

ESTIMATION OF DILUTION, CAPTURE, AND DOSE FOR IAEA REFERENCE BIOSPHERES

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The calculation of radionuclide concentrations in a receptor well of a generic reference aquifer is highly dependent on the mixing that occurs in the aquifer and well. This study tests two simple analytical approaches for estimating well concentrations: a Generic Aquifer Transport and Capture (GATC) approach and an adapted IAEA ERB1B model. These approaches are tested against a 3-D generic repository model simulated numerically using PFLOTRAN. Because the PFLOTRAN model in this study simulates a diffusive release to the aquifer and the ERB1B model is designed for advective releases, the ERB1B model had to be adapted to accept a diffusive release. The results show that for this PFLOTRAN simulation the GATC equations accurately predict the initial breakthrough and peak concentration at the well but somewhat overestimate concentrations between these points. The adapted ERB1B model only predicts the peak accurately. Similar studies with broader aquifer domains and different types of advective and diffusive releases to the aquifer are needed to fully test the ranges of these analytical approaches.

I. INTRODUCTION

A primary source of exposure in IAEA example reference biosphere (ERB) models for the post-closure safety assessment of geologic repositories is groundwater from an aquifer contaminated by radionuclides that have escaped from the repository.¹ ERB1 assumes that dose is due to human consumption of drinking water from a groundwater well. ERB2A assumes that dose is due to radionuclides in groundwater used for irrigation. These models require as input the well water radionuclide concentrations or an accounting of the effective dilution within the aquifer and well. Effects of residence time and decay within the aquifer, which can be especially important for sorbing radionuclides, should also be included. The ERB models do not include these effects and do not account for the effect of the well's capture zone on dilution.

An earlier paper shows how a simple set of algebraic expressions can be used to directly approximate dilution, capture, residence time, retardation, and decay in a reference aquifer over time so that well water radionuclide concentrations and dose rates that are

consistent with aquifer and well properties can be directly predicted.² This set of equations is named the Generic Aquifer Transport and Capture (GATC) equations and is currently developed for four types of sources: vertical borehole (advective), point source (advective), point source (diffusive), and broad area diffusion. For an advective release of radionuclides to the aquifer, the cross-sectional areas and geometries of the plume and well capture zone are directly approximated by the GATC equations. A dilution factor at the well is calculated by comparing the intersection of the cross-sectional areas. Radionuclide concentrations at the well are calculated from the dilution factor after accounting for travel time, decay within the aquifer, and fraction of plume captured. For diffusive releases to the aquifer, dilution cannot be expressed in terms of a dilution factor because a diffusive flux is not a volumetric rate. In this case, the GATC equations are used to directly calculate the diluted concentrations at the well as a function of the mass flux of diffusive releases to the aquifer, travel time, and decay within the aquifer.²

This paper tests the GATC conceptual model (Section II) and equations (Section III) against the results of a 3-D generic repository performance assessment (PA) model simulated using PFLOTRAN (Section IV). The GATC solutions examined are the point source diffusive (PSD) release model and the broad diffusive (BD) release model. The PFLOTRAN model is a salt repository model where radionuclides diffuse upward through a sealed shaft to an overlying aquifer and then migrate to a well 5 km away.

II. CONCEPTUAL MODELS

The GATC conceptual models are specifically designed for generic repository PA modeling. A typical feature of a generic repository PA model is an overlying aquifer that serves as a primary pathway to the biosphere.¹ The GATC equations are used to calculate overall dilution and radionuclide concentrations in a receptor well over time as a function of the fluxes of radionuclides to the aquifer, sorption, radioactive decay, and the properties of the well and aquifer.² The primary assumptions of the GATC equations are:

- The aquifer has uniform saturated thickness and uniform regional specific discharge, except as altered by a pumping receptor well and/or an advective release to the aquifer.
- The receptor well fully penetrates the aquifer thickness and is screened across the aquifer thickness.
- The receptor well is located down-gradient on the same center streamline as the aquifer release point.

Depending on the type of aquifer release (advective, diffusive, point source, non-point source, etc.), fewer or additional assumptions may be applicable.

II.A. Point Source Diffusive (PSD) Release Model

In the PSD release model of the GATC approach, radionuclides emanate diffusively from a small area within the receptor well's capture zone. This might occur for a repository in salt where diffusion up and through a low-permeability shaft seal may be faster than diffusive transport through the surrounding intact salt. The diffusive mass flux into the aquifer is transported through the aquifer to the well as illustrated in Fig. 1.

Dispersion during transport through the aquifer will cause some contamination of adjacent stream tubes and hence some dilution within the aquifer. Dilution caused by mixing contaminated and uncontaminated stream tubes in the well bore is expected to dominate the overall dilution. Regardless, exactly where the dilution takes place is immaterial because the model simply needs to calculate the final concentrations in the well so that dose rates can be calculated. As shown in Section III, this can be done without estimating dilution within the aquifer.

The PSD release model conservatively assumes that all released radionuclides from a diffusive release point are captured by the well. This assumption implies that no radionuclides will escape the capture zone of the well. It also renders moot the assumption that the receptor well is fully penetrating and fully screened across the aquifer thickness. If the plume is actually broader than the capture zone, concentrations in the well will be overestimated and overall dilution will be underestimated. In such a case, the broad diffusive (BD) release model is more appropriate.

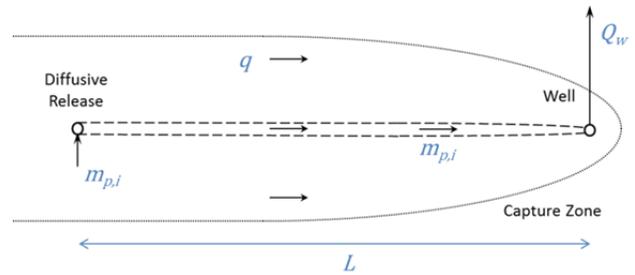


Fig. 1. Point source diffusive (PSD) release model.

II.B. Broad Diffusive (BD) Release Model

For the BD release model, releases are generally spread out over an area larger than the receptor well's capture zone. Such a case could occur for a repository where radionuclides diffuse into an overlying aquifer over an area on the scale of the repository footprint. Alternately, this model may be used when the receptor well has a very low discharge rate and, hence, a very narrow capture zone width compared to the width of a radionuclide plume. This latter case is the case of the PFLOTRAN simulation (Section III). The BD release model is illustrated in Fig. 2.

For the BD release model, vertical dilution within the aquifer is the primary direction of dilution. Mass released to the aquifer is only diluted laterally by spreading across a representative unit width of the aquifer. Vertical dilution is forced by assuming that the length of the well screen spans the entire saturated thickness of the aquifer. If the well does not fully penetrate the aquifer, part of the plume could potentially bypass the well.

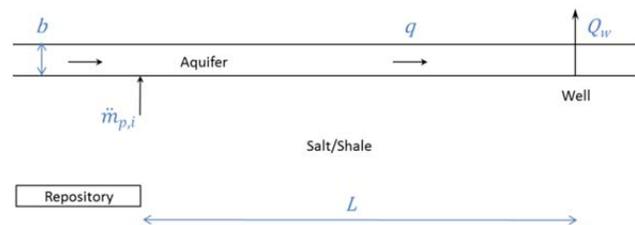


Fig. 2. Broad diffusive (BD) release model.

III. MODEL FORMULATION

For the GATC equations, transport in the aquifer is assumed to be advection-dominated (i.e., the Peclet number is assumed large). The plume is assumed to undergo no dispersion as it migrates through the aquifer toward the receptor well, i.e., radionuclides in the aquifer are assumed to remain within their original stream tubes and migrate as plug flow. Because this assumption prevents transverse and longitudinal spreading of

radionuclides, it prevents attenuation of peak concentrations. Consequently, this assumption ensures that peak dose rates at the well are not underestimated. Preventing longitudinal dispersion will also cause delay in the breakthrough of the leading edge of the plume. In most cases, however, this delay is not expected to be of high concern because initial breakthrough involves low concentrations.

Radionuclide migration through the aquifer and capture at the receptor well is affected by the travel distance, regional darcy flux, sorption, decay, and the size of the well capture zone. The use of each of these parameters is addressed in the following subsections.

III.A. Capture Zone

The plan-view shape of the well's capture zone boundary in an aquifer having constant thickness, b [m], and constant regional specific discharge (darcy velocity), q [m yr⁻¹], is addressed in detail in Mariner (2013) and elsewhere.^{2, 3} The maximum up-gradient width of the capture zone, w_c [m], may be calculated from its relationship to the well discharge rate via the conservation of mass

$$Q_w = w_c b q \quad (1)$$

where Q_w [m³ yr⁻¹] is the well discharge rate. Near the well, the captured streamlines converge and fluid velocities increase, reducing the width of the capture zone.

Although Eq. (1) conceptually represents the capture zone width far from the well, it also can be used to estimate the capture zone width at relatively short distances. For the reference aquifer, the error increases from 1.6% to 5.3% as the distance up gradient decreases from $10w_c$ to $3w_c$.² This error is small compared to the likely spatial variability in q and b in a more realistic representation of an aquifer.

III.B. Sorption and Radioactive Decay

The mean travel time T_i [yr] of radionuclide i to the well from the entry point into the aquifer is estimated as a function of distance, sorption, and regional specific discharge. The GATC model equation for T_i is

$$T_i = \frac{LnR_{f,i}}{q} \quad (2)$$

where L [m] is the travel distance, n [-] is the aquifer porosity, and $R_{f,i}$ [-] is the retardation factor of radionuclide i due to sorption.² For a broad-area diffusive

release, L may be varied over its range to evaluate its effect on predicted dose rates over time.

The GATC equations also account for radioactive decay in the aquifer. The aqueous concentration of radionuclide i in the portion of the plume near the well immediately before the portion enters the well at time t , denoted $C_{nw,i}(t)$ [g L⁻¹], can be approximated from

$$C_{nw,i}(t) = C_{p,i}(t - T_i)e^{-\lambda_i T_i} \quad (3)$$

where $C_{p,i}(t - T_i)$ [g L⁻¹] is the aqueous concentration of radionuclide i released into the aquifer at time $t - T_i$ and λ_i [yr⁻¹] is the decay constant.² Eq. (3) ignores the effects of longitudinal dispersion and ingrowth. Neglecting longitudinal dispersion is addressed at the beginning of Section III.

III.C. Concentrations at the Well

A plume from a point source diffusive release into an aquifer where transport is dominantly advective will only broaden in the aquifer by dispersion. Fig. 1 shows negligible broadening of the plume as it extends to the well. For this plume, the conservative approach is to assume that no radionuclides migrate outside of the capture zone by dispersion.

As noted previously, dilution factors cannot be calculated for diffusive releases, but the effective dilution can still be determined. In the PSD release model, the concentrations at the well are estimated from

$$C_{w,i}(t) = \frac{m_{p,i}(t - T_i)e^{-\lambda_i T_i}}{Q_w} \quad (4)$$

where $m_{p,i}(t - T_i)$ [g yr⁻¹] is the diffusive mass flux of radionuclide i into the aquifer at time $t - T_i$.² Because the entire release in this model is captured in the well discharge, the ratio of the mass flux to the well discharge rate provides a concentration over time that accounts for dilution in the aquifer and well. By using the mass flux at time $t - T_i$, Eq. (4) accounts for travel time through the aquifer. Travel time is a function of sorption and is estimated using Eq. (2). The exponential term accounts for radionuclide decay during transport.

III.D. Broad Diffusive Release and Capture

In the BD release model, the capture zone of the receptor well is smaller than the width of the plume. There is full mixing of all released radionuclides per simulated cross-sectional domain of the well capture zone. The well discharge rate and dilution within the aquifer are immaterial to the calculation of radionuclide

concentrations in the well. For this model, the well concentration over time is calculated from

$$C_{w,i}(t) = \frac{\dot{m}_{p,i}(t - T_i)e^{-\lambda_i T_i}}{qb} \quad (5)$$

where $\dot{m}_{p,i}(t - T_i)$ [$\text{g m}^{-1} \text{yr}^{-1}$] is the diffusive mass flux of radionuclide i into the aquifer per unit width of aquifer at time $t - T_i$.²

III.E. Dose Rate

As described in IAEA (2003), the individual effective dose rate from radionuclide i over time, $H_{E,i}(t)$ [Sv yr^{-1}], can be approximated from

$$H_{E,i}(t) = C_{w,i}(t) * I * dcf_i \quad (6)$$

where I [$\text{m}^3 \text{yr}^{-1}$] is the individual consumption rate and dcf_i [Sv Bq^{-1}] is the ingestion dose coefficient.¹ The value of I is often set at $1.2 \text{ m}^3 \text{yr}^{-1}$, which is the 95th percentile for young adults and approximately twice the mean adult consumption rate recommended by ICRP (1975).⁴ Other exposure pathways are assumed negligible. The committed effective dose per unit intake via ingestion (i.e., the ingestion dose coefficient) for ^{129}I is $1.1 \times 10^{-7} \text{ Sv Bq}^{-1}$.⁵

IV. PFLOTRAN MODEL

For the U.S. Department of Energy's Used Fuel Disposition Campaign, a model of a generic salt repository disposal concept was developed using PFLOTRAN.⁶ The model was developed to demonstrate the capabilities of PFLOTRAN with regard to performance assessment and probabilistic evaluation of the importance of features, processes, and parameters.

The model simulates post-closure radionuclide release and transport from a generic underground repository containing 70,000 MTHM of PWR used nuclear fuel. The repository, depicted in Fig. 3, is located in bedded salt (halite) at a depth of 680 m. The waste packages are placed on the floor of 84 pairs of drifts connected to a central hall. Adjacent drift pairs are located 20 m apart, axis to axis, and waste packages, each 5 m long, are placed 10 m apart, center to center. The drifts are backfilled with crushed salt and sealed with concrete plugs near the central hall. A damaged rock zone (DRZ) extends 12 m beyond the floors, ceilings, and walls of the drifts and central hall. One-meter thick anhydrite interbeds with higher permeability than the halite are located immediately above and below the DRZ. A single central access shaft is located at the center of the repository and is sealed using low permeability materials.

An aquifer lies above the repository at a depth of 435 to 450 m. The aquifer has a porosity of 0.15, a regional specific discharge of $1.58 \times 10^{-9} \text{ m s}^{-1}$, and a well down gradient from the shaft at a distance of 5,805 m.

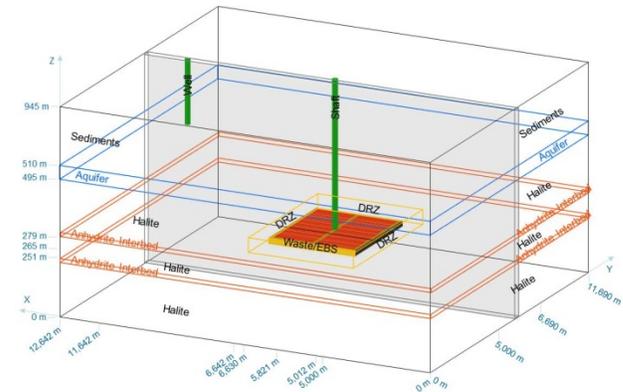


Fig. 3. Generic salt repository conceptual model domain.

The domain simulated in PFLOTRAN does not cover the entire domain of the conceptual model. Instead, it is a 3-D vertical slice through the center of the repository (shown in gray in Fig. 3). The width of the simulated domain is 20 m, extending to the midpoints between the central drift and the two adjacent drifts.

The processes simulated in the PFLOTRAN model include the spatial and temporal flow of heat and water, degradation of the waste form (including instant release fractions), and radionuclide-specific processes, i.e., aqueous solubility limits, sorption, advection, diffusion, decay, and ingrowth. The model is fairly simple and does not include other potentially important features and processes. For example, it does not include explicit representation waste package barriers and degradation processes for these barriers. Instead, the model assumes that waste package barriers do not exist and that at closure all emplaced waste is in contact with groundwater. It also does not simulate infiltration.

The radionuclides included in the PFLOTRAN model are ^{129}I and radionuclides of the Np series. In the model only ^{129}I reaches the aquifer in appreciable quantities. Transport of the other radionuclides is significantly limited by solubility, half-life, sorption, and/or waste degradation. ^{129}I is highly soluble, is assumed not to sorb, has a long half-life ($1.70 \times 10^7 \text{ yr}$), and has an assumed instant release fraction of 11.25%. Because there are no waste package barriers, the instant release fraction causes the simulation to begin with high concentrations of aqueous ^{129}I in groundwater in the vicinity of the waste form. Over the one million year simulation, ^{129}I reaches the receptor well by diffusing up the sealed shaft and into the aquifer where it migrates predominantly by advection

to the well. The calculated mass flux of ^{129}I to the aquifer over time is shown in Fig. 4.

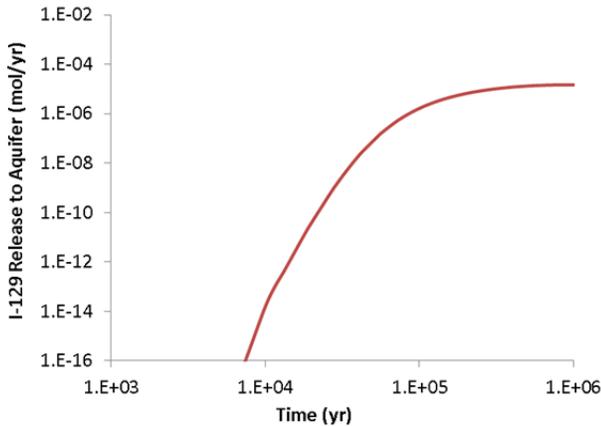


Fig. 4. Mass flux of ^{129}I to aquifer in PFLOTRAN model.

The ^{129}I concentrations at the well were determined in the PFLOTRAN model by numerically simulating advection and dispersion at every location in the aquifer between the shaft and the well. Along the way ^{129}I is allowed to diffuse into overlying sediments and the underlying halite. Fig. 5 shows a snapshot of the dissolved ^{129}I concentrations at 200,000 yr. The aquifer is very thin relative to the vertical extent of the domain, so most of the green band that extends to the left is ^{129}I in the groundwater of the sediments overlying the aquifer. As the figure indicates, a notable fraction of ^{129}I released to the aquifer diffuses into the overlying sediments. The extent of diffusion into the sediments would be reduced if infiltration were included in the simulation.

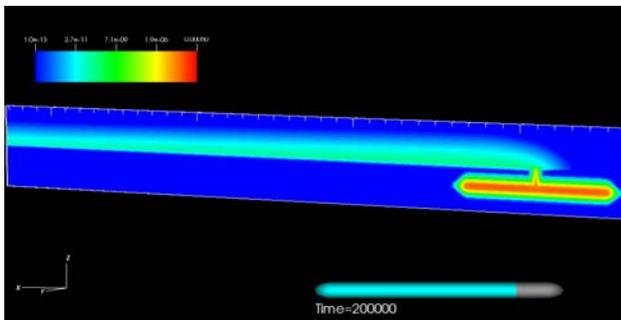


Fig. 5. ^{129}I plume at 200,000 yr in PFLOTRAN model.

For the PFLOTRAN model the reference biosphere is simulated using IAEA ERB1A because the simulation computes the transport of radionuclides through the aquifer to the well. For ERB1A, the geosphere/biosphere interface is the well screen. The only calculation needed for ERB1A is the conversion of well concentrations to

dose rate, i.e., Eq. (6). The results are presented in the Section VII.

V. GATC CALCULATIONS

To test the accuracy of the GATC equations, the PFLOTRAN aquifer was simulated using the BD release model and Eq. (5). The BD release model was chosen over the PSD release model because the well in the PFLOTRAN model is not pumped and therefore has a small capture zone compared to the size of the plume.

For this model, the total diffusive flux of ^{129}I to the aquifer (Fig. 4) was normalized by the width of the model domain (20 m) to calculate $\dot{m}_{p,i}$, the flux into the aquifer per unit width of a quifer. This flux was then offset by travel time T_i to derive the $\dot{m}_{p,i}(t - T_i)$ term of Eq. (5). The travel time for this simulation, calculated from Eq. (2), is 1.75×10^4 yr, which implies a particle velocity of 0.33 m yr^{-1} . Once the concentrations at the well were calculated using Eq. (5), the dose rate was calculated using Eq. (6). Results are presented and compared in Section VII to the results of the PFLOTRAN model.

VI. ERB1B (ADAPTED)

The ERB1B model as presented in IAEA (2003) cannot be used to calculate radionuclide concentrations at the well because it is designed for advective releases. Concentrations are approximated in ERB1B using the following relationship (p. 298)¹

$$C_{nw,i} = \frac{C_{p,i}Q_p}{Q_p + Q_n} \quad (7)$$

where $C_{p,i}$ [g L^{-1}] is the concentration of radionuclide i in the water released to the aquifer, Q_p [$\text{m}^3 \text{ yr}^{-1}$] is the volumetric flux of the contaminated water into the aquifer, and Q_n [$\text{m}^3 \text{ yr}^{-1}$] is the net recharge to the aquifer up-gradient of the region of interest. The ratio $Q_p/(Q_p + Q_n)$ is the effective dilution factor. However, this approach does not work when the flux is diffusive and Q_p is zero.

Eq. (7) can be modified to account for a diffusive release by substituting $m_{p,i}$ for $C_{p,i}Q_p$. Also, because the plume in the PFLOTRAN model is conveniently confined to the cross-sectional area (wb), Q_n can be approximated by qwb . Thus, by adapting to a diffusive flux and to an approximation of the plume cross-sectional area, Eq. (7) becomes

$$C_{w,i} = \frac{m_{p,i}}{qwb} = \frac{\dot{m}_{p,i}}{qb} \quad (8)$$

This equation is similar to Eq. (5) of the BD model, but because it does not account for travel time in the aquifer, it does not account for the delay in breakthrough at the well due to distance (and potentially due to sorption) nor does it account for radionuclide decay during transport.

VII. RESULTS

The concentrations at the well calculated or predicted by the three models are compared in Fig. 6. The secondary y-axis shows the corresponding dose rates. (Regarding the magnitude of the dose rates, recall that the benchmark simulation is an initial simplified model that does not include several engineered barriers and processes that would likely significantly reduce releases and concentrations (Section IV).)

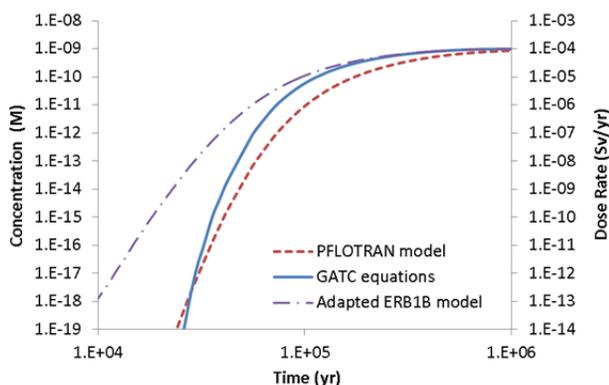


Fig. 6. Comparison of calculations of ^{129}I concentrations at the well and corresponding dose rates over time.

VIII. DISCUSSION

The PFLOTRAN model is more complex than the GATC model in that it computes the transport of radionuclides at all locations in the 3-D aquifer and includes dispersion within the aquifer. In addition, it allows diffusion of radionuclides into the overlying and underlying formations. Because the PFLOTRAN model includes more processes than the analytical models and because the PFLOTRAN code and model have been rigorously tested and reviewed, the results of the PFLOTRAN model serve as the benchmark in this study.

As shown in Fig. 6, the GATC model provides a good approximation of the PFLOTRAN results. The initial breakthrough at the well is predicted accurately and the shape of the curve is closely approximated. The GATC approach overestimates ^{129}I concentrations and dose rate from approximately 30,000 to 1 million years because it does not account for ^{129}I diffusion into overlying and underlying media. Some overestimation of

dose rate may be acceptable because it helps ensure safety. The only time the GATC approach underestimates concentrations and dose rate is during initial breakthrough at the well when concentrations are very low. The underestimation of concentrations at the very leading edge of the plume is due to the exclusion of longitudinal dispersion in the GATC model.

The adapted ERB1B model is less accurate than the GATC model but still does fairly well, partly because ^{129}I in the PFLOTRAN model neither sorbs nor decays appreciably in the aquifer and partly because the narrow PFLOTRAN model boundaries conveniently provide a good estimate of Q_n . Predictions depart notably at early times because the model does not account for travel time in the aquifer. In the ERB1B model, mass released to the aquifer is immediately observed in the well. Also, as in the GATC model, diffusion into overlying and underlying media is assumed not to occur.

IX. CONCLUSIONS

Radionuclide concentrations in a receptor well due to a diffusive release to a generic aquifer can be approximated without numerical simulation. This study shows that for a PFLOTRAN simulation of a generic repository and an overlying aquifer, the GATC equations for diffusive release provided good predictions of initial radionuclide breakthrough and peak concentration at the well. These equations, however, overestimated well concentrations by up to an order of magnitude after breakthrough and before the peak. The primary reason for overestimation during that time is that the equations do not account for diffusion of radionuclides from the aquifer into overlying and underlying formations. This simplification in the GATC equations is conservative.

The ERB1B model could not be used directly for this study because it requires an advective release to the aquifer. The model was therefore adapted for a diffusive release as described in Section VI. The adapted ERB1B model provided a good prediction of the peak radionuclide concentration but a poor prediction of initial breakthrough. It also overestimated concentrations before the peak by not accounting for diffusion into overlying and underlying sediments. The predicted initial breakthrough is too early because the ERB models do not account for travel time through the aquifer. Rather, they assume that radionuclides released to the aquifer immediately enter the well. This simplifying assumption results in significant underestimates in well breakthrough times when the aquifer residence time is long. Long residence times may be due to long distance, low groundwater flow velocities, and/or sorption. In addition, assuming zero residence time will result in significant

overestimates of well concentrations if significant radionuclide decay should occur during aquifer transport.

Modeling issues involving lateral dilution and well capture could not be fully explored in this study because the PFLOTRAN simulation has a modeled aquifer width that is quite narrow (20 m) and a receptor well that is not pumped. Additional studies involving wider simulated domains, wells that are pumped, and different types of advective and diffusive releases are needed to fully test the ranges of the GATC equations and the ERB1B model.

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