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DEVELOPMENT OF A ONE DIMENSIONAL SUBSURFACE CONTAMINANT
TRANSPORT MODEL WITH STOCHASTIC APPLICATIONS

by

BENJAMIN WALDON JOHNSON

A THESIS

Presented to the Faculty of the Graduate School of the
MISSOURI UNIVERSITY OF SCIENCE AND TECHNOLOGY

In Partial Fulfillment of the Requirements for the Degree
MASTER OF SCIENCE IN GEOLOGICAL ENGINEERING

2009

Approved by

Dr. Andrew Curtis Elmore, Advisor
Dr. Jeffrey Cawlfeld
Dr. Cesar Mendoza

PUBLICATION THESIS OPTION

The purpose of Sections 1-3 is to provide detail beyond that presented in the journal manuscript which is included on pages 14-39. The paper in this thesis was submitted on March 6th, 2009 as a journal article in the Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management. Section 4 contains supplementary additions to this submittal and has been added for purposes normal to thesis writing.

ABSTRACT

Contaminant source release history is a necessary element of contaminant mass transport simulation in the saturated zone. The source release history is unknown at many sites, and many inverse models have been developed to define finite release histories. To define a likely domain of three source history parameters, a mass balance-based one dimensional approach is developed and tested by discretizing the spatial domain into bands. By incorporating the approach in a spreadsheet model, observed data is represented by both variable length and constant length bands of analysis. Each representation of observed data was tested to determine the most appropriate band geometry for improved model calibration and constant length bands results were more accurate than results of variable length bands. The source history parameter domains predicted by the model are then evaluated with Monte Carlo analysis. After datasets of artificial transport systems are developed and evaluated with the model, actual site data from a Superfund site is input to the model that provides reasonable results of the source history parameters. Limitations of the model require site characteristics that include negligible diffusion, a uniform regional flow field, and isotropic and homogeneous aquifer media.

ACKNOWLEDGEMENTS

I would like to thank Dr. Curt Elmore for his guidance and motivation throughout my research experience at S&T. He has mentored me through the research process and contributed to my professional development.

I am also grateful for Dr. Jeff Cawlfieid's technical assistance and perspective throughout the development and completion of this research.

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1. INTRODUCTION

1.1 GROUND WATER FLOW AND MASS TRANSPORT CONCEPTS

Ground water is a valuable resource that acts as a primary drinking water source for many Americans. Ground water flow calculation is often necessary to characterize a drinking water source, measure the extent of a polluted aquifer, or perhaps evaluate other ground water parameters of interest. Fetter (2001) evaluates average linear ground water velocity v_x with Darcy's law as

$$v_x = -\frac{K}{n_e} \frac{dh}{dl} \quad (1)$$

where

K = hydraulic conductivity

n_e = effective porosity

$\frac{dh}{dl}$ = hydraulic gradient

Diffusion of subsurface contaminants involves the molecular interaction between ground water and contaminants where higher concentrations of contaminants migrate to areas of lower concentrations (Fetter 2001). This form of contaminant transport is often subordinate to advective transport. Spitz and Moreno (1996) describe advective contaminant transport as the mass transport caused by the movement of groundwater flow. When isotropic homogeneous aquifer properties are present, Fetter (2001) and other texts present the governing equations of advective contaminant transport with Darcy's law in Equation 1.

Contaminant retardation impedes the actual contaminant velocity from transporting at the same rate of ground water velocity. Spitz and Moreno (1996) define retardation R as

$$R = 1 + \frac{(1 - n)\rho_s}{n} K_{oc} f_{oc} \quad (2)$$

where

n = porosity

ρ_s = density of dry matrix material in units of mass per volume

K_{oc} = organic carbon partition coefficient for an organic solute under consideration in units of volume per mass

f_{oc} = weight fraction of organic carbon in soil material

1.2 SUBSURFACE CONTAMINANT MODELING CONCEPTS

Numerical analysis of subsurface contaminant transport is often required to solve problems encountered by Remediation Engineers, and spreadsheet software applications are commonly used among practitioners for transport system evaluation. Typical point source subsurface contaminant modeling involves a release from a given location at a known concentration and known aquifer parameters. In practice however, these parameters are often approximated with a range of values or they are unknown. At many sites, monitoring wells are used to collect observed data to characterize contaminant plumes and estimate the extent of the contaminant system. When the source parameters are unknown, inverse modeling with observed data is a method to approximate the transport system history parameters. Instead of rigorously defining the source history, the method presented in this paper involves mass balance-based inverse modeling that provides a likely domain of source history parameters, which is developed and tested

with both synthetic and real data. The source history parameters of interest include the initial concentration C_0 , the time since the contaminant release began t_r , and the elapsed time of the contaminant release t_s . The method is applicable at sites where the Dupuit theory of equipotential lines may be assumed to describe hydraulic gradient of the phreatic surface and the assumption of isotropic and heterogeneous aquifer media is appropriate.

The Van Genuchten and Alves (1982) solution to the advection dispersion equation for contaminant transport given a rectangular source release of magnitude C_0 for t_s is

$$C(x, t_r) = C_0 \left(\frac{v_x}{v_x + w} \right) \exp \left(\frac{(v_x - w) \cdot x}{2 \cdot D_x} \right) \operatorname{erfc} \left(\frac{R \cdot x - w \cdot t_r}{2 \cdot \sqrt{D_x R} \cdot t_r} \right) \quad (3)$$

for $0 < t_r \leq t_s$

where

$$w = v \cdot \sqrt{1 + \frac{4kD_x R}{v^2}}$$

t_s = time of the contaminant release

and

$$C(x, t_r) = C_0 \left(\frac{v_x}{v_x + w} \right) \exp \left(\frac{(v_x - w) \cdot x}{2 \cdot D_x} \right) \operatorname{erfc} \left(\frac{R \cdot x - w \cdot t_r}{2 \cdot \sqrt{D_x R} \cdot t_r} \right) \quad (4)$$

$$- \operatorname{erfc} \left(\frac{R \cdot x - w \cdot (t_r - t_s)}{2 \cdot \sqrt{D_x R} \cdot (t_r - t_s)} \right)$$

for $t_r \geq t_s$

where t_r is the time since the start of the release. The solution is based on the assumptions that advection and dispersion occur in a uniform regional flow field with constant ground water velocity and homogeneous, isotropic aquifer properties.

Model calibration of inverse model applications is often necessary to provide a reasonable match of observed and predicted data. One method described by Spitz and Moreno (1996) involves a trial and error approach that requires input parameter variation to reduce differences between observed and predicted data. Model error is recorded over a large input parameter domain to locate regions of model accuracy improvement. In practice, modelers often employ automated iteration software to perform otherwise tedious calculation to conduct this procedure.

Calibration accuracy may be assessed with several calibration parameters. Spitz and Moreno (1996) recognize several calibration parameters to assess the goodness of fit between predicted and observed data that include the root mean square error $RMSE$, the coefficient of determination CD , the modeling efficiency EF , and the coefficient of residual mass CRM where

$$RMSE = \left[\sum_{i=1}^n \frac{(P_i - O_i)^2}{n} \right]^{1/2} \left[\frac{100}{\bar{o}} \right] \quad (5)$$

$$CD = \frac{\sum_{i=1}^n (O_i - \bar{o})^2}{\sum_{i=1}^n (P_i - \bar{o})^2} \quad (6)$$

$$EF = \frac{\sum_{i=1}^n (O_i - \bar{o})^2 - \sum_{i=1}^n (P_i - O_i)^2}{\sum_{i=1}^n (O_i - \bar{o})^2} \quad (7)$$

$$CRM = \frac{\sum_{i=1}^n O_i - \sum_{i=1}^n P_i}{\sum_{i=1}^n O_i} \quad (8)$$

where

O = observed value

\bar{o} = mean observed value

P = predicted value

n = number of values

After the model calibration is complete, model verification with observed physical data is used to improve the confidence of the calibration results. For contaminant transport systems, one verification method described by Spitz and Moreno (1996) requires actual field data to be incorporated into the model and contaminant concentrations are predicted at a future time. These predicted values are then compared with actual data collected on the future date used to predict concentration data.

1.3 STATISTICAL CONCEPTS

Summary statistics of sampled datasets that include the median \tilde{X} , the mean \bar{X} , the standard deviation s , and the geometric mean x_{geo} are useful to assess subsurface contaminant transport parameters. These variables are defined by Hensel and Hirsch (2002) as

$$\tilde{X} = X_{(n+1)/2} \quad \text{when } n \text{ is odd} \quad (9)$$

$$\tilde{X} = \frac{1}{2} \left(X_{(n/2)} + X_{(\frac{n}{2}+1)} \right) \quad \text{when } n \text{ is even} \quad (10)$$

$$\bar{X} = \sum_{i=1}^n \frac{X_i}{n} \quad (11)$$

$$s = \sqrt{\sum_{i=1}^n \frac{(X_i - \bar{X})^2}{n-1}} \quad (12)$$

$$x_{geo} = \sqrt[n]{\prod_{i=1}^n (X_i)} \quad (13)$$

where

n = total number of observations in the sample dataset

X_i = the i^{th} observed data value in the sample dataset

The coefficient of variation COV is another useful transport modeling statistic that estimates the relative standard deviation of a dataset as defined by Gilbert (1987) where

$$COV = \frac{s}{\bar{X}} \quad (14)$$

Ground water datasets with small sample sizes pose challenges for engineers to evaluate and compare data. Hensel and Hirsch (2002) recommend testing a dataset for normality prior to statistical comparison of the dataset to other datasets with parametric statistical tests that assume normal distributions for each dataset. If normality of a dataset is rejected, non-parametric statistical comparison is recommended. A common parametric statistical test used to compare two datasets is the student's t-test. According to Navidi (2006), the two tailed student t-test statistic t of two groups with unequal variances is calculated as

$$t = \frac{(\bar{X} - \bar{Y})}{\sqrt{\frac{S_X^2}{n_X} + \frac{S_Y^2}{n_Y}}} \quad (15)$$

where

\bar{X} = the mean value of dataset X

\bar{Y} = the mean value of dataset Y

s_X = the sample standard deviation of sample data within dataset X

s_Y = the sample standard deviation of sample data within dataset Y

n_X = the number of sample data within dataset X

n_Y = the number of sample data within dataset Y

The degrees of freedom df for the student's t-test is approximated as

$$df = \frac{\left(\frac{s_X^2}{n_X} + \frac{s_Y^2}{n_Y}\right)^2}{\frac{(s_X^2/n_X)^2}{n_X - 1} + \frac{(s_Y^2/n_Y)^2}{n_Y - 1}} \quad (16)$$

and rounded down to the nearest integer. The null hypothesis H_0 is defined as not having significant difference between the two datasets X and Y . To reject H_0 at an arbitrary level of confidence α , the following criteria must be met where

$$t > t_{\left(\frac{\alpha}{2}\right), (df)} \quad (17)$$

where

$t_{\left(\frac{\alpha}{2}\right), (df)}$ = the critical student's t-value obtained from a t distribution table at specified values of $\alpha/2$ and df

When comparing two non-normally distributed datasets, Hensel and Hirsch (2002) note that the Mann-Whitney nonparametric test has more power than the student's t-test to recognize significant difference between two independent datasets. According to Navidi (2006), the values of both groups of data are combined and ordered from smallest to largest and assigned a rank from one to $n_Y + n_X$. The larger dataset is denoted as Y_1, Y_2, \dots, Y_{n_Y} and the dataset with the smaller sample size is denoted as X_1, X_2, \dots, X_{n_X} . Next, the Mann-Whitney test statistic W is calculated by summing the ranks

corresponding to the each value within the X dataset. The Mann-Whitney H_0 is defined as not having significant difference between the two datasets and the p-value is determined from a table of Wilcoxon rank-sum critical points. Navidi (2006) defines the p-value as a measurement of H_0 plausibility where smaller p-values suggest more evidence to reject H_0 . Therefore, if the p-value is less than the chosen level of α , H_0 can be rejected.

2. EXPERIMENTAL DESIGN AND WORK NARRATIVE

The initial phase of experimental design involved the Dawson (1995) model, which predicted C_0 at coordinates of t_r and t_s . Bands of analysis were incorporated over a contaminant system modeled with Dawson's model to determine the most appropriate band geometry that led to best model calibration improvement. Microsoft Excel with Visual Basic Application was used as the spreadsheet model software.

Synthetic contaminant system development required generation of artificial datasets that accurately represented a rectangular pulse release transport system. Known values of C_0 , t_r , and t_s were input to the model and 1,000 concentrations were produced in spatial increments of five meters over a 5,000 meter contaminant plume. Subsets of 59 (C, x) data points were chosen with a uniform random number generator to develop uniformly random spatial distributions of observed concentrations along the artificial transport system. This process was repeated three times at different known values of C_0 , t_r , and t_s to synthesize three datasets illustrated in Figures 1-3 that each represent a synthesized rectangular pulse release contaminant transport system.

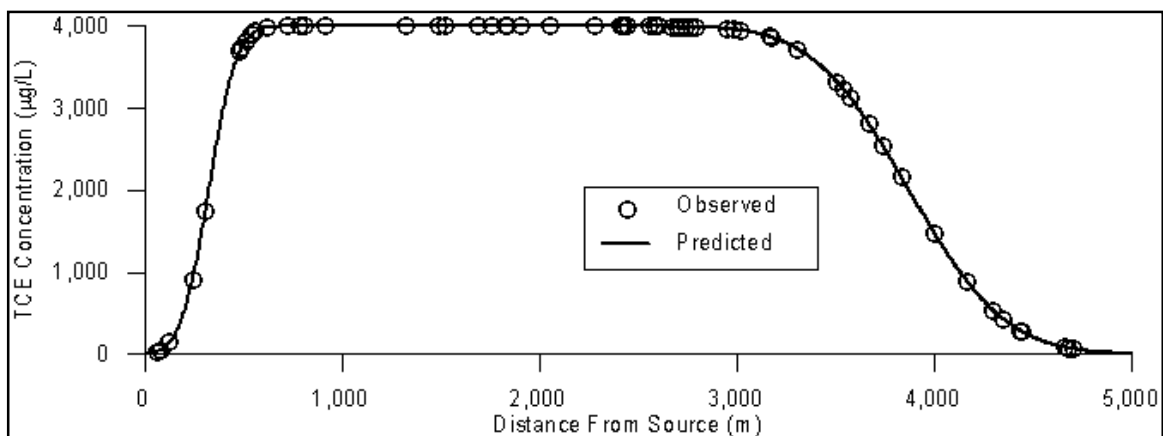


Figure 2.1: Simulated Data where $t_r = 24$ and $t_s = 22$

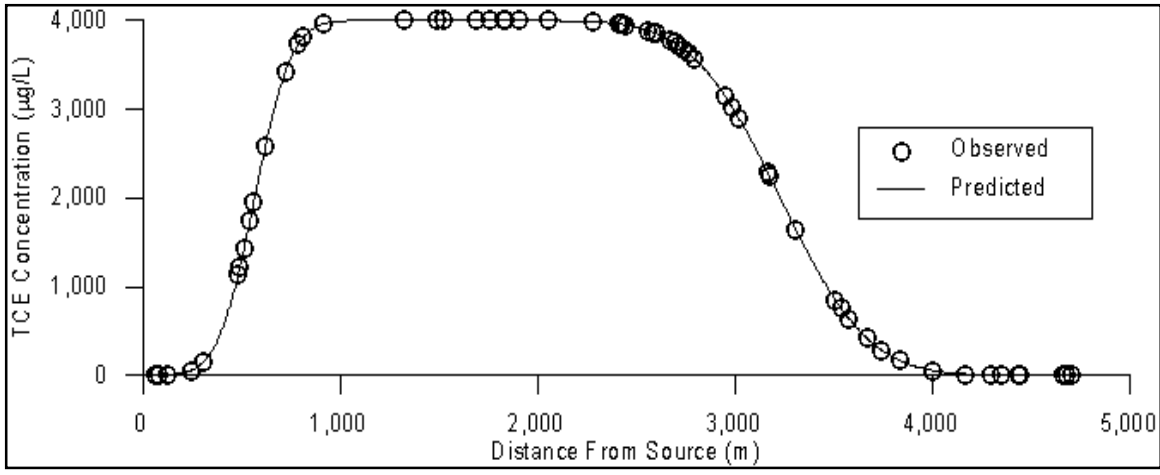


Figure 2.2: Simulated Data where $t_r=20$ and $t_s=16.5$

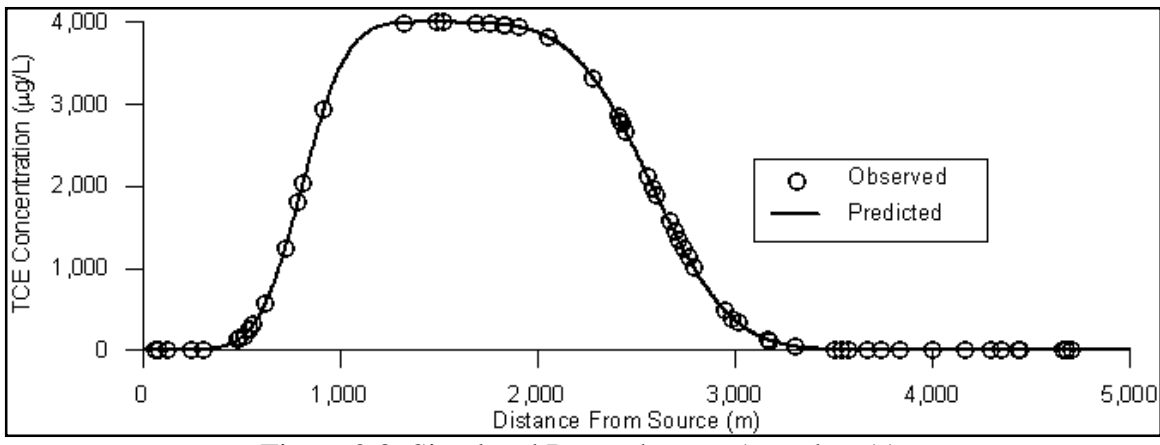


Figure 2.3: Simulated Data where $t_r=16$ and $t_s=11$

Each synthetic contaminant transport system was overlaid by rectangular bands of analysis to summarize observed data along the plume length. Initially, variable length bands were expected to reduce observed data variability by manually adjusting their lengths parallel to the direction of contaminant transport. However, when constant length bands were used, the model calibration results improved.

Spreadsheet automation with Visual Basic for Applications was incorporated into the Microsoft Excel spreadsheet model to develop contours of calibration parameters

described in Equations 6-9 that would otherwise not be feasibly manually calculated. A broad region of t_r and t_s was defined where t_r varied between eighteen and twenty-three years and t_s varied between eleven and sixteen years. Increments of 0.2 years along the ranges of t_r and t_s were defined and values of each calibration parameter was recorded at each time value set of t_r and t_s . The calibration parameter most sensitive to the actual values of t_r and t_s was *RMSE*. Time value pair locations at which *RMSE* was most reduced were within 10% of the actual values of t_r and t_s in each simulation of the three synthetic transport systems.

Monte Carlo (MC) simulation was used for stochastic evaluation of a reasonable domain of the source history time parameters t_r and t_s . Elmore (1996) describes this technique as a useful tool to evaluate the uncertainty of solutions developed with deterministic methods in water resources. Different sets, or realizations, of random independent input variables with assumed log normal distributions are input parameters to a set of governing equations. The ensemble of output parameters are randomly distributed and statistically summarized to predict likely values of t_r and t_s within the domain of t_r and t_s .

Input into the MC model included two coordinates of t_r and t_s that characterize a rectangle used to approximate a contour of *RMSE*. Additional input included the rectangle's length and width, which identify range boundaries for the realization parameters described below and illustrated in Figure 4. Because the contours are best approximated with a rotated rectangle illustrated in Figure 4, rotation/translation equations were developed to input the corresponding values of t_r and t_s into the spreadsheet model as

$$t_r = y'_1 + (X^2 + Y^2)^{1/2} \sin \left[\tan^{-1} \left(\frac{X}{Y} \right) + \tan^{-1}(m') \right] \quad (18)$$

$$t_s = x'_1 + (X^2 + Y^2)^{1/2} \cos \left[\tan^{-1} \left(\frac{X}{Y} \right) + \tan^{-1}(m') \right] \quad (19)$$

where

y'_1 = the value of t_r along the y-axis corresponding to the rectangle's lowest corner

x'_1 = the value of t_s along the x-axis corresponding to the rectangle's lowest corner

X = the rotated x-axis variable

Y = the rotated y-axis variable

and

$$m' = \frac{y'_1 - y'_2}{x'_1 - x'_2} \quad (20)$$

where

m' = the offset slope of the rectangle

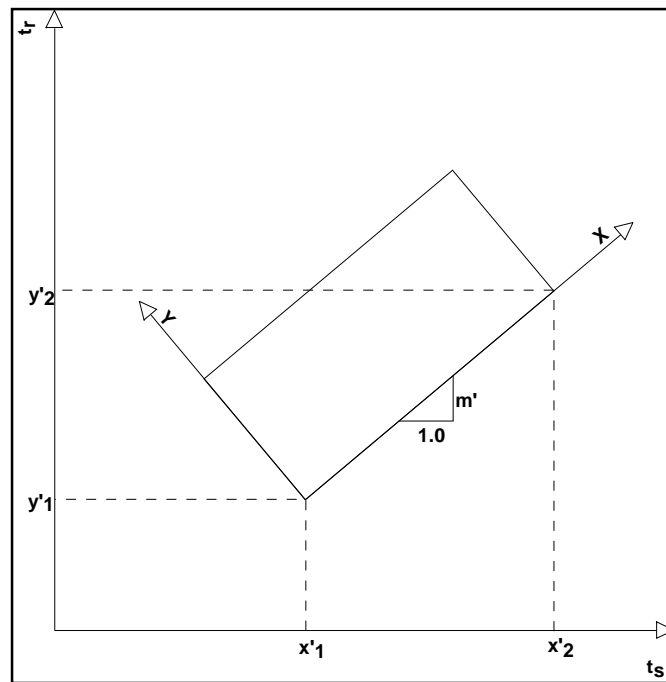


Figure 2.4: Illustration of translation/rotation equation variables

3. OBJECTIVES AND GOALS

The objective of this research was to develop a one dimensional model with Microsoft Excel that provides reasonable estimates of source history parameters in a contaminant transport systems when certain site criteria are met. This approach provides a point of departure for more sophisticated modeling application and reduces the time requirement for modelers to characterize contaminant systems with more complex methods of software evaluation. The model was developed in Microsoft Excel because of its low cost and practitioner familiarity.

The method of analysis band application to summarize observed data was hypothesized to affect model calibration. Both variable length bands and constant length bands were rigorously evaluated to determine which approach would provide more accurate source history parameter estimates. Upon achieving acceptable source history parameter accuracy with synthetic transport systems, the model was applied at a Nebraska Superfund Site to assess an actual contaminant transport system.

PAPER**APPLICATION OF A MASS BALANCE-BASED STOCHASTIC TRANSPORT MODEL**

CE Database Subject Headings: environmental engineering, ground-water flow, mass transport, remedial action, design

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Abstract

The definition of contaminant source release is a necessary element of contaminant mass transport simulation in the saturated zone. At many sites the release history is unknown, and there has been a significant body of research to develop inverse models to define finite release histories. Instead of rigorously defining the source history, a mass balance-based approach is tested to explicitly account for uncertainty in rectangular pulse release variables. The approach has been incorporated into a spreadsheet model which uses a one-dimensional solution to the advection dispersion equation, which readily lends itself

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to Monte Carlo applications. After the model was tested with synthetic datasets, the model was calibrated and verified using a concentration dataset collected at a Superfund site. Model calibration with synthetic and actual data resulted in a reasonable domain of source history parameters and the model provided reasonable results when the mass of contaminant in the aquifer was assumed to be random.

Introduction

The classic approach for modeling mass transport in a groundwater system requires initiating the simulation with the release of the contaminant to the system. Thus the conceptual model and subsequent analytical and numerical transport models require characterization of the source location and the source release function. There is a significant body of literature that addresses the identification of groundwater contamination sources as summarized by Michalak and Kitanidis (2004), Sonnenborg, et al. (1996), and Hill and Tiedeman (2007). Some representative works include Domenico and Robbins (1985) which developed the extended pulse model to identify unknowns including the source concentration, source dimensions, the time of when the contaminant first entered the groundwater, the position of the center of mass, and the groundwater dispersivities. The authors acknowledged that unique solutions were not likely using field data although the model was useful for placing reasonable bounds on the unknowns. Ala and Domenico (1992) developed a two-dimensional analytical method called the two-well solution to find the unknowns associated with a finite plane source. Skaggs and Kabala (1994) evaluated complex contaminant release histories (that is, releases other than continuous or rectangular pulses) using Tikhonov regularization. One of the conclusions of their work was that plume history could be adequately recovered if

dispersion was relatively minor compared to advection even if the concentration measurements contained moderate random errors. A similar conclusion was developed by Alapati and Kabala (2000) who used a non-linear least-squares method without regularization to recover the release history of a source with a known location. Skaggs and Kabala (1998) also provide limitations when developing the release history of a source with observed data. These include the time since the release, which can be better estimated as the actual time since the release decreases. They show that deconvoluting solute transport systems is an ill-posed problem with solutions that improve as the quality of observed data improves. This sensitivity poses a significant issue to pragmatic application of contaminant release history characterization with field datasets. Newman and de Marsily (1976) show that applying least squares to an ill-posed problem will likely contain severe oscillations because of the problem's sensitivity to observed measurement error.

Elmore (2007) used concentration data collected using a direct push drill rig to estimate a subset of the source parameters including the time since the source release t_r , the duration of the release t_s , and the concentration of the source C_0 , for a rectangular pulse. That work introduced a one dimensional (1D) spreadsheet model that could be used to estimate t_r , and t_s , by manual iteration with C_0 as a dependent variable assuming that the source location was known. Much of the previous work focused on developing finite definitions of source history. The focus of this paper is to explicitly account for source history uncertainty through the development and application of stochastic mass transport model without necessarily identifying a single finite source history. The approach initially developed in Elmore (2007) will be expanded to develop a mass-balance basis to

determine a reasonable domain of t_r , t_s , and C_0 with a solution developed with iterative model calibration automation using synthesized observed data. The approach is then applied using site concentration data from a Nebraska Superfund site.

Methodology

Model Development

The governing equation for mass advection, dispersion, degradation, and sorption in a one-dimensional flow field is

$$R \left(\frac{\partial}{\partial t} C \right) = -v_x \cdot \left(\frac{\partial}{\partial x} C \right) + D_x \cdot \left(\frac{\partial^2}{\partial x^2} C \right) - k \cdot R \cdot C \quad (1)$$

where C is the solute concentration, t is time, v_x is the average pore water velocity in longitudinal direction of flow, x is longitudinal distance from source, D_x is the longitudinal dispersion coefficient, and k is the net degradation rate constant. The retardation factor for linear, equilibrium sorption R is defined by Spitz and Moreno (1996).

The Van Genuchten and Alves (1982) solution to Equation 1 was incorporated into a spreadsheet application developed by Dawson (1995) given a rectangular source release of magnitude C_0 for t_r and t_s . The solution is based on the assumptions that advection and dispersion occur in a uniform regional flow field with constant ground water velocity and homogeneous, isotropic aquifer properties.

A dependent relationship between the mass release variables C_0 and t_s may be developed using a mass balance approach by assuming that the groundwater plume has constant rectangular dimensions with width W , thickness b , and length x_{max} , as defined by Elmore (2007). The plume is then divided into analysis bands oriented normal to the

direction of flow. Given a constant concentration C_j for each band, which is the arithmetic average of predicted concentrations in band j , the total mass in solution in the aquifer is

$$M = n \cdot W \cdot b \left[\sum_{j=1}^m (C_j \cdot L_j) \right] \quad (2)$$

where n is the aquifer porosity, m is the total number of analysis bands, and L is the length of each band parallel to the direction of flow. The normalized total mass in the aquifer is defined as

$$M' = \sum_{j=1}^m (C_j \cdot L_j) \quad (3)$$

where

$$M' = \frac{M}{n \cdot W \cdot b}$$

Spitz and Moreno (1996) give the following equation for the total contaminant mass per unit aquifer volume

$$\Delta M = C \cdot n + C_a \cdot (1 - n) \cdot \rho_s \quad (4)$$

where C_a is the adsorbed concentration measured as mass of contaminant per mass of dry aquifer matrix material and ρ_s is the density of solids in the aquifer. The adsorbed fraction is also related to the dissolved concentration according to the linear adsorption isotherm equation which may be expressed as

$$C_a = C \cdot foc \cdot K_{oc} \quad (5)$$

where foc is the fraction organic content and K_{oc} is the organic carbon coefficient. Substituting Equation 5 into Equation 4 and using the Spitz and Moreno (1996) definition of R gives

$$\Delta M = C \cdot n \cdot R \quad (6)$$

The total mass M_T in the aquifer is the product of the rectangular aquifer volume and ΔM which results in

$$M_T = n \cdot R \cdot W \cdot b \cdot \sum_{j=1}^m (C_j \cdot L_j) \quad (7)$$

Thus the normalized total mass in the aquifer at the time that the dissolved concentrations are measured is defined as

$$M'_T = R \cdot \sum_{j=1}^m (C_j \cdot L_j) \quad (8)$$

where

$$M'_T = \frac{M_T}{n \cdot W \cdot b}$$

The total mass released to the aquifer by a rectangular pulse through a finite plane with a height b and a width W is given by

$$M_T = v_x \cdot n \cdot W \cdot b \cdot t_s \cdot C_0 \quad (9)$$

which can be normalized as

$$M'_T = v_x \cdot t_s \cdot C_0 \quad (10)$$

If there is no mass decay, the total mass in the aquifer estimated at the time that the band concentrations are measured is equal to the total mass released to the aquifer, and Equation 8 can be substituted into Equation 10 and solved for the initial concentration

$$C_0 = \frac{R}{v_x \cdot t_s} \cdot \sum_{j=1}^m (C_j \cdot L_j) \quad (11)$$

Solutions to the 1D form of the governing equations are readily available in spreadsheet form, and several spreadsheet models are available for free download at the U.S.

Environmental Protection Agency (USEPA) website. The use of a spreadsheet makes the manipulation of site concentration data to calculate the C_j values relatively trivial, and the model developed by Dawson (1995) solution was modified to incorporate Equation 11.

Following the mass balance approach used to derive Equation 11, two calibration variables are defined as O the normalized mass observed and P the normalized predicted mass by

$$O_j = C_j \cdot L_j \quad (12)$$

$$P_j = \Delta x_j \cdot \sum_{i=1}^m C_p(x_i, t_r) \quad (13)$$

where C_p is the model predicted concentration at the points x_1, x_2, \dots, x_n that land within band j , and the points are separated by the distance

$$\Delta x_j = \frac{L_j}{n} \quad (14)$$

where n denotes the total number of predicted concentration points along the entire plume length. The use of O and P allow the calculation of paired-data calibration parameters including the root mean square error given by Spitz and Moreno (1996) as

$$\text{RMSE} = \frac{100}{\bar{O}} \cdot \sqrt{\sum_{j=1}^m (P_j - O_j)^2} \quad (15)$$

where \bar{O} is the mean of the observed values.

Model calibration consists of the iterative evaluation of model parameters, t_r and t_s in this case, to identify the combination of parameters that result in the best calibration parameter value. One objective of this work is to identify the appropriate band definition

for the 1D model. The parameter $RRMSE$ was defined below to allow the comparison of calibrated t_r and t_s domains for different model band definitions.

$$RRMSE(t_s, t_r) = \frac{RMSE(t_s, t_r)}{\min [RMSE(t_s, t_r)]} \quad (16)$$

where

$RMSE(t_s, t_r)$ is the set of $RMSE$ values calculated for each pair of t_r and t_s values iteratively evaluated for a specific set of simulations. The minimum value of $RMSE$ varies in each simulation in a given domain of t_r and t_s .

Five analysis bands meet the minimum requirements to accommodate the distance concentration curve geometry. The curve has an upwards slope, a plateau, and a downward slope, which is illustrated by the predicted concentration curve in Figure 1. Each slope requires at least two analysis bands and the plateau requires a minimum of one analysis band to best represent the curve's geometry. Three points were chosen as a minimum permissible number of observed points within each band because less than three observed points would degrade the quality of the band's observed data summary statistics. Application of this criterion led to the identification of a maximum number of analysis bands for each simulation.

Model input allowed either constant length bands or variable length bands to represent observed data. An objective of the paper was to determine which of these two band combination better represented the observed values. Model calibration with variable length bands introduces additional complexity depicted by Equation 12. If a single band's length L_j is significantly different from the other variable length bands in a simulation,

the representative quality of O_j is likely decreased. To mitigate this potential, the criteria in Equation 17 were incorporated when selecting variable length band combinations.

$$L_j \leq 2L_{j+1} \text{ and } L_j \leq 1/2 L_{j+1} \quad (17)$$

By simulating concentration values at five meter increments over 5,000 meters using the Dawson spreadsheet model. One dataset represented a roughly rectangular pulse release ($t_r = 24 \text{ years}$, $t_s = 22 \text{ years}$, and $C_0 = 4,000 \mu\text{g}/\text{l}$), the second dataset represented a roughly triangular pulse release ($t_r = 16 \text{ years}$, $t_s = 11 \text{ years}$, and $C_0 = 4,000 \mu\text{g}/\text{l}$), and the third dataset represented an intermediate pulse release ($t_r = 20 \text{ years}$, $t_s = 16.5 \text{ years}$, and $C_0 = 4,000 \mu\text{g}/\text{l}$). Field results were synthesized by selecting 59 (C, x) pairs from each dataset using a uniform random number generator. Two additional sets of 59 synthetic field data representing the intermediate ($t_r = 20 \text{ years}$, $t_s = 16.5 \text{ years}$, and $C_0 = 4,000 \mu\text{g}/\text{l}$) release were similarly developed for further study. This method produced the highest quality observed data without measurement error. Figure 1 illustrates the random spatial distribution of one observed dataset sampled from an intermediate pulse release synthetically developed with the model.

Results

Twenty variable length band combinations with different geometry were evaluated to identify an effective model calibration mechanism. Band lengths were adjusted in each simulation to reduce the observed data coefficient of variation (COV) as defined by Ang & Tang (1975) within each band. By reducing this statistic for each band, variable length bands were expected to improve the bands' representative quality of observed data. When the twenty band combinations were applied to three synthetic datasets with

different values of t_r , t_s , and C_0 , the *COV* of every band's observed data was within 0.66-2.9. The model calibration resulted in model accuracy of ± 25 percent of the actual t_s , t_r values when the $RRMSE = 1.1$ contour was used to determine a reasonable domain of t_s , t_r , and C_0 . Other parameters including the coefficient of determination, the modeling efficiency, the relative percent difference, and the coefficient of residual mass defined by Spitz and Moreno (1996) were insensitive to variable band geometry. The requirement of three observed data points to fall within each band prohibited band length adjustment that would further reduce this range of *COV*. Another restriction for band length adjustment is provided in Equation 17, which suggests that band length variability must be governed to mitigate the negative effect of variable length bands on the normalized observed mass O_j . Variable length bands did not improve calibration. When variable length bands are used in the model, the direct relationship between O_j and the band length L_j shown in Equation 12 may contribute to poor calibration because each calculated O_j value will vary in response to both C_j and L_j .

This issue does not occur when equal length bands are used because each L_j value has the same effect on the calculation of O_j . Equal length band combinations were applied over each dataset; each simulation was defined by its number of analysis bands. The number of bands overlaid on each dataset ranged from five to nine to determine the optimum number of analysis bands for model calibration. Calibration parameters were tested in each simulation and the normalized mass observed and predicted values were best calibrated with $RRMSE$. Several calibration parameters were insensitive to calibration improvement. These included the coefficient of determination, the modeling efficiency, the relative percent difference, and the coefficient of residual mass defined by Spitz and

Moreno (1996). Model calibration provided results within ± 10 percent of the actual solutions when $RRMSE = 1.1$ contours were developed with iterative computations in a specified domain of t_r and t_s . Further analysis with the two additional datasets representing intermediate pulse releases verified this level of accuracy. One dataset's contours of $RRMSE = 1.1$ are graphically represented in Figure 2.

The accuracy of the model was tested for each simulation by comparing the predicted C_0 value to the C_0 value used to generate the synthetic datasets. For all simulations the predicted C_0 was within ± 10 percent of the actual value. The predicted regions of t_s , t_r , and C_0 illustrated in Figures 2 and 3 may be considered reasonable because they compare favorably with Skaggs and Kabala (1998) who used 33 percent as a threshold of accuracy when considering calibration results of synthesized transport systems.

Application at a Former Nebraska Ordnance Site

The spreadsheet model was applied to a trichloroethylene (TCE) dataset collected at a Nebraska Superfund site which has been the subject of prior groundwater modeling studies including Elmore (2007), Miller and Elmore (2005), and Elmore and DeAngelis (2004). The unconfined aquifer at the site consists of glacio-fluvial sands and gravels where flow occurs through primary porosity and it is reasonable to assume that the Dupuit theory of vertical equipotential lines is valid for the aquifer. The plume area of the aquifer has not historically been subject to large stresses such as irrigation pumping, and the plume is relatively far from flow and head boundaries. Thus the assumption of steady uniform flow necessary to apply the Van Genuchten and Alves solution is reasonable for the subject plume. A direct push groundwater investigation was conducted in 2002 and 2003 to characterize the extent of the TCE contamination whose

two dimensional representation can be seen in Figure 4. TCE concentrations above the site action level of $5 \mu\text{g}/\text{l}$ were detected in fifty-nine locations which defined an approximately rectangular plume shape with dimensions of 5,000 m (x_{max}) by 550 m (W). One to six samples were collected at different depth intervals at each location, and no data other than TCE concentrations were collected during the investigation. When multiple depth intervals were sampled at a single location, the arithmetic average of the results was assigned to the location to compress the three dimensional transport system into two dimensions. The plume was then divided into constant length bands for analysis. A remedial action well (RAW) was installed in the approximate center of the plume 2,928 m from the source. The well was sampled several times contemporaneously with the direct push investigation to provide treatment design data, but the remedial action well data were not used included in observed data for model calibration of any simulation.

Site aquifer testing data were used to characterize the aquifer K as $39 \text{ m}/\text{d}$, Elmore and Vandenberg (1997) gave the hydraulic gradient in the area of the subject plume as 0.0023, the site foc average as 0.00028, and the saturated thickness b as 24.7 m. Mehran et al. (1987) estimated the K_{oc} value for TCE as $120 \text{ ml}/\text{g}$, and Piskin (1971) estimated the site n_e as 0.145 and n was assumed to be equal to that value. The density of the solids was assumed to be $2.65 \text{ g}/\text{cm}^3$ based on the typical value given by Freeze and Cherry (1979). Elmore (2007) found that dispersion was relatively minor compared to advection. The model was calibrated using $RRMSE$ by varying t_s and t_r to generate Figure 5. The calibration contours represented in the figure provide relative calibration comparison at values of $RRMSE = 1.1$ between different simulations defined by the

number of analysis bands overlaid the transport system. There is general agreement between the five contours' centroid locations, although the contour boundaries vary as illustrated in Figure 5. When applying the model to field data, any simulations with a number of analysis bands that meet the band number criteria discussed above can be used to produce a reasonable domain of the source history parameters when a $RRMSE = 1.1$ calibration contour is used.

The calibration contours illustrated in Figure 5 provided a domain of t_s and t_r that required further investigation before the results could be considered reasonable. Model verification was necessary to confirm the calibration mechanism used in this study. The RAW installed 2928 m down gradient of the source release was operated briefly five times so that it could be sampled prior to putting it in service, and that concentration dataset provides a means for model verification. The observed data used for model calibration was sampled one year prior to the five samples obtained from the RAW. The RAW was designed to hydraulically capture the entire width and saturated thickness of the plume, so the data collected at the well should be appropriate for comparison to a 1D transport model. The mean TCE concentration measured at the well was 4,060 $\mu\text{g/L}$ with a standard deviation of 577 $\mu\text{g/L}$. A log normal distribution was assumed for the data after the data was tested with the Shapiro-Wilk test for log normality and the resulting p-value was more than 0.10.

When one year was added to the time since the source release t_r , a Monte Carlo (MC) model was developed where t_s , and t_r , and the observed values within each band were random independent variables. The Shapiro-Wilk test indicated the concentration data in six bands fit a log normal probability density function (pdf) with the exception of bands

two and three. However, as discussed below, a nonparametric Mann-Whitney test resulted in little difference in the results of the parametric student's t-test. The arithmetic mean and standard deviation of each band's observed values characterized the band's log-normal distribution. The development of calibration contours of $RRMSE = 1.1$ for each constant length band simulation allowed a rectangular approximation of each contour, all of which are illustrated in Figure 5. The rectangular calibration area of each simulation was used to define two new variables called t'_s and t'_r as illustrated in Figure 7. For the calibration results of the six band simulation, a uniform pdf with limits of 0 and 1.20 yr was assigned to t'_s and t'_r was assigned an independent uniform pdf with limits of 0 and 0.40 yr. A rotation and translation equation converted t'_s and t'_r realizations to t_s and t_r , which were input to the model. This process was repeated with ranges of t'_s and t'_r corresponding to rectangular approximations for $RRMSE = 1.1$ calibration contours for simulations with five through nine constant length bands. Each MC simulation produced 1,000 realizations of the predicted concentration at the location and time of the RAW observed data. The MC realization data was tested for log normality with an α -level of 0.01. Since the minimum p-value of the MC realization datasets was 0.012, log normality of both datasets was assumed and the two tailed student's t-test with a 90 percent confidence level was used to test the assumption of no statistical difference between the two datasets. The nonparametric Mann-Whitney test with a 90 percent confidence level was used to support the assumption, which resulted in similar p-values to those of the student's t-test. The minimum p-value of the student's t-test was 0.06 and the minimum p-value of the Mann-Whitney test was 0.05. Table 1 provides model verification p-values of both hypothesis tests for the tested simulations.

These results support model verification because they do not permit rejection of the null hypothesis, that the two datasets are not significantly different.

The realizations produced by the MC application provide estimations of the source history parameters. The sample mean and sample standard deviation values of C_0 , t_r , and t_s of each simulation's realization ensemble are presented in Table 1. The simulations used for analysis had constant length band combinations between five and nine bands because these simulations met the above discussed criteria. The results in Table 1 suggest that the band number in each eligible simulation was insensitive to model accuracy. Although these point values are results of a stochastic approach to estimate the source history parameters, the primary objective of the model is to provide a likely domain of the source history that defines a point of departure for more sophisticated software applications.

Discussion

The literature review indicated that it is uncommon for a source history investigation to be applied to field data. For example, a table in Michalak and Kitanidis (2004) lists 14 of 19 inverse modeling applications as having been applied at hypothetical sites. The use of field data creates additional challenges due to the variability of the data compared to hypothetical cases. The sources of the variability may include an unsteady release function, the spatial variability of the aquifer hydrogeology, and concentration measurement error. It is assumed that both the direct push dataset and the well dataset include measurement error and release variability. Furthermore, it would seem reasonable to state that the direct push dataset addresses spatial variability while the well dataset addresses temporal variability. Yet there are inaccuracies associated with these

assumptions. While it is convenient to assume that the direct push data represent the TCE concentrations at a single point in time, the data were collected over an approximately 2 year period. Likewise, it is convenient to assume that each time that the RAW was sampled over the 1 year period; it was pumped so that the resulting capture zone was equally developed so as to remove an element of spatial variability from the data. However, there were no data collected characterizing the actual extent of capture zone developed during any of the 5 well sampling events. Therefore, both concentration datasets include elements of all of the sources of variability but there is insufficient information available to systematically evaluate the comparability of the concentration datasets. The two concentration datasets are not atypical of those available to practitioners so model verifications have been performed with the explicit acknowledgement that the data may not be perfect in terms of theoretical comparability.

Conclusions

The mass-balance approach for estimating t_s , t_r , and C_0 provided reasonable results with the synthetic datasets and the observed site dataset when the bandwise predictions were used for deterministic and stochastic simulations. The stochastic results were reasonable given the variability of the well dataset used for verification. Although the bandwise calibration made point predictions less useful, the method provides a reasonable predicted domain of t_s , t_r , and C_0 that acts as a point of departure for more sophisticated multi-dimensional numerical modeling. The model may also reduce effort for additional numerical modeling because the best domain of t_s , t_r , and C_0 is identified.

Although variable length bands did not produce better results relative to constant length bands, the results of both approaches were reasonably accurate. Constant length bands

reduce analysis band incorporation complexity. As long as the consistent method is used to identify the range of minimum and maximum number of bands, the goodness of fit is insensitive to number of bands.

The model may appeal to modelers and designers who can rapidly modify the spreadsheets to evaluate concentration time series at various points as well as calculating the potential mass to be treated and the associated treatment costs. The generation of output ensembles for such additional variables is relatively straightforward given the simplicity of commercial Monte Carlo software.

The apparent utility of the spreadsheet model should not overshadow the limitations of the underlying 1D model. The 1D model will have limited application based on site-specific hydrogeologic conditions where there is a low potential for multiple or complex contaminant sources. The model may be useful for application where the data indicate that the flow field is relatively uniform, has low dispersion, and the assumption of a rectangular release is reasonable. These conditions are encountered as evidenced by the Nebraska Superfund Site data.

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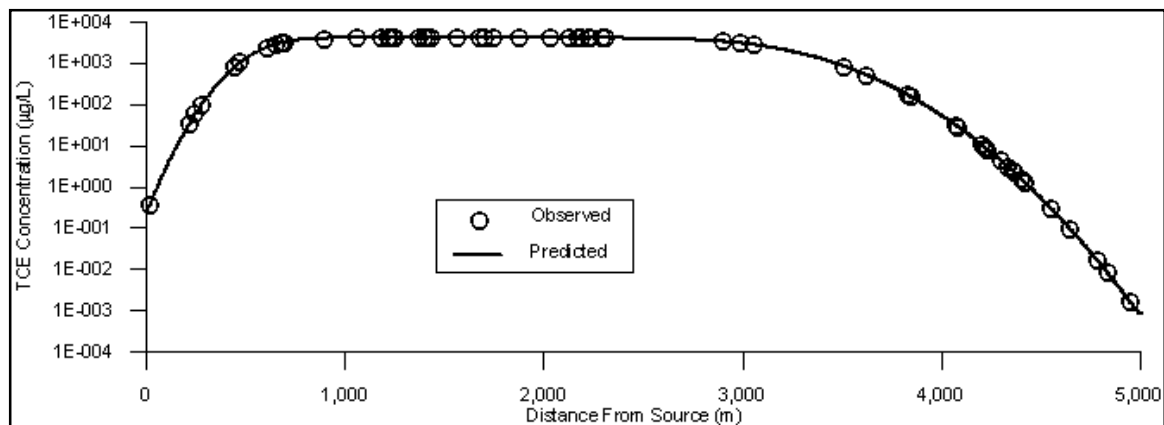


Figure 1: Observed Concentrations with Synthetic Data

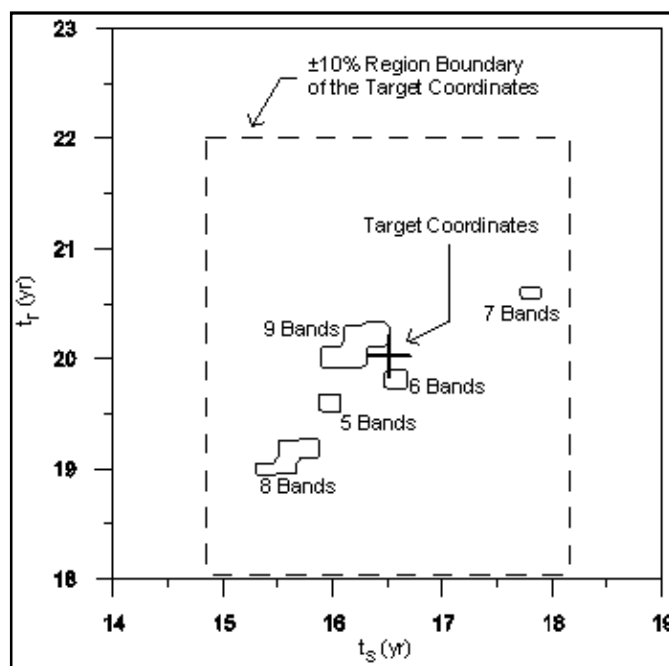


Figure 2: Calibration Results Using Synthetic Data

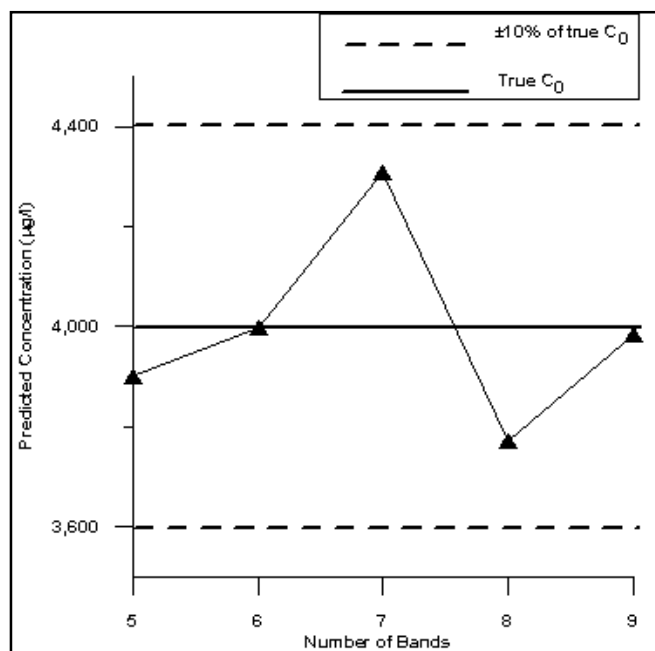


Figure 3: Predictions of C_0 Using Synthetic Data

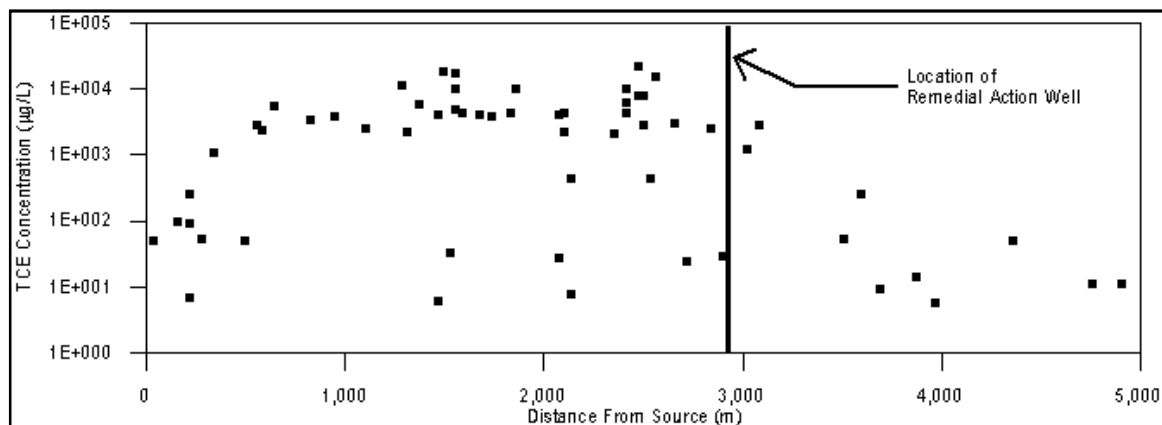
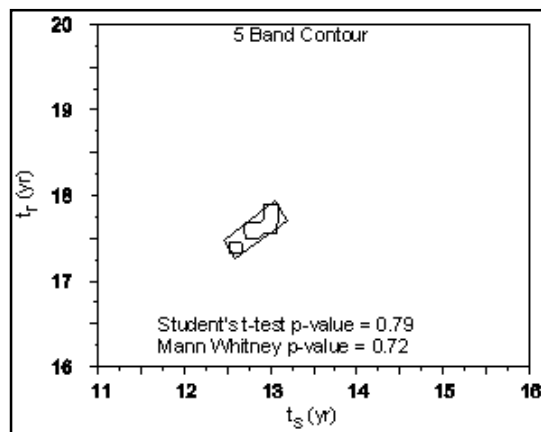
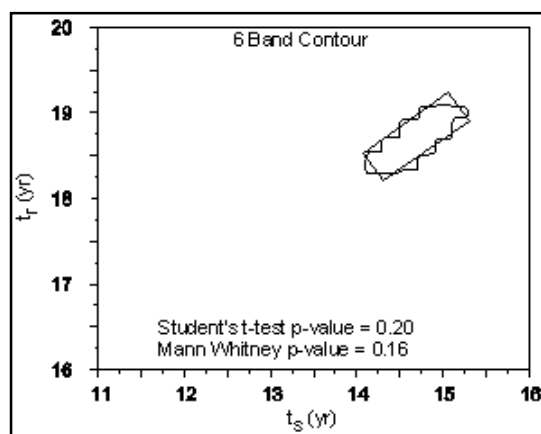


Figure 4: Observed Concentrations with Actual Data

(5a)



(5b)



(5c)

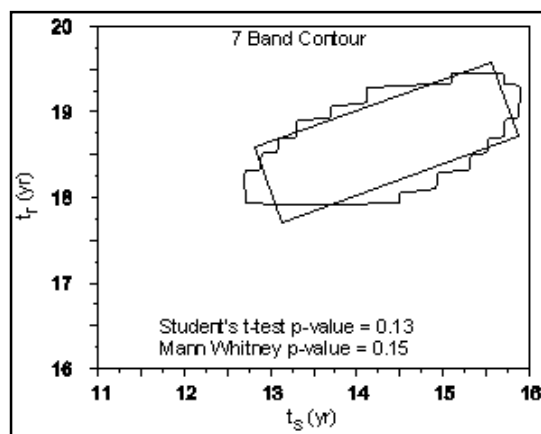
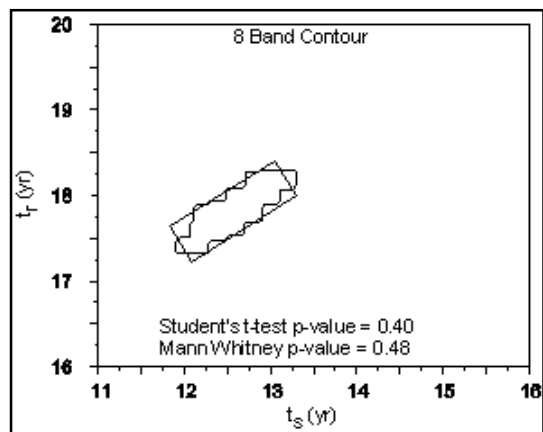


Figure 5: Calibration results with observed data

(5d)



(5e)

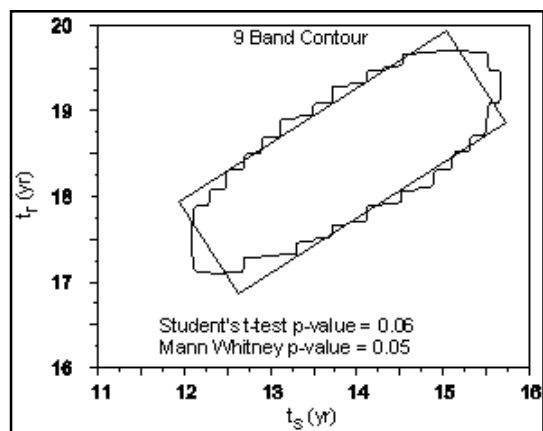


Figure 5 (Continued): Calibration results with observed data

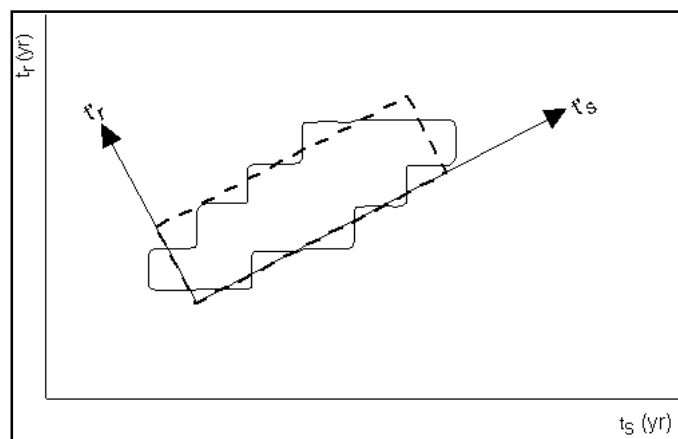
Figure 6: Spatial Definitions of t'_s and t'_r

Table 1: Model Verification Results

Simulation Bands	T_r		T_s		C_0		p-values	
	\bar{X}	S	\bar{X}	S	\bar{X}	S	Student's t	Mann-Whitney
5	18.6	0.154	12.8	0.163	6860	1080	0.79	0.72
6	19.7	0.226	14.7	0.283	5970	1020	0.20	0.16
7	19.7	0.393	14.6	0.830	6220	1170	0.13	0.15
8	18.8	0.274	12.6	0.341	6940	1080	0.40	0.48
9	19.7	0.801	13.6	0.834	6540	1120	0.06	0.05

4. RECOMMENDATIONS FOR FUTURE WORK

The following ideas and topics are recommended to continue this research and to address assumptions made in this paper.

- Instead of using bands of analysis, a grid system can be overlaid on a contaminant plume to develop two-dimensional control volumes to advance the discussed method of contaminant mass evaluation.
- Improve model calibration by studying the mechanism responsible for variation of the $RRMSE = 1.1$ contour within the achieved range of ten percent accuracy predictions of the time source history parameters.
- Synthesize artificial transport systems with different known source release history parameters than those used in this study. Upon achieving acceptable model calibration at larger ranges of the time source history parameters, the site applicability of the model would improve.

APPENDIX A.

**VARIABLE LENGTH BAND MODELS USING SYNTHESIZED DATA
ON DVD-ROM**

Included with this Thesis is a CD-ROM, which contains models using variable length bands. Each module of the VISUAL BASIC CODE has been developed using VISUAL BASIC for Microsoft Excel 2007. All documents have been prepared as Microsoft Office 2007 document files (Windows Vista).

APPENDIX B.

**CONSTANT LENGTH BAND MODELS USING SYNTHESIZED AND
ACTUAL DATA ON DVD-ROM**

Included with this Thesis is a CD-ROM, which contains models using constant length bands. Each module of the VISUAL BASIC CODE has been developed using VISUAL BASIC for Microsoft Excel 2007. All documents have been prepared as Microsoft Office 2007 document files (Windows Vista).

APPENDIX C.

**A CD WITH THE STORED THESIS DEFENCE PRESENTATION AND .PDF
COPY OF THE THESIS ON CD-ROM**

Included with this Thesis is a CD-ROM, which contains a cd with the stored thesis defense presentation and .pdf copy of the thesis. All documents have been prepared as Microsoft Word 2007 document files (Windows Vista).

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VITA

Ben Johnson was born on June 21, 1985 to Frank and Theresa Johnson of Greenfield, Missouri. He was born in Springfield, Missouri and attended Greenfield High School where he played varsity baseball, was an active member of the National Honors Society and Scholar Bowl. Ben is an avid guitar player, outdoorsman, and enjoys fishing and hunting in the Missouri Ozarks. Ben earned a Bachelor of Science degree in Environmental Engineering with a minor in Geological Engineering and received honors of Summa Cum Laude at the Missouri University of Science and Technology in May 2008. During his time at S&T, Ben was an active member of the Water Environment Federation and the Missouri Stream Team. For two summer vacations, Ben worked as a trail construction crew leader in the White Mountain Range of New Hampshire for the Appalachian Mountain Club. He was then commissioned in the United States Public Health Service for two summers as a sanitation engineering intern in central Oklahoma and northern Wisconsin. Ben also worked as an environmental intern at a phosphate mine in central Florida for a semester co-op. In December of 2008, Ben was registered as an Engineer Intern in the State of Missouri and intends to pursue his Professional Engineering license. He received a Master's degree in Geological Engineering from the Missouri University of Science and Technology in August 2009 with emphases in groundwater remediation and water resources. Ben was also involved with a renewable energy demonstration project funded by the State of Missouri to evaluate the feasibility of installing wind and solar energy systems at state owned facilities. Upon completing his graduate career, Ben left academia to pursue a career as a hydraulic engineer with the US Army Corps of Engineers in Kansas City, Missouri.