Numerical simulations of preasymptotic transport in heterogeneous porous media: Departures from the Gaussian limit

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Received 30 November 2001; revised 7 March 2002; accepted 7 March 2002; published 19 March 2003.

[1] The objective of this work is to determine whether conventional solute macrodispersion theories adequately predict the behavior of individual subsurface plumes over time and length scales relevant to practical risk assessment and remediation activities. This issue is studied with a set of high-resolution numerical simulations of conservative tracers moving through saturated two-dimensional heterogeneous conductivity fields. The simulation experiments are designed to mimic long-term field studies. Spatially correlated statistically stationary lognormal conductivity realizations are generated with a Fourier transform procedure. Steady state velocity solutions are calculated for these fields using an accurate Darcian solver. Velocity contour plots reveal the presence of disconnected networks of preferential pathways over a range of correlation lengths. Reverse flow cells are rare. The velocity probability density functions have exponential tails and strong longitudinal asymmetries. Solute concentrations are derived from the simulated velocity fields with an accurate taut-spline transport code that minimizes numerical dispersion. The resulting plumes are tracked for travel distances of over one hundred spatial correlation lengths, corresponding to scales of practical interest. First moments and macrodispersivities, which measure the location and extent of the plume, are reasonably well approximated by conventional Fickian theories but continue to vary after long travel distances. This temporal variability highlights the slow convergence of the plume moments to Gaussian limits. Calculations of the relative dilution index, which is a measure of the plume mixing state, indicate strongly non-Gaussian behavior. Other measures of plume structure suggested by anomalous dispersion theories also reveal the persistence of non-Gaussian behavior after long travel distances. These measures appear to be more sensitive to non-Gaussian behavior than the spatial moments. Taken together, the simulation results suggest that conservative solute plumes moving through statistically stationary random media may not converge to Gaussian limits even after traveling hundreds of log conductivity correlation scales.

INDEX TERMS: 1829 Hydrology: Groundwater hydrology; 1832 Hydrology: Groundwater transport; 1869 Hydrology: Stochastic processes; KEYWORDS: groundwater, solute, stochastic, anomalous transport, simulation


1. Introduction

[2] One of the major topics of subsurface solute transport research has been the effect of geological heterogeneity on the movement of conservative tracers. When viewed at sufficiently large scales heterogeneity tends to disperse or spread solutes, a process commonly called macrodispersion. This process changes significantly over time. At first, when the dimensions of a plume are smaller than local geological anomalies its shape is controlled by individual features such as conductive channels or low permeability barriers. These features induce variations in the plume’s concentration contours as some parts of the plume advance more rapidly than others. If geological anomalies are of limited extent and are scattered throughout space in a random fashion their effects will tend to “average out” and the plume will eventually become more regular.

[3] This conceptual picture of macrodispersion is supported by the Central Limit Theorem, which implies that the
location of a solute particle subject to many small displacements has a Gaussian probability distribution in the limit as the travel time approaches infinity, provided that the displacements are drawn at random from a population with finite moments and a finite correlation scale [Gnedenko and Kolmogorov, 1968]. In the infinite time limit the ensemble mean solute concentration has a Gaussian spatial distribution, with moments that depend on the statistics of the random velocity field [Dagan, 1989]. The difference between the concentration of an individual plume and the ensemble mean concentration is a random quantity with a variance that approaches zero at infinite time if the Central Limit Theorem applies [Dagan, 1990]. It follows that individual plumes should approach the spatially Gaussian ensemble mean in the infinite time limit. This explains the emphasis on the ensemble mean that characterizes stochastic transport theory. Under the assumptions of this theory the ensemble mean should eventually provide an accurate description of any particular real world plume.

A number of investigators have studied how the ensemble mean of a conservative tracer approaches the asymptotic solution to (3) has a Gaussian spatial distribution when the velocity and macrodispersion tensors are constant. The macrodispersivities are generally larger than the corresponding local dispersivities since \( \tau \) is smoother than \( v \) and the \( D_x \) tensor must account for a greater portion of observed solute spreading than \( D_y \). Expressions for the macrodispersivity can be derived from the local dispersivities and the statistics of the small-scale velocity field. The asymptotic solution to (3) has a Gaussian spatial distribution in the infinite time limit. This explains the emphasis on the ensemble mean that characterizes stochastic transport theory. Under the assumptions of this theory the ensemble mean should eventually provide an accurate description of any particular real world plume.

\[ \nabla \cdot (D \nabla C) - v \cdot \nabla C = \frac{\partial C}{\partial t} \quad (1) \]

where \( v \) is a random velocity field with specified statistical properties, \( D \) is a local dispersion tensor that describes the effect of velocity fluctuations at scales smaller than those resolved by \( v \) (emphasized by including the subscript \( v \) and \( C(x, t) \) is the random concentration at the location vector \( x \) and time \( t \). Here, confining attention to two-dimensional processes, we suppose that the principal axes of \( D \) are aligned with the \( x \) and \( y \) coordinates so \( D_x \) is diagonal:

\[ D_x = \begin{pmatrix} v_x A_{L_x} & 0 \\ 0 & v_y A_{T_x} \end{pmatrix} \quad (2) \]

The coefficients \( A_{L_x} \) and \( A_{T_x} \) are the longitudinal and transverse macrodispersivities, respectively. This formulation assumes that the small-scale dispersion process is Fickian (i.e. the dispersive flux is proportional to the concentration gradient).

Under certain assumptions the mean of the solute concentration, \( \overline{C} \), is described by a larger-scale version of (1):

\[ \nabla \cdot (D \nabla \overline{C} - \nabla \cdot \nabla \overline{C} = \frac{\partial \overline{C}}{\partial t} \quad (3) \]

where \( \nabla \) is the mean solute velocity (e.g., a known regional trend) and \( D_x \) is a diagonal macrodispersion tensor that describes the effect of velocity fluctuations at scales smaller than those resolved by \( \nabla \) (emphasized by including the subscript \( \nabla \)):

\[ D_x = \begin{pmatrix} v_x A_{L_x} & 0 \\ 0 & v_y A_{T_x} \end{pmatrix} \quad (4) \]

The coefficients \( A_{L_x} \) and \( A_{T_x} \) are the longitudinal and transverse macrodispersivities, respectively. In (3) macroscopic dispersion is Fickian, in the sense that the macrodispersive flux is proportional to the mean concentration gradient. The macrodispersivities are generally larger than the corresponding local dispersivities since \( \tau \) is smoother than \( v \) and the \( D_x \) tensor must account for a greater portion of observed solute spreading than \( D_y \). Expressions for the macrodispersivity can be derived from the local dispersivities and the statistics of the small-scale velocity field. The asymptotic solution to (3) has a Gaussian spatial distribution when the velocity and macrodispersion tensors are constant.

Spatial moments indicative of Gaussian behavior have been observed in several controlled field studies in reasonably homogeneous media. Examples include the widely cited experiments conducted at Borden, Ontario [Sudicky, 1986] and Cape Cod, Massachusetts [Garabedian et al., 1991; Hess et al., 1992]. Fickian behavior has also been simulated in numerical experiments carried out by Boggs et al. [1992], Tompson and Gelhar [1990], Bellin et al. [1992], Salandin and Fiorotto [1998], and McLaughlin and Ruan [2001]. These field and numerical results lend credibility to the Gaussian model of field-scale transport. On the other hand, evidence at more geologically complex field sites [Boggs et al., 1992] and from two-dimensional multiscale numerical simulations [McLaughlin and Ruan, 2001] suggests that individual plumes may not look Gaussian even after reasonably long travel times. These plumes may be highly skewed with long leading or trailing edges, may have multiple peaks, or may be divided into different sections following separate preferential pathways. Plume geometry and the spatial distribution of concentration influence chemical and biological reactions and can have important implications for risk assessment and remedial design. For these reasons it is important to know how far particular plumes might deviate from the Gaussian ideal.

In this paper we investigate plume convergence with numerical simulations carried out in much the same way as a set of highly controlled field experiments. Numerical simulations are attractive because they enable us to take a close look at individual plumes as they evolve over time. In particular, they allow us to determine when we can adequately approximate individual plumes by an asymptotic Gaussian model based on (3). Since the log hydraulic conductivity and velocity fields used in our experiments have finite moments and correlation scales the assumptions of the Central Limit Theorem and traditional Fickian theory are met. Consequently, our simulated plumes must reach a Gaussian state eventually. The primary question in our investigation is how long it takes for this to occur. A secondary question is whether simple quantitative measures of plume structure can provide insight about non-Fickian behavior. Answers to these questions can help us decide what methods are most appropriate for describing real-world plumes over the finite timescales of most interest for practical applications.

In the investigations that follow we perform a set of single replicate simulations rather than Monte Carlo experiments because we are interested in the behavior of specific plumes rather than ensemble means. Of course, individual plumes generated from random velocity fields will differ
2. Experimental Approach

[10] Numerical solute transport experiments are much easier to perform than field experiments of comparable complexity. Even so, we do not propose that numerical simulations should replace either field experiments or theoretical analyses. Our numerical experiments rely on assumptions that may affect the validity or scope of particular conclusions. Wherever possible, we have tried to anticipate these possibilities in our discussion. However, we believe that our results, when taken as a whole, raise important questions about the applicability of Fickian/Gaussian models. These questions will need to be investigated in more detail in subsequent theoretical, field, and numerical studies.

2.1 Experimental Approach

[11] Our experimental approach is based on a simplified representation of a plume of conservative solute moving through a random time-invariant velocity field. We have confined our attention to two-dimensional transport in a rectangular region lying in the horizontal plane (see Figure 1). We focus on two-dimensional transport primarily for computational reasons. As discussed by McLaughlin and Ruan [2001], there is a tradeoff (for a fixed computational expenditure) between simulating long-term transport in a two-dimensional region versus short-term transport in a three-dimensional region. The two-dimensional approach provides more insight on convergence to Gaussian conditions while the three-dimensional approach accounts for vertical velocity variations and transport paths that are ignored in two dimensions. We were effectively limited in this study to problems having $O(10^6)$ degrees of freedom (feasible to solve on a high-end workstation). This translates to a two-dimensional region extending in the longitudinal direction $O(10^6)$ log transmissivity correlation scales or a three dimensional region extending $O(10^2)$ log hydraulic conductivity correlation scales. Since our goal is to identify long-term trends we have adopted the two-dimensional alternative. Hopefully, improved computational resources and numerical algorithms will soon make it feasible to check our two-dimensional results with three dimensional flow and transport models that can handle $O(10^5)$ or more degrees of freedom.

[12] The velocity fields required in our transport experiments are obtained by solving a steady state flow problem. Fixed head boundary conditions at the upstream and downstream ends of the domain yield a spatially uniform velocity trend (or mean) which is parallel to the longest side of a 2048 by 512 grid of square computational cells with length $\Delta x = \Delta y = 1.0$ (in our experiments length units are meters and time units are days). The other two sides of the domain are no-flow boundaries. The longitudinal hydraulic gradient is $J = -0.02$ and the uniform porosity $n$ is 0.3.

[13] The spatially variable conductivity fields (denoted $K$, with $F = \ln K$) used in our flow simulations are random realizations drawn from lognormal probability distributions with two-dimensional isotropic Gaussian spatial covariance functions [McLaughlin and Ruan, 2001]. The Gaussian covariance yields differentiable conductivity fields that have more persistence and are somewhat smoother than fields obtained from an exponential covariance. The conductivity fields are generated with the Fourier transform procedure described by Ruan and McLaughlin [1998]. Our use of hydraulic conductivities rather than transmissivities is formally equivalent to expanding the two-dimensional problem domain to a three-dimensional domain with unit depth and material properties that are constant in the vertical direction. In the remainder of this paper, we maintain the use of the term conductivity while considering the results of strictly two-dimensional simulations.

[14] In order to examine the connection between heterogeneity and convergence to a Gaussian limit we performed a set of eight simulation experiments which differ only in the values used for the nominal variance $\sigma_F^2$ ($0.25$, $1.0$, $2.5$, and $4.0$, see Table 1) and the log conductivity correlation scale $\lambda_F$ ($2.0$ and $8.0$ m). The variance and correlation scale values used in our simulations are broadly representative of field situations. Reported values of $\sigma_F^2$ in field studies range from below 0.3 [Boggs et al., 1992] to above 4 [Rehfeldt et al., 1992; Christiansen et al., 1998], while $\lambda_F$ spans values ranging from 1 m (our grid cell size) to tens of meters [Jensen et al., 1993]. The number of correlation scales included in our simulation domain is nearly 1024 for $\lambda_F = 2.0$ m and nearly 256 for $\lambda_F = 8.0$ m.

[15] The eight conductivity fields identified in Table 1 were constructed from two realizations, one for each of the chosen $\lambda_F$ values, with nominal means of $\langle F \rangle = \ln 6.25 = 1.8326$. Log conductivity fields associated with the different variances were derived from either of these realizations using a simple scaling procedure. The sample means, variances, and correlation scales obtained for each of the two realizations were within one percent of the nominal values.

[16] The flow model used in our simulation experiments is based on a straightforward two-dimensional steady state finite difference approximation and a preconditioned conjugate gradient solver. The convergence rate of the iterative solver was sensitive to the input field variance: approximately 3000 iterations were sufficient for $\sigma_F^2 = 0.25$, while almost 9000 iterations were required to achieve conver-

Figure 1. Configuration of the simulation domain used in the numerical experiments.
gence at the highest variance level of $\sigma_F^2 = 4$. We checked our flow solver by comparing its simulated head distributions to those calculated by the three-dimensional BIGFLOW code [Ababou et al., 1988], used in two-dimensional mode with the conjugate gradient solver option. For well-converged solutions (Euclidean error norm <10^{-2}) the head distributions were identical to at least 7 digits precision.

For consistency with the earlier approximate simulations of McLaughlin and Ruan [2001], the local dispersivities appearing in (2) are set to the prior values of $A_L = 0.15$ m and $A_T = 0.015$ m, giving a mean grid Peclet number $\tau_c \Delta x / \tau_c A_L \approx 6.7$ in the mean flow (longitudinal) direction. Numerical simulations show that reducing these values further to limit the influence of the local dispersion matrix tends to reinforce the findings of this paper. The mean velocity $\bar{v}_r$ appearing in (2) is set equal to the sample mean of the appropriate velocity realization. All of the sample mean velocity means were close to 0.42 m/day. Numerical dispersion induced by our taut-spline solution algorithm is significantly smaller than the subgrid scale dispersion, especially for local grid Peclet numbers of the order of the mean value above (see Ruan and McLaughlin [1999] for a detailed discussion).

Table 1. Means and Variances of the Log Conductivity and Velocity Fields Used for the Numerical Experiments a

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<th>Log Conductivity Statistics</th>
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<td>Experiment</td>
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aPowers of 10 are indicated in parentheses.

In our experiments, the solute concentration is specified to be zero on the left boundary of the computational domain. Outflow conditions are specified on the remaining boundaries. The outflow conditions are implemented in the dispersive step of the Eulerian-Lagrangian solution algorithm by fixing the outflow boundary concentration at the value obtained from the preceding advective step [Ruan and McLaughlin, 1999]. The initial concentration is zero everywhere in the computational domain, except over the rectangular source region, where it is equal to a specified unit source concentration. The rectangular source has dimensions 10 m by 75 m and is located near the upstream (left) boundary of the simulation domain (see Figure 1).

In our computational experiments, we produced eight different log conductivity fields with our Fourier transform generator, derived eight corresponding two-dimensional steady state velocity fields from our flow model, and then simulated the evolution of the eight corresponding solute plumes with our transport model. Plume concentration distributions were plotted at selected times and various numerical measures of plume structure were computed. In this respect, our results are similar in form to those reported for most field experiments. These results are summarized in the following section.

3. Results

3.1. Velocity Fields

The statistical properties of our eight log conductivity and velocity fields are summarized in Table 1. The longitudinal velocity means are only weakly dependent on $\sigma_F^2$ while the longitudinal and transverse velocity variances scale approximately with $\sigma_F^2$. Note that the velocity means and variances are consistently higher (albeit slightly) for the larger correlation scale. A simple interval estimation method [Greene, 2000, p.146] for the velocity variances shows that the 95% confidence intervals for the respective $\lambda_F = 2$ and $8$ m variances overlap substantially when the effective number of correlation scales (Table 1) is used as a measure of sampling degrees of freedom. In other words, finite-domain effects may account for the observed inflations in velocity statistics.

Figure 2 shows the contours of log conductivity and velocity magnitude $(\nu_x + \nu_y)^{1/2}$ in a 300 m by 300 m portion of the simulation domain for $\lambda_F = 2$ and $8$ m and for $\sigma_F^2 = 0.25$, 1 and 4.0. The velocity solutions reveal the presence of preferential flow channels (shown as red-colored paths), even for moderate $\sigma_F^2$. For $\sigma_F^2 = 0.25$, these
channels are mostly disconnected and of short length. As \( \sigma_F^2 \) increases the number of such channels stays approximately constant but connectivity improves. At high variances, the dominant channels that emerge convey significant fractions of the total flow, although no single channel persists through the length of the grid. In terms of absolute accuracy, Saladin and Fiorotto [1998] give a relevant discussion on the optimal choice of grid spatial increment and correlation scale in discretized models. In particular they show that discretization effects on velocity solutions are worst for small correlation scale and high variance \( F \) fields, but are likely to be small and will tend to suppress extremes in the flow fields. The consistency of development of preferential flow paths with increasing variance between the two columns of Figure 2 gives confidence that grid discretization effects are not unduly influencing the \( \lambda_F = 2 \) m, \( \sigma_F^2 = 4 \) velocity solution.

[24] The probability distributions of the velocity solutions are of particular quantitative interest. Bellin et al. [1992] identify and discuss salient features of velocity probability

Figure 2. Comparison of velocity magnitudes calculated for two different \( F \) realizations with correlation scales of 2 m (left top) and 8 m (right top). The region plotted is a 300 m \( \times \) 300 m portion of the computational domain. Colocated velocity solutions for \( F \) variances of 0.25, 1, and 4 are shown below each \( F \) realization. Mean flow is from left to right. See color version of this figure at back of this issue.
distributions for $\sigma_F^2 \leq 1.6$, showing nonzero third moments for the longitudinal components and Gaussian behavior for the transverse components. That study was performed with log conductivity fields generated with an exponential correlation function. Features of our velocity fields are examined in Figure 3 by plotting sample density (or frequency) functions on a decadic logarithm scale. With this scale a velocity probability density that can be approximated by a decaying exponential function plots as a straight line. A density that can be approximated by a Gaussian function plots as a quadratic with a negative slope of increasing magnitude and a density that can be approximated by a power law at large velocities (e.g., a Pareto density) plots as a logarithmic function with a negative slope of decreasing magnitude.

[25] The $\lambda_F = 2$ and 8 m probability densities match almost exactly for both velocity components shown in Figure 3. The longitudinal densities are strongly asymmetric around the mean and clearly non-Gaussian. At high positive velocities the density functions decay exponentially. Only a small fraction of the longitudinal velocities are negative, indicating that reverse flow cells are rare. The transverse velocity probability densities are symmetric with exponential tails, and cover a greater range of values as $\sigma_F^2$ increases. For comparison purposes, Figures 3b and 3d show ideal Gaussian densities (dotted lines) with variances set equal to the $\lambda_F = 2$ m numerical values (Table 1). It is clear that the exponential transverse densities differ from the Gaussian densities reported by Bellin et al. [1992], and are in general qualitative agreement with the densities calculated by Salandin and Fiorotto [1998]. Furthermore, it is important to note that the asymptotically exponential forms obtained for our velocity probability densities imply that the velocity moments of all orders are finite, so the Central Limit Theorem holds. Consequently, the concentration spatial distributions obtained from our velocities should eventually become Gaussian. The key question is how long it will take for this to occur.

3.2. Conventional Methods For Characterizing the Experimental Solute Plumes

3.2.1. Plume Maps

[26] Contour plots of solute concentration are one of the best ways to interpret the results of field or numerical transport experiments. Figure 4 shows the plumes obtained for all eight of our simulations at time $t = 2000$ days. The plots in each column show the effects of increasing the log conductivity variance (from top to bottom) for a given log conductivity correlation scale ($\lambda_F = 2$ m in the left column and $\lambda_F = 8$ m in the right column). It is apparent that the solute plumes become much more elongated (longitudinal macrodispersion is much greater) when the log conductivity variance increases. Transverse macrodispersion also increases modestly with variance. These effects are consistent with classical macrodispersion theory. Increasing the log conductivity correlation length results in significant distortion of the plume as it moves away from its rectangular source. When the variance and correlation scale are both large the local anomalies dominate. The plume becomes very elongated and dispersed, extending tendrils through preferential flow channels in advance of the plume.

[27] Similar plots could be produced for other random log conductivity realizations. Each of these would reflect the effects of its particular set of geological anomalies. However, our primary purpose here is not to describe the behavior of an ensemble of random replicates but to study the long-term evolution of a single plume. If macrodispersion theories are to be useful for predicting solute transport in practical situations, they must be able to provide reasonable descriptions of individual plumes over the timescales.
of interest for risk assessment and remediation. In order to examine this topic further we consider in the following sections some useful bulk measures of plume structure.

### 3.2.2. Spatial Moments

[28] The first and second spatial moments of solute concentration are widely used to measure the convergence of field plumes to a Gaussian limit. The \((i,j)\)th spatial moment of concentration \(C(x, t)\) is defined as follows:

\[
M_{ij}(t) = \int \int x^i y^j C(x, y, t) \, dx \, dy
\]

The plume mass at any time \(t\) is \(M_{00}(t)\) and the coordinates of its center of mass are \(m_x(t) = M_{10}(t)/M_{00}(t)\) and \(m_y(t) = M_{01}(t)/M_{00}(t)\). In all the simulations performed here, the initial plume mass \(M_{00}(0)\) is conserved to within 5%.

[29] Figure 5 plots versus travel time the deviations (normalized by \(\lambda_F\)) of the \(x\) and \(y\) components of the center of mass from the values \(m_x = v_x t\) and \(m_y = 0\) that would be expected if the plume followed the mean velocity vector. Note that at any given time the plume travels more correlation lengths (but the same absolute distance) when \(\lambda_F = 2\) m than when \(\lambda_F = 8\) m. It takes about 476 days for the center of mass to travel 100 correlation lengths when \(\lambda_F = 2\) m and about 1904 days for it to travel 100 correlation lengths when \(\lambda_F = 8\) m. Figures 5a and 5c show that the longitudinal center of mass coordinates depart from the corresponding mean velocity values by approximately five correlation lengths, although this deviation is variable along the trajectories. In some regions the plumes are ahead of the mean trajectory while elsewhere they lag behind. These discrepancies are affected by \(\sigma_F^2\), but are relatively small.

The transverse center of mass deviations shown in Figures 5b and 5d are no greater than a few correlation lengths, although their magnitude tends to increase with \(\sigma_F^2\). There is no evidence of convergence of the actual center of mass trajectories to the mean values, even after the plumes have traveled over distances of 100 log conductivity correlation scales or more.

[30] The distribution of mass about the plume centroid is described by the central spatial moments:

\[
M_{ij}^c(t) = \int \int x^i y^j C(x, y, t) \, dx \, dy
\]

The second central moments \(M_{20}^c(t)\) and \(M_{02}^c(t)\) are of particular interest here since their square roots provide convenient measures of the distances the plume has spread in the longitudinal and transverse directions, respectively. If the principal axes of the macrodispersion tensor are aligned with the \(x\) and \(y\) coordinates then time-averaged effective (plume-specific) values for the longitudinal and transverse macrodispersivities \(A_{L_v}(t)\) and \(A_{T_v}(t)\) can be computed from the asymptotic relationships [Tompson and Gelhar, 1990]:

\[
A_{L_v}(t) = \frac{1}{t} \int_0^t \frac{1}{2\pi M_{00}(t)} \frac{dM_{20}^c}{dt} \, dt = \frac{M_{20}^c(t) - M_{20}^c(0)}{2\pi M_{00}t}
\]

\[
A_{T_v}(t) = \frac{1}{t} \int_0^t \frac{1}{2\pi M_{00}(t)} \frac{dM_{02}^c}{dt} \, dt = \frac{M_{02}^c(t) - M_{02}^c(0)}{2\pi M_{00}t}
\]

Time histories for both \(A_{L_v}(t)\) and \(A_{T_v}(t)\) (normalized by \(\sigma_F^2\)) are plotted in Figure 6.
Fickian transport theories predict that the longitudinal and transverse macrodispersivities should both be constant when the plume shape is nearly Gaussian. The estimated macrodispersivity values plotted in Figure 6 are characterized by irregular variations, reflecting the effects of fluctuations in the velocity field. These macrodispersivity variations tend to be magnified as $s_F^2$ increases. For comparison, Figures 6a and 6c also show the theoretical Fickian asymptotic values for the longitudinal macrodispersivity. These theoretical values apply for Gaussian-correlated log conductivity fields and are derived by Ruan [1997, p. 209]:

$$\frac{A_{L}}{\sigma_{F}^{2}} = 1.25 \frac{\lambda_{F}}{\gamma}$$

and

$$\frac{A_{T}}{\sigma_{F}^{2}} = \frac{(4 \gamma_{T} + 3 \lambda_{F})}{16 \gamma^{2}}$$

where $\gamma = O(s_F^2)$ is a so-called flow factor, intended to correct for the effects of the linearization process [Gelhar and Axness, 1983; Gelhar, 1993, p. 113]. Dagan [1989, p. 320] questions the mathematical consistency of the flow factor derivation. Here, for simplicity, $\gamma$ is assumed to be unity.

The normalized longitudinal macrodispersivity plotted in Figure 6a and 6c track each other, suggesting that they scale approximately with $s_F^2$. This is in accord with theoretical predictions. However, the curves do not appear to be converging to the theoretical values even after the plume has traveled approximately 100 correlation lengths. The transverse macrodispersivities in Figures 6b and 6d vary even more than the longitudinal values, although this result is of less importance since the theoretical variance-scaled transverse macrodispersivity is very small ($\approx 0.012$ m) in our case. In fact, the major benefit of the transverse macrodispersivity plots is to provide an indirect upper bound on numerical dispersion. Inspection of Figure 6 shows that the variance-scaled transverse macrodispersivities are overestimated by approximately 0.2 m, a discrepancy that arises from both numerical dispersion and the detailed dispersive influences of fluctuations in the Darcian flow field. The contribution of numerical dispersion to this discrepancy is difficult to quantify, since the local

Figure 5. Plume centroid locations plotted as functions of travel time for different variances $\sigma_F^2$. Figures 5a and 5c show departures from the mean longitudinal travel velocity $v_x$ of the plumes while Figures 5b and 5d show lateral deflections of the plumes. A travel distance of 100 correlation lengths corresponds to a travel time of 476 days when $\lambda_F = 2$ m and 1904 days when $\lambda_F = 8$ m. See color version of this figure at back of this issue.
grid Peclet numbers vary spatially with \( v \) (a mean velocity scale is used in the local dispersion tensor (2)). However the total discrepancy is close to the assumed local longitudinal dispersivity \( A_L = 0.15 \) m, suggesting that numerical artifacts account for only a small fraction of the longitudinal dispersion observed in our simulations.

3.2.3. Dilution Index

The lower-order spatial moments discussed above provide helpful information about the location and overall dimensions of a solute plume but are not sensitive to its internal structure. For example, plumes with the same moments could have either single or multiple peaks and could have significantly different shapes. Additional insight about the degree of mixing within a plume can be obtained by examining the dilution (or entropy) index proposed by Kitanidis [1994]. This index is defined as:

\[
I(t) = \frac{E(t)}{E_{\text{max}}(t)}
\]

where

\[
E(t) = \exp \left[ - \sum_{k=1}^{N} p_k \log p_k \right]
\]

and \( E_{\text{max}}(t) \) is the maximum possible value of \( E(t) \), which, for effectively unbounded plumes, is a simple functional of the plume second spatial moments [Kitanidis, 1994]. The ratio \( p_k(t) = C_k(t) \Delta x \Delta y / M_{00}(t) \) is the normalized solute mass at time \( t \) in the cell centered on node \( k \). The dilution index is largest when the plume is well mixed and smallest when the plume mass is highly concentrated. It can be shown that \( E(t) \) is maximized, for a specified set of second central moments, when the spatial distribution of the solute concentration is Gaussian [Kitanidis, 1994]. In this sense \( I(t) \) measures the degree to which the plume has approached the ideal Gaussian limit. The lower the dilution index, the further the plume is from this limit.

Figure 7 shows the temporal evolution of the dilution indices for the eight plumes simulated in our numerical experiment as well as the deterministic \( \lambda_F = 0 \) plume, which provides a convenient reference. The deterministic plume is initially non-Gaussian since it starts from a rectangular source, but its dilution index comes reasonably close to the maximum entropy value of unity after a short time. The dilution indices for nonzero log conductivity variances approach the maximum entropy limit very slowly. The \( \lambda_F = 8 \) m dilution indices are especially variable, suggesting that they are still being influenced by local geological anomalies after thousands of days (hundreds of log conductivity correlation lengths).
3.2.4 Summary

The concentration contour plots, effective macrodispersivities, and dilution indices presented here provide a consistent picture of the long-term behavior of solutes moving through heterogeneous porous media. Generally speaking, our experimental plumes approach the Gaussian limit rather slowly, taking much more than 100 log conductivity correlation scales to attain moment or dilution index values compatible with the classical Gaussian model of macrodispersion. When geological anomalies are more pronounced (larger log conductivity variance) or when they persist for distances comparable to the plume dimensions (larger log conductivity correlation scale) the plume is more irregular and the moments and dilution indices are more variable. The spatial second moments seem to be relatively insensitive to deviations from Gaussian behavior and provide only an aggregate picture of plume dimensions. This is confirmed by examining plume contour plots at times when the longitudinal dispersivities (or second spatial moments) are relatively near theoretical predictions. Such plots often reveal multiple concentration peaks and internal structure that differs significantly from the regular contours of a Gaussian plume. The dilution index appears to be a somewhat more sensitive measure of non-Gaussian behavior, as it increases only gradually toward the ideal value of unity as the plume becomes more well-mixed and Gaussian in appearance.

3.3. Characterization Methods Suggested by Anomalous Dispersion Theory

The results presented above suggest that the travel distance required for convergence to the Gaussian limit in a stationary heterogeneous medium may be longer (hundreds of log conductivity correlation scales or thousands of meters) than generally believed. The practical implications of this slow convergence are problem-dependent. But it is apparent from our simulations that the peak concentrations of the plume must drop by orders of magnitude below the source concentration before the plumes can be reasonably approximated by a Gaussian model. The distinctly non-Gaussian character of our preasymptotic plumes suggests that it may be useful to investigate the behavior of transport measures suggested by non-Fickian (anomalous) transport theories. Although our plumes should eventually become Gaussian they could resemble anomalous plumes in certain respects over the finite timescales simulated in our numerical experiments. We investigate this possibility by evaluating several relatively simple anomalous transport measures.

In order to understand these measures it is helpful to review some basic concepts of anomalous transport. Many transport equations have common origins in probabilistic random walk theories. In these theories, the evolution of a solute plume is modeled by the distribution of a large number of particles that undergo random displacements in space over random intervals in time (pausing times). If each random particle displacement \( \Delta x \) and time \( \Delta t \) is drawn randomly from the same transition probability density \( p_{\Delta x, \Delta t} \), the spatial distribution of particle positions will evolve with time in a well-defined statistical manner. This transition probability density is the basis for the generalized master equation approach used by Berkowitz and Scher [2001] (the reader is referred to Jazwinski [1970] for a useful discussion of random walk theory). This approach gives an anomalous solute transport equation with fractional derivatives:

\[
\mathcal{D} \nabla^\alpha \nabla \cdot \nabla C = \frac{\partial \phi C}{\partial t}
\]

where the constant \( \mathcal{D} \) describes the spreading of the plume in terms of fractional units. The fractional derivatives may be defined in terms of the spectra of the derivative terms. Note that the anomalous mean transport equation reduces to the traditional mean equation of (3) when \( \alpha = 2.0 \) and \( \beta = 1.0 \).
Table 2. Fitted Fractional Indices $\alpha$ and $\beta$ of the Spatial and Temporal Anomaly Models

<table>
<thead>
<tr>
<th>Experiment</th>
<th>$\lambda_F$</th>
<th>$\sigma_F^2$</th>
<th>$\alpha$ ($\beta = 1$)</th>
<th>$(\beta,t)$ ($\alpha = 2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0</td>
<td>2.04</td>
<td>(0.996, 0.44), (0.996, 0.44)</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.25</td>
<td>1.62</td>
<td>(0.992, 0.46), (0.993, 0.46)</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1.0</td>
<td>1.23</td>
<td>(0.973, 0.50), (0.977, 0.48)</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>2.5</td>
<td>1.10</td>
<td>(0.964, 0.55), (0.969, 0.52)</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>4.0</td>
<td>1.15</td>
<td>(0.943, 0.61), (0.949, 0.60)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>0</td>
<td>2.04</td>
<td>(0.996, 0.44), (0.996, 0.44)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>0.25</td>
<td>1.62</td>
<td>(0.984, 0.51), (0.984, 0.48)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>1.0</td>
<td>1.16</td>
<td>(0.958, 0.57), (0.965, 0.54)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>2.5</td>
<td>0.85</td>
<td>(0.918, 0.74), (0.923, 0.71)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>4.0</td>
<td>0.92</td>
<td>(0.897, 0.89), (0.898, 0.90)</td>
<td></td>
</tr>
</tbody>
</table>

*Experiments of Table 1 are referenced by the ensemble correlation lengths and variances of $F$. Uncertainties lie in the second decimal place for $\alpha$ and in the third and second digits for $\beta$ and $t$, respectively.

[38] If all moments of $p_{xt}$ are finite the Central Limit Theorem applies and the asymptotic probability density of a typical particle or, equivalently, the spatial distribution of the mean solute concentration, will be Gaussian. If $p_{xt}$ has finite spatial moments but infinite temporal moments, then the transport is temporally anomalous and $\beta$ will generally differ from 1.0 [Berkowitz and Scher, 2001]. Such fractional (in time) processes, arising from long-tailed pausing time densities, have been shown to give rise to long-tailed breakthrough curves and unusual time dependences of plume lower spatial moments.

[39] Similarly, if $p_{xt}$ has finite temporal moments but infinite spatial moments, then the transport is spatially anomalous and $\alpha$ will generally differ from 2.0 [Benson et al., 2001]. In this case the fractional spatial derivative coefficient can be related to the velocity probability density. Consider a continuous particle velocity process $v$ with a first-order probability density $p(v)$ that approaches a power law of the form $p(v) \sim |v|^{-1-\alpha}$ for large $v$. If the associated index of stability, $\alpha$, is greater than or equal to 2 the velocity process has finite lower-order moments. If $\alpha$ is less than 2 the velocity is called an $\alpha$-stable Lévy process [Meerschaert et al., 1999]. In this case, it has infinite lower-order moments, the Central Limit Theorem does not apply, and the particle displacement does not approach an asymptotic Gaussian limit [Benson et al., 2001]. We discuss below some methods for estimating the coefficients $\alpha$ and $\beta$ from observed (or simulated) concentrations. Estimates of these coefficients provide additional quantitative measures of the degree to which a preasymptotic plumes deviates from the Gaussian limit.

3.3.3. Spatially Anomalous Dispersion

[40] First, we examine the values obtained for the stability index $\alpha$ associated with the spatially anomalous transport model [Benson et al., 2001]. In this case, the mean solute concentration is described by (12), with the dispersion derivative order $\alpha < 2$ and the temporal derivative order $\beta = 1$. The peak mean concentration $C_{max}$ obtained at any given time is given by [Benson et al., 2001]:

\[
C_{max}(t) = \frac{C_0}{(D \tau)^{1/\alpha}} \tag{13}
\]

where $C_0$ is the initial concentration of the instantaneous source, $D$ is a fractional spreading constant (unspecified here) and $t$ is time. This expression suggests that it is possible to estimate the $\alpha$ value associated with a particular experimental plume from a log plot of the maximum simulated concentration versus time. The third column of Table 2 presents the results of fitting (13) to the concentration solutions obtained from our eight numerical experiments, as well as a nominal deterministic experiment with $\sigma_F^2 = 0$. Figure 8 shows the resulting log plots of $C_{max}$ versus time. Since the zero-variance nominal case includes only local (Fickian) dispersion it should yield an asymptotic $\alpha$ value of 2. The value fit over a finite length of the curve is 2.04, a value that is reassuringly close to the Fickian limit. For nonzero log conductivity variances, the fitted $\alpha$ values

![Figure 8](image-url)
are all less than 2, tending to decrease in magnitude with increasing $\sigma_F^2$. The exception to this trend is for the $\sigma_F^2 = 4$ curve, which has a slightly larger $\alpha$ value than the $\sigma_F^2 = 2.5$ curve. This reversal is also seen in the $\lambda_F = 8$ m results (Table 2).

[41] For nonzero variances the peak concentration curves in Figure 8 are not smooth, varying irregularly under the influence of the spatial heterogeneity of the underlying log conductivity fields. The $\lambda_F = 8$ m curves exhibit greater variations, making $\alpha$ fits somewhat more subjective. Nevertheless, it is clear from these simple tests that our simulated plumes do not follow the classical ($\alpha = 2$) Fickian variation of peak concentration with time. Thus it may be concluded that the preasymptotic plumes display characteristics of anomalous Lévy motions, even though the input velocity fields are not Lévy stable.

3.3.2. Temporally Anomalous Dispersion

[42] The previous subsection focuses on plumes obtained from a transport equation with a fractional spatial derivative and a conventional first-order time derivative ($\beta = 1$). In this section, we consider the case where the time derivative is of fractional order and the spatial derivative has the conventional coefficient $\alpha = 2$. Berkowitz and Scher [2001] use a one-dimensional analysis to derive the breakthrough curve $C(t) = f(L, \bar{x}, \beta, t)$ of a Dirac delta of solute at $x = 0$ and $t = 0$ for this case, where $L$ is the breakthrough location and $\bar{x}$ is an unknown transition length [Montroll and Scher, 1973]. The normalized cumulative breakthrough curve $M_{\text{anom}}(t)$ is:

$$M_{\text{anom}}(t) = \int_0^t f(L, \bar{x}, \beta, t')dt'$$

with $\bar{x}$ and $\beta$ treated as fitting parameters. This anomalous breakthrough curve can be compared with the standard Fickian result for lognormal correlated fields [Dagan and Nguyen, 1989]:

$$M_{\text{Fick}}(L, A_L, \bar{v}, t) = \frac{1}{2} \left[ 1 - \text{erf} \left( \frac{L - \bar{v} t}{\sqrt{4A_L \bar{v}}} \right) \right]$$

where $A_L$ is the asymptotic longitudinal macrodispersivity and $\bar{v}$ is the mean flow velocity (in the x direction). In the Fickian case, $\bar{v}$ and $L$ are known, leaving the macrodispersivity as the sole free parameter for fitting.

[43] Figure 9 shows results from fitting $M_{\text{anom}}$ and $M_{\text{Fick}}$ to numerical breakthrough curves calculated at two control sections ($L = 743$ m and $L = 1143$ m downgradient of the source) perpendicular to mean flow in the two-dimensional simulation grids. The numerical breakthrough curves were estimated by integrating the plume mass present in the region downgradient of the control section for each output time, dividing by the total plume mass, and assembling these values into discrete time series (represented by squares in Figure 9). The curves represent best fits to the numerical data points. For $M_{\text{anom}}$, greatest weight was given to matching the breakthrough front, rather than the tail. This weighting process also yields the closest fits to the data in a least squares sense.

[44] Overall, the Fickian curves match the shape of the data points well, with fitted dispersion coefficients ($A_L/\sigma_F^2 = 3.42$ and $2.62$ m for the 743 m and 1143 m control planes, respectively, for $\sigma_F^2 = 1$, and $A_L/\sigma_F^2 = 3.33$ and 3.25 m, respectively, for $\sigma_F^2 = 4$) in good agreement with the effective longitudinal macrodispersivities plotted in Figure 6 and reasonably close to theoretical values derived from (9). The anomalous breakthrough curves do not provide as good a fit. While these curves can reproduce sharp fronts, they are unable to represent the short tails of the breakthrough data. Calculations for all fields show that this disagreement is greatest for small $\lambda_F$ and $\sigma_F^2$ (i.e. near-Fickian conditions), and least for large $\lambda_F$ and $\sigma_F^2$ (non-Fickian conditions).
The final column of Table 2 lists fitted values of $\beta$ and $\tau$ for the range of numerical experiments. Even though these temporal anomaly parameters do not lead to precise agreement with all the breakthrough data, it is useful to try to understand what they signify when fit to breakthrough fronts. The zero-variance Fickian limit experiment results in a lower bound for $\beta$ of 0.996, close to the ideal value of unity. Transition lengths are no more than unity, in qualitative agreement with Berkowitz et al. [2000] who, in a column study, found $\tau$ to be less than the field correlation scale and the corresponding transition velocity to be somewhat higher than the mean velocity $v$. Several trends are noticeable. First, $\beta$ exponents decline and transition lengths increase as $\sigma_F^2$ increases. This may be interpreted as showing the increasingly energetic nature of the velocity as the log conductivity field becomes more heterogeneous. A second trend is that, for a given field, the exponent increases and transition length decreases as attention moves from an earlier control section to a later one. These slight changes reflect the more diffuse nature of the plume as it passes the second control section. A third trend is that exponents for $\lambda_F = 2$ m are somewhat higher (less anomalous) than for $\lambda_F = 8$ m, and the transition lengths are shorter.

The suitability of the temporally anomalous model may also be examined in terms of other measures. For example, it is known that the ratio of the plume mean longitudinal displacement to the standard deviation of the plume varies as $t^{1/2}$ under Fickian conditions while under anomalous conditions the ratio is constant [Berkowitz and Scher, 2001]:

Fickian case \[ \frac{M_{10}(t) - M_{10}(0)}{\sqrt{M_{20}(t)}} \sim t^{1/2} \]  

Anomalous case \[ \frac{M_{10}(t) - M_{10}(0)}{\sqrt{M_{20}(t)}} \sim \text{constant}. \]  

Figure 10 shows plots of this moment ratio for our eight experimental plumes. In Figure 10, all of the large-time moment ratio exponents (slopes on this log-log plot) are reasonably close to the theoretical Fickian value of 1/2, although the slopes vary somewhat for higher variances and correlation scales. Overall, there is no compelling evidence that our preasymptotic experimental plumes resemble temporally anomalous dispersion.

4. Discussion and Conclusions

The numerical experiments described in this paper indicate that simulated plumes may not reach the Gaussian limit predicted by a Fickian macrodispersion model even after travel distances of hundreds of log conductivity correlation scales. This is not to say that the plumes will not or cannot converge to that limit. In fact, we know from the Central Limit Theorem that plumes generated from velocity fluctuations with finite moments and finite correlation scales must approach a Gaussian limit. Since the velocities used in our numerical experiments are of this type, the resulting plumes should eventually become Gaussian. What is at issue is how far a plume must travel before it can be reasonably approximated by a Gaussian transport model. Our experiments suggest that this could be farther than the distances of interest in some risk assessment and remediation problems.

The slow rate of convergence to the Gaussian limit is clearly related to the nature of the heterogeneities encountered by the plume as it travels downgradient. Log conductivity anomalies that deviate significantly from the regional mean and which are comparable in size to the plume dimensions can be expected to have a greater effect than smaller more localized features. During the preasymptotic non-Gaussian regime, significant portions of the plume are diverted into preferential pathways or around low conductivity zones, leading to irregularities that are distinctly non-Gaussian. These irregularities can be seen in
concentration contour plots but are less evident in temporal plots of spatially aggregated quantities such as spatial moments and breakthrough curves. The dilution index and peak concentrations are more sensitive measures of non-Gaussian conditions. In our simulations, both of these measures clearly reveal significant deviations from the Gaussian limit at distances of hundreds of log conductivity correlation scales.

[50] Since our preasymptotic plumes are not Gaussian it is worth considering to what extent their behavior resembles the anomalous dispersion predicted by non-Fickian transport theories. This can be determined by estimating the coefficients $\alpha$ and $\beta$ that appear in the anomalous transport equation given in (12). The $\alpha$ values estimated from our simulated plumes indicate our preasymptotic plumes display behavior that resembles spatially anomalous dispersion ($\alpha < 2$) for a range of log conductivity variance ($\sigma_F^2$) and correlation scale ($\lambda_F$) values. The $\alpha$ values decrease strongly with increasing $\sigma_F^2$ and weakly with increasing $\lambda_F$, although curves for high $\lambda_F$ are more variable. Our estimated $\beta$ values do not provide clear evidence of behavior resembling temporally anomalous dispersion.

[51] The results summarized above and in Figure 8 suggest that our simulated log conductivity and velocity fields include features that are sufficiently persistent or distinctive to produce behavior similar to anomalous dispersion. Although this behavior will eventually be overwhelmed by the leveling effect of the Central Limit Theorem it is still clearly discernible in plumes that have moved hundreds of correlation lengths. If individual plumes exhibit anomalous non-Fickian characteristics over times and distances of interest for risk assessment and remediation, there is a real need for an alternative to traditional Fickian transport models. In particular, it may be useful to consider applying the concepts of anomalous dispersion theory to preasymptotic problems with finite (rather than infinite) velocity moments. If models based on this theory can reproduce observed dilution indices and fractional derivative coefficients, as well as observed spatial moments, they may provide better predictions during the critical early stages of plume movement, when concentrations are highest and the plume is still relatively well contained.

[52] Non-Fickian transport theories may be even more important when the log conductivity fields include more than one distinct scale [McLaughlin and Ruan, 2001]. In such cases, conductivity variations at different scales are likely to interact in complex ways, possibly extending the time it takes for the plume to approach a nearly Gaussian state.

[53] It is evident from our simulations that we need better ways to describe the aggregate behavior of individual preasymptotic plumes, particularly in situations where factors such as peak concentrations and contact areas between contaminated and uncontaminated zones are important. This need for improved characterization of real-world transport phenomena should motivate additional theoretical and numerical investigations of near-source dispersion in heterogeneous media.

Acknowledgments. The authors wish to thank P. Van Bemmelen (Schlumberger GeoQuest) for support and P. L. Bjerg (Technical University of Denmark) for valuable discussions. Comments and criticisms from anonymous reviewers have led to improvements in this work. Part of the work presented in this paper was performed using facilities of the Joint Bureau of Meteorology/CSIRO High Performance Computing and Communications Centre (Melbourne).

References

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Figure 2. Comparison of velocity magnitudes calculated for two different $F$ realizations with correlation scales of 2 m (left top) and 8 m (right top). The region plotted is a 300 m × 300 m portion of the computational domain. Colocated velocity solutions for $F$ variances of 0.25, 1, and 4 are shown below each $F$ realization. Mean flow is from left to right.
Figure 4. Comparison of solute plumes for the simulated range of $\lambda_F$ and $\sigma_F^2$, at time $t = 2000$ days. A black rectangle is drawn to indicate the position of the initial distribution advecting at mean flow velocity $v_x$. Full-grid (2047 m $\times$ 511 m) solutions are plotted using a decadic logarithm concentration scale.
Figure 5. Plume centroid locations plotted as functions of travel time for different variances $\sigma_F^2$. Figures 5a and 5c show departures from the mean longitudinal travel velocity $v_x$ of the plumes while Figures 5b and 5d show lateral deflections of the plumes. A travel distance of 100 correlation lengths corresponds to a travel time of 476 days when $\lambda_F = 2$ m and 1904 days when $\lambda_F = 8$ m.
Figure 6. Effective plume macrodispersivities plotted as functions of travel time (longitudinal, Figures 6a and 6c; transverse, Figures 6b and 6d) calculated using (8). Horizontal black lines in Figures 6a and 6c represent the theoretical asymptotic limits based on linearized flow + transport theories. The theoretical asymptotic limit for Figures 6b and 6d is approximately 0.012 m. A travel distance of 100 correlation lengths corresponds to a travel time of 476 days when $\lambda_F = 2$ m and 1904 days when $\lambda_F = 8$ m.
Figure 7. Dilution index (10) versus time for (a) $\lambda_F = 2$ m and (b) $\lambda_F = 8$ m, each for the four different log conductivity variances indicated. An index of 1 corresponds to the maximum entropy (perfect Gaussian) case. A travel distance of 100 correlation lengths corresponds to a travel time of 476 days when $\lambda_F = 2$ m and 1904 days when $\lambda_F = 8$ m.
Figure 8. Peak plume concentrations, showing the departure from Gaussian conditions as the log conductivity variance $\sigma^2_F$ increases. The fitted asymptotic slopes of the curves are related to the listed spatial anomaly ($\alpha$) indices.
Figure 10. Moment ratios (16) of plumes for varying log conductivity variance $\sigma_F^2$ and correlation length $\lambda_F$. The dashed line indicates the ideal Gaussian-asymptotic slope of 1/2 for the log-log plot.