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The Effect of a Zero-Concentration Sink on Contaminant Transport and Remedial-Action Designs for the Weldon Spring Quarry, Weldon Spring, Missouri

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NOMENCLATURE

The following is a list of acronyms, initialisms, and abbreviations (including units of measure) used in this document.

ACRONYMS, INITIALISMS, AND ABBREVIATIONS

DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
ERFC	complementary error function
ODE	ordinary differential equation
PCB	polychlorinated biphenyl
PDE	partial differential equation
Pe	Peclet number
TNT	trinitrotoluene

UNITS OF MEASURE

d	day(s)
ft	foot (feet)
km	kilometer(s)
L	liter(s)
m	meter(s)
mi	mile(s)
pCi	picocurie(s)
yd	yard(s)
yr	year(s)

FOREWORD

The U.S. Department of Energy (DOE), under its Surplus Facilities Management Program, is responsible for conducting remedial actions at the Weldon Spring site, which is located about 48 km (30 mi) west of St. Louis in St. Charles County, Missouri. The site is included in the National Priorities List of the U.S. Environmental Protection Agency (EPA). The Weldon Spring site became contaminated as a result of processing and disposal activities that took place from the 1940s through the 1960s. The site consists of a quarry and a chemical plant area located about 6.4 km (4 mi) northeast of the quarry. The quarry is surrounded by the Weldon Spring Wildlife Area and is near a well field that constitutes a major source of potable water for St. Charles County; the nearest supply well is located about 0.8 km (0.5 mi) southeast of the quarry. From 1942 to 1969, the quarry was used for the disposal of various radioactively and chemically contaminated materials. As part of overall site remediation, the DOE is proposing to conduct an interim remedial action at the quarry to manage the radioactively and chemically contaminated wastes.

The DOE's currently preferred alternative for managing these wastes is expeditious removal from the quarry with storage at the chemical plant area. Removal of the wastes will permit detailed characterization of the quarry subsurface; detailed characterization is necessary for evaluating the need for follow-up remediation. This report presents the results of solute transport modeling, which will facilitate development of a conceptual hydrogeological model of the quarry area. The hydrogeological model will be useful for developing remedial action plans.

**THE EFFECT OF A ZERO-CONCENTRATION SINK ON CONTAMINANT
TRANSPORT AND REMEDIAL-ACTION DESIGNS FOR THE
WELDON SPRING QUARRY, WELDON SPRING, MISSOURI**

by

David Tomasko

ABSTRACT

One-dimensional analytical expressions are developed to simulate two processes in a homogeneous porous medium: (1) contaminant transport through a porous medium that has a zero-concentration sink located at a finite distance from a step-function source; and (2) contaminant transport through a porous medium that has an initial steady-state distribution corresponding to a constant strength source and zero-concentration sink separated by a finite distance. For general applicability, the governing equations are cast in dimensionless form, making use of the flow system's Peclet number. Evaluation of the analytical expressions is accomplished by numerical inversion of Laplace-space concentrations using either a full Fourier series approach with acceleration, or the Stehfest algorithm; the method used depends on the presence or absence of abrupt discontinuities in the source term. The analytical expressions are used to evaluate possible contaminant conditions at the Weldon Spring quarry near Weldon Spring, Missouri. For the assumed conditions of the calculations and site-specific hydrogeological parameters, the following results have been found: contaminant concentrations should be at or near steady-state conditions; the spatial distribution of contaminants should be a function of the flow system's Peclet number; contaminant concentrations near the Femme Osage Slough, located approximately 213 m (700 ft) south of the quarry pond, should approach zero; contaminant concentrations near the quarry during dewatering and bulk-waste removal should monotonically decrease with time; and the spatial distribution of contaminants during remedial activities should be relatively flat, especially near the dewatering pumps.

Future work will entail evaluating existing radionuclide or chemical concentration data to determine the applicability of the proposed contaminant transport model and to improve the hydrogeological conceptualization of the quarry area and vicinity.

1 BACKGROUND

1.1 OBJECTIVE

The U.S. Department of Energy (DOE) plans to perform a response action at the Weldon Spring quarry near St. Louis, Missouri (Fig. 1.1). Specific activities that are a part of this action can be divided into five areas: (1) removal and treatment of contaminated water from the quarry pond and other sources, (2) removal of existing bulk wastes, (3) potential removal of any residual materials remaining after bulk-waste removal, (4) potential restoration of the groundwater, and (5) potential restoration of surrounding properties. This report presents one-dimensional analytical solutions designed to investigate the potential effects of a zero-concentration sink at the Femme Osage Slough.

1.2 SITE DESCRIPTION

The Weldon Spring quarry is part of a DOE surplus facility located in St. Charles County, Missouri, approximately 6.4 km (4 mi) south of the Weldon Spring Chemical Plant and 48 km (30 mi) west of St. Louis (Fig. 1.1). The quarry is located near Route 94, about 213 m (700 ft) from the Femme Osage Slough. The quarry is 762 m (2500 ft) from the nearest well in the St. Charles municipal well field and 1.6 km (1 mi) from the Missouri River (Fig. 1.2). Originally the quarry was used to supply limestone and sand for construction. The nine-acre site was later used for disposal of soils and other materials contaminated with trinitrotoluene (TNT) during the 1940s and 1950s and the disposal of low-level radioactive wastes during the 1950s and 1960s. The quarry is presently filled with about 72,637 m³ (95,000 yd³) of radioactively or chemically contaminated wastes, including steel drums, concrete and steel rubble, machinery, process residues, uncontained wastes, and contaminated soil.

1.2.1 Geologic Setting

The Weldon Spring quarry is located in low limestone hills near the west bank of the Missouri River. The mid-Ordovician bedrock of the quarry area is predominantly limestone and dolomite. In the vicinity of the quarry, the carbonate rocks dip to the northeast at a gradient of 11 to 15 m/km (60 to 80 ft/mi) (Berkeley Geosciences Associates 1984).

The bedrock is overlain in the upland areas by 3 to 12 m (10 to 40 ft) of silty clay derived primarily from glacial loess and till. Figure 1.3 shows a generalized stratigraphic column for the Weldon Spring site area; Fig. 1.4 shows an idealized geological cross-section for the quarry for a line passing through the quarry in a northwest to southeast direction. As indicated in Fig. 1.4, the walls of the quarry expose the Ordovician Kimmswick Formation, while the bedrock floor of the quarry, presently covered with debris, lies in the upper portion of the Decorah Formation.

FIGURE 1.1 Location of the Weldon Spring Site, Weldon Spring, Missouri (Source: Argonne National Laboratory 1990)

FIGURE 1.2 Surface-Water Features near the Weldon Spring Quarry (Source: Argonne National Laboratory 1990)

The Kimmswick Limestone, mined during quarry operation, is predominantly a crystalline limestone about 21 m (70 ft) thick. It is characterized by solution-enlarged features associated with the intersection of vertical joints and bedding planes.

The Decorah Formation is 6 to 12 m (20 to 40 ft) thick; its upper portion is predominantly composed of fossiliferous limestone with shaley partings (Berkeley Geosciences Associates 1984). The Decorah Formation contains shale beds and is considered to be a leaky confining layer on a regional scale (Kleeschulte et al, 1986). At the quarry, however, its confining capabilities may be limited by the presence of vertical joints and fractures and quarrying operations that penetrated the top 5 m (15 ft) of the formation (MK-Ferguson Company and Jacobs Engineering Group 1989). Underlying the Kimmswick and Decorah formations is the Platin Formation, a thinly bedded, very fine crystalline, dolomitic limestone. Below the Platin Limestone are the Ordovician Joachim Dolomite and the St. Peter Sandstone.

East and south of the quarry (Fig. 1.4), fine- to coarse-grained Quarternary alluvial deposits, sand, and gravels unconformably contact the Platin, Decorah, and Kimmswick formations. Locally, the alluvium is composed of a surficial layer of 3 m

System	Series	Stratigraphic Unit	Typical Thickness (ft)	Physical Characteristics	Aquifer
Quaternary	Holocene	Alluvium	0.5 - 4	Gravelly, silty loam	
	Pleistocene	Loess and Glacial Drift	15 - 55	Silty clay, gravelly clay, silty loam, clay, or loam over residuum from weathered bedrock	
Mississippian	Osagean	Burlington and Keokuk Limestones	100 - 200	Cherty limestone, very fine to very coarsely crystalline, fossiliferous, thickly bedded to massive	Shallow Bedrock Aquifer
		Fern Glen Limestone	45 - 70	Cherty limestone, dolomitic in part, very fine to very coarsely crystalline, medium to thickly bedded	
	Kinderhookian	Chouteau Limestone	20 - 50	Dolomitic, argillaceous limestone; finely crystalline, thin to medium bedded	
Devonian	Upper	Bushberg Sandstone	40 - 55	Quartz arenite, fine to medium grained, friable	
		Lower Part of Sullur Spring Undifferentiated		Calcareous siltstone, sandstone, oolitic limestone, and hard carbonaceous shale	
	Cincinnati	Maquoketa Shale	10 - 30	Calcareous to dolomitic silty shale and mudstone, thinly laminated to massive	
Ordovician	Champlainian	Kimmswick Limestone	70 - 100	Limestone, coarsely crystalline, medium to thickly bedded, fossiliferous and cherty near base	Leaky Confining Layer
		Decorah Formation	30 - 60	Shale with thin interbeds of very finely crystalline limestone	
		Plattin Limestone	100 - 130	Dolomitic limestone, very finely crystalline, fossiliferous, thinly bedded	
		Joachim Dolomite	80 - 105	Interbedded very finely crystalline, thinly bedded dolomite, limestone, and shale; sandy at base	
		St. Peter Sandstone	120 - 150	Quartz arenite, fine to medium grained, massive	
		Powell Dolomite	50 - 60	Sandy dolomite, medium to finely crystalline, minor chert and shale	
		Cotter Dolomite	200 - 250	Argillaceous, cherty dolomite; fine to medium crystalline; interbedded with shale	
Cambrian	Canadian	Jefferson City Dolomite	160 - 180	Dolomite, fine to medium crystalline	Deep Bedrock Aquifer
		Roubidoux Formation	150 - 170	Dolomitic sandstone	
		Gasconade Dolomite	250	Cherty dolomite and arenaceous dolomite (Gunter Member)	
	Upper	Eminence Dolomite	200	Dolomite, medium to coarsely crystalline, medium bedded to massive	
		Potosi Dolomite	100	Dolomite, fine to medium crystalline, thickly bedded to massive; drusy quartz common	

FIGURE 1.3 Generalized Stratigraphic Column (Source: Argonne National Laboratory 1990)

FIGURE 1.4 Idealized Geologic Cross Section for the Weldon Spring Quarry (Source: Argonne National Laboratory 1990)

(10 ft) of silt underlain by about 6 m (20 ft) of sand. The thickness of the silt layer increases toward the river. This water-bearing alluvium is a major contributor to the domestic water supplies of St. Charles County.

The alluvium north of the Femme Osage Slough consists of very fine-grained clays and silts of apparently low hydraulic conductivity and extends downward to the limestone bedrock. South of the slough, the alluvial aquifer becomes thicker and more permeable with correspondingly higher values of hydraulic conductivity and groundwater velocity (MK-Ferguson Company and Jacobs Engineering Group 1989).

1.2.2 Groundwater

Groundwater flow in the vicinity of the Weldon Spring quarry occurs in alluvium, fractured limestone and dolomite, and the St. Peter Sandstone (Berkeley Geosciences Associates 1984) (Fig. 1.3). Water-table conditions (i.e., unconfined aquifers) are typically found in areas of significant alluvial deposits; semiconfined conditions (confined to leaky aquifers) occur where layers of varying permeability are present. Flow in the limestone is primarily through secondary porosity provided by fractures and solution features. The St. Peter Sandstone, about 91 m (300 ft) below the floor of the quarry, contains a confined groundwater aquifer. The degree of connection between the St. Peter Sandstone and the overlying formations is not fully understood.

In the vicinity of the quarry, groundwater flows primarily from north to south; however, water-level measurements taken in 1987 indicate a possible mounding of groundwater at the quarry due to local recharge from precipitation and surface runoff (MK-Ferguson Company and Jacobs Engineering Group 1989) (Fig. 1.5). Even with this local mounding feature, contaminated water from the quarry is expected to flow toward the Femme Osage Slough and St. Charles County municipal well field. The well field is completed in the alluvium near the Missouri River (Fig. 1.2). Near the quarry, flow is likely to take place through secondary porosity features in the fractured limestone formations. Near the well field, flow is through a porous Missouri River alluvium.

Groundwater flow in the alluvial aquifer near the Femme Osage Slough is not well defined (MK-Ferguson Company and Jacobs Engineering Group 1989). Groundwater levels in the bedrock consistently measure about 3 m (10 ft) higher than elevations in the alluvium near the slough. This abrupt change in water level at the interface of the two groundwater regimes indicates a poor hydraulic connection between the limestone bedrock aquifer and the alluvium.

Currently, 28 DOE monitoring wells are located at the Weldon Spring quarry and surrounding properties (Fig. 1.6). Eleven of these wells monitor bedrock water quality at and very near the quarry (MK-Ferguson Company and Jacobs Engineering Group 1989). The remaining monitoring wells (17) are completed in the alluvium/soils above the bedrock, south and east of the slough. In addition, four St. Charles County monitoring wells (RMW-1 through 4) completed in the alluvium are also monitored by the DOE.

FIGURE 1.5 Water Levels Measured near the Weldon Spring Quarry in 1987 (Source: Argonne National Laboratory 1990)

Since 1960, groundwater in the quarry bedrock has been sampled for radiological parameters (MK-Ferguson Company and Jacobs Engineering Group 1989). Total uranium concentration ranges from 3 to 18,700 pCi/L, with the highest concentrations found in the southeast region of the quarry site (MW-1003 [Fig. 1.6] in 1977). Elevated uranium concentrations have been observed in four monitoring wells installed north of the Femme Osage Slough (MW-1013, MW-1014, MW-1015, and MW-1016) in 1987; however, no above-background radionuclide concentrations were found in monitoring wells (MW-1017, MW-1018, and MW-1019) south of the slough (MK-Ferguson Company and Jacobs Engineering Group 1987). These results indicate that contaminated groundwater has migrated into the fine-grained alluvium north of the slough but not beyond it.

The first comprehensive chemical groundwater evaluation for the Weldon Spring quarry was performed in March 1987 (MK-Ferguson Company and Jacobs Engineering Group 1987). Ten monitoring wells in the quarry area were sampled and analyzed for radiological parameters, priority pollutants, pesticides, polychlorinated biphenyls (PCBs), nitroaromatics, metals, and inorganic anions (MK-Ferguson Company and Jacobs Engineering Group 1989). Nitrate levels in excess of drinking water standards were

FIGURE 1.6 Groundwater Monitoring Locations at the Weldon Spring Quarry (Source: Argonne National Laboratory 1990)

observed in monitoring wells MW-1004 and MW-1005; elevated sulfate concentrations were observed in numerous monitoring wells. In general, high-level concentrations of chemicals were found in the same wells that exhibited high radionuclide concentrations.

1.3 PROBLEM

Both hydrological and chemical information from the Weldon Spring quarry and vicinity properties indicate that the Femme Osage Slough is acting as an effective barrier to contaminant transport. In Sec. 2, a one-dimensional analytical solution is developed to investigate the effects of a zero-concentration boundary condition at the slough on upstream contaminant transport and the effects of dewatering the quarry pond.

2 TRANSPORT MODEL

2.1 THEORY: NATURAL FLOW

Freeze and Cherry (1979) present the following partial differential equation (PDE) to represent one-dimensional advection and dispersion of a solute in a porous medium:

$$\frac{\partial C}{\partial t} + V \frac{\partial C}{\partial X} = D \frac{\partial^2 C}{\partial X^2} \quad (2.1)$$

Equation 2.1 can be cast into dimensionless form by making the following substitutions:

$$X = X/X_L \quad (2.2)$$

$$C = C/C_0 \quad (2.3)$$

$$t = \frac{tD}{X_L^2} \quad (2.4)$$

and

$$Pe = \frac{VX_L}{D} = \text{Peclet number} = \text{ratio of advective to dispersive transport} \quad (2.5)$$

where:

X_L = arbitrary scaling distance,

X = longitudinal spatial coordinate,

C_0 = peak concentration at the source,

C = solute concentration,

V = specific discharge (Darcy velocity/effective porosity),

t = time, and

D = coefficient of dispersion.

The resultant dimensionless advection-dispersion equation is

$$\frac{\partial C}{\partial t} + Pe \frac{\partial C}{\partial X} = \frac{\partial^2 C}{\partial X^2} \quad (2.6)$$

Applying the Laplace transform method (Kreyszig 1967) to Eq. 2.6, under an assumed condition of zero initial solute concentration, yields the following result:

$$s\bar{C} + Pe \frac{d\bar{C}}{dX} = \frac{d^2 \bar{C}}{dX^2} \quad (2.7)$$

The general solution to the above ordinary differential equation (ODE) is given by the expression

$$\bar{C} = C_1 e^{\frac{PeX}{2}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} + C_2 e^{\frac{PeX}{2}} e^{\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} \quad (2.8)$$

where C_1 and C_2 are arbitrary constants.

To eliminate the arbitrary constants in Eq. 2.8 in Laplace space, two spatial boundary conditions are required. First, the concentration of the contaminant is assumed to go to zero as X goes to infinity. That is,

$$\lim_{X \rightarrow \infty} \bar{C} = 0 \quad (2.9)$$

Equation 2.8 then reduces to

$$\bar{C} = C_1 e^{\frac{PeX}{2}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} \quad (2.10)$$

A second spatial boundary condition is then applied at the location of the source ($X = 0$). The concentration is assumed to equal C_0 at time zero and remain equal to C_0 until time Δt , at which point the concentration returns to zero. For generality, a time-dependent step function is assumed. Hildebrand (1976) writes this function as

$$\frac{C}{C_0} = U(t - 0) - U(t - \Delta t) \quad (2.11)$$

where U is the unit function.

In Laplace space, the above boundary condition transforms to

$$\bar{C} \Big|_{X=0} = \frac{1}{S}(1 - e^{-\Delta t S}) \quad (2.12)$$

The value of the arbitrary constant C_1 in Eq. 2.10 can then be found by substituting Eq. 2.12 into Eq. 2.10. The result is

$$C_1 = \frac{1}{S}(1 - e^{-\Delta t S}) \quad (2.13)$$

The solution for the dimensionless solute concentration in Laplace space (Eq. 2.10) can then be rewritten as

$$\bar{C} = \left(\frac{1 - e^{-\Delta t S}}{S} \right) e^{\frac{PeX}{2}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + SX^2}} \quad (2.14)$$

The terms of Eq. 2.14 can be inverted using the following relationship presented by Ditkin and Prudnikov (1967):

$$\begin{aligned} L^{-1} \left(\frac{e^{-\sqrt{a(b+S)}}}{S} \right) &= \frac{1}{2} \left[e^{-\sqrt{ab}} \text{ERFC} \left(\frac{1}{2} \sqrt{\frac{a}{t} - \sqrt{bt}} \right) \right. \\ &\quad \left. + e^{\sqrt{ab}} \text{ERFC} \left(\frac{1}{2} \sqrt{\frac{a}{t} + \sqrt{bt}} \right) \right] \end{aligned} \quad (2.15)$$

where ERFC is the complementary error function given by Hildebrand (1976) as

$$\text{ERFC}(\gamma) = 1 - \text{ERF}(\gamma) = 1 - \frac{2}{\sqrt{\pi}} \int_0^\gamma e^{-\lambda^2} d\lambda \quad (2.16)$$

and the following shift identity for Laplace transforms (Hildebrand 1976):

$$e^{-\Delta t S} f(S) = L(f(t - \Delta t)H(t - \Delta t)) \quad (2.17)$$

The solution to Eq. 2.14 in real time and space is then

$$C = \frac{1}{2} \left[\text{ERFC} \left(\frac{1}{2} \sqrt{\frac{X^2}{t} - \frac{Pe^2 t}{4}} \right) + e^{PeX} \text{ERFC} \left(\frac{1}{2} \sqrt{\frac{X^2}{t} + \frac{Pe^2 t}{4}} \right) - \frac{H(t-\Delta t)}{2} \left(\text{ERFC} \left(\frac{1}{2} \sqrt{\frac{X^2}{t-\Delta t} - \frac{Pe^2(t-\Delta t)}{4}} \right) + e^{PeX} \text{ERFC} \left(\frac{1}{2} \sqrt{\frac{X^2}{t-\Delta t} + \frac{Pe^2(t-\Delta t)}{4}} \right) \right) \right] \quad (2.18)$$

where H is the Heaviside function (Hildebrand 1976), such that

$$\begin{aligned} H(t-\Delta t) &= 0 & \text{for } t < \Delta t \\ &= 1 & \text{for } t \geq \Delta t \end{aligned} \quad (2.19)$$

Parameters of interest for the Weldon Spring quarry include the following:

$$X = \frac{x}{L} \quad (2.20)$$

$$t = \frac{tD}{L^2} \quad (2.21)$$

$$\text{and } Pe = \frac{VL}{D} \quad (2.22)$$

The coefficient of dispersion, D , is defined by Bear (1979) as

$$D = \alpha V + D^* \quad (2.23)$$

where D^* is the coefficient of diffusion (assumed to be negligible in the following calculations due to the magnitude of advective flow and dispersion) and α is the dispersivity.

For small distances, dispersivity is scale dependent and given by the following approximation (Lallemand-Barres and Peaudecerf 1978):

$$\alpha = 0.1L \quad (2.24)$$

where L is the distance from the quarry pond to the slough (approximately 213 m [700 ft]).

Using the Lallemard-Barres and Peaudecerf (1978) approximation, the Peclet number for the Weldon Spring Quarry is simply given by the following value:

$$Pe = \frac{VL}{0.1LV} = 10 \quad (2.25)$$

The average linear velocity (Darcy velocity/effective porosity) used in the above relations can range from about 30 to 305 m/d (100 to 1000 ft/d) in the vicinity of the quarry, depending on the characteristics of the porous medium (alluvium or limestone). A base-case value of 91 m/d (300 ft/d) was assumed to be representative for all subsequent calculations. This base-case value is consistent with point-dilution measurements made in the fractured limestone near the quarry (Berkeley Geosciences Associates 1984) and is a reasonable compromise between the high fluid velocities expected in the fractured limestone and the low fluid velocities expected in the floodplain alluvium north of the slough (Tomasko 1989).

Figures 2.1 and 2.2 show the results of calculations performed for step functions of 20- and 30-yr durations. The simplifying assumptions and site-specific hydrological parameters discussed previously were used in the calculations. The longer the duration of the source at $X = 0$, the higher the spatial concentration of the solute. For very long times, t , the pulse of contaminant moves longitudinally along the X -axis toward $X = \text{infinity}$ as demonstrated in Fig. 2.3. The broadening observed in the initial square-wave source term is caused by longitudinal dispersion in the advective direction. The peak amplitude of the contaminant "slug," however, is time invariant due to the assumed conservative nature of the solute (i.e., no nonreversible sorption processes).

The zero-concentration model is constrained to allow the solute concentration to go to zero as the spatial separation goes to infinity. For the case of interest, the Femme Osage Slough located at $X = 1$, it is assumed that the dimensionless solute concentration is held at zero. That is,

$$\lim_{X \rightarrow 1} C = 0 \quad (2.26)$$

For the Femme Osage Slough, the solution to Eq. 2.7 in Laplace space is again given by the general solution presented in Eq. 2.8. The two arbitrary constants that appear in Eq. 2.8 can be eliminated by using spatial boundary conditions at $X = 0$ and $X = 1$.

For a step-function source at $X = 0$, the appropriate boundary conditions are given by

$$\bar{C} \Big|_{X=0} = \frac{1}{S} (1 - e^{-\Delta t S}) \quad (2.27)$$

and at $X = 1$, the Laplace-space concentration is identically equal to zero.

FIGURE 2.1 Concentration versus Distance for a Zero-Concentration Boundary at $X = \infty$;
 $Pe = 10$, $\Delta t = 20$ yr

FIGURE 2.2 Concentration versus Distance for a Zero-Concentration Boundary at $X = \infty$;
 $Pe = 10$, $\Delta t = 30$ yr

FIGURE 2.3 Longitudinal Contaminant Migration as a Function of Space and Time for a Zero-Concentration Boundary at $X = \text{Infinity}$; $Pe = 10$, $\Delta t = 30$ yr

Use of the above two relations in Eq. 2.8 yields the following result:

$$\bar{C} = \frac{e^{\frac{PeX}{2}}}{S} \left(\frac{1 - e^{-\Delta t S}}{1 - e^{\sqrt{\frac{Pe^2}{4} + 4S}}} \right) \quad (2.28)$$

$$\left(e^{\sqrt{\frac{Pe^2 X^2}{4} + SX^2}} - e^{\sqrt{\frac{Pe^2}{4} + 4S}} - e^{\sqrt{\frac{Pe^2 X^2}{4} + SX^2}} \right)$$

A search of standard Laplace transform tables did not produce any simple analytic inversion of the required solution. Applying the Stehfest algorithm (Stehfest 1970a, 1970b) did not produce useful results because the algorithm cannot replicate abrupt amplitude transitions, such as those in the assumed square-wave source term (Davies and Martin 1979). This behavior is demonstrated in Fig. 2.4 for a step function of 10-yr duration calculated with the Stehfest algorithm. Significant, artificial dispersion is introduced, thereby negating the method's applicability.

FIGURE 2.4 Reproduction of a Square-Wave Source Using the Stehfest Algorithm

In 1968, Dubner and Abate introduced the concept of numerically inverting Laplace transforms using the real portion of a Fourier series. This method is particularly well suited for functions with abrupt amplitude transitions. Crump (1976) improved on the method by making use of both the real and imaginary terms of a Fourier series. In this approach,

$$f(t) \approx \frac{e^{ct}}{2T_m} \left(\frac{1}{2} (F(c) + \sum_{K=1}^{\infty} \left[\operatorname{Re} F\left(c + \frac{K\pi i}{2T_m}\right) \cos\left(\frac{K\pi t}{2T_m}\right) - \operatorname{Im} F\left(c + \frac{K\pi i}{2T_m}\right) \sin\left(\frac{K\pi t}{2T_m}\right) \right] \right) \right) \quad (2.29)$$

where:

Re = real part of the complex function,

Im = imaginary part of the complex function,

$c = 5/T_m$, and

T_m = maximum time of the calculation.

In implementing Eq. 2.29, the Laplace variable S is replaced with its complex counterpart:

$$S = c + di \quad (2.30)$$

where:

$$c = 5/T_m,$$

$$d = \frac{K\pi}{2T_m}, \text{ and}$$

$$i = \sqrt{-1}.$$

To implement Eq. 2.29, $F(c)$ and $F(S)$ must be formed from Eq. 2.29. The results are

$$F(c) = \frac{PeX}{c} \left(\frac{1 - e^{-\Delta t c}}{1 - e^{\sqrt{Pe^2 + 4c}}} \left(e^{\sqrt{\frac{Pe^2 X^2}{4} + cX^2}} - e^{\sqrt{Pe^2 + 4c}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + cS^2}} \right) \right) \quad (2.31)$$

and

$$F(S) = F(c + di) = \frac{(c - di)}{c^2 + d^2} e^{\frac{PeX}{2}} \left(\frac{1 - e^{-\Delta t(c + di)}}{1 - e^{\sqrt{Pe^2 + 4(c + di)}}} \left(e^{\sqrt{\frac{Pe^2 X^2}{4} + (c + di)X^2}} - e^{\sqrt{Pe^2 + 4(c + di)}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + (c + di)X^2}} \right) \right) \quad (2.32)$$

where the denominator of Eq. 2.32 has been rationalized.

Substituting Eqs. 2.31 and 2.32 in Eq. 2.29 gives the following complex expression for the concentration in real time and space:

$$C(t) = f(t) \sim$$

$$\begin{aligned} & \frac{e^{ct}}{2T_m} \left(\frac{1}{2} \left(\frac{e^{\frac{PeX}{2}}}{c} \left(\frac{1 - e^{-\Delta t c}}{1 - e^{-\sqrt{Pe^2 + 4c}}} \right) \left(e^{\sqrt{\frac{Pe^2 X^2}{4} + cX^2}} - e^{\sqrt{Pe^2 + 4c}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + cX^2}} \right) \right. \right. \\ & \quad + \sum_{K=1}^{\infty} \left[\operatorname{Re} \left(\frac{(c - di)}{c^2 + d^2} \left(\frac{1 - e^{-\Delta t(c + di)}}{1 - e^{-\sqrt{Pe^2 + 4(c + di)}}}} \right) e^{\frac{PeX}{2}} \right. \right. \\ & \quad \left. \left(e^{\sqrt{\frac{Pe^2 X^2}{4} + (c + di)X^2}} - e^{\sqrt{Pe^2 + 4(c + di)}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + (c + di)X^2}} \right) \right. \\ & \quad \left. \left. \cos \left(\frac{K\pi t}{T_m} \right) - \operatorname{IM} \left(\frac{(c - di)}{c^2 + d^2} \left(\frac{1 - e^{-\Delta t(c + di)}}{1 - e^{-\sqrt{Pe^2 + 4(c + di)X^2}}} \right) e^{\frac{PeX}{2}} \right. \right. \right. \\ & \quad \left. \left(e^{\sqrt{\frac{Pe^2 X^2}{4} + (c + di)X^2}} - e^{\sqrt{Pe^2 + 4(c + di)}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + (c + di)X^2}} \right) \right. \\ & \quad \left. \left. \left. \sin \left(\frac{K\pi t}{2T_m} \right) \right] \right) \right) \right) \end{aligned} \quad (2.33)$$

For computation, terms of the form,

$$e^{\sqrt{r_1 + r_2 i}} \quad (2.34)$$

are inconvenient. Such terms can be managed, however, by making use of the following transformation from Wunsch (1983):

$$e^{\sqrt{X + iY}} = e^{a + ib} \quad (2.35)$$

for

$$a = \pm \frac{\sqrt{X + \sqrt{X^2 + Y^2}}}{\sqrt{2}} \quad (2.36)$$

and

$$b = \pm \frac{\sqrt{-X + \sqrt{X^2 + Y^2}}}{\sqrt{2}} \quad (2.37)$$

(Note: $\pm$ signs on the radicals must be paired [i.e., both must be plus, or both must be negative]. Identical results are obtained for either both plus or both minus.)

The relationship

$$e^{a + ib} \quad (2.38)$$

can be more conveniently managed by using the following property of the complex exponential function (Wunsch 1983):

$$e^{a + ib} = e^a (\cos b + i \sin b) \quad (2.39)$$

A computer program was written to solve Eq. 2.33. Absolute convergence of the solute concentration to within 0.001 was typically achieved by using a K value of 3000. With this many terms in the Fourier series, computation time is lengthy. To eliminate this difficulty, convergence of the solution was accelerated (Macdonald 1964) by employing the epsilon algorithm of Wynn (1956). To decrease computer storage, the algorithm that was developed used a three-column approach with continuous updating. Use of the epsilon algorithm reduced the number of terms in the series to fewer than 100 without significantly affecting numerical accuracy. In addition, to ensure functional smoothness, additional terms in the Fourier series were retained if the inverted function was rapidly varying.

Figure 2.5 shows the dimensionless concentration as a function of time for various X locations for a Pe of 10 and a step-function duration of 20 yr. For X = 0, excellent reproduction of the step-function is seen. At larger separations from the

FIGURE 2.5 Concentration versus Time for a Zero-Concentration Sink at $X = 1$ and $Pe = 10$, Using a Fourier Series Approach

source, a lag time is necessary before contaminant concentration rises above the initial value.

The results of decreasing the Pe from 10 to 5 are shown in Fig. 2.6. At lower Pe values, the lag times are increased; the peak amplitude reached is substantially less than that achieved for higher advective transport (i.e., a higher Pe). Again, reproduction of the step-function source at $X = 0$ is excellent.

Figures 2.7, 2.8, and 2.9 show the buildup of dimensionless solute concentration as a function of spatial coordinate and time for Peclet numbers of 5, 10, and 20, respectively, and a step-function duration of 20 yr. At early time, longitudinal dispersion broadens the contaminant slug profile and the concentration quickly moves into a steady-state condition in which the concentration profile no longer changes with time. As expected, the quickest concentration buildup occurs for the highest Pe .

For steady-state conditions to exist,

$$\frac{\partial C}{\partial t} = 0 \quad (2.40)$$

and

$$Pe \frac{dC}{dX} = \frac{d^2 C}{dX^2} \quad (2.41)$$

FIGURE 2.6 Concentration versus Time for a Zero-Concentration Sink at $X = 1$ and $Pe = 5$, Using a Fourier Series Approach

FIGURE 2.7 Concentration Buildup for $Pe = 5$

FIGURE 2.8 Concentration Buildup for $Pe = 10$

FIGURE 2.9 Concentration Buildup for $Pe = 20$

For a constant strength source at $X = 0$, the solution to Eq. 2.41 is given by

$$\frac{C}{C_0} = \frac{1 - e^{-Pe(X-1)}}{1 - e^{-Pe}} \quad (2.42)$$

Plots of the behavior of C/C_0 versus X for various Peclet numbers are shown in Fig. 2.10. As expected, higher Pe flows produce higher spatial solute concentrations. When Pe equals 50, the steady-state concentration is nearly equal to 1.0 for all X .

Once the step-function source is exhausted, the spatial distribution of concentration decays as shown in Figs. 2.11, 2.12, and 2.13, where the Peclet numbers are 5, 10, and 20, respectively. The higher the Pe , the quicker the flow system returns to initial conditions.

Differences between spatial concentrations calculated with a model that assumes a zero-concentration sink at infinity, and one in which the zero-concentration sink is assumed to be at the slough (702 m [2500 ft]) or the Missouri River (1524 m [5000 ft]) are shown in Figs. 2.14 through 2.17 for 1, 5, 10, and 20 yr, respectively. In all cases, the duration of the step function was assumed to be 20 yr and the Pe equal to 20. At early times (Fig. 2.14), the spatial concentrations of the contaminant are very similar. As time progresses, the concentrations predicted with the second model go to a steady-state profile, while the first model continues to predict transient concentrations with an ever changing profile. After 20 yr, the model-predicted concentrations are again very similar

FIGURE 2.10 Steady-State Solutions for a Zero-Concentration Sink at $X = 1$ as a Function of Space and Pe

FIGURE 2.11 Concentration Decay for $Pe = 5$

FIGURE 2.12 Concentration Decay for $Pe = 10$

FIGURE 2.13 Concentration Decay for $Pe = 20$

FIGURE 2.14 Comparison of Concentration versus Distance for Zero Boundary Conditions at $X = 2500$ ft, 5000 ft, and Infinity at 1 yr; $Pe = 20$, $\Delta t = 20$ yr

FIGURE 2.15 Comparison of Concentration versus Distance for Zero Boundary Conditions at $X = 2500$ ft, 5000 ft, and Infinity at 5 yr; $Pe = 20$, $\Delta t = 20$ yr

FIGURE 2.16 Comparison of Concentration versus Distance for Zero Boundary Conditions at $X = 2500$ ft, 5000 ft, and Infinity at 10 yr; $Pe = 20$, $\Delta t = 20$ yr

FIGURE 2.17 Comparison of Concentration versus Distance for Zero Boundary Conditions at $X = 2500$ ft, 5000 ft, and Infinity at 20 yr; $Pe = 20$, $\Delta t = 20$ yr

(Fig. 2.17); however, the concentrations predicted with the finite-sink locations go to zero at prescribed separations. Differences between the models are also a function of the Pe ; the lower the Pe , the greater the difference in spatial distributions for late-time behavior.

2.2 THEORY: PUMP AT THE QUARRY

As part of the response action planned at the Weldon Spring quarry, the DOE plans to dewater the quarry pond and remove the wastes. During the waste-removal process, pumps installed in the vicinity of the quarry will maintain a negative gradient so that groundwater will flow toward the quarry. This induced flow will inhibit any further migration of contaminants from the quarry toward the St. Charles municipal well field.

In Sec. 2.1, it was shown that a zero-concentration sink at the Femme Osage Slough produces nearly steady-state spatial distributions in a relatively short time for site-specific hydrological parameters. For step functions of 20- to 30-yr durations, such as those likely to have been produced by disposal activities at the quarry in the late 1950s and early 1960s, current solute concentrations should be nearly time invariant.

Remedial pump-back activities at the quarry will, thus, induce contaminant migration toward the quarry from an initial steady-state spatial distribution. In the remainder of this section, a one-dimensional pump-back theory will be developed and the effects of this activity evaluated.

For simplicity, pump-back activities are assumed to produce an instantaneous gradient from the Femme Osage Slough to the quarry pond. It is also assumed that this gradient will produce a constant specific discharge equal to the Darcy velocity divided by the effective porosity.

In a natural coordinate system for the above pump-back geometry, the slough is located at a dimensionless distance, $X = 0$; the quarry is located at $X = 1$. To avoid confusion with Sec. 2.1, calculational results for the pump-back geometry are shown plotted with the same orientation as that used for natural flow conditions, that is, $X = 0 = \text{pond}$, and $X = 1 = \text{slough}$. The initial contaminant concentration is assumed to be equal to the steady-state value achieved under no-pumping conditions. That is,

$$C(t = 0) = \frac{1 - e^{-PeX}}{1 - e^{-Pe}} \quad (2.43)$$

The governing PDE for contaminant migration for the pump-back geometry is the same as that given in Eq. 2.1; however, application of the Laplace transform technique gives the following ODE for the concentration in Laplace space:

$$s\bar{C} - \bar{C}(t = 0) + Pe \frac{d\bar{C}}{dX} = \frac{d^2\bar{C}}{dX^2} \quad (2.44)$$

Equation 2.44 is identical to Eq. 2.7, except for the inclusion of the initial concentration term (Eq. 2.43).

Grouping terms in Eq. 2.44 gives the following nonhomogeneous second-order ODE:

$$\frac{d^2\bar{C}}{dX^2} - Pe \frac{d\bar{C}}{dX} - s\bar{C} = -a + ae^{-PeX} \quad (2.45)$$

where:

$$a = \frac{1}{1 - e^{-Pe}} \quad (2.46)$$

The general solution to Eq. 2.45 is given as the sum of a particular solution (Kreyszig 1967), C_p , and the homogeneous solution, C_h .

$$\bar{C} = C_p + C_h \quad (2.47)$$

Using the variations-of-parameters technique (Kreyszig 1967), the particular solution, C_p , is given by the expression

$$C_p = \frac{a}{s} + \frac{a}{2Pe^2 - s} e^{-PeX} \quad (2.48)$$

The homogeneous solution, C_h , is given by

$$C_h = C_1 e^{\frac{PeX}{2}} e^{\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} + C_2 e^{\frac{PeX}{2}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} \quad (2.49)$$

Adding Eqs. 2.48 and 2.49 gives the following general expression for the contaminant concentration in Laplace space:

$$\begin{aligned} \bar{C} = & C_1 e^{\frac{PeX}{2}} e^{\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} + C_2 e^{\frac{PeX}{2}} e^{-\sqrt{\frac{Pe^2 X^2}{4} + sX^2}} \\ & + \frac{a}{s} + \frac{a}{2Pe^2 - s} e^{-PeX} \end{aligned} \quad (2.50)$$

To solve Eq. 2.50, two boundary conditions are required to eliminate the two arbitrary constants. The first boundary condition occurs at the location of the slough, $X = 0$. At this location, the concentration is again assumed to equal zero. At the quarry, use of the pumps will produce a symmetric dropoff in concentration, like that expected for a zero-flux condition, that is,

$$\left. \frac{\partial \bar{C}}{\partial X} \right|_{X=1} = 0 \quad (2.51)$$

Manipulation of Eq. 2.50 with the two boundary conditions gives the following results for the two arbitrary constants:

$$C_2 = - \left(C_1 + \frac{a}{s} + \frac{a}{2Pe^2 - s} \right) \quad (2.52)$$

and

$$C_1 = \frac{aPe e^{-Pe}}{2Pe^2 - s} + \left(\frac{a}{s} + \frac{a}{2Pe^2 - s}\right) \left(\frac{Pe}{2} - \sqrt{\frac{Pe^2}{4} + s}\right) e^{\frac{Pe}{2}} e^{-\sqrt{\frac{Pe^2}{4} + s}} \quad (2.53)$$

$$\left(\frac{Pe}{2} + \sqrt{\frac{Pe^2}{4} + s}\right) e^{\frac{Pe}{2}} e^{-\sqrt{\frac{Pe^2}{4} + s}} - \left(\frac{Pe}{2} - \sqrt{\frac{Pe^2}{4} + s}\right) e^{\frac{Pe}{2}} e^{-\sqrt{\frac{Pe^2}{4} + s}}$$

Because there are no abrupt discontinuities in the functional form for the contaminant source strength, the Stehfest (1970a, 1970b) algorithm was used to numerically invert Eq. 2.50. With this method, the inverse Laplace transform is obtained using the expression

$$\bar{L}(t) = \frac{\ln(2)}{t} \sum_{i=1}^N V_i L\left(\frac{\ln 2i}{t}\right) \quad (2.54)$$

where, N determines the accuracy of the approximation (machine-dependent value), and the values for V_i are constants dependent on N in the following manner:

$$V_i = (-1)^{\left(\frac{N}{2} + i\right)} \sum_{k=(i+1)/2}^{\max(\frac{N}{2}, i)} \frac{K^{\left(\frac{N}{2} + 1\right)} (2K)!}{\left(\frac{N}{2} - K\right)! (K!)^2 (i - K)! (2K - i)!} \quad (2.55)$$

Figures 2.18 and 2.19 show the results of calculations performed for the pump-back geometry and Peclet numbers of 5 and 10, respectively. Concentrations at the quarry pond ($X = 0$, on the plot) are seen to decrease smoothly in time to zero, with higher Pe values producing faster changes in the concentration. Application of the zero-flux boundary condition is also evident by the symmetrical functional form of the concentration at the quarry pond.

The time-dependent behavior of the contaminant concentration for specified distances is shown in Figures 2.20 and 2.21 for Pe values of 5 and 10, respectively. For all cases examined, the concentration at the quarry pond ($X = 0$, on the plot) monotonically decreases with time to zero. Faster decreases are produced by higher Pe values.

FIGURE 2.18 Pump-Back Concentrations as a Function of Space and Time for $Pe = 5$

FIGURE 2.19 Pump-Back Concentrations as a Function of Space and Time for $Pe = 10$

FIGURE 2.20 Pump-Back Concentrations as a Function of Time and Space for $Pe = 5$

FIGURE 2.21 Pump-Back Concentrations as a Function of Time and Space for $Pe = 10$

3 IMPLICATIONS FOR REMEDIAL-ACTION DESIGNS

In Sec. 2, analytical expressions were developed to evaluate the effect of a zero-concentration sink at the Femme Osage Slough on spatial and temporal distributions for a migrating contaminant in a one-dimensional porous medium. In this conceptualization, spatial contaminant concentrations quickly approach a steady-state distribution that is functionally dependent on the flow system's Pe . For lengthy step functions, the spatial concentrations predicted by the finite-sink model are significantly less than those predicted by a model having a zero concentration at infinity, especially near the location of the slough. Field measurements in the vicinity of the Weldon Spring quarry for conservative solutes, such as nitrate or sorbing solutes with reversible kinetics, can be used to assist in differentiating the two models and developing an improved conceptual model for the quarry vicinity.

During dewatering of the quarry and bulk-waste removal, pumps will be used to maintain a flow toward the quarry. If assuming a zero-concentration sink at the Femme Osage Slough is appropriate, concentrations measured near the location of the quarry pond should decrease monotonically with time to zero and have relatively flat profiles near the location of the pumps. The actual rate of contaminant concentration decrease will be a function of the advective Pe . However, if the slough does not act as a zero-concentration sink, concentrations near the location of the pumps may increase with time as the dispersion-broadened slug of contaminant is brought back toward the quarry. With time, higher contaminant concentrations at selected monitoring wells may reflect a returning contaminant slug rather than an increase due to a perturbation in source strength at the quarry.

Future work entails evaluating existing radionuclide concentration data for the Weldon Spring quarry vicinity to determine the appropriateness of assuming a zero-concentration boundary at the Femme Osage Slough.

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