Estimation of Contaminant Transport in Groundwater Beneath Radioactive Waste Disposal Facilities

J. C. Wang, J. D. Tauxe, and D. W. Lee

Energy Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee, USA

May 25, 1995

Work sponsored by the U.S. Department of Energy under contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
Estimation of Contaminant Transport in Groundwater Beneath Radioactive Waste Disposal Facilities

J. C. Wang, J. D. Tauxe, and D. W. Lee

Energy Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee

ABSTRACT

Performance assessments are required for low-level radioactive waste disposal facilities to demonstrate compliance with the performance objectives, and consider human exposures from water, air, and inadvertent intruder pathways. Among these, the groundwater pathway analysis usually involves complex numerical simulations with results which are often difficult to verify and interpret. This paper presents a technique to identify and simplify the essential parts of the groundwater analysis. The transport process of radionuclides including infiltration of precipitation, leachate generation, and advection and dispersion in the groundwater is divided into several steps. For each step, a simple analytical model is constructed and refined to capture the dominant phenomena represented in the complex analysis included in a site-specific performance assessment. This step-wise approach provides a means for gaining insights into the transport process and obtaining reasonable estimates of relevant quantities for facility design and site evaluation.
I. INTRODUCTION

Performance assessments (PAs) are required for low-level radioactive waste disposal facilities in order to demonstrate compliance with the performance objectives contained in either 10 CFR 61, Licensing Requirements for Land Disposal of Radioactive Waste, or U.S. Department of Energy Order 5820.2A, Radioactive Waste Management. The purpose of a PA is to provide detailed, site-specific analyses of all credible pathways by which radionuclides could escape the disposal facility and enter the environment. Generally, these assessments consider human exposures from water, air, and inadvertent intruder pathways. Among these, the groundwater pathway analysis usually involves complex numerical simulations with results which are difficult to verify and interpret. This paper demonstrates that the use of simpler analytical models avoids the complexity and opacity of heavily discretized numerical simulations while capturing the essential characteristics and behavior of a site. While not a substitute for a detailed PA, this approach provides insight by breaking the problem down into manageable parts, facilitates the construction of an appropriate conceptual site model, and yields reasonable estimates of relevant quantities for facility design and site evaluation.

In the following sections, we develop an example conceptual model for a DOE low-level waste site, and construct simple, analytical equations to capture the dominant processes. While the processes identified in this example are likely to be present in some form at most sites, there exists no single model which is applicable to all locations, because hydrogeological phenomena are strongly site-specific. The example continues with the presentation of values obtained from the site for the necessary parameters and calculation of dilution factors during the dominant transport processes. For a contaminant with an arbitrary starting concentration, the potential dose to an individual at a down-gradient groundwater compliance location is first compared to a specified dose limit. A back calculation then yields the permissible concentration in the waste which conforms to the dose limit. The concentration changes during contaminant transport from the facility to the compliance point are, therefore, the main quantities to evaluate.
II. CONCEPTUAL MODEL DEVELOPMENT

As the illustrative example for this paper, we use the Radioactive Waste Management Complex (RWMC) site at Idaho National Engineering Laboratory (INEL). The site has been the subject of a PA in which a conceptually straightforward hydrogeological model was proposed. Here we use that conceptual model as the basis for the development of simple mathematical models for contaminant transport.

Radionuclides in the waste disposal facility are carried out of the facility by infiltration of precipitation. Infiltration increases as engineered flow barriers, such as a liner and cap, gradually deteriorate. Leachate produced by the infiltration, if uncollected, flows through the vadose zone and enters the underlying aquifer as contaminated recharge (see Fig. 1). Contaminants travel in the aquifer by advection and dispersion.

Infiltration and leachate generation are complicated processes controlled by factors such as precipitation, waste properties, and facility structures and their deterioration, all of which vary with time and location. Analyzing such a complex problem requires simplifications, such as averaging values over a given time period and/or over a facility area. For example, we assume that for each specified period in the analysis, the infiltration rate through the waste remains constant and is uniform over the area of the facility. This infiltration rate is controlled by the degree of deterioration of the facility structures, and when the structures are fully deteriorated it returns to that of the average regional groundwater recharge.

A simple waste leaching model may be constructed by relating the concentration of a contaminant in the dry waste, $C_w$, and that in the water flux through the waste, $C_i$, by the use of a sorption coefficient $K_w$, commonly applied in a linear form

\[ C_w = K_w^c C_i \quad (1) \]

where $C_w$ is in pCi/g, $K_w^c$ is in mL/g, and $C_i$ is in pCi/mL. (1 Ci = 3.7×10$^{10}$ Bq.) Assuming that the concentration in the leachate emerging from the waste equals that of the equilibrated water within the waste, and that $C_w^e$ is defined as the average concentration in the wetted waste in units of pCi/cm$^3$, then
Figure 1. Schematic diagram of a disposal facility and the underlying groundwater pathway.
where $\theta_w$ is the volumetric water content (volume fraction of the water in the wetted waste) and $\rho_w$ is the dry bulk density of the waste. As can be seen from Eq. (2) the concentration of a contaminant in the leachate is a factor of $\theta_w + \rho_w K_d^w$ different from that in the wetted waste. In this factor, $K_d^w$ varies widely between chemical species and depends on the contaminant concentration, waste composition, and the chemistry of the water contained in the waste. An example of laboratory results for uranium can be found in a report by Collins et al.\textsuperscript{4}

As leachate migrates from a deteriorating facility, it enters the unsaturated (vadose) zone below the facility and above the water table. Unsaturated flow processes are complicated and difficult to describe quantitatively, involving relations among varying water content, suction, and conductivity.\textsuperscript{5} However, since our interest in these processes is mainly to determine the change of contaminant concentration due to radioactive decay (neglecting the dispersion effect), we shall only estimate the contaminant travel time through the vadose zone. Beneath the RWMC lies an extensive series of basaltic lava flows with sedimentary interbeds. Since flow through the vesicular and fractured basalts is difficult to model and probably relatively fast, we can make the conservative assumption (as was done in the RWMC PA\textsuperscript{3}) that the basalt flow contribute little to the residence time of infiltrating water migrating to the aquifer below. Thus, only the cumulative thicknesses of the interbeds are considered in vadose zone travel time calculations. Ignoring the complexities of unsaturated flow, a simple estimate of the average water pore velocity is given by

$$v_v = \frac{I}{\theta_v},$$

(3)

where $I$ is the average infiltration rate and $\theta_v$ is a representative volumetric water content in the vadose zone. According to Eq. (3), the pore velocity is greater than the average infiltration rate because water flows through only that part of the soil which is occupied by water. The travel time for a given contaminant is given by
where \( b_t \) is the thickness from the base of the disposal facility to the underlying water table (Fig. 1), \( b_t/v \) is the travel time for water, and \( R_v \) is the retardation factor for the contaminant in the vadose zone. The retardation factor is defined as

\[
R_v = 1 + \frac{\rho_v K_d^v}{\theta_v},
\]

where \( \rho_v \) is the bulk density of the soil and \( K_d^v \) is the sorption coefficient for the contaminant in the vadose zone. As can be seen from Eqs. (3)-(5), the travel time through the vadose zone depends on the travel distance and the retarded velocity. The latter is controlled by the infiltration rate, the volumetric water content, and the retardation factor. As in the waste, in general, the value of \( K_d^v \) in the vadose zone depends on the chemical species involved and its concentration, the soil composition, and the water chemistry in the soil. Laboratory results for several radionuclides were reported by Seeley and Kelmers.6 A collection of \( K_d \) values for elements in various soils has been compiled by Sheppard and Thibault.7 In our example, we choose uranium as the element of interest, and adopt the \( K_d \) values presented in the RWMC PA.3 In the vadose zone underlying the RWMC, the \( K_d^v \) for uranium is 1000 mL/g.

Contaminants entering the groundwater undergo advection and dispersion during transport toward a down-gradient compliance point. Analytical equations8-10 for the simple case illustrated in Fig. 1 are available for calculating the concentration in the aquifer for an instantaneous release of contaminant mass \( M \) at \( t = 0 \) at the water table. Assuming the facility has a length \( L \) parallel to the groundwater flow direction and a width \( W \), the concentration in the aquifer can be written as

\[
C_a(x,y,t) = \frac{M}{4LWb_t n R_v} \left[ \text{erf}(x+) - \text{erf}(x-) \right] \left[ \text{erf}(y+) + \text{erf}(y-) \right],
\]

with the error functions given by
In these equations, \( x \) is the horizontal distance from the center of the facility to the groundwater compliance point, \( y \) the horizontal displacement perpendicular to the \( x \) direction, \( b \) the thickness of the aquifer, \( v_p \) the groundwater pore velocity, \( n \) the porosity of the aquifer, \( R_s \) the retardation factor for the contaminant in the aquifer, and \( D \) the coefficient of hydrodynamic dispersion, with components \( D_x, D_y, \) and \( D_z \). Equation (6) gives the average concentration at \((x,y,t)\) over the thickness of the aquifer \( b \). For the three-dimensional solution, Eq. (6) is multiplied by a \( z \)-component term:

\[
erf(x_{\pm}) = erf\left(\frac{x_{\pm} - \frac{v_p t}{2R_s}}{\sqrt{\frac{4D_x t}{R_s}}}\right),
\]

and

\[
erf(y_{\pm}) = erf\left(\frac{W_{\pm} y}{2\sqrt{\frac{4D_z t}{R_s}}}ight).
\]

In these equations, \( x \) is the horizontal distance from the center of the facility to the groundwater compliance point, \( y \) the horizontal displacement perpendicular to the \( x \) direction, \( b \) the thickness of the aquifer, \( v_p \) the groundwater pore velocity, \( n \) the porosity of the aquifer, \( R_s \) the retardation factor for the contaminant in the aquifer, and \( D \) the coefficient of hydrodynamic dispersion, with components \( D_x, D_y, \) and \( D_z \). Equation (6) gives the average concentration at \((x,y,t)\) over the thickness of the aquifer \( b \). For the three-dimensional solution, Eq. (6) is multiplied by a \( z \)-component term:

\[
C_a(x,y,z,t) = C_a(x,y,t) \left(1 + 2 \sum_{m=1}^{\infty} \exp\left(-\frac{m^2\pi^2D_z t}{b^2R_s}\right) \cos\frac{m\pi z}{b} \right),
\]

where \( z \) is the depth measured from the water table. Equations (6) and (7) are used in this work to verify and refine some of the simpler analytical models presented in this section, such as the models for the contaminant travel time in the aquifer and the mixing thickness at the groundwater compliance point. In both cases, the total concentration resulting from a continuous release is given by the convolution of the source term and the concentration given by Eq. (6) or (7).

Because of the sorption effect of the aquifer material, the contaminant travels at a retarded velocity \( v_s \) which is related to the mean groundwater pore velocity \( v_p \) by
where $v_D$ is the Darcy velocity. The retardation factor $R_a$ for the contaminant in the aquifer is related to the sorption coefficient $K_d$ in a way similar to Eq. (5):

$$R_a = 1 + \frac{\rho_a K_d}{n},$$

where $n$ is used because the porosity and the volumetric water content are equal in a saturated medium. The dispersion of the contaminant in the aquifer is also retarded by a factor of $R_a$:

$$D' = \frac{D}{R_a}.$$  

The reduction of the contaminant velocity and dispersion rate by the retardation factor $R_a$ also is seen in Eq. (6).

For the purpose of calculating concentration change due to radioactive decay, contaminant travel time in the aquifer from the facility to the compliance point is needed and can be estimated with

$$t_a = \frac{x}{v_a} = \frac{x}{v_p} R_a = t_g R_a,$$  

where $t_g$ is the travel time for groundwater and $x$, as defined before, is the horizontal distance from the center of the facility to the groundwater compliance point. The justification of using $x$ instead of the smallest horizontal distance from the facility to the compliance point is that although the contaminant released at the edge reaches the compliance point first, it takes longer times for the releases at other locations to reach the same point. The validity of Eq. (11) was verified by numerically calculating the convolution of a time-dependent release with a peak and $C_s(x,y,t)$ from Eq. (6), and determining the time for the peak to reach the compliance point.

During the travel time given by Eq. (11), the vertical dispersion in a thick aquifer
results in a mixing thickness \( b_m \) (see Fig. 1) which can be estimated using the diffusion length from diffusion theory by\(^{12}\)

\[
b_m = \sqrt{2D \frac{t_a}{R_a}} = \sqrt{\frac{2D_z}{v_p}} = \sqrt{2D_\ell z_w},
\]

(12)

where the last step suggests that the mixing thickness is independent of \( R_a \) and, therefore, \( K_\ell \). The dispersion coefficient \( D \) can be estimated from the dispersivity \( \alpha \) by\(^{13}\)

\[
D = v_p \alpha,
\]

(13)

neglecting the usually small coefficient of molecular diffusion. The validity of Eq. (12) for our purpose was demonstrated by the use of Eq. (7) and numerical integration as follows: A continuous contaminant release was applied over the facility area at the water table beneath the facility, and the concentration at \( x \) as a function of \( z \) was calculated to estimate the mixing thickness (see Fig. 1). If the mixing thickness calculated with Eq. (12) is greater than the thickness of the aquifer \( b_a \), the latter quantity should be used as the mixing thickness.

Contaminants in the recharge, with a concentration \( C_r \), are diluted on entering the aquifer and during the transport to the groundwater compliance point. By the time the contaminants reach the compliance point, the mixing thickness will have a value \( b_m \) (or \( b_a \) if \( b_m > b_a \)). The dilution can be estimated from

\[
C_a = \frac{ILW}{ILW+WB_m v_D} C_r = \frac{IL}{IL+b_m v_D} C_r,
\]

(14)

where \( I-L-W \) is the recharge flux through the disposal facility area, and \( W-b_m v_D \) is the water flux in the aquifer through the cross section \( W-b_m \) beneath the facility. Eq. (14) suggests that a greater mixing thickness or groundwater flow velocity will yield greater dilution during this part of contaminant transport. In addition, because the width of the facility cancels out in Eq. (14), the relevant facility dimension for dilution is the length \( L \), which is parallel to the groundwater flow direction.
III. APPLICATION TO A SITE

In this section, we shall use an example to demonstrate the methodology described above. To simplify the matter, we assume that uranium is the only contaminant in the waste and a constant concentration $C_w = 10^6$ pCi/g is maintained in the dry waste for the period under consideration. The isotopes being considered are $^{238}$U and $^{235}$U. Both are long-lived, so the concentration change due to radioactive decay is insignificant and is not considered in this example. We shall assume that the hydrogeological setting is similar to that of INEL's RWMC. The facility overlies the Snake River Plain Aquifer formed by highly fractured basalts. Water is assumed to infiltrate at the land surface, contact emplaced waste, percolate downward through the surficial sediments, continue percolating downward through the fractured basalts (which are ignored in the modeled thickness of the vadose zone) and sedimentary interbeds, and finally enter the aquifer. The depth to groundwater is about 180 m, while the total interbed thickness is 12.5 m. It is assumed that at $t=0$, the disposal facility has completely deteriorated and the infiltration $I$ is identical to the regional average groundwater recharge, which is reported to be 0.07 m/y. Values of this and other parameters are summarized as follows:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_w$</td>
<td>$10^6$ pCi/g</td>
<td>(assumed starting concentration)</td>
</tr>
<tr>
<td>$I$</td>
<td>0.07 m/y</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$\theta_w$</td>
<td>0.33</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$\theta_v$</td>
<td>0.168</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$n$</td>
<td>0.10</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$b_v$</td>
<td>12.5 m</td>
<td>(Ref. 3, neglecting ~200 m of basalt)</td>
</tr>
<tr>
<td>$\rho_w$</td>
<td>1.5 g/cm$^3$</td>
<td>(assumed)</td>
</tr>
<tr>
<td>$\rho_v$</td>
<td>1.5 g/cm$^3$</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$\rho_a$</td>
<td>1.9 g/cm$^3$</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$K_d^w$</td>
<td>1000 mL/g</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$K_d^v$</td>
<td>1000 mL/g</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$K_d^a$</td>
<td>100 mL/g</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$L$</td>
<td>257 m</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$W$</td>
<td>122 m</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$x$</td>
<td>228.5 m</td>
<td>(Ref. 3, 100 m from facility)</td>
</tr>
<tr>
<td>$v_p$</td>
<td>560 m/y</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$b_a$</td>
<td>&gt; 100 m</td>
<td>(Ref. 3)</td>
</tr>
<tr>
<td>$\alpha_z$</td>
<td>4 m</td>
<td>(Ref. 3)</td>
</tr>
</tbody>
</table>
With these given values, other quantities are calculated to be:

\[
\begin{align*}
C_1 &= 1000 \text{ pCi/mL} \\
C_w' &= 1.5 \times 10^6 \text{ pCi/cm}^3 \\
v_v &= 0.417 \text{ m/y} \\
R_v &= 8930 \\
t_v &= 2.68 \times 10^5 \text{ y} \\
C_r &= 1000 \text{ pCi/mL} \\
R_\alpha &= 1900 \\
v_D &= 56 \text{ m/y} \\
v_a &= 0.295 \text{ m/y} \\
t_s &= 776 \text{ y} \\
t_{ew} &= 0.408 \text{ y} \\
b_\alpha &= 42.8 \text{ m} \quad \text{(a lower value of 12 m was assumed in Ref. 3)} \\
C_\alpha &= 7.45 \text{ pCi/mL}
\end{align*}
\]

The calculated changes in concentration during transport from the facility to the groundwater compliance point, 100 m down-gradient from the edge of the facility, are summarized in Table 1:

**Table 1. Summary of calculated concentrations in the example.**

<table>
<thead>
<tr>
<th>Location</th>
<th>Notation</th>
<th>Concentration</th>
<th>Explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste</td>
<td>(C_w)</td>
<td>(10^6) pCi/g</td>
<td>Assumed starting value</td>
</tr>
<tr>
<td></td>
<td>(C_w')</td>
<td>(1.5 \times 10^6) pCi/cm(^3)</td>
<td>Eq. (2)</td>
</tr>
<tr>
<td>Water in waste</td>
<td>(C_1)</td>
<td>1000 pCi/mL</td>
<td>Eq. (1)</td>
</tr>
<tr>
<td>Leachate from waste</td>
<td>(C_1)</td>
<td>1000 pCi/mL</td>
<td>Eq. (1)</td>
</tr>
<tr>
<td>Contaminated recharge</td>
<td>(C_r)</td>
<td>1000 pCi/mL</td>
<td>Eq. (6)</td>
</tr>
<tr>
<td>Compliance point</td>
<td>(C_\alpha)</td>
<td>7.45 pCi/mL</td>
<td>Eq. (15)</td>
</tr>
</tbody>
</table>

The overall concentration of uranium changed from \(10^6\) pCi/g \((1.5 \times 10^6\) pCi/cm\(^3\)) in the bulk
waste to 7.45 pCi/mL in the groundwater at the compliance point. The total dilution factor is $1.34 \times 10^5 \text{ (pCi/g)/(pCi/mL)}$, where the unit comes from the different ways of expressing concentrations in waste and in groundwater. The corresponding value extracted from the RWMC PA is $4.5 \times 10^4 \text{ (pCi/g)/(pCi/mL)}$. The difference comes mainly from the fact that a smaller mixing thickness in the aquifer, $b_a = 12 \text{ m}$, was assumed in Reference 3. The dilution factor obtained in the present example has two major parts: $1000 \text{ (pCi/g)/(pCi/mL)}$ from partitioning between the water and solid phases in the waste, and the rest, 134, from the dilution of the contaminated recharge when mixed with groundwater.

If the drinking water protection criterion is interpreted as 4 mrem/y from drinking 2 L/d of contaminated groundwater (as adopted in References 1-3), the activity of uranium in the groundwater at the compliance point should not exceed 0.022 pCi/mL. From Table 1, one finds that the limit for the uranium concentration in the waste at the facility assumed in the example is: $1.34 \times 10^5 \text{ (pCi/g)/(pCi/mL)} \times 0.022 \text{ pCi/mL} = 2900 \text{ pCi/g}$.

IV. DISCUSSION AND CONCLUSIONS

The analysis of human exposures to radioactivity from the groundwater pathway forms an important part of the PA of a low-level waste disposal facility. Because of the complex nature of the contaminant transport process via this pathway, analyses are computationally expensive and provide large amounts of data which are difficult to verify and interpret. In this paper, a step-wise approach using simple analytical models is adopted to capture the dominant physical phenomena represented by the complex calculations. This step-wise approach is not meant to replace the PAs required by government regulations, but to provide a means for gaining insights into the transport process and obtaining reasonable estimates of relevant quantities for facility design and site evaluation.

The conceptual models adopted in this paper are similar to those used in the recent PAs. However, the analytical models constructed for the various steps to estimate the concentration changes are much simpler than the calculations made for these assessments. When the same site specific data are used as inputs, the concentration changes estimated using the technique developed in this paper are generally comparable to those obtained in
the PAs. Corresponding results are usually within an order of magnitude of each other.

From the models presented in this paper, it can be seen that a major concentration change is caused by the partitioning of a contaminant between the waste and the interstitial waters. For a contaminant with a large $K_W^*$, the concentration in leachate can be much smaller than that in the waste [see Eqs. (1) and (2), and Table 1]. As a result, for a given infiltration rate, a contaminant with a greater $K_W^*$ will be released at a slower rate. These results should be taken under consideration when choosing waste form and facility design.

As mentioned in Section II, Eq. (14) suggests that a greater mixing thickness or groundwater flow velocity will yield a greater dilution during contaminant transport in the aquifer. This implies that with other conditions equal, a facility above a more productive aquifer can receive waste with greater contaminant concentration and still comply with groundwater protection requirements. On the other hand, to protect groundwater, one may be reluctant to build a facility above such an aquifer. This is a dilemma which must be rationalized when choosing a disposal site.

For a contaminant with a smaller $K_W^*$, the concentration in leachate is greater [Eq. (1)] and the total release period is smaller. In such cases, leachate collection before significant deterioration of the liners has occurred may remove a large part of the contamination. In our treatment, only vertical dispersion is considered in terms of saturated zone mixing thickness [Eq. (12)]. If the release period is short, the dispersion along the y- and x-direction may result in a greater dilution factor than that shown in Eq. (14).

Concentration changes due to radioactive decay can be estimated from the various travel times and the half-lives of the radionuclides involved. These changes can be included for each part of the transport process by multiplying the resultant concentration at each step with a concentration reduction factor due to decay. For short-lived radionuclides such as $^3$H, this reduction factor can be important in choosing facility design and/or the institutional control duration.

The simple models constructed in this paper are based on the ideal situation shown in Fig. 1. In situations which are different from the one discussed here, modification of the models will be necessary. For example, perched water tables will complicate the unsaturated zone travel time calculations, and the groundwater velocity between the facility and the
compliance point may not be constant due to gradients produced by infiltration or aquifer structure. In those cases the models become less reliable unless proper modifications are made.

ACKNOWLEDGEMENT

This work was sponsored by the U. S. Department of Energy under contract No. DE-AC05-84OR-21400 with Martin Marietta Energy Systems, Inc. The authors wish to thank A. H. Curtis and R. O. Johnson for their critical reading of the manuscript.

REFERENCES


