PATHRAE reference case that compensate for the non-conservative effect of neglecting vadose zone dispersion: the use of a steady infiltration rate and vadose zone velocity, and the way equilibrium sorption is used to represent transport in the fractured material of the saturated aquifer. With more realistic representations of all three processes, the PATHRAE reference case was found to be either pessimistic (conservative) or to be a reasonably good approximation for all parameter combinations considered.

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