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The effects of non-linear bromide adsorption on apparent macrodispersivity

Eron J. Dodak
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THE EFFECTS OF NON-LINEAR BROMIDE ADSORPTION ON
APPARENT MACRODISPERSIVITY

by

Eron J. Dodak

Bachelor of Science
Portland State University, 1993

A Thesis

Submitted to the Graduate Faculty

of the

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in partial fulfillment of the requirements

for the degree of

Master of Science

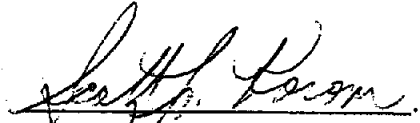
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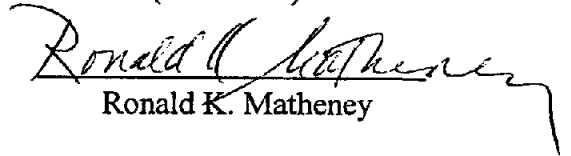
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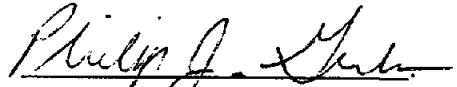
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This thesis meets the standards for appearance, conforms to the style and format requirements of the Graduate School of the University of North Dakota, and is hereby approved.

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To Robert and Darlene Dodak

ABSTRACT

Macrodispersivity estimates are crucial in predicting the subsurface transport of contaminants. These predictions have become increasingly important as the number of contaminated sites continues to increase. In addition, the longevity of many contaminants can affect water quality over extended periods of time.

Anionic tracers such as bromide and chloride are commonly used as groundwater tracers in dispersivity experiments because they are inexpensive, easily detected, and are commonly believed to behave conservatively. It has been found, however, that anionic tracers do not always behave conservatively; sediments with a net positive surface charge can adsorb some of the tracer.

A weakly non-linear, experimentally derived Freundlich sorption isotherm was found to describe bromide adsorption in sediments from the Savannah River Site in South Carolina. The aim of this research was to quantify the effects of this sorption on apparent dispersivities relative to a conservative tracer. In addition, the effects on apparent dispersivities of injecting different amounts of tracer and omitting tracer concentrations below detection limits were evaluated.

Monte Carlo simulations were performed using SUTRA and WATSUTRA. The models were developed to simulate natural gradient tracer tests. Two types of hydraulic conductivity fields were generated using the Fast Fourier Transform method. One represented a weakly heterogeneous flow field, with physical parameters similar to the Borden aquifer in Ontario, Canada; the other represented a strongly heterogeneous flow

field with physical parameters similar to the Columbus aquifer in Mississippi. These hydraulic conductivity fields represent two extremes of aquifer conditions.

Forty simulations were performed in this study for eight cases. Five realizations were performed for each case; each realization was simulated for two years. Apparent dispersivities were calculated by the method of moments. Ensemble means were estimated by averaging the apparent longitudinal or horizontal transverse dispersivity values for the five realizations in each case. Null hypothesis testing was performed with a standard t-test to test the significance between the estimates for the ensemble means for various cases.

The following simplifying assumptions were used in this research:

- 1) Sorption according to the Freundlich isotherm was assumed to be a reversible equilibrium process. Thus, desorption followed the same isotherm as adsorption, only with decreasing concentrations, and both adsorption and desorption were rapid in comparison to advective flow.
- 2) The Freundlich isotherm was constant throughout the model domain.
- 3) The groundwater model employed was two-dimensional; vertical heterogeneity was not represented.

The results indicated the experimentally derived Freundlich isotherm, given the assumptions and limited number of realizations used in this study, did not significantly affect apparent plume dispersivity values. Thus, bromide was not found to be an unsuitable groundwater tracer. The quantity of tracer mass injected and omission of tracer concentrations below detection limits also did not have a significant effect on apparent dispersivity values. It was found that using tracer concentrations below detection level can adversely affect high-order spatial moments; this finding may, however, be unique to SUTRA and WATSUTRA or similar finite-element programs.

INTRODUCTION

The quantification of macrodispersivity is important in assessing the spreading of a plume as it travels through an aquifer. Macrodispersivity affects the lateral, vertical, and longitudinal extent of a contaminant plume. The way in which a plume disperses as it moves through an aquifer will affect the type of remediation strategy best suited for a site. This has become increasingly important as the number of contaminated sites continues to increase. In addition, the longevity of many contaminants can affect groundwater quality over extended periods of time.

Over the past 40 years dispersivity research has moved from small-scale laboratory column tests, to larger-scale single-well tracer tests, and finally to large-scale natural gradient tracer tests. The advantage of the natural gradient tracer test is that it can be used to calculate longitudinal and transverse dispersivities on a field scale of hundreds of meters under natural groundwater flow conditions. Contaminant plumes found in nature are generally of this scale. The results of a natural gradient tracer test can be used as a predictive tool in subsurface transport and in transport model testing.

Conservative tracers are an important tool in natural gradient tests because they do not react with the solid or liquid phases in an aquifer; thus, the movement and spreading of the tracer is caused by groundwater flow. Anionic tracers such as bromide and chloride are commonly used as groundwater tracers because they are inexpensive, easily detected, and are commonly believed to behave conservatively. The conservative nature of anionic tracers is due to the net negative surface charge on many sediments, which repels anions and limits interaction between the anion and mineral surfaces.

There are two types of surface charges found on minerals, 1) constant surface charge minerals, which dominate in temperate climates, and 2) variable surface charge minerals, which dominate in tropical climates (Uehara and Gillman, 1981, p. 2-3). The charge on minerals with a constant surface charge is permanent. A perfectly formed crystal lattice will have no net surface charge because the atoms in the crystal are electrically balanced (Uehara and Gillman, 1981, p. 31). Some crystals do, however, possess defects within the crystal lattices. This is generally the result of isomorphic substitution, which is the substitution of an ion at a particular site in the crystal lattice with one of a different, generally lower, valence. The result is a charge imbalance in the crystal lattice that is permanent. This generally leads to a net negative surface charge, which is balanced by counterions in solution (Uehara and Gillman, 1981, p. 2). Anionic tracers such as bromide and chloride are generally conservative in sediments with constant charge minerals because the net negative surface charge of the sediments repels anionic tracers.

Some highly weathered sediments found in tropical climates, however, may have a net positive surface charge that may lead to adsorption of some of the anionic tracer. In such climates the constant surface charge minerals are generally severely altered or completely weathered out (Uehara and Gillman, 1981, p. 3). Sediments containing significant quantities of variably charged minerals such as Fe and Al oxides tend to adsorb anionic tracers (Uehara and Gillman, 1981, p. 43). The surface charge of the oxide is generated by potential-determining ions in solution, principally H^+ and OH^- . Thus, the pH governs the sign and magnitude of the charge. The pH where the net surface charge is zero is the zero point of charge (ZPC). This indicates that equal amounts of H^+ and OH^- have been adsorbed onto the oxide's surface. The ZPC of Fe and Al oxides is generally within the range of pH 7-9, and depends on the composition and degree of crystallinity of the oxide (Uehara and Gillman, 1981, p. 46). At pH values

below ZPC, more H^+ is adsorbed than OH^- ; thus the mineral surface charge will be positive and an anionic tracer may be adsorbed onto the surface of the oxides to maintain charge neutrality. As a result, some of the tracer may be lost from solution.

Sediments with a net positive surface charge are present at the Savannah River Site (SRS) in South Carolina (Seaman et al., 1996). SRS is a 775 km² Department of Energy complex on the Atlantic Coastal Plain. Column studies were conducted on sediment samples from SRS (Korom, 1994, personal communication). The sediments are part of the Dry Branch Formation and consist primarily of medium- to fine-grained sands with less than one percent silt and clay. Undisturbed horizontal sediment cores were taken from an erosional cut approximately 60 feet deep (Korom, 1994, personal communication).

The column studies were performed with the samples by injecting known concentrations of tritium (3H) and bromide (Br^-) through the column at an average rate of 0.103 m/day. The bromide had calcium as the carrier cation ($CaBr_2$). Tritium was used as the conservative tracer. The concentration of the influent was increased in order-of-magnitude increments and was then reduced sequentially to the initial low concentrations. Bromide concentrations of almost 4 orders of magnitude were tested. At each concentration, the amount of bromide adsorbed was calculated relative to tritium breakthrough curves (BTC), which were fitted using CXTFIT (Parker and van Genuchten, 1984). The difference between the tritium and bromide BTCs is related to the quantity of bromide adsorbed onto the sediment. The mass of bromide adsorbed per mass of sediment, C_s , was plotted against the mass of bromide per mass of water, C , on log-log axes. The resulting linear regression (95% confidence intervals on the regression) yielded the following equilibrium Freundlich isotherm:

$$C_s = 8.64 \times 10^{-5} (\rho C)^{(1/1.12)} \quad (1)$$

where ρ is the density of water (M/L^3). The sorption coefficient in the Freundlich isotherm, 8.64×10^{-5} , represents the y-intercept value of the line in the graph of $\log C_s$ versus $\log C$. The Freundlich exponent, $n = 1/1.12$, represents the slope of the line, and is defined for values $0 < n < 1$; a value of 1 indicates linear sorption. An implicit assumption of a Freundlich isotherm is that sorption equilibrium is rapid in comparison to advective groundwater flow (Fetter, 1993, p. 117).

The aim of this research was to quantify, using some simplifying assumptions, the effects of the non-linear sorption isotherm in Eq. (1) on apparent dispersivities relative to that of a conservative tracer. A numerical groundwater model was developed to quantify the effects because actual natural gradient tracer tests are costly to perform. This will provide guidelines on whether anionic tracers can be used in sediments onto which they are adsorbed following a similar sorption isotherm. This study also examined the effects of an order-of-magnitude difference in the amount of tracer mass used in the simulated natural gradient tests. In addition, the effects of omitting concentrations below a detection level, set at 0.1 part per million (ppm), were examined.

BACKGROUND

The purpose of this section is to provide the reader with a general knowledge of solute transport and stochastic groundwater modeling. The basic concepts of solute transport will be discussed with an emphasis on dispersion, the focus of this study. Stochastic groundwater modeling, the approach to solute transport modeling used this study, will also be discussed.

Solute Transport

Solute transport is the result of advection, mechanical dispersion, molecular diffusion, and chemical reactions. Advection is the movement of groundwater as a result of a hydraulic gradient. Mechanical dispersion is caused by variations in flow velocity at the pore and field scales. Molecular diffusion is the movement of a solute as a result of a concentration gradient. Mechanical dispersion requires advection; molecular diffusion does not. Mechanical dispersion and molecular diffusion are commonly combined into a quantity called hydrodynamic dispersion because they cannot be evaluated independently in flowing groundwater (Fetter, 1993, p. 51). Chemical reactions include sorption, transformation, and precipitation/dissolution. Adsorption of anionic tracers, the focus of this study, is commonly a result of ionic attractions between the tracer and aquifer sediments.

Advection

Advection is described by Darcy's law, which states that the flow rate through a porous medium is proportional to the hydraulic gradient; the proportionality constant is

hydraulic conductivity. The hydraulic conductivity is a measure of the permeability of sediments for a given fluid (generally water). Permeability is a measure of how readily fluids can move through a porous medium and is an intrinsic property of the porous medium. Darcy's law can be used to estimate groundwater velocity; the form used to calculate average linear velocity in the i direction is:

$$v_i = - (K/n) * (dh/dl) \quad (2)$$

where K is the hydraulic conductivity (L/T), n is effective porosity (L^3/L^3), and dh/dl is the hydraulic gradient (L/L).

Hydrodynamic Dispersion

Hydrodynamic dispersion, as stated above, consists of two terms, mechanical dispersion and molecular diffusion. Mechanical dispersion is defined as dispersivity times average linear velocity. Dispersivity is a mixing length; it is an intrinsic property of the porous medium. It results from variations in flow velocity at the pore and field scales. If dispersion did not occur, there would be an abrupt interface between the solute and uncontaminated groundwater; the solute would move like a plug. The molecular diffusion component of hydrodynamic dispersion is a result of concentration gradients. Solutes in water spontaneously travel from areas of high concentration toward areas of low concentration. Molecular diffusion is a result of mixing caused by random molecular motions due to the thermal kinetic energy of the solute in solution (Domenico and Schwartz, 1990, p. 367).

The hydrodynamic dispersion coefficients in the longitudinal, D_L , and horizontal transverse, D_T , directions are:

$$D_L = \alpha_L v_i + D^* \quad (3)$$

$$D_T = \alpha_T v_i + D^* \quad (4)$$

where α_L is the longitudinal dispersivity (L), α_T is the transverse dispersivity (L), and D^* is the effective molecular diffusion coefficient (L^2/T).

The longitudinal direction is parallel to the principal direction of flow; the horizontal transverse direction is in the horizontal plane perpendicular to the direction of flow. A third component, the vertical transverse direction, is commonly neglected because mixing in the vertical plane is very small in naturally stratified sediments (Gelhar et al., 1992).

In most groundwater systems the molecular diffusion term of hydrodynamic dispersion can be neglected because it is insignificant in comparison to mechanical dispersion. The only cases where diffusion would dominate are where concentration gradients are high and hydraulic gradients and/or hydraulic conductivities are low (Perkins et al., 1963).

Mechanical dispersion occurs at the pore and field scales. At the pore scale, mechanical dispersion is mainly due to heterogeneities in the sediment (Wang and Anderson, 1982, p. 175). The result of the heterogeneities (i.e. sediment grains) is varying pore size, flow path length, and velocity distributions in the pore (Fetter, 1993, p. 149). The pore size affects the velocity at which the water will travel; the smaller the pore, the more friction the water encounters with the grains and the slower it travels. Water flows along paths of varying lengths as it moves around individual sediment grains to go a given linear distance. The final cause of dispersion at the pore scale is the velocity distribution in the pore. Water travels fastest in the center of the pore throat and slower where it interacts with the grain surfaces. The factors listed above all cause the

water to mix or disperse. Dispersivities at the pore scale are on the order of centimeters (Wang and Anderson, 1982, p. 175).

At the field scale dispersivity, also known as macrodispersivity, is the spreading of a plume at scales of tens to thousands of meters. It is caused by variations in aquifer parameters contained in Darcy's law (Eq. 2). Hydraulic conductivity is by far the most important of these parameters because it can vary several orders of magnitude in a distance of a few meters. Porosity and hydraulic gradient generally vary little in comparison. Thus, macrodispersivity is primarily the result of the variance in hydraulic conductivity of an aquifer. Dispersivities at the field scale are on the order of meters (Wang and Anderson, 1982, p. 175).

Groundwater flows preferentially along paths of the highest hydraulic conductivities. A low conductivity zone, such as a clay lens, will deflect groundwater around the zone. As a plume travels longer distances, more heterogeneities are encountered, and the macrodispersivity of the formation increases. This is known in the literature as the scale effect of dispersivity.

Neuman (1990) performed a fractal analysis on the scaling of longitudinal dispersivities. In the analysis, Neuman derived a semivariogram and fractal dimension that can be viewed as a universal scaling rule. This scaling rule essentially accounts for the self-similarity of natural log transformed hydraulic conductivity values from a wide range of geologic materials, flow conditions, and flow lengths. The universal scaling rule could therefore be used without describing any particular site, other than its length.

Gelhar et al. (1992) believe it is not appropriate to represent longitudinal dispersivity data by a single universal line. They argued the scale effect of dispersivity was less clear when the reliability of the data was considered. In the study they examined dispersivity calculations from 59 different field sites. The dispersivity values were rated by the type of test and method of data analysis. Highly reliable data were only available

up to scales of about 300 m. They acknowledged a scale dependence of longitudinal dispersivity, but concluded that more reliable large-scale tests must be performed to determine the nature of the dependence.

Engesgaard et al. (1996) reported a field scale dispersion test on a scale of 1000 m. The researchers reported a longitudinal dispersivity value in the range of 1 to 10 m. The study results apparently do not support the hypothesis that longitudinal dispersivity increases indefinitely with scale.

Methods of Calculating Dispersivity

There are principally three ways to estimate dispersivities at three different scales. At the pore scale, dispersivity can be estimated by performing laboratory column tests. This method only takes into account dispersion due to pore scale heterogeneities. At a scale of meters, single-well tracer tests can be performed which produce in situ dispersivity values representing the large-scale heterogeneities in the area affected by the test as well as those at the pore scale. At the scale of tens to thousands of meters (field scale), natural gradient tracer tests can be performed to produce in situ dispersivity estimates that take into account the large-scale heterogeneities over tens to thousands of meters as well as those at the pore scale. The natural gradient tracer test is the only method with which transverse dispersivities can be estimated and is the focus of this study.

The natural gradient tracer test involves a pulse injection of a tracer at a constant rate and concentration into an aquifer. Inherent in the test is the assumption that the injection rate is slow; the natural gradient must not be affected by the injection. Once the injection is complete, the tracer plume is allowed to move through the aquifer under natural gradient conditions. A three-dimensional array of sample wells is installed to measure the concentrations of the tracer plume through time. The result is a database of spatial and temporal concentration data providing "snapshots" of the plume through time.

Generally, the plume is monitored for a few years (Mackay et al., 1986; LeBlanc et al., 1991; Boggs et al., 1992).

Apparent macrodispersivities in the longitudinal and transverse directions are calculated from the concentration data by the method of moments. This involves summing the tracer concentrations and finding the variances about the center of mass of the plume. The average rate of change of the plume variance with respect to the center of mass is used to calculate apparent dispersivities (these calculations will be discussed in the Methods section).

Stochastic Groundwater Modeling

There are two basic approaches to groundwater modeling, deterministic and stochastic. A deterministic model solves a partial differential equation (Eq. 5 below) for a given set of input values, aquifer parameters, and boundary conditions (Fetter, 1993, p. 77). The output variable, such as solute concentrations, have specific values at every point in the aquifer. An assumption in this approach is that the distribution of aquifer parameters is known. Thus, in deterministic modeling an "exact" output parameter is provided. The problem with such a model is the uncertainty in the characterization of the aquifer parameters. It is not possible to know the hydraulic conductivity of the aquifer sediments at every point in the system. Even if it were possible to test the hydraulic conductivity at every location in an aquifer, additional uncertainty results from experimental error.

A stochastic approach to groundwater modeling recognizes the statistical uncertainties involved in parameter estimation and provides a range of possible outcomes rather than an "exact" solution. This is important because knowledge of aquifer parameters will always be limited and therefore cannot provide an exact solution to the problem.

The type of stochastic groundwater modeling used in this study was a limited Monte Carlo analysis. Monte Carlo simulations are performed by generating random, but statistically equivalent, hydraulic conductivity fields over the entire model domain. The advection-dispersion equation is then solved using the generated aquifer parameters. The one-dimensional form of the advection-dispersion equation is (Fetter, 1993, p. 115):

$$\partial C/\partial t = D_L(\partial^2 C/\partial x^2) - v_i(\partial C/\partial x) - (\rho_d/n)(\partial C^*/\partial t) \quad (5)$$

where ρ_d is the bulk density of the aquifer (M/L^3), and C^* is the amount of solute sorbed per unit weight of solid (M/M).

The advection-dispersion equation states that the rate of change in concentration with respect to time is equal to dispersion minus advection and sorption (this simplified form of the equation assumes there are no sources, sinks, or chemical reactions occurring other than sorption).

Hydraulic conductivity is one of the most important parameters in solute transport modeling because conductivities can vary several orders of magnitude for geologic materials. As such it is a parameter with great uncertainty. The hydraulic conductivity field is treated as a random variable with a given covariance structure. Each simulation conducted with a random hydraulic conductivity field constitutes a realization. The best estimate of the plume concentration solution is the ensemble mean. The ensemble mean is the mean of all possible random but equivalent populations (i.e. realizations) (Fetter, 1993, p. 81). A small number of realizations (e.g. five) is used to approximate the ensemble mean in a limited Monte Carlo analysis.

The covariance structure of the natural log transformed hydraulic conductivity ($\ln K$) field consists of the mean, variance, and correlation lengths. The mean is the average of all $\ln K$ values. The variance is a measure of the spread of values about the mean $\ln K$;

it is a measure of the heterogeneity in an aquifer. The correlation length is a measure of the distance that values are correlated from a given point in space. The correlation of two values decreases with increasing distance between them. There is a separate correlation length for every dimension used in the random field.

The correlation length can be thought of as a measure of the size of heterogeneities in an aquifer. The heterogeneities in a natural depositional system are the result of different local environments of deposition. In the horizontal plane they may represent deposits such as point bars and channel deposits in a meandering stream system. In the vertical plane the correlation length generally represents stratification.

Limited Monte Carlo simulations have been used to estimate the ensemble mean by using a small number of realizations. Burr et al. (1994) performed such a study with the geostatistical parameters of the Borden aquifer in Ontario, Canada. The ensemble mean in the study was estimated using only five realizations. The researchers performed an additional 20 realizations and found essentially identical results.

PREVIOUS WORK

There are two main categories of previous work relevant to this study; field studies, which focus on natural gradient tracer tests, and numerical studies, which focus on stochastic groundwater modeling with sorbing solutes in heterogeneous aquifers. Both are reviewed below. In addition, a new groundwater modeling trend is discussed.

Field Studies

Natural Gradient Tracer Tests

Two natural gradient tracer tests are discussed in this section, the Borden and Columbus natural gradient tracer tests. These were chosen because they have well documented statistical parameters of hydraulic conductivity and they represent two extremes in aquifer parameters; the Borden site represents a weakly heterogeneous aquifer, the Columbus site represents a strongly heterogeneous aquifer.

Borden Site

A large-scale natural gradient tracer test was conducted in the unconfined sand aquifer underlying an inactive sand quarry at the Canadian Forces Base Borden in Ontario, Canada (Mackay et al., 1986). The purpose of this study was to produce a detailed and accurate data base describing the transport, transformation, and fate of conservative tracers (bromide and chloride) and halogenated organic contaminants in the saturated zone (Mackay et al., 1986). The data could then be used to evaluate dispersive transport models. This experiment was ideal for such evaluation because of the controlled initial conditions, long plume travel distance, and detailed three-dimensional

resolution of concentration distributions and hydraulic conductivity fields. Detailed three-dimensional monitoring of solute concentrations through time provided "snapshots" of plume development, which could be used to calculate spatial moments to quantify the movement and spreading of the plumes through time.

The aquifer is composed of clean, well-sorted, fine- to medium-grained sand of glaciofluvial origin. Quartz, feldspar, and carbonates dominate the mineralogy of the sediments. The grains are subangular to well-rounded (Mackay et al., 1986).

The aquifer extends about 9 m below the horizontal quarry floor and is underlain by a thick silty clay deposit. The lower 2-3 m of the aquifer contains a landfill leachate plume. The water table depth is generally about 1.0 m below the quarry floor, but fluctuates seasonally. The average horizontal hydraulic gradient throughout the year in the study area is approximately 0.0043 (Sudicky, 1986). The aquifer is quite homogeneous compared to most aquifers. The bedding is approximately horizontal and parallel, but some cross and convolute bedding are also found. The main forms of spatial heterogeneity in the aquifer are thin lenses (0.02-0.1 m) of 2-5 m lateral extent composed of sediments of contrasting particle size distribution and hydraulic conductivity (Sudicky, 1986).

Sudicky (1986) examined the spatial structure of the hydraulic conductivity at the Borden site using 32 cores from two core transects. One transect was oriented in the direction of groundwater flow, the other was transverse to groundwater flow. Each core was approximately 2 m long and was obtained from depths of approximately 2.5 (top) to 4.5 m (bottom) below the quarry surface. The horizontal spacing of the cores was 1 m. Each core was divided into 0.05 m vertical intervals; each relatively homogeneous interval was dried and subjected to a permeameter test to determine its hydraulic conductivity. A total of 1279 hydraulic conductivity measurements were performed on the cores.

The geometric mean of the hydraulic conductivity measurements was found to be 9.75×10^{-3} cm/s. Each hydraulic conductivity value was transformed into a natural log value ($\ln K$); the geometric mean of $\ln K$ was -4.63. The variance of the mean $\ln K$ was found to be 0.29 with correlation lengths of 2.8 m in both horizontal directions and 0.12 m in the vertical direction (Sudicky, 1986). A reevaluation of the data by Woodbury and Sudicky (1991), which examined how assumptions and interpretations may affect the geostatistical parameters, yielded in the direction of flow, a variance of 0.24 and a correlation length of 5.1 m in the horizontal direction and 0.21 m in the vertical direction. The values in the direction transverse to flow were 0.37, 8.3 m, and 0.34 m, respectively.

The goal of the injection system was to create plumes of well defined geometry with a uniform distribution of solutes (Mackay et al., 1986). In addition, the injection was desired to be essentially instantaneous with little disturbance to the natural flow. A total of 12 m^3 of solution containing the nonreactive tracers and contaminants was injected into the aquifer over a 14.75-hour period. Nine wells were used to inject the solution.

A dense network of multilevel sampling wells was installed with horizontal spacing ranging from 1.0 to 4.0 m. Figure 1 shows the three-dimensional array of multilevel sampling wells. The vertical spacing of the sampling points ranged from 0.2 to 0.3 m. Samples were taken from the multilevel wells and analyzed for solute concentrations to provide "snapshots" in time of solute distributions. Over 11,000 samples were taken during 14 sampling sessions. The tracer test was monitored for 1038 days and the nonreactive plumes traveled more than 110 m.

Freyberg (1986) used the nonreactive tracer data (bromide and chloride) to calculate plume spatial moments, zeroth through second. The moments were used to calculate the total mass of solute, average plume velocity, and dispersivities. The moments were calculated in two dimensions; the vertical concentration values were

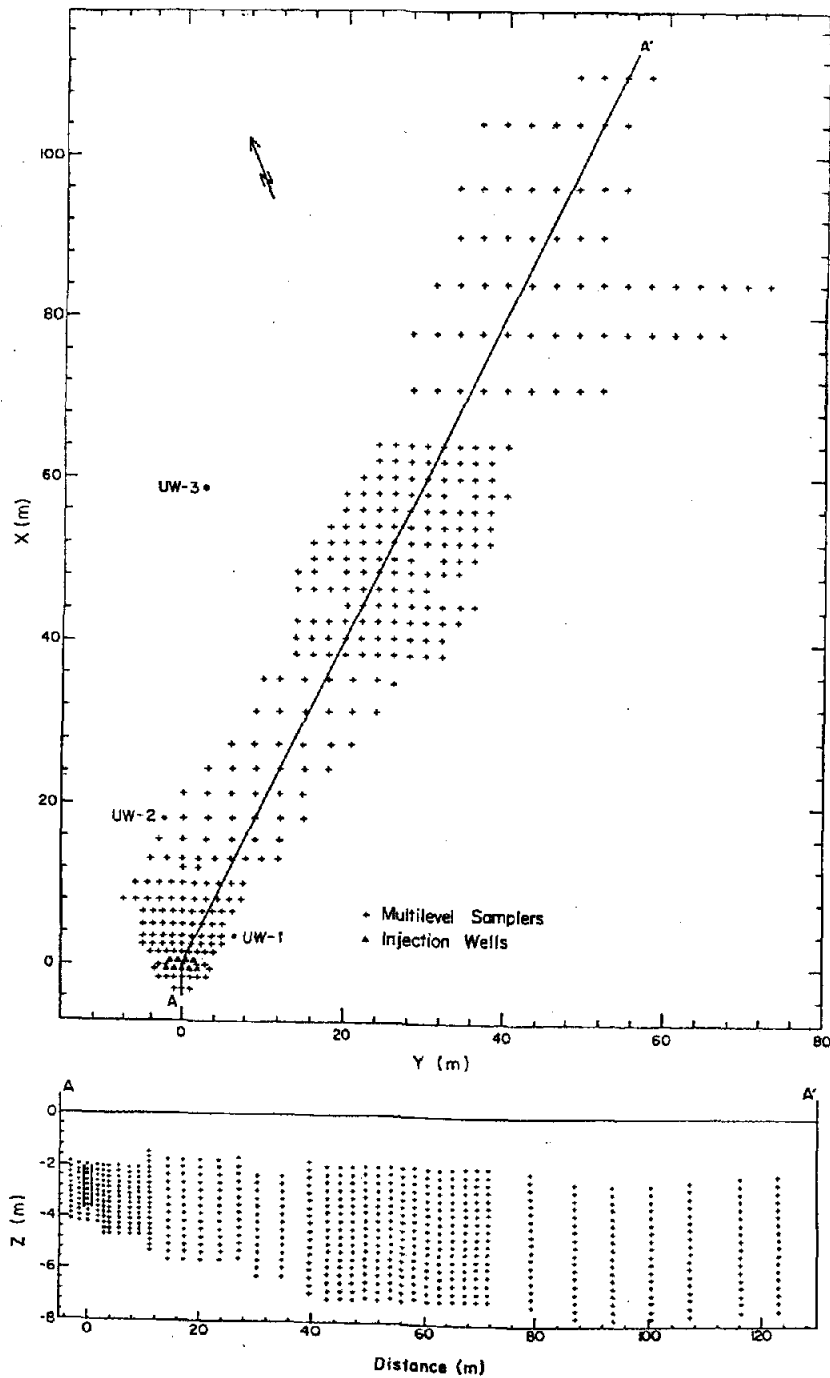


Figure 1. Three-dimensional array of multilevel sampling wells used to monitor the Borden natural gradient tracer test. Plus symbols represent multilevel sampling wells; triangles represent injection wells. Position labeled UW-3 is the location of the 32 cores used in calculating the geostatistical parameters of hydraulic conductivity. (Figure 5 of Mackay et al., 1986).

averaged. The average linear velocity of the plume was estimated at 0.091 m/day by examining the plume's center of mass as a function of time. The apparent dispersivities were calculated using the average plume velocity and the rate of change of plume variance with respect to time. The apparent longitudinal and horizontal transverse dispersivities were 0.36 m and 0.039 m, respectively.

Columbus Site

A large-scale natural gradient tracer experiment was conducted in an unconfined aquifer at Columbus Air Force Base in northeastern Mississippi. The purpose of the experiment was to increase the existing information gathered from natural gradient tracer tests for a very heterogeneous aquifer (Boggs et al., 1992). The study would expand the spectrum of aquifer conditions for transport model testing. In addition, this study examined practical field methods for estimating the spatial variability of hydraulic conductivity. As with the Borden experiment, detailed three-dimensional monitoring of solute concentrations was performed to provide "snapshots" of plume development through time.

The aquifer is composed of poorly to well-sorted sandy gravel and gravelly sand with small amounts of silt and clay. The mineralogy of the sands and gravels consists of chert, feldspar and mica. The fine-grained materials (< 0.074 mm) consist of quartz, potassium feldspar, muscovite, and clay minerals. The clay minerals are predominantly kaolinites and illites with small amounts of montmorillonites and vermiculites (actually altered biotite). The sediments are commonly coated with iron oxides (Boggs et al., 1992).

The unconfined aquifer consists of an alluvial terrace deposit and averages approximately 11 m in thickness. An aquifer exposure adjacent to the site indicated the facies occur as irregular lenses and layers of up to 8 m wide and 1 m thick. The aquifer is underlain by an aquitard consisting of fine-grained marine sediments belonging to the

Eutaw Formation (Boggs et al., 1992). The hydraulic head field of the site exhibits complex temporal and spatial variability as a result of the heterogeneity of the aquifer and seasonal fluctuations of the water table. The average horizontal hydraulic gradient is approximately 0.003, and exhibits seasonal periodicity as a result of water table fluctuations. At many locations the vertical hydraulic gradient is greater than the horizontal gradient.

Rehfeldt et al. (1992) examined the spatial variability of hydraulic conductivity at the site. A total of 2187 hydraulic conductivity measurements were taken throughout the site. Many methods were employed in taking the conductivity measurements including the borehole flowmeter method, slug tests, and laboratory permeameter tests. Estimates of covariance parameters were found using indirect measures of hydraulic conductivity such as soil grain size distributions, surface geophysical surveys, and mapping of sediment facies in outcrop.

The values of hydraulic conductivity varied considerably throughout the site. Near the injection site the mean value of hydraulic conductivity was approximately 10^{-3} cm/s, while the mean value of the far downgradient region of the test site was 1-2 orders of magnitude larger (Boggs et al., 1992), for a mean range $\ln K$ of -6.9 to -2.3 (a single mean $\ln K$ value was not given for the site). The covariance parameters were estimated by two different methods. The first method assumed second-order stationarity of the hydraulic conductivity field. This method indicates the covariance between two points depends only on the distance between them and not on their spatial location; thus, the hydraulic conductivity field was assumed to be uniformly heterogeneous. In other words, it was assumed that there are no spatial trends in hydraulic conductivity values; features such as abandoned channel deposits were treated as large-scale heterogeneities. The values of $\ln K$ variance and correlation scales in the horizontal and vertical directions were found by this method to be 4.5, 12.8 m, and 1.6 m, respectively (Rehfeldt et al.,

1992). The hydraulic conductivity field was found, however, to have a nonstationary character (i.e. spatial trends in hydraulic conductivity exist).

The second method used to calculate the covariance parameters assumed a nonstationary character and involved trend removal and the subsequent covariance analysis of the stationary residuals. Trend analysis is a mathematical method of separating a map into two components; that of a regional nature (trend), and local fluctuations (residuals) (Davis, 1986, p. 405). Thus, residuals are deviations from the general trend. The resulting values of variance and correlation scales in the horizontal and vertical directions using the trend removal method were found to be 2.7, 4.8 m, and 0.8 m, respectively (Rehfeldt et al., 1992).

A total of 10.07 m³ of groundwater containing bromide and three organic tracers was pulse injected into the aquifer over a period of 48.5 hours. The slow injection rate was chosen to keep the disturbance of the natural flow system to a minimum. Five injection wells spaced 1 m apart in a linear array were used in the injection.

The monitoring system consisted of an array of 258 multilevel sampling wells (Boggs et al., 1992), having 6000 three-dimensional sampling points with which to monitor the plumes. The emphasis of the monitoring program, as with the Borden site, was to provide snapshots of plume development through time. Eight sampling sessions were performed during the experiment at intervals of five to 19 weeks. The experiment was monitored for 594 days.

Adams and Gelhar (1992) performed a spatial moment analysis on the bromide concentration data. An apparent longitudinal dispersivity value of 50-75 m was calculated from second-order spatial moments of the plume. The relationship between the second-order spatial moment and dispersivity assumes a uniform flow field with a Gaussian concentration distribution were present. The site, however, was shown to have a nonuniform flow field and non-Gaussian plume concentration distributions. The flow

field was found to be accelerating in a downgradient direction; the groundwater velocity increased tenfold 100 m downgradient from the injection site. The researchers therefore performed a dispersivity calculation which assumed a nonuniform flow field. A longitudinal dispersivity value of 5-10 m was calculated using this approach, which is an order of magnitude larger than that of the weakly heterogeneous Borden site.

A decreasing mass balance for bromide throughout the experiment prompted studies to be performed to determine if bromide was being adsorbed onto the aquifer sediments (Boggs and Adams, 1992). As discussed earlier, researchers have shown that sediments containing significant quantities of variably charged minerals such as Fe and Al oxides may adsorb anionic tracers (Uehara and Gillman, 1981, p. 46). The pH governs the sign and magnitude of the oxide surface charge. The presence of iron oxides and kaolinite, in conjunction with the relatively low pH at the site (approximately 4.8), indicated that anion adsorption may be a factor (Boggs and Adams, 1992).

A column study was performed to estimate the effects of bromide adsorption relative to the conservative tracer, tritium. A linear sorption isotherm described the sorption that occurred in the column studies. The results of the bromide column studies indicated that approximately 20% of the bromide was being adsorbed. A retardation factor of 1.20 was estimated for bromide (Boggs and Adams, 1992). Boggs and Adams (1992) did not, however, test if the sorption was a function of concentration; one bromide concentration of 500 mg/l was used in the column studies.

Sorption was tested at the Columbus site because it can have profound effects on plume spreading. The effects of sorption were well illustrated in a natural gradient tracer test at Cape Cod, Massachusetts (LeBlanc et al., 1991). Figure 2 shows plume "snapshots" of bromide, lithium, and molybdate through time. It is clear from Figure 2 that the longitudinal spreading of the molybdate plume at 461 days was much larger than

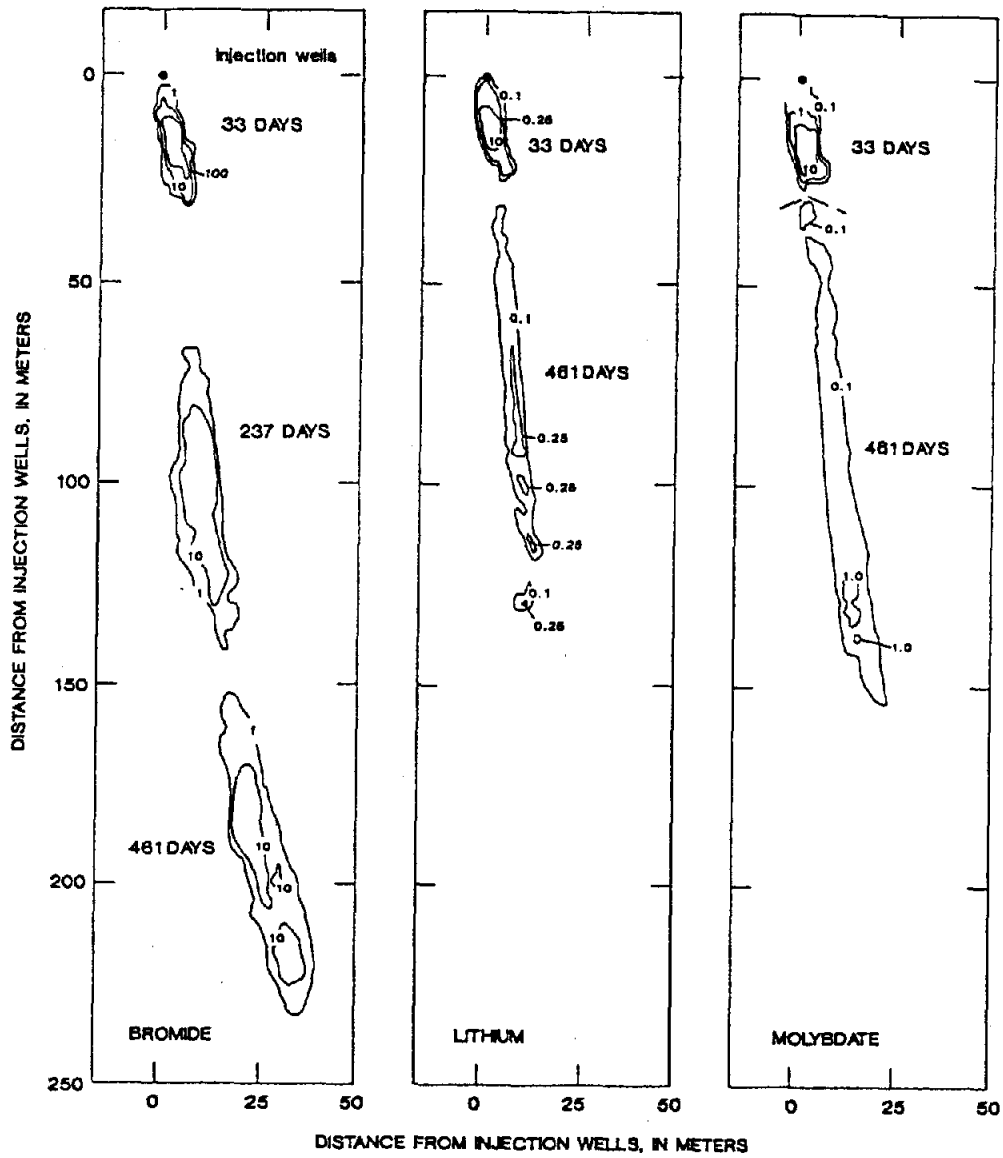


Figure 2. Plume "snapshots" of bromide, lithium, and molybdate at the Cape Cod natural gradient tracer test (Figure 10 of LeBlanc et al., 1991).

the conservative bromide plume. The enhanced longitudinal spreading was attributed to sorption (LeBlanc et al., 1991).

Natural gradient tracer tests are an important tool in examining the transport of solutes in a subsurface environment. They are, however, quite costly to perform. Many researchers are turning to numerical groundwater modeling to quantify solute transport because it is a cost-effective alternative to field natural gradient tracer tests.

Numerical Studies

The Effect of Sorption on Dispersion

Sorption of contaminants or tracers can profoundly affect plume spreading and dispersivities. Recently, researchers have been modeling the effects of heterogeneous linear and non-linear sorption in heterogeneous aquifers (e.g. Burr et al., 1994; Brusseau, 1995; Bosma et al., 1996). They have found that sorption can enhance longitudinal dispersion. Transverse dispersion, however, does not seem to be affected as much.

Burr et al. (1994) performed a limited Monte Carlo analysis with geostatistical parameters similar to the Borden aquifer. The aim of the study was to examine the effects of linear sorption in which the distribution coefficient was assumed to be negatively correlated with hydraulic conductivity. This indicates the solute will be strongly sorbed in areas of low hydraulic conductivity. This correlation was chosen because it would maximize field-scale nonequilibrium effects on solute movement; there is, however, no evidence to suggest such a correlation exists at the Borden site (Burr et al., 1994). The researchers found that the longitudinal macrodispersivity was more than 3 times greater for the reactive tracer than that of a conservative tracer. In the horizontal and vertical transverse directions, dispersivities were only slightly greater.

Bosma et al. (1996) performed a number of two-dimensional Monte Carlo simulations in aquifers with homogeneous and heterogeneous flow fields and Freundlich isotherms. They performed simulations with random hydraulic conductivity fields of

different variances and different Freundlich isotherm parameters. In addition, they performed simulations where the Freundlich sorption coefficient was heterogeneous; that is, the sorption coefficient was treated as a random variable with a given natural log mean and variance. In the heterogeneous flow fields with homogeneous sorption, Bosma et al. found the longitudinal and horizontal transverse variances increased with increasing non-linearity of the Freundlich isotherm. As stated above, the Freundlich exponent can take on values between 0 and 1. They found that exponent values as large as 0.8 produced enhanced longitudinal variance relative to the linear model ($n = 1$ in the Freundlich isotherm). Chemical heterogeneity only made a small difference in longitudinal variance, and virtually no difference in the transverse variance. The authors did not, however, compare the results to a conservative tracer.

Brusseau (1995) examined the effects of non-linear sorption and rate-limited transformation on the transport of contaminants through a porous medium. Numerical simulations that take into account non-linear sorption and rate-limited transformation were performed in one dimension. Breakthrough curves were derived from the simulations to assess the effects of sorption and rate-limited transformation of a contaminant as it moves past a point in the system. The study compared non-linear sorption and rate-limited transformation relative to that of linear sorption. The results of the study indicated that rate-limited transformation and transport of non-linearly sorbing solutes with exponents less than 0.9 cannot be accurately simulated with a linear sorption isotherm model.

Apparently, none of the studies to date has compared the effects of weakly non-linear sorption relative to that of a conservative tracer. Generally, researchers have compared non-linear sorption to linear sorption (Brusseau, 1995; Bosma et al., 1996), or linear sorption relative to a conservative tracer (Burr et al., 1994). In addition, many previous studies considered sorption isotherms which were designed to show the effects

of varying the sorption coefficients, exponents, and chemical heterogeneity; the sorption isotherms were not experimentally derived (Burr, et al., 1994; Brusseau, 1995; Bosma et al., 1996). Most of the sorption isotherms used in previous studies have been moderately strong. Burr et al. (1994) calculated a retardation factor of approximately 4; Bosma et al. (1996) calculated values as large as 26 (numerous simulations were performed with varying degrees of sorption and retardation).

In contrast to the work reviewed above, the aim of this research was to quantify the effects of a tracer undergoing sorption according to a weakly non-linear, experimentally derived Freundlich isotherm relative to a conservative tracer.

Recent Modeling Trend

Many researchers have performed Monte Carlo simulations with stationary random hydraulic conductivity fields (e.g. Burr et al., 1994; Bosma et al., 1996). This assumes that the statistical parameters do not vary spatially. Webb and Anderson (1996) argue that the problem with this assumption is that the presence of high conductivity pathways in a random field are not correlated to any known geologic structures; they are a function of the variability of the statistical distribution.

Recently, researchers have been generating geometric hydraulic conductivity fields in three dimensions which more realistically represent a given geologic system and its depositional environments (Webb and Anderson, 1996; Scheibe and Freyberg, 1995). Webb and Anderson (1996) argue that the use of a random hydraulic conductivity field underutilizes the existing knowledge of geologic systems. They proposed a method for modeling a braided stream system which takes into account the geometry of features common to the braided stream depositional environment. The subsurface features used in the model were inferred from surface topographical features.

Scheibe and Freyberg (1995) performed a similar study on point bar sediments of the Wabash River in the midwestern United States. They generated a model with aquifer

parameters of features at the microform to macroform scale. The microforms include such small-scale features as current ripples; the macroforms are large features which represent the cumulative effect of multiple events over long periods of time, such as point bar deposits. The grid spacing of the model was 0.00381 m to insure that the resolution of the model was sufficient for the smallest-scale features. The resulting high-resolution model was said to accurately characterize the depositional features found in a point bar system.

METHODS

This study utilized the groundwater modeling programs SUTRA (Voss, 1984) and WATSUTRA (VanderKwaak et al., 1996) to perform a limited Monte Carlo analysis. The Fast Fourier Transform (FFT) method of random field generation (Gutjahr et al., 1995) was used to generate random hydraulic conductivity fields with given covariance parameters. The numerical models were tested to confirm they worked properly. Apparent dispersivities were calculated by the method of moments. The ensemble mean estimates for various simulation cases were tested for significance using null hypothesis testing with a standard t-test.

Transport Modeling Using SUTRA and WATUSTRA

The numerical simulations in this study were done using SUTRA and WATSUTRA. SUTRA, an acronym for Saturated-Unsaturated TRansport, is a United States Geological Survey finite element, two-dimensional simulation model. This program simulates fluid movement and the transport of either energy or dissolved substances in the saturated or unsaturated zones. SUTRA also contains routines for modeling sorption, including the Freundlich isotherm. WATSUTRA is a modified version of SUTRA with an iterative sparse matrix solver, WATSOLVE (VanderKwaak et al., 1995), which speeds up calculations and conserves memory. The codes for SUTRA and WATSUTRA were written in ANSI-Standard FORTRAN-77.

WATSUTRA was found to be about 15 times faster than SUTRA for simulations requiring an iterative solution. WATSUTRA performed the simulations reported herein

in approximately 20 hours. The iterative solution is required to resolve non-linearities, such as for a non-linear sorption isotherm. The conservative runs were done using SUTRA with a non-iterative solution. The simulations were performed on 100 and 120 megahertz (MHz) Pentium PCs, both equipped with 32 megabytes (Mb) of random access memory (RAM). The simulations were run on FORTRAN Powerstation version 1.0 for Windows.

Model Parameters

The simulations in this study were designed to replicate natural gradient tracer tests in two different flow fields, weakly and strongly heterogeneous. Figure 3 shows the basic model design used in all simulations in this study. The model consisted of 9425 nodes and 9216 elements (145 by 65 nodes, 144 by 64 elements). This size was chosen because it was big enough to facilitate a large amount of plume spreading without losing tracer mass through the constant-pressure boundaries. Each element was 1 m by 1 m and was of unit thickness. All model boundaries were constant-pressure (head). The left and right sides had constant pressure nodes of $103524.7 \text{ kg/m} \cdot \text{s}^2$ and $97886.4 \text{ kg/m} \cdot \text{s}^2$, respectively. The top and bottom constant pressure nodes decreased at a constant rate from the left to right sides. This gave a uniform hydraulic gradient from left to right of 0.004. Other researchers (e.g. Burr et al., 1994; Bosma et al., 1996) have modeled with no-flow boundaries on the top and bottom to allow flow parallel to the x-axis. The constant head boundaries were chosen to better represent a field situation. All elements had a porosity of 0.35. The experimentally derived Freundlich isotherm in Eq. (1) was used over the entire model domain for the reactive simulations (i.e. chemically homogeneous). A chemically homogeneous model was used in this study because there is little sorption data available at the Savannah River Site to model chemical heterogeneity.

Constant Head Boundary
Pressure = $103524.7 \text{ kg/m}^3 \cdot \text{s}^2 - (39.1549) \cdot (J)$
where J is the distance in meters from the left boundary

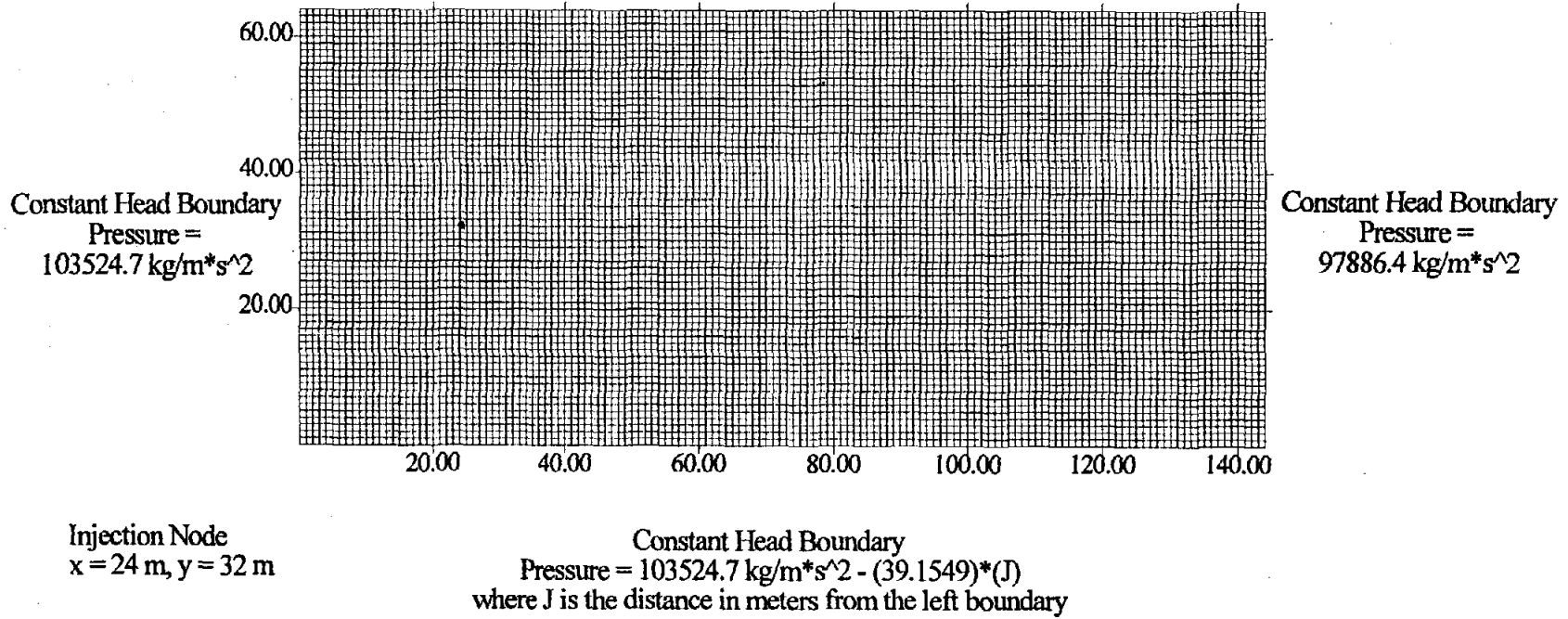


Figure 3. Two-dimensional model used for all simulations in this study.

Local dispersivity values (model input values which represent pore-scale dispersivity) in the longitudinal and horizontal transverse directions were 0.6 m and 0.03 m, respectively, for all simulations. The local dispersivity values used in this study are larger than those used by other researchers (e.g. Burr et al., 1994 used 0.1 m and 0.005 m, respectively). The local dispersivity values were chosen to facilitate enough plume spreading so that the plume front covered a minimum of five elements, a discretization rule of thumb for SUTRA (Voss, 1984, p. 231). It was also found that large discrepancies occurred in the moment calculations between using all concentrations and only those greater than 0.1 ppm when local dispersivity values were small. For example, realization 1 in a weakly heterogeneous flow field was simulated using local longitudinal and horizontal transverse dispersivities of 0.2 m and 0.001 m, respectively. The zeroth and second moment calculations using concentrations greater than 0.1 ppm were found to be as much as twice as large as the same calculations using all concentrations. When the local longitudinal and horizontal transverse dispersivities were given values of 0.6 m and 0.03 m, respectively, the zeroth and second moment calculations using all concentrations and only those above 0.1 ppm generally agreed within a few percent. The larger local dispersivity values will not affect the results of this study because the same values were used in the reactive and conservative cases.

The user must specify the convergence parameters for pressure and concentration in SUTRA and WATSUTRA for simulations requiring an iterative solution. The simulations in this study were designed to quantify the effects of the Freundlich isotherm in Eq. (1) on apparent dispersivities. The concentration convergence parameter should be small enough so that all significant figures in the Freundlich isotherm are utilized. In addition, the convergence criteria should be accurate enough to facilitate sorption of the smallest concentration values used in the dispersivity calculations (detection limit set at 0.1 ppm). A convergence parameter of 1.00×10^{-8} was found to accurately characterize

the Freundlich isotherm in Eq. (1) for the concentrations used in this study. This value is small enough to include all significant figures in the Freundlich isotherm and will accommodate sorption of the smallest concentration values used in this study. The same value was used for pressure convergence. Thus, the change in concentration or pressure from the previous to the current iteration had to be less than or equal to 1.00×10^{-8} at all nodes for convergence.

The permeability values were generated for each element using a FFT random field generator (Gutjahr, et al., 1995). This random field generator uses the turning bands method of field generation. The turning bands method generates the random field along lines in space rather than synthesizing the multidimensional field directly (Bras and Rodriguez-Iturbe, 1985, p. 310). This is an efficient method of generating a random field. The fields on average are of mean zero, with the variance specified by the user (Gutjahr, et al., 1995).

The random $\ln K$ fields generated in this study were stationary; that is, the covariance parameters did not vary spatially. The input parameters in the FFT random field generator consisted of a $\ln K$ variance, correlation lengths in the x and y directions, spectral model type, size and length of the field in the x and y directions, and a seed number. The input parameters were chosen to be similar to the Borden and Columbus aquifer parameters to represent two extreme aquifer conditions, weakly and strongly heterogeneous, respectively. The variance of the weakly and strongly heterogeneous flow fields, 0.35 and 3.5, respectively, were chosen to differ by an order of magnitude. Both fields had the same correlation length of 5 m in the x and y directions. The anisotropic exponential covariance model (spectral model) was chosen in the random field generator because the statistical characterization of hydraulic conductivities at the Borden and Columbus sites assumed an exponential covariance function (Sudicky, 1986; Woodbury and Sudicky, 1991; Rehfeldt et al., 1992). In addition, Monte Carlo simulations with the

geostatistical characteristics of the Borden aquifer performed by Burr et al. (1994) also used an exponential covariance function in generating random hydraulic conductivity fields. The size of the random field was chosen to be 256 by 64, in the x and y directions, respectively, because the random field generator required the number of values in the x and y directions to be a power of 2. The field was therefore truncated to attain the model dimensions of 144 by 64. Finally, the seed number tells the random field generator where to begin the random field generation. Holding all other input parameters constant, a unique seed number will generate a unique realization. Each realization generated by the random field generator can therefore be replicated by using the same input parameters. The mean and variance of the entire field and truncated field, and the seed numbers used are shown in Tables 1 and 2 for the weakly and strongly heterogeneous fields, respectively.

A FORTRAN program was written to shift the $\ln K$ mean from about 0 to about -5, convert $\ln K$ to permeability, and print the results in the proper format for SUTRA's input files (see appendix E). The mean hydraulic conductivity value used in both fields was 6.74×10^{-3} cm/s ($\ln K = -5$). This value is characteristic of a clean sand (Freeze and Cherry, 1979, p. 29) and is the approximate mean value found at the Borden and Columbus sites.

Ten random hydraulic conductivity fields were generated in this study; five weakly heterogeneous and five strongly heterogeneous. Each of the ten realizations was used for four simulations: conservative tracer with 1 kg of bromide, reactive tracer with 1 kg of bromide, conservative tracer with 0.1 kg of bromide, and reactive tracer with 0.1 kg of bromide. Forty simulations were performed. Each simulation consisted of three smaller simulations: injection (600 s time-steps), first 100 hours of simulation (200 s time-steps, a size necessary for convergence when implementing the Freundlich isotherm), and the two year simulation (8640 s time-steps).

Table 1. Random field generator input and output parameters for a weakly heterogeneous flow field.

Realization Number	Seed Number	Output Variance	Truncated Variance	Output Mean	Truncated Mean
1	267913	0.335	0.317	0.059	2.71E-04
2	70269	0.316	0.317	-0.095	-0.075
3	392279	0.322	0.346	0.088	0.102
4	693471	0.373	0.410	-0.035	-0.040
5	7779433	0.327	0.272	-0.018	0.012
Average	na*	0.335	0.333	-3.34E-04	-9.68E-05

* denotes not applicable

Table 2. Random field generator input and output parameters for a strongly heterogeneous flow field.

Realization Number	Seed Number	Output Variance	Truncated Variance	Output Mean	Truncated Mean
1	70269	3.161	3.173	-0.300	-0.237
2	112643	3.332	3.733	-0.335	-0.398
3	11262	3.157	3.102	-0.247	-0.349
4	267913	3.355	3.166	0.185	0.001
5	693471	3.735	4.105	-0.110	-0.125
Average	na*	3.348	3.456	-0.161	-0.222

* denotes not applicable

The ensemble means were estimated by averaging the apparent longitudinal or horizontal transverse dispersivities in the five realizations for each simulation case. As stated in the Background section, Burr et al. (1994) found this number to be sufficient to provide a good estimate of the ensemble mean in a weakly heterogeneous flow field. This estimate will probably not be as good in the strongly heterogeneous flow field because plume variance is much greater. This number of realizations will, however, give some insight into the effects of the Freundlich isotherm in Eq. (1) on apparent dispersivities.

The following simplifying assumptions were used in this study:

- 1) Sorption according to the Freundlich isotherm was assumed to be a reversible equilibrium process. Thus, desorption followed the same isotherm as adsorption, only with decreasing concentrations, and both adsorption and desorption were rapid in comparison to advective flow.
- 2) The Freundlich isotherm was constant throughout the model domain.
- 3) The groundwater model employed was two-dimensional; vertical heterogeneity was not represented.

Model Testing

Model testing in solute transport is crucial in determining if the output parameters are reasonable. A homogeneous simulation can be used to test the local dispersivity values put into the modeling program; the local dispersivity values should be equal to the calculated dispersivity values. Heterogeneous models can be tested for grid spacing and time-step size. If the grid spacing and time-step size are too large, a phenomenon called numerical dispersion will cause the dispersivity values to be overestimated.

A homogeneous natural gradient tracer simulation was performed with SUTRA. The homogeneous model had a single value of permeability, $1.025 \times 10^{-11} \text{ m}^2$, throughout the model domain. The model had dimensions of 316 m by 20 m in the x and

y directions, respectively, with a grid spacing of 1 m. A local longitudinal dispersivity value of 4.000 m was used in SUTRA; the value calculated by the method of moments, 3.979 m, was within 0.5 percent of the local dispersivity value (see Appendix F for spatial moment estimates). A local horizontal transverse dispersivity value of 0.1000 m was put into SUTRA and an apparent horizontal transverse dispersivity value of 0.1005 m was calculated by the method of moments. Again, the calculated value was with 0.5 percent of the local value. Therefore SUTRA was performing as expected.

The type of comparison outlined above cannot be performed in heterogeneous aquifers because the heterogeneities in the flow field enhance dispersivity. Therefore the heterogeneous model was tested for spatial and temporal discretization. Discretization is a measure of the fineness of the grid spacing and the size of the time steps. An inadequate model discretization may lead to numerical dispersion. Numerical dispersion is a result of difficulties in solving the advection-dispersion equation (Eq. 5) numerically (Anderson and Woessner, 1992, p. 327). It is minimized by using a fine grid size and small time steps (dx and dt , respectively, in Eq. 5). Increased accuracy must be balanced with the increased CPU time required for the simulations. A finer grid size and smaller time steps will require more calculations for a given simulation time.

A pecllet number can be calculated to estimate a grid spacing that minimizes numerical dispersion. The pecllet number for grid spacing is defined as the length of the node spacing divided by the characteristic dispersivity value, $\Delta L/\alpha$. Acceptable solutions may be obtained with pecllet numbers as high as 10 (Huyakorn and Pinder, 1983). Burr et al. (1994) found that using a grid spacing in the x and y directions of 0.5 m and a horizontal transverse dispersivity of 0.005 m (pecllet number of 100) appeared to accurately characterize dispersivities. The length of the node spacing for the model used in this research was 1 m, the characteristic dispersivity was 0.03 m (the local horizontal transverse dispersivity). The pecllet number was therefore 33.3.

A more direct way to test the effect of grid spacing on numerical dispersion for a specific model is to increase the number of nodes, repeat the simulation, and compare the dispersivity values (Sudicky, 1996, personal communication). If numerical dispersion is occurring, the dispersivity values will be larger in the model with a smaller number of nodes.

The grid size discretization was tested by breaking each element into four elements of the same permeability (see Appendix E for the FORTRAN program written to do this). The number of nodes and elements was increased by a factor of four, to 37,281 nodes and 36,864 elements. This test was performed on a simulation where plume dispersion was small, a reactive simulation in the weakly heterogeneous flow field (realization 1 with 1 kg of bromide). Numerical dispersion is maximized in cases where the plume occupies a minimum number of nodes.

The grid size test was run on WATSUTRA. The CPU run-time for the grid size test was 93 hours using a 100 MHz Pentium PC equipped with 56 Mb of RAM. Using all concentrations in the moment calculations, the simulation with the finer discretization yielded dispersivity values 5.7 percent smaller in the longitudinal direction and 3.0 percent smaller in the horizontal transverse direction than the 9425 node model (see Appendix E for grid-size test moment programs and Appendix F for moment calculations). Because numerical dispersion apparently accounted for a relatively small amount of the dispersivity, the original model of 9425 nodes was assumed to be adequate to compare the spreading of the plumes of conservative tracers to weakly adsorbed tracers.

The Courant number can be calculated to estimate a time step size that minimizes numerical dispersion. The Courant number is defined as the average velocity times the time step divided by the node spacing, $v_i \Delta t / \Delta L$. The Courant number should be less than or equal to one (Anderson and Woessner, 1992, p. 327). Using the largest time step in

the simulations (8640 seconds) and the largest value of velocity (7.5×10^{-7} m/s), the Courant number was 6.48×10^{-3} , which is ≤ 1 . Thus, the time-step size of 8640 seconds was small enough to minimize numerical dispersion.

The temporal discretization was also tested by running a 7-day simulation and examining the concentration at a given node as the time step size was decreased. At some point, the concentration at the node should level off as the time step size is decreased, thus converging on the true value. This test was simulated with realization 1 in a strongly heterogeneous flow field with dimensions of 196 m by 36 m, respectively, in the x and y directions. The 8640 second time step was chosen to be sufficiently small (see Table 3).

Table 3. The effects of time step size on concentration values at a given node for a 1 week simulation in a strongly heterogeneous flow field.

Time-step size (s)	Number of time steps	Concentration at node 574 (mass fraction)
6.048×10^5	1	4.847×10^{-5}
8.640×10^4	7	4.553×10^{-5}
4.320×10^4	14	4.521×10^{-5}
1.728×10^4	35	4.502×10^{-5}
8.640×10^3	70	4.495×10^{-5}
4.320×10^3	140	4.492×10^{-5}

The reactive simulations were performed using WATSUTRA. This modified version of SUTRA has only been tested for transient solute transport and flow (VanderKwaak, 1996, personal communication). WATSUTRA was tested to confirm it would provide the same solute solution as SUTRA. Details are given in Appendix F.

Method of Moments

Calculations of dispersivity and velocity were done using the method of moments. The method of moments can be used to estimate many parameters describing the solute plume through time: total mass of solute in solution, average linear velocity, longitudinal and horizontal transverse dispersivities, skewness, and kurtosis. These parameters are important in quantifying how the solute plume changes through time. The moments, zeroth through fourth, were calculated using a FORTRAN program (see Appendix E).

The general spatial moment equation for the zeroth and first moment in two dimensions is (Freyberg, 1986):

$$M_{ij}(t) = \iint n C(x,y,t) x^i y^j dx dy \quad (6)$$

where $C(x,y,t)$ is the mass concentration of the solute at a given node at time t , x and y are the spatial coordinates, and $0 \leq i + j \leq 1$ are integers that define the moment. The zeroth moment, M_{00} , is obtained when $i + j = 0$ and is the total mass of solute in solution. The first moment is defined when $i + j = 1$. When normalized by the total solute mass in solution, M_{00} , this represents the location of the center of mass of the plume in the x -direction, x_c , or y -direction, y_c :

$$x_c = M_{10} / M_{00} \quad y_c = M_{01} / M_{00} \quad (7)$$

Average linear velocity in the x -direction (direction of flow) can be estimated from the rate of change of the center of mass with respect to time:

$$v = dx_c / dt \quad (8)$$

The average velocity can be estimated by plotting x_c as a function of time. A linear regression can then be performed to find the slope of the best-fit line through the points.

The retardation factor for a reactive simulation can be calculated from the average linear velocities of the conservative and reactive cases for a given realization. The retardation factor, R , is the ratio of the conservative velocity to the reactive velocity:

$$R = v_c / v_r \quad (9)$$

where v_c is the average linear velocity of the conservative plume and v_r is the average linear velocity of the reactive plume. The larger the retardation factor, the slower the reactive plume moves relative to the conservative plume.

The generic spatial moment equation for the second and higher moments in two dimensions is (Farrell et al., 1994):

$$M_{ij}(t) = \iint n C(x,y,t) (x-x_c)^i (y-y_c)^j dx dy \quad (10)$$

The second spatial moment is defined when $i + j = 2$. When normalized by the total mass in solution, the spatial variance about the center of mass is obtained:

$$\sigma_{xx}^2 = M_{20} / M_{00} \quad \sigma_{yy}^2 = M_{02} / M_{00} \quad (11)$$

Apparent dispersivities in the longitudinal (A_L) and horizontal transverse (A_T) directions can be obtained from the following relationships (Freyberg, 1986):

$$A_L = (1 / |2v|) (d\sigma_{xx}^2 / dt) \quad A_T = (1 / |2v|) (d\sigma_{yy}^2 / dt) \quad (12)$$

The quantity $d\sigma^2/dt$ in Eq. (12) can be estimated by plotting $d\sigma^2$ as a function of time. A linear regression can then be performed to find the slope of the best fit line through the points. As will be discussed later, however, many of the plots were not linear. Freyberg (1986) coined the term "apparent dispersivity" because the relationships in Eq. (12) only produce a true dispersivity value for a Gaussian (normal) solute distribution. When the solute distribution is not normal, the relationships in Eq. (12) are only estimates of true dispersivities. Loaiciga (1988) pointed out the relationship in Eq. (12), without the velocity term, was originally derived by Einstein (1905) to describe Brownian motion of particles immersed in a fluid solution. Equivalent equations for longitudinal and horizontal transverse dispersivities are (Garabedian et al., 1991; Adams and Gelhar, 1992):

$$A_L = (1/2)(d\sigma_{xx}^2 / dx_c) \quad A_T = (1/2)(d\sigma_{yy}^2 / dx_c) \quad (13)$$

since average velocity, v , is defined as dx_c/dt (the dt terms cancel out). This is a more straightforward way to calculate apparent dispersivities because only one linear regression is required.

The third moment is obtained when $i + j = 3$. The coefficient of skewness of the plume in the x and y directions can be estimated from the third moment through the following equations:

$$SK_x = M_{30} / (M_{00}(\sigma_{xx}^2)^{3/2}) \quad SK_y = M_{03} / (M_{00}(\sigma_{yy}^2)^{3/2}) \quad (14)$$

The coefficient of skewness is a unitless measure of asymmetry (Yevjevich, 1972, p. 110). The coefficient of skewness, because it is an odd-valued moment, can have positive or negative values. A value of zero indicates the plume is symmetrical. A

positive skewness indicates tailing to the right of the mode (in a down-gradient direction); a negative skewness indicates tailing to the left of the mode (in an up-gradient direction) (Haan, 1977, p. 51).

The fourth moment is obtained when $i + j = 4$. The kurtosis of the concentration distribution in the x and y directions can be estimated using the fourth moment in the following equations:

$$KR_x = M_{40} / (M_{00}(\sigma_{xx}^2)^2) \quad KR_y = M_{04} / (M_{00}(\sigma_{yy}^2)^2) \quad (15)$$

The kurtosis of a concentration distribution is a measure of peakedness or flatness. A value of 3 indicates a normal distribution (Haan, 1977, p. 51). A kurtosis value greater than three indicates a distribution more peaked than a normal distribution. The value of kurtosis, because it is based on positive moments, will always be positive (Yevjevich, 1972, p. 109).

Values for skewness and kurtosis are susceptible to error because they are based on quantities raised to the third or fourth power. Thus, a small error in concentration values due to numerical dispersion will be magnified when it is multiplied by a moment arm raised to the third or fourth power.

Moment Program Testing

The FORTRAN programs written to calculate the spatial moments were tested for each of the moments. The zeroth moment was tested by comparing its value to that of the mass of bromide injected. A nearly identical mass was obtained. The first moment was tested by plotting the bromide plume in SURFER (Golden Software, 1994) and estimating the center of mass. The plume in the homogeneous simulation was symmetrical and it was therefore easy to determine its center of mass. The calculated and visual center of mass were found to be in agreement.

The second moment was tested in a homogeneous aquifer by calculating the longitudinal and horizontal transverse dispersivities and comparing them against those put into SUTRA. In homogeneous sediments dispersivities are simply a result of the local dispersivity values put into SUTRA. As stated earlier, the input and calculated values of longitudinal and horizontal transverse dispersivities agreed within 0.5 percent. The weakness in this method of verification is that the second moment calculations were tested simultaneously with the performance of SUTRA. The second moment equation, however, is straightforward and has the same basic form as the higher-order moments.

The third and fourth moments were tested by inserting concentrations of 1×10^{-4} at 41 nodes in a symmetrical diamond-shaped pattern. All arrays and parameters in the moment program were declared as double-precision (14 significant figures). The skewness values in the x and y directions were found to be approximately zero (-0.4×10^{-13} and -0.6×10^{-13} , respectively). The kurtosis values in the x and y directions had a value of 2.5. This seemed reasonable since the pattern looked similar to a normal distribution (kurtosis of 3). In addition, the center of mass of the plume in the x and y directions was found to be in the exact middle of the diamond-shaped pattern. The plume variances in the x and y direction were equal, as would be expected for a symmetrical pattern. It was concluded that the moment programs accurately calculated the moments.

Null Hypothesis Testing

Null hypothesis testing can be employed to test if the ensemble mean estimates are significantly different. The null hypothesis states the two ensemble means are equal: the mean of the population from which the first sample was drawn is the same as the mean of the parent population of the second sample. The ensemble mean estimates can be tested with a standard two-tailed t-test. This test uses the difference between the means and a pooled standard deviation to calculate the t value. If the calculated t value is

greater than the table value for a given number of degrees of freedom, the null hypothesis is rejected.

There are three assumptions in using the t-test (Davis, 1986, p. 66). The first is that both samples were selected at random. This assumption is believed to hold for this study because the results are based on randomly generated hydraulic conductivity fields. The second assumption is that the populations from which the samples were drawn is normally distributed. This assumption is impossible to test with a limited number (i.e. five) of realizations. The third assumption is that the two samples have the same variance. This can be tested with an F-test and confirmed before the t-test is employed.

It was concluded that, based on tests previously described, SUTRA and the method of moments FORTRAN program were giving acceptable results for the tasks required for this research.

RESULTS AND DISCUSSION

Forty simulations were performed in this study for eight cases. Five realizations were performed for each case. The eight cases were:

strongly heterogeneous flow field with

- (1) conservative simulations with 1 kg of bromide;
- (2) reactive simulations with 1 kg of bromide;
- (3) conservative simulations with 0.1 kg of bromide;
- (4) reactive simulations with 0.1 kg of bromide;

weakly heterogeneous flow field with

- (5) conservative simulations with 1 kg of bromide;
- (6) reactive simulations with 1 kg of bromide;
- (7) conservative simulations with 0.1 kg of bromide;
- (8) reactive simulations with 0.1 kg of bromide.

Each realization was simulated for two years (with two exceptions). All four cases in the weakly heterogeneous flow field were performed with the same five generated hydraulic conductivity fields. Similarly, all four cases in the strongly heterogeneous flow field were performed with each of the five generated hydraulic conductivity fields. This allowed direct comparisons between the reactive and conservative simulations, and the 0.1 and 1 kg simulations. In addition, the moment calculations were performed two ways

for each case, using (1) all concentrations and (2) only those above the detection level (set at 0.1 ppm).

Simulations with 1 kg of Bromide

Weakly Heterogeneous Flow Field

Ten simulations were performed in the weakly heterogeneous flow field with 1 kg of bromide, five conservative and five reactive. SURFER contour plots of plume "snapshots" are given to provide a visual perspective of plume behavior (see Appendix B). All contour plots of the plumes were done using concentrations greater than 0.1 ppm. The reactive simulations are only shown at $T = 2$ years (with two exceptions) because the plumes are similar to the conservative simulations at $T = 1$ year. SURFER contour plots of the weakly heterogeneous natural log hydraulic conductivity fields, given in Appendix C, are of the same scale as the plume "snapshots" and can be overlaid to examine the effects of heterogeneities on plume spreading.

Spatial Moment Estimates of Plume Behavior

Mass of Bromide in Solution. Moment calculations, zeroth through fourth, are given in Appendix A. In all conservative realizations, the zeroth moments are reasonably close to 1 kg. As expected, there is a slight difference (a few percent) between using all concentrations and omitting those below 0.1 ppm. The zeroth moments for the reactive simulations showed a steady decline in mass through time. This was predominantly a result of concentration mass loss through adsorption according to the Freundlich isotherm in Eq. (1). At 2 years about 42 percent of the bromide injected was adsorbed onto the aquifer sediments.

Center of Mass. The first moments, when normalized by the total mass in solution, yields the plume center of mass. Plots of the center of mass in the x-direction as a function of time are shown in Appendix D. The average linear velocity for each realization was calculated from the slope of a best-fit line through the points on the graph.

Table 4. Dispersivities, velocities, and retardation factors for simulations in a weakly heterogeneous flow field with 1 kg of bromide.

Simulation	AL (m)	AT (m)	V (m/yr)	Retardation Factor
Case 9 = 1.0 kg, all concentrations, conservative				
9.1	0.429	0.069	23.243	na
9.2	0.276	0.094	22.377	na
9.3	0.634	0.052	18.063	na
9.4	0.464	0.090	24.959	na
9.5	0.661	0.036	22.777	na
Ensemble mean*	0.493	0.068	22.284	na
Case 10 = 1.0 kg, all concentrations, reactive				
10.1	0.402	0.066	13.910	1.671
10.2	0.300	0.076	13.640	1.641
10.3	0.529	0.048	10.808	1.671
10.4	0.474	0.077	14.781	1.689
10.5	0.607	0.032	13.598	1.675
Ensemble mean*	0.462	0.060	13.347	1.669
Case 11 = 1.0 kg, concentrations > 0.1 ppm, conservative				
11.1	0.448	0.074	23.096	na
11.2	0.298	0.094	22.256	na
11.3	0.641	0.057	17.931	na
11.4	0.482	0.094	24.776	na
11.5	0.670	0.039	22.697	na
Ensemble mean*	0.508	0.072	22.151	na
Case 12 = 1.0 kg, concentrations > 0.1 ppm, reactive				
12.1	0.387	0.068	13.949	1.656
12.2	0.294	0.075	13.641	1.632
12.3	0.536	0.052	10.772	1.665
12.4	0.461	0.078	14.790	1.675
12.5	0.615	0.039	13.643	1.664
Ensemble mean*	0.459	0.062	13.359	1.658

* denotes estimate

'na' denotes not applicable

heterogeneous flow field will be discussed in the Strongly Heterogeneous Flow Field section.

The graphs of longitudinal and horizontal transverse variances as a function of center of mass (x-coordinate) for realization 2, using all concentrations in the calculations, are shown in Figures 4 and 5, respectively. Figures 6 and 7 are the same graphs with only concentrations greater than detection limit (0.1 ppm) used in the calculations. The conservative and reactive curves in Figures 4-7 are similar. In all cases, the conservative variance was slightly greater than the reactive variance. This indicates the spreading of the conservative plumes was slightly greater than that of the reactive plume.

Apparent dispersivities for all simulations were calculated using Eq. (13) and the slope of the best-fit line through the points in the graph of variance as a function of center of mass (x-coordinate). It is evident from these graphs that the conservative simulation plumes traveled a greater distance than the reactive plumes. The scale effect of dispersivity suggests that as a plume travels longer distances, more heterogeneities are encountered, and the macrodispersivity of the formation increases. The main goal of this study was to quantify the effects of the Freundlich isotherm in Eq. (1) relative to a conservative tracer. Thus, the best fit line used to calculate the apparent dispersivities only included data points common to both the reactive and conservative simulations. It was found in all simulations that the center of mass of the reactive plume after 2 years had traveled the same distance as the conservative plume after it had traveled from 1 to 1.25 years. Linear interpolation was performed on the conservative data to provide an overlap with the reactive data. Data points from 0 and 0.0114 years were not used in the dispersivity calculations because the grid-size test, described earlier, revealed a significant amount of numerical dispersion at these times (compare Appendices A and F).

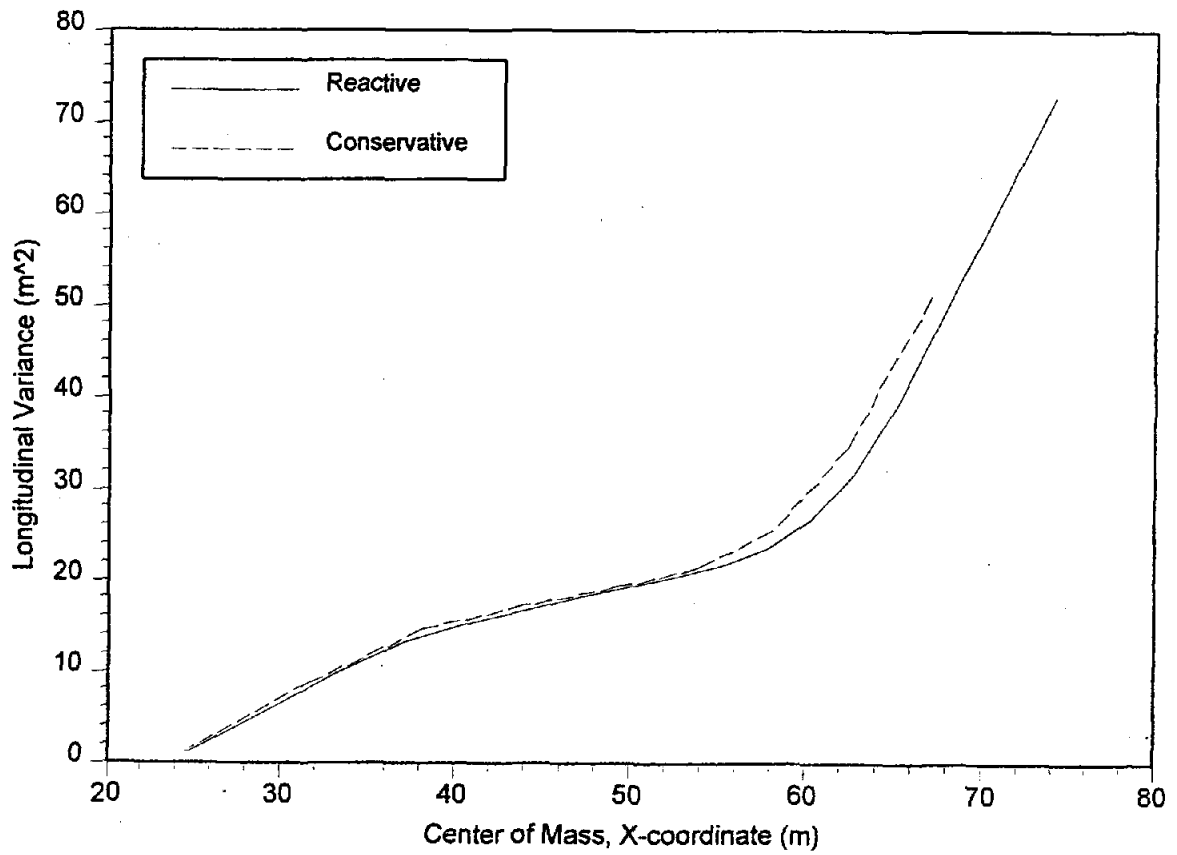


Figure 4. Reactive and conservative curves for realization 2 in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.

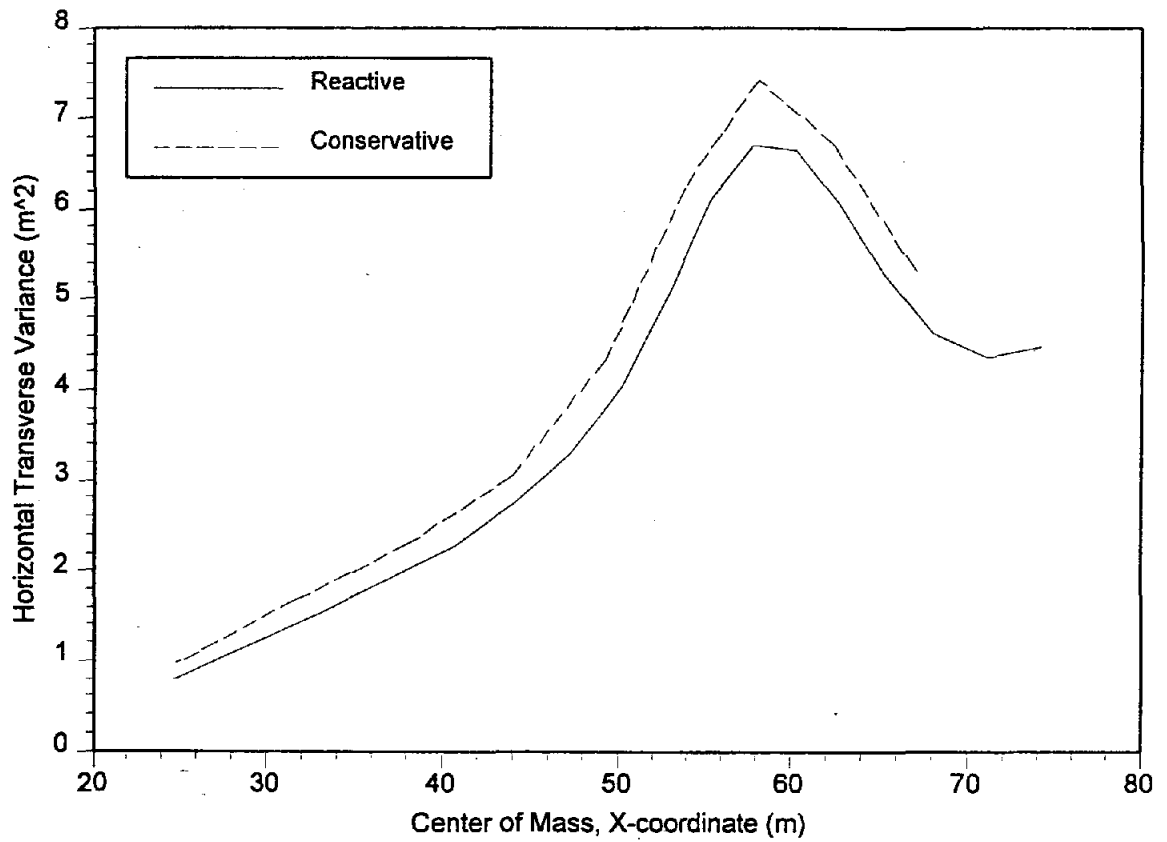


Figure 5. Reactive and conservative curves for realization 2 in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.

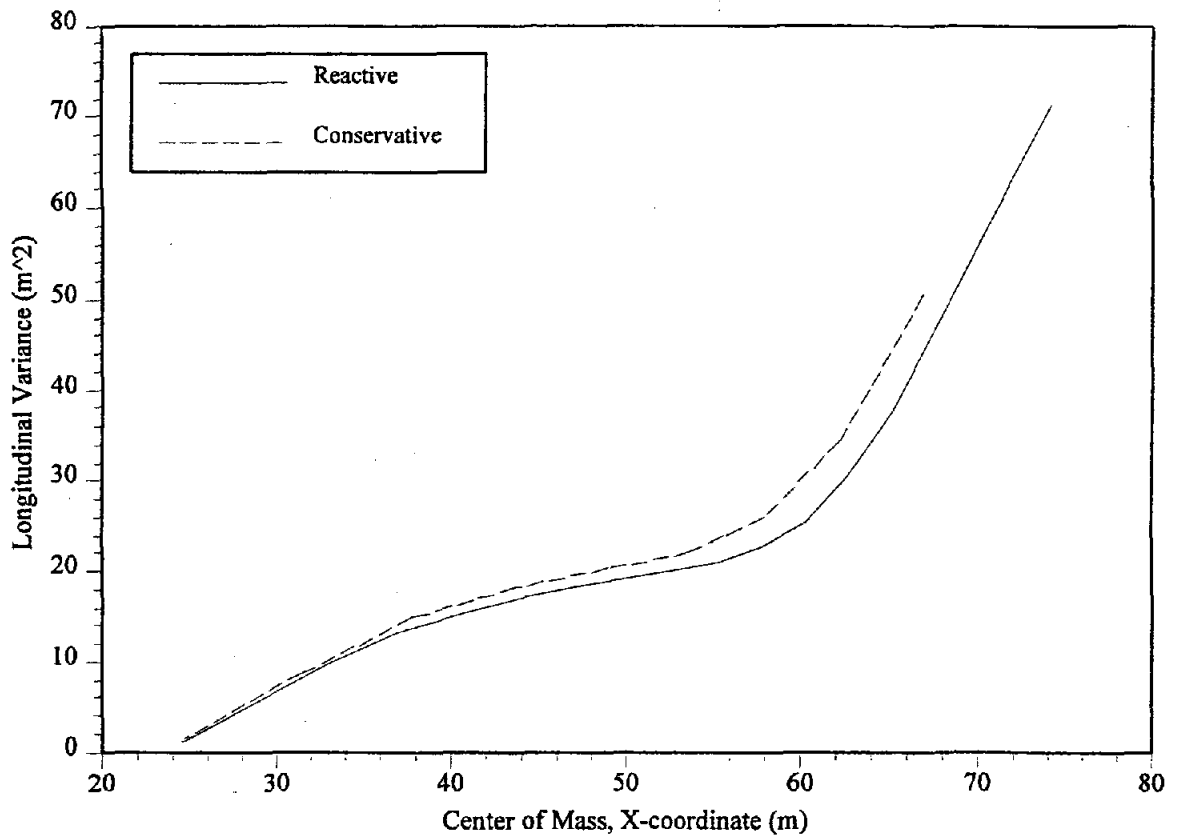


Figure 6. Reactive and conservative curves for realization 2 in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.

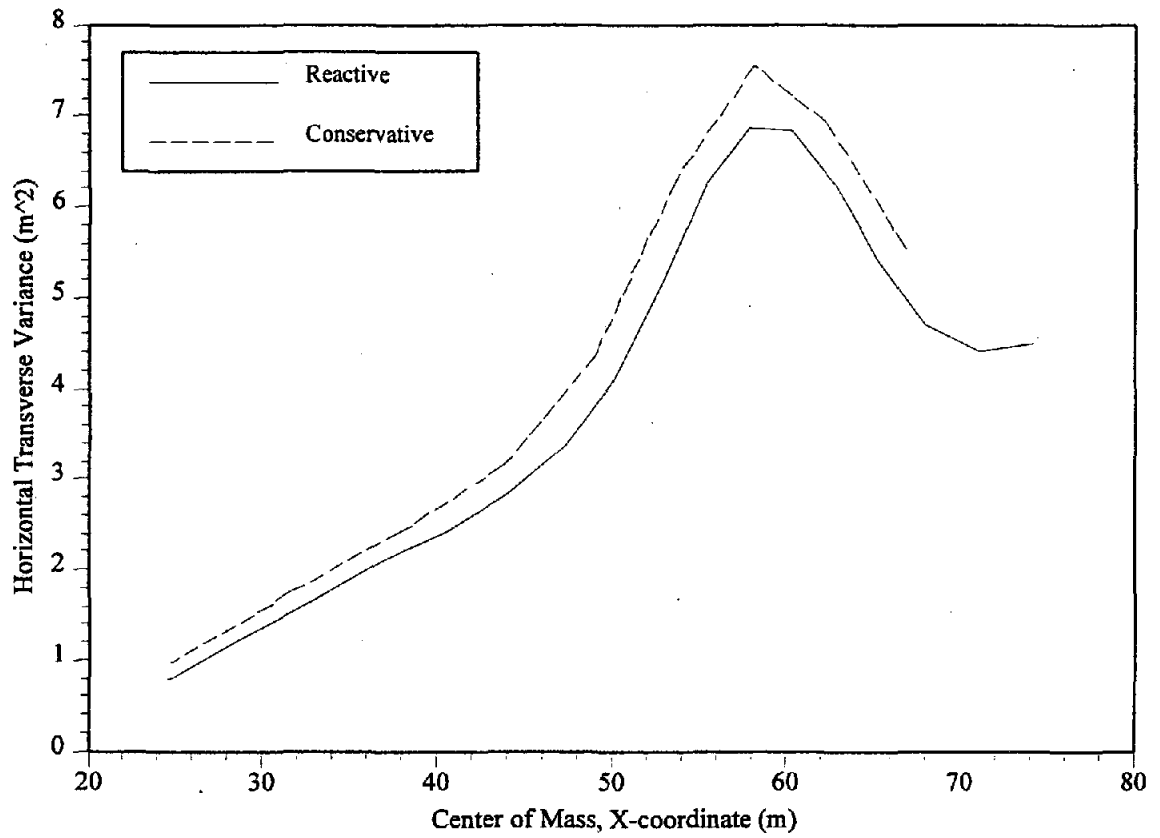


Figure 7. Reactive and conservative curves for realization 2 in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.

Apparent dispersivities for simulations in a weakly heterogeneous flow field with 1 kg of bromide are shown in Table 4. The apparent longitudinal dispersivity values for conservative simulations using all concentrations ranged from 0.276 m to 0.661 m. The same calculations performed on concentrations greater than 0.1 ppm ranged from 0.298 m to 0.670 m.

The ensemble mean estimates were calculated by averaging the dispersivity values for the five realizations in each case. Null hypothesis testing was employed to test if the ensemble mean estimates were significantly different. The null hypothesis in all cases states the two ensemble means are equal: the mean of the population from which the first sample was drawn is the same as the mean of the parent population of the second sample. A significance level of 5 percent was used in all statistical tests. This indicates there is a 5 percent chance of rejecting a true hypothesis (type I error). Results of increasing the significance level are discussed at the end of the Results and Discussion section.

The null hypothesis tests were set up in such a way as to isolate the affects of the parameter in question. For example, when testing the affects of the Freundlich isotherm, ensemble means with identical simulations and calculations for the reactive and conservative cases were compared. The ensemble mean estimate in a weakly heterogeneous flow field with a conservative tracer and 1 kg of bromide using all concentrations was tested with the ensemble mean estimate in a weakly heterogeneous flow field with a reactive tracer and 1 kg of bromide using all concentrations.

The ensemble mean estimates of apparent longitudinal dispersivities for the conservative simulations using all concentrations and those greater than 0.1 ppm were 0.493 m and 0.508 m, respectively. The null hypothesis was not rejected (see Table 5). The ensemble mean estimate of apparent longitudinal dispersivities for the reactive

Table 5. Statistics for hypothesis testing of ensemble means using all concentrations and omitting concentrations below 0.1 ppm. The significance level was 5 percent.

Cases tested	Test F	Table F	Null Hyp.	Test t	Table t	Null Hyp.
1,3 AL*	1.099	6.39	not rej.	-0.031	+/- 2.306	not rej.
2,4 AL	1.048	6.39	not rej.	0.103	+/- 2.306	not rej.
5,7 AL	1.180	6.39	not rej.	-0.163	+/- 2.306	not rej.
6,8 AL	1.398	6.39	not rej.	0.309	+/- 2.306	not rej.
9,11 AL	1.085	6.39	not rej.	0.153	+/- 2.306	not rej.
10,12 AL	1.129	6.39	not rej.	-0.049	+/- 2.306	not rej.
13,15 AL	1.093	6.39	not rej.	-0.007	+/- 2.306	not rej.
14,16 AL	2.175	6.39	not rej.	-0.782	+/- 2.306	not rej.
1,3 AT**	1.014	6.39	not rej.	0.043	+/- 2.306	not rej.
2,4 AT	1.018	6.39	not rej.	0.037	+/- 2.306	not rej.
5,7 AT	1.054	6.39	not rej.	-0.055	+/- 2.306	not rej.
6,8 AT	1.203	6.39	not rej.	-0.130	+/- 2.306	not rej.
9,11 AT	1.068	6.39	not rej.	0.221	+/- 2.306	not rej.
10,12 AT	1.385	6.39	not rej.	0.228	+/- 2.306	not rej.
13,15 AT	1.449	6.39	not rej.	-0.225	+/- 2.306	not rej.
14,16 AT	4.104	6.39	not rej.	-0.337	+/- 2.306	not rej.

Key: Case 1 = 1.0 kg, strongly heterogeneous, all concentrations, conservative

Case 2 = 1.0 kg, strongly heterogeneous, all concentrations, reactive

Case 3 = 1.0 kg, strongly heterogeneous, concentrations > 0.1 ppm, conservative

Case 4 = 1.0 kg, strongly heterogeneous, concentrations > 0.1 ppm, reactive

Case 5 = 0.1 kg, strongly heterogeneous, all concentrations, conservative

Case 6 = 0.1 kg, strongly heterogeneous, all concentrations, reactive

Case 7 = 0.1 kg, strongly heterogeneous, concentrations > 0.1 ppm, conservative

Case 8 = 0.1 kg, strongly heterogeneous, concentrations > 0.1 ppm, reactive

Case 9 = 1.0 kg, weakly heterogeneous, all concentrations, conservative

Case 10 = 1.0 kg, weakly heterogeneous, all concentrations, reactive

Case 11 = 1.0 kg, weakly heterogeneous, concentrations > 0.1 ppm, conservative

Case 12 = 1.0 kg, weakly heterogeneous, concentrations > 0.1 ppm, reactive

Case 13 = 0.1 kg, weakly heterogeneous, all concentrations, conservative

Case 14 = 0.1 kg, weakly heterogeneous, all concentrations, reactive

Case 15 = 0.1 kg, weakly heterogeneous, concentrations > 0.1 ppm, conservative

Case 16 = 0.1 kg, weakly heterogeneous, concentrations > 0.1 ppm, reactive

* denotes apparent longitudinal dispersivity

** denotes apparent horizontal transverse dispersivity

simulations using all concentrations and those greater than 0.1 ppm were 0.462 m and 0.459 m, respectively. Again, the null hypothesis was not rejected (see Table 5).

The null hypothesis was also not rejected for the horizontal transverse dispersivities in the reactive and conservative cases. Therefore, with the assumptions and limited number of realizations used in this study, omitting concentrations below 0.1 ppm did not have a significant effect on apparent dispersivity values in a weakly heterogeneous flow field.

Hypothesis testing was also performed on the reactive and conservative runs. None of the null hypotheses was rejected (see Table 6). It was concluded, with the assumptions and limited number of realizations used in this study, that the Freundlich isotherm in Eq. (1) did not have a significant effect on apparent dispersivity values in a weakly heterogeneous flow field when compared to a conservative plume.

The reactive simulation for realization 2, as discussed above, was simulated for an additional 2 years. All data from the 2 year conservative simulation were used in the apparent dispersivity calculations. The apparent dispersivities in the longitudinal and horizontal transverse direction for the conservative simulation using concentrations greater than 0.1 ppm were 0.487 m and 0.078 m, respectively. The same calculations for the reactive simulation were 0.397 m and 0.073 m, respectively. Thus, the similarities in apparent longitudinal and horizontal transverse dispersivities between the conservative and reactive simulations were also observed for a longer plume travel distance in a weakly heterogeneous flow field.

Skewness and Kurtosis. The significance of skewness and kurtosis is that they are measures of how close a concentration distribution is to a Gaussian distribution. The skewness and kurtosis values through time for each simulation are listed in Appendix A.

Table 6. Statistics for hypothesis testing of reactive and conservative ensemble means. The significance level was 5 percent.

Cases tested	Test F	Table F	Null Hyp.	Test t	Table t	Null Hyp.
1,2 AL*	1.068	6.39	not rej.	-1.673	+/- 2.306	not rej.
3,4 AL	1.231	6.39	not rej.	-1.553	+/- 2.306	not rej.
5,6 AL	1.199	6.39	not rej.	-1.034	+/- 2.306	not rej.
7,8 AL	1.011	6.39	not rej.	-0.472	+/- 2.306	not rej.
9,10 AL	1.804	6.39	not rej.	-0.345	+/- 2.306	not rej.
11,12 AL	1.473	6.39	not rej.	-0.559	+/- 2.306	not rej.
13,14 AL	3.928	6.39	not rej.	0.397	+/- 2.306	not rej.
15,16 AL	1.653	6.39	not rej.	-0.205	+/- 2.306	not rej.
1,2 AT**	1.156	6.39	not rej.	-0.079	+/- 2.306	not rej.
3,4 AT	1.161	6.39	not rej.	-0.082	+/- 2.306	not rej.
5,6 AT	1.261	6.39	not rej.	-0.306	+/- 2.306	not rej.
7,8 AT	1.438	6.39	not rej.	-0.386	+/- 2.306	not rej.
9,10 AT	1.618	6.39	not rej.	-0.598	+/- 2.306	not rej.
11,12 AT	2.098	6.39	not rej.	-0.708	+/- 2.306	not rej.
13,14 AT	1.256	6.39	not rej.	-0.859	+/- 2.306	not rej.
15,16 AT	3.557	6.39	not rej.	-1.269	+/- 2.306	not rej.

Key: Case 1 = 1.0 kg, strongly heterogeneous, all concentrations, conservative
 Case 2 = 1.0 kg, strongly heterogeneous, all concentrations, reactive
 Case 3 = 1.0 kg, strongly heterogeneous, concentrations > 0.1 ppm, conservative
 Case 4 = 1.0 kg, strongly heterogeneous, concentrations > 0.1 ppm, reactive
 Case 5 = 0.1 kg, strongly heterogeneous, all concentrations, conservative
 Case 6 = 0.1 kg, strongly heterogeneous, all concentrations, reactive
 Case 7 = 0.1 kg, strongly heterogeneous, concentrations > 0.1 ppm, conservative
 Case 8 = 0.1 kg, strongly heterogeneous, concentrations > 0.1 ppm, reactive
 Case 9 = 1.0 kg, weakly heterogeneous, all concentrations, conservative
 Case 10 = 1.0 kg, weakly heterogeneous, all concentrations, reactive
 Case 11 = 1.0 kg, weakly heterogeneous, concentrations > 0.1 ppm, conservative
 Case 12 = 1.0 kg, weakly heterogeneous, concentrations > 0.1 ppm, reactive
 Case 13 = 0.1 kg, weakly heterogeneous, all concentrations, conservative
 Case 14 = 0.1 kg, weakly heterogeneous, all concentrations, reactive
 Case 15 = 0.1 kg, weakly heterogeneous, concentrations > 0.1 ppm, conservative
 Case 16 = 0.1 kg, weakly heterogeneous, concentrations > 0.1 ppm, reactive

* denotes apparent longitudinal dispersivity

** denotes apparent horizontal transverse dispersivity

The skewness and kurtosis values vary, but are relatively close to that of a normal distribution. This indicates that the apparent dispersivities are probably close to true dispersivities.

When using all concentrations in the calculations, the last three kurtosis values in the y-direction are negative for the reactive simulation of realization 5. As discussed earlier, kurtosis values are always positive because every term in Eq. (15) is positive or is raised to an even power. SUTRA and WATSUTRA provide non-zero concentration values for all nodes in the model domain. The majority of the values outside the plume are given extremely small negative numbers. Clearly, negative concentration values are not possible. These numbers are important because the bromide plume only occupies a small percentage of nodes in the model domain. The method of moments FORTRAN program uses all concentration values in the model domain (see Appendix E). When higher-order moments are calculated, such as third or fourth order, the solute concentration at a given node is multiplied by the third or fourth power of the quantity $(x-x_c)$ or $(y-y_c)$ (see Eq. 10). In addition, the concentration is multiplied by the density of water (998.23 kg/m^3) to convert mass fraction (kg/kg) to mass since each element has a volume of 1 m^3 (the moment programs take porosity into account). Clearly, if the quantity $(x-x_c)$ or $(y-y_c)$ is large, the fourth power of the quantity may produce a significant number when multiplied by the small negative concentration. Therefore the negative values for kurtosis are likely a result of the small negative concentrations. This interpretation is reinforced by the absence of negative kurtosis values when concentrations below 0.1 ppm are omitted.

It is not clear why SUTRA and WATSUTRA have small negative concentrations at many of the nodes when this model is simulated. It may be a result of numerical dispersion.

As a result of the negative concentration numbers at many of the nodes in the model domain, it is probably best to examine the moment calculations when the values below detection limit are omitted. One must exercise caution when interpreting the differences between moment values with and without omitting concentrations below detection limit. The presence of small negative values may only be a phenomenon observed in SUTRA and WATSUTRA or in similar finite-element programs. Thus, the findings may not apply to all solute transport modeling programs.

Strongly Heterogeneous Flow Field

Ten simulations were performed in the strongly heterogeneous flow field with 1 kg of bromide, five conservative and five reactive. SURFER contour plots of the plumes are given in Appendix B. SURFER contour plots of the strongly heterogeneous natural log hydraulic conductivity fields are given in Appendix C. The difference in plume spreading between the weakly and strongly heterogeneous flow fields is striking. The strong heterogeneities caused enhanced spreading in the longitudinal and horizontal transverse directions.

Spatial Moment Estimates of Plume Behavior

Mass of Bromide. Moment calculations, zeroth through fourth, are given in Appendix A. The same basic trends of tracer mass for the weakly heterogeneous flow field are observed in the strongly heterogeneous flow field. Again, about 42% of the tracer is adsorbed onto the sediments in the reactive simulations at 2 years.

Center of Mass. Graphs of center of mass as a function of time are shown in Appendix D. The average linear velocities show more variability than those for the weakly heterogeneous flow field; the velocities varied by as much as a factor of 2 (see Table 7). A greater range of velocities is expected for a greater variance in the hydraulic conductivity field. The retardation factor averaged approximately 1.7.

Table 7. Dispersivities, velocities, and retardation factors for simulations in a strongly heterogeneous flow field with 1 kg of bromide.

Simulation	AL (m)	AT (m)	V (m/yr)	Retardation Factor
Case 1 = 1.0 kg, all concentrations, conservative				
1.1	1.919	0.701	14.198	na
1.2	1.230	0.477	10.181	na
1.3	1.188	0.168	17.538	na
1.4	1.430	0.118	22.266	na
1.5	1.555	0.301	19.889	na
Ensemble mean*	1.464	0.353	16.814	na
Case 2 = 1.0 kg, all concentrations, reactive				
2.1	1.447	0.722	8.140	1.744
2.2	0.682	0.468	5.966	1.706
2.3	1.113	0.143	10.400	1.686
2.4	1.099	0.098	13.159	1.692
2.5	1.395	0.272	11.705	1.699
Ensemble mean*	1.147	0.341	9.874	1.705
Case 3 = 1.0 kg, concentrations > 0.1 ppm, conservative				
3.1	1.850	0.708	14.121	na
3.2	1.151	0.484	10.086	na
3.3	1.222	0.175	17.332	na
3.4	1.520	0.117	21.780	na
3.5	1.551	0.314	19.761	na
Ensemble mean*	1.459	0.360	16.616	na
Case 4 = 1.0 kg, concentrations > 0.1 ppm, reactive				
4.1	1.418	0.733	8.134	1.736
4.2	0.653	0.472	5.963	1.691
4.3	1.137	0.146	10.314	1.680
4.4	1.220	0.103	12.940	1.683
4.5	1.408	0.279	11.659	1.695
Ensemble mean*	1.167	0.347	9.802	1.697

* denotes estimate

'na' denotes not applicable

Plume Variance. Graphs of plume variance in the longitudinal and horizontal transverse directions as a function of center of mass (x-coordinate) are given in Appendix D. Note the scales for the reactive and conservative simulations are the same. The curves for the reactive simulations are similar to the conservative simulation curves. In most cases, the conservative simulations exhibited slightly enhanced plume variance.

The reactive simulation of realization 4 was run for an addition two years to see if the plume variance similarities hold for longer travel distances. The longitudinal and horizontal transverse variance as a function of center of mass using all concentrations are shown in Figures 8 and 9, respectively. The same quantities are graphed in Figures 10 and 11, respectively, using concentrations greater than 0.1 ppm in the calculations. These graphs indicate the reactive and conservative curves are similar. The conservative plume variances were slightly greater than the reactive variances.

Apparent dispersivities were calculated after performing a linear interpolation as previously described. The apparent dispersivities for the 1 kg simulations in a strongly heterogeneous flow field are given in Table 7. The values are approximately 2 to 3 times larger than those in the weakly heterogeneous flow field. This is the result of greater heterogeneity in the flow fields.

The ensemble mean estimates of apparent longitudinal dispersivities for the conservative simulations using all concentrations and only those greater than 0.1 ppm were 1.464 m and 1.459 m, respectively. Null hypothesis testing was employed to test if they were significantly different. The null hypothesis was not rejected (see Table 5). The same hypothesis test was performed on the ensemble mean estimates of the apparent horizontal transverse dispersivities. Again, the null hypothesis was not rejected.

The ensemble mean estimates of apparent longitudinal dispersivities for the reactive simulations using all concentrations and only those greater than 0.1 ppm were 1.147 m and 1.167 m, respectively. The null hypothesis was not rejected. The same

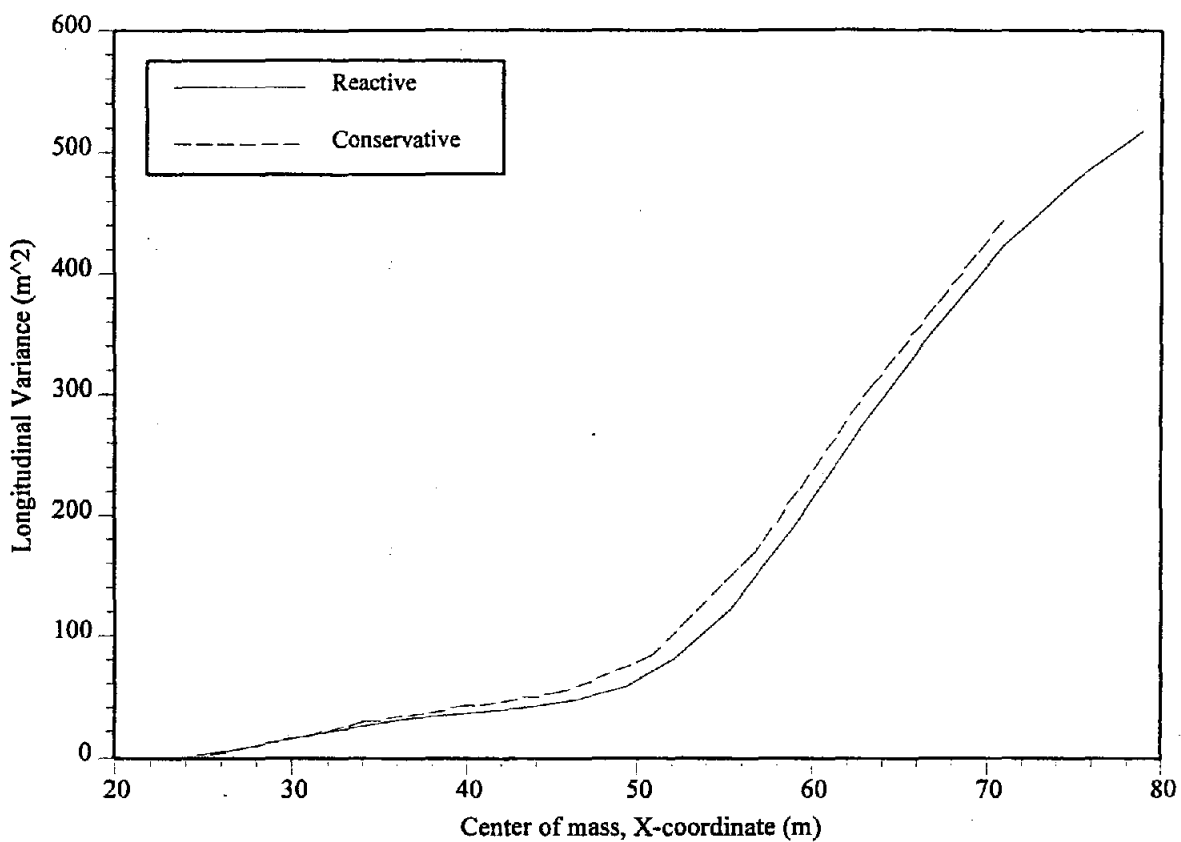


Figure 8. Reactive and conservative curves for realization 4 in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations were used in calculations.

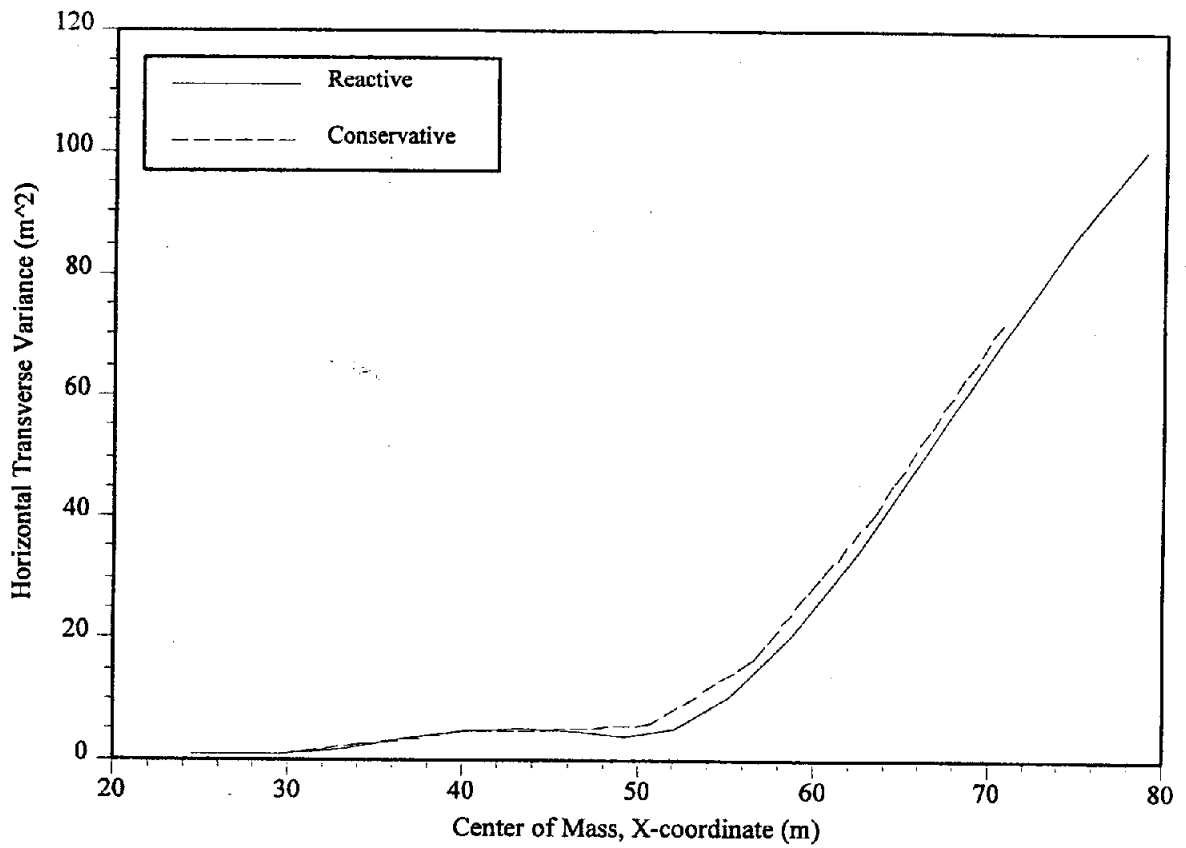


Figure 9. Reactive and conservative curves for realization 4 in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations were used in calculations.

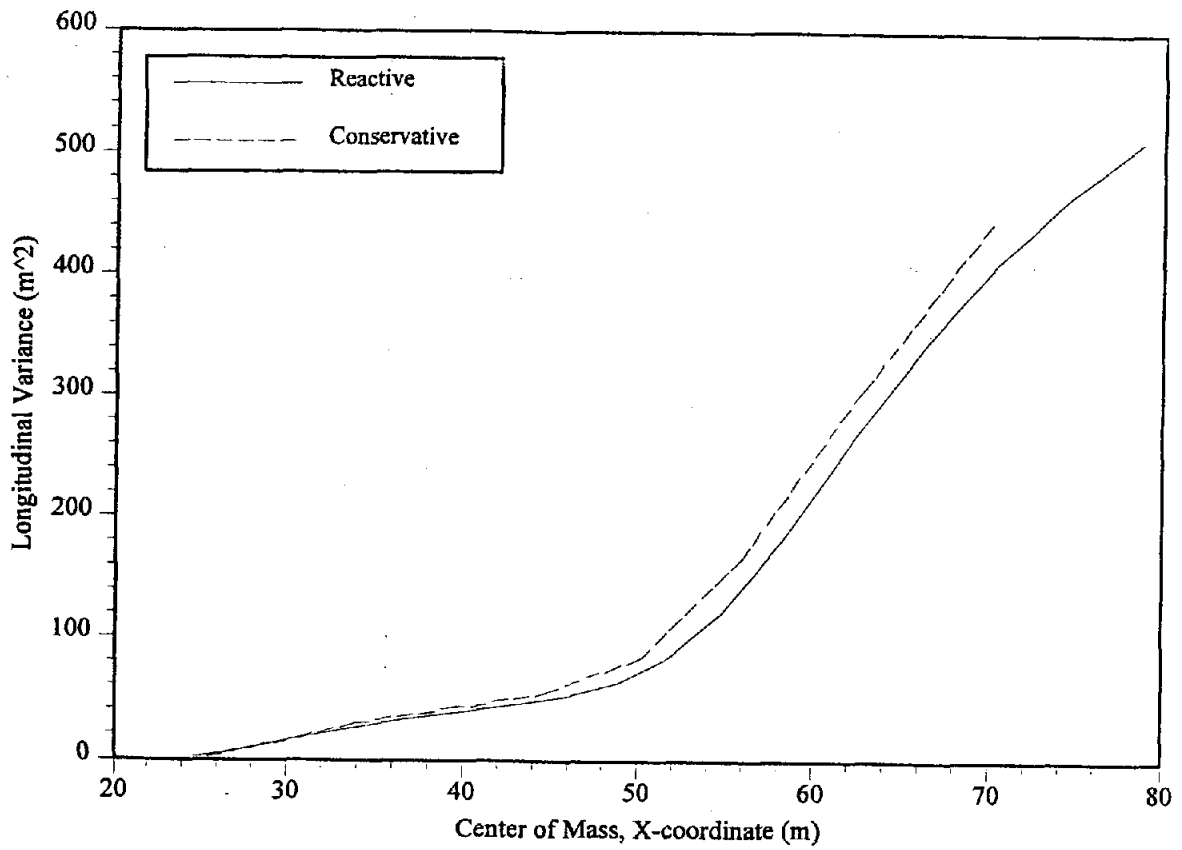


Figure 10. Reactive and conservative curves for realization 4 in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.

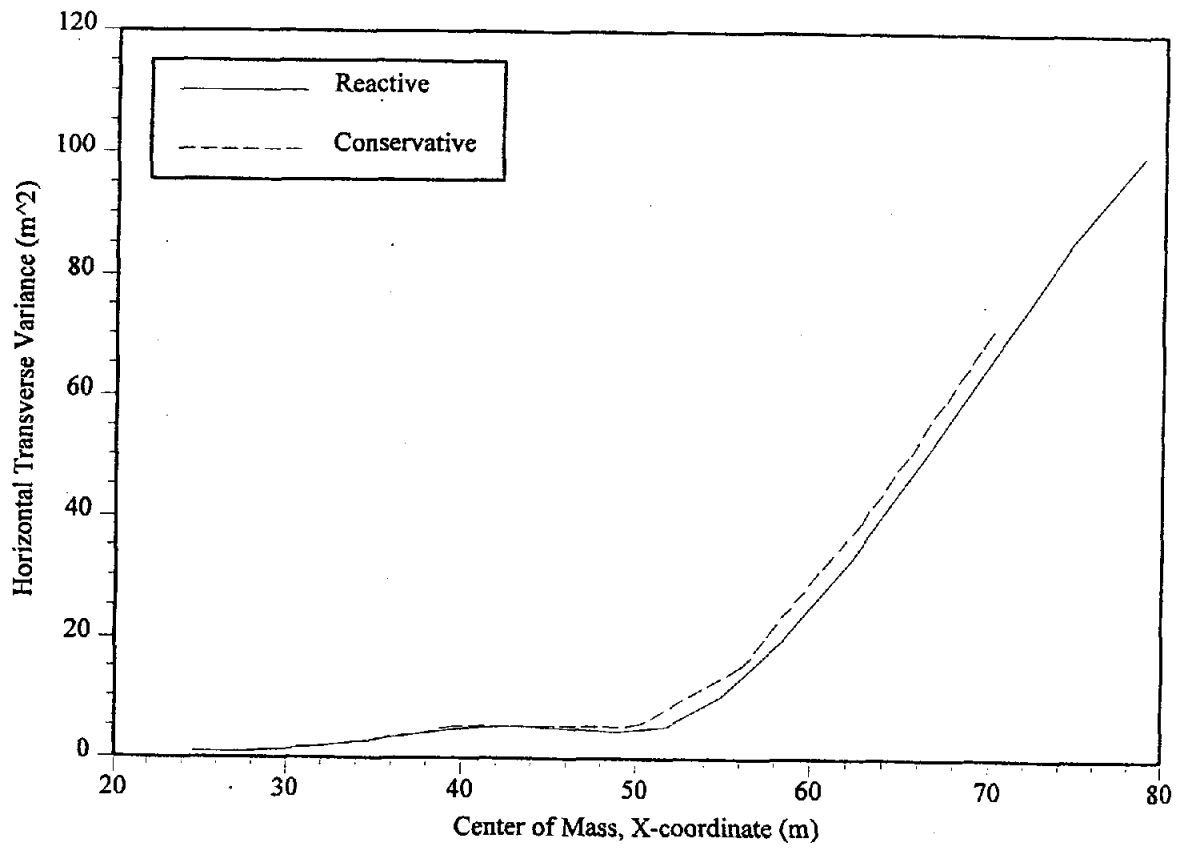


Figure 11. Reactive and conservative curves for realization 4 in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.

hypothesis test was performed on the ensemble mean estimates of the horizontal transverse dispersivities. It was not rejected. This indicates that, with the assumptions and limited number of realizations used in this study, there is no significant difference between the apparent dispersivity ensemble mean estimates using all concentrations and only those greater than 0.1 ppm in a strongly heterogeneous flow field.

The ensemble mean estimates for the reactive simulations were compared to those with the same parameters for the conservative simulations. Again, none of the null hypotheses was rejected. Thus, with the assumptions and limited number of realizations used in this study, there was no significant difference between the apparent dispersivity ensemble mean estimates of the conservative and reactive simulations in a strongly heterogeneous flow field.

As stated above, the reactive simulation of realization 4 was run for an additional two years to see if the similarities between the reactive and conservative cases hold for longer travel distances. All data from the 2 year conservative simulation were used in the apparent dispersivity calculations. The apparent longitudinal and horizontal transverse dispersivities for the conservative simulation using concentrations greater than 0.1 ppm were 5.018 m and 0.744 m, respectively. The same calculations for the reactive simulation yielded 4.274 m and 0.622 m, respectively. Thus, the similarities between the reactive and conservative apparent longitudinal and horizontal transverse dispersivities were observed for a longer plume travel distance in a strongly heterogeneous flow field.

Skewness and Kurtosis. The values of skewness and kurtosis through time for the strongly heterogeneous flow field are given in Appendix A. In contrast to the weakly heterogeneous flow field, the skewness and kurtosis values in many simulations indicate concentration distributions much different than a Gaussian distribution.

Simulations with 0.1 kg of Bromide

The purpose of performing simulations with 0.1 kg of bromide was to examine the effects of tracer mass on apparent dispersivities. The only difference between the simulations with 0.1 kg of bromide and those with 1 kg was the injection time. The injection time for the 0.1 kg simulations was 1/10 that of the 1 kg simulations; the injection rate and bromide concentration in the injected water were unchanged. The simulations in the weakly and strongly heterogeneous flow fields will be discussed together because most of the same trends are observed in both flow fields.

SURFER contour plots of plumes using 0.1 kg of bromide are shown in Appendix B. For comparison, the contour intervals in the 0.1 kg plots were exactly one order of magnitude smaller than the 1 kg plots. The plumes with 0.1 kg of bromide were similar to the 1 kg plumes. In most cases the 0.1 kg plumes were slightly smaller than the 1 kg plumes, but the same general characteristics are present.

Spatial Moment Estimates of Plume Behavior

Mass of Bromide. The same basic trends of tracer mass in the 1 kg simulations were observed in the 0.1 kg simulations. The main difference between them is that the reactive 0.1 kg simulations experience more mass loss due to adsorption. Approximately 48 to 50 percent of the total tracer mass was lost due to adsorption at 2 years; the reactive 1 kg simulations lost about 42 percent to adsorption.

Center of Mass. Graphs of center of mass as a function of time are given in Appendix D. Average linear velocities for the weakly and strongly heterogeneous flow fields are given in Tables 8 and 9, respectively. In all cases, with the exception of the strongly heterogeneous conservative cases, the ensemble mean velocities are slightly smaller in the 0.1 kg simulations than the equivalent 1 kg simulations. The retardation factor averaged approximately 1.85 in the weakly and strongly heterogeneous flow fields.

Table 8. Dispersivities, velocities, and retardation factors for simulations in a weakly heterogeneous flow field with 0.1 kg of bromide.

Simulation	AL (m)	AT (m)	V (m/yr)	Retardation Factor
Case 13 = 0.1 kg, all concentrations, conservative				
13.1	0.379	0.073	23.782	na
13.2	0.227	0.079	23.393	na
13.3	0.596	0.055	18.240	na
13.4	0.494	0.086	25.389	na
13.5	0.737	0.033	22.657	na
Ensemble mean*	0.487	0.065	22.692	na
Case 14 = 0.1 kg, all concentrations, reactive				
14.1	0.470	0.064	12.659	1.879
14.2	0.381	0.061	12.655	1.848
14.3	0.585	0.042	9.592	1.902
14.4	0.567	0.076	13.208	1.922
14.5	0.625	0.028	11.928	1.899
Ensemble mean*	0.526	0.054	12.008	1.890
Case 15 = 0.1 kg, concentrations > 0.1 ppm, conservative				
15.1	0.350	0.073	23.536	na
15.2	0.263	0.073	23.000	na
15.3	0.633	0.054	17.810	na
15.4	0.472	0.077	25.040	na
15.5	0.711	0.035	22.453	na
Ensemble mean*	0.486	0.062	22.368	na
Case 16 = 0.1 kg, concentrations > 0.1 ppm, reactive				
16.1	0.354	0.059	12.839	1.833
16.2	0.286	0.053	12.772	1.801
16.3	0.588	0.045	9.663	1.843
16.4	0.467	0.060	13.361	1.874
16.5	0.625	0.038	12.130	1.851
Ensemble mean*	0.464	0.051	12.153	1.840

* denotes estimate

'na' denotes not applicable

Table 9. Dispersivities, velocities, and retardation factors for simulations in a strongly heterogeneous flow field with 0.1 kg of bromide.

Simulation	AL (m)	AT (m)	V (m/yr)	Retardation Factor
Case 5 = 0.1 kg, all concentrations, conservative				
5.1	1.397	0.555	13.644	na
5.2	0.562	0.480	10.075	na
5.3	1.100	0.150	18.034	na
5.4	1.069	0.127	22.686	na
5.5	1.608	0.267	18.781	na
Ensemble mean*	1.147	0.316	16.644	na
Case 6 = 0.1 kg, all concentrations, reactive				
6.1	0.904	0.517	7.260	1.879
6.2	0.283	0.402	5.790	1.740
6.3	1.168	0.141	9.302	1.939
6.4	0.999	0.123	11.974	1.895
6.5	1.144	0.219	9.722	1.932
Ensemble mean*	0.900	0.280	8.810	1.877
Case 7 = 0.1 kg, concentrations > 0.1 ppm, conservative				
7.1	1.258	0.532	13.373	na
7.2	0.388	0.480	9.849	na
7.3	1.160	0.144	17.473	na
7.4	1.173	0.125	21.770	na
7.5	1.544	0.265	18.498	na
Ensemble mean*	1.105	0.309	16.193	na
Case 8 = 0.1 kg, concentrations > 0.1 ppm, reactive				
8.1	1.115	0.448	7.464	1.792
8.2	0.241	0.419	5.526	1.782
8.3	1.119	0.121	9.192	1.901
8.4	1.054	0.129	11.684	1.863
8.5	1.355	0.217	9.856	1.877
Ensemble mean*	0.977	0.267	8.744	1.843

* denotes estimate

'na' denotes not applicable

Plume Variance. The graphs of plume variances versus center of mass for the 0.1 kg simulations are shown in Appendix D. The variances in the 0.1 kg simulations follow the same trends as the 1 kg simulations, but are slightly smaller.

Apparent dispersivities for the 0.1 kg simulations in the weakly and strongly heterogeneous flow fields are given in Tables 8 and 9, respectively. Note the total mass of tracer does not affect the apparent dispersivity calculations since the second moment is normalized by the total mass of tracer (see Appendix E). All ensemble mean estimates in the strongly heterogeneous flow field were smaller than the 1 kg simulations. The ensemble mean estimates for the 0.1 kg simulations in the weakly heterogeneous flow field were mixed; most were smaller than the equivalent 1 kg ensemble means, but a few were greater. The reason for this finding is unclear, but may be partly due to small numerical errors in SUTRA and WATSUTRA.

The ensemble mean estimates of the longitudinal and horizontal transverse dispersivities for the 0.1 kg simulations were tested against the same simulations with 1 kg of bromide. The results of the null hypothesis testing are shown in Table 10. None of the hypotheses was rejected. Therefore, with the assumptions and limited number of realizations used in this study, there is no evidence to suggest tracer mass has any significant effect on apparent dispersivities in weakly and strongly heterogeneous flow fields.

Skewness and Kurtosis. The skewness and kurtosis values for the 0.1 kg simulations differed from the 1 kg simulations, especially kurtosis values in the reactive simulations using all concentrations in the calculations. Many of the kurtosis values for the reactive simulations using all concentrations were negative. This result is probably due to the small negative concentrations in the model domain, as previously discussed. This is supported by the observation that negative kurtosis values only occur in the moment calculations using all concentrations.

Table 10. Statistics for hypothesis testing of tracer mass ensemble means.
The significance level was 5 percent.

Cases tested	Test F	Table F	Null Hyp.	Test t	Table t	Null Hyp.
1,5 AL*	1.801	6.39	not rej.	-1.438	+/- 2.306	not rej.
2,6 AL	1.406	6.39	not rej.	-1.172	+/- 2.306	not rej.
3,7 AL	2.334	6.39	not rej.	-1.543	+/- 2.306	not rej.
4,8 AL	1.876	6.39	not rej.	-0.805	+/- 2.306	not rej.
9,13 AL	1.535	6.39	not rej.	-0.055	+/- 2.306	not rej.
10,14 AL	1.419	6.39	not rej.	0.919	+/- 2.306	not rej.
11,15 AL	1.524	6.39	not rej.	-0.204	+/- 2.306	not rej.
12,16 AL	1.358	6.39	not rej.	0.063	+/- 2.306	not rej.
1,5 AT**	1.527	6.39	not rej.	-0.270	+/- 2.306	not rej.
2,6 AT	2.226	6.39	not rej.	-0.435	+/- 2.306	not rej.
3,7 AT	1.633	6.39	not rej.	-0.369	+/- 2.306	not rej.
4,8 AT	2.727	6.39	not rej.	-0.588	+/- 2.306	not rej.
9,13 AT	1.338	6.39	not rej.	-0.205	+/- 2.306	not rej.
10,14 AT	1.039	6.39	not rej.	-0.460	+/- 2.306	not rej.
11,15 AT	1.815	6.39	not rej.	-0.691	+/- 2.306	not rej.
12,16 AT	3.077	6.39	not rej.	-1.342	+/- 2.306	not rej.

Key: Case 1 = 1.0 kg, strongly heterogeneous, all concentrations, conservative

Case 2 = 1.0 kg, strongly heterogeneous, all concentrations, reactive

Case 3 = 1.0 kg, strongly heterogeneous, concentrations > 0.1 ppm, conservative

Case 4 = 1.0 kg, strongly heterogeneous, concentrations > 0.1 ppm, reactive

Case 5 = 0.1 kg, strongly heterogeneous, all concentrations, conservative

Case 6 = 0.1 kg, strongly heterogeneous, all concentrations, reactive

Case 7 = 0.1 kg, strongly heterogeneous, concentrations > 0.1 ppm, conservative

Case 8 = 0.1 kg, strongly heterogeneous, concentrations > 0.1 ppm, reactive

Case 9 = 1.0 kg, weakly heterogeneous, all concentrations, conservative

Case 10 = 1.0 kg, weakly heterogeneous, all concentrations, reactive

Case 11 = 1.0 kg, weakly heterogeneous, concentrations > 0.1 ppm, conservative

Case 12 = 1.0 kg, weakly heterogeneous, concentrations > 0.1 ppm, reactive

Case 13 = 0.1 kg, weakly heterogeneous, all concentrations, conservative

Case 14 = 0.1 kg, weakly heterogeneous, all concentrations, reactive

Case 15 = 0.1 kg, weakly heterogeneous, concentrations > 0.1 ppm, conservative

Case 16 = 0.1 kg, weakly heterogeneous, concentrations > 0.1 ppm, reactive

* denotes apparent longitudinal dispersivity

** denotes apparent horizontal transverse dispersivity

Significance Levels of Null Hypothesis Testing

A 5 percent significance level was used in all null hypothesis testing in this study. This significance level was chosen because it is widely used in statistics. A 5 percent significance level for 8 degrees of freedom corresponds to a table t value of 2.306. A total of 48 null hypothesis were tested in this study (see Tables 5, 6, and 10). None of the hypotheses was rejected.

The significance level was increased to see at what point the hypotheses would be rejected. A 10 percent significance level for 8 degrees of freedom corresponds to a table t value of 1.860. None of the hypotheses was rejected at this level. A 20 percent significance level corresponds to a table t value of 1.397. Only 4 of the 48 hypotheses were rejected at this level.

The acceptance of the null hypotheses in this study is reasonable because the significance level must be increased to 20 percent in order to reject some of the hypotheses (4 out of 48). At a 20 percent significance level there is a 20 percent chance of rejecting a true hypothesis (type I error).

Practical Application of the Results

Bromide is commonly used as a tracer because it is inexpensive, easily detected, and is commonly believed to behave conservatively. In recent years, however, bromide has been found to be non-conservative in anion-sorbing sediments. This study has shown, with several assumptions and a limited number of realizations, that bromide is a suitable groundwater tracer for macrodispersivity experiments in anion-sorbing sediments.

One of the assumptions used in this study is that the adsorption and desorption isotherms are equal. There are apparently no data on desorption rates for bromide. If, for example, desorption follows a more strongly non-linear isotherm than adsorption, the apparent longitudinal dispersivity would be overestimated because of plume "tailing".

Testing the desorption isotherm with column studies would, however, be difficult and costly to perform.

Another assumption used in this study is that the aquifer is chemically homogeneous. There are not enough data available at the present time to model chemical heterogeneity. Adsorption of bromide at the Savannah River Site is primarily a function of the iron oxide content of the sediments (Seaman et al., 1996) and has apparently no correlation with hydraulic conductivity. Chemical heterogeneity has been examined by several researchers. Bosma et al. (1996) found the effects of chemical heterogeneity in strongly non-linear sorbing solutes to be small on plume variance. It has apparently not been explored in weakly sorbing solutes. A more sophisticated model than SUTRA and WATSUTRA would be required to perform such modeling.

Recently, researchers have been using dissolved noble gases as tracers because they are truly conservative (Sanford et al., 1996). Dissolved gas tracers, in contrast to bromide, are cumbersome and expensive to sample and analyze. However, researchers are developing simplified sampling and analyzing techniques for dissolved gas tracers to lower these costs (e.g. Sanford et al., 1996).

The results of this study are encouraging for the use of bromide as a groundwater tracer in anion-sorbing sediments. The desorption isotherm, however, needs to be examined before one can definitively conclude that bromide can be used as a tracer for macrodispersivity experiments in anion-sorbing sediments.

SUMMARY AND CONCLUSIONS

A weakly non-linear, experimentally derived Freundlich sorption isotherm was found to describe bromide adsorption in sediments from the Savannah River Site in South Carolina. The aim of this research was to quantify the effects of this sorption isotherm on apparent dispersivities relative to a conservative tracer. In addition, the effects of an order-of-magnitude difference in tracer mass injected into the simulated aquifer and omitting concentrations below detection level (set at 0.1 ppm) were evaluated.

Monte Carlo simulations were performed using SUTRA and WATSUTRA. The models were developed to simulate natural gradient tracer tests. Two different hydraulic conductivity fields were modeled; a weakly heterogeneous field with physical parameters similar to the Borden aquifer in Ontario, Canada and a strongly heterogeneous field with physical parameters similar to the Columbus aquifer in Mississippi. The variance of the natural log transformed hydraulic conductivity was chosen to differ by an order-of-magnitude. These hydraulic conductivity fields represent two extremes in aquifer conditions.

Forty simulations were performed in this study for eight cases. Five realizations were performed for each case; each realization was simulated for 2 years (with two exceptions). Apparent dispersivities were calculated by the method of moments. Ensemble means were estimated by averaging the five realizations for each case. Null hypothesis testing was performed with a standard t-test to test the significance between the ensemble mean estimates for various cases.

The following simplifying assumptions were used in this research:

- 1) Sorption according to the Freundlich isotherm was assumed to be a reversible equilibrium process. Thus, desorption followed the same isotherm as adsorption, only with decreasing concentrations, and both adsorption and desorption were rapid in comparison to advective flow.
- 2) The Freundlich isotherm was constant throughout the model domain.
- 3) The groundwater model employed was two-dimensional; vertical heterogeneity was not represented.

With the above assumptions and the limited number of realizations used in this study, the following conclusions can be drawn:

- 1) The Freundlich isotherm did not significantly affect apparent dispersivity values; thus, tracers which are adsorbed following a Freundlich isotherm similar to the one used in this study are not unsuitable as groundwater tracers.
- 2) An order-of-magnitude difference in tracer mass did not significantly affect apparent dispersivity values.
- 3) Omitting concentrations below detection limit did not significantly affect apparent dispersivity values.
- 4) In numerical simulations, moment calculations should be performed with concentrations above detection level because using all concentrations can have adverse effects on higher-order moments. This finding may, however, be unique to SUTRA and WATSUTRA or to similar finite-element programs. Omitting low concentrations is reasonable because numerical studies are meant to replicate field studies.

The results of this study are encouraging for the use of bromide as a groundwater tracer in anion-sorbing sediments. The desorption isotherm, however, needs to be examined before one can definitively conclude that bromide can be used as a tracer for macrodispersivity experiments in anion-sorbing sediments.

APPENDICES

APPENDIX A

MOMENT CALCULATIONS

Weakly Heterogeneous
 Realization I
 1 Kilogram of Bromide
 All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.5665	32.0971	1.2214	0.9216	0.5494	0.0923	3.4211	2.8443
0.0114	1.0001	24.8011	32.1255	1.5614	0.8995	0.6812	0.0942	3.4981	2.8221
0.2614	1.0001	31.0896	32.3770	11.6149	1.0121	0.6800	0.1422	3.0487	3.1483
0.5113	1.0001	37.8965	32.3370	18.2535	1.9780	-0.0015	0.0529	3.0266	3.9360
0.7613	1.0001	43.8887	32.0349	20.7139	3.1311	0.4685	-0.5231	3.0548	3.9826
1.0113	1.0001	49.1625	31.5683	25.1115	3.8143	1.4815	-1.1464	3.2990	3.2473
1.2612	1.0001	54.2330	31.1266	34.7925	3.8319	3.5578	-0.7456	3.9337	2.7762
1.5112	1.0001	59.6777	30.9365	55.4922	3.9826	5.1825	1.0769	3.4503	4.6405
1.7612	1.0001	65.9357	31.3664	83.0534	6.2282	3.8587	3.1753	2.7429	5.4858
2.0114	1.0001	72.7905	32.4377	103.0710	9.6304	1.1864	3.0417	2.6234	3.7905

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6574	24.3978	32.0631	0.9055	0.7668	0.3537	0.0658	3.2124	2.8273
0.0114	0.6545	24.5454	32.0823	1.1002	0.7570	0.4525	0.0763	3.3010	2.7868
0.2614	0.6229	28.3377	32.3356	6.8112	0.7072	0.4469	0.1631	2.8130	2.1765
0.5113	0.6059	32.5868	32.3748	12.3618	0.9783	-0.1720	0.1393	2.9529	2.9817
0.7613	0.5971	36.7364	32.3462	16.1052	1.5366	-0.8912	0.0274	3.0662	3.4222
1.0113	0.5916	40.4959	32.2255	18.1348	2.1781	-1.3121	-0.1099	3.2945	3.5129
1.2612	0.5885	43.8592	32.0111	19.9494	2.6992	-1.3642	-0.4867	3.3862	3.3650
1.5112	0.5858	46.9701	31.7508	22.1862	3.1453	-1.3491	-0.9201	3.4109	3.0674
1.7612	0.5836	49.9294	31.4872	24.7929	3.5176	-1.2188	-0.9692	3.5039	2.8885
2.0114	0.5816	52.8136	31.2133	28.5866	3.6094	-0.4644	-0.6897	3.6699	2.7628

Key: XC and YC are the center of mass values in the x and y directions, respectively
 Var X and Var Y are the plume variances in the x and y directions, respectively
 SKX and SKY are the plume skewness values in the x and y directions, respectively
 KRX and KRY are the plume kurtosis values in the x and y directions, respectively

Weakly Heterogeneous
 Realization 2
 1 Kilogram of Bromide
 All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.7014	31.9337	1.2126	0.9598	0.5369	-0.0622	3.3521	2.9688
0.0114	1.0001	25.0255	31.9340	1.5930	0.9910	0.5687	-0.0938	3.2899	3.0097
0.2614	1.0001	31.6779	32.1099	8.6523	1.6780	0.8432	0.2081	3.2255	3.1882
0.5113	1.0001	38.0680	33.1034	14.5517	2.3131	0.3725	0.5461	2.9758	2.6179
0.7613	1.0001	43.9162	33.3759	17.2021	3.0528	0.6745	0.2212	2.9847	2.9339
1.0113	1.0001	49.1518	33.4190	19.1857	4.3149	0.9602	0.6268	2.8192	3.5682
1.2612	1.0001	53.8699	33.9824	21.3472	6.2377	1.5744	1.3574	3.1475	3.6383
1.5112	1.0001	58.2344	34.8836	25.5015	7.4140	2.8823	1.1386	3.8119	2.8907
1.7612	1.0001	62.5090	35.6643	34.4842	6.7100	4.8180	0.5034	4.1806	2.3870
2.0114	1.0001	67.0663	36.0747	50.9757	5.3197	5.7660	0.1897	3.5566	2.3296

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6574	24.4979	31.9508	0.8965	0.7810	0.3844	-0.0349	3.2112	2.9031
0.0114	0.6533	24.7111	31.9542	1.1367	0.7998	0.4270	-0.0514	3.1920	2.9186
0.2614	0.6195	29.0534	31.9319	5.3541	1.1630	0.2460	-0.0624	3.1377	2.7195
0.5113	0.6086	33.0779	32.3388	9.5860	1.5189	0.2530	0.4893	2.9785	2.4602
0.7613	0.6013	37.0259	33.0140	13.1244	1.9143	-0.4320	0.6555	2.9305	2.2171
1.0113	0.5960	40.7136	33.3661	15.0496	2.2624	-0.7923	0.4128	3.2347	2.5197
1.2612	0.5918	44.1096	33.3758	16.6516	2.7389	-0.8016	0.1415	3.2564	2.7556
1.5112	0.5886	47.2537	33.3457	18.0814	3.2866	-1.0085	0.1566	3.2176	2.8771
1.7612	0.5857	50.1717	33.4918	19.2188	4.0409	-1.2471	0.5293	3.3184	3.2158
2.0114	0.5832	52.8938	33.8540	20.3486	5.0747	-1.3560	1.0285	3.4756	3.4548
2.2614	0.5808	55.4588	34.3653	21.6658	6.1187	-1.2877	1.2344	3.6235	3.2224
2.5113	0.5787	57.9102	34.9199	23.4893	6.7200	-0.9338	1.0327	3.7520	2.7808
2.7613	0.5768	60.3105	35.4224	26.4807	6.6519	-0.1801	0.6249	3.8081	2.4493
3.0113	0.5751	62.7461	35.8127	31.5074	6.0633	0.8623	0.2349	3.7098	2.2927
3.2612	0.5736	65.3098	36.0591	39.2814	5.2804	1.8032	-0.0443	3.4284	2.2590
3.5112	0.5720	68.0753	36.1591	49.7830	4.6247	2.1583	-0.2033	3.0147	2.2713
3.7612	0.5704	71.0607	36.1286	61.7203	4.3440	1.6367	-0.3146	2.6055	2.3700
4.0114	0.5688	74.2276	36.0111	72.9324	4.4894	0.2902	-0.3434	2.3329	2.5734

Weakly Heterogeneous
 Realization 3
 1 Kilogram of Bromide
 All Concentrations Used in Calculations

 Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.3185	31.8779	1.0567	1.0856	0.3917	-0.0540	3.1358	3.0304
0.0114	1.0001	24.5242	31.8719	1.2717	1.1360	0.4370	-0.0808	3.2164	3.0719
0.2614	1.0001	28.7287	31.7802	6.5568	1.4412	1.5151	-0.1327	3.5965	2.7972
0.5113	1.0001	33.3194	31.9527	15.1665	1.6861	1.1503	-0.0869	2.6183	4.2749
0.7613	1.0001	38.0788	31.9892	19.8447	2.6462	-0.4115	-0.1776	2.9459	4.0656
1.0113	1.0001	42.4145	31.8365	22.6142	3.1226	0.8669	-0.3466	4.0686	3.2065
1.2612	1.0001	46.6807	31.6709	32.6010	2.8532	4.3840	-0.4499	4.5220	2.6254
1.5112	1.0001	51.5336	31.5743	55.1046	2.3926	6.2521	-0.2585	3.6632	2.4466
1.7612	1.0001	57.3352	31.5730	86.7325	2.2627	5.2197	0.2315	2.7930	3.0361
2.0114	1.0001	63.9836	31.6617	116.7307	2.5747	2.7153	0.6647	2.5312	3.6982

 Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6567	24.2030	31.9029	0.8290	0.8539	0.2451	-0.0359	3.0058	2.9839
0.0114	0.6538	24.3367	31.9011	0.9651	0.8824	0.2885	0.0494	3.0704	3.0190
0.2614	0.6276	26.9964	31.8166	3.5852	1.1707	0.6704	-0.1490	3.2930	2.7167
0.5113	0.6173	29.5784	31.8370	7.2842	1.1214	1.1821	-0.0489	3.1925	2.1380
0.7613	0.6080	32.4083	31.9699	12.4877	1.2147	0.9499	0.1170	2.5982	2.8852
1.0113	0.6007	35.3710	32.0540	16.5561	1.6549	-0.3381	0.1863	2.4154	3.6747
1.2612	0.5943	38.1989	32.0323	18.1604	2.1787	-1.4284	0.0920	2.8529	3.4128
1.5112	0.5906	40.7868	31.9481	18.9043	2.5325	-1.5648	-0.0487	3.3744	2.9556
1.7612	0.5885	43.2230	31.8488	20.7064	2.6443	-0.6655	-0.2007	3.6935	2.6512
2.0114	0.5868	45.6771	31.7555	25.2417	2.5616	1.0118	-0.3062	3.7664	2.5605

Weakly Heterogeneous

Realization 4

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.5255	31.9387	1.2325	0.9237	0.5364	0.0437	3.3341	2.8758
0.0114	1.0001	24.7905	31.9704	1.5686	0.9335	0.6120	0.0598	3.3389	2.8840
0.2614	1.0001	30.7025	32.7252	9.5570	1.2316	0.9900	0.4702	3.1483	2.9705
0.5113	1.0001	37.0674	33.8865	20.2732	1.8332	1.2673	1.1681	3.0226	3.4015
0.7613	1.0001	43.8226	34.9987	28.7955	2.8739	-0.0533	1.3600	2.7789	4.2744
1.0113	1.0001	49.9215	35.8246	29.6833	4.4048	-0.3514	1.2607	3.2908	3.7884
1.2612	1.0001	55.1106	36.5624	32.2449	5.6833	1.5111	0.5012	3.6051	3.2312
1.5112	1.0001	59.9408	36.9820	40.0071	7.0379	2.9189	-0.5826	3.5617	3.2356
1.7612	1.0001	64.6715	36.9478	49.6706	8.1167	3.8709	-0.8359	3.7613	2.8937
2.0114	1.0001	69.3119	36.6297	62.8012	7.9397	5.9715	-0.3317	4.2149	2.4860

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6573	24.3579	31.9432	0.9167	0.7645	0.3521	0.0203	3.1807	2.8414
0.0114	0.6540	24.5288	31.9647	1.1197	0.7720	0.4229	0.0337	3.2211	2.8361
0.2614	0.6222	28.2513	32.4166	5.5346	0.9205	0.5121	0.3123	2.9985	2.5078
0.5113	0.6087	31.9682	32.9566	10.4568	1.1187	0.3993	0.6519	2.9009	2.5645
0.7613	0.5990	35.8333	33.6933	16.4218	1.5036	0.3177	0.9857	2.9260	2.9551
1.0113	0.5916	39.8614	34.4554	22.9693	1.9756	-0.2747	1.0076	2.7566	3.4322
1.2612	0.5870	43.8273	35.0451	27.1938	2.6313	-1.4505	1.0169	2.8395	3.9086
1.5112	0.5836	47.4702	35.5326	28.2635	3.4056	-2.3453	1.1465	3.2340	3.7965
1.7612	0.5811	50.7090	36.0163	28.1018	4.1141	-2.4624	1.0810	3.6241	3.2532
2.0114	0.5793	53.6520	36.4891	28.7334	4.7387	-1.8666	0.6913	3.7759	2.7810

Weakly Heterogeneous

Realization 5

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.6342	31.8751	1.2954	0.9306	0.6270	-0.0662	3.4826	2.8993
0.0114	1.0001	24.9910	31.8421	1.7537	0.9533	0.7087	-0.1011	3.4130	2.9902
0.2614	1.0001	32.1609	31.4260	8.1997	2.6431	0.3067	-0.2825	3.3005	3.2935
0.5113	1.0001	37.6702	32.2051	13.7678	3.2853	1.3779	1.1126	3.2480	2.6728
0.7613	1.0001	43.1958	33.5891	23.0628	4.0064	1.3951	0.7823	3.0308	2.6101
1.0113	1.0001	49.1041	34.6100	31.7245	3.9321	0.6257	0.5527	2.9064	2.5197
1.2612	1.0001	54.9706	34.8276	35.8996	4.3929	-0.1582	0.1082	2.9869	2.7981
1.5112	1.0001	60.4190	34.5522	37.0424	5.3742	0.2259	-0.4641	3.3737	2.8135
1.7612	1.0001	65.4908	34.1566	41.2988	5.9095	2.2458	-0.4725	3.5998	2.5924
2.0114	1.0001	70.5739	33.8998	52.7726	5.9384	3.8321	0.0756	3.3107	2.7435

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6570	24.4295	31.9101	0.9346	0.7670	0.4180	-0.0461	3.2930	2.8442
0.0114	0.6527	24.6572	31.8873	1.2059	0.7765	0.4974	-0.0563	3.2985	2.8448
0.2614	0.6156	29.4571	31.5118	5.9141	1.6262	-0.1519	-0.7476	2.8911	3.6570
0.5113	0.6025	33.2531	31.5257	7.9238	2.2312	-0.1185	0.4467	3.2614	2.0005
0.7613	0.5967	36.4824	31.9880	10.8013	2.5554	0.2382	1.0398	2.9945	1.9709
1.0113	0.5925	39.6755	32.7706	15.1210	3.1116	0.2315	0.9803	2.8091	1.9284
1.2612	0.5885	43.0268	33.6584	20.4603	3.4169	-0.0506	1.0191	2.7266	0.8364
1.5112	0.5852	46.5126	34.3722	25.7618	3.2714	-0.5896	1.3547	2.7170	-1.3911
1.7612	0.5820	50.0369	34.7699	29.9933	3.0734	-1.2925	1.7984	2.7394	-3.5859
2.0114	0.5793	53.4833	34.8759	32.5293	3.2820	-2.1461	1.6399	2.7792	-2.6667

Weakly Heterogeneous

Realization 1

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0002	24.5650	32.0972	1.2190	0.9206	0.5320	0.0929	3.3641	2.8365
0.0114	1.0001	24.7998	32.1253	1.5555	0.9013	0.6620	0.0925	3.4230	2.8177
0.2614	1.0162	31.0191	32.3710	11.6889	1.1085	0.6393	0.0832	2.9609	3.3187
0.5113	1.0229	37.7214	32.3361	19.0946	2.0918	-0.3219	0.1156	2.9613	3.6916
0.7613	1.0223	43.6802	32.0459	22.0251	3.2795	-0.3200	-0.3909	3.1429	3.8732
1.0113	1.0210	48.9598	31.5878	26.2434	4.0740	0.5177	-0.9232	3.3045	3.4657
1.2612	1.0174	54.0406	31.1383	35.1399	4.0926	2.7005	-0.6385	3.6869	2.9555
1.5112	1.0145	59.4781	30.9321	55.2603	4.1459	4.5371	0.7875	3.2629	4.0273
1.7612	1.0131	65.7145	31.3478	82.6696	6.2863	3.3751	2.7933	2.6347	4.9552
2.0114	1.0105	72.5541	32.4079	102.5586	9.6419	0.6849	2.8137	2.5172	3.6358

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6574	24.3967	32.0628	0.9041	0.7662	0.3409	0.0624	3.1742	2.8111
0.0114	0.6545	24.5444	32.0815	1.0967	0.7563	0.4419	0.0696	3.2558	2.7810
0.2614	0.6321	28.3023	32.3279	6.7853	0.8072	0.4518	0.0404	2.7623	3.1102
0.5113	0.6233	32.4651	32.3653	12.5884	1.1373	-0.1506	0.1078	2.8448	3.4194
0.7613	0.6127	36.5847	32.3423	16.6705	1.6958	-0.9823	0.0998	2.9645	3.5140
1.0113	0.6047	40.3534	32.2287	18.7527	2.3449	-1.4814	-0.0051	3.2168	3.5788
1.2612	0.5987	43.7431	32.0168	20.4055	2.8908	-1.5347	-0.3754	3.3158	3.6954
1.5112	0.5936	46.8980	31.7559	22.2081	3.4021	-1.3627	-0.7971	3.2367	3.6640
1.7612	0.5892	49.8910	31.4891	24.4428	3.7719	-1.1571	-0.8993	3.2785	3.3664
2.0114	0.5850	52.7969	31.2068	27.9337	3.7797	-0.2921	-0.7255	3.4070	3.0169

Weakly Heterogeneous

Realization 2

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0004	24.6993	31.9334	1.2108	0.9600	0.5189	-0.0582	3.2932	2.9481
0.0114	1.0001	25.0241	31.9339	1.5868	0.9911	0.5515	-0.0921	3.2247	3.0002
0.2614	1.0088	31.6313	32.1070	8.7334	1.7535	0.7638	0.1356	3.1444	3.3052
0.5113	1.0111	37.9747	33.0916	14.9573	2.4249	0.1515	0.3641	2.9295	2.8744
0.7613	1.0113	43.7866	33.3662	18.0770	3.1418	0.1250	0.1316	3.0509	2.8657
1.0113	1.0097	49.0082	33.4132	20.2036	4.3569	0.0973	0.5324	2.9461	3.3351
1.2612	1.0081	53.7280	33.9759	22.1761	6.2980	0.5022	1.2654	3.1490	3.5417
1.5112	1.0070	58.1001	34.8734	25.9198	7.5389	1.7511	1.0552	3.5431	2.9368
1.7612	1.0057	62.3809	35.6558	34.3747	6.9106	3.8322	0.4248	3.8596	2.5961
2.0114	1.0052	66.9377	36.0697	50.4426	5.5353	5.0468	0.0822	3.3631	2.6331

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6575	24.4963	31.9505	0.8956	0.7809	0.3721	-0.0355	3.1722	2.8869
0.0114	0.6534	24.7099	31.9540	1.1325	0.7994	0.4159	-0.0538	3.1442	2.9172
0.2614	0.6263	29.0196	31.9280	5.3795	1.2582	0.2298	-0.1132	3.0675	3.2133
0.5113	0.6168	33.0165	32.3301	9.6775	1.6607	0.2251	0.2829	2.9075	3.1707
0.7613	0.6096	36.9431	32.9991	13.3192	2.0849	-0.4431	0.3876	2.8322	2.8716
1.0113	0.6038	40.6181	33.3521	15.3408	2.4149	-0.8267	0.2692	3.0891	2.7864
1.2612	0.5978	44.0186	33.3706	16.9681	2.8342	-0.8624	0.1169	3.1166	2.7713
1.5112	0.5935	47.1735	33.3404	18.3236	3.3721	-1.0376	0.1267	3.0464	2.8555
1.7612	0.5890	50.1167	33.4832	19.1488	4.1160	-1.1525	0.4335	3.0438	3.1584
2.0114	0.5854	52.8557	33.8412	19.9433	5.1594	-1.1387	0.8943	3.0752	3.4259
2.2614	0.5822	55.4352	34.3509	20.9668	6.2228	-0.9559	1.1105	3.1097	3.2681
2.5113	0.5793	57.8999	34.9089	22.5838	6.8578	-0.5438	0.9685	3.2120	2.9372
2.7613	0.5771	60.3122	35.4190	25.4197	6.8371	0.2438	0.6300	3.2945	2.7012
3.0113	0.5751	62.7566	35.8147	30.2946	6.2384	1.2854	0.2926	3.2861	2.6040
3.2612	0.5732	65.3255	36.0674	37.8731	5.4171	2.2151	0.0735	3.1222	2.6138
3.5112	0.5712	68.0944	36.1691	48.2472	4.7056	2.5402	-0.0403	2.8077	2.6319
3.7612	0.5695	71.0834	36.1390	60.0471	4.3977	2.0297	-0.1217	2.4572	2.7462
4.0114	0.5676	74.2608	36.0232	71.1852	4.4939	0.7076	-0.1371	2.2238	2.9654

Weakly Heterogeneous

Realization 3

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.3169	31.8781	1.0553	1.0838	0.3742	-0.0505	3.0832	3.0103
0.0114	0.9999	24.5234	31.8718	1.2678	1.1349	0.4247	-0.0792	3.1624	3.0560
0.2614	1.0052	28.7091	31.7798	6.5092	1.4997	1.4331	-0.1245	3.4697	2.9686
0.5113	1.0113	33.2696	31.9518	15.1130	1.7892	1.0809	-0.1207	2.5626	4.1804
0.7613	1.0151	37.9814	31.9891	19.8872	2.7686	-0.5580	-0.1935	2.8169	4.0055
1.0113	1.0147	42.2918	31.8374	22.7102	3.2999	0.4180	-0.3102	3.7836	3.3501
1.2612	1.0132	46.5432	31.6704	32.3860	3.0797	3.7046	-0.3741	4.1836	3.0354
1.5112	1.0116	51.3891	31.5728	54.3600	2.6140	5.6868	-0.2500	3.4517	2.9599
1.7612	1.0098	57.1682	31.5688	85.4977	2.4247	4.8033	0.1306	2.6544	3.1798
2.0114	1.0077	63.7836	31.6568	115.1971	2.6508	2.2424	0.5186	2.4072	3.4136

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6567	24.2016	31.9027	0.8286	0.8528	0.2313	-0.0384	2.9712	2.9564
0.0114	0.6535	24.3361	31.9012	0.9618	0.8801	0.2794	-0.0478	3.0262	3.0010
0.2614	0.6305	26.9853	31.8163	3.5698	1.2202	0.6300	-0.1520	3.1984	3.0374
0.5113	0.6225	29.5579	31.8372	7.2451	1.2180	1.1304	-0.0805	3.1248	2.9520
0.7613	0.6158	32.3731	31.9685	12.4517	1.3427	0.9175	-0.0113	2.5738	3.7065
1.0113	0.6092	35.3149	32.0550	16.5577	1.7956	-0.3718	0.0634	2.3892	4.1702
1.2612	0.6044	38.1202	32.0302	18.2412	2.3488	-1.4220	0.0152	2.8067	3.7908
1.5112	0.5994	40.7007	31.9436	19.1124	2.7285	-1.6474	-0.0662	3.3305	3.3887
1.7612	0.5954	43.1527	31.8456	20.8686	2.8446	-0.7908	-0.1605	3.6908	3.1335
2.0114	0.5922	45.6185	31.7500	25.3039	2.7467	0.8658	-0.2712	3.7898	3.0278

Weakly Heterogeneous

Realization 4

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.5239	31.9387	1.2303	0.9228	0.5176	0.0441	3.2767	2.8626
0.0114	1.0000	24.7896	31.9703	1.5635	0.9333	0.5977	0.0572	3.2858	2.8772
0.2614	1.0126	30.6546	32.7086	9.5729	1.3320	0.9221	0.2911	3.0484	3.2463
0.5113	1.0214	36.9359	33.8421	20.5697	2.0259	1.1232	0.7680	2.9364	3.4809
0.7613	1.0230	43.6307	34.9415	29.5482	3.0696	-0.3153	0.9899	2.7241	4.0490
1.0113	1.0210	49.7077	35.7693	30.7724	4.6020	-0.9928	1.0551	3.2363	3.6585
1.2612	1.0173	54.9401	36.5142	32.6324	5.9494	0.7977	0.3672	3.4878	3.1669
1.5112	1.0149	59.7941	36.9459	39.5921	7.3705	2.3047	-0.6111	3.3579	3.1596
1.7612	1.0120	64.5352	36.9284	48.5561	8.4341	3.2349	-0.8279	3.4573	2.9002
2.0114	1.0094	69.1830	36.6206	61.1276	8.3064	5.2173	-0.3013	3.8768	2.6718

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6572	24.3564	31.9430	0.9154	0.7628	0.3381	0.0185	3.1469	2.8083
0.0114	0.6539	24.5276	31.9646	1.1150	0.7705	0.4068	0.0317	3.1520	2.8220
0.2614	0.6295	28.2228	32.4029	5.5173	1.0103	0.4859	0.1493	2.9345	3.0769
0.5113	0.6210	31.8988	32.9301	10.4980	1.2800	0.3698	0.3582	2.8305	3.2654
0.7613	0.6137	35.7244	33.6491	16.5428	1.7215	0.3045	0.5846	2.8502	3.3449
1.0113	0.6052	39.7452	34.4055	23.0840	2.1951	-0.2848	0.5990	2.6926	3.5149
1.2612	0.5980	43.7200	35.0017	27.1916	2.8053	-1.4344	0.7332	2.7592	3.7513
1.5112	0.5925	47.3860	35.4911	28.1232	3.5860	-2.3234	0.8952	3.1195	3.7533
1.7612	0.5874	50.6574	35.9841	27.7661	4.3215	-2.4569	0.8998	3.4812	3.3649
2.0114	0.5835	53.6279	36.4694	27.9893	4.9526	-1.8024	0.6176	3.5530	2.9621

Weakly Heterogeneous

Realization 5

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0006	24.6312	31.8747	1.2938	0.9329	0.5988	-0.0624	3.4036	2.8824
0.0114	1.0012	24.9874	31.8413	1.7492	0.9607	0.6807	-0.1010	3.3334	3.0033
0.2614	1.0178	32.0923	31.4080	8.2861	2.8097	0.1847	-0.3922	3.1830	3.3869
0.5113	1.0208	37.5736	32.1638	13.9128	3.6459	1.0794	0.5999	3.0999	3.1832
0.7613	1.0188	43.0746	33.5447	23.1841	4.3633	1.1284	0.3339	2.9083	2.9405
1.0113	1.0162	48.9616	34.5727	32.0508	4.2854	0.3290	0.0560	2.8095	3.2418
1.2612	1.0128	54.8234	34.8100	36.3911	4.6957	-0.5816	-0.2217	2.9103	3.5075
1.5112	1.0098	60.2779	34.5465	37.3722	5.6574	-0.4182	-0.6122	3.2679	3.2386
1.7612	1.0072	65.3641	34.1575	41.1136	6.1792	1.4840	-0.5507	3.4240	2.9120
2.0114	1.0061	70.4427	33.9018	52.0813	6.1791	3.0866	-0.0526	3.1318	2.9587

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6573	24.4276	31.9097	0.9352	0.7680	0.4018	-0.0461	3.2620	2.8409
0.0114	0.6532	24.6549	31.8866	1.2042	0.7810	0.4828	-0.0610	3.2644	2.8801
0.2614	0.6260	29.4149	31.4994	5.9202	1.7641	-0.1713	-0.7692	2.8321	3.8365
0.5113	0.6156	33.1961	31.5013	8.0562	2.5573	-0.1907	-0.0127	3.1833	3.2172
0.7613	0.6089	36.4414	31.9534	10.9872	2.9629	0.2099	0.4676	2.9671	3.1940
1.0113	0.6026	39.6492	32.7395	15.4279	3.4916	0.3323	0.4908	2.8461	2.9114
1.2612	0.5970	43.0114	33.6208	20.8760	3.8464	0.1132	0.2448	2.7637	2.7766
1.5112	0.5917	46.5108	34.3341	26.2016	3.7581	-0.3985	0.0931	2.7506	2.8298
1.7612	0.5871	50.0495	34.7328	30.4424	3.6252	-1.0559	0.1327	2.7943	2.9723
2.0114	0.5831	53.5093	34.8435	33.0080	3.8112	-1.8114	0.1536	2.8844	2.9786

Strongly Heterogeneous

Realization 1

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.8139	31.6708	1.1525	1.0193	0.5189	-0.4007	3.2702	3.4637
0.0114	1.0001	25.1470	31.5712	1.4665	1.1782	0.5016	-0.6011	3.1785	3.8109
0.2614	1.0001	30.3676	29.8174	5.8701	5.4237	1.7517	-2.4155	4.0764	4.5921
0.5113	1.0001	34.3059	29.4057	13.9741	13.7167	2.5575	-3.0027	3.4013	3.3978
0.7613	1.0001	37.8555	29.0756	25.1664	21.0076	3.5156	-3.2834	3.8932	2.6015
1.0113	1.0001	41.2363	28.8377	42.6498	22.5423	5.1986	-2.8632	4.2241	2.3082
1.2612	1.0003	44.4694	29.0222	64.7651	23.2109	6.3768	-1.7201	4.1187	2.5893
1.5112	1.0004	47.5164	29.6311	90.6536	26.7601	7.3464	0.4111	3.8880	3.1473
1.7612	1.0004	50.4920	30.5089	123.6033	33.4786	8.3854	2.2194	3.6198	3.1655
2.0114	1.0004	53.5500	31.4322	167.5898	41.5332	9.3993	2.7412	3.3439	2.8205

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6585	24.5934	31.7687	0.8675	0.7944	0.4033	-0.2309	3.1434	3.1419
0.0114	0.6535	24.8210	31.7112	1.0806	0.8709	0.4224	-0.3469	3.1017	3.3592
0.2614	0.6137	28.5980	30.4080	3.5269	2.8367	0.5991	-1.6094	3.4362	4.2035
0.5113	0.6017	31.2150	29.8550	6.7433	5.0668	1.5400	-1.7735	3.5723	3.5185
0.7613	0.5942	33.5272	29.7291	11.3770	9.4240	1.5110	-2.0776	2.1918	3.1828
1.0113	0.5885	35.6363	29.6160	16.0266	14.6001	0.8933	-2.8908	1.3270	2.9949
1.2612	0.5839	37.5842	29.3816	21.3858	18.3261	0.9009	-3.3973	1.5086	2.7528
1.5112	0.5798	39.4513	29.1425	28.2821	20.1993	1.2834	-3.4886	1.9368	2.5457
1.7612	0.5758	41.2668	29.0158	36.5955	20.9133	1.6788	-3.2378	2.2889	2.3707
2.0114	0.5719	43.0204	29.0481	45.9365	21.3254	2.0984	-2.7109	2.5973	2.3138

Strongly Heterogeneous

Realization 2

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.3087	32.0510	1.0621	1.0961	0.4120	-0.0559	3.2416	2.9995
0.0114	1.0001	24.4333	32.1192	1.2283	1.1256	0.5398	-0.0271	3.4373	2.9490
0.2614	1.0001	27.7548	33.0043	6.2312	1.4894	0.5584	0.0627	2.6681	3.3587
0.5113	1.0001	31.0098	33.3241	8.4257	3.0756	0.2262	0.4208	4.1312	3.9662
0.7613	1.0001	33.5665	33.7597	10.8693	6.0091	3.8887	0.9050	7.9335	3.0784
1.0113	1.0001	35.7230	34.3738	19.2552	8.7946	10.0032	1.1767	12.0611	2.5915
1.2612	1.0001	37.9197	35.0619	41.4495	10.6894	17.2670	1.1580	12.6780	2.3879
1.5112	1.0001	40.5060	35.6720	88.3845	11.8399	23.0354	1.0046	9.4975	2.3086
1.7612	1.0001	43.6102	36.0930	166.2334	12.8736	26.4021	0.6407	6.7305	2.4012
2.0114	1.0000	47.1862	36.2954	276.0819	14.3475	28.7077	-0.2982	5.2242	3.0223

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6568	24.1902	32.0443	0.8237	0.8664	0.2502	-0.0223	3.0653	2.9706
0.0114	0.6548	24.2679	32.0889	0.9119	0.8920	0.3330	0.0080	3.2191	2.9559
0.2614	0.6310	26.2882	32.8143	3.8427	1.1447	0.8576	0.0423	2.8095	2.7276
0.5113	0.6178	28.5330	33.1376	6.4202	1.4136	-0.0846	0.2237	2.5255	3.3870
0.7613	0.6088	30.5365	33.3264	7.2552	2.3087	-0.7638	0.4306	3.1937	3.8864
1.0113	0.6030	32.1783	33.5631	7.5225	3.8862	-0.2581	0.6046	4.4738	3.4308
1.2612	0.5988	33.5387	33.8496	8.4822	5.7996	1.5473	0.8093	5.9155	2.9013
1.5112	0.5952	34.7304	34.1722	10.7869	7.6623	3.8928	0.9912	6.8844	2.5866
1.7612	0.5918	35.8468	34.5303	14.9895	9.2370	6.3578	1.0795	7.7091	2.4191
2.0114	0.5881	36.9709	34.9182	22.1449	10.4076	9.3926	1.0761	8.9653	2.3211

Strongly Heterogeneous

Realization 3

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.4630	31.9121	1.1409	1.0042	0.3787	-0.2129	3.1336	3.0753
0.0114	1.0001	24.5748	31.8671	1.2690	1.0292	0.4382	-0.2515	3.1889	3.1243
0.2614	1.0001	27.1275	30.8872	5.4109	1.3626	2.1869	-0.1155	4.6711	2.7098
0.5113	1.0001	30.6175	31.1846	16.7618	2.5248	3.0754	1.3121	3.3461	3.9358
0.7613	1.0001	35.1706	32.5381	30.9994	4.3883	2.0776	0.9293	3.0171	2.9466
1.0113	1.0001	40.0072	34.0128	40.9598	5.4209	1.4315	1.0892	3.2101	3.6700
1.2612	1.0001	44.3581	35.5647	44.2034	7.3733	1.7052	1.7714	3.5778	3.5267
1.5112	1.0001	47.7724	37.2660	45.0055	9.8518	3.9803	1.4474	4.2515	2.7474
1.7612	1.0001	50.4368	38.9796	51.4292	11.2645	7.4386	0.5042	4.7886	2.4221
2.0114	1.0001	52.9298	40.5023	67.5162	11.1569	9.5718	-0.3284	4.5569	2.5441

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6571	24.3355	31.9696	0.8782	0.7989	0.2661	-0.1221	3.0650	2.9744
0.0114	0.6554	24.4088	31.9429	0.9602	0.8127	0.3094	-0.1477	3.0995	2.9987
0.2614	0.6334	26.0016	31.2299	2.7664	1.1231	0.8078	-0.4044	3.5241	2.6449
0.5113	0.6204	27.7133	30.7495	6.1308	1.1754	1.9920	0.0089	3.9659	2.3681
0.7613	0.6081	29.8751	30.9696	12.5942	1.8865	2.3458	1.1071	3.0622	4.0196
1.0113	0.5970	32.4976	31.7325	20.5298	3.2518	1.6002	1.2043	2.6071	3.0950
1.2612	0.5894	35.3669	32.6322	27.7786	4.0715	0.7944	0.6706	2.7391	2.6996
1.5112	0.5840	38.2867	33.4667	33.7583	4.3834	0.3520	0.4727	2.9629	3.0981
1.7612	0.5797	41.0957	34.2882	37.9064	4.9564	0.0828	0.8326	3.1265	3.5477
2.0114	0.5763	43.6267	35.1849	39.7290	6.1542	0.0195	1.2318	3.3373	3.4365

Strongly Heterogeneous

Realization 4

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.6657	32.0905	1.2748	0.8626	0.6566	0.1443	3.6938	2.7816
0.0114	1.0001	24.7894	32.1040	1.5096	0.8340	0.8229	0.1280	3.8733	2.7439
0.2614	1.0001	28.7187	32.3177	12.5055	0.8126	2.0385	0.5717	3.2965	4.2947
0.5113	1.0001	34.1884	32.9527	29.4587	2.4653	1.3341	1.8522	2.7746	4.6815
0.7613	1.0001	40.0053	33.9561	42.2776	4.8103	0.3102	1.4601	2.9903	2.7270
1.0113	1.0001	45.4969	34.4380	53.8106	5.0338	2.9150	0.7105	4.0165	2.8289
1.2612	1.0001	50.8024	34.3418	85.2176	5.8781	11.4516	3.3705	5.8600	9.6845
1.5112	1.0000	56.6938	34.8208	170.6140	16.4549	19.8898	8.7768	5.3342	8.6222
1.7612	0.9992	63.5846	36.3979	309.7285	41.0308	20.2738	9.3432	3.2971	4.2334
2.0114	0.9941	70.8837	38.5874	444.5628	71.9351	15.0073	7.8423	2.1241	2.5599

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6584	24.4852	32.0393	0.9192	0.7332	0.4178	0.0994	3.3776	2.7793
0.0114	0.6568	24.5587	32.0481	1.0353	0.7171	0.5100	0.0908	3.4692	2.7367
0.2614	0.6308	26.7009	32.2002	5.9570	0.5237	1.4432	0.1245	3.0275	1.4384
0.5113	0.6135	29.6480	32.3467	13.9395	0.8142	1.2350	0.7133	2.9155	4.1596
0.7613	0.5989	33.0856	32.7415	23.3795	1.6846	0.7603	1.4592	2.7623	4.6327
1.0113	0.5902	36.6703	33.3598	31.4702	3.1898	-0.2108	1.8221	2.7022	3.6997
1.2612	0.5842	40.1453	34.0128	37.1346	4.5846	-0.8404	1.4847	2.9182	2.6274
1.5112	0.5794	43.3975	34.4301	41.5726	4.9288	-0.4373	0.9340	3.2560	2.2252
1.7612	0.5756	46.4124	34.4844	47.9712	4.4504	1.4704	0.5622	3.5775	2.3504
2.0114	0.5727	49.2845	34.2941	59.9831	3.9329	4.5431	0.5114	3.8632	3.3579
2.2614	0.5693	52.1786	34.1740	82.7090	5.0255	8.7818	3.0997	4.4130	9.3675
2.5113	0.5652	55.3095	34.4097	124.3444	10.3420	13.9232	6.9092	4.8324	9.5177
2.7613	0.5607	58.8176	35.0439	189.8753	20.6248	17.7377	8.3383	4.3681	6.0570
3.0113	0.5562	62.6720	35.9757	270.4983	34.7306	18.4577	8.4741	3.4380	4.0714
3.2612	0.5512	66.7147	37.1158	350.6550	51.4707	16.5728	8.1029	2.6451	3.0481
3.5112	0.5454	70.8260	38.4182	420.5270	69.4004	13.2609	7.3077	2.1220	2.4314
3.7612	0.5382	74.9496	39.8394	476.8072	86.3054	9.2701	6.0513	1.8145	2.0227
4.0114	0.5289	79.0435	41.3195	518.6431	100.0927	4.9434	4.4164	1.6599	1.7636

Strongly Heterogeneous

Realization 5

1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0001	24.4826	31.7554	1.3514	0.8243	0.5705	0.0224	3.4158	2.8348
0.0114	1.0001	24.7155	31.7969	1.6523	0.8385	0.6539	0.0398	3.3972	2.8460
0.2614	1.0001	29.4282	33.0220	9.4973	1.1083	1.6447	0.6941	3.3526	2.5894
0.5113	1.0001	34.0508	34.4884	24.6371	1.8372	2.5247	1.5976	3.0659	4.5194
0.7613	1.0001	39.1165	36.1270	44.0461	4.7009	1.6027	2.2770	2.5976	5.0638
1.0113	1.0001	44.2890	38.0652	59.3952	9.4440	-0.2230	2.0606	2.7044	3.1516
1.2612	1.0001	49.1355	39.9262	66.8584	12.2138	-1.6990	0.5607	3.3114	2.4826
1.5112	1.0001	53.4863	41.2840	69.1851	12.4011	-1.7597	-0.1700	4.4026	2.5223
1.7612	1.0001	57.4433	42.1896	72.0681	13.1180	0.9583	-0.1236	6.4432	2.5251
2.0114	1.0001	61.1802	42.8298	79.8718	15.9681	7.1712	-0.3562	9.8525	2.8605

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6579	24.3199	31.7904	0.9799	0.6989	0.3657	-0.0072	3.2624	2.7867
0.0114	0.6551	24.4707	31.8183	1.1609	0.7097	0.4415	0.0074	3.2786	2.7882
0.2614	0.6247	27.5306	32.5003	5.1084	0.8800	0.8362	0.4945	3.0490	2.3634
0.5113	0.6106	30.2093	33.3802	10.4661	1.0016	1.3983	0.8851	2.9594	2.0239
0.7613	0.6016	32.9072	34.2403	18.4842	1.3885	1.7092	1.1121	2.8094	3.4139
1.0113	0.5949	35.7717	35.1562	28.3491	2.4057	1.5314	1.2624	2.5792	4.2576
1.2612	0.5899	38.8001	36.1773	38.5346	4.1497	0.6828	1.8611	2.3642	4.4305
1.5112	0.5835	41.9042	37.3585	47.5833	6.8196	-0.6177	2.4235	2.3159	3.5868
1.7612	0.5780	44.9494	38.6118	54.1740	9.5922	-2.1805	1.9636	2.4270	2.5597
2.0114	0.5739	47.8258	39.7587	57.9371	11.1326	-3.7428	1.0484	2.6839	2.1023

Strongly Heterogeneous

Realization 1

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	1.0005	24.8122	31.6704	1.1518	1.0203	0.5056	-0.3923	3.2277	3.4171
0.0114	1.0004	25.1452	31.5715	1.4637	1.1778	0.4861	-0.5840	3.1412	3.7292
0.2614	1.0138	30.3054	29.8360	5.9936	5.4678	1.5056	-2.2133	3.8943	4.2157
0.5113	1.0119	34.2251	29.4184	13.7761	13.8914	2.1396	-3.0568	2.8453	3.4246
0.7613	1.0078	37.7677	29.0843	24.3126	21.2452	2.8546	-3.3706	3.1658	2.6392
1.0113	1.0067	41.1287	28.8393	41.1865	22.8278	4.5318	-3.0132	3.7754	2.3465
1.2612	1.0056	44.3207	29.0080	62.3537	23.2611	5.6724	-2.0271	3.8370	2.4925
1.5112	1.0049	47.3347	29.6097	87.0589	26.7032	6.5216	0.0074	3.6361	3.0145
1.7612	1.0038	50.2730	30.4750	118.1014	33.1930	7.4029	1.6674	3.3771	3.0673
2.0114	1.0028	53.2591	31.3936	159.0339	40.9364	8.1992	2.1824	3.0816	2.8076

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6587	24.5923	31.7683	0.8677	0.7955	0.3951	-0.2269	3.1215	3.1202
0.0114	0.6537	24.8194	31.7121	1.0783	0.8708	0.4068	-0.3320	3.0713	3.3192
0.2614	0.6217	28.5537	30.4222	3.6407	2.9043	0.4264	-1.4617	3.4496	4.0255
0.5113	0.6120	31.1576	29.8793	6.8337	5.3030	1.4481	-1.8314	3.5759	3.8071
0.7613	0.6020	33.4927	29.7373	11.5692	9.7631	1.8293	-2.3213	2.8218	3.5141
1.0113	0.5939	35.6160	29.6298	16.3164	14.9018	1.5825	-3.0928	2.3879	3.2292
1.2612	0.5876	37.5630	29.3993	21.5455	18.6131	1.5290	-3.5846	2.2871	2.9647
1.5112	0.5825	39.4185	29.1590	28.1556	20.5209	1.6074	-3.6521	2.2802	2.7155
1.7612	0.5782	41.2160	29.0243	36.1224	21.3338	1.7608	-3.4047	2.3932	2.5368
2.0114	0.5734	42.9538	29.0446	45.1148	21.7264	2.0368	-2.9618	2.6153	2.4656

Strongly Heterogeneous

Realization 2

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.9998	24.3077	32.0511	1.0584	1.0934	0.3968	-0.0537	3.1811	2.9702
0.0114	0.9998	24.4327	32.1192	1.2247	1.1220	0.5276	-0.0248	3.3799	2.9183
0.2614	1.0050	27.7349	32.9987	6.2443	1.5132	0.5030	0.0051	2.6064	3.3436
0.5113	1.0094	30.9528	33.3109	8.5278	3.1086	-0.0916	0.3610	3.8029	3.8541
0.7613	1.0089	33.4920	33.7398	10.7877	6.0530	2.8595	0.8422	6.4030	3.0450
1.0113	1.0071	35.6300	34.3440	18.0750	8.8764	7.8679	1.0989	8.7548	2.5862
1.2612	1.0054	37.8192	35.0316	38.5900	10.7604	15.3207	1.0980	11.1424	2.3853
1.5112	1.0047	40.3640	35.6446	82.2951	11.8690	21.3558	1.0005	8.8735	2.2907
1.7612	1.0036	43.3973	36.0772	155.0772	12.7780	24.6470	0.7677	6.2276	2.2650
2.0114	1.0013	46.8234	36.3053	254.8609	14.0198	26.3174	0.0887	4.6579	2.5943

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6567	24.1892	32.0443	0.8213	0.8653	0.2365	-0.0209	3.0087	2.9570
0.0114	0.6547	24.2674	32.0889	0.9101	0.8897	0.3250	0.0084	3.1835	2.9281
0.2614	0.6343	26.2747	32.8111	3.8523	1.1709	0.8271	-0.0160	2.7836	2.8578
0.5113	0.6244	28.4928	33.1268	6.4999	1.4628	-0.1487	0.1426	2.5125	3.3849
0.7613	0.6167	30.4700	33.3128	7.4661	2.3549	-0.9597	0.3354	3.1246	3.7531
1.0113	0.6100	32.1114	33.5485	7.7571	3.9375	-0.6404	0.5250	4.2943	3.8310
1.2612	0.6039	33.4758	33.8313	8.6036	5.8551	0.9761	0.7266	5.5222	2.8863
1.5112	0.5990	34.6819	34.1499	10.7280	7.7434	3.2572	0.9099	6.2115	2.5864
1.7612	0.5946	35.8078	34.5085	14.6665	9.3130	5.5643	1.0203	6.6208	2.4138
2.0114	0.5904	36.9368	34.8970	21.2775	10.4705	8.1548	1.0382	7.2848	2.3099

Strongly Heterogeneous

Realization 3

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.9999	24.4620	31.9127	1.1380	1.0018	0.3641	-0.2057	3.0836	3.0427
0.0114	0.9998	24.5740	31.8674	1.2661	1.0267	0.4264	-0.2438	3.1479	3.0758
0.2614	1.0071	27.1125	30.8940	5.3949	1.4125	2.0302	-0.0825	4.4284	2.8152
0.5113	1.0175	30.5782	31.1755	16.5752	2.6194	2.9377	1.1614	3.2305	3.7241
0.7613	1.0243	35.0562	32.4851	30.7419	4.4966	1.9954	0.7238	2.9243	2.8357
1.0113	1.0244	39.8311	33.9307	41.2984	5.5982	1.2936	0.8062	3.1001	3.4944
1.2612	1.0206	44.1585	35.4695	44.9554	7.6021	1.2180	1.4741	3.4176	3.4333
1.5112	1.0152	47.5930	37.1754	45.5015	10.1069	3.0916	1.2270	4.0170	2.7376
1.7612	1.0107	50.2803	38.9018	51.1893	11.4912	6.3660	0.3327	4.4706	2.4086
2.0114	1.0067	52.7886	40.4410	65.9300	11.3258	8.5922	-0.4769	4.2240	2.4996

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6570	24.3346	31.9699	0.8760	0.7977	0.2551	-0.1192	3.0248	2.9536
0.0114	0.6553	24.4085	31.9430	0.9576	0.8108	0.3028	-0.1446	3.0713	2.9753
0.2614	0.6370	25.9887	31.2382	2.7826	1.1641	0.7040	-0.3273	3.4196	2.8480
0.5113	0.6298	27.6967	30.7650	6.1049	1.2937	1.8579	0.0620	3.8298	2.7988
0.7613	0.6227	29.8450	30.9713	12.4896	2.0349	2.2676	0.9468	3.0052	3.8014
1.0113	0.6149	32.4340	31.7025	20.3194	3.3815	1.5677	1.0070	2.5684	2.9999
1.2612	0.6076	35.2553	32.5742	27.6613	4.2202	0.8043	0.4904	2.6813	2.6785
1.5112	0.5998	38.1487	33.3932	33.8936	4.5494	0.3376	0.2605	2.8825	3.0230
1.7612	0.5927	40.9552	34.2142	38.3230	5.1240	0.0085	0.5949	3.0356	3.4312
2.0114	0.5865	43.4968	35.1124	40.2342	6.2830	-0.1689	0.9939	3.2386	3.3289

Strongly Heterogeneous

Realization 4

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.9999	24.6652	32.0904	1.2722	0.8614	0.6469	0.1429	3.6476	2.7738
0.0114	0.9999	24.7886	32.1041	1.5039	0.8326	0.8042	0.1283	3.7924	2.7419
0.2614	1.0147	28.6898	32.3193	12.3432	0.8947	1.9877	0.4485	3.2292	4.1888
0.5113	1.0267	34.0128	32.9265	29.7251	2.5733	1.3282	1.6784	2.6952	4.4273
0.7613	1.0257	39.7193	33.9125	43.9741	4.9583	-0.0395	1.3785	2.8562	2.6857
1.0113	1.0250	45.0887	34.3925	56.5190	5.1739	1.1967	0.5925	3.5360	2.5391
1.2612	1.0216	50.2968	34.2935	85.7572	5.6428	7.7277	2.4200	4.6771	7.3812
1.5112	1.0202	56.1151	34.7447	168.8713	15.4344	17.3892	7.9217	5.0609	7.8764
1.7612	1.0181	62.9614	36.2939	306.7125	39.3595	19.0822	8.9987	3.3136	4.1361
2.0114	1.0117	70.2471	38.4707	443.6142	70.7171	14.4995	7.9109	2.1744	2.6040

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6582	24.4845	32.0386	0.9148	0.7313	0.4050	0.0924	3.3046	2.7535
0.0114	0.6569	24.5590	32.0485	1.0382	0.7186	0.5175	0.0954	3.5110	2.7469
0.2614	0.6402	26.6935	32.1963	5.8912	0.6174	1.4056	0.0267	2.9688	2.9206
0.5113	0.6310	29.5716	32.3374	13.8139	0.9530	1.2794	0.5421	2.8825	4.2143
0.7613	0.6205	32.8903	32.7055	23.7277	1.8509	0.8374	1.2468	2.6804	4.3793
1.0113	0.6102	36.3911	33.3058	32.7369	3.3877	-0.2701	1.6558	2.6019	3.6181
1.2612	0.6027	39.8074	33.9528	39.5872	4.8429	-1.1883	1.3784	2.8178	2.6730
1.5112	0.5964	43.0227	34.3774	45.1289	5.2438	-1.2633	0.8700	3.1931	2.3435
1.7612	0.5900	46.0444	34.4525	52.0296	4.7596	0.1110	0.5793	3.5676	2.4960
2.0114	0.5842	48.9159	34.2685	63.2680	4.1159	2.5317	0.4156	3.7062	2.9728
2.2614	0.5791	51.8243	34.1456	84.3861	4.9569	6.4866	2.4321	4.0316	7.6063
2.5113	0.5728	54.9703	34.3684	122.9311	9.8799	11.8358	6.3535	4.4920	9.0178
2.7613	0.5668	58.4809	34.9803	185.6970	19.6474	16.3858	7.9742	4.2823	5.9277
3.0113	0.5604	62.3325	35.8799	264.0420	33.1567	17.7685	8.1694	3.4626	3.9703
3.2612	0.5539	66.3923	37.0107	342.2943	49.8069	16.2447	7.9265	2.6723	3.0009
3.5112	0.5465	70.5330	38.3194	410.8095	67.9949	13.2583	7.2518	2.1383	2.4231
3.7612	0.5379	74.6934	39.7598	466.7721	85.4241	9.4041	6.0746	1.8153	2.0273
4.0114	0.5276	78.8320	41.2734	509.5995	99.7803	5.1196	4.4479	1.6508	1.7644

Strongly Heterogeneous

Realization 5

1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.9998	24.4817	31.7551	1.3467	0.8220	0.5566	0.0181	3.3609	2.8063
0.0114	0.9999	24.7147	31.7968	1.6461	0.8374	0.6376	0.0387	3.3349	2.8339
0.2614	1.0134	29.3943	32.9825	9.4551	1.2289	1.5249	0.3435	3.2170	3.1242
0.5113	1.0197	33.9758	34.4171	24.3186	2.0340	2.3668	1.0227	2.9686	4.2686
0.7613	1.0194	39.0182	36.0499	43.5169	4.8872	1.3448	2.0451	2.4849	4.7997
1.0113	1.0194	44.1565	37.9629	58.8478	9.8050	-0.6964	1.8526	2.5444	3.1201
1.2612	1.0195	48.9710	39.7962	66.3742	12.8804	-2.5134	0.2925	3.0679	2.5293
1.5112	1.0162	53.2752	41.1590	68.0212	13.1633	-3.5126	-0.5610	3.7650	2.6509
1.7612	1.0102	57.1551	42.1000	68.8642	13.5972	-2.7304	-0.5522	4.7760	2.6873
2.0114	1.0057	60.7776	42.7700	71.2557	16.0947	-0.7257	-0.6139	5.5518	2.8677

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6578	24.3194	31.7904	0.9777	0.6984	0.3573	-0.0070	3.2259	2.7687
0.0114	0.6550	24.4702	31.8182	1.1567	0.7093	0.4298	0.0065	3.2240	2.7768
0.2614	0.6320	27.5105	32.4708	5.1263	0.9709	0.7717	0.2173	3.0184	3.0214
0.5113	0.6259	30.1604	33.3104	10.3860	1.2071	1.2916	0.2817	2.8906	3.1589
0.7613	0.6148	32.8571	34.1710	18.3050	1.5883	1.5976	0.5518	2.7539	3.7067
1.0113	0.6059	35.7299	35.0949	28.0947	2.5414	1.3904	0.9646	2.5255	3.9554
1.2612	0.5993	38.7468	36.1183	38.3135	4.2793	0.5004	1.6351	2.3048	4.2460
1.5112	0.5926	41.8336	37.2871	47.6073	7.0848	-0.8173	2.1242	2.2524	3.6289
1.7612	0.5869	44.8462	38.5282	54.2872	9.9511	-2.4440	1.6883	2.3499	2.6663
2.0114	0.5819	47.7186	39.6789	58.2733	11.5249	-4.0086	0.8154	2.6077	2.2283

Weakly Heterogeneous

Realization 1

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0832	32.0109	0.3985	0.3862	0.1827	0.0267	2.9466	2.8026
0.0114	0.1003	24.3017	32.0327	0.6831	0.3894	0.5071	0.0773	3.7969	2.7401
0.2614	0.1003	30.5664	32.3266	10.3335	0.7665	0.5869	0.0706	3.1047	3.7219
0.5113	0.1003	37.4771	32.3044	16.6069	1.7066	0.1974	-0.0119	3.0806	4.2019
0.7613	0.1003	43.5077	32.0083	19.2034	2.8097	0.7482	-0.6043	2.9738	4.1469
1.0113	0.1003	48.7633	31.5447	23.9195	3.4570	1.3932	-1.3352	3.2561	3.3065
1.2612	0.1003	53.7912	31.1070	33.1011	3.5148	3.1614	-0.9237	3.9904	2.7447
1.5112	0.1003	59.1493	30.8867	52.7506	3.5693	5.0201	0.8998	3.5598	4.6698
1.7612	0.1003	65.3205	31.2515	80.1170	5.5776	3.9321	3.1874	2.7904	5.9430
2.0114	0.1003	72.1358	32.2773	100.9021	8.9329	1.2820	3.1600	2.6264	4.0181

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0486	32.0061	0.3347	0.3276	0.1280	0.0170	2.4937	2.4268
0.0114	0.0635	24.1756	32.0184	0.4882	0.3298	0.3522	0.0542	3.4032	2.3588
0.2614	0.0550	27.5965	32.2760	5.5969	0.3959	0.4755	0.1186	2.7171	-0.5375
0.5113	0.0531	31.4300	32.3473	10.8688	0.6496	-0.3219	0.0110	2.9516	1.7499
0.7613	0.0528	35.1600	32.3482	15.1896	1.0778	-1.0532	-0.0894	3.0819	2.9192
1.0113	0.0527	38.5749	32.2667	18.2966	1.6170	-1.8085	-0.1786	3.3325	3.1158
1.2612	0.0526	41.6637	32.1147	20.5885	2.0511	-2.2345	-0.3557	3.5751	2.8009
1.5112	0.0525	44.5212	31.9064	22.7510	2.4071	-2.4345	-0.7349	3.7023	2.4430
1.7612	0.0523	47.2276	31.6837	25.0383	2.7981	-2.6037	-0.9416	3.7688	2.5970
2.0114	0.0522	49.8347	31.4560	27.3609	3.1393	-2.6521	-0.8502	3.8581	2.8135

Weakly Heterogeneous

Realization 2

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.1047	31.9880	0.3973	0.3857	0.2274	-0.0247	2.9407	2.8168
0.0114	0.1003	24.4493	32.0060	0.8060	0.4218	0.5022	-0.0079	3.5276	3.0217
0.2614	0.1003	31.3631	32.2113	7.8565	0.9914	0.8664	0.6134	3.2192	3.6783
0.5113	0.1003	37.9670	33.2445	13.4047	1.6244	0.2937	1.2054	3.0574	2.6477
0.7613	0.1003	43.9776	33.5434	14.9847	2.4710	1.0881	0.1738	3.0268	3.3794
1.0113	0.1003	49.2690	33.5608	16.6404	3.6130	1.6351	0.6049	2.6829	3.9469
1.2612	0.1003	53.9866	34.1488	19.0083	5.2656	2.0418	1.6871	3.2041	4.0204
1.5112	0.1003	58.3194	35.0978	23.4166	6.1613	3.0523	1.6856	4.0317	2.8857
1.7612	0.1003	62.5364	35.8997	32.5463	5.3390	4.8305	1.1485	4.3843	1.8628
2.0114	0.1003	67.0265	36.2973	49.3916	4.1113	5.7248	0.7146	3.6538	1.6206

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0643	24.0611	31.9929	0.3339	0.3271	0.1605	-0.0183	2.4848	2.4249
0.0114	0.0627	24.2701	32.0103	0.5707	0.3402	0.3910	0.0214	3.3190	2.4516
0.2614	0.0549	28.4338	32.0689	4.5855	0.4960	0.1644	0.5434	3.1053	-3.8318
0.5113	0.0539	32.0983	32.3506	8.8320	0.6605	-0.0374	1.7817	3.0045	-4.7059
0.7613	0.0531	35.7531	32.9696	12.9211	1.0930	-0.8926	1.6093	2.9936	0.1438
1.0113	0.0529	39.2017	33.4196	15.2979	1.5412	-1.7114	0.7517	3.4206	2.2215
1.2612	0.0524	42.3770	33.5402	17.0525	1.8888	-2.0356	0.2294	3.7715	2.3927
1.5112	0.0523	45.2848	33.4949	18.9325	2.2320	-2.2955	0.0901	3.8356	2.1227
1.7612	0.0521	47.9894	33.5224	20.5867	2.6215	-2.6854	0.4006	3.9262	2.2077
2.0114	0.0520	50.5092	33.7162	22.0512	3.1962	-3.0413	0.9914	4.0689	2.7692

Weakly Heterogeneous

Realization 3

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0345	31.9750	0.3900	0.3960	0.0872	-0.0472	2.8706	2.8889
0.0114	0.1003	24.2462	31.9791	0.6294	0.4225	0.3425	-0.0534	3.4385	3.1445
0.2614	0.1003	28.3510	31.9281	5.5290	0.5648	1.5340	-0.1365	3.7740	1.3935
0.5113	0.1003	32.8064	32.1001	14.3972	0.8609	1.5315	0.3898	2.6617	6.5859
0.7613	0.1003	37.6120	32.1907	20.0780	1.6737	-0.4089	0.2377	2.7549	4.8189
1.0113	0.1003	42.0243	32.0719	22.3165	2.0814	0.3463	-0.0272	3.9861	2.8924
1.2612	0.1003	46.3062	31.9115	30.9101	1.9194	4.0003	-0.1720	4.6102	1.6854
1.5112	0.1003	51.1486	31.7944	51.9961	1.6419	6.1989	-0.0221	3.7428	1.5913
1.7612	0.1003	56.9693	31.7559	82.2673	1.6656	5.2555	0.3834	2.8159	3.1092
2.0114	0.1003	63.6787	31.8086	110.1756	2.0740	2.7987	0.7210	2.5479	4.0902

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0195	31.9850	0.3290	0.3326	0.0549	-0.0373	2.4560	2.4853
0.0114	0.0634	24.1451	31.9883	0.4691	0.3460	0.2578	-0.0324	3.1750	2.6472
0.2614	0.0561	26.5985	31.9687	2.7863	0.2711	0.5900	-0.1921	3.3093	-22.1321
0.5113	0.0544	28.7922	31.9660	5.6795	0.2260	1.0647	0.7502	3.2839	-54.6425
0.7613	0.0537	31.1546	32.0799	10.3290	0.3215	1.2992	1.9205	2.8532	-31.3147
1.0113	0.0527	33.7388	32.2069	15.4866	0.5400	0.3642	2.4218	2.2893	-16.5518
1.2612	0.0523	36.3154	32.2562	18.6252	0.8204	-1.1390	2.3679	2.3213	-11.6469
1.5112	0.0523	38.7108	32.2251	19.7961	1.0950	-2.3348	1.9843	2.6443	-8.1605
1.7612	0.0523	40.9390	32.1617	20.2513	1.3282	-3.0061	1.4800	2.7369	-4.7656
2.0114	0.0522	43.0902	32.0849	21.3565	1.5227	-3.1058	0.9148	2.2380	-1.4019

Weakly Heterogeneous

Realization 4

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0699	31.9825	0.3984	0.3859	0.1614	-0.0293	2.9607	2.7902
0.0114	0.1003	24.3359	32.0201	0.7231	0.4037	0.4733	0.0513	3.6469	2.8838
0.2614	0.1003	30.2382	32.7856	8.3620	0.8027	1.0904	0.8778	3.1847	3.0865
0.5113	0.1003	36.5508	33.9180	18.5653	1.5129	1.4217	1.4863	3.0427	3.7144
0.7613	0.1003	43.2704	35.0752	27.2691	2.5941	0.2409	1.4601	2.7366	4.5107
1.0113	0.1003	49.3693	35.9411	28.4225	3.9672	-0.1956	1.4636	3.2214	3.9557
1.2612	0.1003	54.5373	36.7114	30.7446	5.0162	1.5023	0.7807	3.5818	3.2318
1.5112	0.1003	59.3308	37.1974	37.8856	6.0871	2.7791	-0.3831	3.5369	3.3198
1.7612	0.1003	64.0098	37.2193	46.3926	7.0476	3.5648	-0.8203	3.7562	3.0292
2.0114	0.1003	68.5613	36.9007	57.5580	7.0633	5.5993	-0.4527	4.3419	2.4909

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0405	31.9893	0.3337	0.3274	0.1093	-0.0258	2.5033	2.4173
0.0114	0.0631	24.1982	32.0157	0.5179	0.3341	0.3448	0.0522	3.3478	2.4101
0.2614	0.0547	27.6192	32.4806	4.4518	0.4063	0.5164	1.3177	3.0180	-4.4263
0.5113	0.0534	30.8517	32.9389	8.9641	0.6136	0.4413	1.2699	2.8366	-0.8048
0.7613	0.0527	34.1574	33.5291	14.3070	1.0396	0.2866	1.1751	2.8248	2.8376
1.0113	0.0525	37.5890	34.1956	20.3783	1.6736	0.0223	0.7872	2.7758	3.7006
1.2612	0.0524	41.0879	34.7985	25.6763	2.2420	-0.8296	0.6645	2.7117	3.8857
1.5112	0.0522	44.4643	35.3023	28.3676	2.7268	-2.0020	0.9872	2.9257	3.8565
1.7612	0.0521	47.5699	35.7454	28.7066	3.2531	-2.9041	1.1981	3.3598	3.5173
2.0114	0.0520	50.3843	36.1819	28.2241	3.7687	-3.2844	1.0957	3.7719	2.8332

Weakly Heterogeneous

Realization 5

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0826	31.9809	0.3970	0.3852	0.1923	-0.0392	2.9850	2.7985
0.0114	0.1003	24.4313	31.9374	0.8232	0.4101	0.5483	-0.1248	3.7036	2.9179
0.2614	0.1003	31.6991	31.4884	7.2110	1.7213	0.3205	-0.0532	3.3346	3.4698
0.5113	0.1003	37.1390	32.1598	12.6585	2.3058	1.5227	1.7271	3.1729	2.6506
0.7613	0.1003	42.6258	33.5397	22.7023	3.1130	1.3493	1.5373	2.9806	2.1952
1.0113	0.1003	48.6017	34.6496	31.9128	2.9145	0.4061	1.5345	2.9204	0.9046
1.2612	0.1003	54.5710	34.9218	35.8093	3.1720	-0.4321	0.8157	3.0519	1.6105
1.5112	0.1003	60.1012	34.6569	35.9565	4.1032	0.0207	-0.2275	3.4423	2.3369
1.7612	0.1003	65.1963	34.2585	39.0141	4.6812	2.3415	-0.3947	3.6432	2.2629
2.0114	0.1003	70.2530	33.9843	49.6706	4.7468	4.1676	0.1620	3.3141	2.5929

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0643	24.0477	31.9888	0.3324	0.3274	0.1300	-0.0289	2.5014	2.4092
0.0114	0.0627	24.2553	31.9545	0.5719	0.3315	0.4000	-0.1182	3.4251	2.2863
0.2614	0.0538	28.7366	31.5402	5.1388	0.6754	-0.1612	-0.7534	2.8556	-0.7598
0.5113	0.0532	32.1603	31.4914	7.1901	1.1006	-0.7469	0.9603	3.2085	-5.7646
0.7613	0.0529	34.9667	31.7560	9.4935	1.2484	-0.8595	1.2147	2.5041	-5.8248
1.0113	0.0529	37.6993	32.3299	12.8805	1.6920	-1.1282	1.2942	1.8227	-2.3367
1.2612	0.0527	40.5842	33.1260	17.3098	2.1541	-1.3850	1.8082	1.7397	-3.5767
1.5112	0.0525	43.6243	33.9133	22.2715	2.2058	-1.8119	2.5730	1.8026	-9.3375
1.7612	0.0522	46.7823	34.5113	26.6550	1.8926	-2.5757	4.0820	1.8286	-22.2444
2.0114	0.0519	49.9529	34.8373	29.9011	1.6770	-3.6287	5.3684	1.7426	-34.2540

Weakly Heterogeneous

Realization 1

0.1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1001	24.0811	32.0100	0.3941	0.3814	0.1607	0.0164	2.8214	2.6839
0.0114	0.1002	24.2992	32.0318	0.6689	0.3883	0.4660	0.0619	3.5089	2.7598
0.2614	0.1052	30.3360	32.3075	10.3453	0.9072	0.4493	0.0503	2.8007	3.4213
0.5113	0.1034	37.1308	32.2942	17.3890	1.7306	-0.5264	0.0688	2.8960	3.3836
0.7613	0.1027	43.0978	32.0249	20.2569	2.7671	-0.7298	-0.3310	3.0345	3.6360
1.0113	0.1017	48.3908	31.5886	23.0447	3.5893	-0.1677	-0.9397	2.8805	3.4644
1.2612	0.1011	53.3648	31.1285	29.8779	3.6791	1.2933	-0.7489	3.2021	2.9410
1.5112	0.1005	58.6250	30.8401	46.7236	3.3723	3.1836	0.0526	2.9381	3.0094
1.7612	0.0997	64.6939	31.1610	71.8787	4.8945	2.4840	2.0327	2.4122	4.2966
2.0114	0.0985	71.4281	32.1614	91.7358	7.8439	0.0552	2.3105	2.3350	3.2734

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0472	32.0059	0.3313	0.3268	0.1087	0.0140	2.3656	2.3824
0.0114	0.0635	24.1737	32.0175	0.4813	0.3304	0.3190	0.0408	3.1665	2.4070
0.2614	0.0587	27.4487	32.2511	5.4270	0.6066	0.5096	0.0414	2.5481	3.3636
0.5113	0.0562	31.1777	32.3138	10.7794	0.8699	-0.2876	-0.0034	2.6907	3.3727
0.7613	0.0544	34.9330	32.3230	14.9189	1.2125	-1.0283	-0.0056	2.7959	3.2613
1.0113	0.0534	38.4275	32.2594	17.2342	1.7201	-1.7033	-0.0204	3.0037	3.2878
1.2612	0.0526	41.5947	32.1224	18.5421	2.0991	-1.8139	-0.1677	3.1078	3.2393
1.5112	0.0520	44.5259	31.9232	19.7963	2.4883	-1.6520	-0.5317	3.0670	3.4209
1.7612	0.0514	47.2752	31.7067	21.1897	2.8485	-1.5499	-0.8140	2.9452	3.4403
2.0114	0.0510	49.9014	31.4702	22.5527	3.1953	-1.4887	-0.8580	2.9071	3.2806

Weakly Heterogeneous

Realization 2

0.1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1002	24.1037	31.9885	0.3955	0.3816	0.2181	-0.0186	2.8753	2.6787
0.0114	0.1001	24.4455	32.0071	0.7877	0.4188	0.4554	0.0016	3.2418	2.9506
0.2614	0.1048	31.1286	32.1700	8.1907	1.1433	0.5813	0.2574	2.9436	3.4112
0.5113	0.1052	37.5366	33.1579	14.8528	1.9124	-0.4521	0.5565	2.8366	2.9623
0.7613	0.1041	43.4710	33.4799	17.3084	2.5734	-0.6662	0.0512	3.1462	2.8245
1.0113	0.1030	48.7726	33.5002	18.6196	3.5115	-0.4401	0.2034	2.8399	3.0138
1.2612	0.1020	53.4973	34.0644	19.6471	5.1602	-0.2542	1.1577	2.8214	3.4906
1.5112	0.1011	57.8212	34.9945	22.1459	6.2930	0.4453	1.1948	2.9617	2.9284
1.7612	0.1006	62.0118	35.8123	28.7901	5.7831	2.1662	0.6176	3.1912	2.5224
2.0114	0.0998	66.4297	36.2422	42.9053	4.4117	3.7731	0.2363	3.0336	2.4346

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0593	31.9931	0.3300	0.3262	0.1375	-0.0165	2.3261	2.3720
0.0114	0.0628	24.2642	32.0075	0.5569	0.3430	0.3401	0.0114	3.0270	2.5068
0.2614	0.0582	28.2748	32.0194	4.5938	0.7677	0.1441	-0.0494	2.8137	3.3039
0.5113	0.0569	31.8990	32.2820	8.6501	1.0521	0.0180	0.3579	2.7304	3.4601
0.7613	0.0559	35.5166	32.8856	12.6333	1.5130	-0.6725	0.5397	2.6612	3.0810
1.0113	0.0549	38.9710	33.3607	14.8182	1.8217	-1.3859	0.4593	2.9772	2.8285
1.2612	0.0538	42.1723	33.4966	16.0012	2.0852	-1.6049	0.1729	3.2062	2.7494
1.5112	0.0528	45.1786	33.4594	16.8131	2.3720	-1.4517	-0.0273	3.0638	2.7149
1.7612	0.0521	47.9492	33.4795	17.4322	2.7077	-1.3429	0.0269	2.8650	2.7084
2.0114	0.0515	50.5276	33.6543	17.8761	3.2265	-1.4806	0.3274	2.8433	2.9339

Weakly Heterogeneous

Realization 3

0.1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1001	24.0323	31.9758	0.3849	0.3890	0.0625	-0.0362	2.7417	2.6988
0.0114	0.1001	24.2442	31.9782	0.6163	0.4177	0.3192	-0.0628	3.2202	3.0280
0.2614	0.1057	28.1730	31.9169	5.4249	0.7738	1.1495	-0.0655	3.1966	3.2763
0.5113	0.1065	32.4526	32.0708	14.5871	1.0461	1.2179	0.1763	2.4948	4.4291
0.7613	0.1056	37.1294	32.1604	21.0789	1.7954	-1.0330	0.1397	2.5006	3.9527
1.0113	0.1042	41.5004	32.0404	23.1800	2.2661	-1.3375	0.0344	3.3765	3.1892
1.2612	0.1025	45.8028	31.8779	29.4415	2.1818	1.5508	-0.1761	3.8383	2.9195
1.5112	0.1015	50.5933	31.7661	47.5587	1.8591	4.2737	-0.1756	3.1967	2.7969
1.7612	0.1005	56.2840	31.7300	75.1787	1.7212	3.7292	0.1284	2.4230	2.8266
2.0114	0.1000	62.8792	31.7752	102.1359	1.9815	1.1087	0.4752	2.2202	3.1370

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0187	31.9853	0.3276	0.3313	0.0444	-0.0338	2.3814	2.4124
0.0114	0.0634	24.1436	31.9888	0.4646	0.3427	0.2455	-0.0240	3.0559	2.4956
0.2614	0.0601	26.4811	31.9543	2.7605	0.5800	0.4890	-0.1280	2.9120	3.1097
0.5113	0.0590	28.6238	31.9269	5.6852	0.6423	0.8471	-0.1417	2.9749	3.3291
0.7613	0.0577	30.9490	32.0270	10.2452	0.7200	1.0966	0.0027	2.7050	3.8367
1.0113	0.0561	33.4996	32.1470	15.3192	0.9508	0.2638	0.1636	2.2234	4.1933
1.2612	0.0549	36.0981	32.1758	18.8086	1.2871	-1.1542	0.1894	2.3421	3.9081
1.5112	0.0542	38.5595	32.1391	20.0766	1.5994	-2.1871	0.1465	2.8149	3.3024
1.7612	0.0535	40.8658	32.0746	20.5314	1.8246	-2.5600	0.1257	3.3019	2.9751
2.0114	0.0529	43.0923	32.0149	21.4856	1.9169	-2.0606	0.0851	3.6273	2.8288

Weakly Heterogeneous

Realization 4

0.1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1001	24.0678	31.9828	0.3937	0.3808	0.1391	-0.0275	2.8415	2.6812
0.0114	0.1002	24.3339	32.0204	0.7097	0.4024	0.4435	0.0534	3.4050	2.8598
0.2614	0.1055	30.0302	32.7024	8.3230	1.0286	0.8441	0.2661	2.8517	3.5483
0.5113	0.1048	36.2039	33.8002	18.4876	1.8296	0.8517	0.5382	2.7749	3.6068
0.7613	0.1038	42.8014	34.9479	27.7389	2.7457	-0.4347	0.6663	2.5601	3.7464
1.0113	0.1019	48.9375	35.8508	27.9422	3.8734	-1.5035	1.0437	3.0027	3.4432
1.2612	0.1008	54.1458	36.6675	27.9431	4.8750	-0.2125	0.6491	3.0636	2.8214
1.5112	0.1000	58.9140	37.1810	32.6953	5.9306	1.0965	-0.4336	2.8104	2.9351
1.7612	0.0995	63.5523	37.2193	39.5655	6.9485	1.4631	-0.9002	2.8722	2.8924
2.0114	0.0987	68.0172	36.9230	46.9191	6.9914	2.5328	-0.4695	3.2003	2.6040

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0391	31.9894	0.3306	0.3264	0.0906	-0.0251	2.3795	2.3768
0.0114	0.0631	24.1961	32.0152	0.5092	0.3336	0.3101	0.0361	3.1200	2.4391
0.2614	0.0588	27.4704	32.3910	4.3494	0.7053	0.4570	0.1530	2.7560	3.4447
0.5113	0.0570	30.6657	32.8247	8.5828	0.9690	0.3171	0.1625	2.6244	3.4130
0.7613	0.0554	33.9735	33.4095	13.5381	1.4059	0.1283	0.2549	2.6247	3.6330
1.0113	0.0540	37.4415	34.1105	19.0927	1.8817	-0.1011	0.2623	2.5905	3.5258
1.2612	0.0528	40.9862	34.7550	23.5520	2.2260	-0.8024	0.3635	2.5016	3.4310
1.5112	0.0517	44.4297	35.2732	25.4198	2.5058	-1.8243	0.6659	2.6708	3.2154
1.7612	0.0513	47.6015	35.7254	25.2065	3.0097	-2.5049	0.9141	2.9973	3.1217
2.0114	0.0508	50.4684	36.1735	23.9358	3.5439	-2.4575	0.8913	3.1795	2.8813

Weakly Heterogeneous

Realization 5

0.1 Kilogram of Bromide

Only Concentrations Greater than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1002	24.0810	31.9814	0.3937	0.3805	0.1767	-0.0308	2.8939	2.6816
0.0114	0.1002	24.4260	31.9386	0.8002	0.4108	0.4911	-0.1208	3.3485	2.9027
0.2614	0.1055	31.4352	31.5208	7.6589	1.9270	-0.1636	-0.2866	2.9845	3.5540
0.5113	0.1047	36.8207	32.1184	12.9066	2.6542	0.5271	0.7844	2.8873	3.1933
0.7613	0.1036	42.2901	33.4775	22.1373	3.4651	0.7247	0.6818	2.6903	2.7035
1.0113	0.1027	48.1541	34.5718	31.4587	3.2490	-0.3303	0.3573	2.6958	2.7130
1.2612	0.1017	54.1094	34.8767	35.4901	3.4093	-1.4854	0.1617	2.8072	2.9452
1.5112	0.1006	59.6718	34.6415	34.3906	4.2488	-1.4383	-0.3943	3.0513	2.8802
1.7612	0.1001	64.7649	34.2459	35.8918	4.8060	0.4298	-0.4914	3.1020	2.6673
2.0114	0.0994	69.7412	33.9541	44.3272	4.7017	2.0126	-0.1773	2.7933	2.6045

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0461	31.9891	0.3289	0.3259	0.1079	-0.0260	2.3508	2.3714
0.0114	0.0628	24.2481	31.9606	0.5562	0.3384	0.3423	-0.0792	3.0786	2.4461
0.2614	0.0583	28.5340	31.6141	5.2582	1.0493	-0.1186	-0.5804	2.5986	4.2195
0.5113	0.0562	31.9991	31.5118	7.3846	1.6772	-0.7014	-0.1362	3.0136	3.2725
0.7613	0.0551	34.9185	31.7377	9.6974	1.9077	-0.2948	0.3468	2.9186	3.1082
1.0113	0.0542	37.7238	32.2900	13.1357	2.2572	0.0193	0.5968	2.7223	2.8457
1.2612	0.0535	40.6641	33.0600	17.4903	2.8023	0.0232	0.5038	2.5823	2.5857
1.5112	0.0529	43.7453	33.8372	22.6545	2.9806	-0.3616	0.2854	2.5519	2.4848
1.7612	0.0522	46.8950	34.4210	27.1973	2.7575	-0.9363	0.1016	2.6023	2.5530
2.0114	0.0516	50.0800	34.7483	30.4543	2.5554	-1.5825	0.1641	2.7106	2.5499

Strongly Heterogeneous

Realization 1

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.1305	31.9503	0.3994	0.3836	0.2708	-0.1109	2.8912	2.8368
0.0114	0.1003	24.5097	31.8821	0.7961	0.4825	0.5130	-0.3255	3.3885	3.6734
0.2614	0.1003	30.3498	30.3579	4.7511	2.7441	1.9449	-1.8107	3.9363	5.3021
0.5113	0.1003	34.3672	30.3219	12.3575	7.5473	2.2057	-2.0912	2.7481	4.1676
0.7613	0.1003	37.7812	30.3280	21.3696	12.2836	1.6927	-3.2821	2.4963	3.7556
1.0113	0.1003	40.7858	30.0825	32.8313	14.3335	1.7441	-3.5900	2.6785	3.5483
1.2612	0.1003	43.5550	30.0647	46.6237	16.7357	2.1334	-2.4311	3.0956	3.6197
1.5112	0.1003	46.1944	30.4616	64.7731	21.8977	3.6565	0.0702	3.6486	3.8349
1.7612	0.1003	48.8632	31.2092	91.8597	29.8972	6.0826	2.0113	3.8795	3.5285
2.0114	0.1003	51.6896	32.1111	131.1336	38.5751	8.3452	2.5526	3.6997	2.9965

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0643	24.0763	31.9710	0.3361	0.3264	0.1954	-0.0776	2.4812	2.4153
0.0114	0.0623	24.3168	31.9382	0.5668	0.3690	0.4374	-0.1893	3.1956	2.9453
0.2614	0.0549	28.2733	30.8510	2.8742	1.2625	0.5424	-1.0838	3.0866	2.4097
0.5113	0.0539	30.7809	30.4214	5.4509	1.6999	1.0132	1.7788	3.3309	-16.2163
0.7613	0.0534	32.9037	30.4485	9.2040	3.3158	0.6196	2.6006	0.6459	-9.8325
1.0113	0.0530	34.8414	30.5829	12.5463	6.0245	-1.4291	0.4680	-3.3324	1.2195
1.2612	0.0526	36.5885	30.5748	15.2512	8.6656	-4.2499	-1.6136	-7.6975	1.9851
1.5112	0.0524	38.1959	30.4227	18.3042	10.6091	-6.5994	-2.8925	-10.6767	3.0243
1.7612	0.0520	39.7153	30.2384	22.2019	11.9711	-7.9463	-3.4878	-11.1466	3.1660
2.0114	0.0517	41.1531	30.1046	26.5014	13.1690	-9.0030	-3.6112	-10.9278	3.0986

Strongly Heterogeneous

Realization 2

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0291	32.0120	0.3851	0.3969	0.0782	0.0195	2.8618	2.9065
0.0114	0.1003	24.1478	32.0776	0.4998	0.4492	0.3404	0.1591	3.6257	3.1342
0.2614	0.1003	27.4915	33.0545	5.1865	1.0059	0.7338	0.4530	2.5885	3.3337
0.5113	0.1003	30.8505	33.4105	6.9522	2.3360	-0.3520	0.8175	3.7315	4.4868
0.7613	0.1003	33.3516	33.8630	7.7483	5.1743	2.4599	1.1101	7.8781	3.3022
1.0113	0.1003	35.3346	34.4278	12.7204	8.1674	8.2521	1.2792	13.4464	2.6563
1.2612	0.1003	37.2638	35.0313	27.3068	10.3275	15.4967	1.2812	15.8779	2.3949
1.5112	0.1003	39.4895	35.5863	60.8976	11.5648	22.1520	1.1530	12.5335	2.2854
1.7612	0.1003	42.1883	35.9946	121.6045	12.3592	26.4635	0.8728	8.7240	2.3025
2.0114	0.1003	45.3626	36.2156	212.4221	13.2549	29.3355	0.1854	6.4961	2.6928

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0160	32.0073	0.3263	0.3331	0.0467	0.0174	2.4233	2.4964
0.0114	0.0637	24.0841	32.0447	0.3865	0.3638	0.2169	0.1124	3.0267	2.7552
0.2614	0.0565	24.9633	32.7867	2.6716	0.6919	1.1986	0.6282	3.0394	1.5341
0.5113	0.0549	28.0720	33.1392	5.2923	0.8613	0.1734	0.7596	2.3670	2.2766
0.7613	0.0544	29.9730	33.3110	6.3586	1.4234	-0.9318	0.9341	2.8999	3.7129
1.0113	0.0540	31.5254	33.5207	6.4673	2.5280	-1.5064	1.0730	3.9082	3.6963
1.2612	0.0538	32.7776	33.7767	6.4608	4.0970	-1.3091	1.0601	4.7949	3.2393
1.5112	0.0537	33.8360	34.0277	6.9065	5.9055	-0.4051	1.0451	4.4834	2.8570
1.7612	0.0535	34.7878	34.2770	8.1408	7.6456	0.7391	1.0990	2.0626	2.5986
2.0114	0.0532	35.7029	34.5482	10.4592	9.0862	2.0251	1.1746	-0.4388	2.4365

Strongly Heterogeneous

Realization 3

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0730	32.0061	0.3976	0.3859	0.1574	-0.0016	2.9174	2.8366
0.0114	0.1003	24.1943	31.9759	0.5387	0.3980	0.3384	-0.0874	3.4068	2.9843
0.2614	0.1003	26.8615	30.9294	4.0002	0.8532	1.8388	-0.7526	4.9532	2.0143
0.5113	0.1003	30.3409	31.0050	13.8751	1.9380	3.1868	1.5338	3.5160	5.0305
0.7613	0.1003	35.0390	32.4145	26.8111	3.9856	2.2496	1.1577	3.0626	2.9364
1.0113	0.1003	40.0800	33.9578	35.1844	4.7510	2.1730	1.2830	3.2587	3.6524
1.2612	0.1003	44.6129	35.5213	37.8140	6.4405	3.1717	2.1799	3.3748	3.5781
1.5112	0.1003	48.1201	37.2427	38.8262	8.9254	5.7513	1.8352	3.9107	2.6483
1.7612	0.1003	50.7665	38.9934	45.8957	10.4065	8.7783	0.7881	4.6419	2.2794
2.0114	0.1003	53.1932	40.5511	62.6942	10.3237	10.2059	-0.0975	4.5506	2.4032

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0430	32.0048	0.3338	0.3273	0.1125	0.0085	2.4916	2.4275
0.0114	0.0638	24.1151	31.9893	0.4149	0.3321	0.2476	-0.0407	3.0448	2.4988
0.2614	0.0563	25.7367	31.3335	2.0101	0.5270	0.5620	-1.4915	3.3565	1.5723
0.5113	0.0540	27.2817	30.7168	4.1944	0.6423	1.6476	-1.3393	4.2382	-1.8377
0.7613	0.0534	29.1084	30.7115	8.9325	1.0602	2.4391	0.6918	3.4088	4.5933
1.0113	0.0526	31.3682	31.2952	15.7252	2.2862	1.9321	1.5880	2.6081	4.0884
1.2612	0.0521	33.9121	32.1357	22.4394	3.3940	0.9933	1.1700	2.5424	2.8385
1.5112	0.0518	36.5478	32.9343	28.2650	3.8018	0.3943	0.6917	2.7998	2.7488
1.7612	0.0516	39.1486	33.6428	33.3629	4.0236	0.1044	0.6710	2.9875	3.2532
2.0114	0.0515	41.6130	34.3639	37.2489	4.6601	-0.1634	1.0400	3.1075	3.6072

Strongly Heterogeneous

Realization 4

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.1086	31.9985	0.4009	0.3818	0.2284	0.0086	2.9413	2.7714
0.0114	0.1003	24.2005	32.0008	0.5328	0.3746	0.4619	0.0135	3.8500	2.6586
0.2614	0.1003	27.5982	32.1779	9.4017	0.5197	2.0464	0.4135	3.3586	4.3678
0.5113	0.1003	32.9469	32.7149	25.0817	1.8735	1.5178	1.7857	2.9583	5.4460
0.7613	0.1003	38.8994	33.7445	37.0593	4.3081	0.5738	1.6943	3.0266	3.0633
1.0113	0.1003	44.5670	34.4433	45.1065	5.0007	2.9415	0.8054	3.8691	2.6255
1.2612	0.1003	49.8832	34.4010	67.1620	5.1000	11.1047	2.0577	5.8673	7.5298
1.5112	0.1003	55.5478	34.6226	136.3299	12.5085	20.0768	8.0238	6.0302	10.2356
1.7612	0.1002	62.2119	35.9574	266.3285	34.3021	21.6056	9.4801	3.7697	4.9268
2.0114	0.0999	69.5050	38.0711	407.4573	64.8460	16.7487	8.2405	2.3134	2.8064

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0643	24.0641	31.9980	0.3364	0.3259	0.1650	-0.0018	2.5053	2.4012
0.0114	0.0639	24.1151	31.9983	0.4027	0.3220	0.2985	0.0026	3.1961	2.3096
0.2614	0.0578	25.7570	32.0620	3.7254	0.2877	1.5777	-0.1068	3.4610	-1.5647
0.5113	0.0545	28.2482	32.1854	10.1853	0.4679	1.1081	0.3163	2.7518	3.0067
0.7613	0.0530	31.2299	32.4706	18.0048	1.0365	0.6515	1.1263	2.8723	4.8341
1.0113	0.0524	34.4245	32.9410	25.8832	2.0263	-0.0107	1.6463	2.8042	4.4147
1.2612	0.0520	37.6283	33.5481	32.0411	3.4180	-0.8596	1.8103	2.8876	3.4109
1.5112	0.0517	40.6991	34.1231	36.3214	4.5037	-1.2441	1.4434	3.1343	2.5034
1.7612	0.0514	43.5614	34.4733	40.0114	4.7305	-0.7804	0.9518	3.4362	2.1536
2.0114	0.0512	46.2230	34.5252	45.4859	4.3394	0.8304	0.6093	3.7089	2.2366

Strongly Heterogeneous

Realization 5

0.1 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1003	24.0582	31.9396	0.4038	0.3781	0.1413	-0.1095	3.0174	2.7028
0.0114	0.1003	24.2849	31.9864	0.6804	0.4026	0.4519	-0.0062	3.6708	2.8318
0.2614	0.1003	28.6623	33.1060	7.2124	0.7829	2.0476	1.1340	3.7433	2.2566
0.5113	0.1003	32.7784	34.4305	19.6326	1.4673	3.4870	1.4538	3.5069	4.6268
0.7613	0.1003	37.4601	35.9347	36.8650	3.7434	3.1879	2.3380	2.8196	5.5936
1.0113	0.1003	42.5420	37.8543	52.6208	7.7379	1.4045	2.7571	2.4993	3.4518
1.2612	0.1003	47.5767	39.8785	60.7425	10.3088	-0.9832	1.5920	2.6837	2.1030
1.5112	0.1003	52.2236	41.4860	61.2783	10.0256	-2.6842	1.0575	3.3992	1.4804
1.7612	0.1003	56.4149	42.5705	59.0495	9.9520	-2.3102	1.1657	4.9129	1.3400
2.0114	0.1003	60.2492	43.3099	58.3035	12.0990	0.8768	0.5236	7.6212	2.4980

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0334	31.9635	0.3364	0.3247	0.0919	-0.0903	2.5453	2.3491
0.0114	0.0633	24.1669	31.9932	0.4925	0.3363	0.3190	-0.0010	3.3279	2.4190
0.2614	0.0554	26.8062	32.6295	3.5706	0.4670	0.9995	1.3747	3.2367	-0.7052
0.5113	0.0542	28.8896	33.3147	6.9196	0.7636	1.8356	1.0319	3.4552	1.9220
0.7613	0.0539	30.9608	33.9827	11.4854	1.1242	2.3100	0.6099	3.2777	3.3715
1.0113	0.0538	33.2059	34.6531	16.9673	1.7464	2.0328	0.6210	2.8204	4.0834
1.2612	0.0533	35.6645	35.4597	23.5038	2.5732	1.2238	1.3546	2.1416	4.0975
1.5112	0.0528	38.2982	36.4114	30.4530	3.9128	-0.4127	2.3934	1.1238	4.1453
1.7612	0.0522	41.0443	37.5135	36.3509	5.9603	-3.1971	3.0944	-0.2910	3.2415
2.0114	0.0516	43.7938	38.7063	40.2236	8.0202	-7.0630	2.9961	-2.0622	1.8872

Strongly Heterogeneous

Realization 1

0.1 Kilogram of Bromide

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1002	24.1293	31.9503	0.3959	0.3811	0.2554	-0.1057	2.7876	2.7704
0.0114	0.1003	24.5028	31.8852	0.7870	0.4763	0.4618	-0.2743	3.1717	3.3213
0.2614	0.1040	30.1542	30.4257	5.1310	2.8300	1.0975	-1.2617	3.7717	3.9564
0.5113	0.1020	34.1844	30.3423	12.2176	7.4899	1.9722	-1.9241	2.6239	3.7805
0.7613	0.1009	37.5749	30.3778	20.5280	12.0421	1.4386	-3.2673	2.2550	3.7662
1.0113	0.0993	40.5019	30.1568	30.0311	13.9608	0.8158	-3.8900	2.0076	3.7475
1.2612	0.0980	43.0842	30.1188	39.7027	15.8345	-0.1915	-3.4588	1.8531	3.4811
1.5112	0.0960	45.3380	30.3925	49.1570	19.3491	-0.9543	-2.0249	1.8943	3.2439
1.7612	0.0938	47.5017	31.0362	63.6224	26.5374	0.0241	0.4130	2.3359	3.2686
2.0114	0.0923	49.7946	31.8703	89.1476	34.7876	2.7228	1.2385	2.8174	2.7532

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0643	24.0743	31.9717	0.3316	0.3240	0.1697	-0.0684	2.2977	2.3471
0.0114	0.0625	24.3060	31.9402	0.5628	0.3718	0.3635	-0.1748	2.9472	2.9195
0.2614	0.0576	28.1050	30.9153	3.1579	1.5158	0.0360	-0.7015	3.1486	3.3014
0.5113	0.0561	30.6333	30.4203	5.4713	2.5367	0.8191	-0.8282	3.4349	3.2986
0.7613	0.0545	32.8391	30.3694	9.1338	4.5013	1.6968	-0.9105	2.8399	3.2759
1.0113	0.0535	34.8778	30.5357	13.4988	7.0970	1.5388	-1.3926	2.2854	3.1661
1.2612	0.0527	36.6914	30.5474	17.6589	9.0814	1.2072	-2.1828	2.1235	3.3051
1.5112	0.0517	38.3354	30.4451	22.0494	10.4353	1.0575	-2.9805	2.0746	3.6036
1.7612	0.0512	39.8615	30.2929	27.1974	11.4838	0.7658	-3.5533	1.9730	3.8079
2.0114	0.0504	41.2898	30.1902	32.3563	12.3913	0.3456	-3.8931	1.8643	3.9243

Strongly Heterogeneous

Realization 2

0.1 Kilogram of Bromide

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1001	24.0269	32.0118	0.3806	0.3897	0.0553	0.0170	2.7303	2.7105
0.0114	0.1001	24.1451	32.0781	0.4876	0.4459	0.2895	0.1644	3.2447	3.0732
0.2614	0.1046	27.3220	33.0148	5.3947	1.1082	0.4741	0.1532	2.4733	3.2213
0.5113	0.1035	30.6259	33.3875	7.4841	2.3130	-1.1501	0.6153	3.2270	3.9060
0.7613	0.1023	33.0990	33.8242	7.6934	5.0898	-0.0089	0.8869	5.1875	3.1314
1.0113	0.1009	35.0663	34.3667	10.5545	8.0931	3.9960	1.0721	6.6085	2.5767
1.2612	0.0999	36.8228	34.9611	17.7126	10.2412	7.4983	1.1322	6.8254	2.3295
1.5112	0.0984	38.5427	35.5556	32.0896	11.5015	12.2093	1.0251	8.5262	2.2277
1.7612	0.0971	40.7191	36.0053	69.8959	11.9458	19.6762	0.8655	8.7125	2.1594
2.0114	0.0960	43.1891	36.3126	127.3922	12.2054	23.2531	0.6796	6.4504	2.1114

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0153	32.0072	0.3246	0.3315	0.0370	0.0165	2.3587	2.4242
0.0114	0.0636	24.0814	32.0427	0.3806	0.3574	0.1894	0.0818	2.8428	2.5549
0.2614	0.0605	25.8195	32.7244	2.8440	0.8712	0.8757	0.1224	2.9525	2.8960
0.5113	0.0583	27.8462	33.0910	5.7295	1.0792	-0.1143	0.1468	2.3707	3.3061
0.7613	0.0568	29.7591	33.2821	6.9575	1.5491	-1.2411	0.4521	2.8467	3.4810
1.0113	0.0556	31.3447	33.5035	7.0128	2.5368	-1.9183	0.7405	3.7597	3.4169
1.2612	0.0545	32.6292	33.7637	6.6411	4.0228	-1.8826	0.7998	4.6354	3.1239
1.5112	0.0539	33.7425	34.0095	6.8306	5.8253	-0.5067	0.8069	5.3617	2.7483
1.7612	0.0532	34.6937	34.2633	7.7111	7.4858	1.3405	0.9130	5.5136	2.4874
2.0114	0.0524	35.6144	34.5352	9.4161	8.9900	3.4683	1.0568	5.4010	2.3584

Strongly Heterogeneous

Realization 3

0.1 Kilogram of Bromide

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1001	24.0715	32.0070	0.3935	0.3807	0.1411	0.0107	2.8147	2.6810
0.0114	0.1001	24.1917	31.9779	0.5275	0.3926	0.2951	-0.0607	3.1386	2.8498
0.2614	0.1061	26.7342	31.0206	3.9056	1.0920	1.4743	-0.2501	4.1249	2.8005
0.5113	0.1060	30.0577	31.0314	13.3312	2.0565	2.5961	1.1259	3.0237	3.9034
0.7613	0.1056	34.5465	32.3063	26.6295	3.8972	1.4560	0.7556	2.6588	2.6466
1.0113	0.1036	39.4873	33.7622	35.9575	4.6897	0.6927	0.5737	2.9196	3.0940
1.2612	0.1019	44.0395	35.3060	38.5873	6.3302	0.8406	1.3358	3.1674	3.1581
1.5112	0.1003	47.5419	37.0500	36.6337	8.7810	2.3509	1.1688	3.4188	2.4862
1.7612	0.0991	50.1256	38.8505	38.9591	10.2624	5.0167	0.3158	3.6947	2.2341
2.0114	0.0979	52.4583	40.4643	50.4064	9.9578	6.3746	-0.5848	3.4221	2.3168

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0418	32.0050	0.3311	0.3261	0.0963	0.0117	2.3789	2.3761
0.0114	0.0637	24.1140	31.9900	0.4125	0.3300	0.2328	-0.0246	2.9509	2.4018
0.2614	0.0610	25.6244	31.4405	1.9963	0.7935	0.4341	-0.4891	2.9534	3.4066
0.5113	0.0587	27.0749	30.8502	4.0797	1.0933	1.1633	-0.0297	3.5475	3.0338
0.7613	0.0578	28.8684	30.7849	8.7005	1.4035	2.0032	0.6010	3.1078	3.6265
1.0113	0.0563	31.0549	31.2781	15.2318	2.4152	1.6038	1.1097	2.4305	3.4223
1.2612	0.0548	33.5371	32.0455	21.8261	3.4703	0.6183	0.8234	2.3313	2.6935
1.5112	0.0534	36.1759	32.7955	27.6024	3.8153	-0.0172	0.3126	2.5976	2.5711
1.7612	0.0523	38.7896	33.4897	32.3501	3.9142	-0.4669	0.1335	2.7926	2.8795
2.0114	0.0513	41.3453	34.2099	34.7893	4.3566	-0.6752	0.4045	2.8663	3.0925

Strongly Heterogeneous

Realization 4

0.1 Kilogram of Bromide

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1001	24.1067	31.9976	0.3964	0.3782	0.2070	-0.0033	2.8046	2.6761
0.0114	0.1002	24.1993	31.9996	0.5255	0.3737	0.4300	-0.0004	3.5984	2.6653
0.2614	0.1038	27.4873	32.1835	8.8932	0.5930	1.7862	0.3145	2.9704	3.4947
0.5113	0.1042	32.5929	32.6633	24.7715	1.8660	1.3345	1.4323	2.7149	4.5919
0.7613	0.1033	38.3408	33.6596	38.3229	4.3647	-0.2855	1.6263	2.6918	2.9708
1.0113	0.1027	43.8092	34.3783	47.8767	5.0955	-0.0481	0.8714	3.2577	2.3268
1.2612	0.1012	48.7461	34.2858	61.5924	4.0885	3.1541	0.5163	3.5169	2.6904
1.5112	0.0988	53.6808	34.2228	96.7571	7.0537	9.5305	4.8227	3.8285	9.3355
1.7612	0.0962	59.4266	35.1944	195.4067	22.7004	17.0575	8.1561	3.9597	5.1892
2.0114	0.0942	66.1250	37.0093	332.0241	49.7571	16.5235	7.8399	2.6470	2.8511

Reactive Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0630	31.9985	0.3318	0.3247	0.1450	-0.0026	2.3321	2.3566
0.0114	0.0639	24.1126	31.9978	0.3953	0.3213	0.2637	-0.0050	2.9670	2.3284
0.2614	0.0602	25.6911	32.0983	3.5660	0.4051	1.4759	0.1585	3.2103	2.8789
0.5113	0.0579	28.0418	32.1792	9.5524	0.6219	0.9896	0.2336	2.4836	3.3507
0.7613	0.0559	30.8939	32.4170	17.6158	1.1458	0.7146	0.7833	2.6637	4.0503
1.0113	0.0545	34.0077	32.8477	25.9678	2.0742	-0.0274	1.3131	2.5806	3.9512
1.2612	0.0533	37.1390	33.4522	32.8107	3.5137	-1.1192	1.6740	2.6350	3.4058
1.5112	0.0526	40.1303	34.0299	37.7151	4.7333	-1.9095	1.3875	2.8690	2.5818
1.7612	0.0519	43.0076	34.4222	41.1699	4.9820	-1.9964	0.9219	3.1697	2.2545
2.0114	0.0511	45.6463	34.5223	45.2681	4.5114	-0.8402	0.6852	3.4439	2.3295

Strongly Heterogeneous

Realization 5

0.1 Kilogram of Bromide

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.1002	24.0575	31.9403	0.4012	0.3748	0.1322	-0.1001	2.9336	2.6072
0.0114	0.1001	24.2821	31.9853	0.6674	0.3979	0.4176	-0.0198	3.4484	2.7242
0.2614	0.1050	28.5347	32.9795	6.9261	1.0458	1.6644	0.1863	3.2998	3.3631
0.5113	0.1038	32.5826	34.2881	17.8887	1.7412	2.5526	0.3598	2.9411	4.1872
0.7613	0.1022	37.1646	35.8078	34.0064	3.6210	2.1803	1.7094	2.4419	4.6250
1.0113	0.1014	42.1643	37.7159	49.8081	7.7591	0.1855	2.4206	2.1425	3.3721
1.2612	0.1011	47.1935	39.7223	58.7476	10.6698	-2.3194	1.1597	2.3124	2.2077
1.5112	0.1008	51.7933	41.3101	60.3902	10.7273	-4.4994	0.0839	2.8275	2.2058
1.7612	0.1002	55.9512	42.3944	57.4724	10.4279	-5.6928	-0.1233	3.4632	2.3168
2.0114	0.0993	59.6844	43.1464	54.3917	11.7527	-5.4260	-0.3429	4.1120	2.3528

Reactive Run

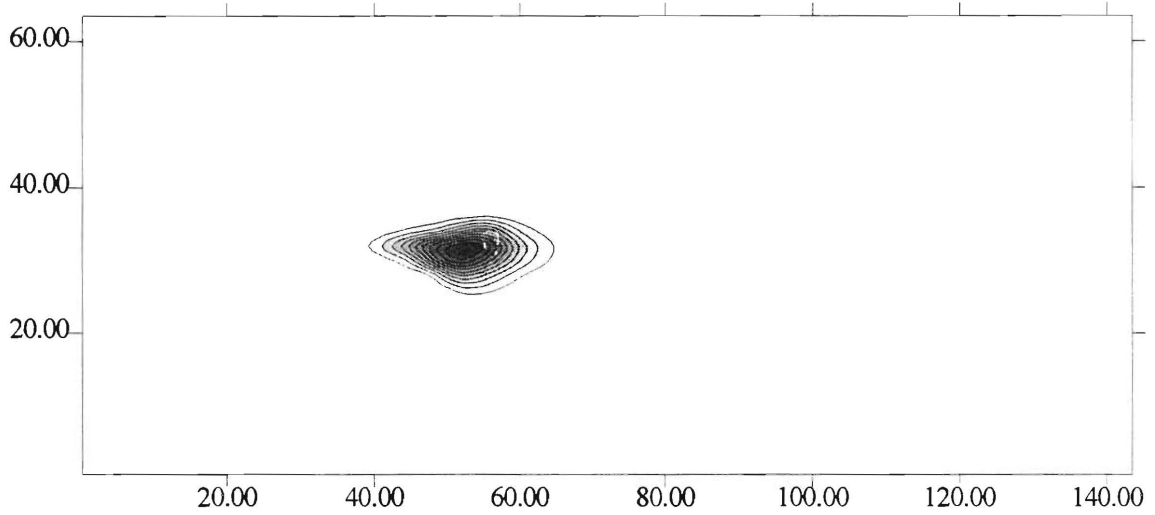
Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.0642	24.0328	31.9630	0.3320	0.3225	0.0767	-0.0909	2.4031	2.3072
0.0114	0.0633	24.1657	31.9919	0.4859	0.3361	0.2905	-0.0176	3.1421	2.4547
0.2614	0.0590	26.7015	32.4954	3.4793	0.7423	0.7817	0.0657	3.0504	3.5186
0.5113	0.0569	28.8003	33.1860	6.6254	1.0591	1.4323	0.0110	3.1080	3.6222
0.7613	0.0558	30.9093	33.8515	10.8983	1.3353	1.7305	-0.1973	2.8346	3.8380
1.0113	0.0550	33.1915	34.5789	16.9367	1.7775	1.8039	0.2503	2.7127	3.7862
1.2612	0.0539	35.6685	35.3815	24.1157	2.4707	1.5423	0.8638	2.4546	3.4792
1.5112	0.0531	38.3181	36.2822	32.6130	3.7642	0.9572	1.4284	2.2672	3.6567
1.7612	0.0524	41.1259	37.3615	41.3313	6.1489	0.0835	2.1884	2.1834	3.5761
2.0114	0.0518	43.9356	38.5305	48.1416	8.5658	-1.3105	1.8898	2.1817	2.6807

APPENDIX B

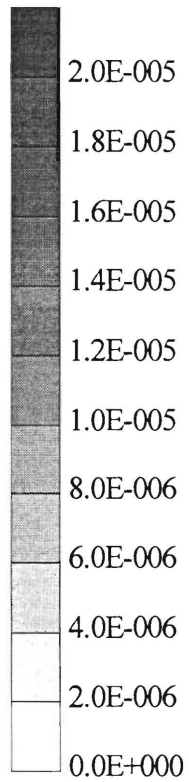
PLUME "SNAPSHOTS"

Weakly Heterogeneous Flow Field
Realization 1
Reactive Run
1 kg of Bromide

T = 2 Years

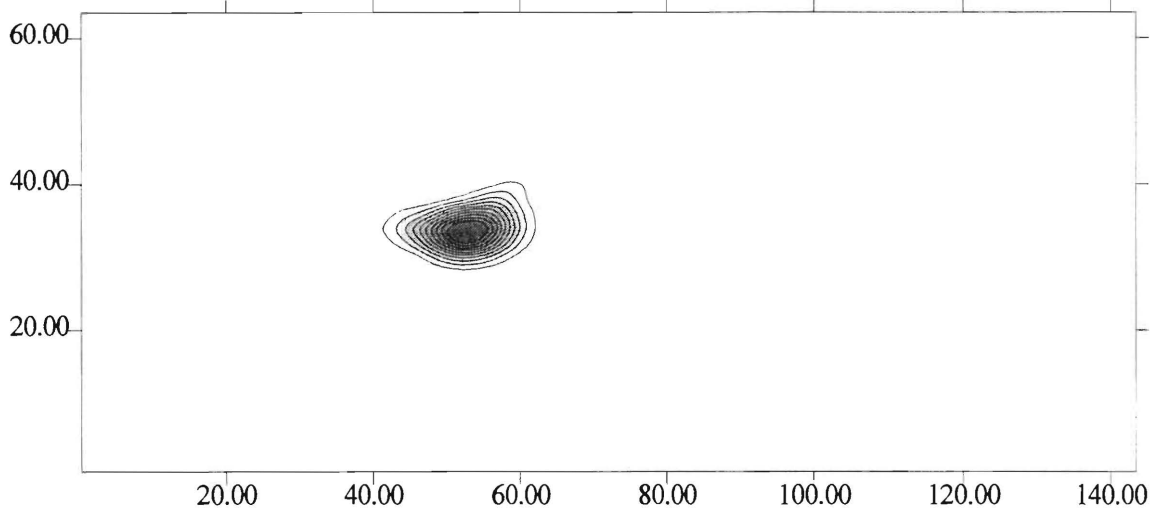


Bromide concentration scale
for realizations in a weakly
heterogeneous flow field with
1 kg of bromide.

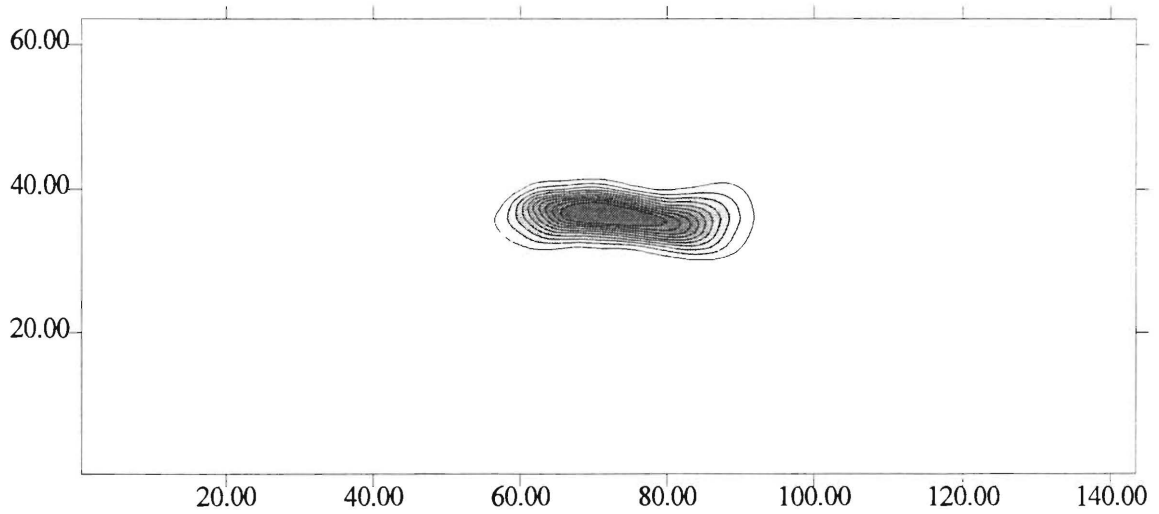


Weakly Heterogeneous Flow Field
Realization 2
Reactive Run
1 kg of Bromide

T = 2 Years



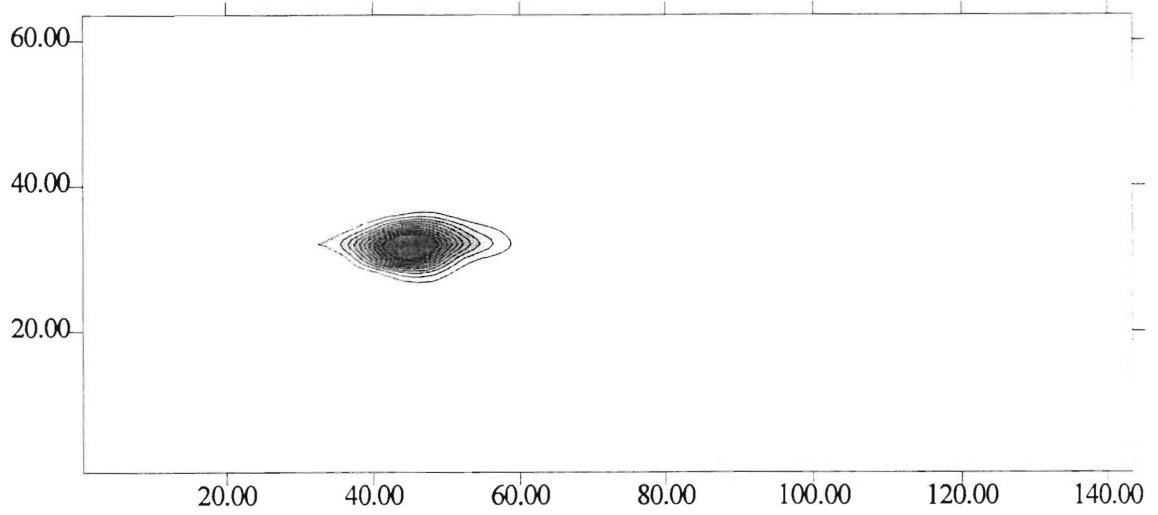
T = 4 Years *



* Contour interval for this plot is 1.0E-006 due to low concentration values

Weakly Heterogeneous Flow Field
Realization 3
Reactive Run
1 kg of Bromide

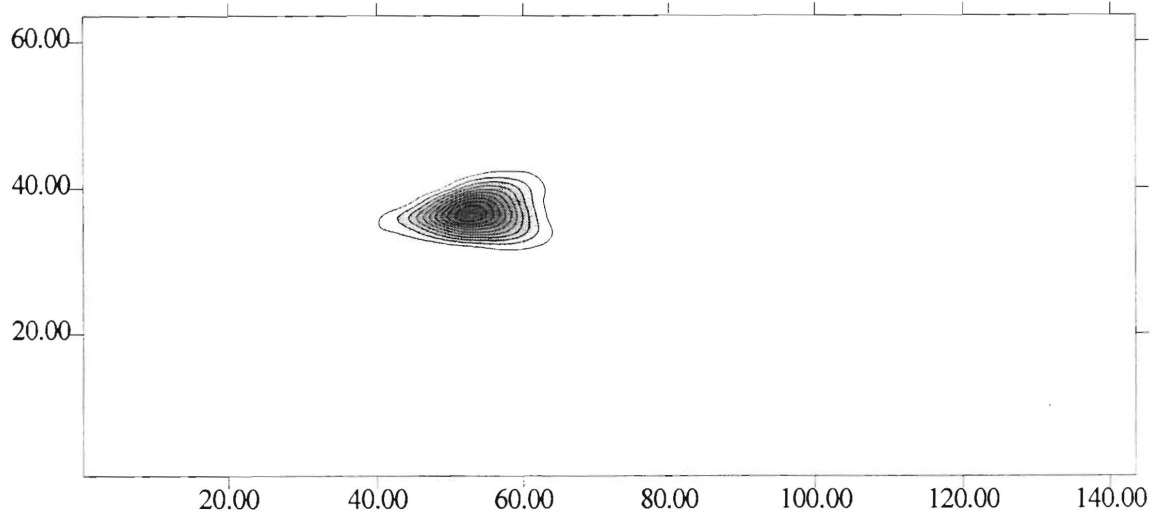
T = 2 Years



120

Weakly Heterogeneous Flow Field
Realization 4
Reactive Run
1 kg of Bromide

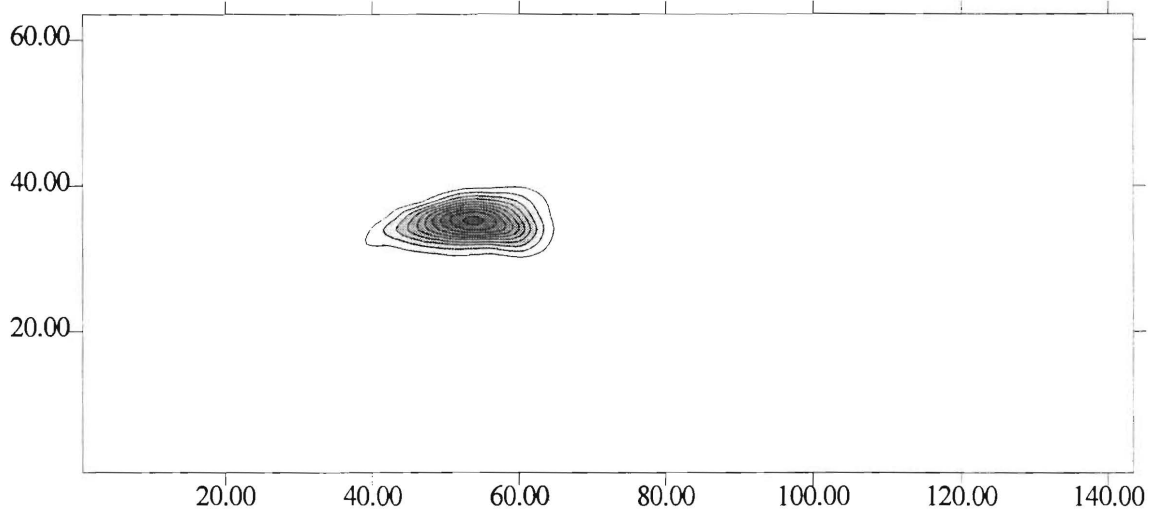
T = 2 Years



121

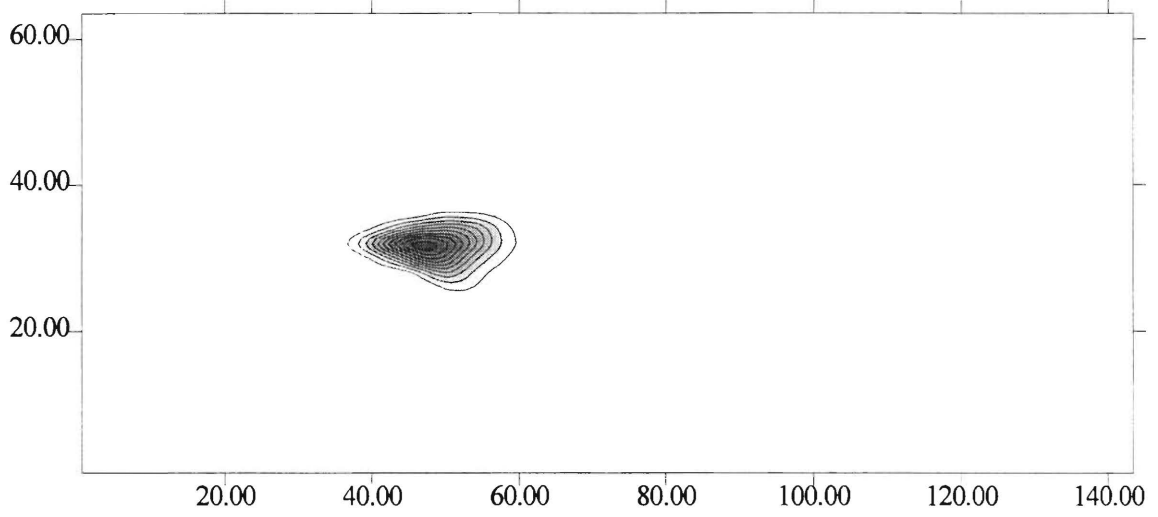
Weakly Heterogeneous Flow Field
Realization 5
Reactive Run
1 kg of Bromide

T = 2 Years

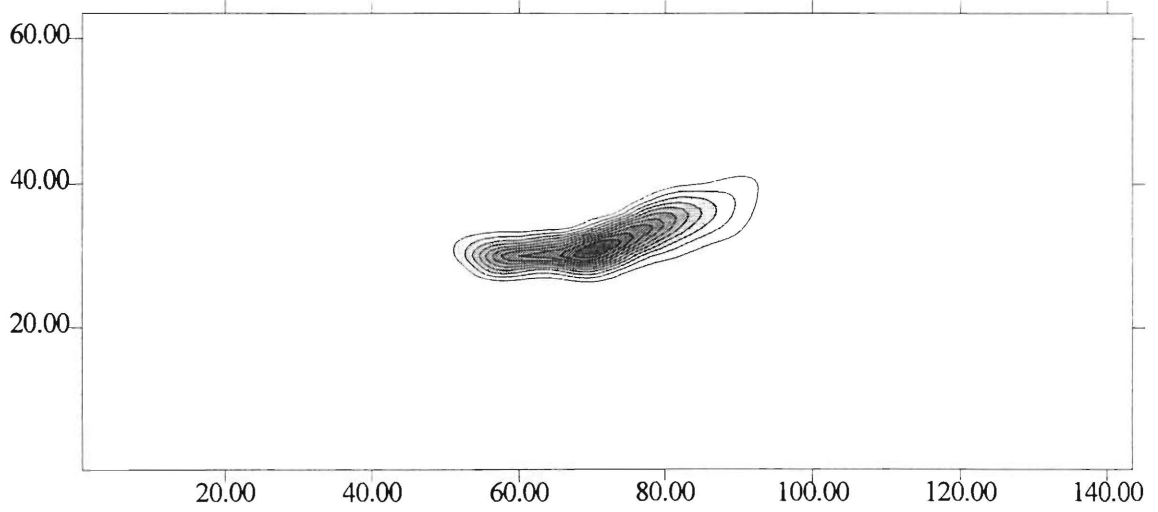


Weakly Heterogeneous Flow Field
Realization 1
Conservative Run
1 kg of Bromide

T = 1 Year



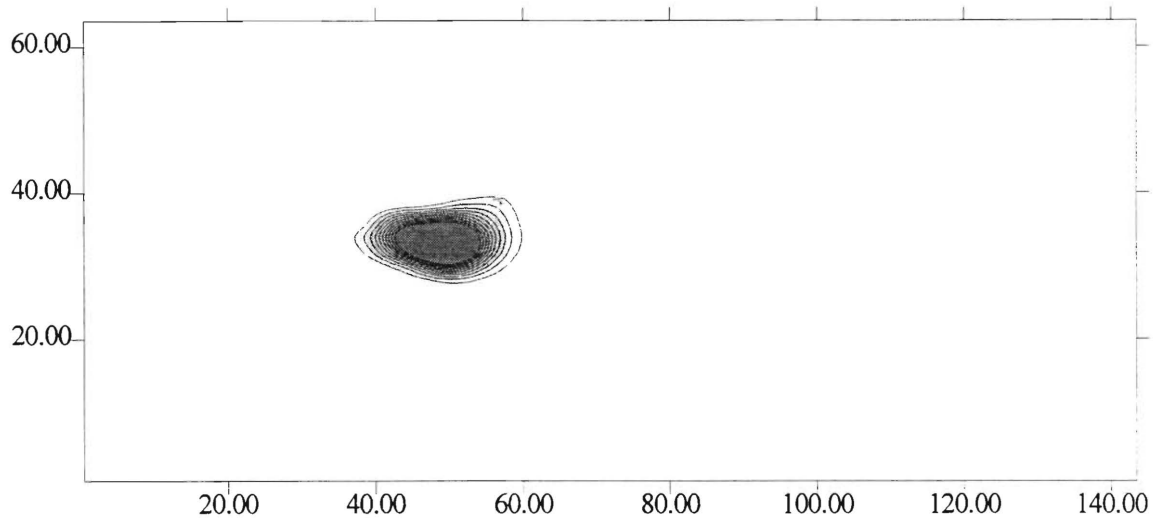
T = 2 Years



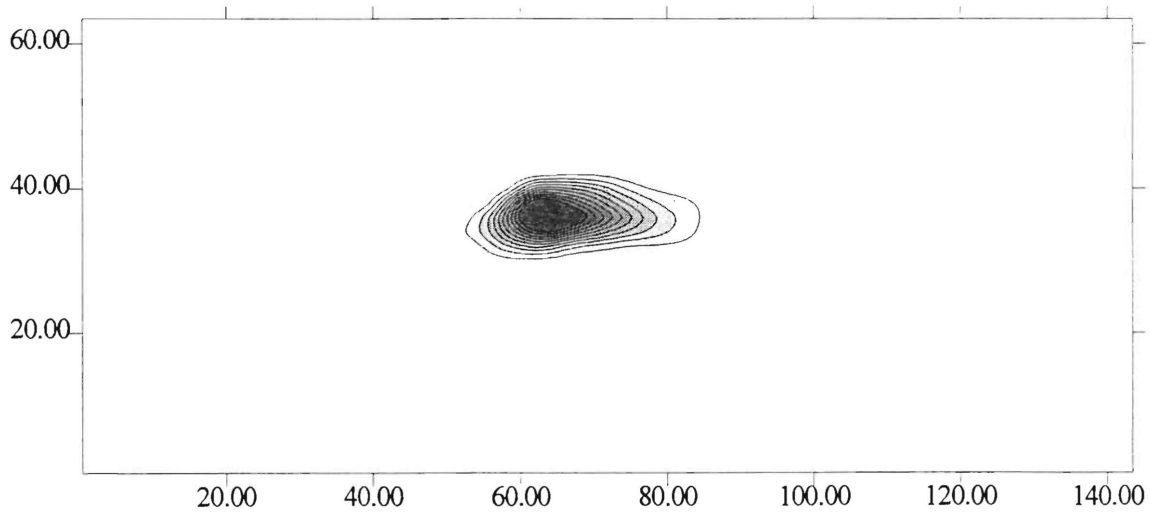
123

Weakly Heterogeneous Flow Field
Realization 2
Conservative Run
1 kg of Bromide

T = 1 Year

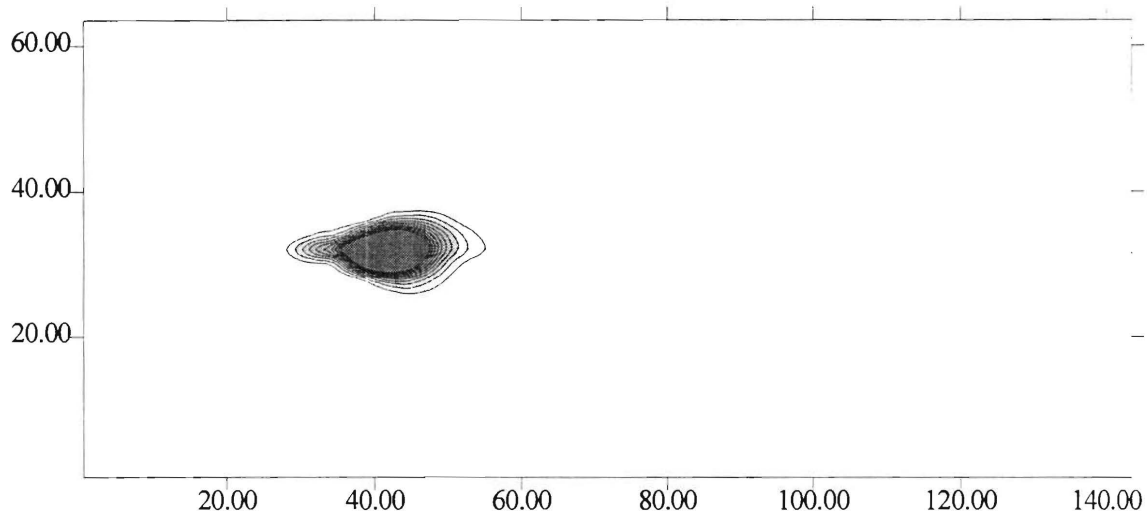


T = 2 Years

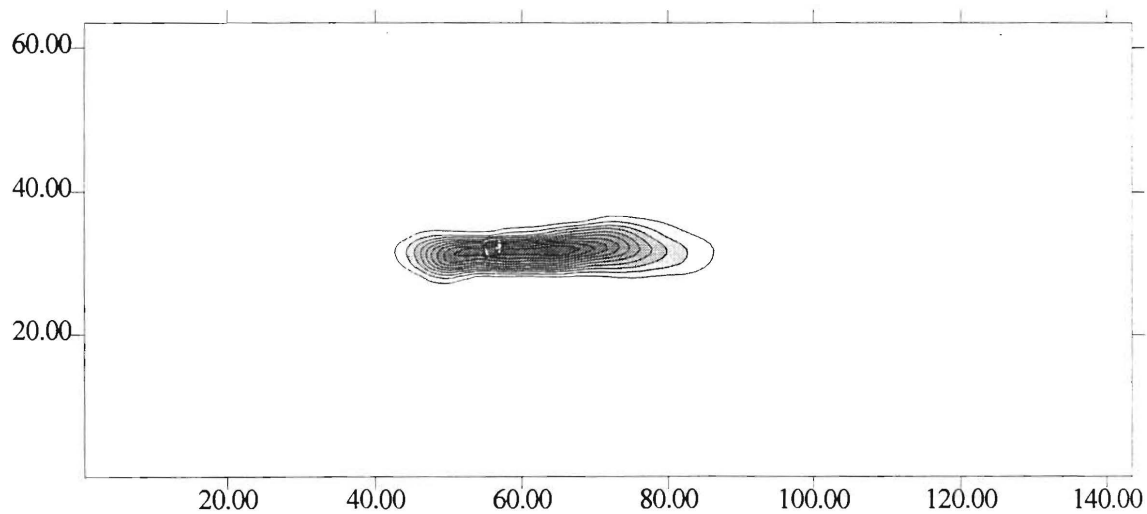


Weakly Heterogeneous Flow Field
Realization 3
Conservative Run
1 kg of Bromide

T = 1 Year



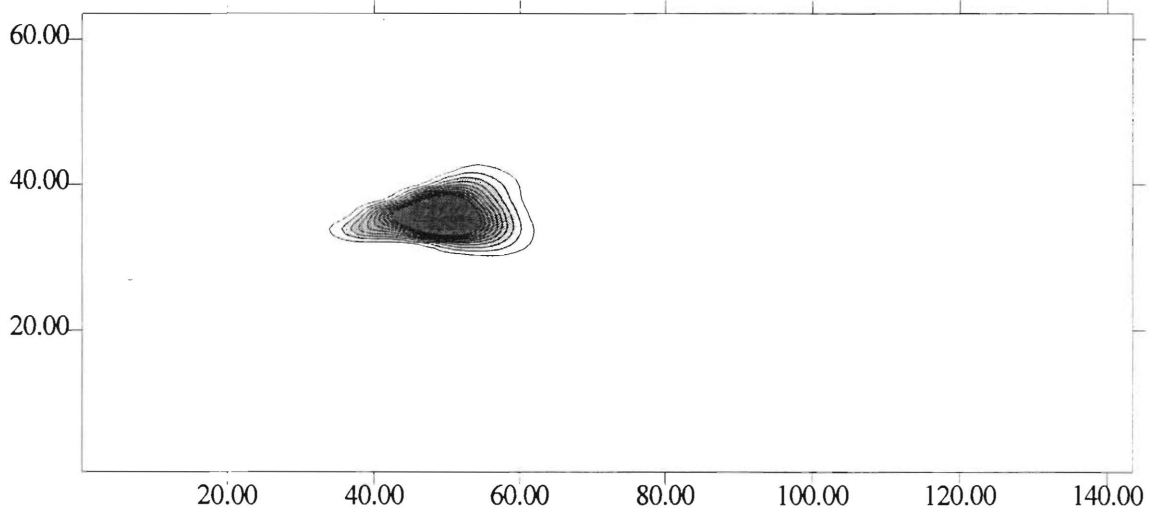
T = 2 Years



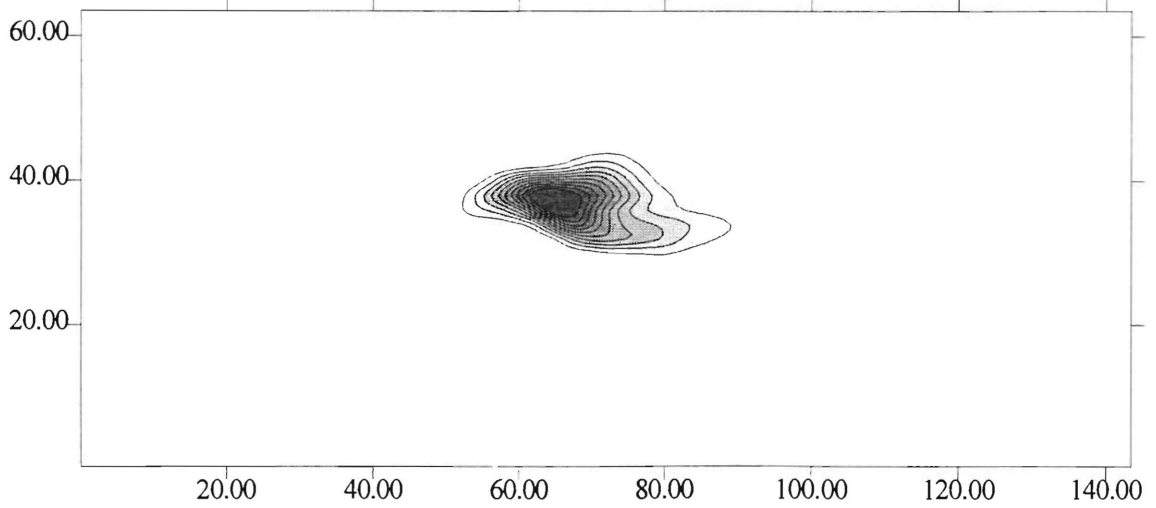
125

Weakly Heterogeneous Flow Field
Realization 4
Conservative Run
1 kg of Bromide

T = 1 Year

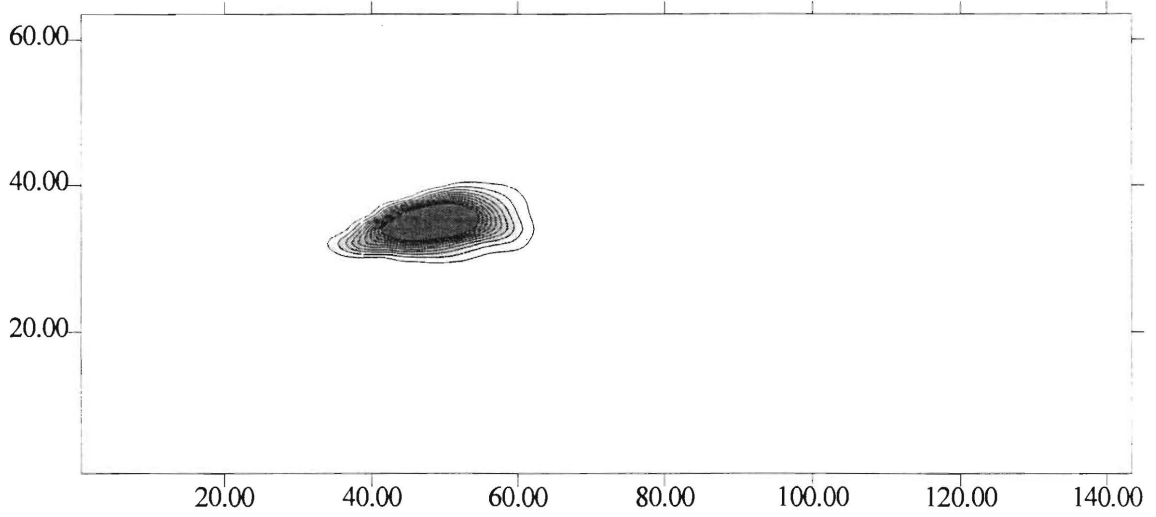


T = 2 Years

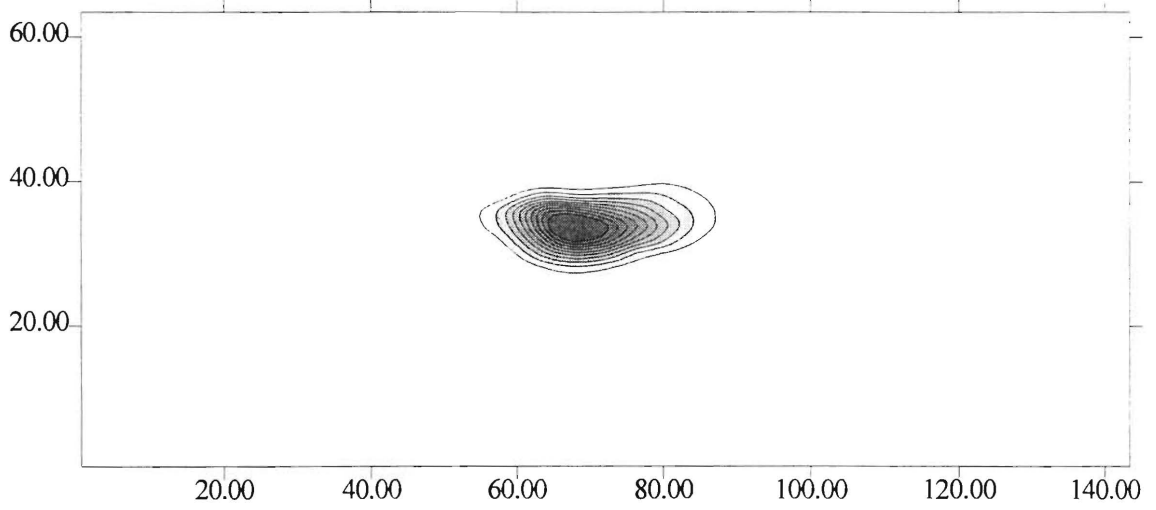


Weakly Heterogeneous Flow Field
Realization 5
Conservative Run
1 kg of Bromide

T = 1 Year

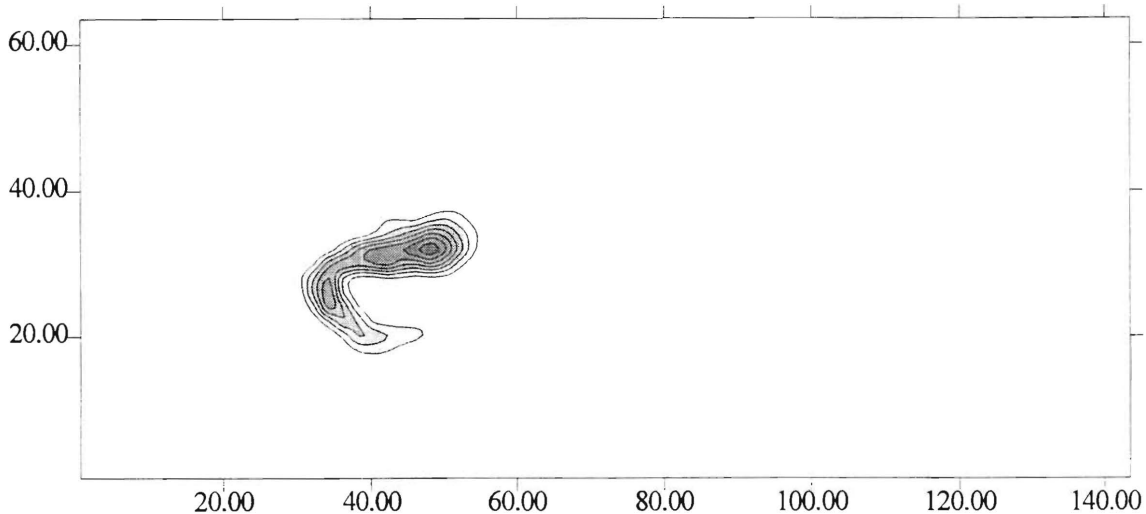


T = 2 Years

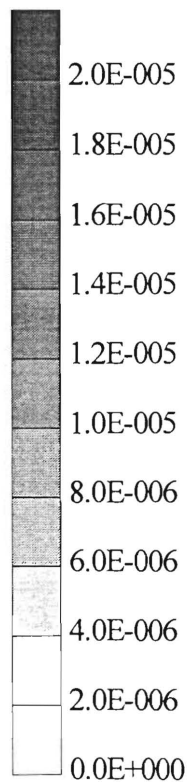


Strongly Heterogeneous Flow Field
Realization 1
Reactive Run
1 kg of Bromide

T = 2 Years

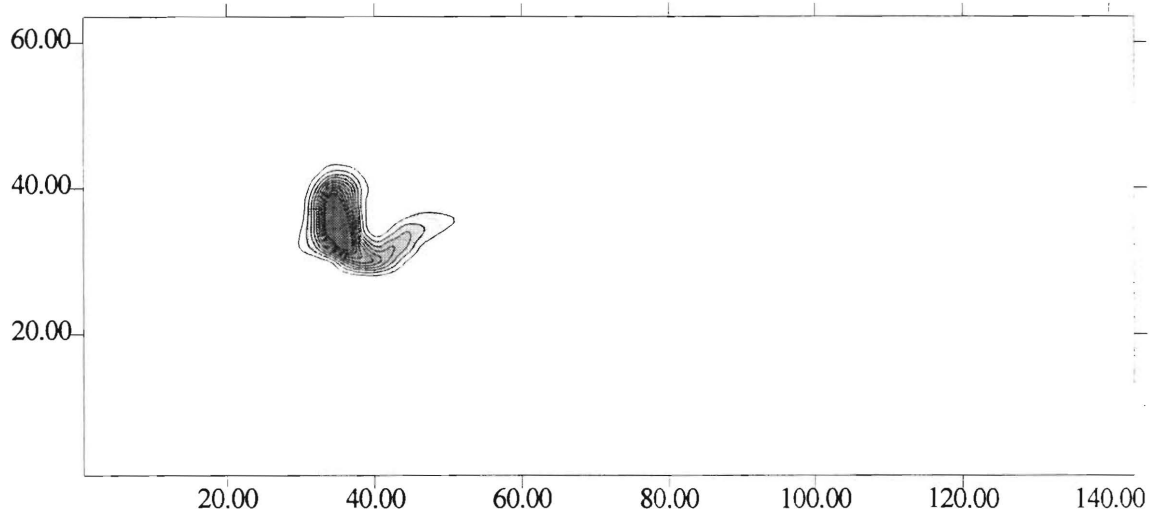


Bromide concentration scale
for realizations in a strongly
heterogeneous flow field with
1 kg of bromide.



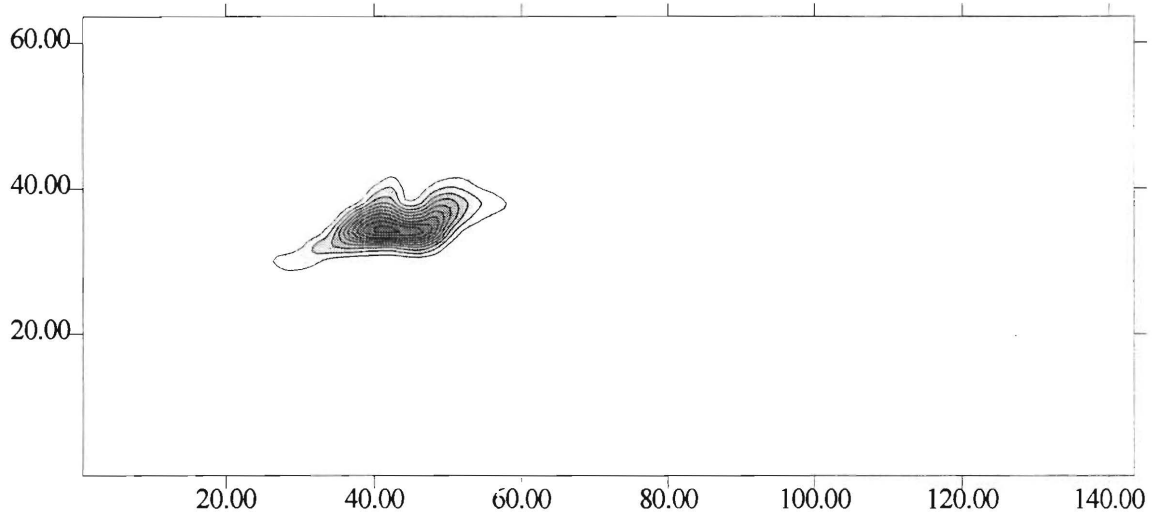
Strongly Heterogeneous Flow Field
Realization 2
Reactive Run
1 kg of Bromide

T = 2 Years



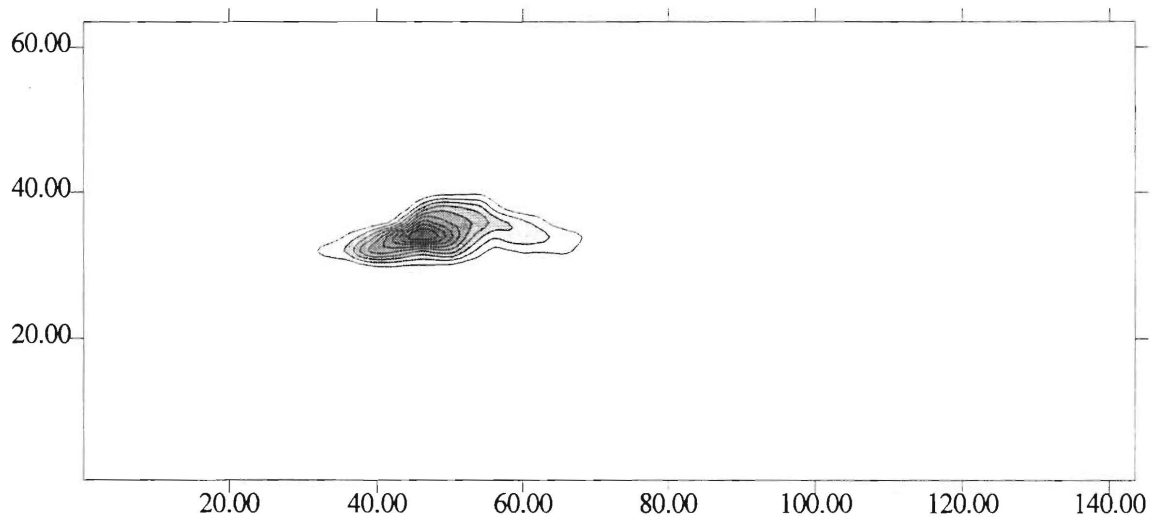
Strongly Heterogeneous Flow Field
Realization 3
Reactive Run
1 kg of Bromide

T = 2 Years

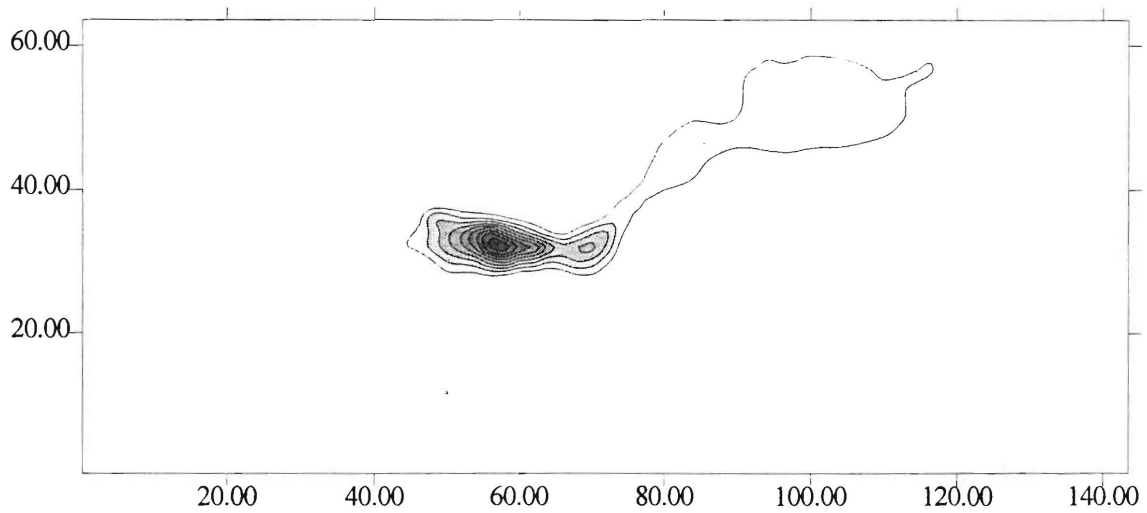


Strongly Heterogeneous Flow Field
Realization 4
Reactive Run
1 kg of Bromide

T = 2 Years



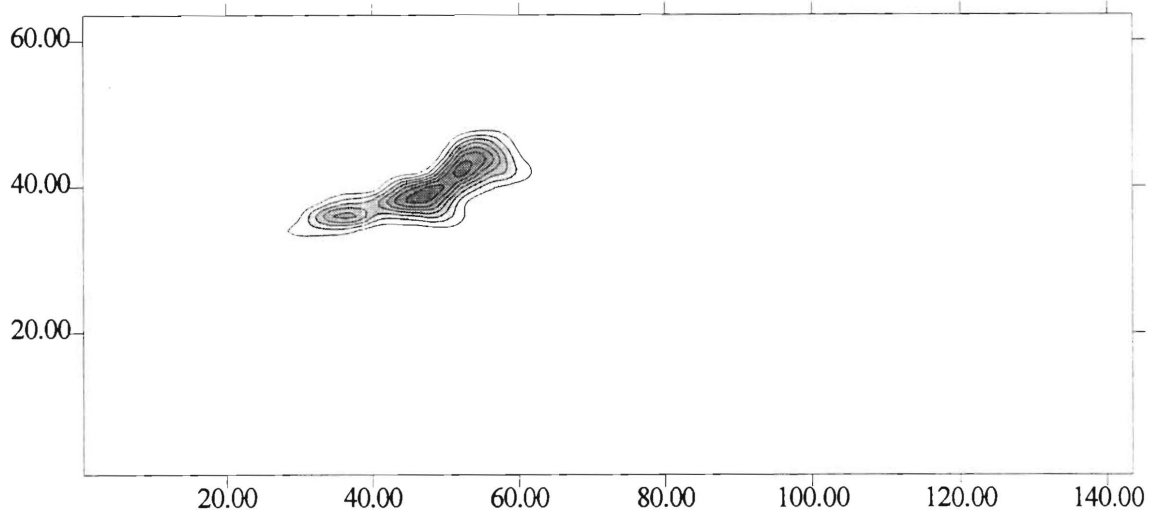
T = 4 Years *



* Concentration interval for this plot is 1.0E-006
due to low bromide concentrations

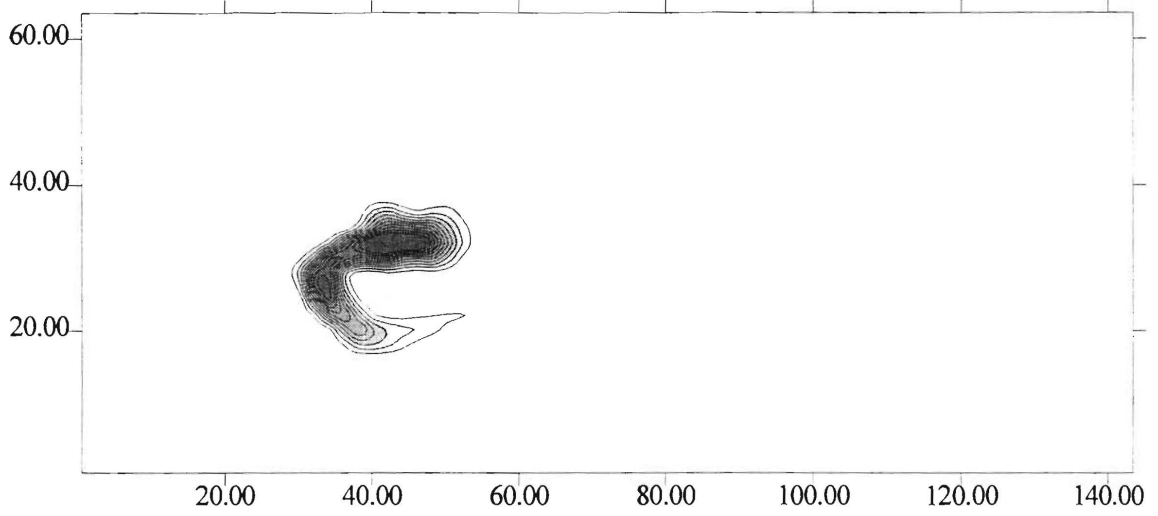
Strongly Heterogeneous Flow Field
Realization 5
Reactive Run
1 kg of Bromide

T = 1 Year

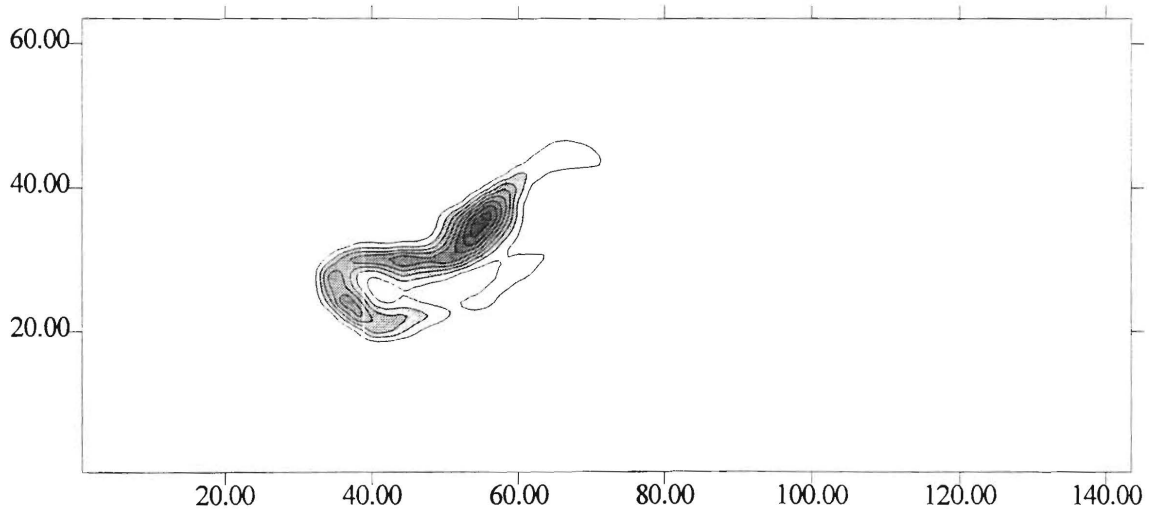


Strongly Heterogeneous Flow Field
Realization 1
Conservative Run
1 kg of Bromide

T = 1 Year

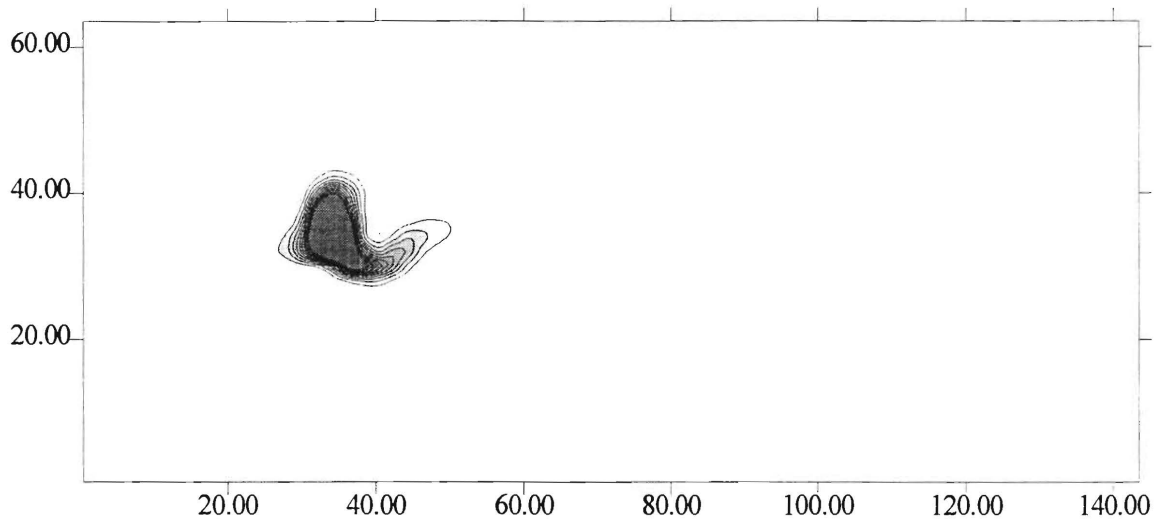


T = 2 Years

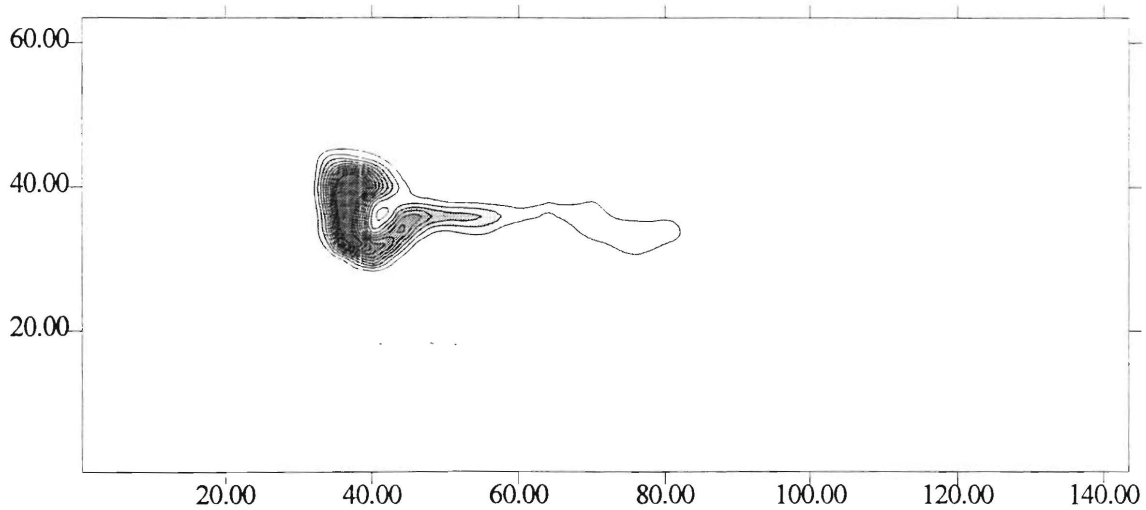


Strongly Heterogeneous Flow Field
Realization 2
Conservative Run
1 kg of Bromide

T = 1 Year

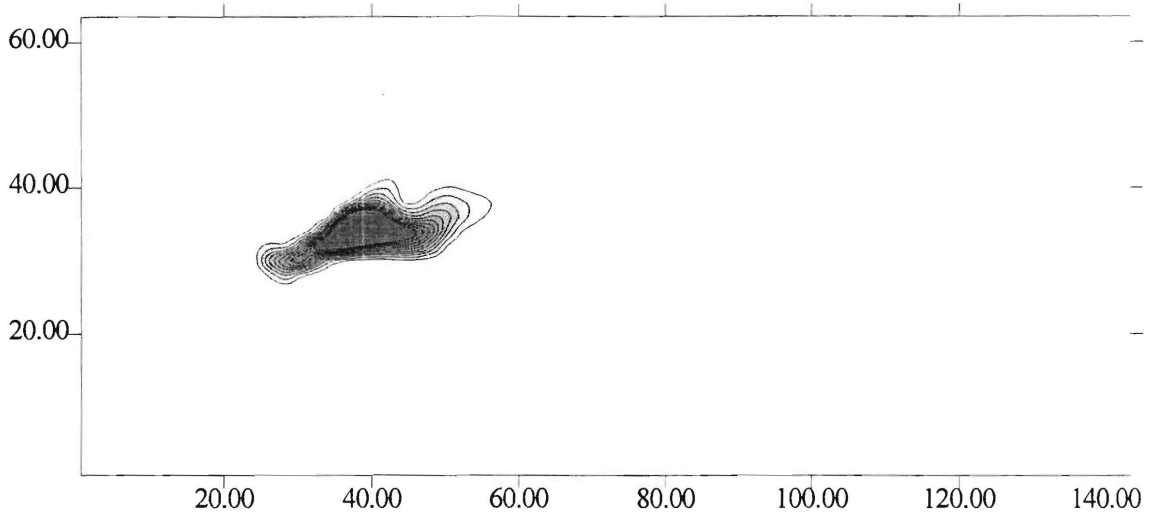


Time = 2 Years

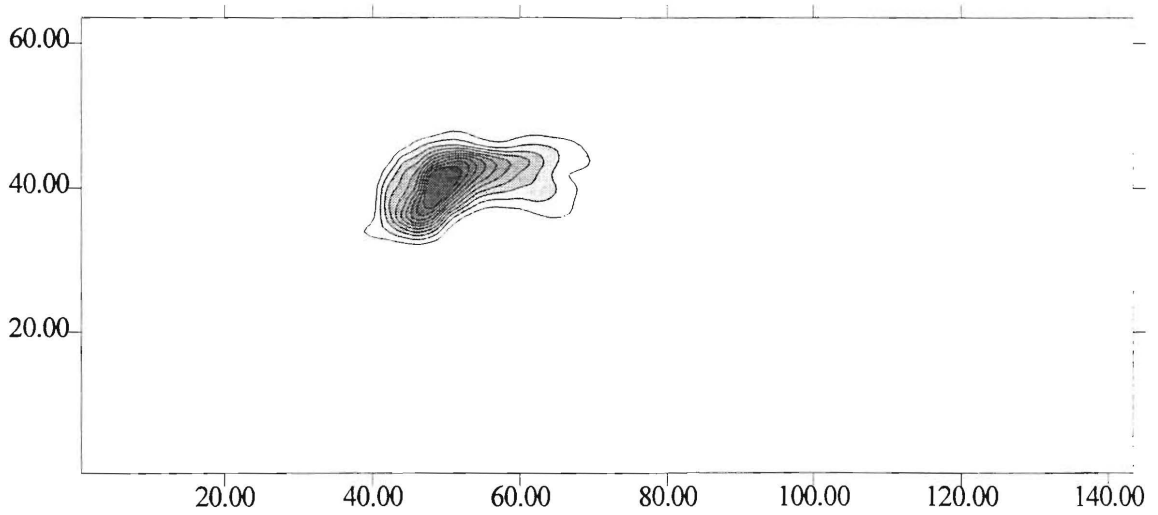


Strongly Heterogeneous Flow Field
Realization 3
Conservative Run
1 kg of Bromide

T = 1 Year

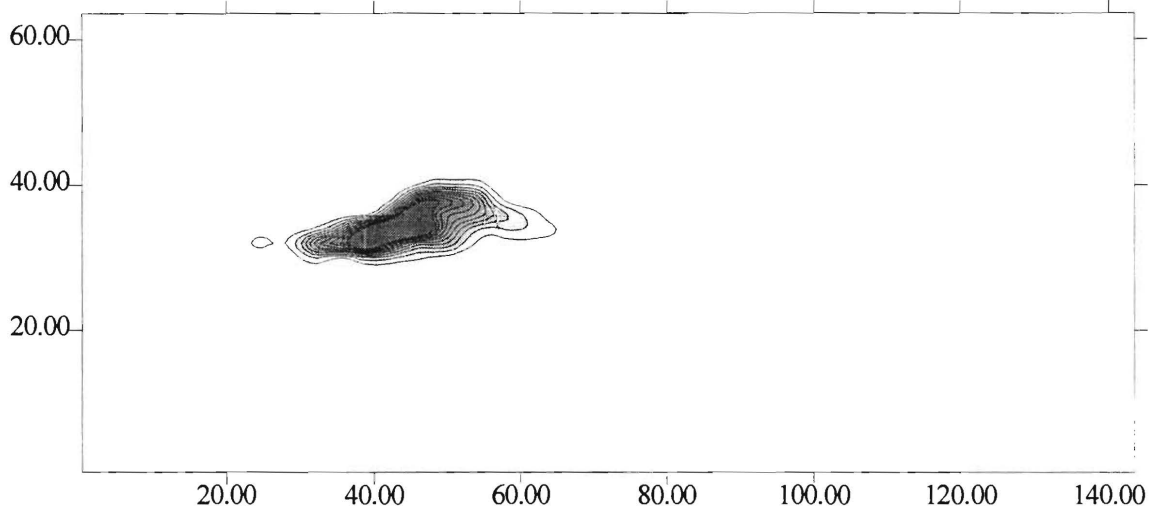


T = 2 Years

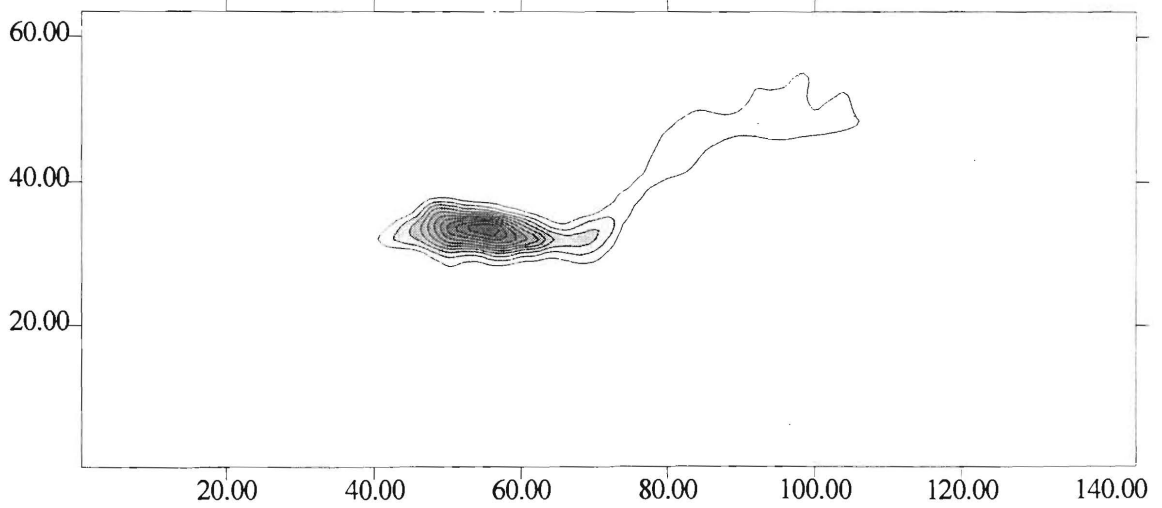


Strongly Heterogeneous Flow Field
Realization 4
Conservative Run
1 kg of Bromide

T = 1 Year

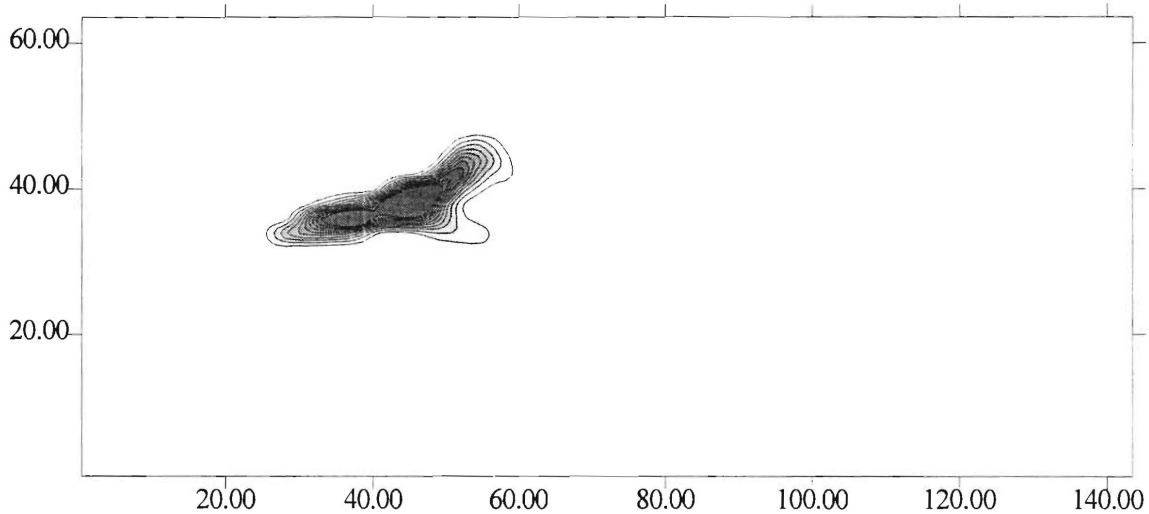


T = 2 Years

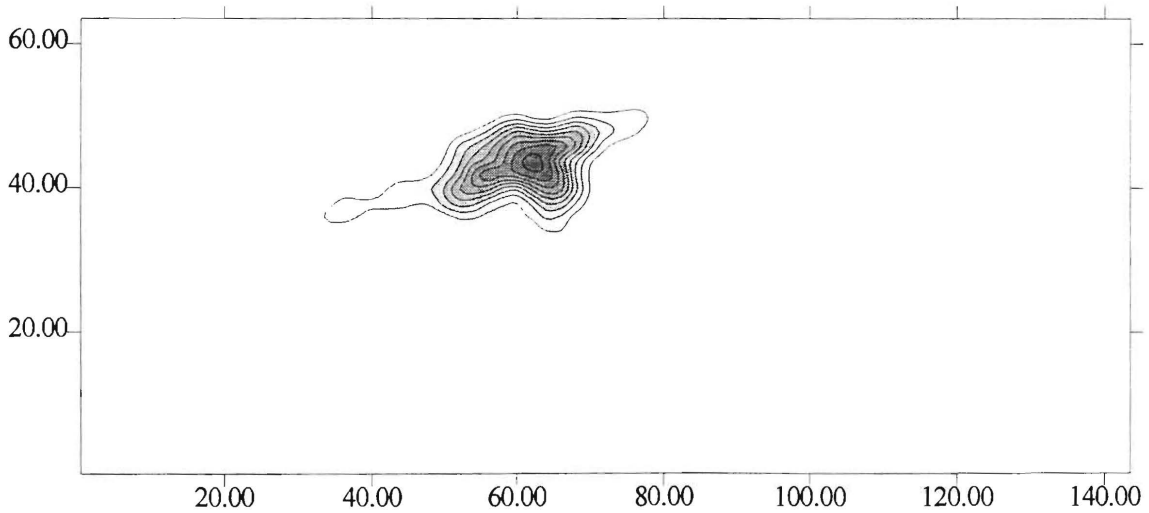


Strongly Heterogeneous Flow Field
Realization 5
Conservative Run
1 kg of Bromide

T = 1 Year

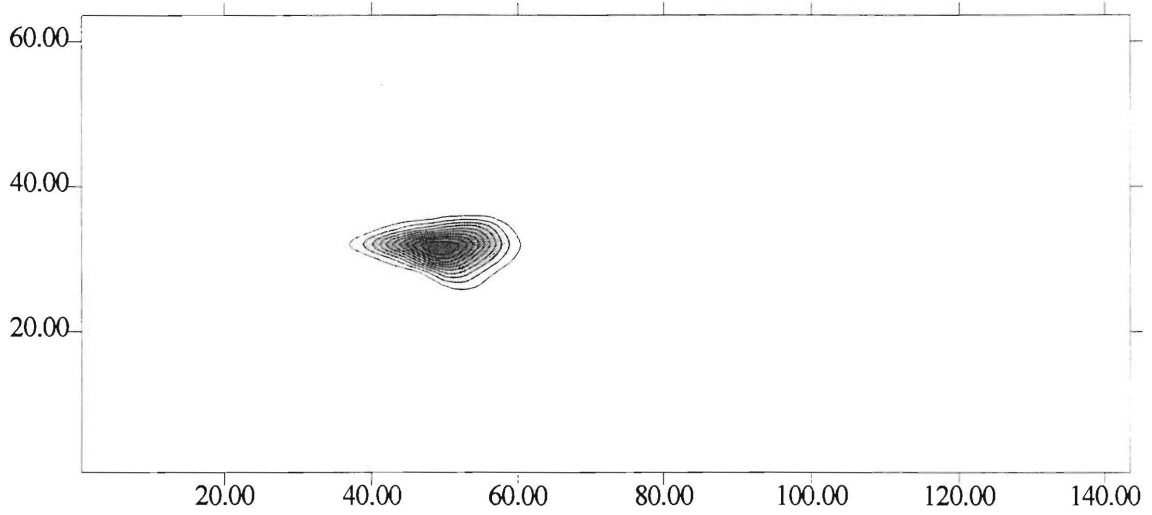


T = 2 Years

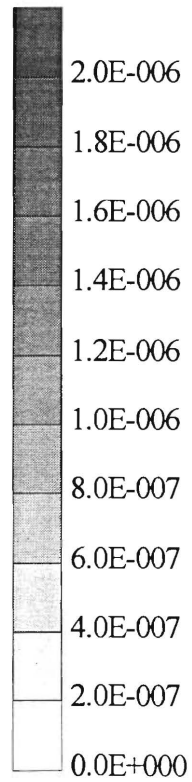


Weakly Heterogeneous Flow Field
Realization 1
Reactive Run
0.1 kg of Bromide

T = 2 Years

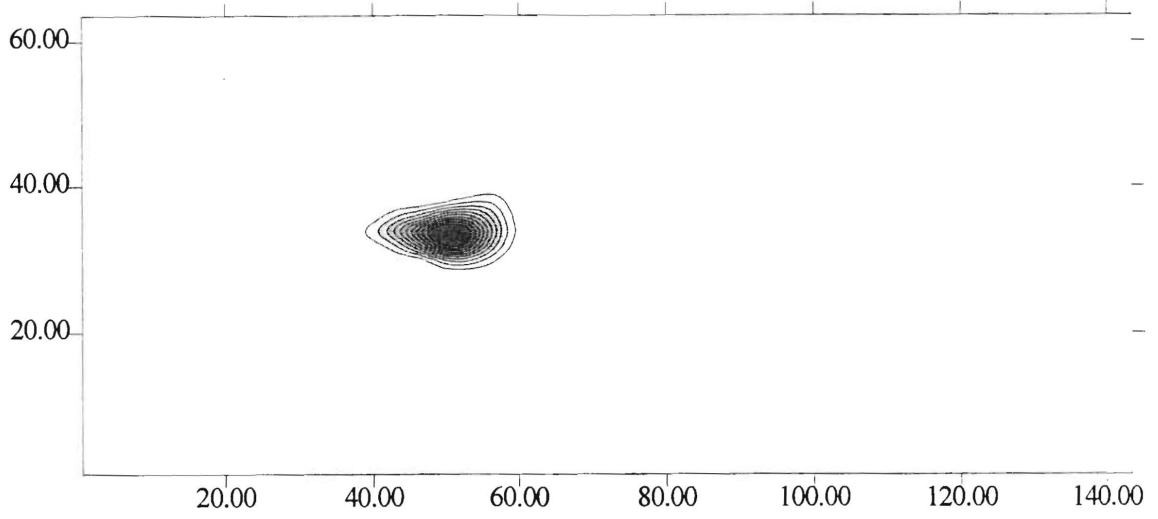


Bromide concentration scale
for realizations in a weakly
heterogeneous flow field with
0.1 kg of bromide.



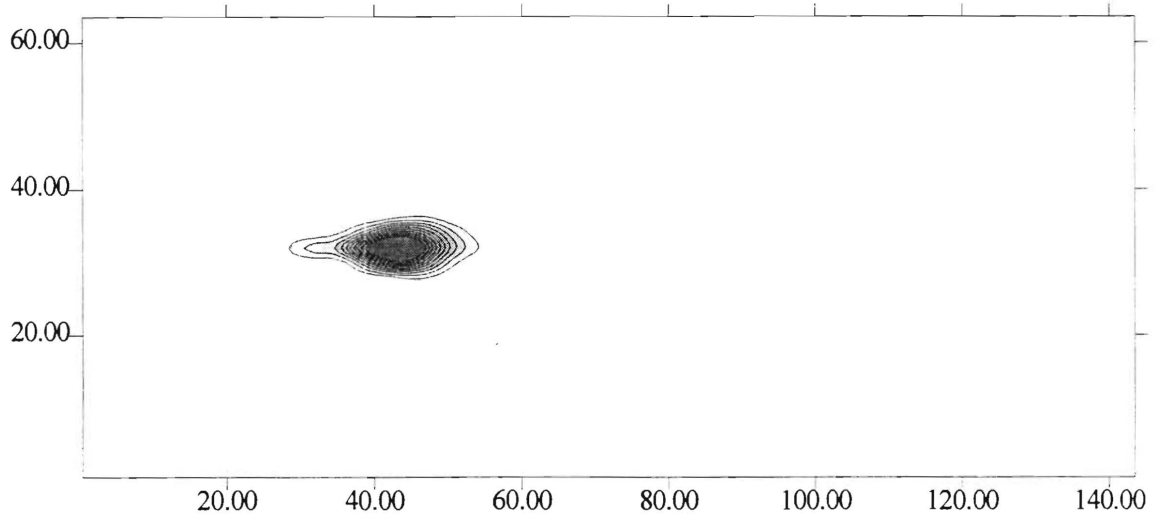
Weakly Heterogeneous Flow Field
Realization 2
Reactive Run
0.1 kg of Bromide

T = 2 Years



Weakly Heterogeneous Flow Field
Realization 3
Reactive Run
0.1 kg of Bromide

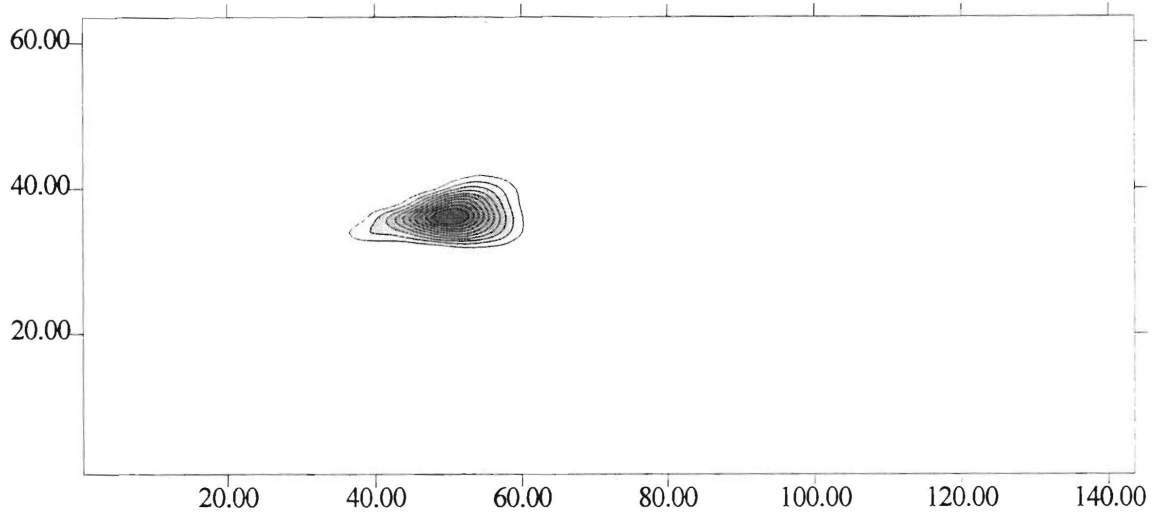
T = 2 Years



140

Weakly Heterogeneous Flow Field
Realization 4
Reactive Run
0.1 kg of Bromide

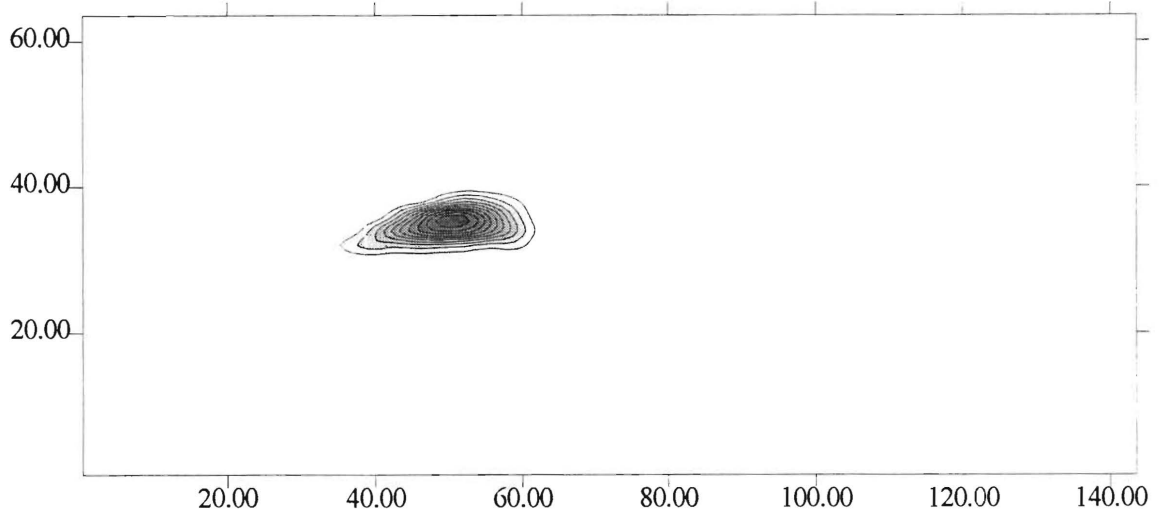
T = 2 Years



141

Weakly Heterogeneous Flow Field
Realization 5
Reactive Run
0.1 kg of Bromide

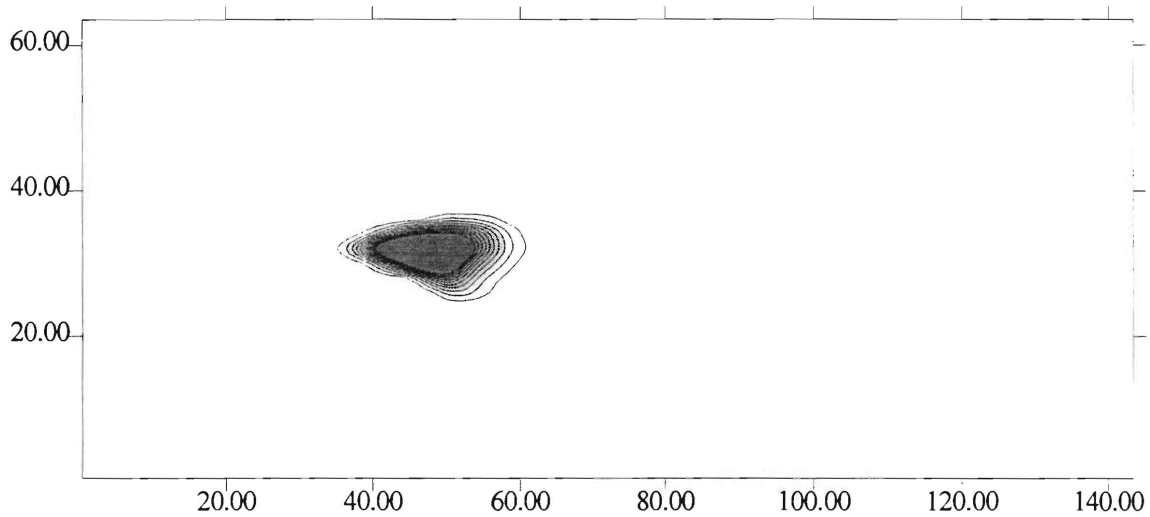
T = 2 Years



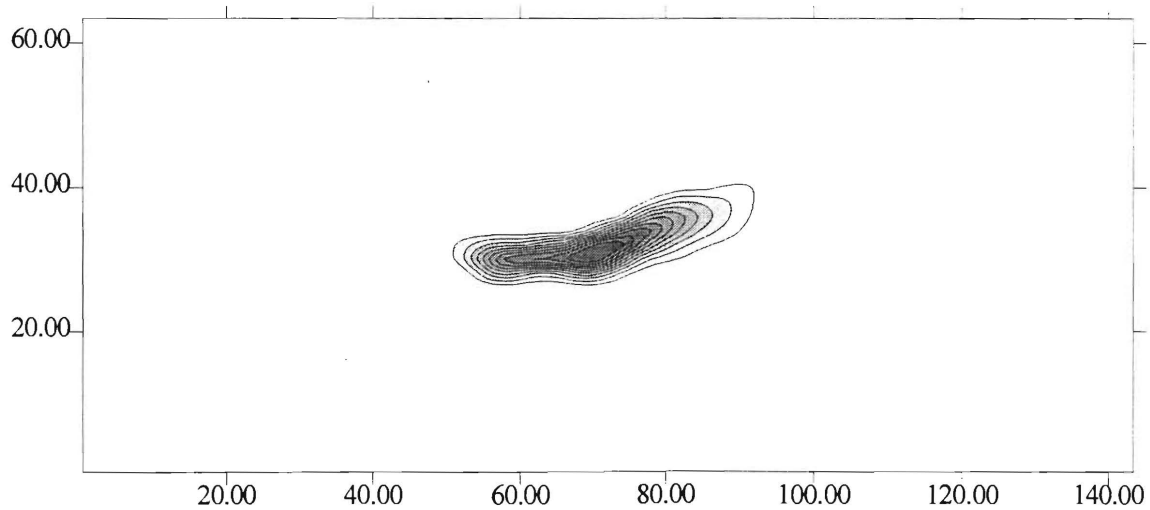
142

Weakly Heterogeneous Flow Field
Realization 1
Conservative Run
0.1 kg of Bromide

T = 1 Year

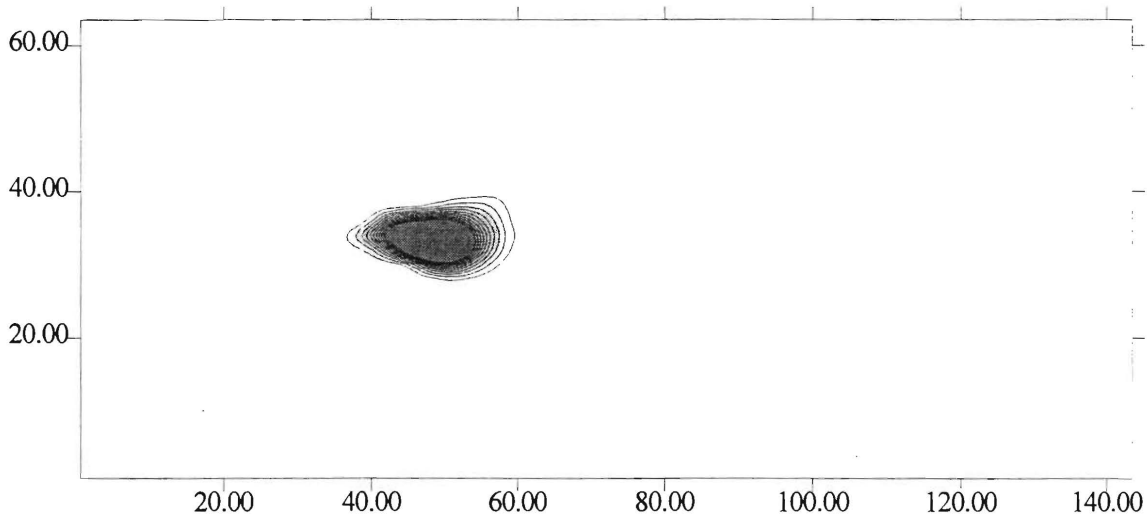


T = 2 Years

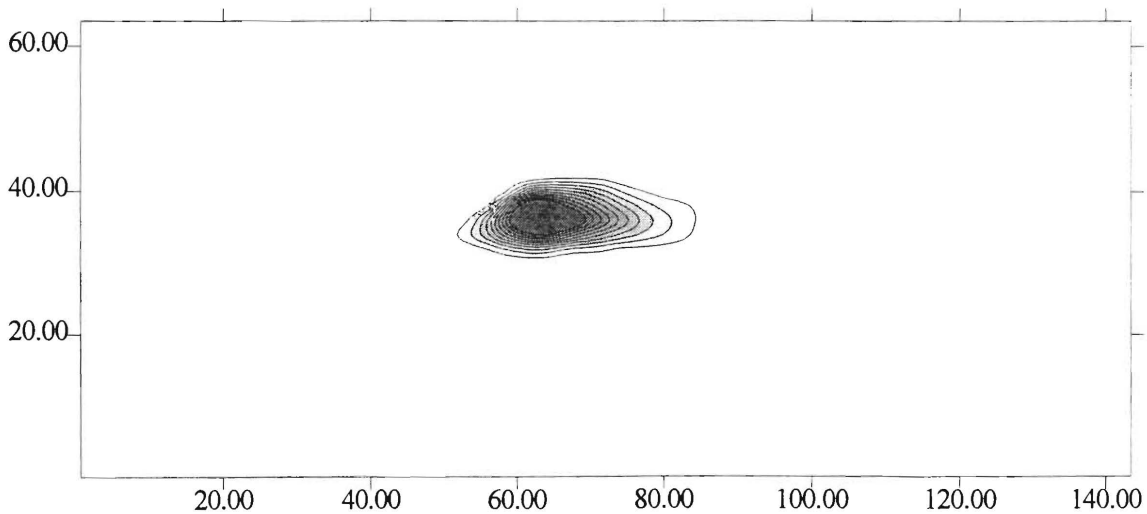


Weakly Heterogeneous Flow Field
Realization 2
Conservative Run
0.1 kg of Bromide

T = 1 Years

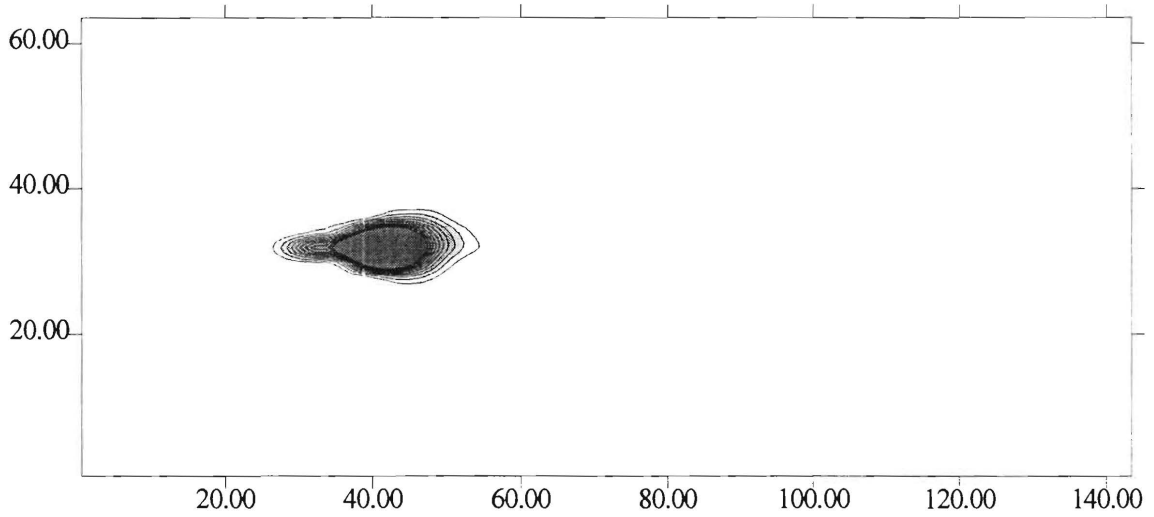


T = 2 Years

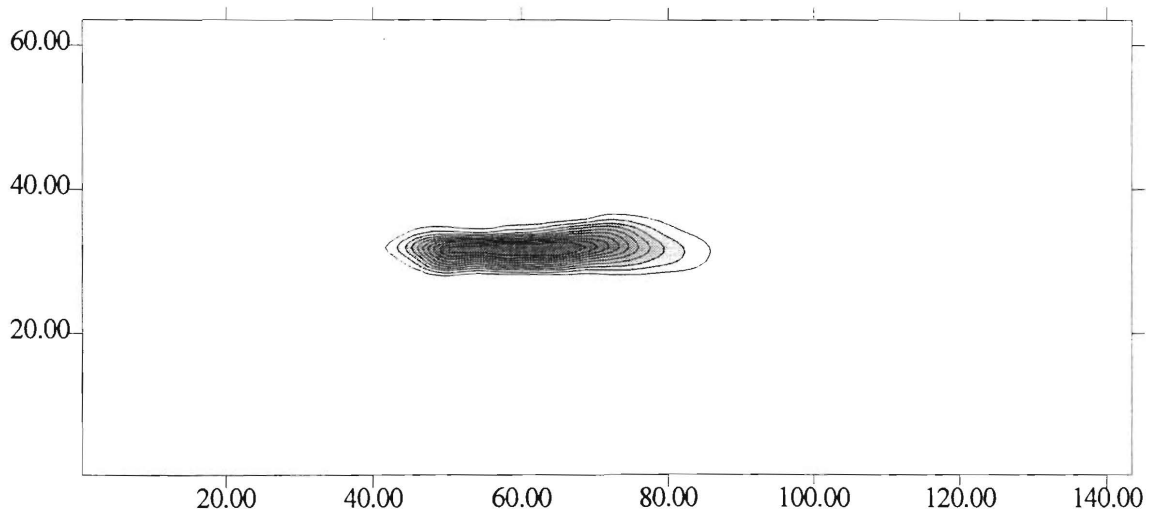


Weakly Heterogeneous Flow Field
Realization 3
Conservative Run
0.1 kg of Bromide

T = 1 Year

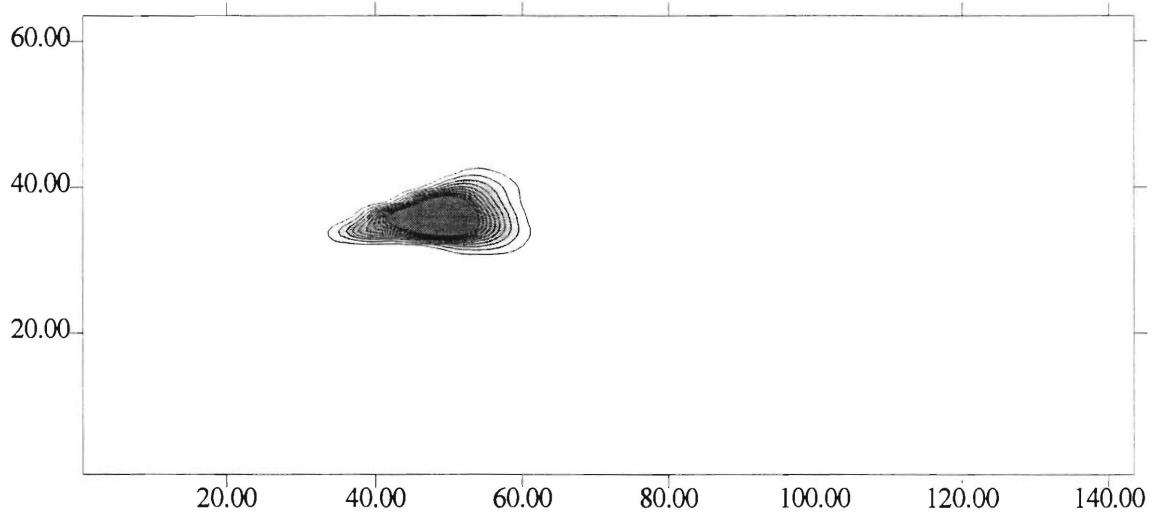


T = 2 Years

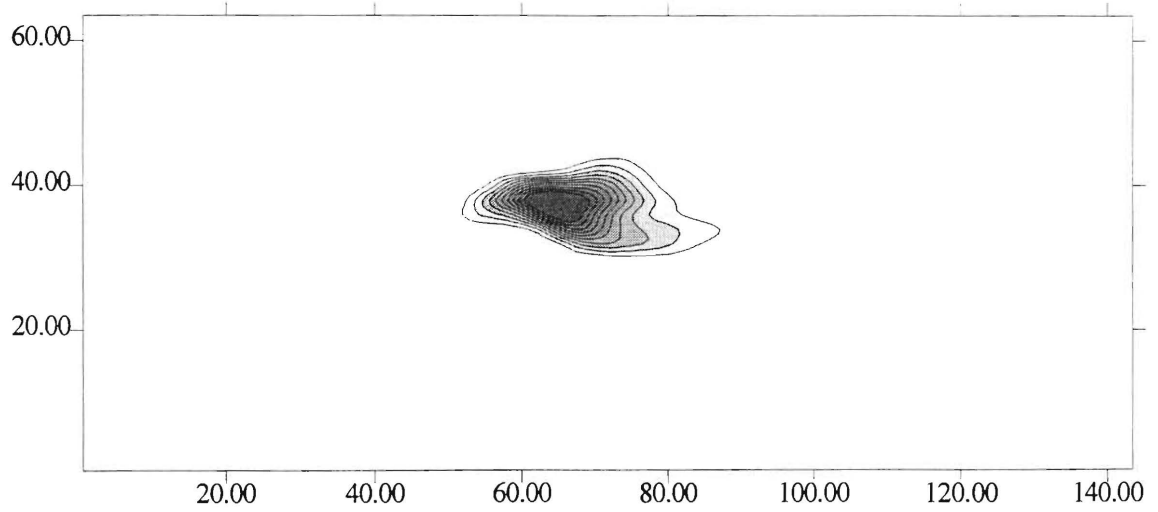


Weakly Heterogeneous Flow Field
Realization 4
Conservative Run
0.1 kg of Bromide

T = 1 Year



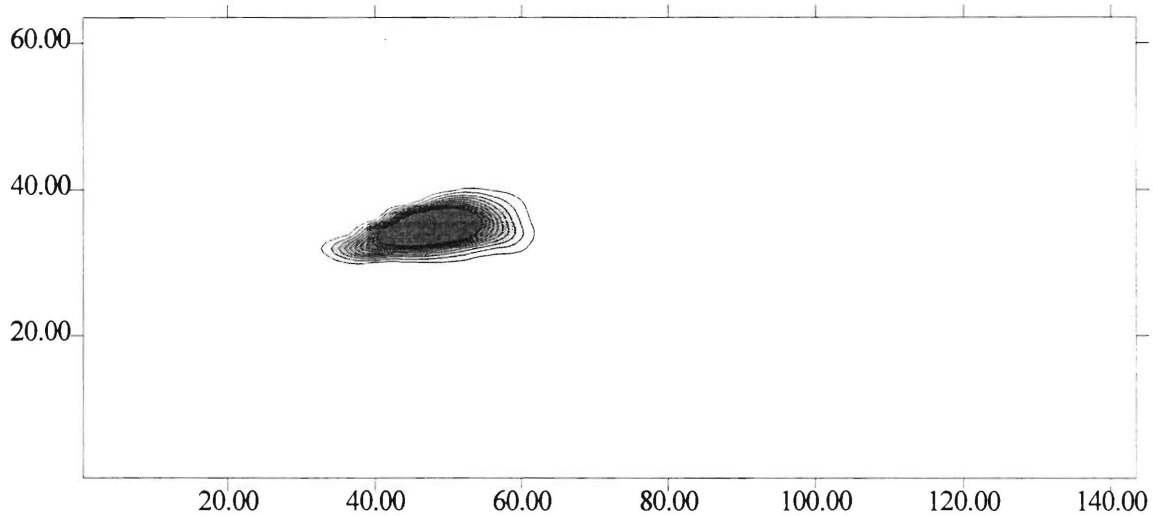
T = 2 Years



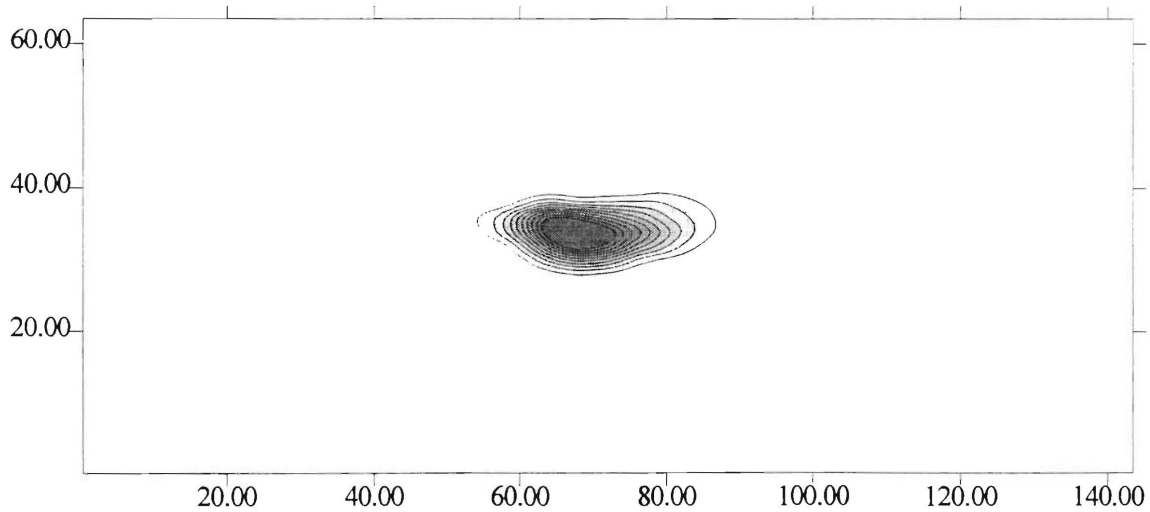
146

Weakly Heterogeneous Flow Field
Realization 5
Conservative Run
0.1 kg of Bromide

T = 1 Year

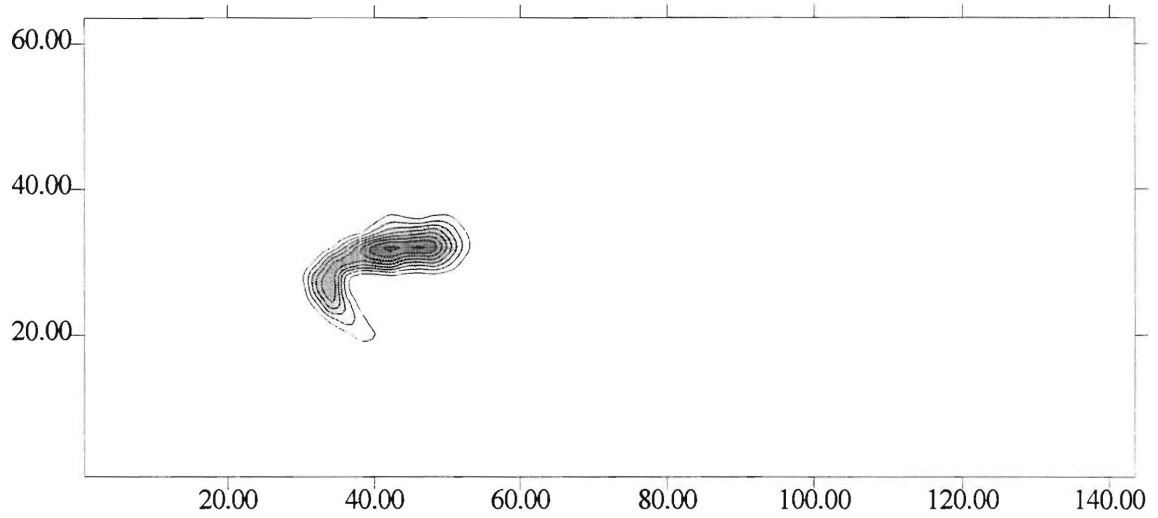


T = 2 Years

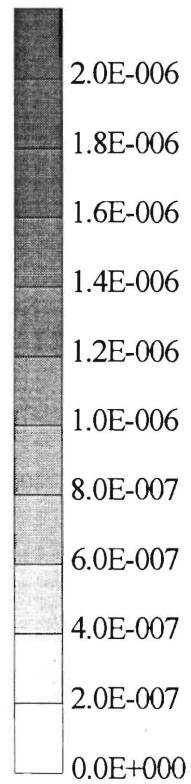


Strongly Heterogeneous Flow Field
Realization 1
Reactive Run
0.1 kg of Bromide

T = 2 Years



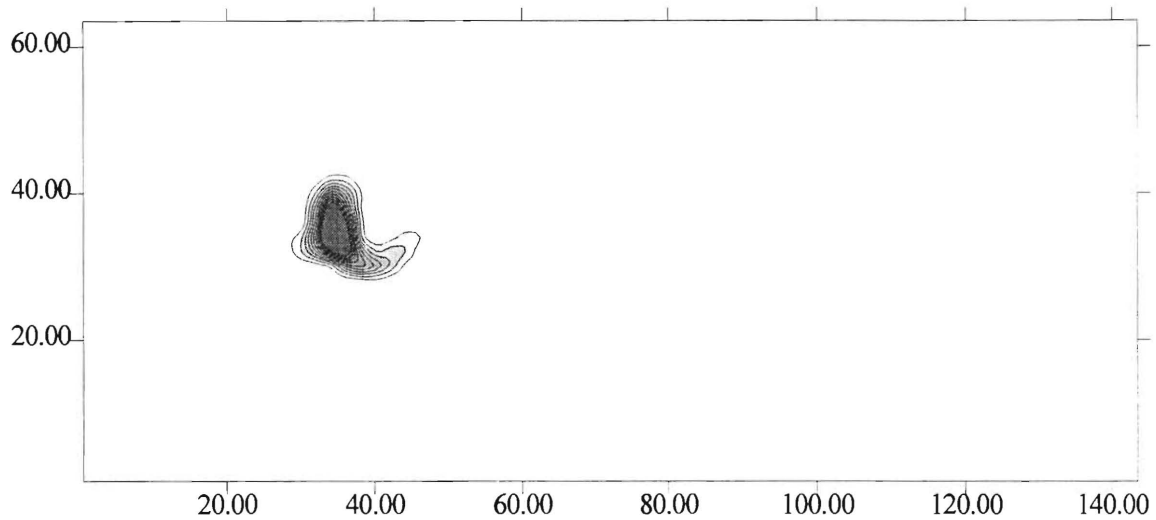
Bromide concentration scale
for realizations in a strongly
heterogeneous flow field with
0.1 kg of bromide.



148

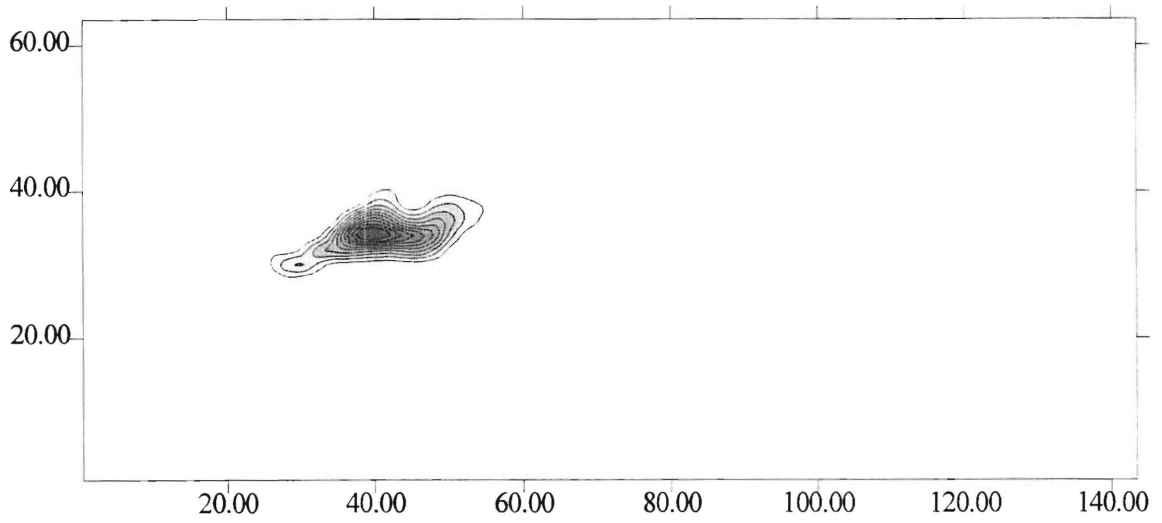
Strongly Heterogeneous Flow Field
Realization 2
Reactive Run
0.1 kg of Bromide

T = 2 Years



Strongly Heterogeneous Flow Field
Realization 3
Reactive Run
0.1 kg of Bromide

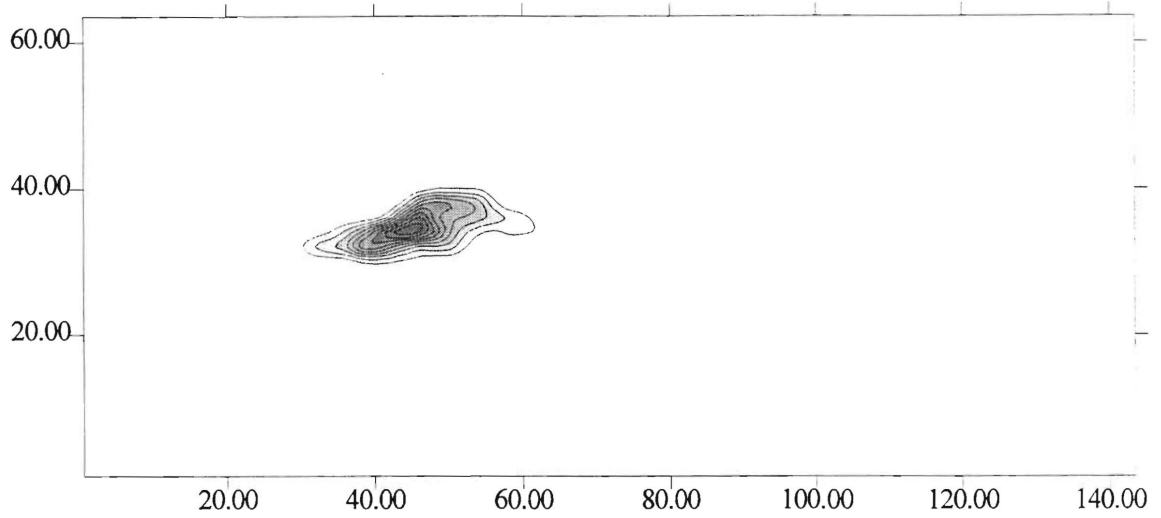
T = 2 Years



150

Strongly Heterogeneous Flow Field
Realization 4
Reactive Run
0.1 kg of Bromide

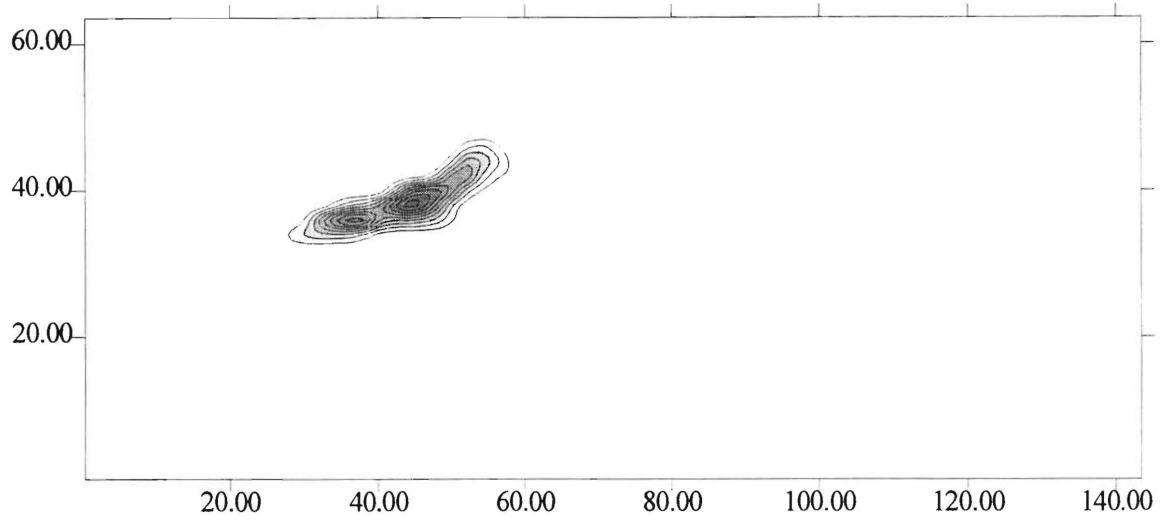
T = 2 Years



151

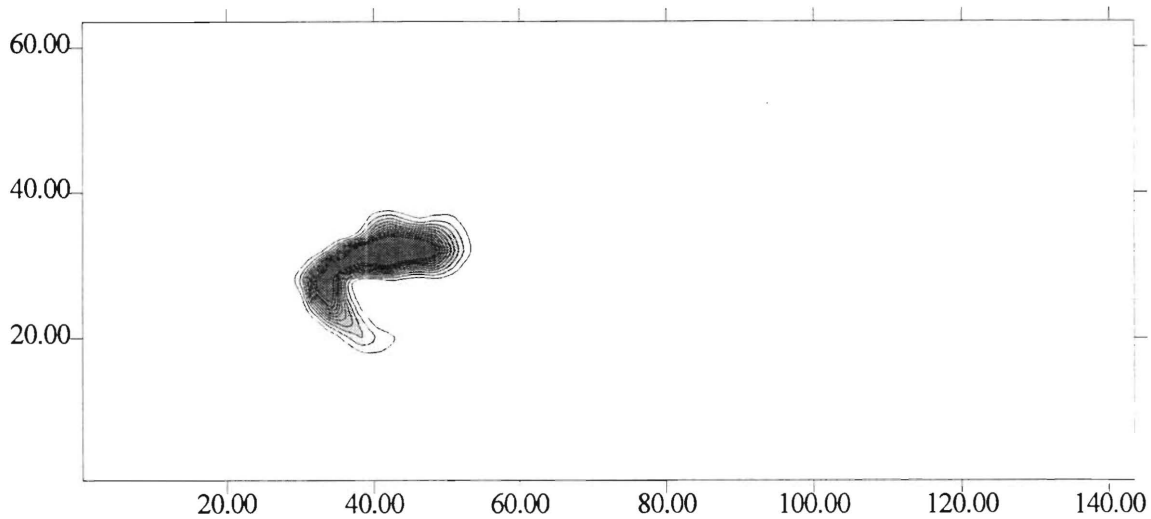
Strongly Heterogeneous Flow Field
Realization 5
Reactive Run
0.1 kg of Bromide

T = 2 Years

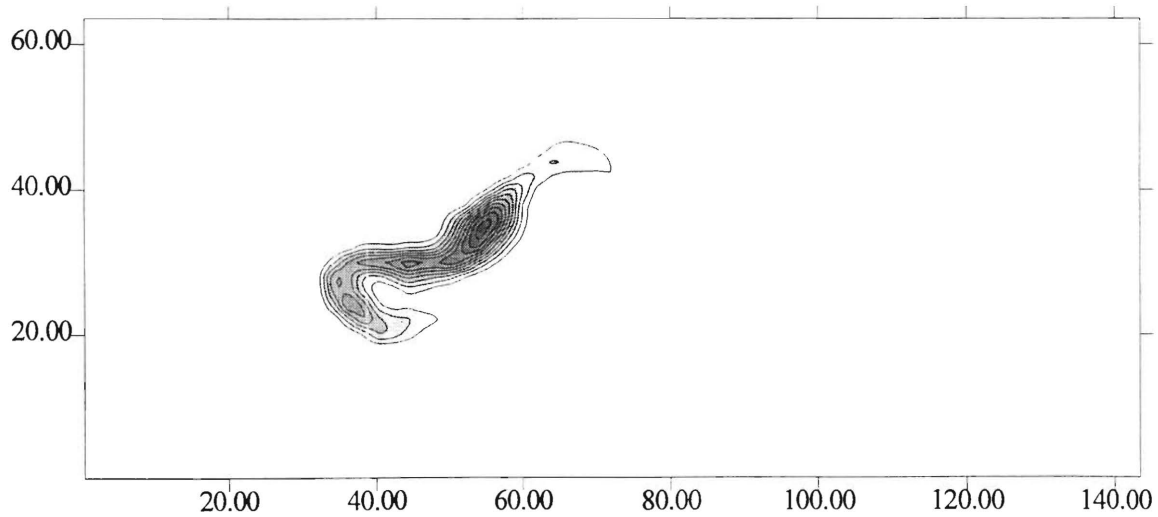


Strongly Heterogeneous Flow Field
Realization 1
Conservative Run
0.1 kg of Bromide

T = 1 Year

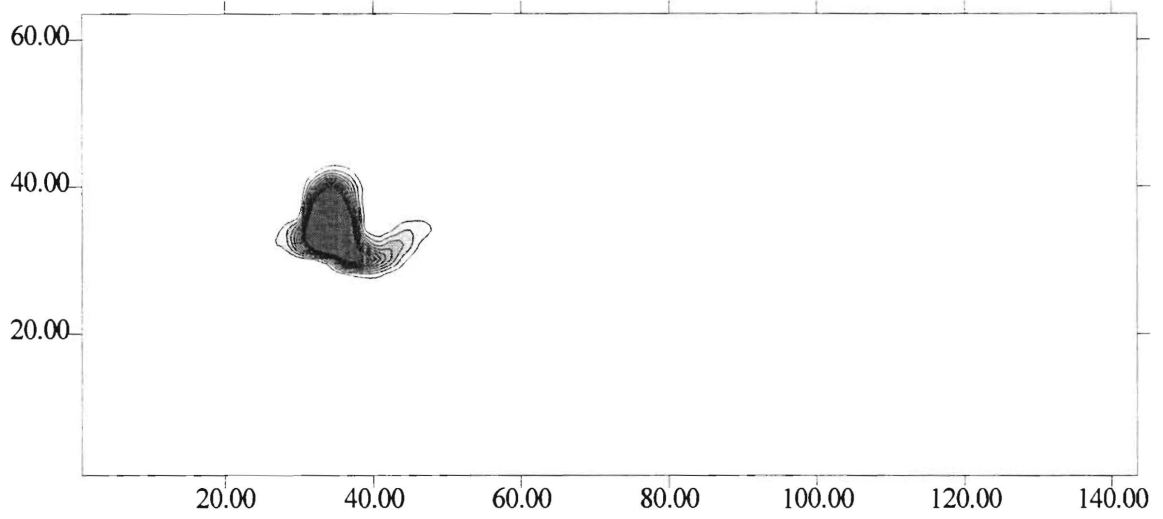


T = 2 Years

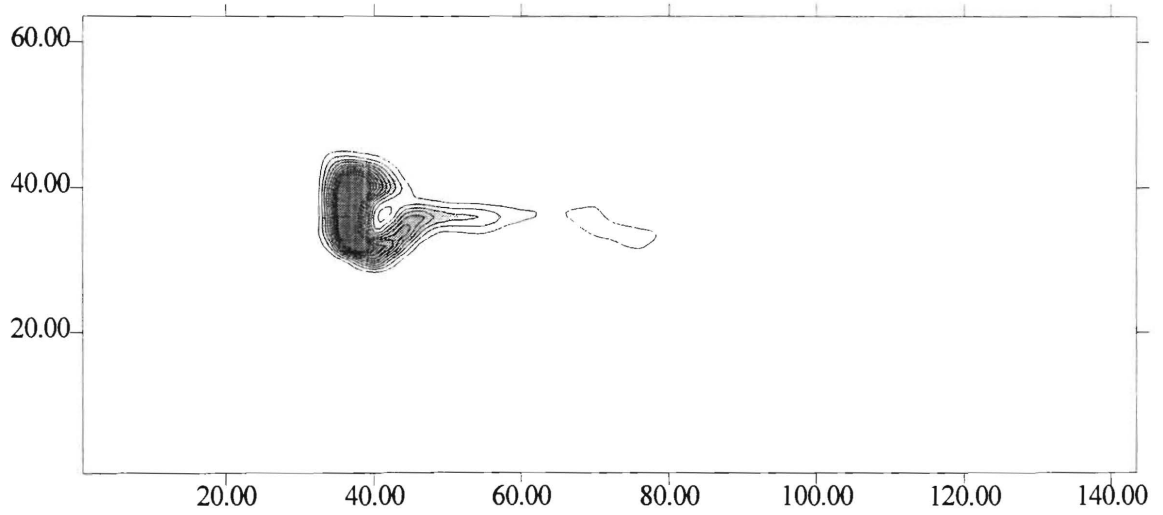


Strongly Heterogeneous Flow Field
Realization 2
Conservative Run
0.1 kg of Bromide

T = 1 Year

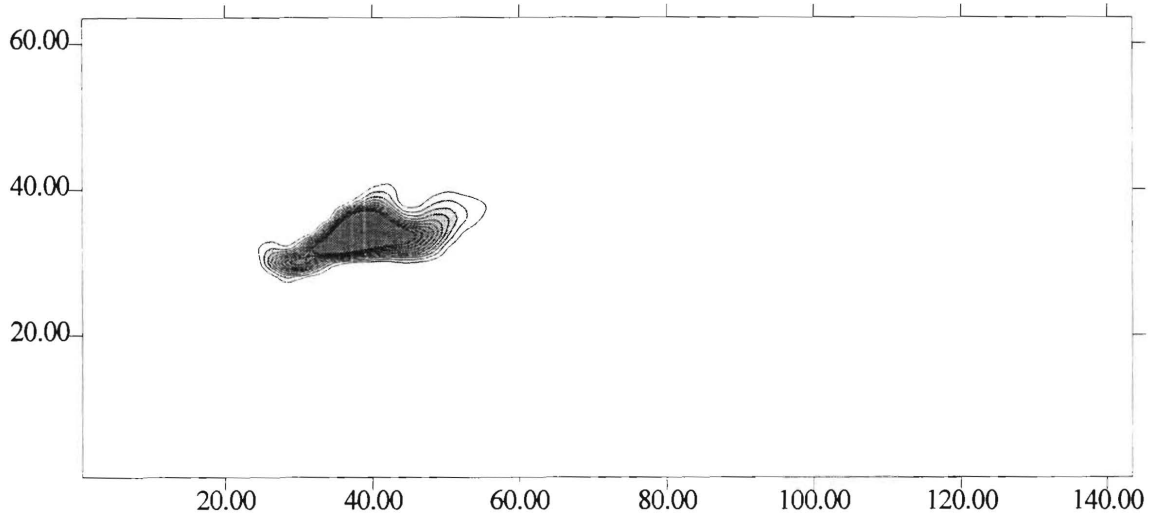


T = 2 Years

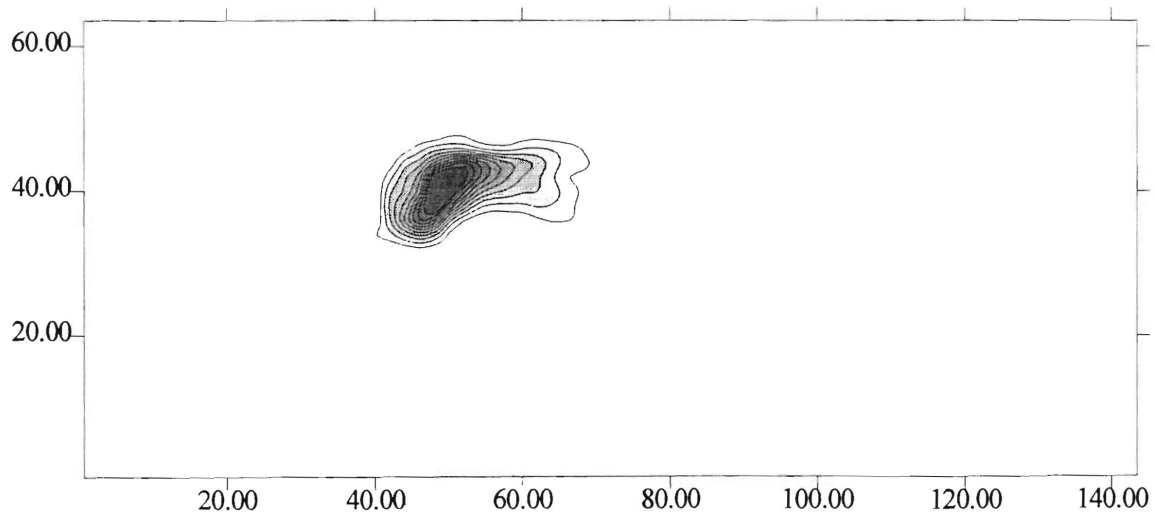


Strongly Heterogeneous Flow Field
Realization 3
Conservative Run
0.1 kg of Bromide

T = 1 Year

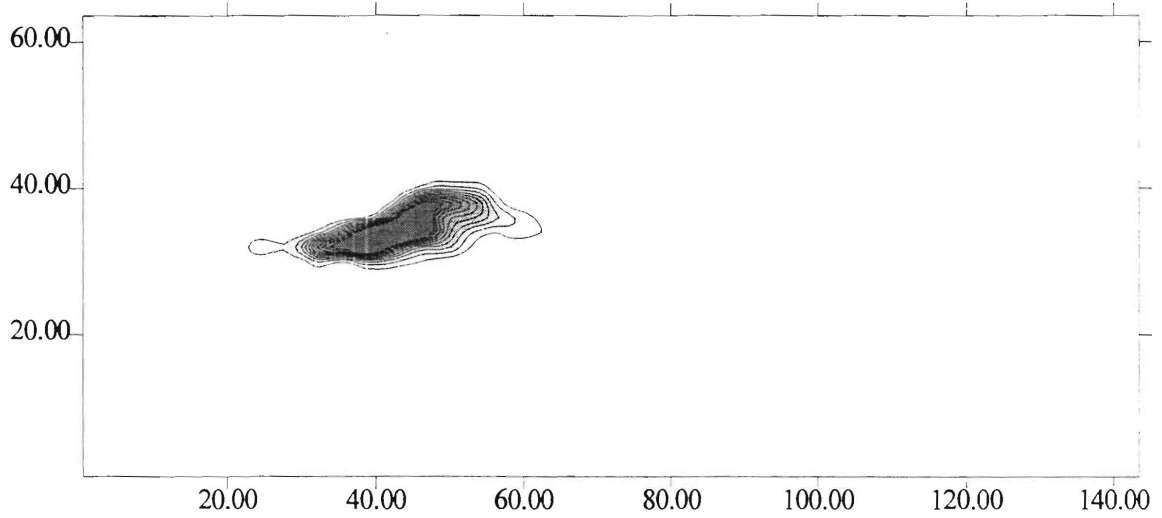


T = 2 Years

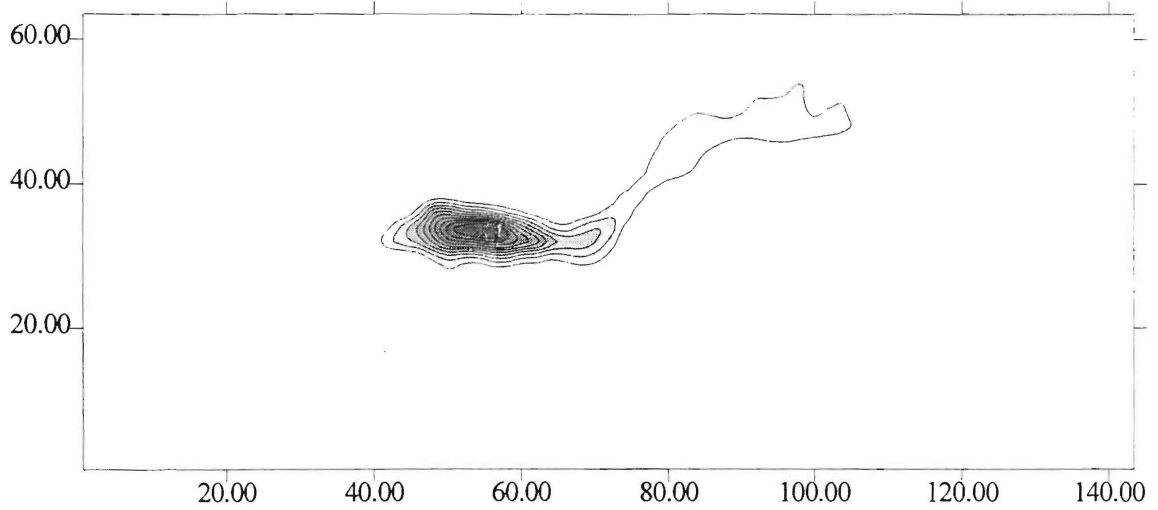


Strongly Heterogeneous Flow Field
Realization 4
Conservative Run
0.1 kg of Bromide

T = 1 Year

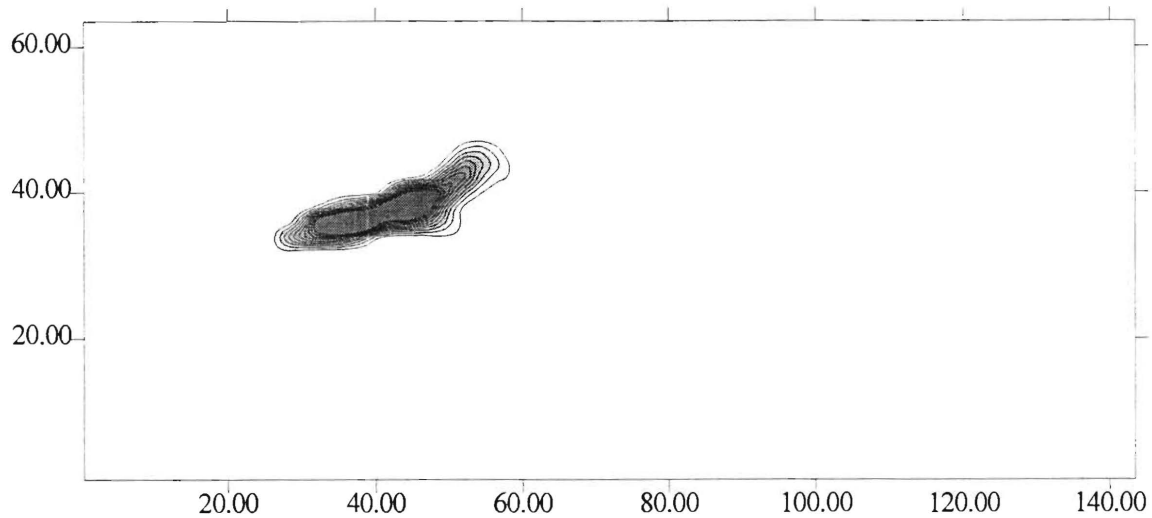


T = 2 Years

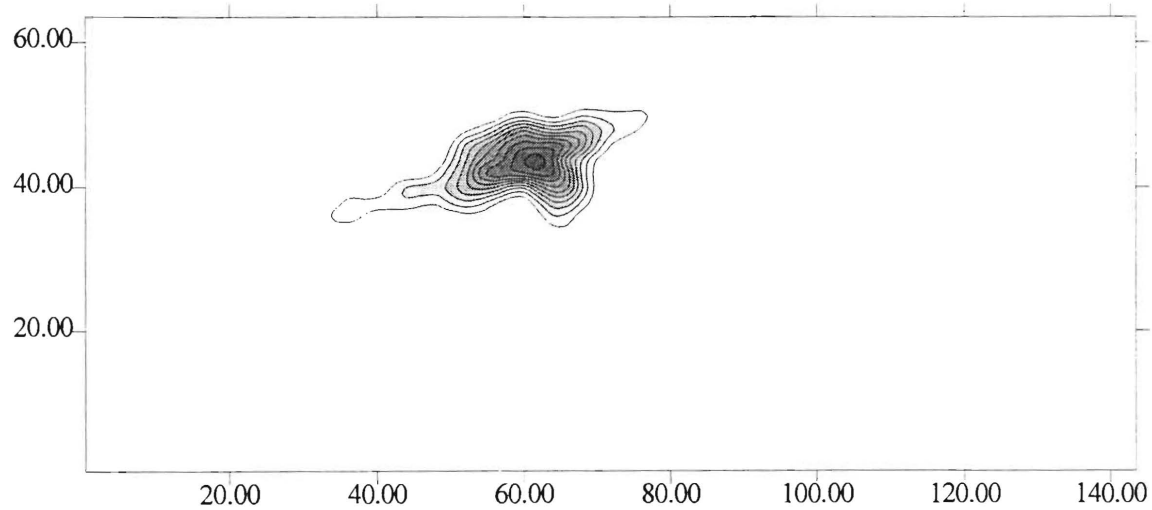


Strongly Heterogeneous Flow Field
Realization 5
Conservative Run
0.1 kg of Bromide

T = 1 Year



T = 2 Years

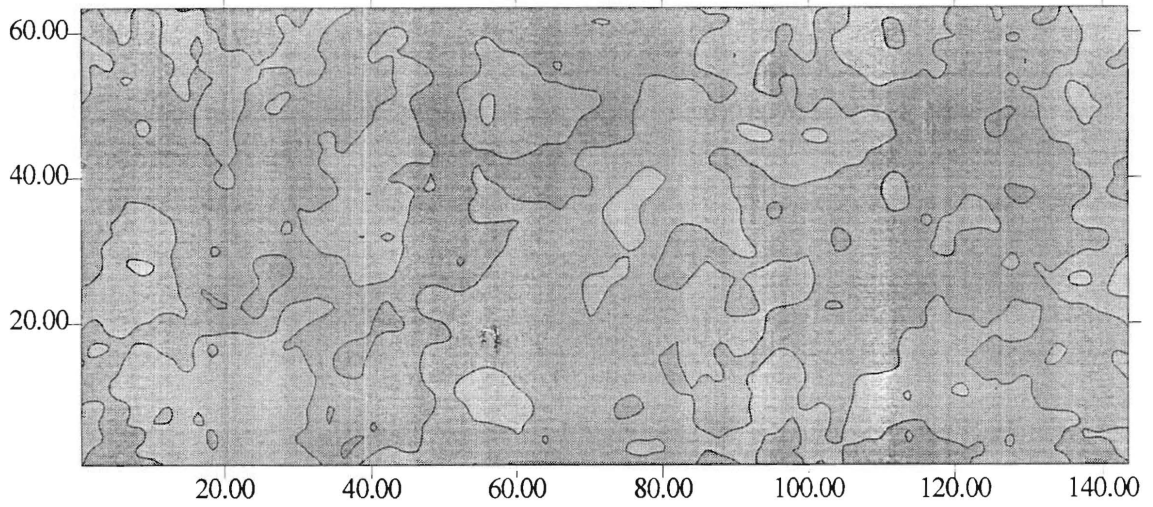


APPENDIX C

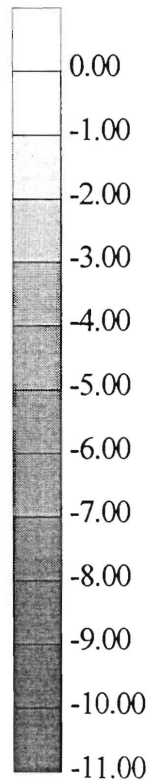
HYDRAULIC CONDUCTIVITY FIELDS

Hydraulic Conductivity Field
Weakly Heterogeneous Flow Field

Realization 1

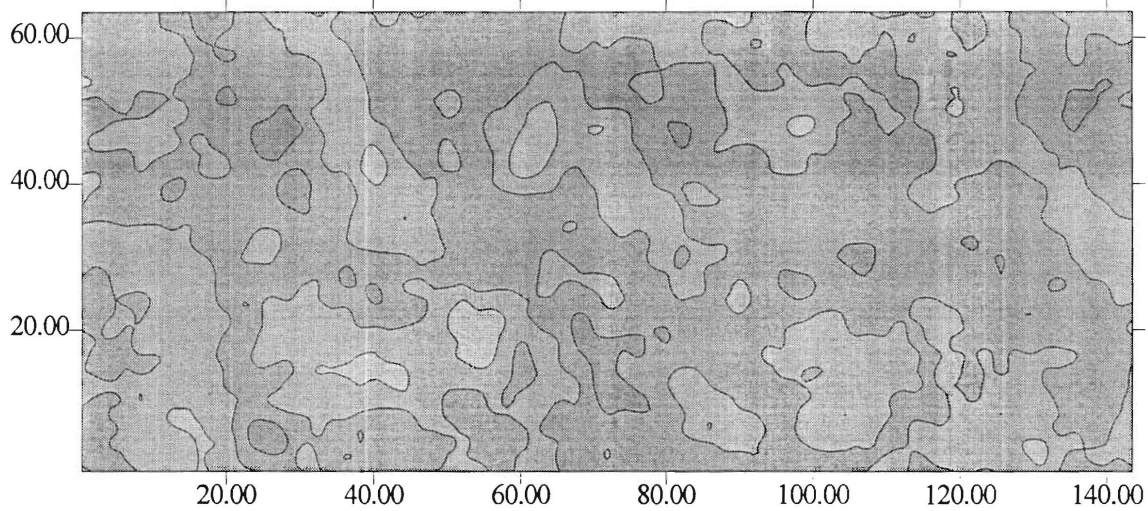


Natural log hydraulic conductivity
scale for all realizations.

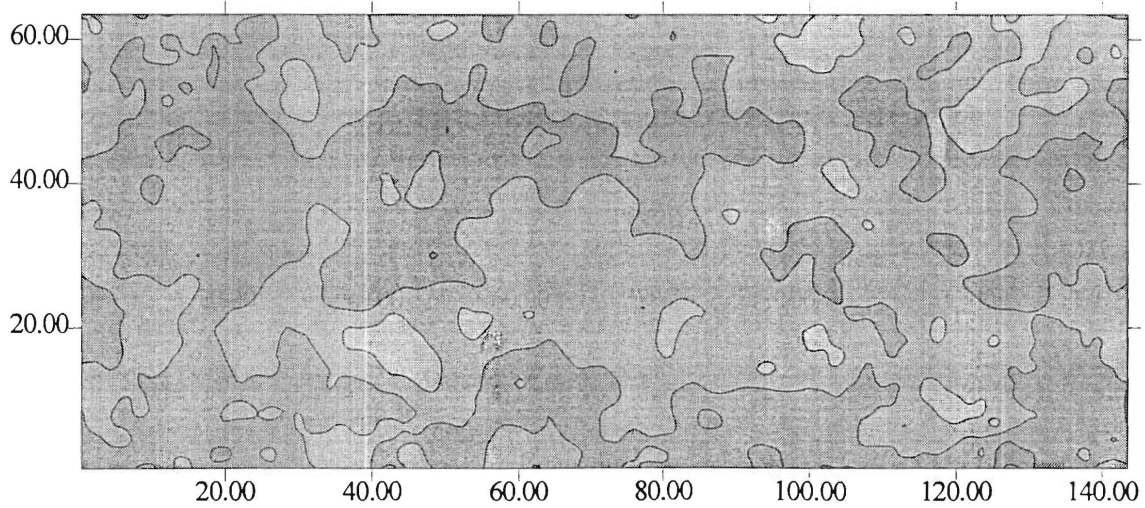


Hydraulic Conductivity Field
Weakly Heterogeneous Flow Field

Realization 2

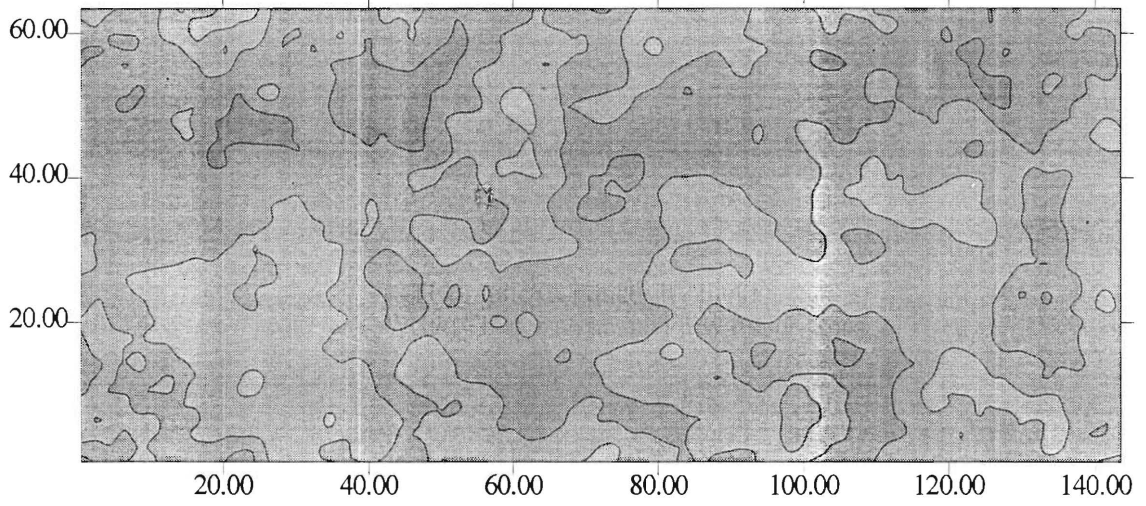


Realization 3

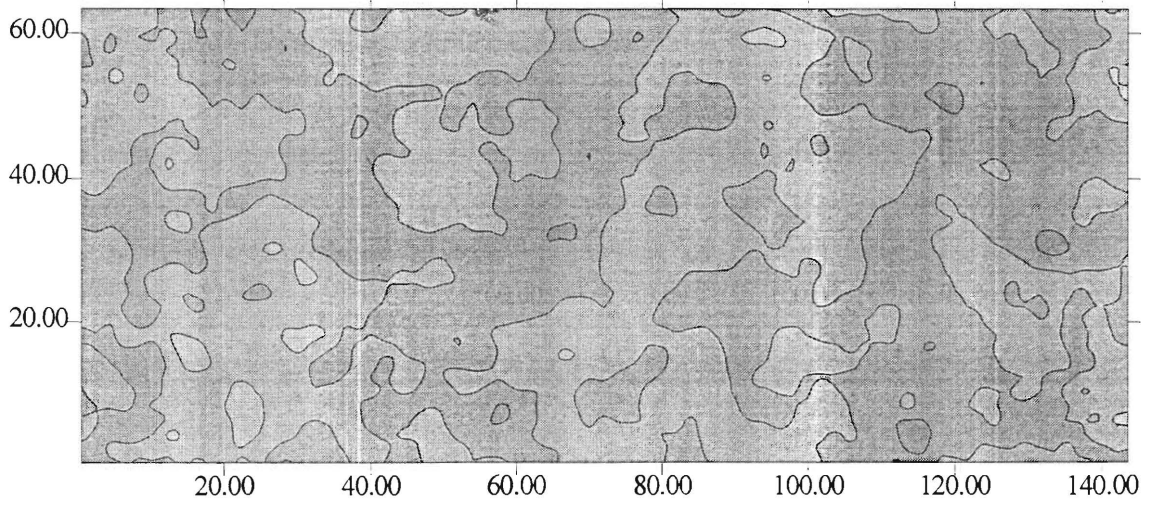


Hydraulic Conductivity Field
Weakly Heterogeneous Flow Field

Realization 4

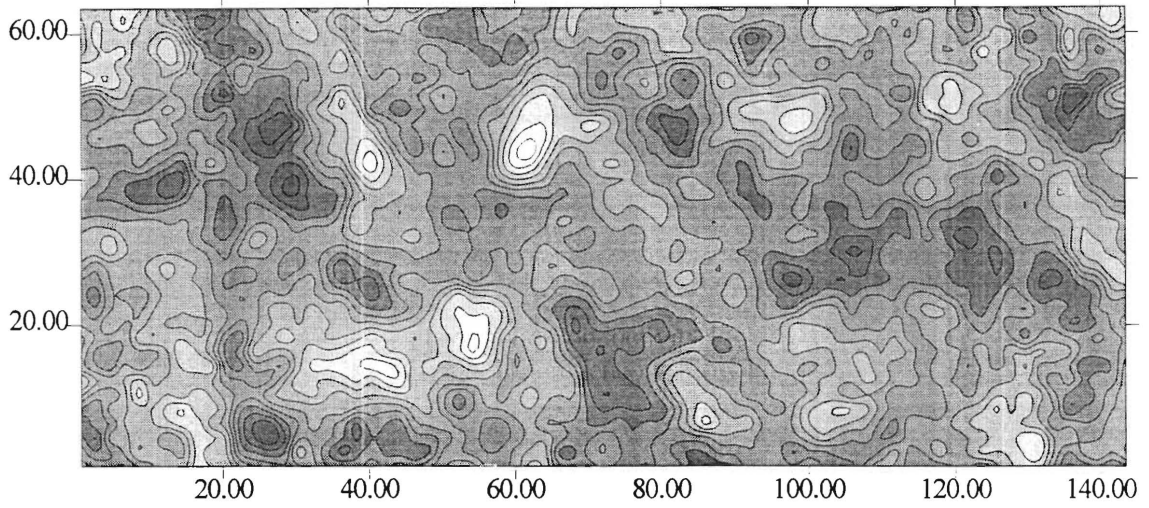


Realization 5

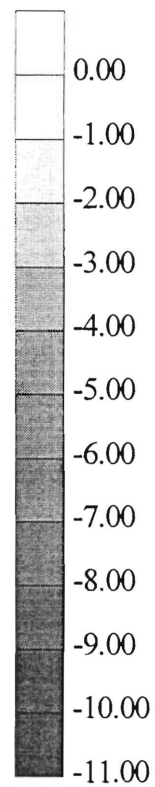


Hydraulic Conductivity Field
Strongly Heterogeneous Flow Field

Realization 1

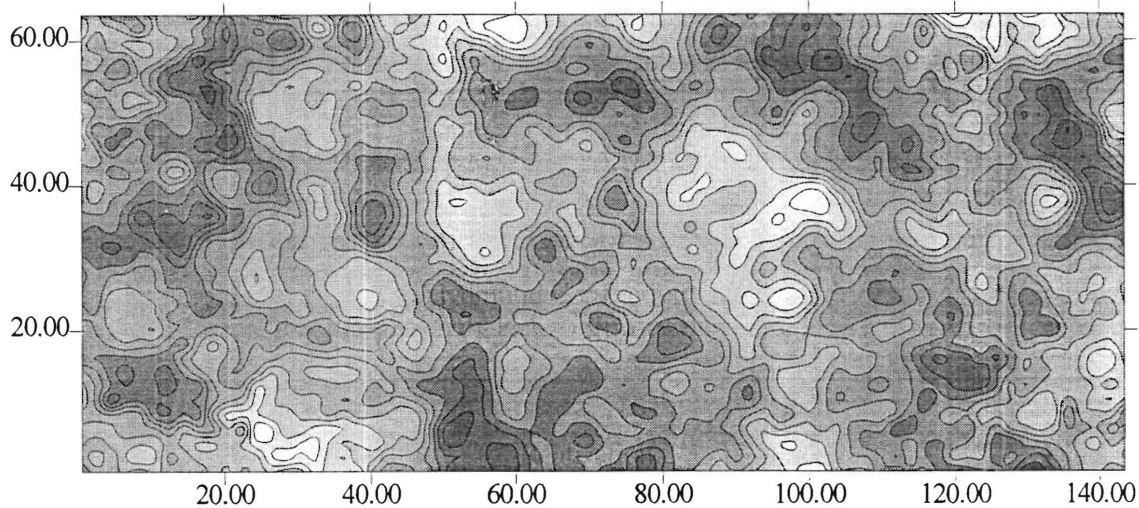


Natural log hydraulic conductivity
scale for all realizations.

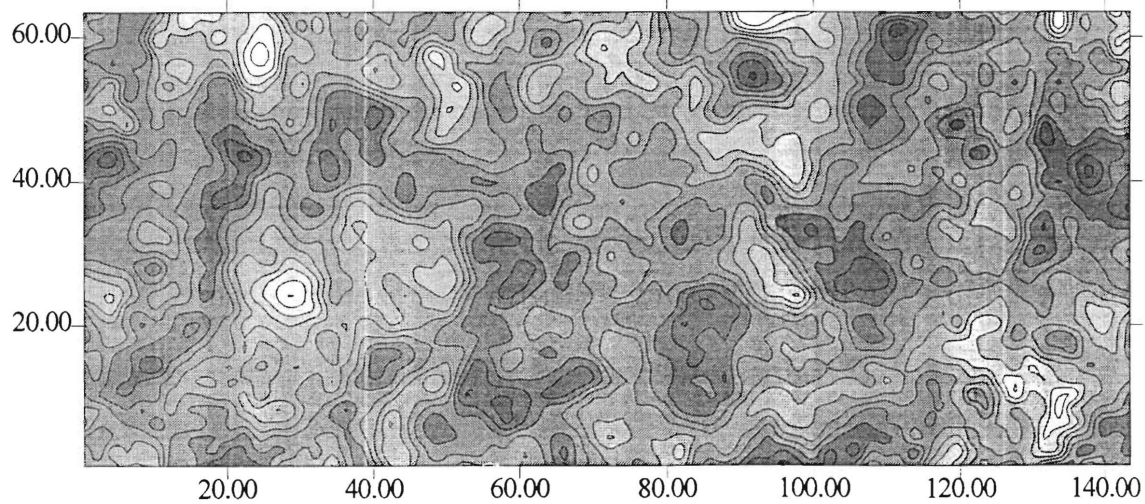


Hydraulic Conductivity Field
Strongly Heterogeneous Flow Field

Realization 2

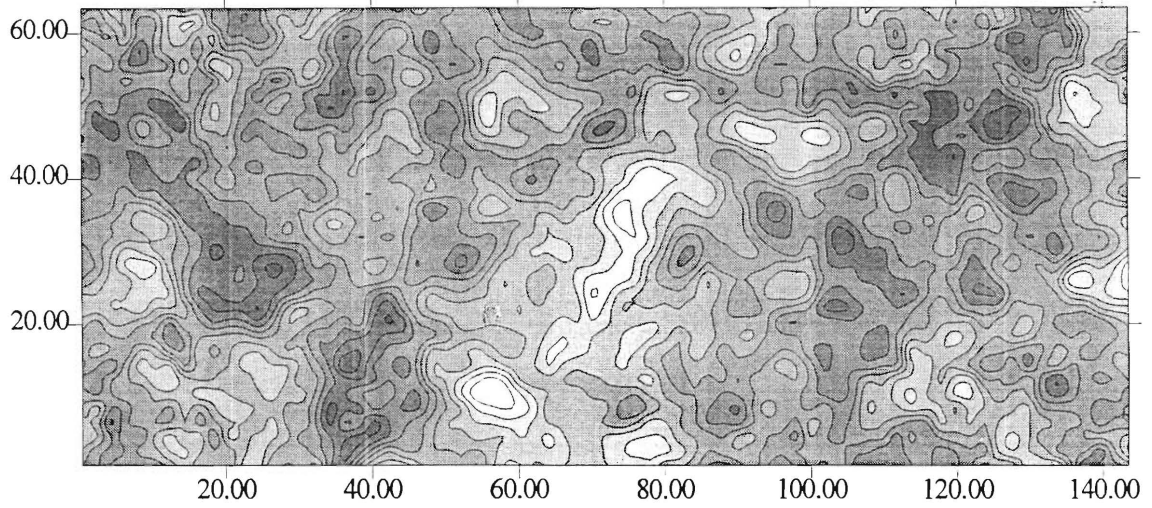


Realization 3

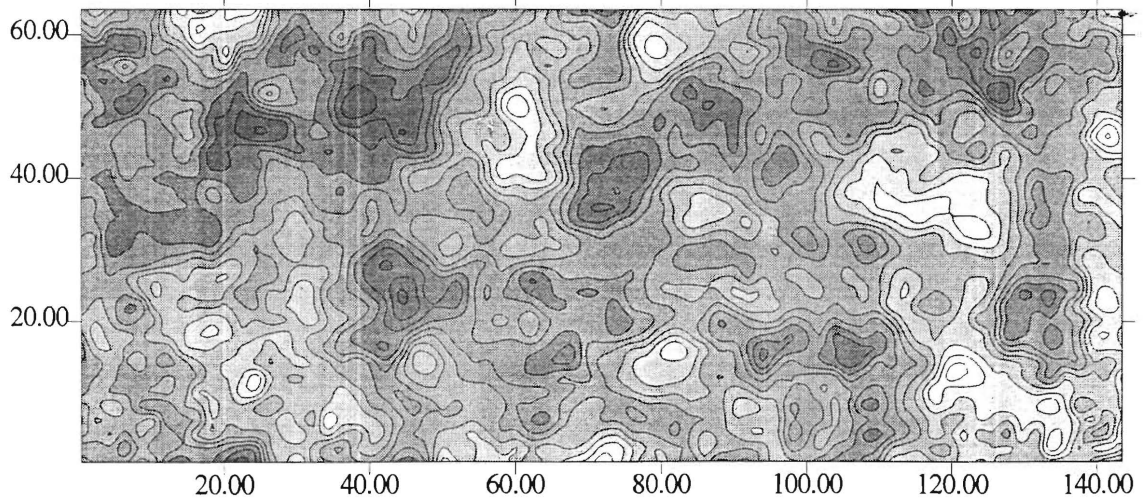


Hydraulic Conductivity Field
Strongly Heterogeneous Flow Field

Realization 4

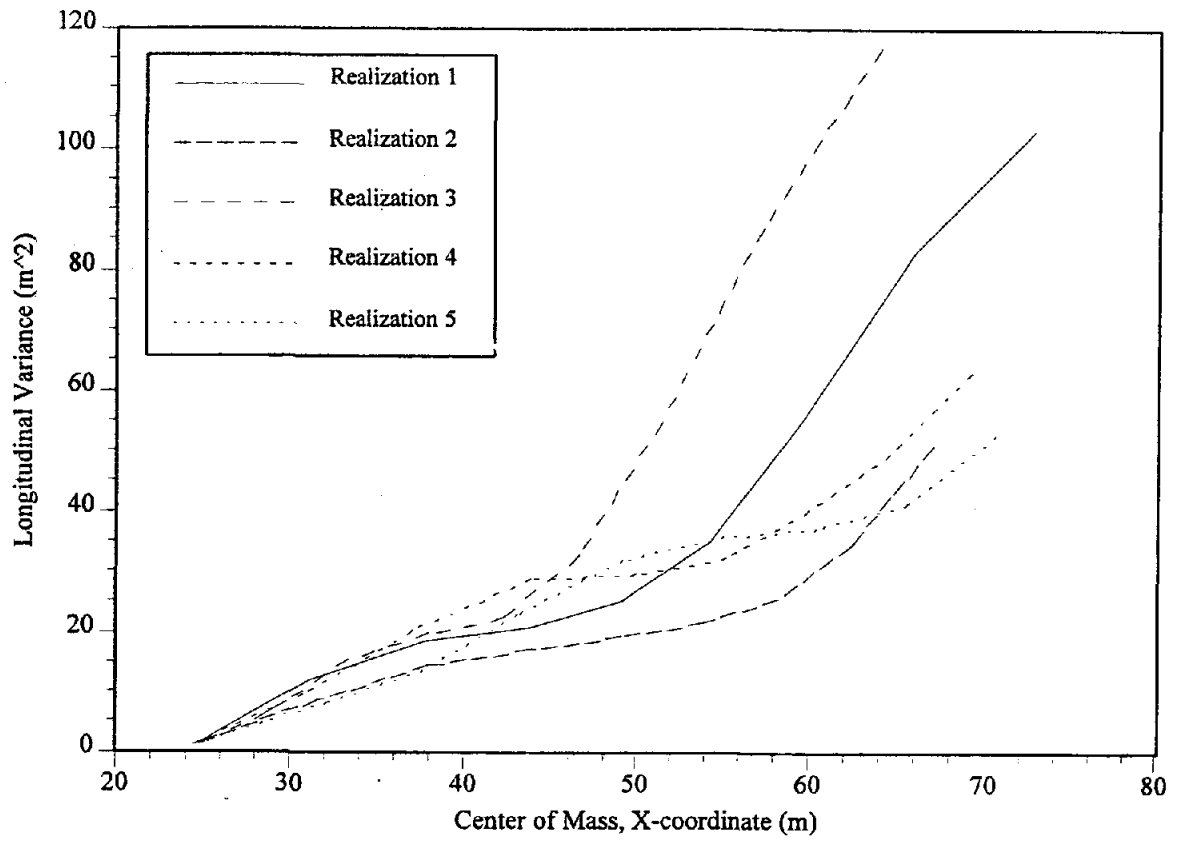


Realization 5

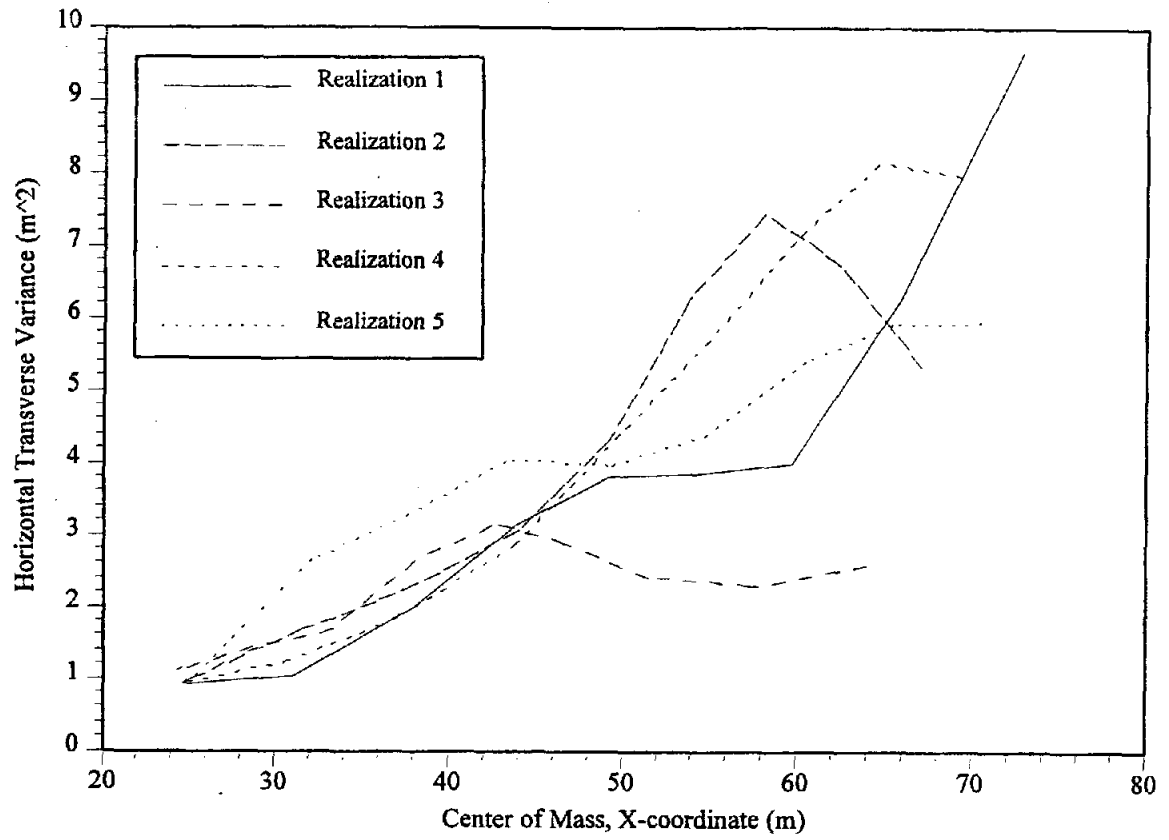


APPENDIX D

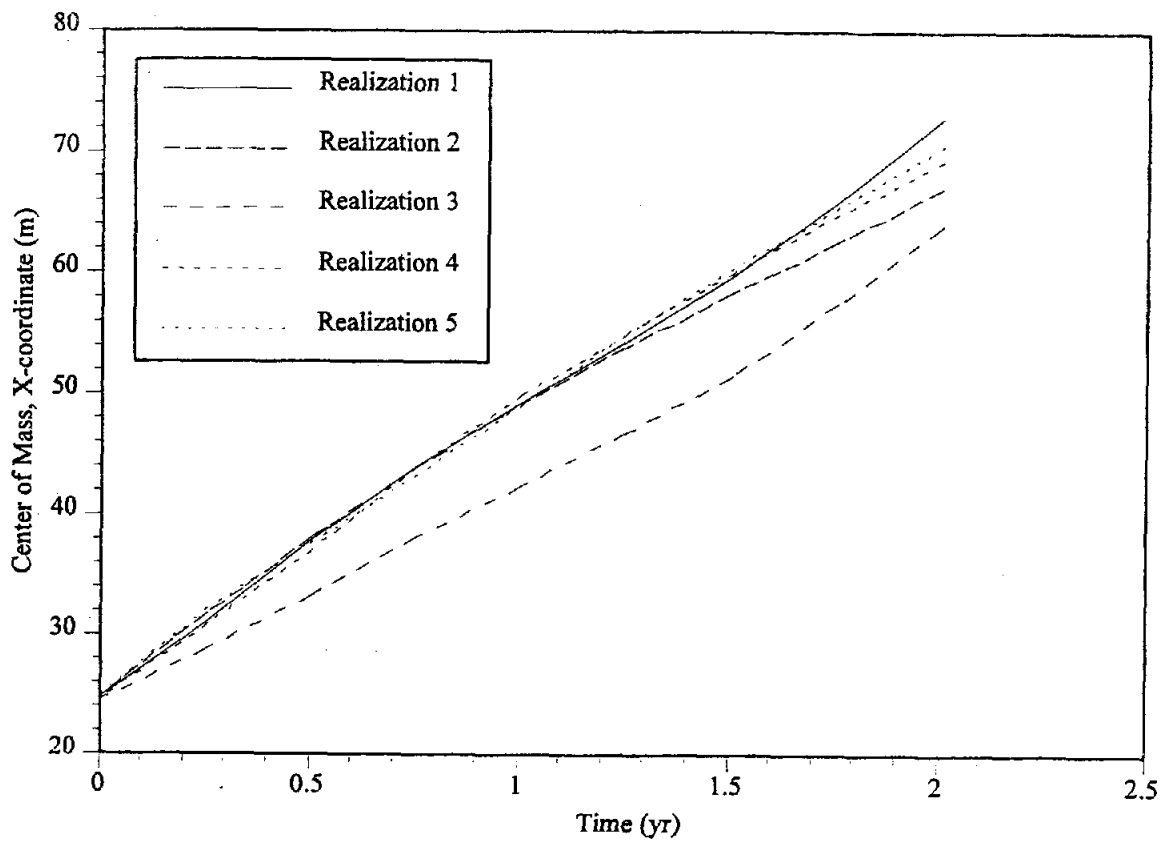
GRAPHS



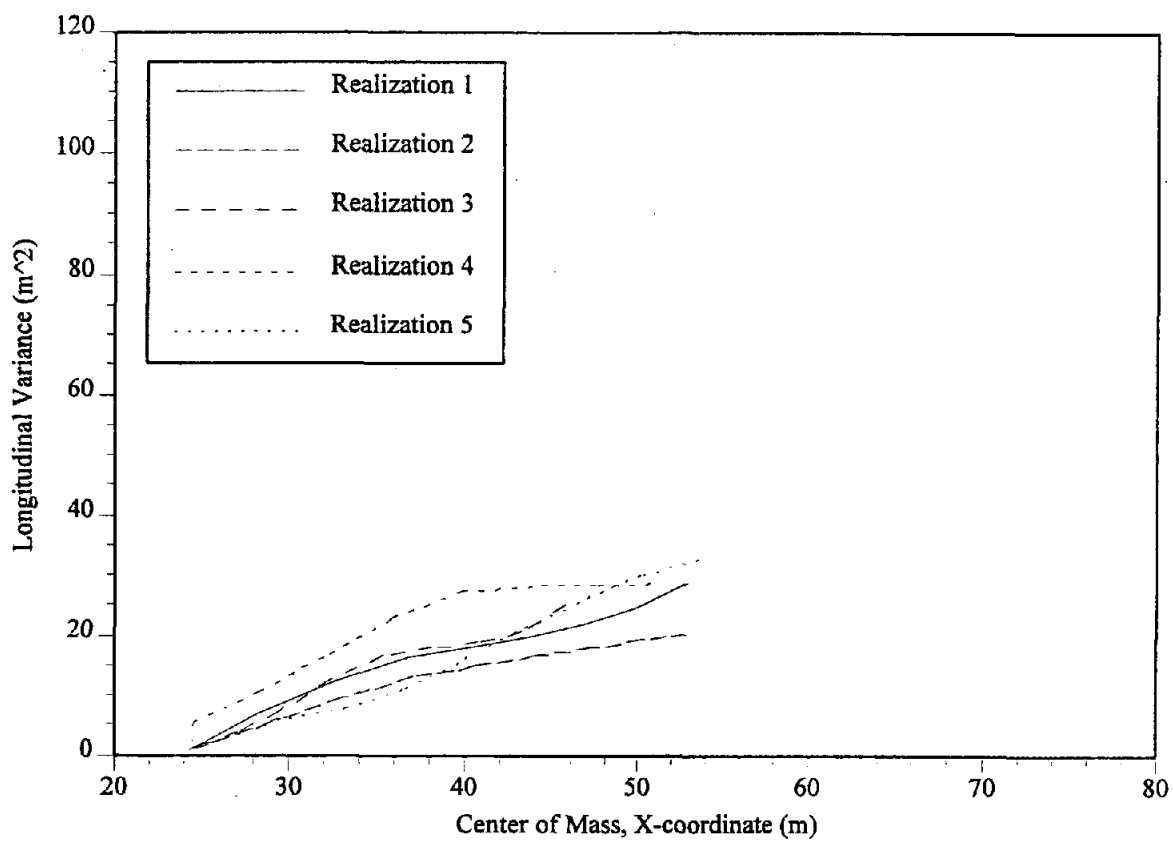
Conservative simulations in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



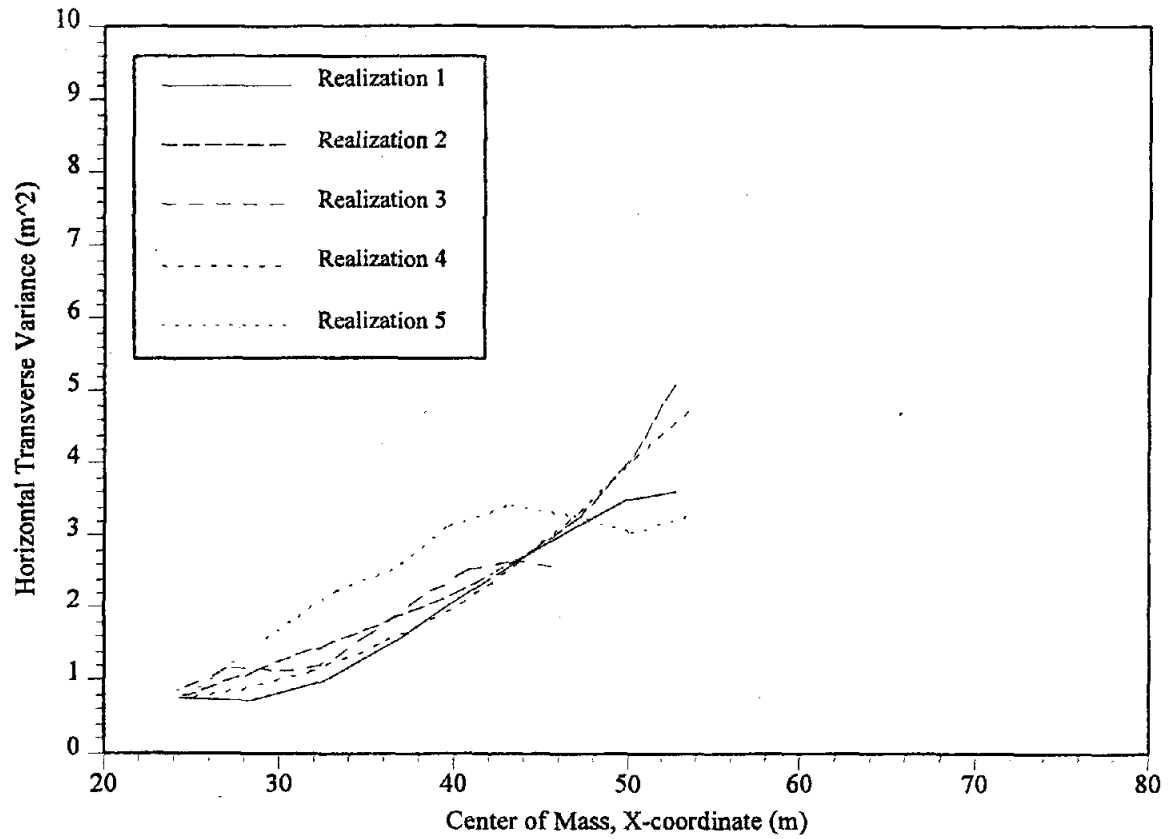
Conservative simulations in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



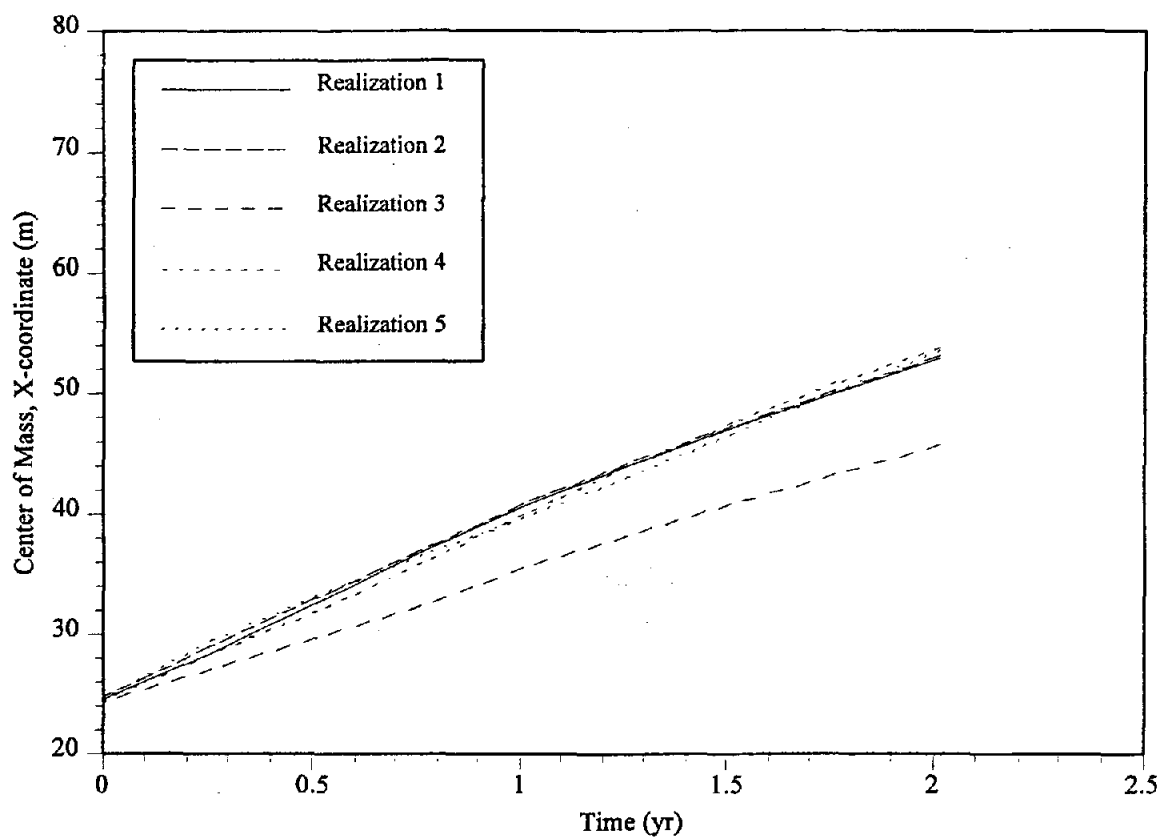
Conservative simulations in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



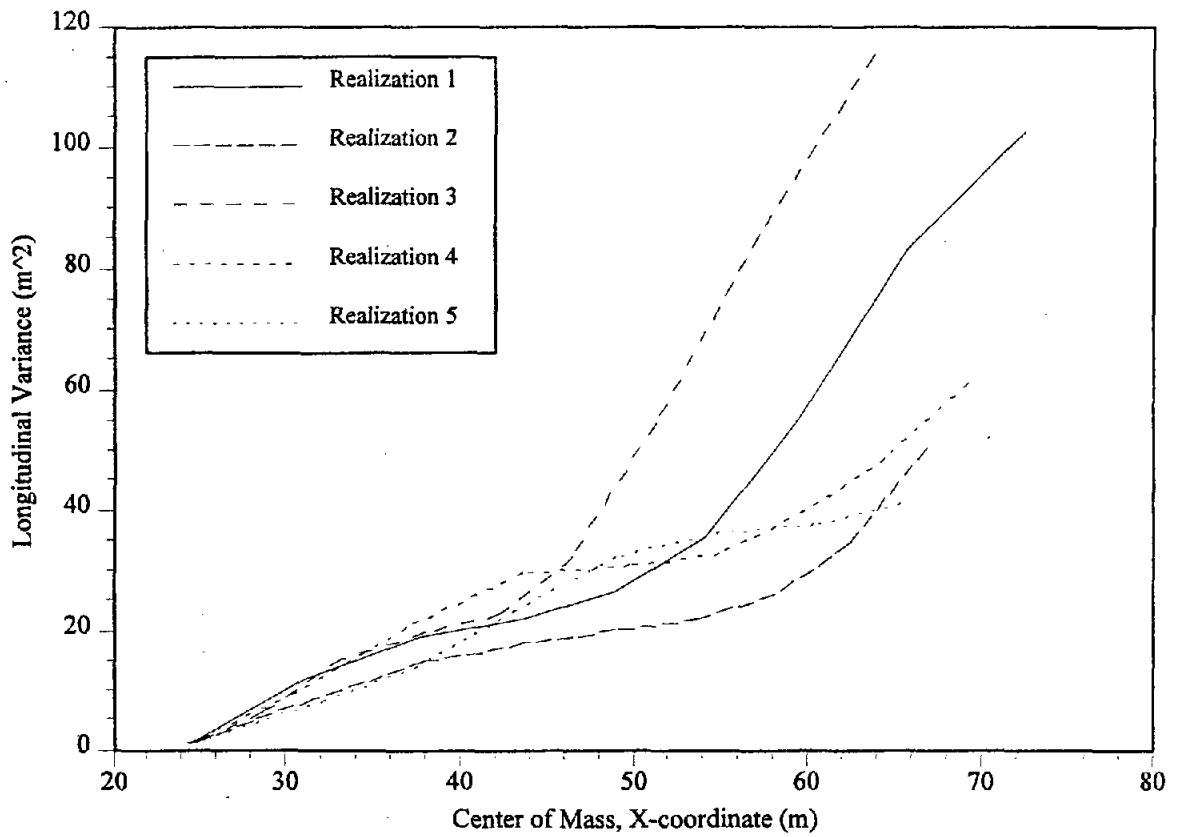
Reactive simulations in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



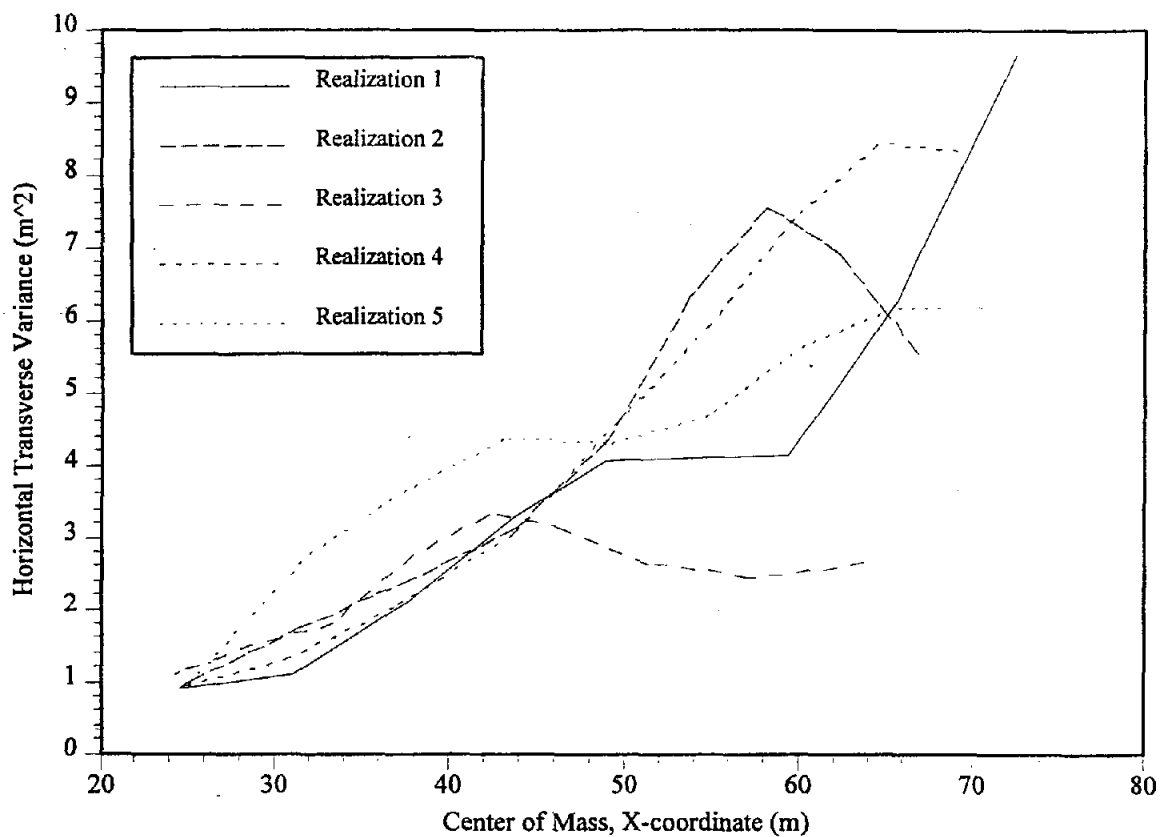
Reactive simulations in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



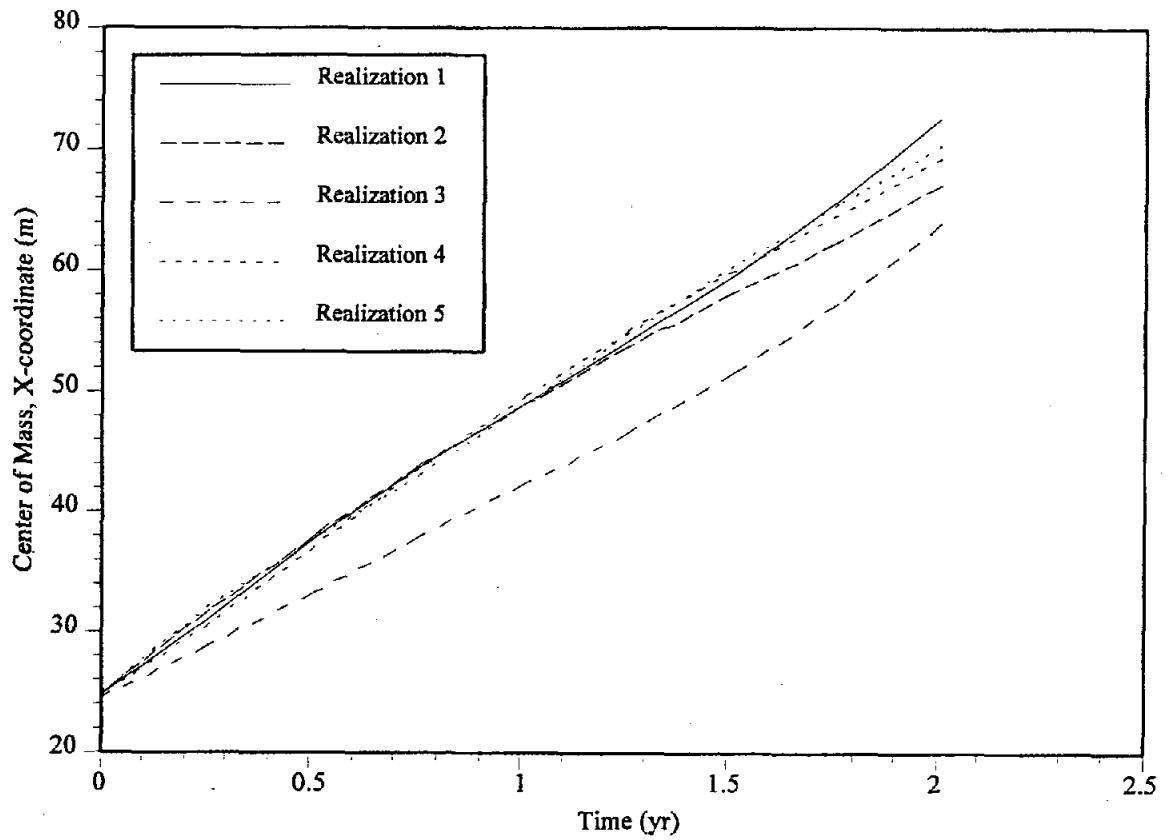
Reactive simulations in a weakly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



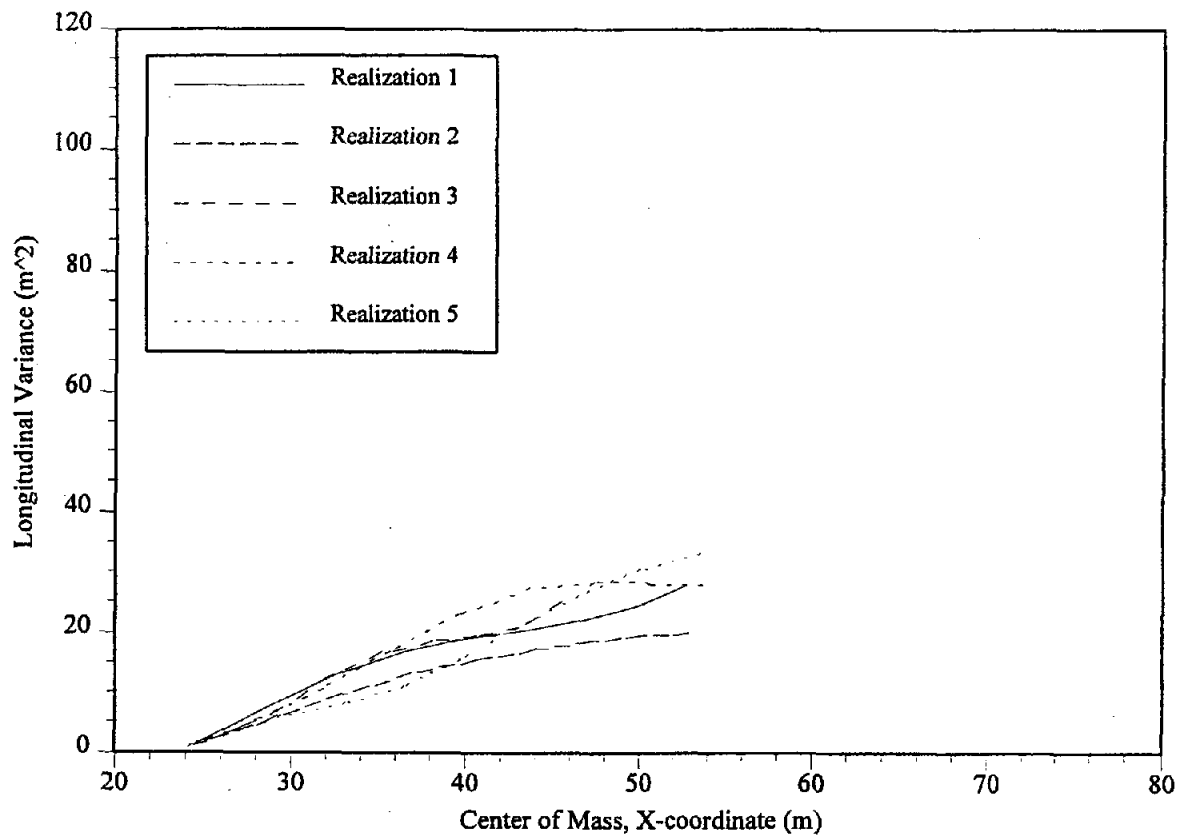
Conservative simulations in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



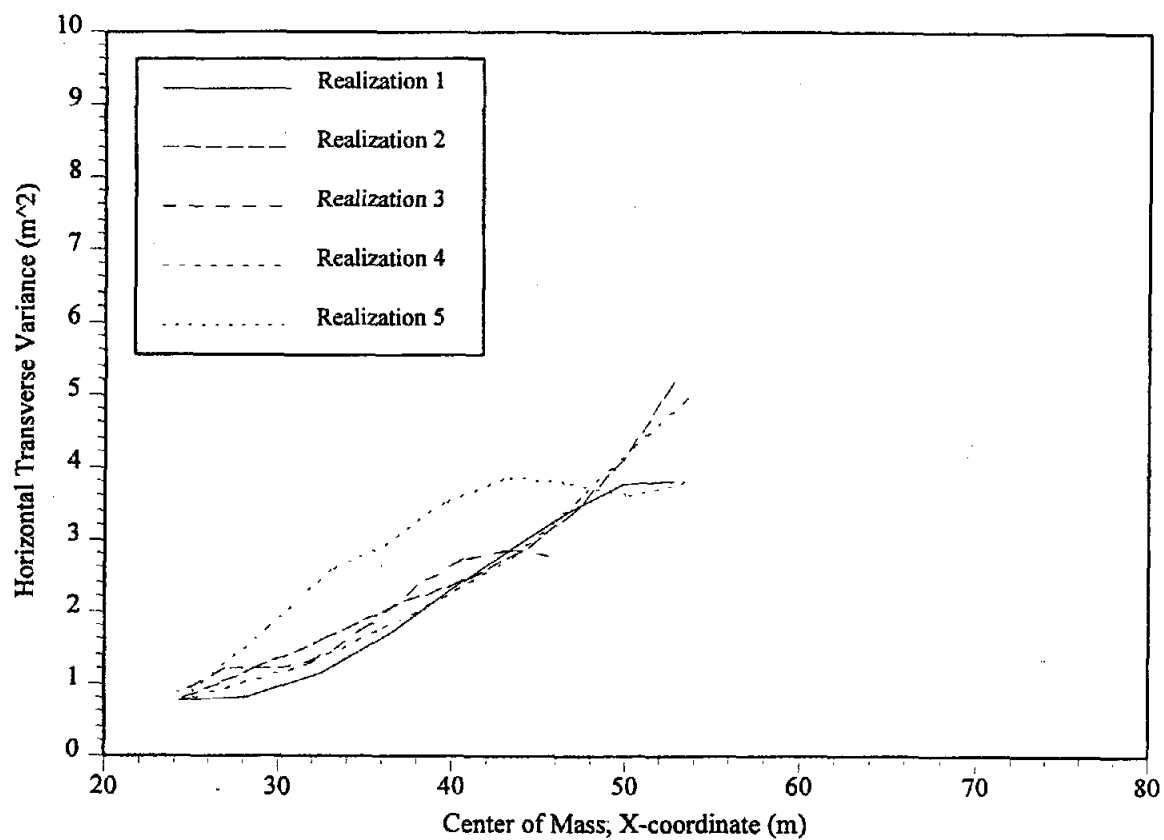
Conservative simulations in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



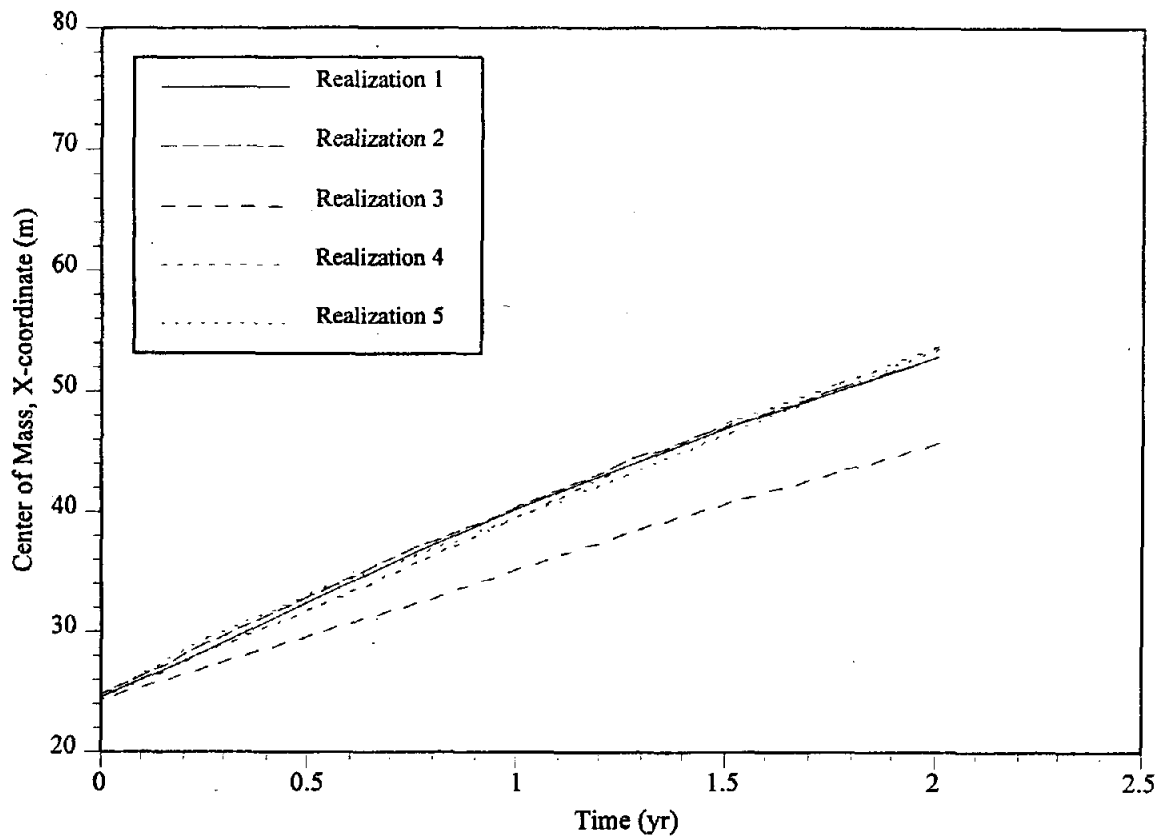
Conservative simulations in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



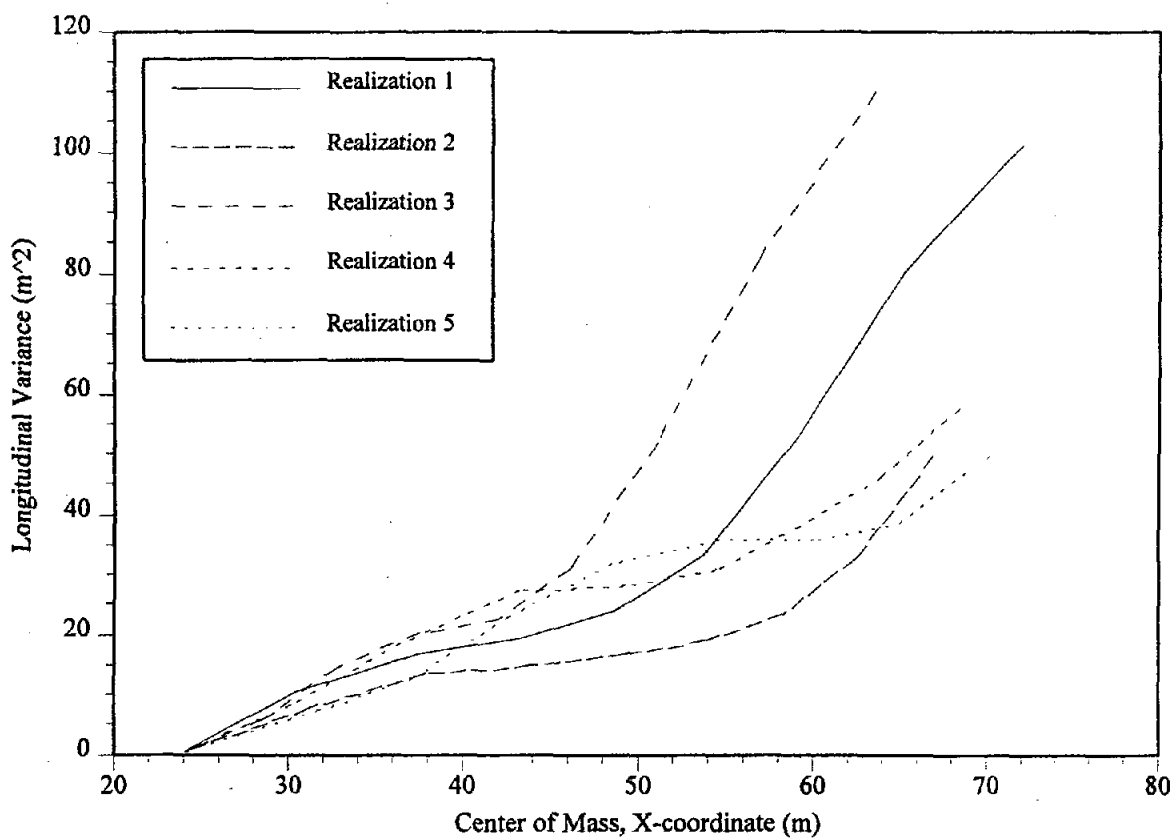
Reactive simulations in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



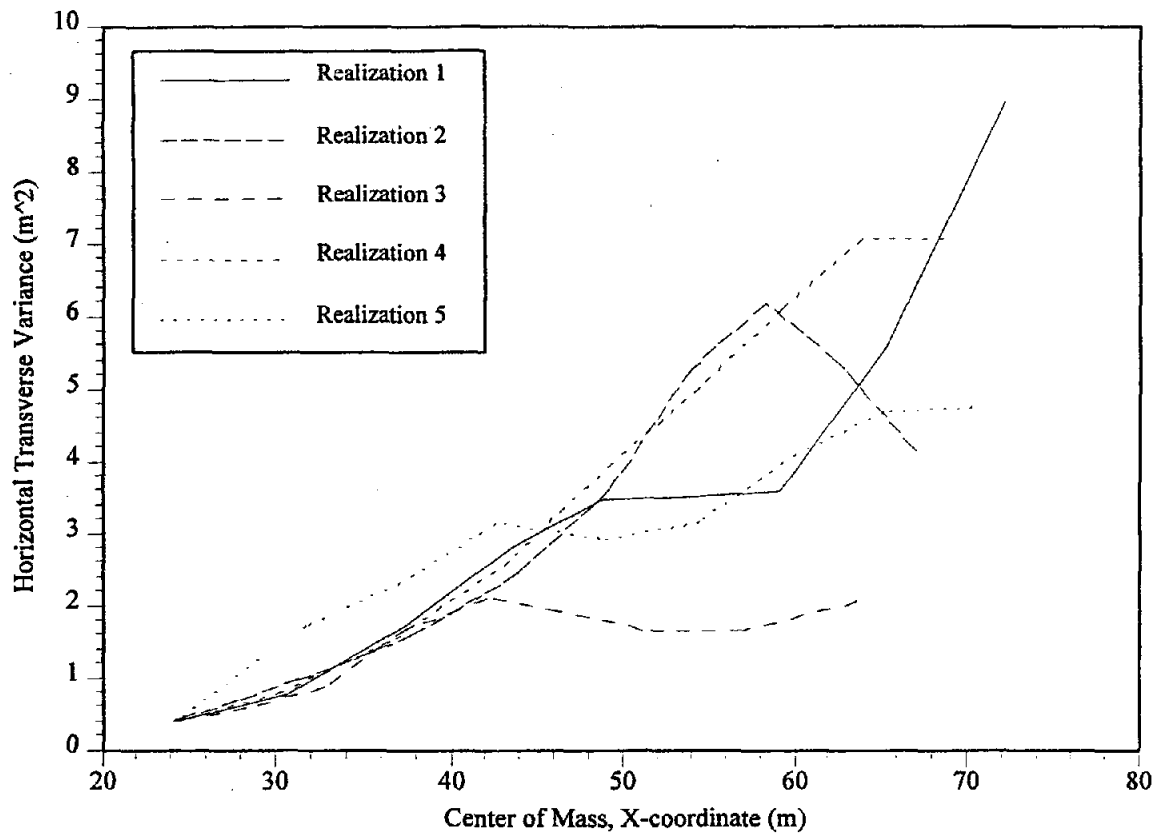
Reactive simulations in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



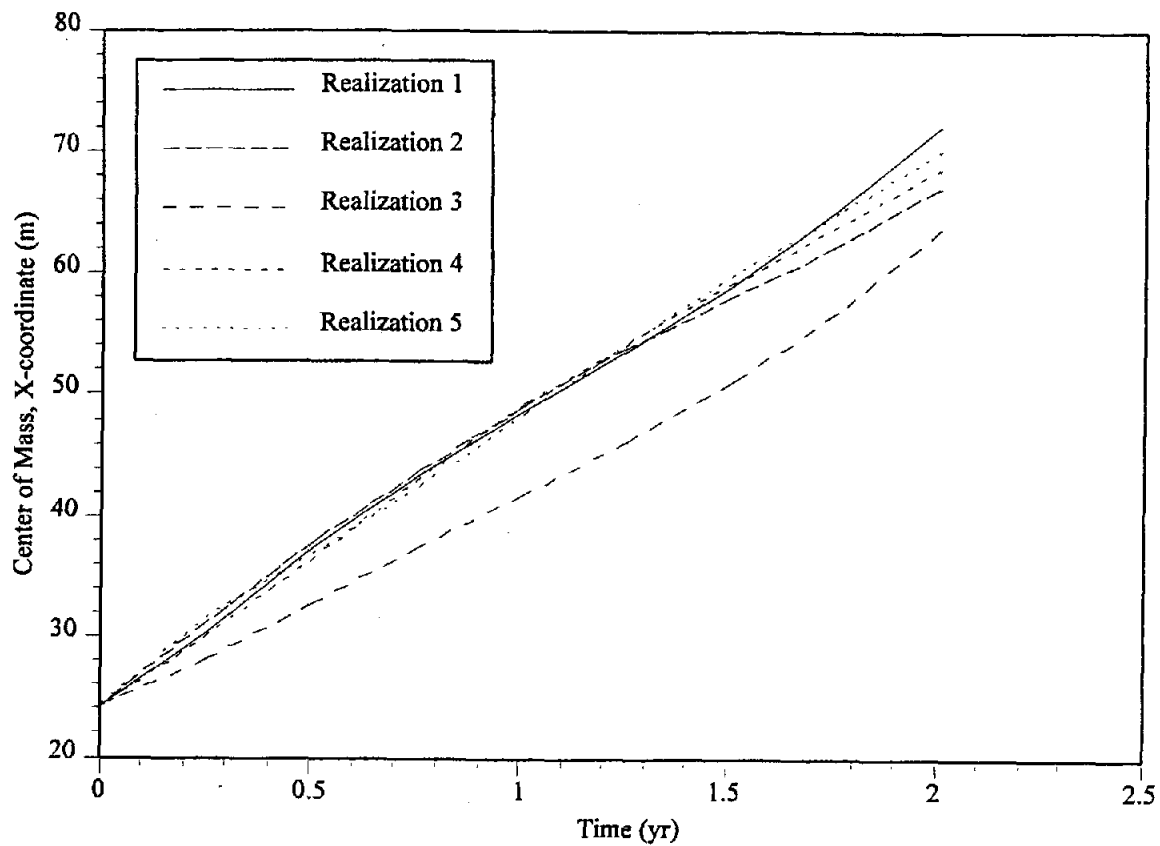
Reactive simulations in a weakly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



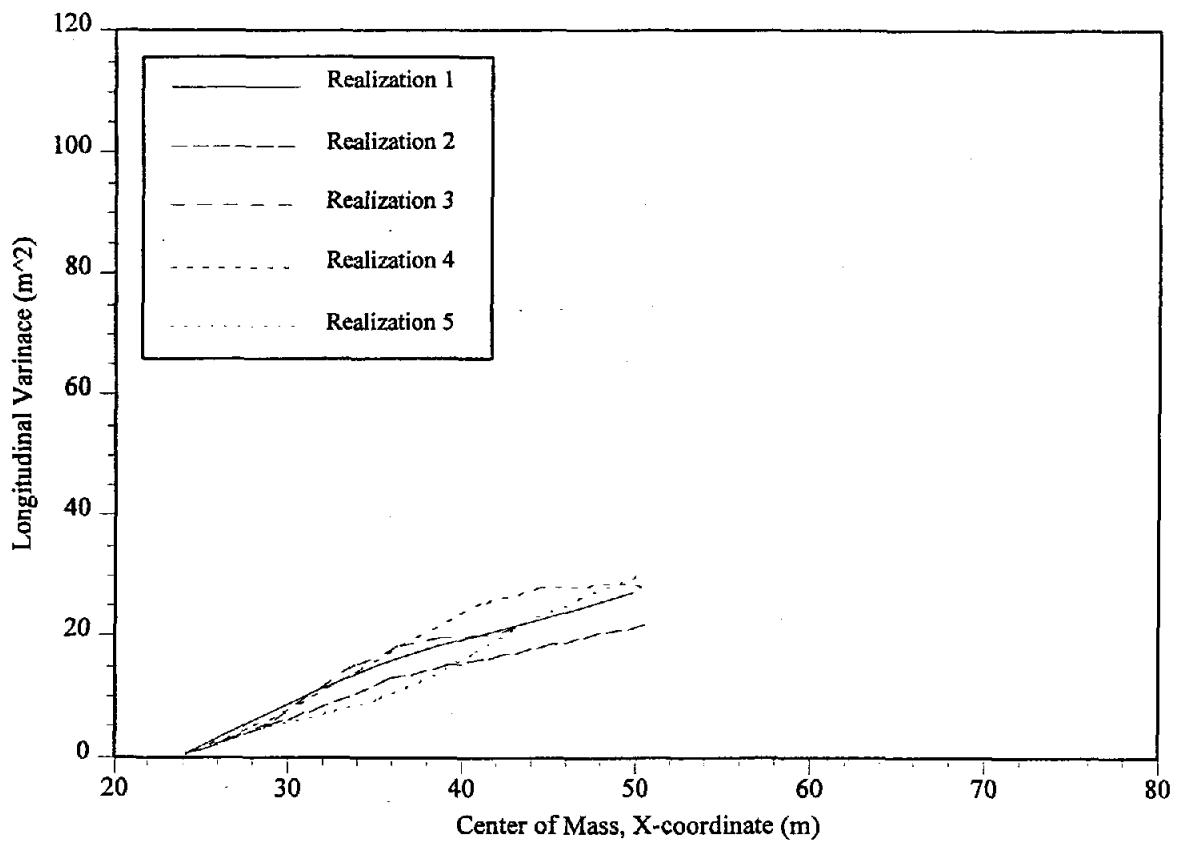
Conservative simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. All Concentrations used in calculations.



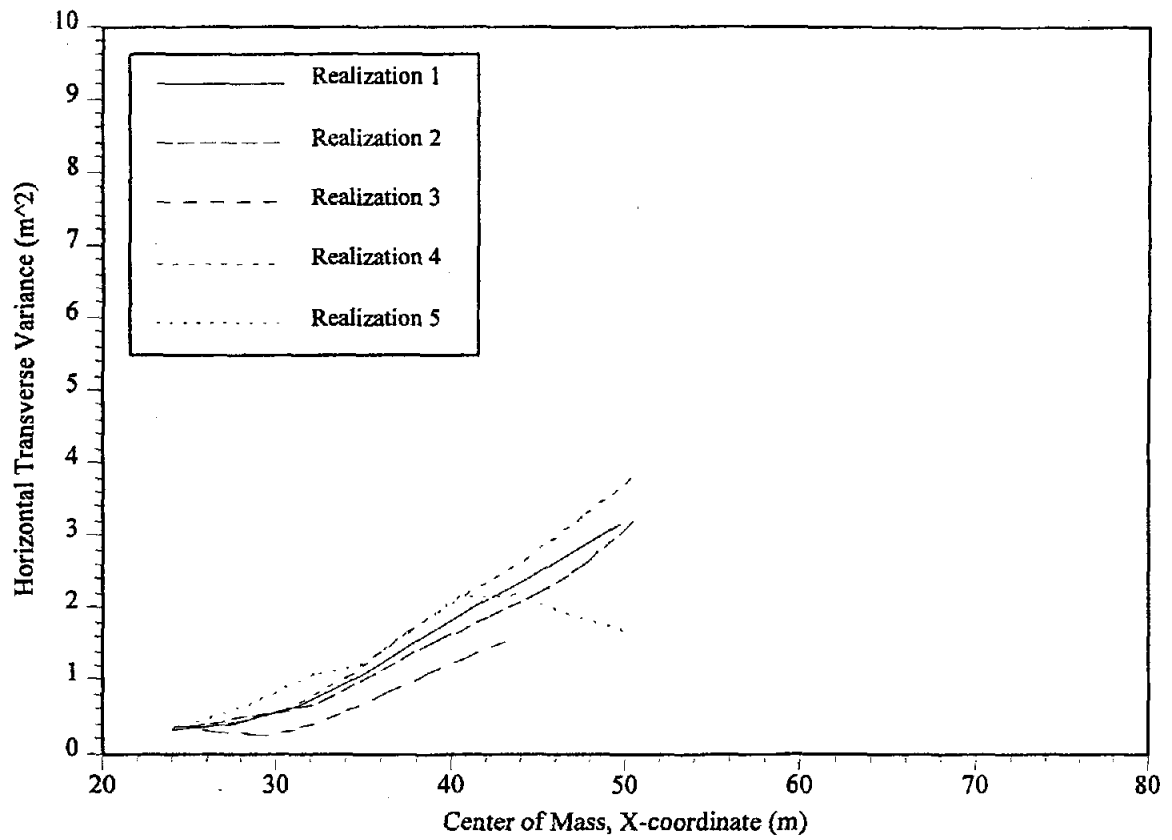
Conservative simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



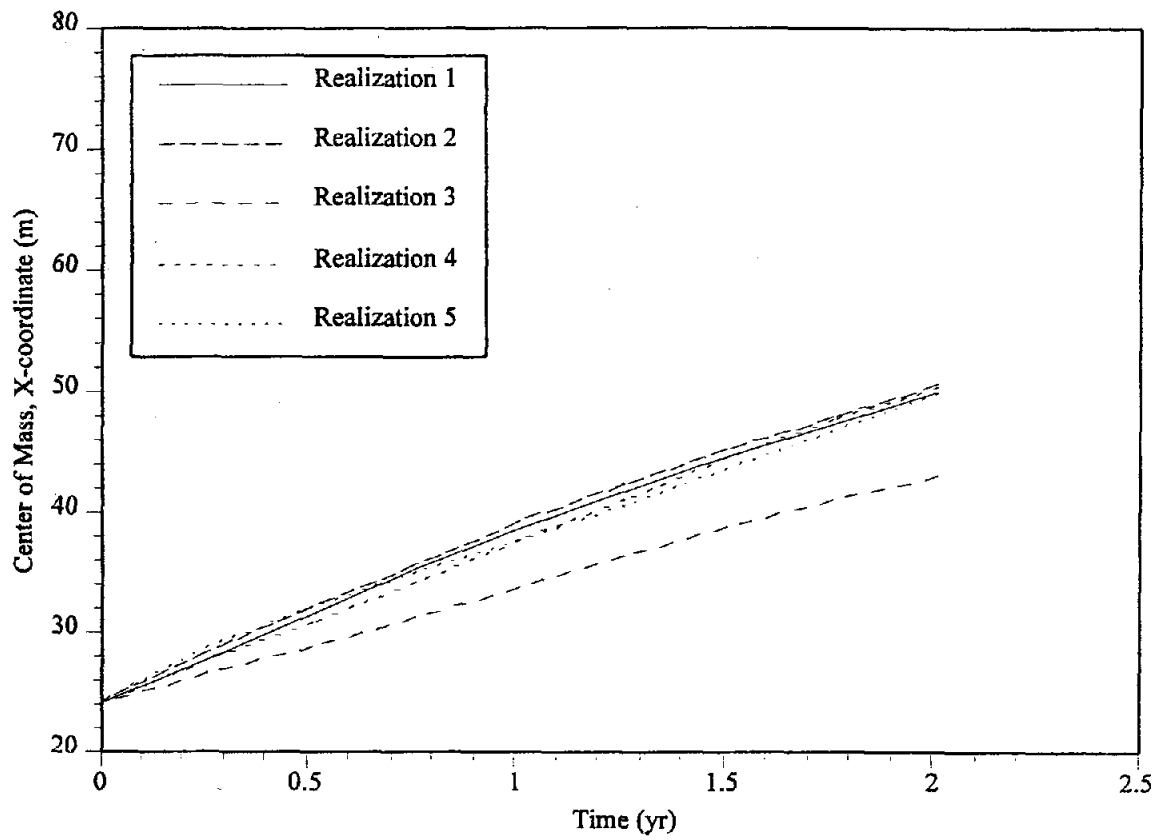
Conservative simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



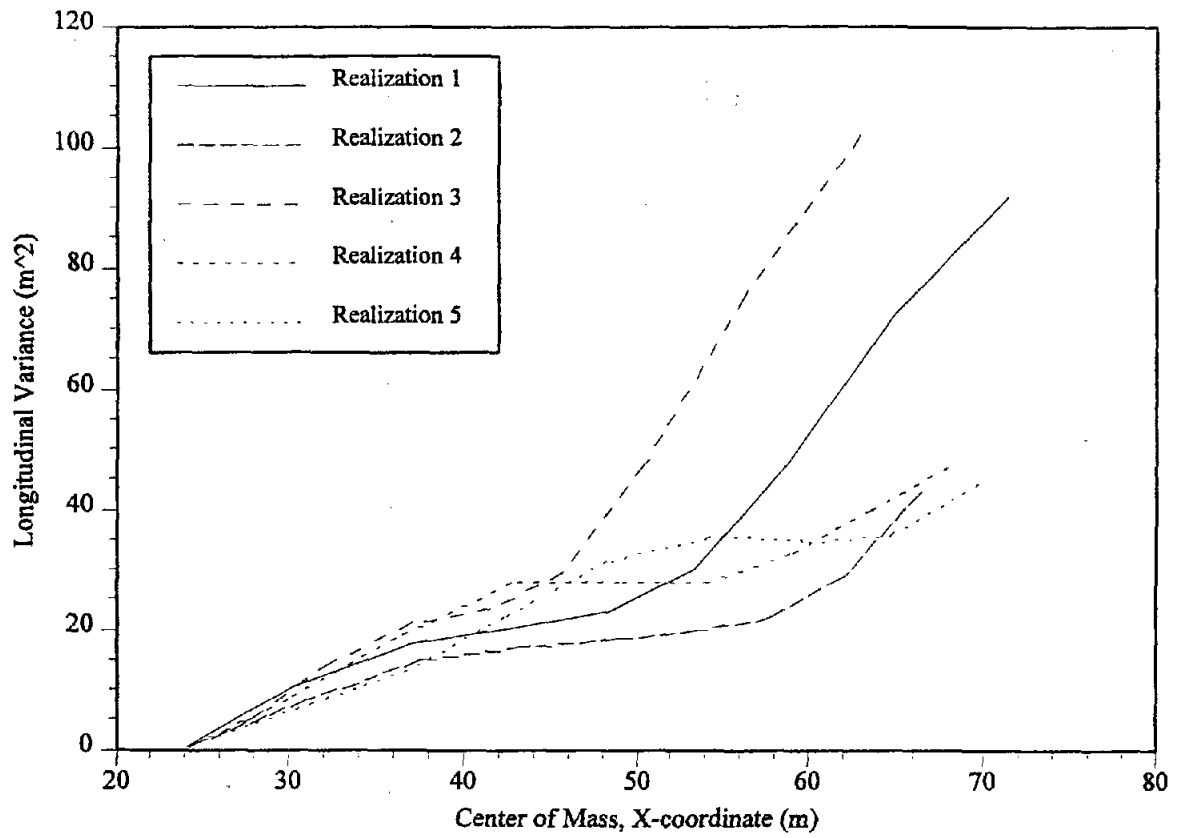
Reactive simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



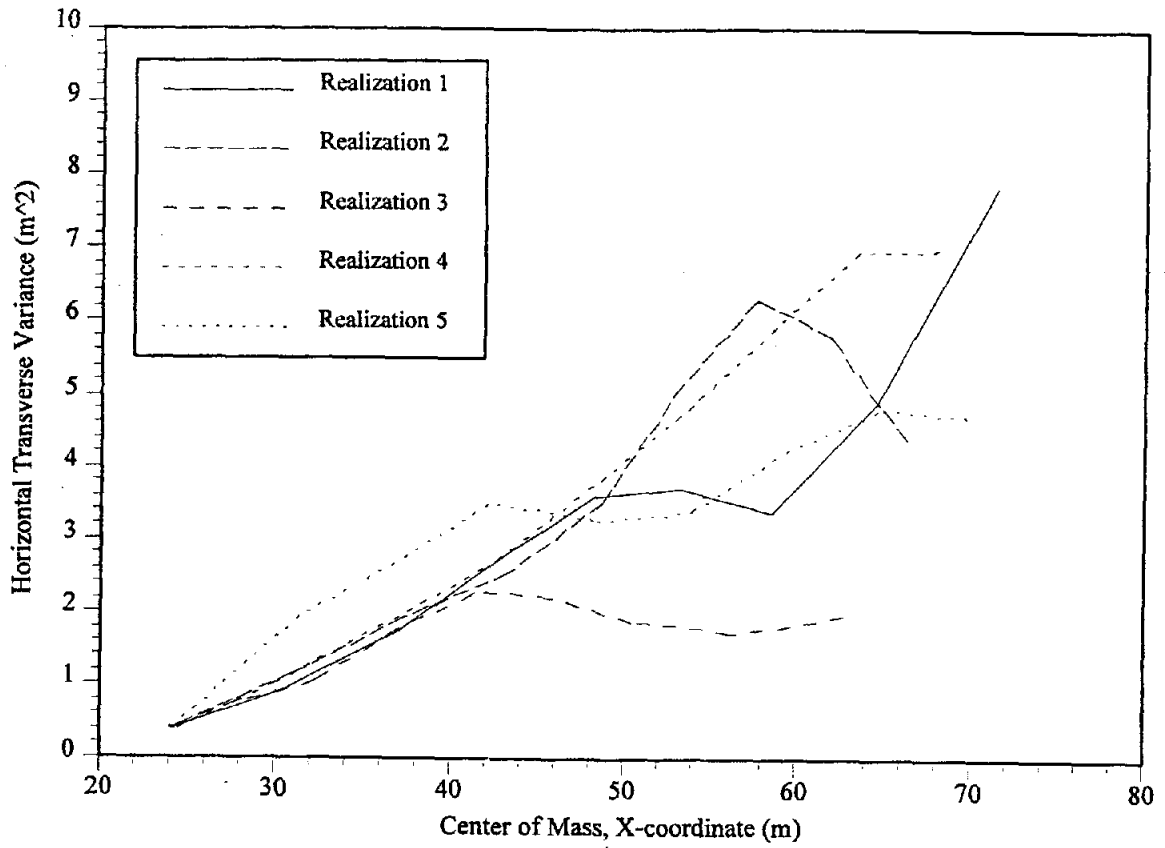
Reactive simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



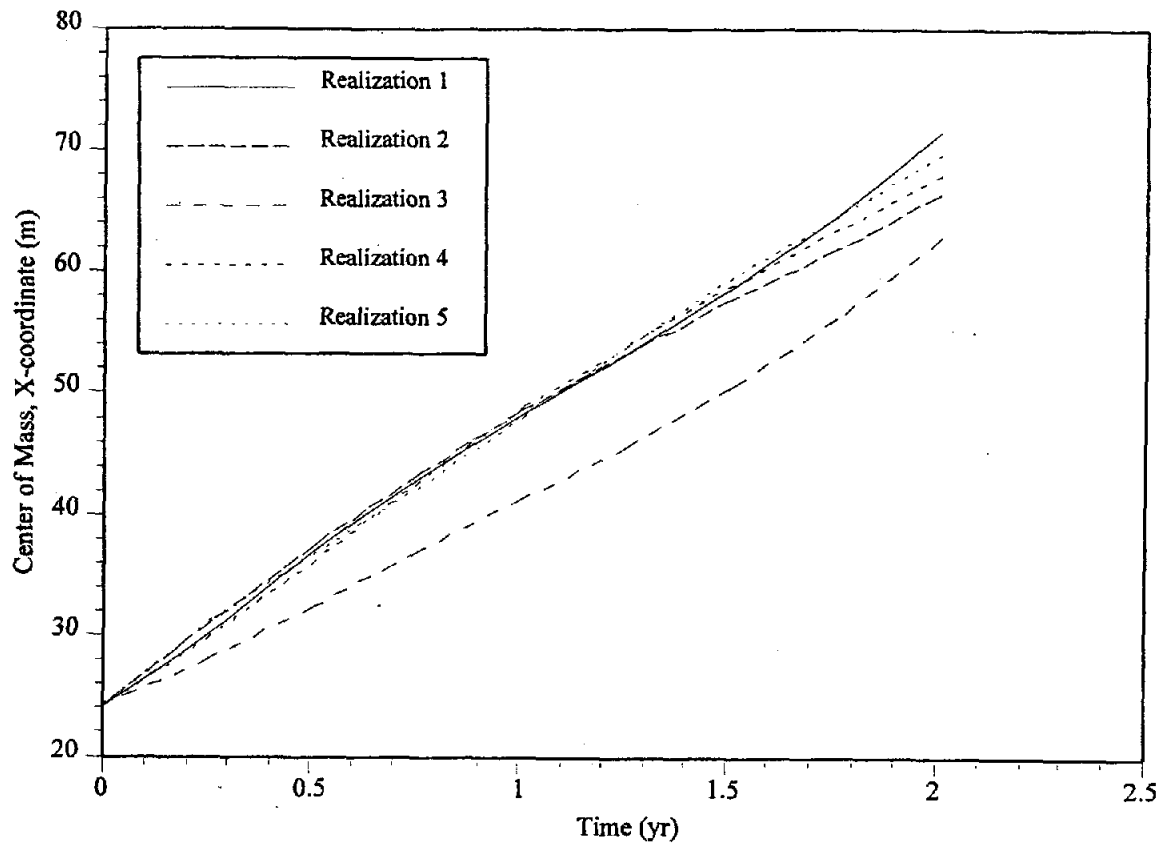
Reactive simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



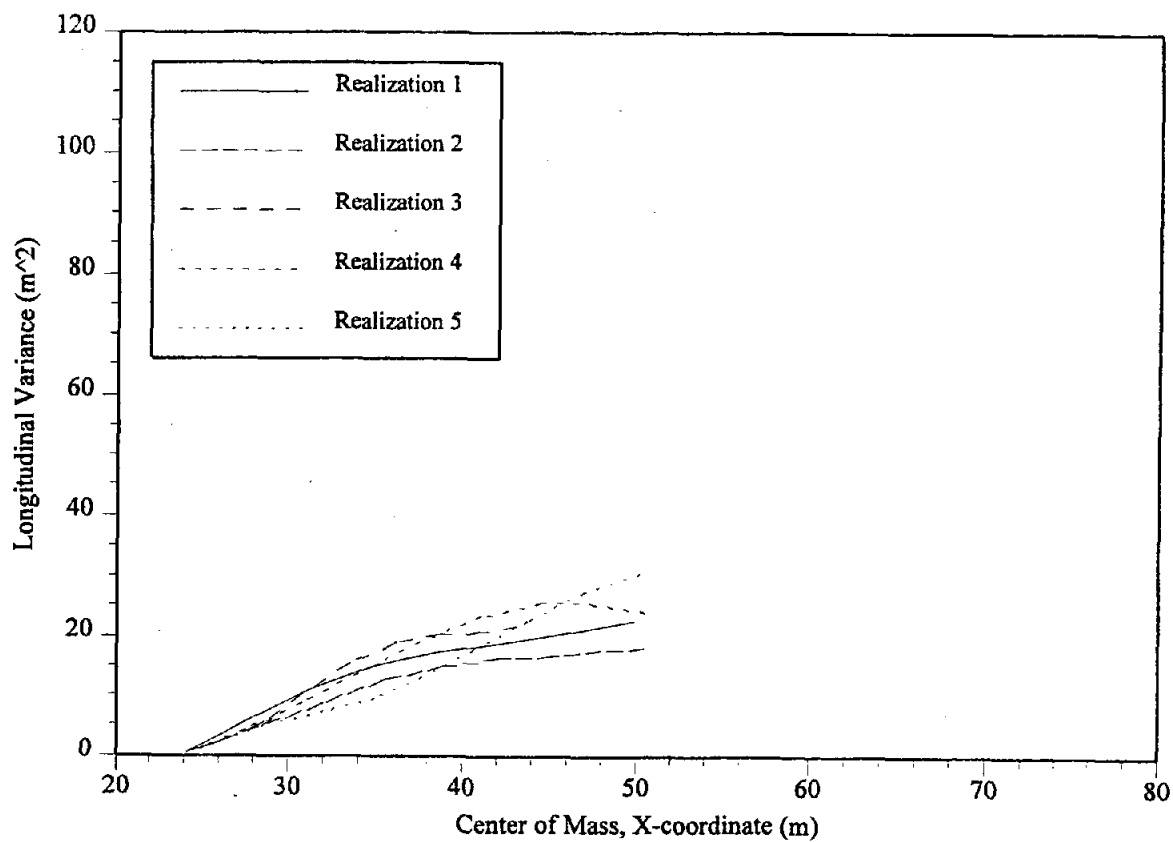
Conservative simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



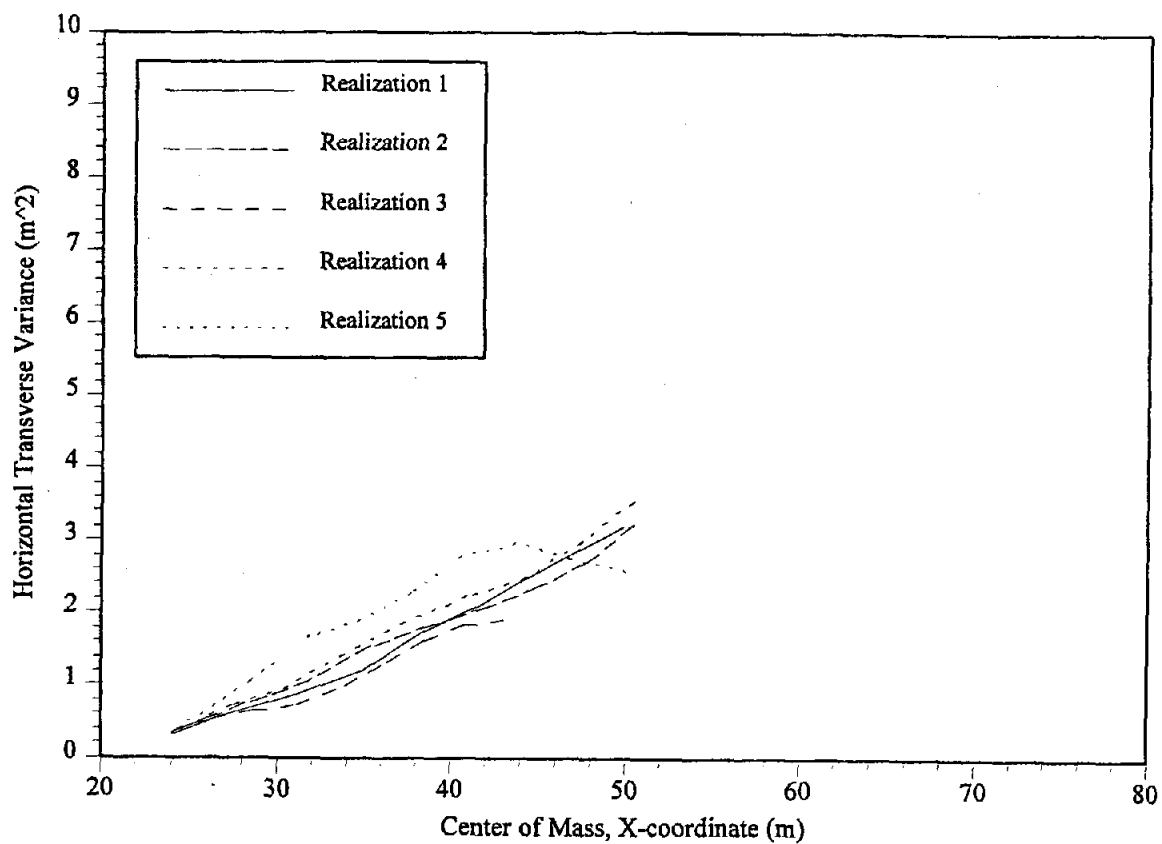
Conservative simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



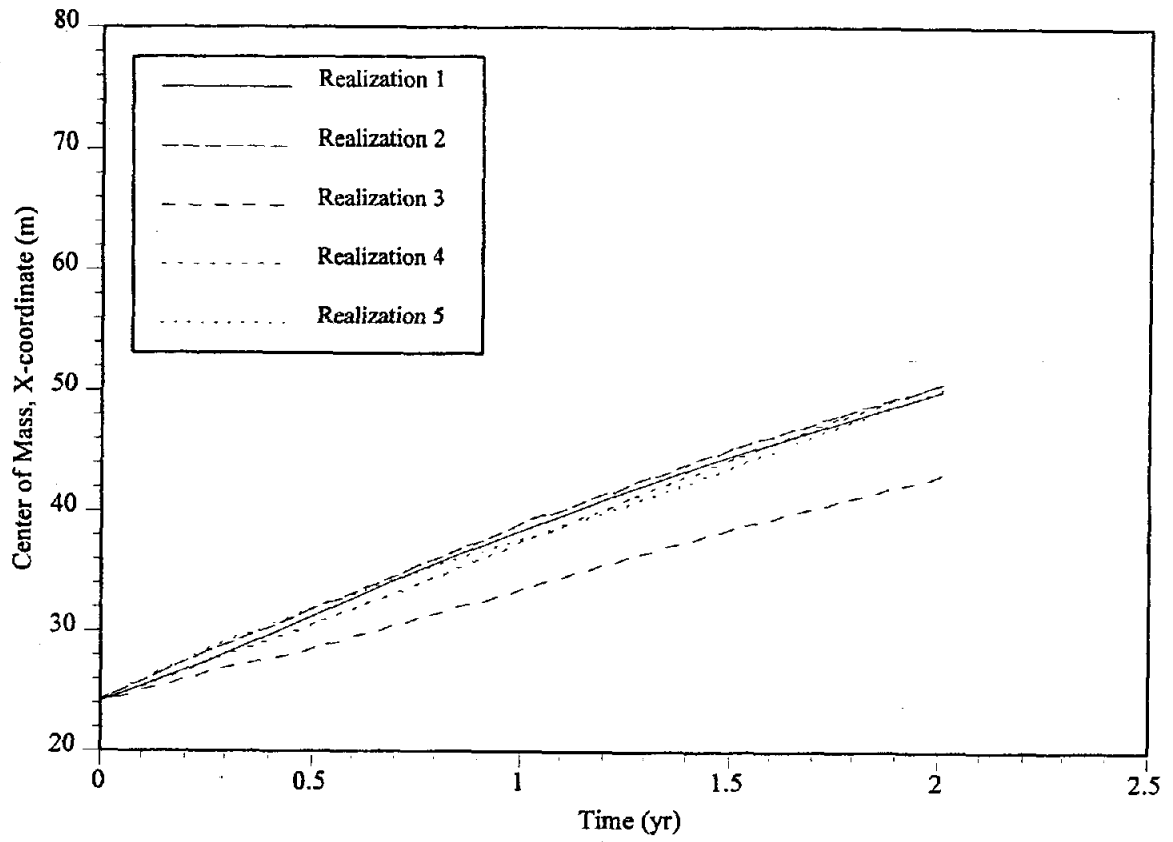
Conservative simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



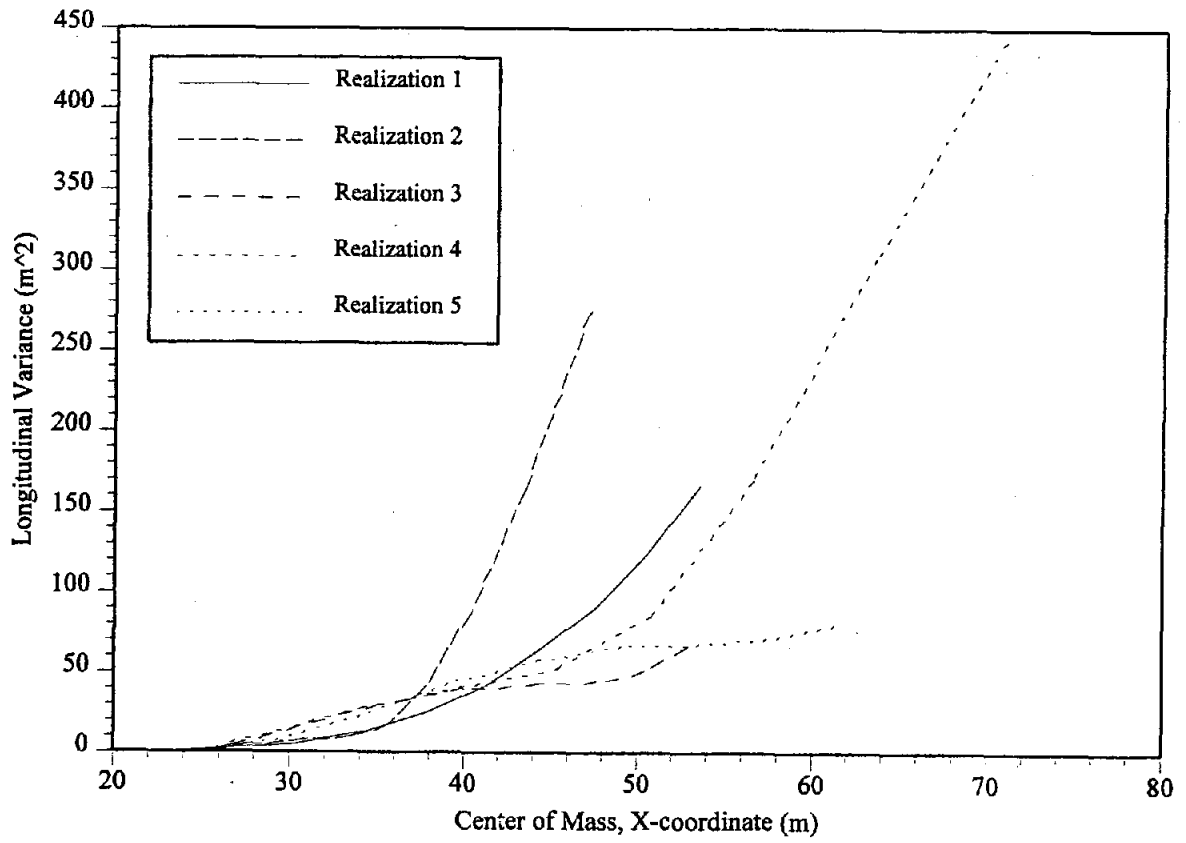
Reactive simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



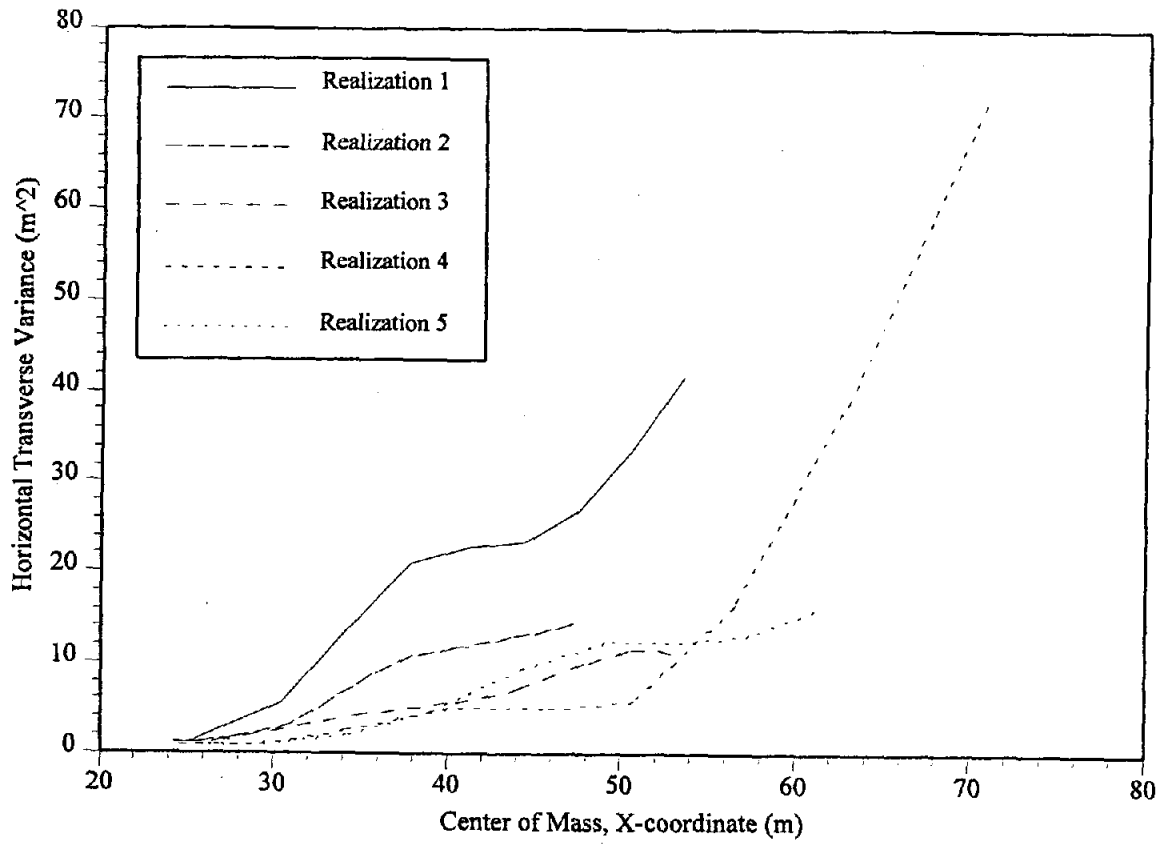
Reactive simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



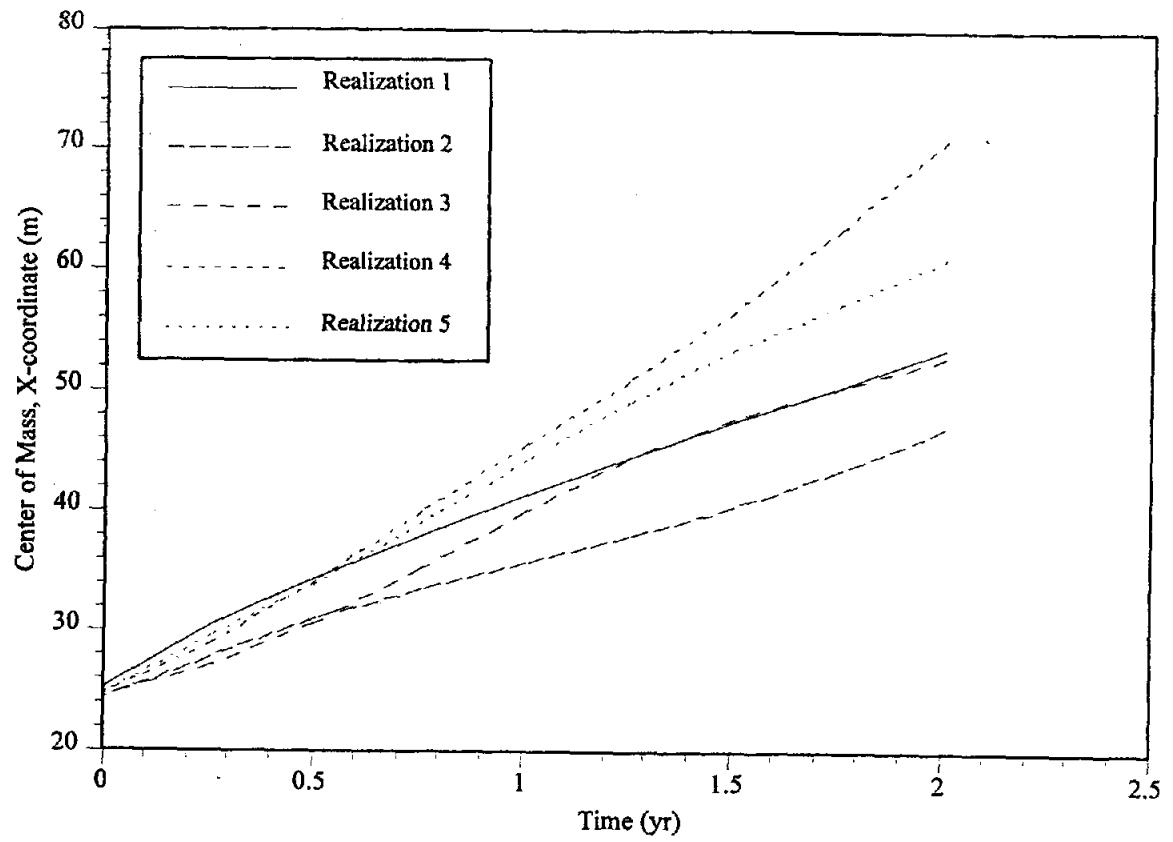
Reactive simulations in a weakly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



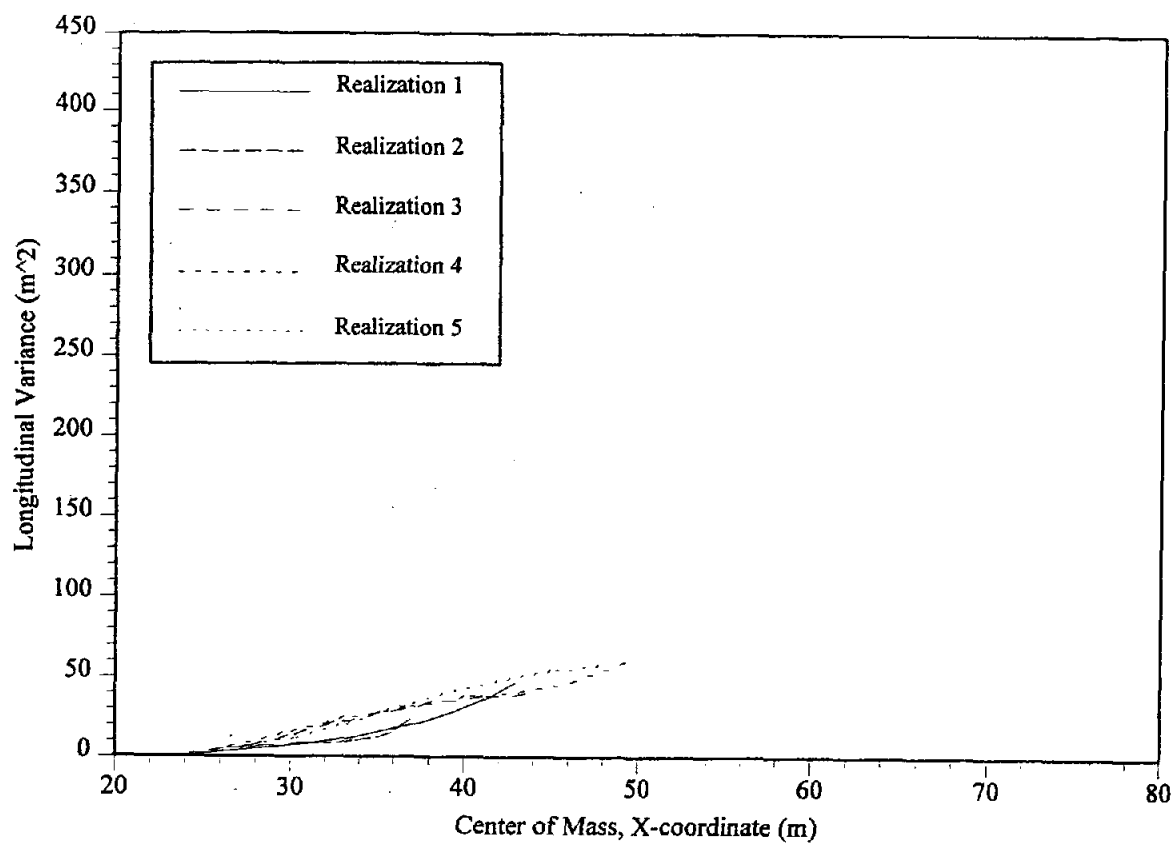
Conservative simulations in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



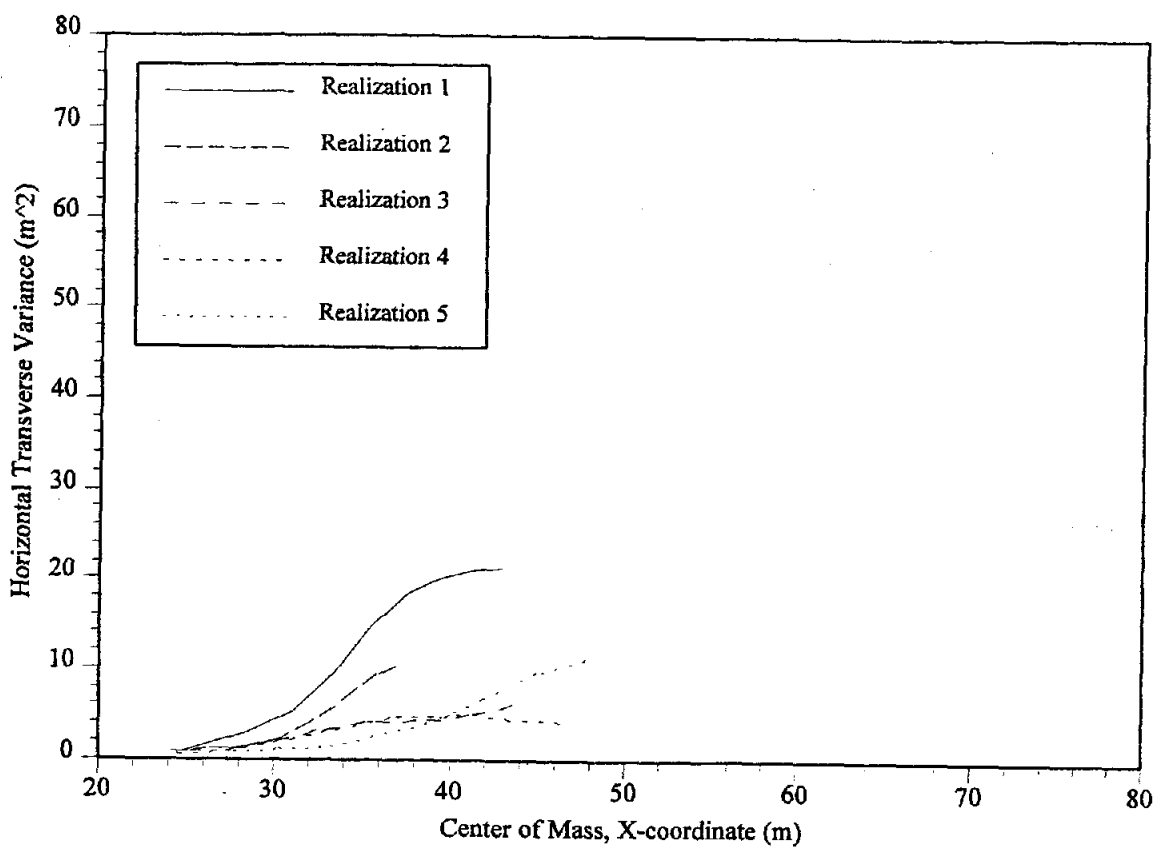
Conservative simulations in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



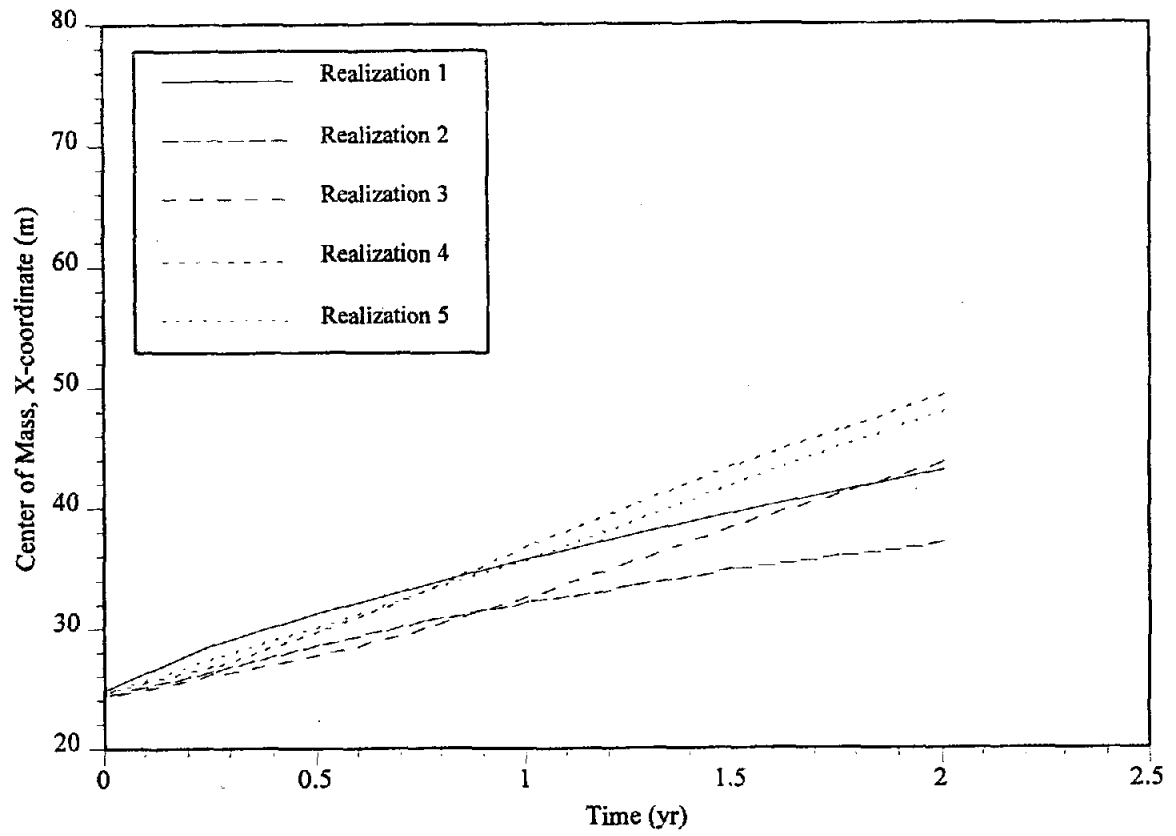
Conservative simulations in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



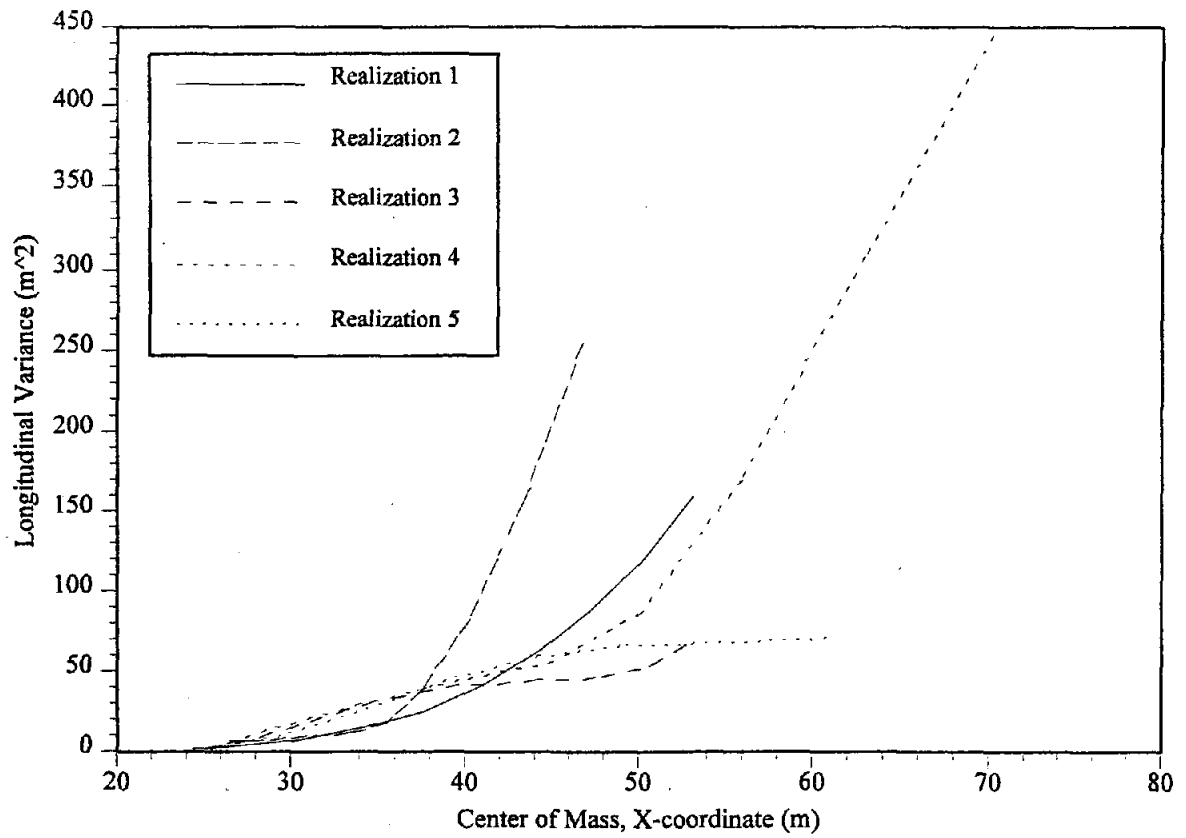
Reactive simulations in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



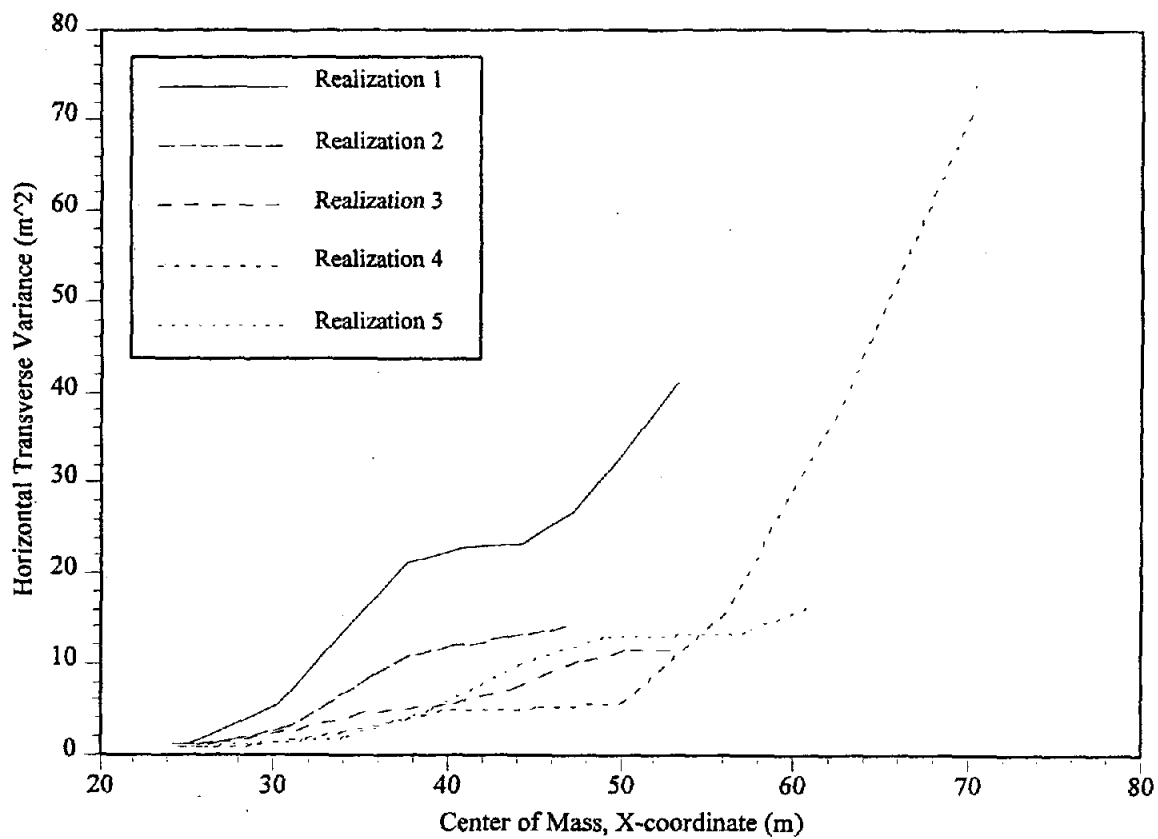
Reactive simulations in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



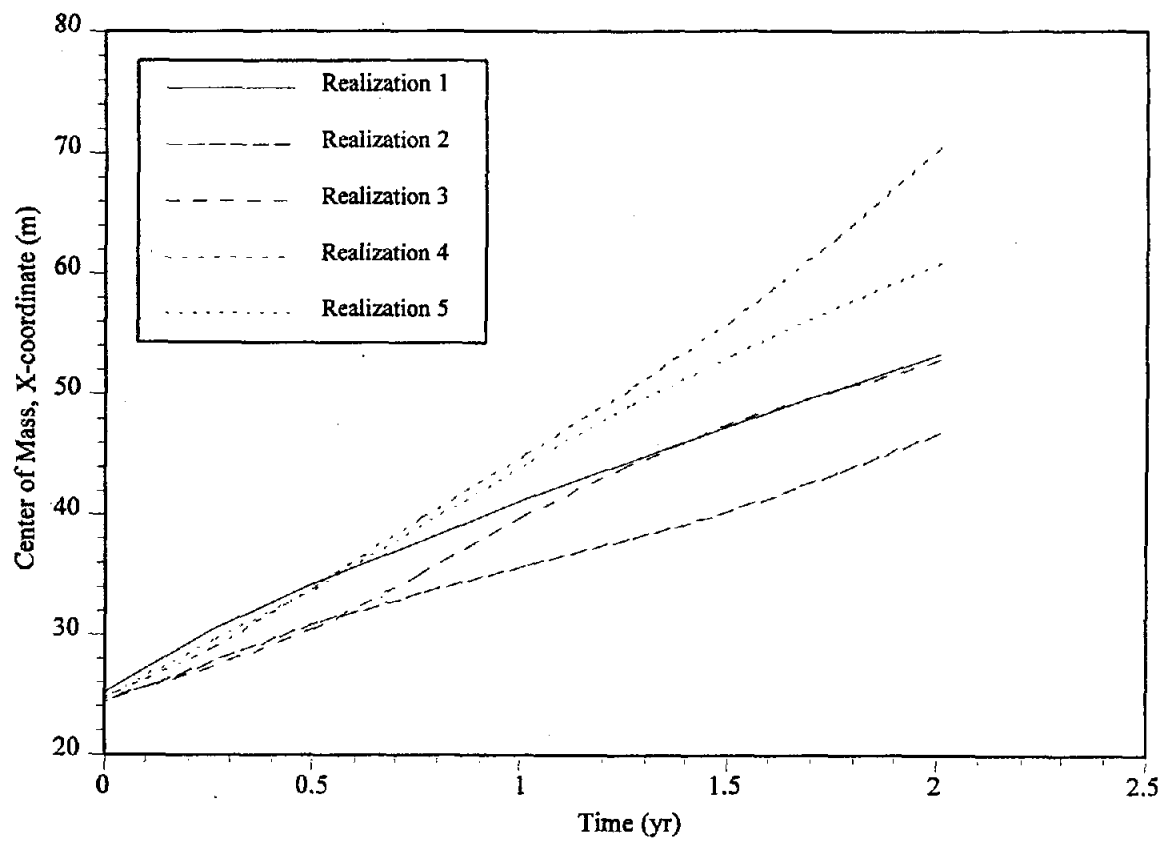
Reactive simulations in a strongly heterogeneous flow field with 1 kg of bromide. All concentrations used in calculations.



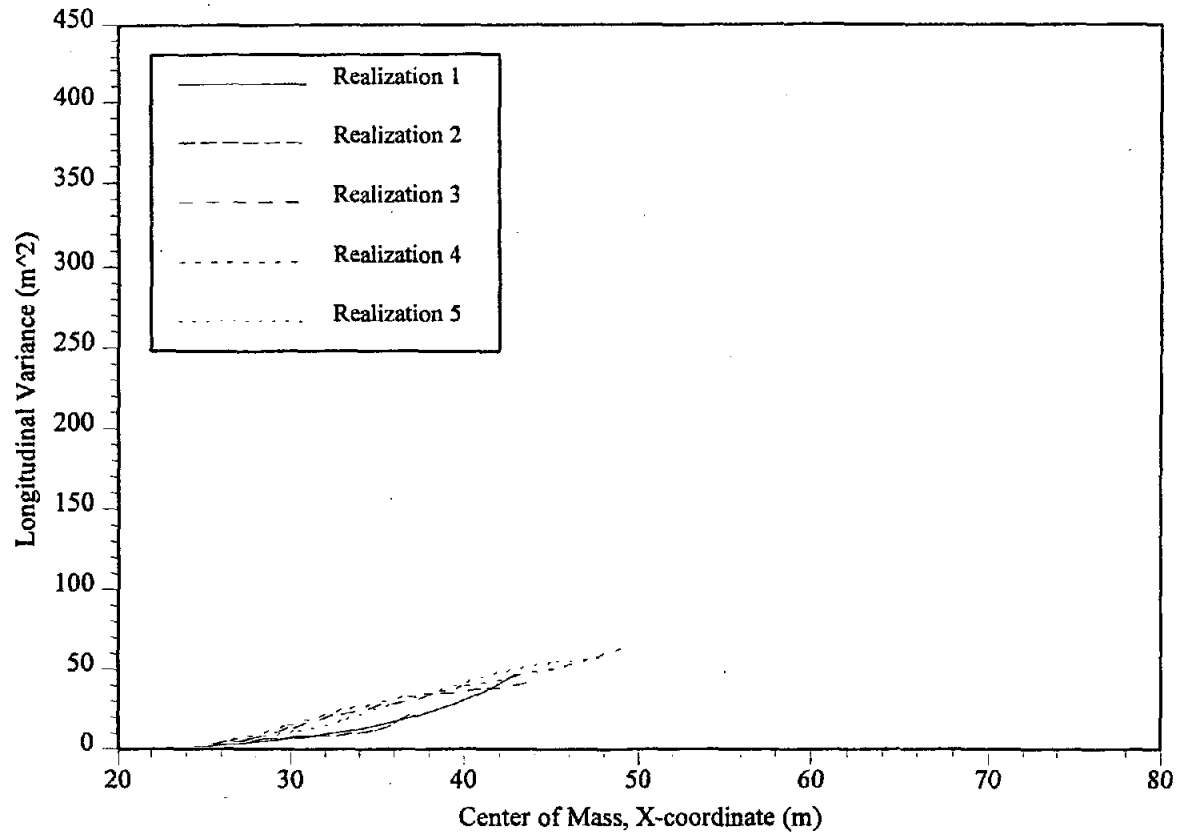
Conservative simulations in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



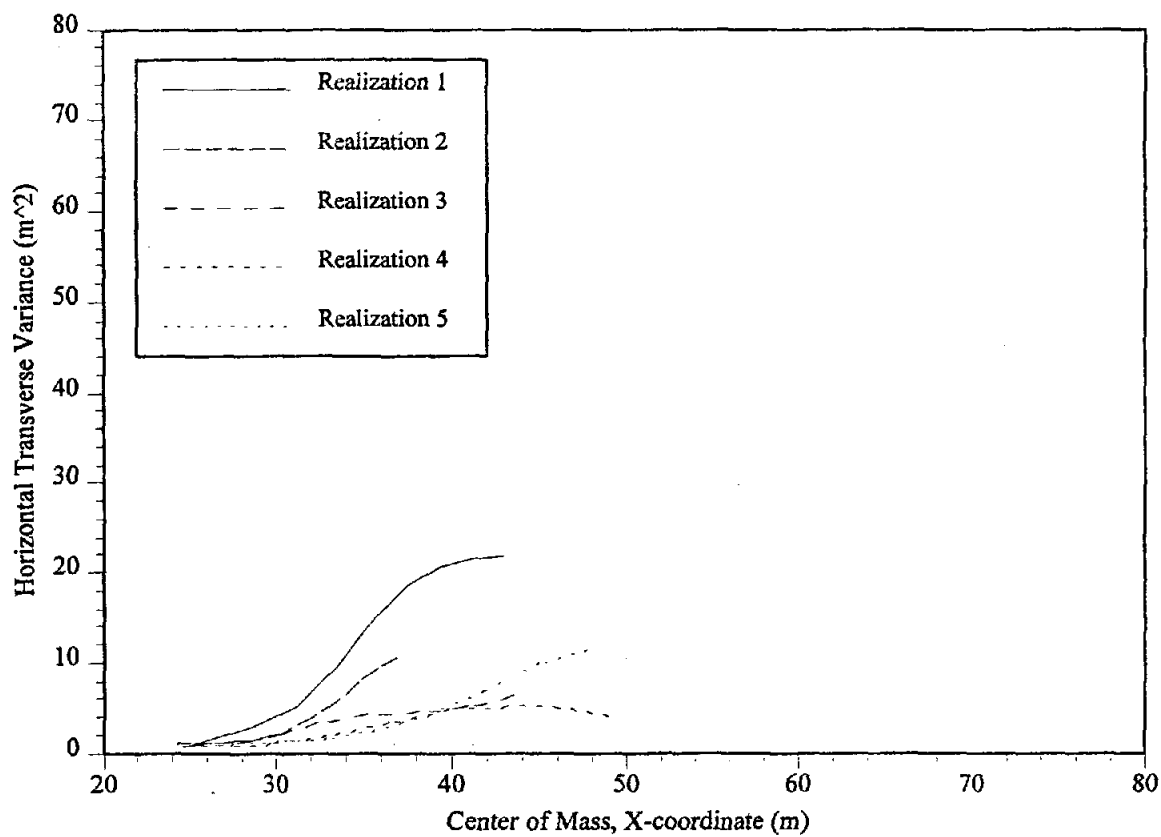
Conservative simulations in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



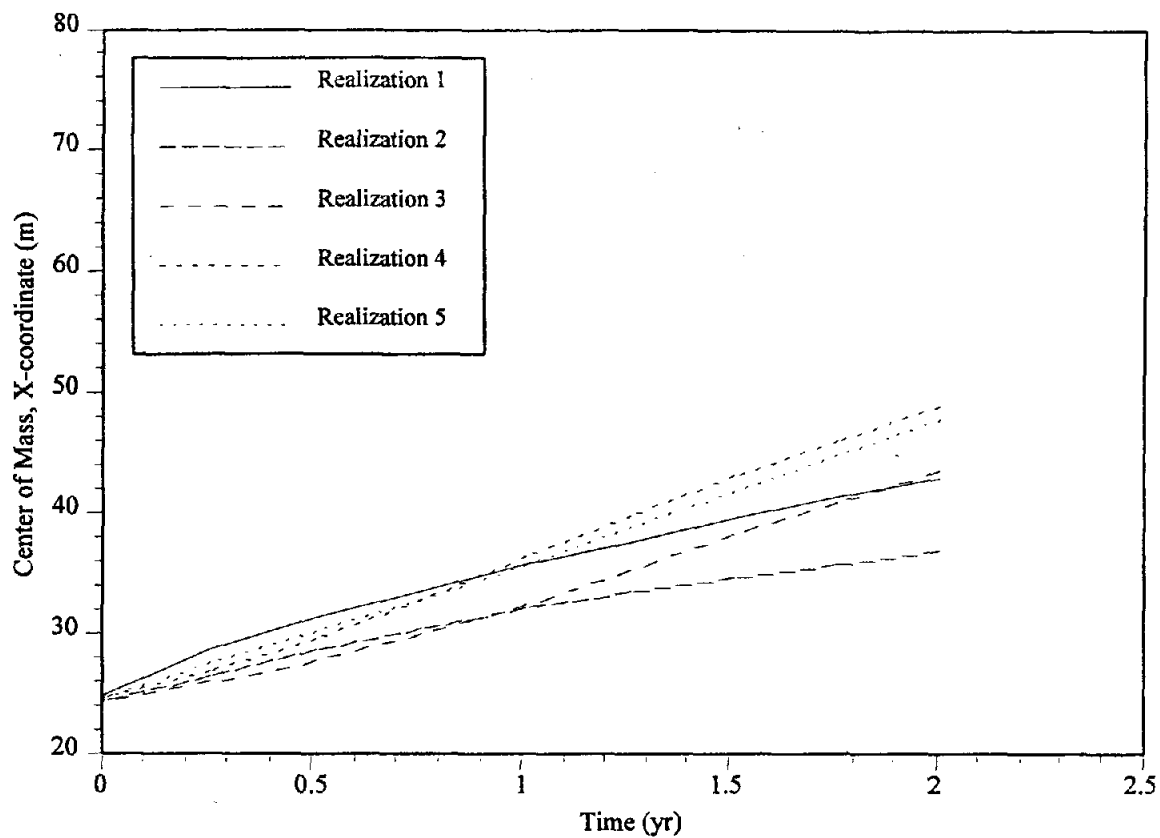
Conservative simulations in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



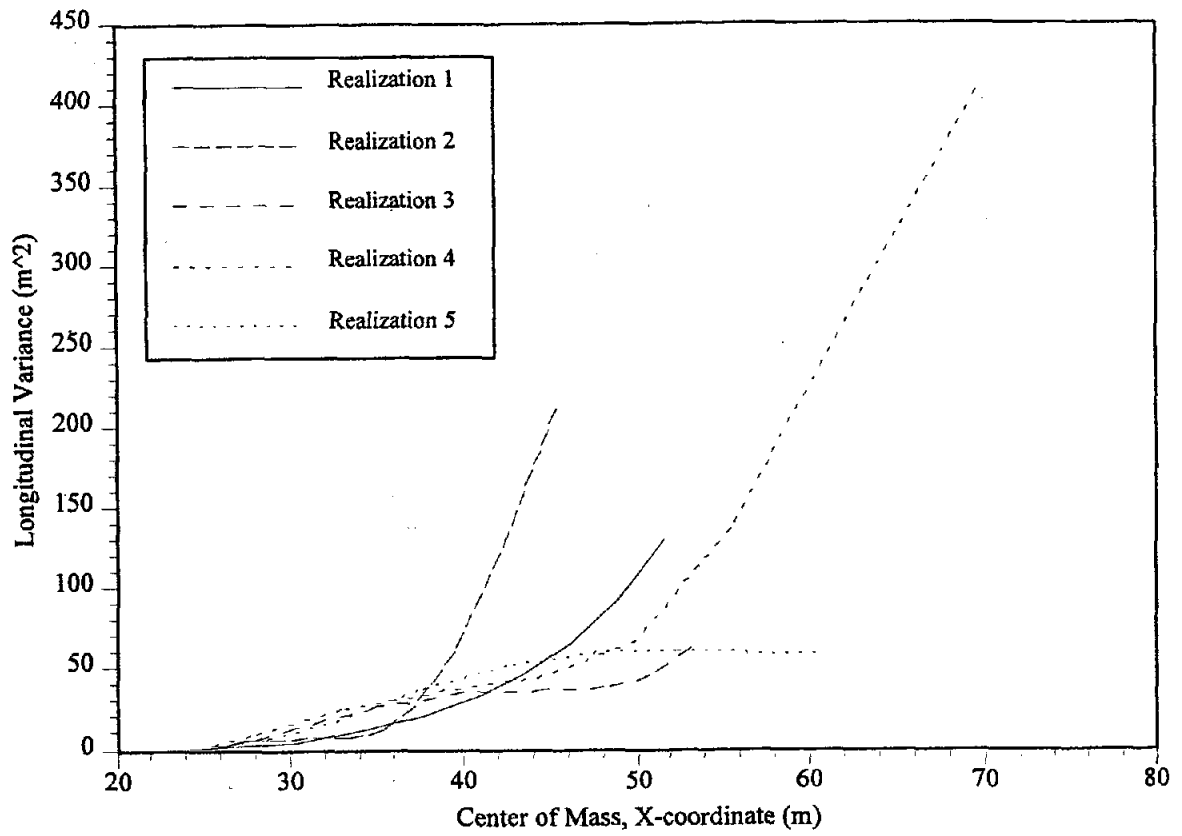
Reactive simulations in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



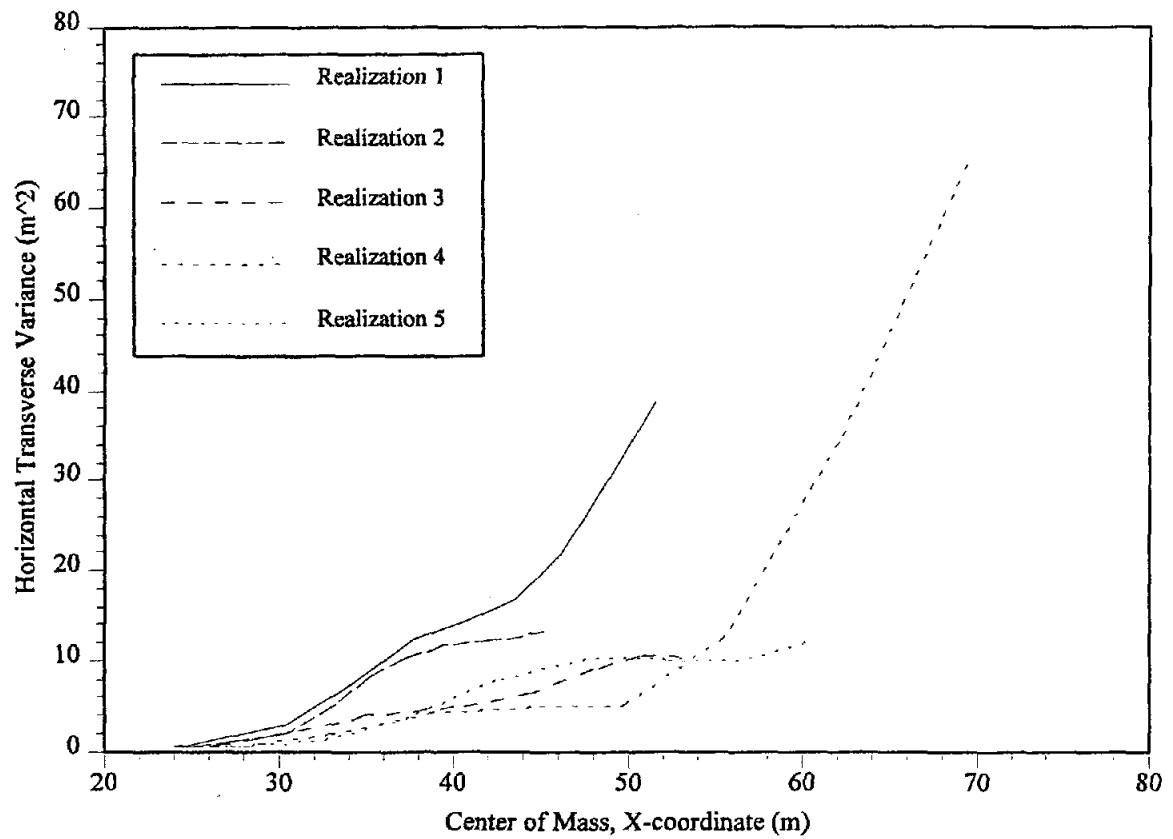
Reactive simulations in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



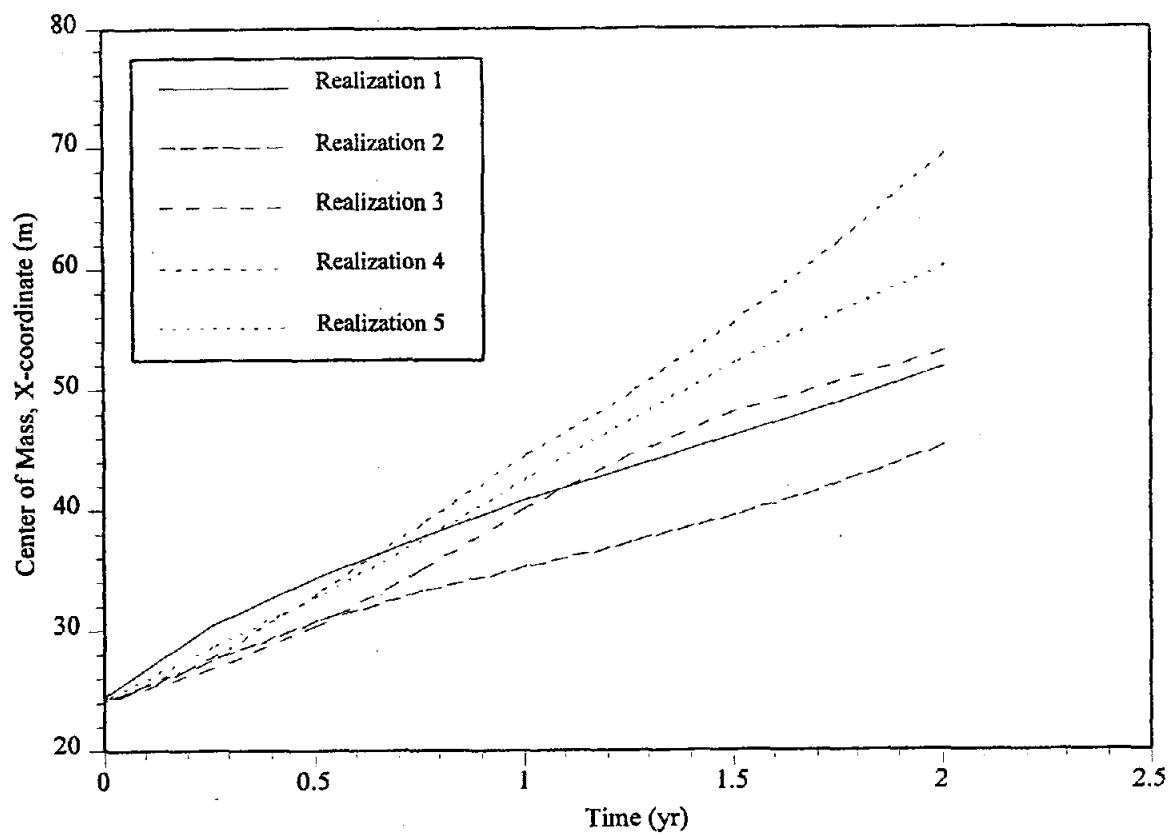
Reactive simulations in a strongly heterogeneous flow field with 1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



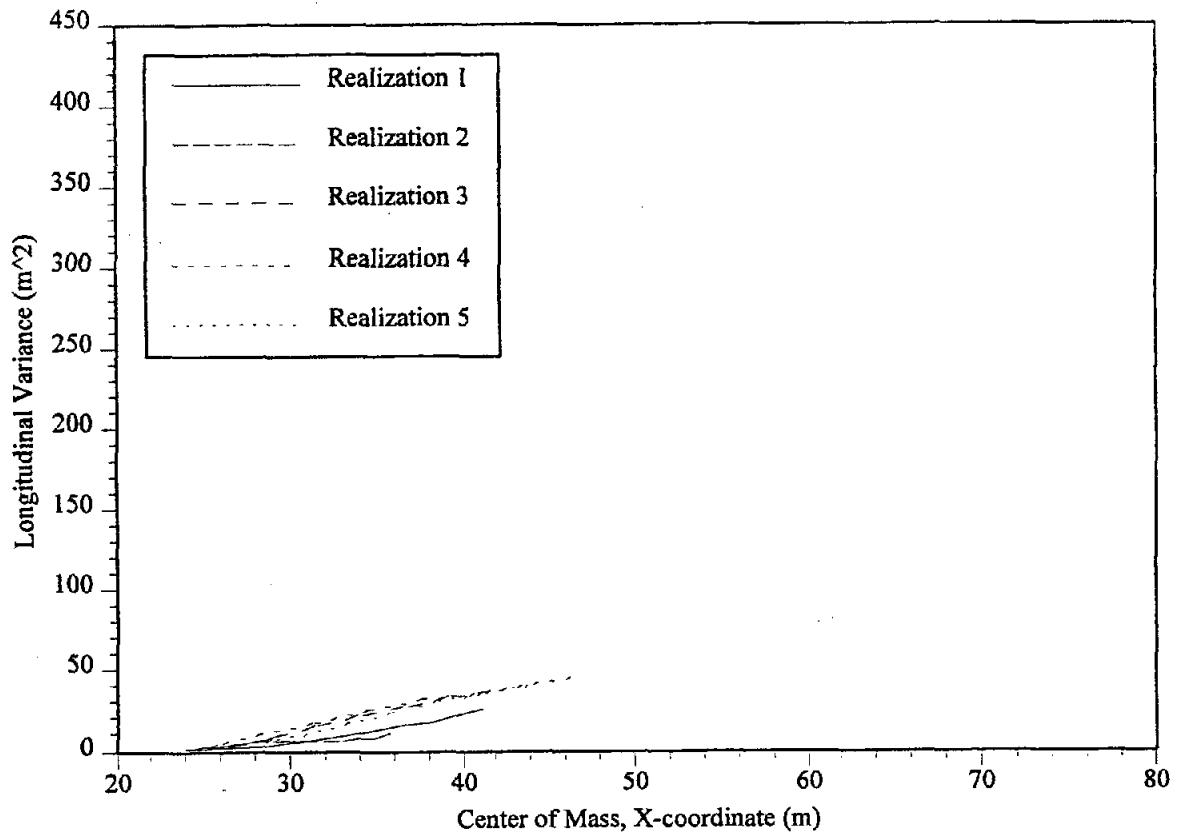
Conservative simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



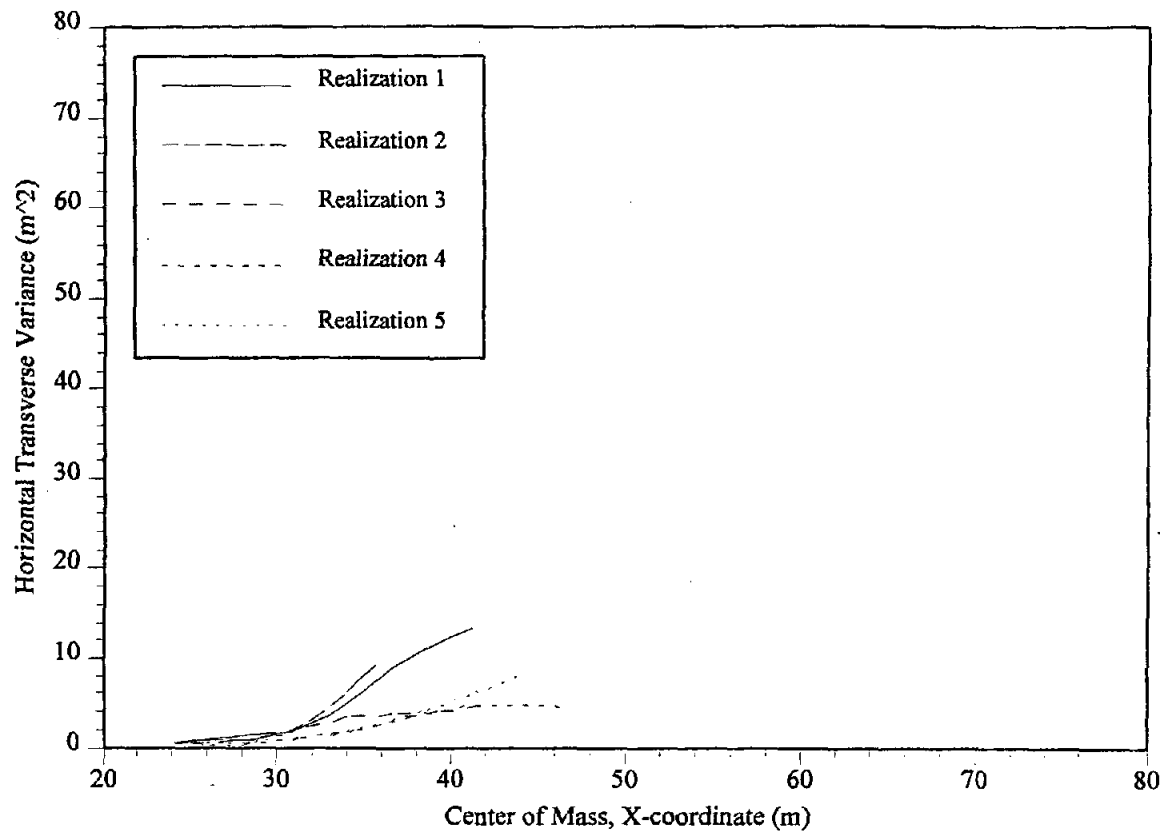
Conservative simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



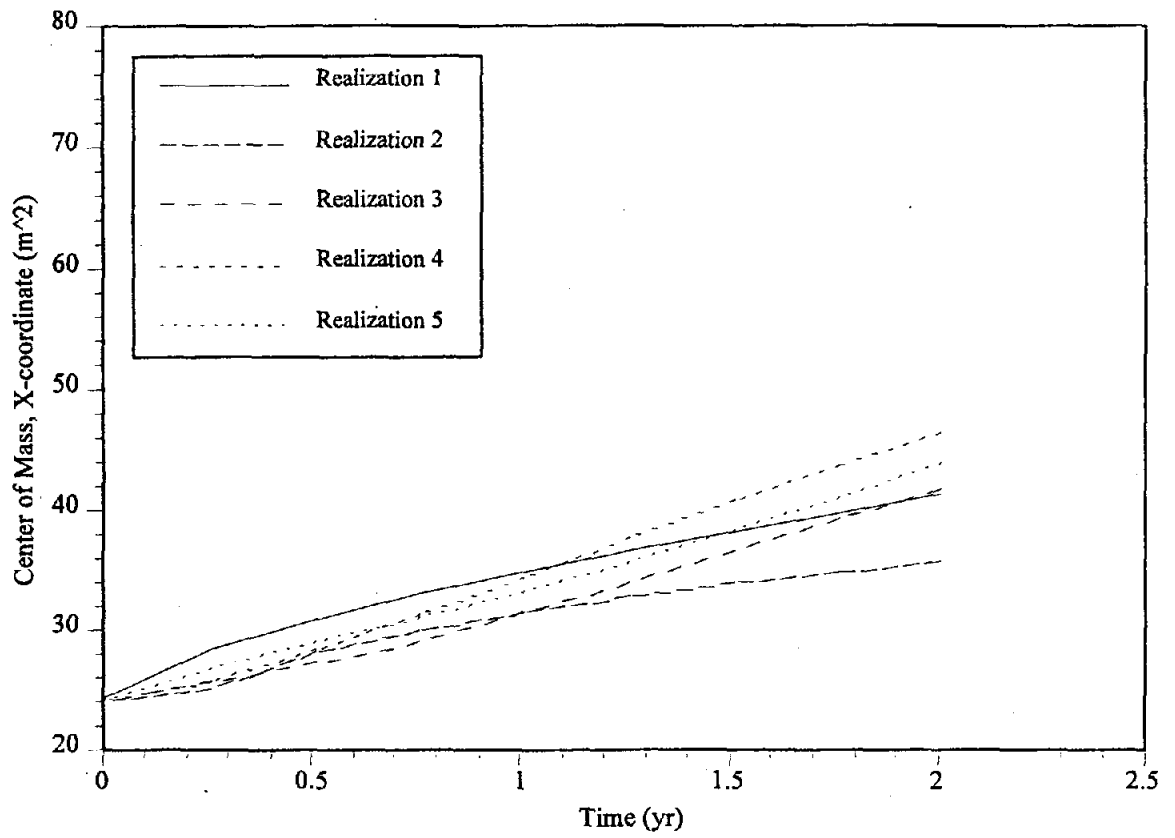
Conservative simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



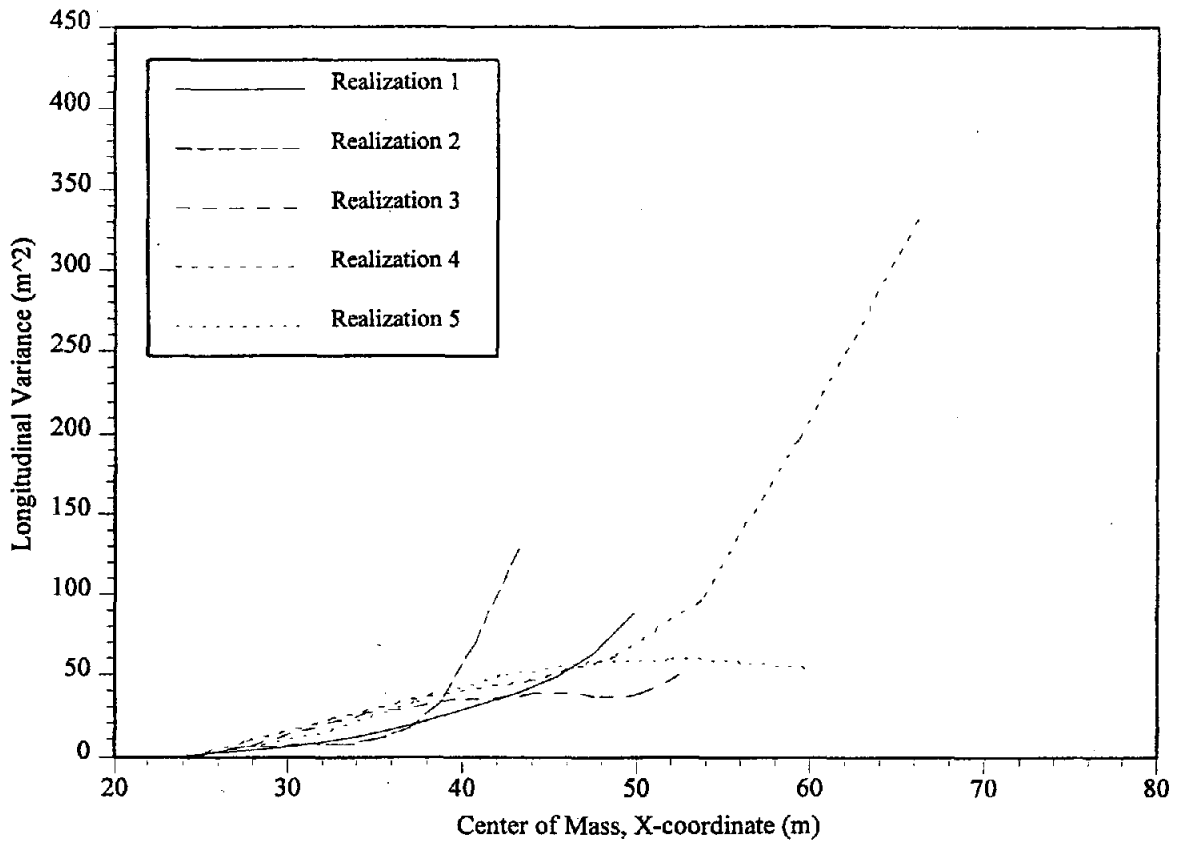
Reactive simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



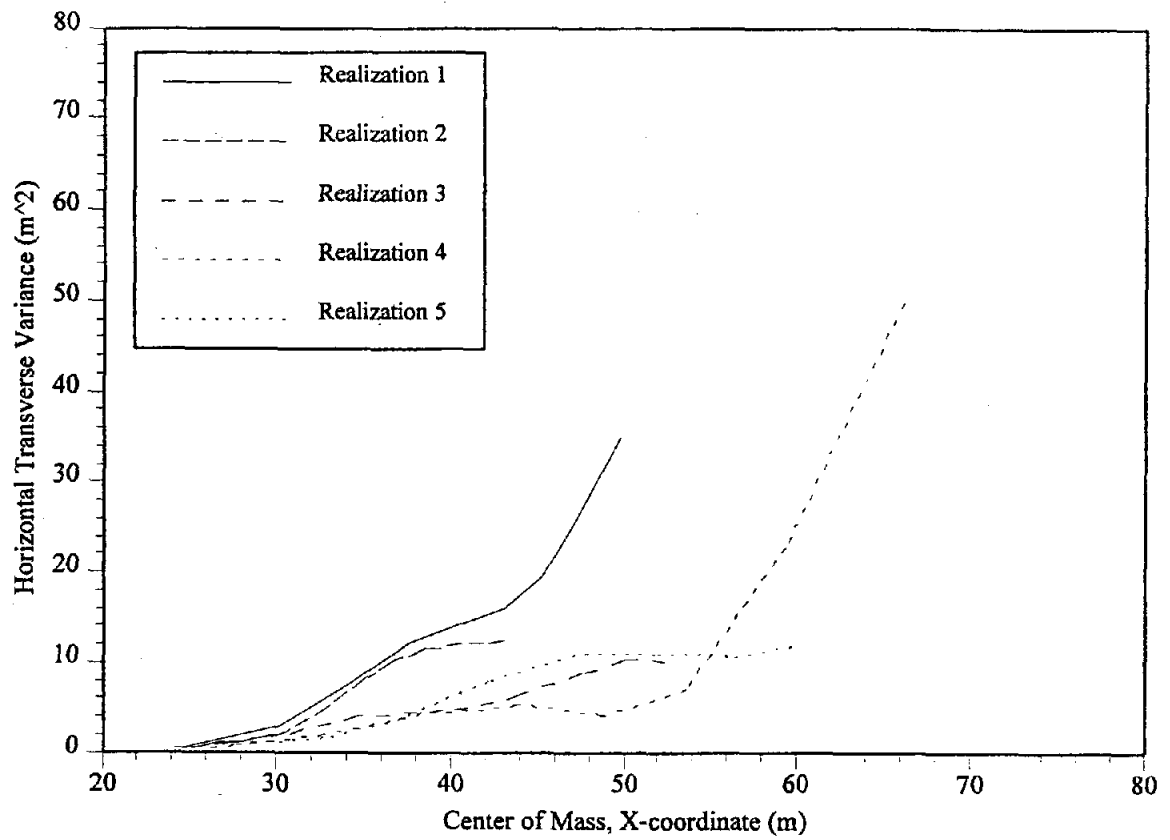
Reactive simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



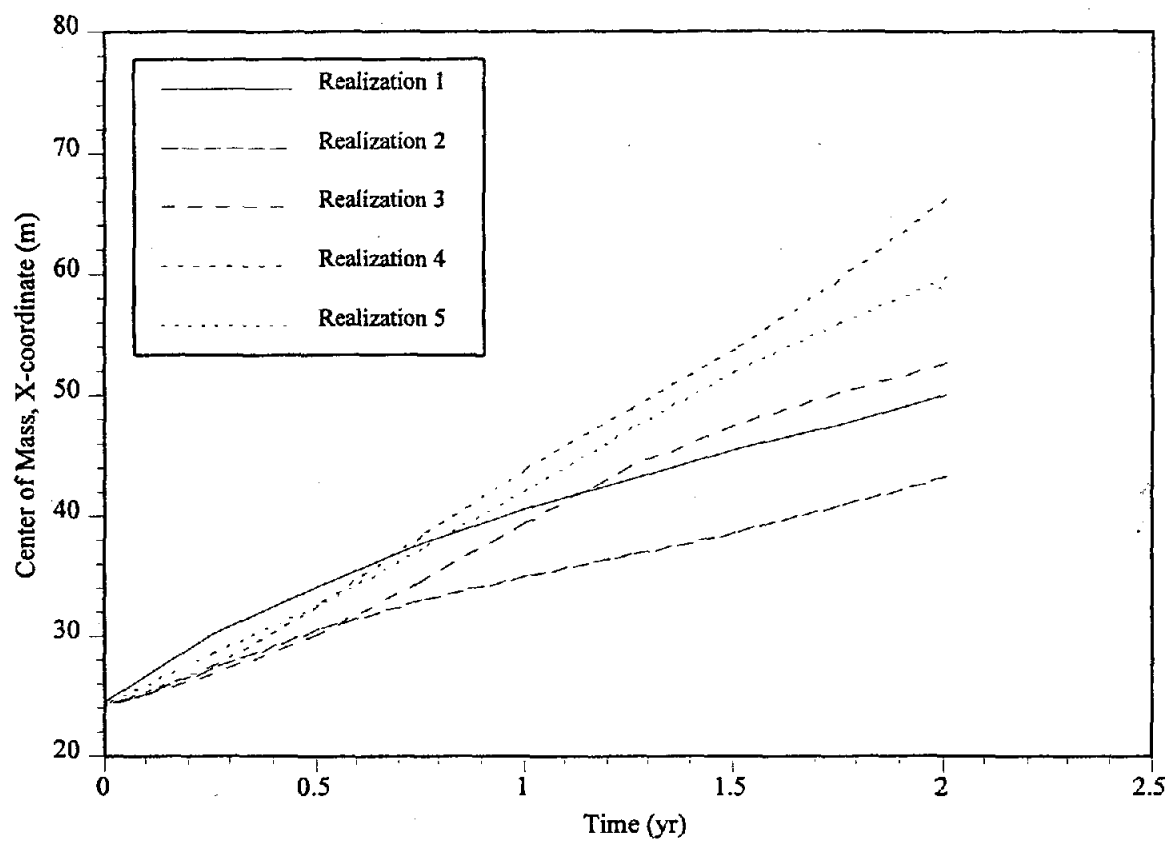
Reactive simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. All concentrations used in calculations.



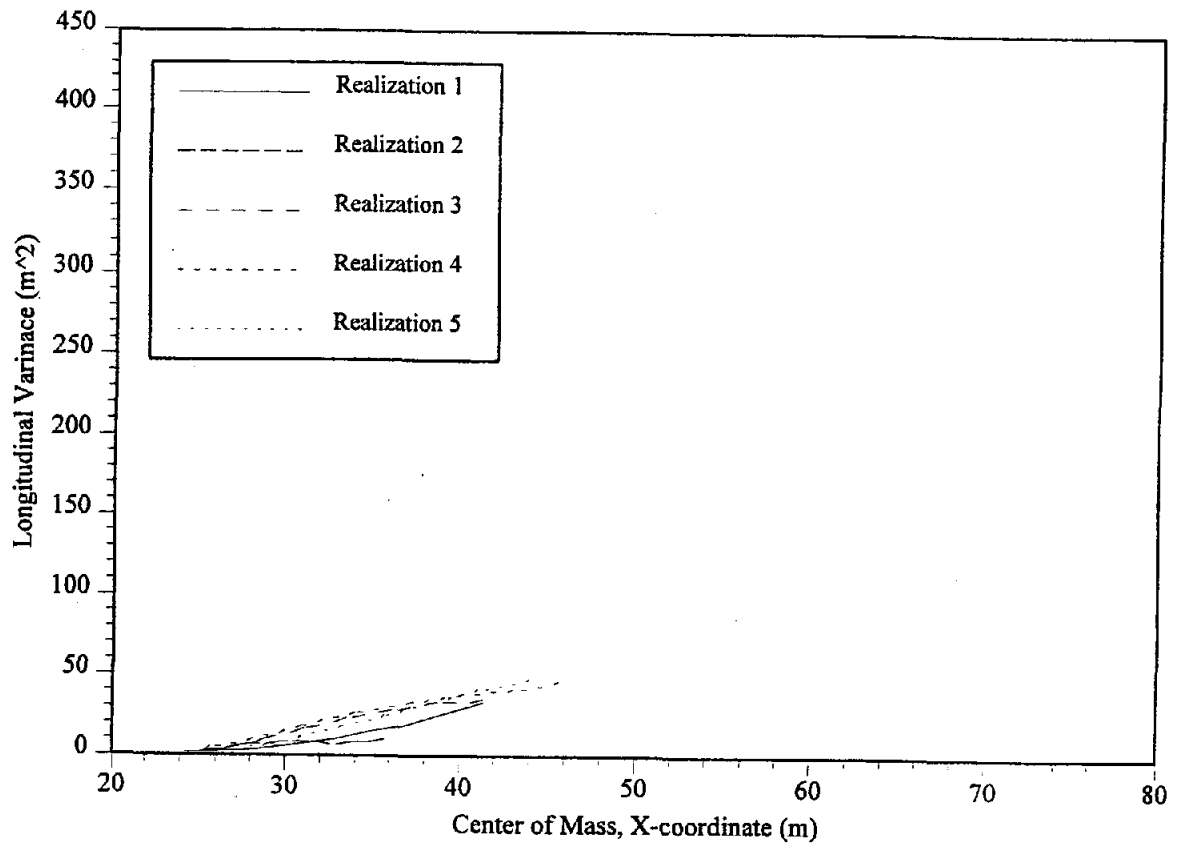
Conservative simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



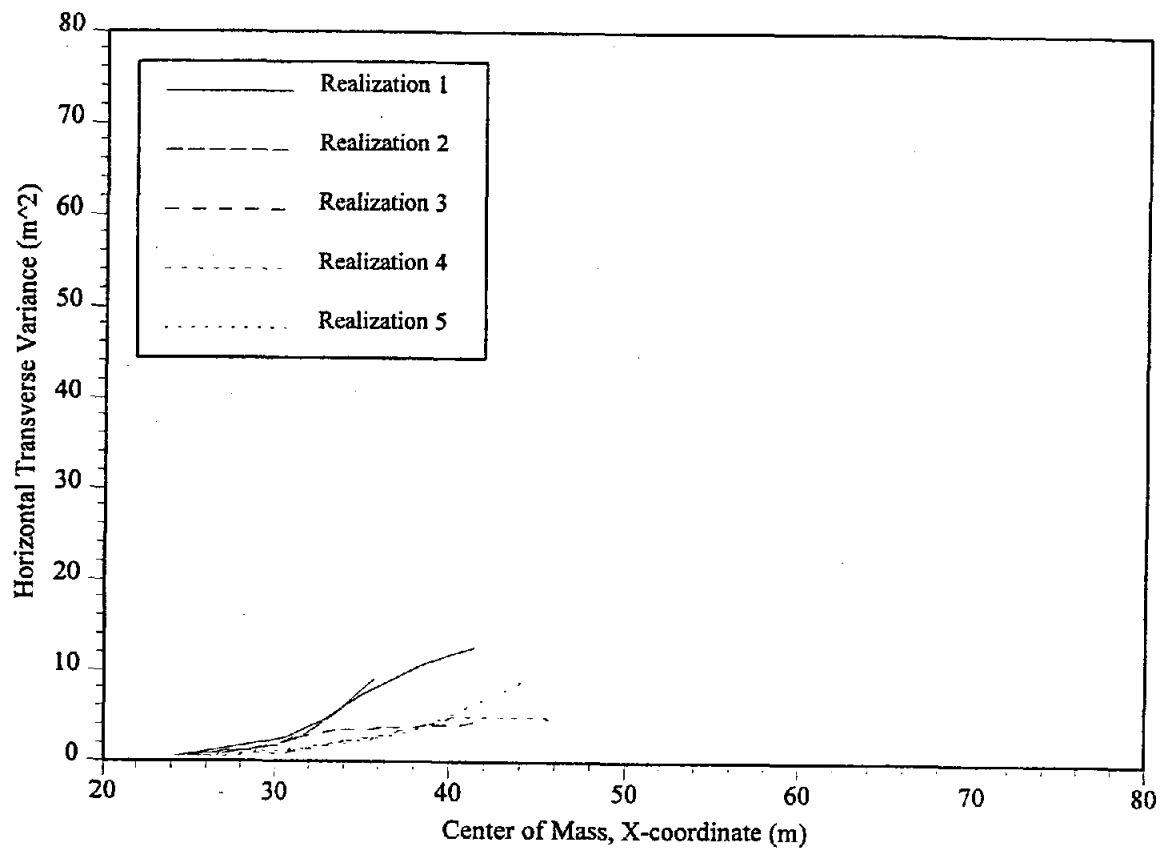
Conservative simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



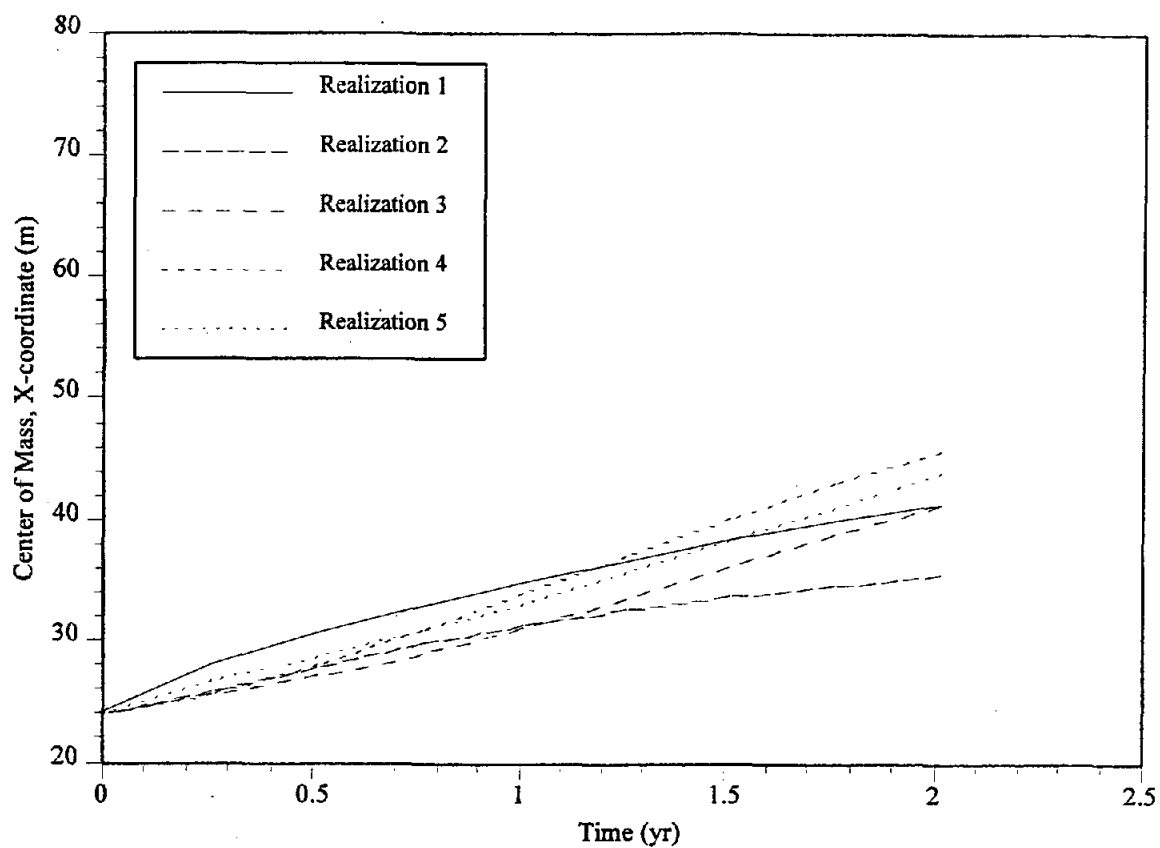
Conservative simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



Reactive simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



Reactive simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.



Reactive simulations in a strongly heterogeneous flow field with 0.1 kg of bromide. Only concentrations greater than 0.1 ppm used in calculations.

APPENDIX E

FORTRAN PROGRAMS

- * THIS PROGRAM TAKES THE RAW NODE CONCENTRATION OUTPUT FROM SUTRA AND
- * CALCULATES THE SPATIAL MOMENTS, ZERO TH THROUGH FOURTH.
- * A GRID FILE IS ALSO CREATED FOR CONTOURING THE RESULTS IN SURFER.
- * ONLY CONCENTRATIONS GREATER THAN 0.1 PPM ARE USED IN THE CALCULATIONS.
- * THIS PROGRAM IS DIMENSIONED FOR 9216 ELEMENTS (COLUMBUS & BORDEN AQUIFERS)
- * REVISED JULY 8, 1996

```

PROGRAM SURFHET
REAL M000,M100,M010,M110,M200,M020,M300,M030,M400,M040,
+ XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
DIMENSION C(9500),N(9500),CIJ(9500),XIJ(9500),YIJ(9500)
OPEN (UNIT=7,FILE='CONC.IN',STATUS='OLD')
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
OPEN (UNIT=9,FILE='MOMENTS',STATUS='UNKNOWN')
P=0.35
V=1
D=998.2
C
C REDUCE THE RAW SUTRA NODE CONCENTRATION OUTPUT FROM 6 COLUMNS TO 1
C
DO 10 K=1,9420,6
  READ (7,200)N(K),C(K),N(K+1),C(K+1),N(K+2),C(K+2),N(K+3),
+   C(K+3),N(K+4),C(K+4),N(K+5),C(K+5)
200  FORMAT (7X,I5,G16.8,I5,G16.8,I5,G16.8,I5,G16.8,I5,
+   G16.8)
10  CONTINUE
  READ (7,300)N(9421),C(9421),N(9422),C(9422),N(9423),C(9423),
+   N(9424),C(9424),N(9425),C(9425)
300  FORMAT (7X,I5,G16.8,I5,G16.8,I5,G16.8,I5,G16.8,I5,G16.8)
C
C AVERAGE THE 4 NODE CONCENTRATIONS AROUND EACH ELEMENT TO PERFORM
C THE MOMENT CALCULATIONS ON THE ELEMENTS (RATHER THAN THE NODES) AND
C CALCULATE THE X AND Y-COORDINATES OF EACH ELEMENT
C
DO 20 I=1,144,1
DO 30 J=1,64,1
  C1=((I-1)*65+J)
  C2=(I*65+J)
  C3=(I*65+1+J)
  C4=((I-1)*65+1+J)
  CIJ(K)=(C(C1)+C(C2)+C(C3)+C(C4))/4
  IF (CIJ(K).LT.1.0E-7) THEN
    CIJ(K)=0.0
  ENDIF
  XIJ(K)=(I-0.5)
  YIJ(K)=(J-0.5)
  WRITE (8,400)XIJ(K),YIJ(K),CIJ(K)
400  FORMAT (E16.8,E16.8,E16.8)
30  CONTINUE
20  CONTINUE
C
C READ IN THE ELEMENT CONCENTRATIONS AND COORDINATES CALCULATED ABOVE
C
CLOSE (8)
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
DO 35 K=1,9216,1

```

Program SURFHET (continued)

```

      READ (8,400)XIJ(K),YIJ(K),CIJ(K)
35  CONTINUE
C
C ZEROTH MOMENT (TOTAL MASS)
C
      SUM=0.0
      DO 40 K=1,9216,1
          SUM=SUM+CIJ(K)*P*V*D
40  CONTINUE
      M000=SUM
C
C FIRST MOMENT, X (CENTER OF MASS IN THE X DIRECTION)
C
      SUM=0.0
      DO 50 K=1,9216,1
          SUM=SUM+CIJ(K)*P*V*D*XIJ(K)
50  CONTINUE
      M100=SUM
      XC=M100/M000
C
C FIRST MOMENT, Y (CENTER OF MASS IN THE Y DIRECTION)
C
      SUM=0.0
      DO 55 K=1,9216,1
          SUM=SUM+CIJ(K)*P*V*D*YIJ(K)
55  CONTINUE
      M010=SUM
      YC=M010/M000
C
C SECOND MOMENT, XX (SPATIAL VARIANCE IN THE X DIRECTION)
C
      SUM=0.0
      DO 60 K=1,9216,1
          SUM=SUM+CIJ(K)*P*V*D*(XIJ(K)**2)
60  CONTINUE
      M200=SUM
      SIGMAXX=(M200/M000)-(XC**2)
C
C SECOND MOMENT, YY (SPATIAL VARIANCE IN THE Y DIRECTION)
C
      SUM=0.0
      DO 70 K=1,9216,1
          SUM=SUM+CIJ(K)*P*V*D*(YIJ(K)**2)
70  CONTINUE
      M020=SUM
      SIGMAYY=(M020/M000)-YC**2
C
C SECOND MOMENT, XY (SPATIAL COVARIANCE)
C
      SUM=0.0
      DO 80 K=1,9216,1
          SUM=SUM+CIJ(K)*P*V*D*XIJ(K)*YIJ(K)
80  CONTINUE
      M110=SUM
      SIGMAXY=(M110/M000)-(XC*YC)

```

Program SURFHET (continued)

C THIRD MOMENT, X (SKEWNESS OF THE PLUME IN THE X DIRECTION)

```
C
  SUM=0.0
  DO 90 K=1,9216,1
    SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**3)
90  CONTINUE
  M300=SUM
  SKX=M300/(M000*(SIGMAXX**(3/2)))
```

C THIRD MOMENT, Y (SKEWNESS OF THE PLUME IN THE Y DIRECTION)

```
C
  SUM=0.0
  DO 100 K=1,9216,1
    SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**3)
100 CONTINUE
  M030=SUM
  SKY=M030/(M000*(SIGMAYY**(3/2)))
```

C FOURTH MOMENT, X (KURTOSIS OF THE PLUME IN THE X DIRECTION)

```
C
  SUM=0.0
  DO 110 K=1,9216,1
    SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**4)
110 CONTINUE
  M400=SUM
  KRX=M400/(M000*(SIGMAXX**2))
```

C FOURTH MOMENT, Y (KURTOSIS OF THE PLUME IN THE Y DIRECTION)

```
  SUM=0.0
  DO 120 K=1,9216,1
    SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**4)
120 CONTINUE
  M040=SUM
  KRY=M040/(M000*(SIGMAYY**2))
  WRITE (9,500)M000,XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
500 FORMAT (//,'****SPATIAL MOMENT CALCULATIONS****',//,
  A 'LATIONS****',//,
  B '#####',
  C '# CONCENTRATIONS < 0.1 PPM OMITTED #',//,
  D '#####',
  E 'MASS OF BROMIDE (KG)'/2X,'M000=',G14.7,//,
  F 'CENTER OF MASS, X-COORDINATE (M)'/2X,'XC=',G14.7,//,
  G 'CENTER OF MASS, Y-COORDINATE (M)'/2X,'YC=',G14.7,//,
  H 'SPATIAL VARIANCE, XX'/2X,'SIGMAXX=',G14.7,//,
  I 'SPATIAL VARIANCE, YY'/2X,'SIGMAYY=',G14.7,//,
  J 'SPATIAL COVARIANCE, XY'/2X,'SIGMAXY=',G14.7,//,
  K 'SKEWNESS OF THE PLUME, X'/2X,'SKX=',G14.7,//,
  L 'SKEWNESS OF THE PLUME, Y'/2X,'SKY=',G14.7,//,
  M 'KURTOSIS OF THE PLUME, X'/2X,'KRX=',G14.7,//,
  N 'KURTOSIS OF THE PLUME, Y'/2X,'KRY=',G14.7)
```

CLOSE (7)

CLOSE (8)

CLOSE (9)

STOP

END

* THIS PROGRAM TAKES THE RAW NODE CONCENTRATION OUTPUT FROM SUTRA AND
 * CALCULATES THE SPATIAL MOMENTS, ZERO TH THROUGH FOURTH. A GRID FILE IS
 * ALSO CREATED FOR CONTOURING THE RESULTS IN SURFER. ALL CONCENTRATIONS
 * ARE USED IN THE CALCULATIONS. THIS PROGRAM IS DIMENSIONED FOR 9216
 * ELEMENTS (COLUMBUS AND BORDEN AQUIFERS). REVISED JULY 8, 1996
 *

```

PROGRAM SURFHETZ
REAL M000,M100,M010,M110,M200,M020,M300,M030,M400,M040,
+ XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
DIMENSION C(9500),N(9500),CIJ(9500),XIJ(9500),YIJ(9500)
OPEN (UNIT=7,FILE='CONC.IN',STATUS='OLD')
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
OPEN (UNIT=9,FILE='MOMENTS',STATUS='UNKNOWN')
P=0.35
V=1
D=998.2
C
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C
DO 10 K=1,9420,6
  READ (7,200)N(K),C(K),N(K+1),C(K+1),N(K+2),C(K+2),N(K+3),
+   C(K+3),N(K+4),C(K+4),N(K+5),C(K+5)
200  FORMAT (7X,I5,G16.8,I5,G16.8,I5,G16.8,I5,G16.8,I5,
+   G16.8)
  10  CONTINUE
  READ (7,300)N(9421),C(9421),N(9422),C(9422),N(9423),C(9423),
+   N(9424),C(9424),N(9425),C(9425)
300  FORMAT (7X,I5,G16.8,I5,G16.8,I5,G16.8,I5,G16.8,I5,G16.8)
C
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C THE MOMENT CALCULATIONS ON THE ELEMENTS (RATHER THAN THE NODES) AND
C CALCULATE THE X AND Y-COORDINATES OF EACH ELEMENT
C
DO 20 I=1,144,1
DO 30 J=1,64,1
  C1=((I-1)*65+J)
  C2=(I*65+J)
  C3=(I*65+1+J)
  C4=((I-1)*65+1+J)
  CIJ(K)=(C(C1)+C(C2)+C(C3)+C(C4))/4
  XIJ(K)=(I-0.5)
  YIJ(K)=(J-0.5)
  WRITE (8,400)XIJ(K),YIJ(K),CIJ(K)
400  FORMAT (E16.8,E16.8,E16.8)
  30  CONTINUE
  20  CONTINUE
C
C READ IN THE ELEMENT CONCENTRATIONS AND COORDINATES CALCULATED ABOVE
C
CLOSE (8)
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
DO 35 K=1,9216,1
  READ (8,400)XIJ(K),YIJ(K),CIJ(K)
35  CONTINUE
C
C ZERO TH MOMENT (TOTAL MASS)

```

Program SURFHET2 (continued)

```

SUM=0.0
DO 40 K=1,9216,1
  SUM=SUM+CIJ(K)*P*V*D
40  CONTINUE
M000=SUM
C
C FIRST MOMENT, X (CENTER OF MASS IN THE X DIRECTION)
C
SUM=0.0
DO 50 K=1,9216,1
  SUM=SUM+CIJ(K)*P*V*D*XIJ(K)
50  CONTINUE
M100=SUM
XC=M100/M000
C
C FIRST MOMENT, Y (CENTER OF MASS IN THE Y DIRECTION)
C
SUM=0.0
DO 55 K=1,9216,1
  SUM=SUM+CIJ(K)*P*V*D*YIJ(K)
55  CONTINUE
M010=SUM
YC=M010/M000
C
C SECOND MOMENT, XX (SPATIAL VARIANCE IN THE X DIRECTION)
C
SUM=0.0
DO 60 K=1,9216,1
  SUM=SUM+CIJ(K)*P*V*D*(XIJ(K)**2)
60  CONTINUE
M200=SUM
SIGMAXX=(M200/M000)-(XC**2)
C
C SECOND MOMENT, YY (SPATIAL VARIANCE IN THE Y DIRECTION)
C
SUM=0.0
DO 70 K=1,9216,1
  SUM=SUM+CIJ(K)*P*V*D*(YIJ(K)**2)
70  CONTINUE
M020=SUM
SIGMAYY=(M020/M000)-YC**2
C
C SECOND MOMENT, XY (SPATIAL COVARIANCE)
C
SUM=0.0
DO 80 K=1,9216,1
  SUM=SUM+CIJ(K)*P*V*D*XIJ(K)*YIJ(K)
80  CONTINUE
M110=SUM
SIGMAXY=(M110/M000)-(XC*YC)
C
C THIRD MOMENT, X (SKEWNESS OF THE PLUME IN THE X DIRECTION)
C
SUM=0.0
DO 90 K=1,9216,1

```

Program SURFHET2 (continued)

```

      SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**3)
90  CONTINUE
      M300=SUM
      SKX=M300/(M000*(SIGMAXX**(3/2)))
C
C THIRD MOMENT, Y (SKEWNESS OF THE PLUME IN THE Y DIRECTION)
C
      SUM=0.0
      DO 100 K=1,9216,1
        SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**3)
100 CONTINUE
      M030=SUM
      SKY=M030/(M000*(SIGMAYY**(3/2)))
C
C FOURTH MOMENT, X (KURTOSIS OF THE PLUME IN THE X DIRECTION)
C
      SUM=0.0
      DO 110 K=1,9216,1
        SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**4)
110 CONTINUE
      M400=SUM
      KRX=M400/(M000*(SIGMAXX**2))
C
C FOURTH MOMENT, Y (KURTOSIS OF THE PLUME IN THE Y DIRECTION)
C
      SUM=0.0
      DO 120 K=1,9216,1
        SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**4)
120 CONTINUE
      M040=SUM
      KRY=M040/(M000*(SIGMAYY**2))
      WRITE (9,500)M000,XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
500 FORMAT (//,'*****SPATIAL MOMENT CALCUL',
A 'ATIONS****',//,
B '#####',//,
C '# ALL CONCENTRATIONS USED IN CALCULATIONS #',//,
D '#####',//,
E 'MASS OF BROMIDE (KG)',/2X,'M000=',G14.7,/,
F 'CENTER OF MASS, X-COORDINATE (M)',/2X,'XC=',G14.7,/,
G 'CENTER OF MASS, Y-COORDINATE (M)',/2X,'YC=',G14.7,/,
H 'SPATIAL VARIANCE, XX',/2X,'SIGMAXX=',G14.7,/,
I 'SPATIAL VARIANCE, YY',/2X,'SIGMAYY=',G14.7,/,
J 'SPATIAL COVARIANCE, XY',/2X,'SIGMAXY=',G14.7,/,
K 'SKEWNESS OF THE PLUME, X',/2X,'SKX=',G14.7,/,
L 'SKEWNESS OF THE PLUME, Y',/2X,'SKY=',G14.7,/,
M 'KURTOSIS OF THE PLUME, X',/2X,'KRX=',G14.7,/,
N 'KURTOSIS OF THE PLUME, Y',/2X,'KRY=',G14.7)
      CLOSE (7)
      CLOSE (8)
      CLOSE (9)
      STOP
      END

```

* THIS PROGRAM TAKES THE OUTPUT FROM GUTJAHR'S RANDOM
* FIELD GENERATOR AND PUTS IT IN THE FORMAT REQUIRED
* TO GRID IN SURFER
*

```
DIMENSION OUT(9500),XIJ(9500),YIJ(9500),SURF(9500)
OPEN (UNIT=8,FILE='FIELD',STATUS='OLD')
OPEN (UNIT=9,FILE='COND.TXT',STATUS='UNKNOWN')
```

* READ THE VALUES GENERATED BY GUTJAHR'S PROGRAM
*

```
DO 10 K=1,9216,1
  READ (8,15) OUT(K)
15  FORMAT (E21.14)
10  CONTINUE
```

* CALCULATE THE X AND Y COORDINATES FOR EACH ELEMENT, SHIFT THE
* MEAN TO -5.0 AND PRINT IT OUT
*

```
DO 30 I=1,144,1
DO 40 J=1,64,1
  SURF(K)=OUT((I-1)*64+J)-5.0
  XIJ(K)=(I-0.5)
  YIJ(K)=(J-0.5)
  WRITE (9,100)XIJ(K),YIJ(K),SURF(K)
100  FORMAT (E16.8,E16.8,E16.8)
40  CONTINUE
30  CONTINUE
  CLOSE (8)
  CLOSE (9)
  STOP
  END
```

* This program calculates the mean and variance of the random hydraulic
 * conductivity fields used in SUTRA. August 6, 1996
 *

```

PROGRAM MEANVAR
DOUBLE PRECISION MEAN,VARIANCE,SUM,MEANF,VARF
DIMENSION COND(20000)
OPEN (UNIT=8,FILE='FIELD',STATUS='OLD')
OPEN (UNIT=9,FILE='MEAN',STATUS='UNKNOWN')
DO 10 I=1,16384,1
  READ (8,100) COND(I)
100 FORMAT (D21.14)
10 CONTINUE
  SUM=0.0
  DO 20 I=1,16384,1 ! Calculate mean of entire field
    SUM=SUM+COND(I)
20 CONTINUE
  MEANF=(SUM/16384)
  SUM=0.0
  DO 30 I=1,16384,1 ! Calculate variance of entire field
    SUM=SUM+((COND(I))-MEANF)**2
30 CONTINUE
  VARF=(SUM/16383)
  SUM=0.0
  DO 40 I=1,9216,1 ! Calculate mean of field used in SUTRA
    SUM=SUM+COND(I)
40 CONTINUE
  MEAN=(SUM/9216)
  SUM=0.0
  DO 50 I=1,9216,1 ! Calculate variance of field used in SUTRA
    SUM=SUM+((COND(I))-MEAN)**2
50 CONTINUE
  VARIANCE=(SUM/9215)
  WRITE (9,200) MEANF,VARF,MEAN,VARIANCE
200 FORMAT (//,2X,'MEAN OF ENTIRE FIELD.....= ',D14.8,
+ /,2X,'VARIANCE OF ENTIRE FIELD....= ',D14.8,
+ ///,2X,'MEAN OF TRUNCATED FIELD.....= ',D14.8,
+ /,2X,'VARIANCE OF TRUNCATED FIELD.= ',D14.8)
CLOSE (8)
CLOSE (9)
STOP
END

```

* This program puts the *.d6 output file of pressures and concentrations
 * in the proper format for the *.d55 file to use as initial conditions for
 * simulation restart.
 *

```

PROGRAM PRESCONC
DOUBLE PRECISION CONC(9500),PRES(9500)
DIMENSION NOD(9500),NOD(9500) ! CAN'T DECLARE INTEGERS AS DP
OPEN (UNIT=7,FILE='CONCPRES.IN',STATUS='OLD')
OPEN (UNIT=8,FILE='RESTART.D55',STATUS='UNKNOWN')
TIME=6.34752D+07 ! SIMULATION ET
*
* READ PRESSURES FOR EACH NODE
*
DO 10 I=1,9415,6
  READ (7,100)NOD(I),PRES(I),NOD(I+1),PRES(I+1),NOD(I+2),
+   PRES(I+2),NOD(I+3),PRES(I+3),NOD(I+4),PRES(I+4),
+   NOD(I+5),PRES(I+5)
100  FORMAT (7X,I5,D16.10,I5,D16.10,I5,D16.10,I5,D16.10,I5,D16.10,
+   I5,D16.10)
10  CONTINUE
  READ (7,200)NOD(9421),PRES(9421),NOD(9422),PRES(9422),NOD(9423),
+   PRES(9423),NOD(9424),PRES(9424),NOD(9425),PRES(9425)
200  FORMAT (7X,I5,D16.10,I5,D16.10,I5,D16.10,I5,D16.10,I5,D16.10)
*
* READ CONCENTRATIONS FOR EACH NODE
*
DO 20 I=1,9415,6
  READ (7,100)NOD(I),CONC(I),NOD(I+1),CONC(I+1),NOD(I+2),
+   CONC(I+2),NOD(I+3),CONC(I+3),NOD(I+4),CONC(I+4),
+   NOD(I+5),CONC(I+5)
20  CONTINUE
  READ (7,200)NOD(9421),CONC(9421),NOD(9422),CONC(9422),
+   NOD(9423),CONC(9423),NOD(9424),CONC(9424),
+   NOD(9425),CONC(9425)
*
* WRITE THE ET AND PRESSURES / CONCENTRATIONS FOR EACH NODE
*
  WRITE (8,350)TIME
350  FORMAT (D20.10)
  DO 30 I=1,9421,4
    WRITE (8,300)PRES(I),PRES(I+1),PRES(I+2),PRES(I+3)
300  FORMAT (4D20.10)
30  CONTINUE
    WRITE (8,400)PRES(9425)
400  FORMAT (D20.10)
    DO 40 I=1,9421,4
      WRITE (8,300)CONC(I),CONC(I+1),CONC(I+2),CONC(I+3)
40  CONTINUE
      WRITE (8,400)CONC(9425)
    CLOSE (7)
    CLOSE (8)
    STOP
  END

```

```
* THIS PROGRAM TAKES THE OUTPUT FROM GUTJAHR'S RANDOM FIELD
* GENERATOR AND PUTS IT IN THE FORMAT REQUIRED FOR DATASET
* # 15B IN SUTRA'S D5 FILE
*
  DIMENSION OUT(9500),COND(9500),PERM(9500)
  OPEN (UNIT=8,FILE='FIELD',STATUS='OLD')
  OPEN (UNIT=9,FILE='15B',STATUS='UNKNOWN')
*
* READ THE VALUES GENERATED BY GUTJAHR'S PROGRAM
*
  DO 10 I=1,9216,1
    READ (8,15) OUT(I)
15  FORMAT (E21.14)
10  CONTINUE
*
* CONVERT FROM ln HYDRAULIC CONDUCTIVITY TO HYDRAULIC CONDUCTIVITY AND
* CHANGE THE MEAN FROM 0.0 TO -5.0
*
  DO 30 I=1,9216,1
    COND(I)=2.7182818**(OUT(I)-5.0)
30  CONTINUE
*
* CONVERT FROM HYDRAULIC CONDUCTIVITY TO INTRINSIC PERMEABILITY,
* CENTIMETERS TO METERS (K) AND PRINT OUT THE RESULTS IN THE PROPER
* FORMAT FOR DATASET 15B IN SUTRA
*
  DO 40 I=1,9216,1
    PERM(I)=((COND(I))*(1.024687595E-7))/100
    WRITE (9,45) I,PERM(I),PERM(I),1.0,1.0,1.0,1.0,1.0
45  FORMAT (I5,5X,D10.4,D10.4,F10.3,F10.3,F10.3,F10.3,F10.3)
40  CONTINUE
    CLOSE (8)
    CLOSE (9)
    STOP
    END
```

```

* THIS PROGRAM TAKES THE OUTPUT FROM GUTJAHR'S RANDOM FIELD
* GENERATOR AND PUTS IT IN THE FORMAT REQUIRED FOR DATASET
* # 15B IN SUTRA'S D5 FILE ##### 36,864 ELEMENTS #####
  INTEGER A,B,C,D,E
  DIMENSION OUT(9500),COND(9500),PERM(9500),PERME(40000)
  OPEN (UNIT=8,FILE='FIELD',STATUS='OLD')
  OPEN (UNIT=9,FILE='15B',STATUS='UNKNOWN')
*
* READ THE VALUES GENERATED BY GUTJAHR'S PROGRAM
*
  DO 10 I=1,9216,1
    READ (8,15) OUT(I)
15  FORMAT (E21.14)
10  CONTINUE
*
* CONVERT FROM ln HYDRAULIC CONDUTIVITY TO HYDRAULIC CONDUCTIVITY AND
* CHANGE THE MEAN FROM 0.0 TO -5.0
*
  DO 30 I=1,9216,1
    COND(I)=2.7182818**(OUT(I)-5.0)
30  CONTINUE
*
* CONVERT FROM HYDRAULIC CONDUCTIVITY TO INTRINSIC PERMEABILITY,
* CENTIMETERS TO METERS (K) AND PRINT OUT THE RESULTS IN THE PROPER
* FORMAT FOR DATASET 15B IN SUTRA
*
  DO 40 I=1,9216,1
    PERM(I)=((COND(I))*(1.024687595E-7))/100
40  CONTINUE
* BREAK EACH OF THE 9216 ELEMENTS INTO 4 ELEMENTS OF THE SAME
* PERMEABILITY FOR A TOTAL OF 36,864 ELEMENTS
  E=1
  DO 50 K=1,287,2
    DO 60 J=1,127,2
      I=E
      A=(J+(K-1)*128)
      B=(J+1+(K-1)*128)
      C=(J+(K)*128)
      D=(J+1+(K)*128)
      PERME(A)=PERM(I)
      PERME(B)=PERM(I)
      PERME(C)=PERM(I)
      PERME(D)=PERM(I)
      WRITE (9,45) A,PERME(A),PERME(A),1.0,1.0,1.0,1.0,1.0
45  FORMAT (15,5X,D10.4,D10.4,F10.3,F10.3,F10.3,F10.3,F10.3)
      WRITE (9,45) B,PERME(B),PERME(B),1.0,1.0,1.0,1.0,1.0
      WRITE (9,45) C,PERME(C),PERME(C),1.0,1.0,1.0,1.0,1.0
      WRITE (9,45) D,PERME(D),PERME(D),1.0,1.0,1.0,1.0,1.0
      WRITE (*,*) E
      E=E+1
60  CONTINUE
50  CONTINUE
  CLOSE (8)
  CLOSE (9)
  STOP
  END

```


* THIS PROGRAM SETS UP ALL NO-FLOW BOUNDARIES IN THE PROPER FORMAT
* FOR SUTRA'S DATASET 19. MODEL DISCRETIZATION TEST, DIMENSIONED
* FOR 289 X 129 NODES (TOTAL CONSTANT HEAD NODES = 832)
*

```
PROGRAM DDATA19
INTEGER NODE
REAL PRES,CONC
OPEN (UNIT=8,FILE='19',STATUS='UNKNOWN')
DO 10 J=1,289,1
DO 20 I=1,129,128
  NODE=129*(J-1)+I
  PRES=103524.7-((39.15486111)/2)*(J-1)
  CONC=0.0
  WRITE (8,100)NODE,PRES,CONC
100  FORMAT (I5,F20.2,F20:1)
20  CONTINUE
10  CONTINUE
DO 30 I=2,128,1
  NODE=I
  PRES=103524.7
  CONC=0.0
  WRITE (8,100)NODE,PRES,CONC
30  CONTINUE
DO 40 I=37154,37280,1
  NODE=I
  PRES=97886.4
  CONC=0.0
  WRITE (8,100)NODE,PRES,CONC
40  CONTINUE
CLOSE (8)
STOP
END
```

* THIS PROGRAM SETS UP ALL NO-FLOW BOUNDARIES IN THE PROPER FORMAT
* FOR SUTRA'S DATASET 19 (DIMENSIONED FOR 145 X 65 NODES)
*

```
PROGRAM DATA19
INTEGER NODE
REAL PRES,CONC
OPEN (UNIT=8,FILE='19',STATUS='UNKNOWN')
DO 10 J=1,145,1
DO 20 I=1,65,64
  NODE=65*(J-1)+I
  PRES=103524.7-(39.15486111)*(J-1)
  CONC=0.0
  WRITE (8,100)NODE,PRES,CONC
100  FORMAT (I5,F20.2,F20.1)
20  CONTINUE
10  CONTINUE
DO 30 I=2,64,1
  NODE=I
  PRES=103524.7
  CONC=0.0
  WRITE (8,100)NODE,PRES,CONC
30  CONTINUE
DO 40 I=9362,9424,1
  NODE=I
  PRES=97886.4
  CONC=0.0
  WRITE (8,100)NODE,PRES,CONC
40  CONTINUE
CLOSE (8)
STOP
END
```

- * THIS PROGRAM TAKES THE RAW NODE CONCENTRATION OUTPUT FROM SUTRA AND
- * CALCULATES THE SPATIAL MOMENTS, ZERO TH THROUGH FOURTH.
- * A GRID FILE IS ALSO CREATED FOR CONTOURING THE RESULTS IN SURFER.
- * ONLY CONCENTRATIONS GREATER THAN 0.1 PPM ARE USED IN THE CALCULATIONS.
- * THIS PROGRAM IS DIMENSIONED FOR 36,864 ELEMENTS (DISCRETIZATION TEST)
- * THE NODE INTEGER FIELD WAS INCREASED TO 6 TO ACCOMODATE 37,281 NODES
- * REVISED JULY 8, 1996

```

PROGRAM DSURFHET
REAL M000,M100,M010,M110,M200,M020,M300,M030,M400,M040,
+ XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
DIMENSION C(38000),N(38000),CIJ(38000),XIJ(38000),YIJ(38000)
OPEN (UNIT=7,FILE='CONC.IN',STATUS='OLD')
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
OPEN (UNIT=9,FILE='MOMENTS',STATUS='UNKNOWN')
P=0.35
V=0.25
D=998.2

C
C REDUCE THE RAW SUTRA NODE CONCENTRATION OUTPUT FROM 6 COLUMNS TO 1
C
DO 10 K=1,37278,6
  READ (7,200)N(K),C(K),N(K+1),C(K+1),N(K+2),C(K+2),N(K+3),
+ C(K+3),N(K+4),C(K+4),N(K+5),C(K+5)
200  FORMAT (7X,I6,G16.8,I6,G16.8,I6,G16.8,I6,G16.8,I6,
+ G16.8)
10  CONTINUE
  READ (7,300)N(37279),C(37279),N(37280),C(37280),N(37281),
+ C(37281)
300  FORMAT (7X,I6,G16.8,I6,G16.8,I6,G16.8)

C
C AVERAGE THE 4 NODE CONCENTRATIONS AROUND EACH ELEMENT TO PERFORM
C THE MOMENT CALCULATIONS ON THE ELEMENTS (RATHER THAN THE NODES) AND
C CALCULATE THE X AND Y-COORDINATES OF EACH ELEMENT
C
DO 20 I=1,288,1
DO 30 J=1,128,1
  C1=((I-1)*129+J)
  C2=(I*129+J)
  C3=(I*129+1+J)
  C4=((I-1)*129+1+J)
  CIJ(K)=(C(C1)+C(C2)+C(C3)+C(C4))/4
  IF (CIJ(K).LT.1.0E-7) THEN
    CIJ(K)=0.0
  ENDIF
  XIJ(K)=(I*0.5)-0.25
  YIJ(K)=(J*0.5)-0.25
  WRITE (8,400)XIJ(K),YIJ(K),CIJ(K)
400  FORMAT (E16.8,E16.8,E16.8)
30  CONTINUE
20  CONTINUE

C
C READ IN THE ELEMENT CONCENTRATIONS AND COORDINATES CALCULATED ABOVE
C
CLOSE (8)
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')

```

Program DSURFHET (continued)

```

      DO 35 K=1,36864,1
        READ (8,400)XIJ(K),YIJ(K),CIJ(K)
35    CONTINUE
      C
      C ZEROTH MOMENT (TOTAL MASS)
      C
      SUM=0.0
      DO 40 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D
40    CONTINUE
      M000=SUM
      C
      C FIRST MOMENT, X (CENTER OF MASS IN THE X DIRECTION)
      C
      SUM=0.0
      DO 50 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*XIJ(K)
50    CONTINUE
      M100=SUM
      XC=M100/M000
      C
      C FIRST MOMENT, Y (CENTER OF MASS IN THE Y DIRECTION)
      C
      SUM=0.0
      DO 55 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*YIJ(K)
55    CONTINUE
      M010=SUM
      YC=M010/M000
      C
      C SECOND MOMENT, XX (SPATIAL VARIANCE IN THE X DIRECTION)
      C
      SUM=0.0
      DO 60 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*(XIJ(K)**2)
60    CONTINUE
      M200=SUM
      SIGMAXX=(M200/M000)-(XC**2)
      C
      C SECOND MOMENT, YY (SPATIAL VARIANCE IN THE Y DIRECTION)
      C
      SUM=0.0
      DO 70 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*(YIJ(K)**2)
70    CONTINUE
      M020=SUM
      SIGMAYY=(M020/M000)-YC**2
      C
      C SECOND MOMENT, XY (SPATIAL COVARIANCE)
      C
      SUM=0.0
      DO 80 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*XIJ(K)*YIJ(K)
80    CONTINUE
      M110=SUM

```

Program DSURFHET (continued)

```

      SIGMAXY=(M110/M000)-(XC*YC)
C
C THIRD MOMENT, X (SKEWNESS OF THE PLUME IN THE X DIRECTION)
C
      SUM=0.0
      DO 90 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**3)
90  CONTINUE
      M300=SUM
      SKX=M300/(M000*(SIGMAXX**(3/2)))
C
C THIRD MOMENT, Y (SKEWNESS OF THE PLUME IN THE Y DIRECTION)
C
      SUM=0.0
      DO 100 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**3)
100 CONTINUE
      M030=SUM
      SKY=M030/(M000*(SIGMAYY**(3/2)))
C FOURTH MOMENT, X (KURTOSIS OF THE PLUME IN THE X DIRECTION)
      SUM=0.0
      DO 110 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**4)
110 CONTINUE
      M400=SUM
      KRX=M400/(M000*(SIGMAXX**2))
C FOURTH MOMENT, Y (KURTOSIS OF THE PLUME IN THE Y DIRECTION)
      SUM=0.0
      DO 120 K=1,36864,1
        SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**4)
120 CONTINUE
      M040=SUM
      KRY=M040/(M000*(SIGMAYY**2))
      WRITE (9,500)M000,XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
500  FORMAT (//,'***SPATIAL MOMENT CALCULATION'
A  ' N S***',, 36,864 ELEMENTS (Discretization test)',//,
B  '#####',,
C  '# CONCENTRATIONS < 0.1 PPM OMITTED #',,
D  '#####',//,
E  'MASS OF BROMIDE (KG)',/2X,'M000=',G14.7,//,
F  'CENTER OF MASS, X-COORDINATE (M)',/2X,'XC=',G14.7,//,
G  'CENTER OF MASS, Y-COORDINATE (M)',/2X,'YC=',G14.7,//,
H  'SPATIAL VARIANCE, XX',/2X,'SIGMAXX=',G14.7,//,
I  'SPATIAL VARIANCE, YY',/2X,'SIGMAYY=',G14.7,//,
J  'SPATIAL COVARIANCE, XY',/2X,'SIGMAXY=',G14.7,//,
K  'SKEWNESS OF THE PLUME, X',/2X,'SKX=',G14.7,//,
L  'SKEWNESS OF THE PLUME, Y',/2X,'SKY=',G14.7,//,
M  'KURTOSIS OF THE PLUME, X',/2X,'KRX=',G14.7,//,
N  'KURTOSIS OF THE PLUME, Y',/2X,'KRY=',G14.7)
      CLOSE (7)
      CLOSE (8)
      CLOSE (9)
      STOP
      END

```

* THIS PROGRAM TAKES THE RAW NODE CONCENTRATION OUTPUT FROM SUTRA AND
 * CALCULATES THE SPATIAL MOMENTS, ZERO TH THROUGH FOURTH.
 * A GRID FILE IS ALSO CREATED FOR CONTOURING THE RESULTS IN SURFER.
 * ALL CONCENTRATIONS ARE USED IN THE CALCULATIONS.
 * THIS PROGRAM IS DIMENSIONED FOR 36,864 ELEMENTS (DISCRETIZATION TEST)
 * THE NODE INTEGER FIELD WAS INCREASED TO 6 TO ACCOMODATE 37,281 NODES
 * REVISED JULY 8, 1996
 *

```

PROGRAM DSURFHE2
REAL M000,M100,M010,M110,M200,M020,M300,M030,M400,M040,
+ XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
DIMENSION C(38000),N(38000),CIJ(38000),XIJ(38000),YIJ(38000)
OPEN (UNIT=7,FILE='CONC.IN',STATUS='OLD')
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
OPEN (UNIT=9,FILE='MOMENTS',STATUS='UNKNOWN')
P=0.35
V=0.25
D=998.2

C
C REDUCE THE RAW SUTRA NODE CONCENTRATION OUTPUT FROM 6 COLUMNS TO 1
C
DO 10 K=1,37278,6
  READ (7,200)N(K),C(K),N(K+1),C(K+1),N(K+2),C(K+2),N(K+3),
+   C(K+4),N(K+4),C(K+4),N(K+5),C(K+5)
200  FORMAT (7X,I6,G16.8,I6,G16.8,I6,G16.8,I6,G16.8,I6,G16.8,I6,
+   G16.8)
10  CONTINUE
  READ (7,300)N(37279),C(37279),N(37280),C(37280),N(37281),
+   C(37281)
300  FORMAT (7X,I6,G16.8,I6,G16.8,I6,G16.8)
C
C AVERAGE THE 4 NODE CONCENTRATIONS AROUND EACH ELEMENT TO PERFORM
C THE MOMENT CALCULATIONS ON THE ELEMENTS (RATHER THAN THE NODES) AND
C CALCULATE THE X AND Y-COORDINATES OF EACH ELEMENT
C
DO 20 I=1,288,1
DO 30 J=1,128,1
  C1=((I-1)*129+J)
  C2=(I*129+J)
  C3=(I*129+1+J)
  C4=((I-1)*129+1+J)
  CIJ(K)=(C(C1)+C(C2)+C(C3)+C(C4))/4
  XIJ(K)=(I*0.5)-0.25
  YIJ(K)=(J*0.5)-0.25
  WRITE (8,400)XIJ(K),YIJ(K),CIJ(K)
400  FORMAT (E16.8,E16.8,E16.8)
30  CONTINUE
20  CONTINUE
C
C READ IN THE ELEMENT CONCENTRATIONS AND COORDINATES CALCULATED ABOVE
C
CLOSE (8)
OPEN (UNIT=8,FILE='CONC.TXT',STATUS='UNKNOWN')
DO 35 K=1,36864,1
  READ (8,400)XIJ(K),YIJ(K),CIJ(K)
35  CONTINUE

```

Program DSURFHET2 (continued)

```

C
C ZEROTH MOMENT (TOTAL MASS)
C
  SUM=0.0
  DO 40 K=1,36864,1
    SUM=SUM+CIJ(K)*P*V*D
40  CONTINUE
  M000=SUM
C
C FIRST MOMENT, X (CENTER OF MASS IN THE X DIRECTION)
C
  SUM=0.0
  DO 50 K=1,36864,1
    SUM=SUM+CIJ(K)*P*V*D*XIJ(K)
50  CONTINUE
  M100=SUM
  XC=M100/M000
C
C FIRST MOMENT, Y (CENTER OF MASS IN THE Y DIRECTION)
C
  SUM=0.0
  DO 55 K=1,36864,1
    SUM=SUM+CIJ(K)*P*V*D*YIJ(K)
55  CONTINUE
  M010=SUM
  YC=M010/M000
C
C SECOND MOMENT, XX (SPATIAL VARIANCE IN THE X DIRECTION)
C
  SUM=0.0
  DO 60 K=1,36864,1
    SUM=SUM+CIJ(K)*P*V*D*(XIJ(K)**2)
60  CONTINUE
  M200=SUM
  SIGMAXX=(M200/M000)-(XC**2)
C
C SECOND MOMENT, YY (SPATIAL VARIANCE IN THE Y DIRECTION)
C
  SUM=0.0
  DO 70 K=1,36864,1
    SUM=SUM+CIJ(K)*P*V*D*(YIJ(K)**2)
70  CONTINUE
  M020=SUM
  SIGMAYY=(M020/M000)-YC**2
C
C SECOND MOMENT, XY (SPATIAL COVARIANCE)
C
  SUM=0.0
  DO 80 K=1,36864,1
    SUM=SUM+CIJ(K)*P*V*D*XIJ(K)*YIJ(K)
80  CONTINUE
  M110=SUM
  SIGMAXY=(M110/M000)-(XC*YC)
C
C THIRD MOMENT, X (SKEWNESS OF THE PLUME IN THE X DIRECTION)

```

Program DSURFHET2 (continued)

```

SUM=0.0
DO 90 K=1,36864,1
  SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**3)
90 CONTINUE
M300=SUM
SKX=M300/(M000*(SIGMAXX**(3/2)))
C
C THIRD MOMENT, Y (SKEWNESS OF THE PLUME IN THE Y DIRECTION)
C
SUM=0.0
DO 100 K=1,36864,1
  SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**3)
100 CONTINUE
M030=SUM
SKY=M030/(M000*(SIGMAYY**(3/2)))
C
C FOURTH MOMENT, X (KURTOSIS OF THE PLUME IN THE X DIRECTION)
C
SUM=0.0
DO 110 K=1,36864,1
  SUM=SUM+CIJ(K)*P*V*D*((XIJ(K)-XC)**4)
110 CONTINUE
M400=SUM
KRX=M400/(M000*(SIGMAXX**2))
C
C FOURTH MOMENT, Y (KURTOSIS OF THE PLUME IN THE Y DIRECTION)
C
SUM=0.0
DO 120 K=1,36864,1
  SUM=SUM+CIJ(K)*P*V*D*((YIJ(K)-YC)**4)
120 CONTINUE
M040=SUM
KRY=M040/(M000*(SIGMAYY**2))
WRITE (9,500)M000,XC,YC,SIGMAXX,SIGMAYY,SIGMAXY,SKX,SKY,KRX,KRY
500 FORMAT(/,'***SPATIAL MOMENT',
a ' CALCULATIONS***',
A /,' 36,864 ELEMENTS (Discretization test)',//,
B '#####',/
C '# ALL CONCENTRATIONS USED IN CALCULATIONS #',/
D '#####',///,
E 'MASS OF BROMIDE (KG)',/2X,'M000=',G14.7,//,
F 'CENTER OF MASS, X-COORDINATE (M)',/2X,'XC=',G14.7,//,
G 'CENTER OF MASS, Y-COORDINATE (M)',/2X,'YC=',G14.7,//,
H 'SPATIAL VARIANCE, XX',/2X,'SIGMAXX=',G14.7,//,
I 'SPATIAL VARIANCE, YY',/2X,'SIGMAYY=',G14.7,//,
J 'SPATIAL COVARIANCE, XY',/2X,'SIGMAXY=',G14.7,//,
K 'SKEWNESS OF THE PLUME, X',/2X,'SKX=',G14.7,//,
L 'SKEWNESS OF THE PLUME, Y',/2X,'SKY=',G14.7,//,
M 'KURTOSIS OF THE PLUME, X',/2X,'KRX=',G14.7,//,
N 'KURTOSIS OF THE PLUME, Y',/2X,'KRY=',G14.7)
CLOSE (7)
CLOSE (8)
CLOSE (9)
STOP
END

```


APPENDIX F

MODEL TESTING DATA

WATSUTRA was tested by running a reactive simulation and comparing the solute solution with that of SUTRA. It was found that a nearly identical solution was obtained. The method of moment calculations was the same as SUTRA's to the third or fourth significant figure. The longitudinal and horizontal transverse apparent dispersivities were identical to those calculated using SUTRA. WATSUTRA was run with both transient flow and solute transport because the routine for steady-state flow was not working properly. The model is actually steady-state flow; it was found that running transient flow did not change the pressure distribution (the pressure distribution at the start of the simulation was identical to the distribution at 2 years). The only difference in running transient flow rather than steady-state flow is more calculations since there are no stresses on the system (e.g. no pumping periods).

WatSutra Varification Test

Weakly Heterogeneous

Realization I

1 Kilogram of Bromide

Reactive Run

All Concentrations Used in Calculations

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6574	24.3978	32.0631	0.9055	0.7668	0.3537	0.0658	3.2124	2.8273
0.0114	0.6545	24.5454	32.0823	1.1002	0.7572	0.4525	0.0763	3.3010	2.7850
0.2614	0.6229	28.3377	32.3356	6.8112	0.7072	0.4469	0.1631	2.8130	2.1765
0.5113	0.6059	32.5868	32.3748	12.3621	0.9783	-0.1720	0.1393	2.9527	2.9817
0.7613	0.5971	36.7364	32.3462	16.1052	1.5368	-0.8912	0.0274	3.0662	3.4211
1.0113	0.5916	40.4959	32.2255	18.1346	2.1779	-1.3121	-0.1099	3.2946	3.5137
1.2612	0.5885	43.8592	32.0111	19.9495	2.6995	-1.3642	-0.4867	3.3862	3.3644
1.5112	0.5858	46.9701	31.7508	22.1861	3.1452	-1.3491	-0.9202	3.4109	3.0674
1.7612	0.5836	49.9294	31.4872	24.7928	3.5175	-1.2188	-0.9692	3.5039	2.8887
2.0114	0.5816	52.8136	31.2133	28.5865	3.6093	-0.4644	-0.6897	3.6699	2.7628

Reactive Run

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6574	24.3967	32.0628	0.9041	0.7661	0.3409	0.0624	3.1742	2.8118
0.0114	0.6545	24.5444	32.0815	1.0967	0.7563	0.4419	0.0696	3.2558	2.7817
0.2614	0.6321	28.3023	32.3279	6.7853	0.8072	0.4518	0.0404	2.7623	3.1102
0.5113	0.6233	32.4651	32.3653	12.5884	1.1373	-0.1506	0.1078	2.8448	3.4194
0.7613	0.6127	36.5847	32.3423	16.6705	1.6960	-0.9823	0.0998	2.9645	3.5130
1.0113	0.6047	40.3534	32.2287	18.7524	2.3447	-1.4814	-0.0051	3.2169	3.5795
1.2612	0.5987	43.7431	32.0168	20.4053	2.8909	-1.5347	-0.3754	3.3158	3.6951
1.5112	0.5936	46.8980	31.7559	22.2077	3.4018	-1.3627	-0.7972	3.2369	3.6646
1.7612	0.5892	49.8910	31.4891	24.4428	3.7719	-1.1571	-0.8993	3.2785	3.3664
2.0114	0.5850	52.7969	31.2068	27.9335	3.7796	-0.2921	-0.7255	3.4070	3.0171

Discretization Test (37,281 nodes)

Weakly Heterogeneous

Realization 1

1 Kilogram of Bromide

Reactive Run

All Concentrations Used in Calculations

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6626	24.4528	32.0746	0.6328	0.4726	0.3619	0.0746	3.1293	2.7652
0.0114	0.6585	24.6080	32.0946	0.8333	0.4698	0.3735	0.0632	3.1358	2.7742
0.2614	0.6288	28.5530	32.3284	7.0723	0.5193	-0.0146	-0.0203	2.6402	2.8937
0.5113	0.6154	32.7883	32.3655	12.9947	0.7736	-0.7983	0.0277	2.8893	2.9650
0.7613	0.6063	36.8552	32.3226	16.8916	1.2237	-1.7196	0.0563	3.1530	3.0763
1.0113	0.5998	40.5552	32.1997	18.9012	1.7363	-2.1290	0.0364	3.5276	3.1717
1.2612	0.5949	43.9463	31.9981	20.6747	2.2371	-2.0411	-0.1244	3.6290	3.4296
1.5112	0.5909	47.1158	31.7575	22.4982	2.7544	-1.9239	-0.3378	3.5673	3.5350
1.7612	0.5875	50.1001	31.4895	24.3352	3.2037	-1.6949	-0.4141	3.5794	3.3992
2.0114	0.5847	52.9873	31.1914	27.6487	3.4042	-0.6702	-0.2797	3.6872	3.2259

Reactive Run

Only Concentrations Greater Than 0.1 ppm Used in Calculations

Time (yr)	Mass (kg)	XC (m)	YC (m)	Var X (m ²)	Var Y (m ²)	SKX	SKY (Unitless values)	KRX	KRY
0	0.6626	24.4525	32.0745	0.6314	0.4725	0.3581	0.0738	3.1134	2.7575
0.0114	0.6584	24.6078	32.0945	0.8312	0.4688	0.3694	0.0628	3.1165	2.7677
0.2614	0.6285	28.5520	32.3281	7.0529	0.5179	-0.0223	-0.0239	2.6206	2.8740
0.5113	0.6150	32.7878	32.3652	12.9339	0.7696	-0.7993	0.0247	2.8660	2.9234
0.7613	0.6056	36.8544	32.3224	16.7716	1.2135	-1.7072	0.0535	3.1153	3.0130
1.0113	0.5989	40.5572	32.1998	18.7050	1.7197	-2.0732	0.0389	3.4518	3.0975
1.2612	0.5938	43.9510	31.9980	20.3895	2.2153	-1.9216	-0.1200	3.4935	3.3547
1.5112	0.5895	47.1238	31.7571	22.0718	2.7276	-1.7416	-0.3379	3.3614	3.4813
1.7612	0.5859	50.1092	31.4885	23.7692	3.1712	-1.4772	-0.4194	3.3097	3.3576
2.0114	0.5828	52.9988	31.1884	26.9363	3.3632	-0.4288	-0.2957	3.4217	3.1741

Homogeneous Simulation

0.5 Kilogram of Bromide

All Concentrations Used in Calculations

Conservative Run

Time (yr)	Mass (kg)	XC (m)	Var X (m ²)	Var Y (m ²)
0.0000	0.4973	15.8751	2.2226	1.4826
0.5006	0.4973	33.8704	143.6283	5.1080
1.0011	0.4973	51.8464	285.3527	8.7318
1.5016	0.4973	69.8028	428.4481	12.3541
2.0022	0.4973	87.7548	572.0367	15.9757
2.5000	0.4973	105.6075	714.9858	19.5768
3.0005	0.4973	123.5578	858.8140	23.1948
3.5011	0.4973	141.5085	1002.5800	26.8059
4.0016	0.4973	159.4587	1146.3390	30.4030
4.5022	0.4973	177.4012	1289.0420	33.9765
5.0000	0.4970	195.1709	1421.9960	37.4934

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