WATER RESOURCES RESEARCH, VOL. 33, NO. 6, PAGES 1485–1494, JUNE 1997

The first-order reliability method of predicting cumulative mass flux in heterogeneous porous formations

Todd H. Skaggs
U.S. Salinity Laboratory, Riverside, California

D. A. Barry
Department of Environmental Engineering, University of Western Australia, Nedlands, Australia

Abstract. Previous studies have proposed the first-order reliability method (FORM) as an approach to quantitative stochastic analysis of subsurface transport. Most of these considered only simple analytical models of transport in homogeneous media. Studies that looked at more-complex, heterogeneous systems found FORM to be computationally demanding and were inconclusive as to the accuracy of the method. Here we show that FORM is poorly suited for computing point concentration cumulative distribution functions (cdfs) except in the case of a constant or monotonically increasing solute source. FORM is better equipped to predict transport in terms of the cumulative mass flux across a control surface. As a demonstration, we use FORM to estimate the cumulative mass flux cdf in two-dimensional, random porous media. Adjoint sensitivity theory is employed to minimize the computational burden. In addition, properties of the conductivity covariance and distribution are exploited to improve efficiency. FORM required eight times less CPU time than Monte Carlo simulation to generate the results presented. The accuracy of FORM is found to be minimally affected by the size of the initial solute body and the solute travel distance. However, the accuracy is significantly influenced by the degree of heterogeneity, providing an accurate estimate of the cdf when there is mild heterogeneity ($\sigma_{\ln K} = 0.5$) but a less accurate estimate when there is stronger heterogeneity ($\sigma_{\ln K} = 1.0$).

Introduction

A standard approach to modelling heterogeneity in the subsurface environment is to treat heterogeneous formation properties as realizations of random spatial functions (RSF) [e.g., *Dagan*, 1989; *Christakos*, 1992; *Gelhar*, 1993]. With this conceptualization, fluid flow and mass transport are random processes, and a central problem for geohydrologists is to calculate the moments or distributions of dependent quantities such as hydraulic head on the basis of the RSF model used for the formation property.

Two common approaches to the problem are analytical methods and Monte Carlo simulation, both with well-known advantages and disadvantages. Analytical methods produce closed-form solutions for the ensemble moments of the quantity of interest (e.g., hydraulic head, contaminant concentration). The solutions provide insight into the processes that occur in the field and have been shown to be in agreement with some field-scale experimental observations [e.g., *Barry et al.*, 1988]. However, analytical solutions typically rely on a series of simplifying assumptions about the heterogeneity and geometry of the porous medium and consequently are not always appropriate. Monte Carlo simulation, on the other hand, generates the full distribution of the quantity of interest and is extremely flexible in terms of the type of problem for which it may be used, but its high computational cost often prevents routine application.

Paper number 97WR00660.
0043-1397/97/97WR-00660$9.00

Because of the limitations of these approaches, there has been interest in developing alternative methodologies. Generally, these methods obtain only approximations to the distributions or moments, but aim to do so with high computational efficiency and flexibility. One such method is the first-order reliability method (FORM), which apparently originated in structural engineering and has recently been applied to both surface [e.g., *Melching*, 1990; *Melching et al.*, 1992] and subsurface [e.g., *Sitar et al.*, 1987; *Jang et al.*, 1994] hydrologic problems.

The objective of this work is to evaluate FORM as an approach to predicting transport in heterogeneous porous media. Previous studies have focused primarily on simple analytical models of transport, and it remains to be demonstrated that FORM is a useful methodology for treating more complex problems. In limited applications to heterogeneous transport, FORM has performed poorly in terms of computational efficiency, and a thorough investigation of FORM's accuracy has not been possible. Herein we present a FORM implementation with much greater efficiency and provide the first detailed assessment of FORM's accuracy in predicting transport in heterogeneous media.

This article is organized as follows. FORM is first presented as a general methodology for computing the distribution of a scalar function of random variables. Next, previous applications of FORM to transport problems are reviewed and, on the basis of the literature and description of FORM given herein, general conclusions are drawn as to the utility of FORM in transport analysis. Last, we use FORM to predict the cumulative mass flux across a control surface in two-dimensional het-

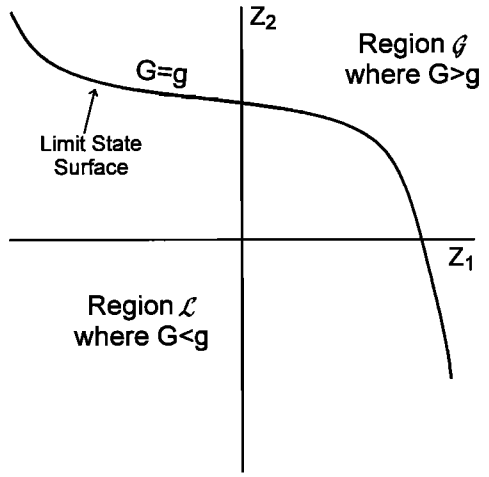

Figure 1. Hypothetical limit state surface and regions $\mathcal{G}$ and $\mathcal{L}$.

erogeneous porous media and assess the accuracy of the methodology in several simulations.

First-Order Reliability Method (FORM)

Theory

In this section FORM is presented as a general methodology for computing the cumulative distribution of a scalar function of a random vector. In the standard presentation of FORM, the focus is on computing the probability that a physical system will enter a state defined as "failure," with the complimentary probability (one minus the probability of failure) serving as a measure of the "reliability" or "safety" of the system. The safety/failure paradigm is useful in environmental problems that can be formulated in terms of compliance/noncompliance with environmental standards, and this perspective has been presented in the hydrologic literature [e.g., *Sitar et al.*, 1987]. Here we take a different approach, presenting FORM as a general stochastic method.

Let $\mathbf{Z} = [Z_1, \ldots, Z_n]'$ be a length n random vector with probability density f_Z, where the prime indicates vector transpose. Throughout we follow the convention that a random variable is represented by an upper case letter and a particular realization is represented by the same letter written in lower case. For example, the scalar function $G(\mathbf{Z})$ is a random variable and g is a realization. By definition, the probability that G is less than or equal to g, Pr $[G \leq g]$, is given by the cumulative distribution function (cdf):

$$F_G(g) = \Pr[G \leq g] = \int_{G \leq g} f_Z \, dZ_1 \cdots dZ_n. \qquad (1)$$

The n-fold integral is over the region of Z space where $G \leq g$. For a general function G and distribution f_Z, this integral is difficult if not impossible to compute because it is hard to make explicit the area of integration and because of well-known numerical difficulties associated with multifold integrals. The objective of FORM is to estimate F_G by obtaining an approximate solution to the integral in (1). Note that G may be computed either analytically or numerically. For example, in the context of subsurface transport, G could be a concentration computed with either an analytical or numerical transport model, with $\mathbf{Z}$ containing random model parameters.

To apply FORM we make the following restriction on G. We require G to be such that $G = g$ defines a hypersurface that divides Z space into two regions: region $\mathcal{L}$ where $G < g$ and region $\mathcal{G}$ where $G > g$. The hypersurface, called the limit state surface, is illustrated in Figure 1 for the case of $n = 2$. The figure is a contour plot of a hypothetical function G with only the $G = g$ contour shown; for $n = 2$ the limit state "surface" is this contour. Although we have restricted the allowable functions, the integral in (1) is in general no easier to compute because the limit state surface is still difficult to make explicit, and the integral is still multifold. The implications of this restriction in terms of FORM's applicability to transport simulation is discussed below.

To motivate the methodology, it is useful to consider initially a special case for which an exact solution to (1) is easily obtained. Suppose f_Z is the multivariate standard normal distribution (i.e., the elements of $\mathbf{Z}$ are uncorrelated normal variates with zero mean and unit variance) and the limit state surface is a hyperplane. In this case F_G is exactly [*Madsen et al.*, 1986],

$$F_G = \Phi(\beta), \qquad (2)$$

where $\beta = \mathbf{n}' \cdot \mathbf{Z}^*$, $\mathbf{n}$ is a unit vector normal to the limit state surface and directed toward $\mathcal{G}$, $\mathbf{Z}^*$ is the point on the limit state surface closest to the origin, and $\Phi(\ )$ is the univariate standard normal cdf. The situation is illustrated in Figure 2, again for the case of $n = 2$. It is straightforward to show $|\beta| = (\mathbf{Z}^{*\prime}\mathbf{Z}^*)^{1/2}$; that is, $|\beta|$ is the distance between the limit state surface and the origin.

Because of the relative ease of evaluating F_G for multivariate standard normal f_Z and a hyperplane limit state surface, the strategy of FORM is to transform (at least approximately) more general problems to this special case. In other words, transform $\mathbf{Z}$ to the space of uncorrelated standard normal variables and then approximate the transformed limit state surface with a first-order series expansion. The approximation of F_G is then given by (2), where $\mathbf{Z}^*$ and $\mathbf{n}$ are defined for the linearized surface.

In more detail, the first step in FORM is to transform $\mathbf{Z}$,

$$\mathbf{U} = T(\mathbf{Z}), \qquad (3)$$

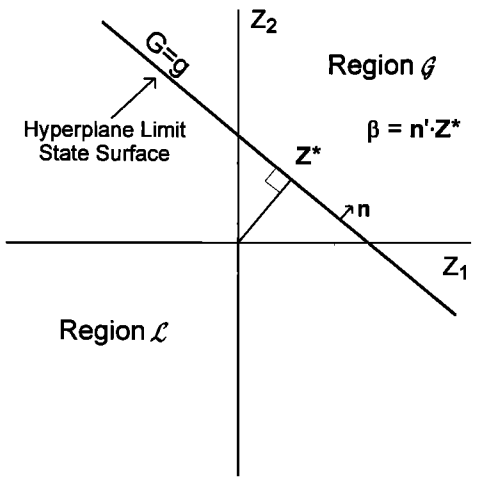

Figure 2. Hyperplane limit state surface.

where $\mathbf{U}$ is a vector of uncorrelated standard normal variates. The exact nature of the one-to-one transformation depends on f_Z. For example, if f_Z is the multivariate normal distribution, T is the linear transformation [e.g., *Madsen et al.*, 1986]

$$\mathbf{U} = T(\mathbf{Z}) = \mathbf{L}^{-1}(\mathbf{Z} - \bar{\mathbf{Z}}) \qquad \mathbf{C} = \mathbf{LL}', \tag{4}$$

where $\mathbf{C}$ is the covariance matrix of $\mathbf{Z}$, $\mathbf{L}$ is a lower triangle matrix, and $\bar{\mathbf{Z}}$ is the mean of $\mathbf{Z}$. If f_Z is the multivariate log-normal distribution, the transformation is

$$U = T(\mathbf{Z}) = \mathbf{L}^{-1}(\ln \mathbf{Z} - \bar{\mathbf{Z}}) \qquad \mathbf{C} = \mathbf{LL}', \tag{5}$$

where $\ln \mathbf{Z} = [\ln Z_1, \ldots, \ln Z_n]'$ and $\mathbf{C}$ and $\bar{\mathbf{Z}}$ are, respectively, the covariance and mean of $\ln \mathbf{Z}$. In principal, the required transformation can be made for any f_Z using the Rosenblatt transform [e.g., *Madsen et al.*, 1986], but this requires knowledge of conditional distributions and is computationally burdensome when $\mathbf{Z}$ contains more than a few elements. *Der Kiureghian and Liu* [1986] discuss a method for linearizing T and iteratively transforming more general distributions.

The transformation also maps the limit state surface into U space,

$$G(T^{-1}(\mathbf{U})) \equiv \hat{G}(\mathbf{U}) = g. \tag{6}$$

Likewise, the regions $\mathcal{L}$ and $\mathcal{G}$ are mapped into $\mathcal{L}_u$ and $\mathcal{G}_u$, respectively.

The next step is to approximate the limit state surface to first order,

$$\hat{G} \approx \hat{G}(\mathbf{U}_0) + \nabla\hat{G} \cdot (\mathbf{U} - \mathbf{U}_0) = g, \tag{7}$$

where $\mathbf{U}_0$ is the point about which $\hat{G}$ is linearized and $\nabla\hat{G} = [\partial\hat{G}/\partial U_1, \ldots, \partial\hat{G}/\partial U_n]$ is evaluated at $\mathbf{U}_0$. The gradient term is computed by applying the chain rule,

$$\nabla\hat{G} = \nabla G \cdot \frac{\partial T^{-1}}{\partial \mathbf{U}}, \tag{8}$$

where $\nabla G = [\partial G/\partial Z_1, \ldots, \partial G/\partial Z_n]$ is evaluated at $\mathbf{Z} = T^{-1}(\mathbf{U}_0)$ and $\partial T^{-1}/\partial \mathbf{U}$ is the Jacobian of the inverse transformation.

The problem is now transformed so that F_G can be approximated using (2). Straightforward application of the Lagrange multiplier method shows that the point on the linearized surface closest to the origin is

$$\mathbf{U}^* = [\nabla\hat{G} \cdot \mathbf{U}_0 - \hat{G}(\mathbf{U}_0) + g](\nabla G \cdot \nabla\hat{G}')^{-1}\nabla\hat{G}'. \tag{9}$$

The cdf is then $F_G(g) = \Phi(\beta)$, with $\beta = \mathbf{n}' \cdot \mathbf{U}^*$ and $\mathbf{n}$ directed towards $\mathcal{G}_u$. Note that this produces an approximation of F_G at g; the entire distribution can be constructed by repeating the procedure for a series of g values.

There remains the question of choosing $\mathbf{U}_0$. One possibility is to linearize about the transformed mean, $\mathbf{U}_0 = T(\bar{\mathbf{Z}})$, but this typically produces poor results near the tails of F_G. Previous studies have shown that the best results are obtained if the linearization is done at the point on the transformed limit state surface that is closest to the origin [*Madsen et al.*, 1986]. This point is called the "design point," or $\mathbf{U}^{DP}$, and is illustrated in Figure 3. To use this approach, however, $\mathbf{U}^{DP}$ must be located by solving the optimization problem

$$\min \mathbf{U}'\mathbf{U} \tag{10a}$$

subject to

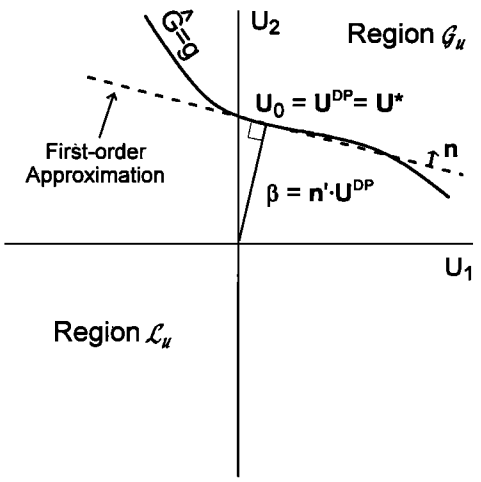

Figure 3. First-order approximation of the limit state surface in transformed parameter space.

$$\hat{G}(\mathbf{U}) = g \tag{10b}$$

The constraint (10b) is nonlinear for problems of interest and any optimization algorithm suitable for nonlinearly constrained problems can be used. A relatively simple iterative method that has been shown to be effective in solving (10) is based on sequentially linearizing the constraint [*Liu and Der Kiureghian*, 1991]. When (10b) is linearized, the solution to (10) is given by (9). Thus a sequential linearization algorithm is obtained by iterating on (9),

$$\mathbf{U}_{i+1} = [\nabla\hat{G} \cdot \mathbf{U}_i - \hat{G}(\mathbf{U}_i) + g](\nabla\hat{G} \cdot \nabla\hat{G}')^{-1}\nabla\hat{G}', \tag{11}$$

with the convergence point being $\mathbf{U}^{DP}$. Usually $\mathbf{U}_1 = T(\bar{\mathbf{Z}})$ is taken as the first point in the sequence. Although convergence is not guaranteed, experience has shown that the method works well. *Liu and Der Kiureghian* [1991] discuss other optimization algorithms available for solving (10). When $\mathbf{U}_0 = \mathbf{U}^{DP}$, $\mathbf{U}^* = \mathbf{U}^{DP}$ and we have $F_G(g) = \Phi(\beta)$, where $\beta = \mathbf{n}' \cdot \mathbf{U}^{DP}$ and $\mathbf{n}$ is defined as before. In a typical FORM calculation, solving (10) consumes the majority of the computational time. In particular, when gradient-based optimization algorithms such as (11) are used, it is the calculation of the constraint gradient, $\nabla\hat{G}$, that is costly.

In summary, an estimate of the cdf of $G(\mathbf{Z})$ is obtained by transforming $\mathbf{Z}$ to the space of standard normal variates and then linearizing the transformed limit state surface. The linearization is done at the design point, which must be located by solving the optimization problem (10). Each solution of (10) yields a single point, $F_G(g)$, and the entire distribution can be constructed by repeating the FORM calculations for a series of g values. The accuracy of the FORM approximation depends on the accuracy of the first-order expansion. If the transformed limit state surface is a hyperplane, FORM produces exact results. The more the limit state deviates from linearity, the worse FORM is expected to perform.

FORM With Incomplete Distribution Information

In our presentation of FORM we have assumed that f_Z and associated distribution parameters are known. Such full distribution information is often not available for hydrologic problems. Consequently, much of the FORM research in the hydrology literature has focused on the situation where only

partial knowledge of f_Z is available. For example, the marginal distributions of various subsets of $\mathbf{Z}$ might be known, but the joint distribution is unknown. In this case F_G cannot be computed because full distribution information cannot be obtained in a problem where less than full distribution information is input. However, FORM researchers have investigated the practice of assuming a joint distribution that is consistent with whatever information is available and then performing the FORM analysis using the constructed distribution [*Der Kiureghian and Liu*, 1986]. In structural engineering, results obtained in this manner are not usually regarded as a cdf; instead, they are treated as an index and used to rank the relative safety of different structures. Such an interpretation may also prove useful in environmental problems, although this has not yet been explored. Hydrologic researchers have tended to treat the results as a cdf and have shown in some simple example problems that this may be a reasonable interpretation.

It is known that the most important uncertainty in predicting subsurface solute transport is that associated with the spatial heterogeneity of hydraulic conductivity. Although it would be interesting to investigate the performance of FORM when only partial information on the heterogeneity is available or to consider additional uncertainties such as measurement errors, it should first be demonstrated that FORM is accurate when used with full distribution information. Since this has not yet been done, we will focus on problems where hydraulic conductivity is taken to be a spatial random field with known distribution and all other parameters are regarded as sure variables.

Sensitivity Analysis

At the design point the sensitivity of β with respect to model parameters is

$$\frac{\partial \beta}{\partial \mathbf{Z}} = \mathbf{n}' \cdot \frac{\partial T}{\partial \mathbf{Z}} \tag{12}$$

where $\partial T/\partial \mathbf{Z}$ is the Jacobian of the parameter transformation evaluated at the design point. This provides with little or no additional computational cost a measure of the sensitivity of β, and hence $F_G(g)$, to individual model parameters in the neighborhood of the design point. This information may be useful, for example, in determining the relative importance of different model parameters. Other sensitivity measures can also be obtained [see, e.g., *Madsen et al.*, 1986; *Sitar et al.*, 1987].

FORM and Subsurface Transport Simulation

Previous Studies

Sitar et al. [1987] give an overview of FORM and possible applications to subsurface flow and contaminant transport. As examples, they compute probability density functions (pdf) for three analytical models, including a one-dimensional convective-dispersive transport model in which specific discharge, porosity, and dispersivity are random variables. It is shown how the computed concentration pdf varies for different assumptions about model parameter distributions and correlations. The accuracy of the transport results is not assessed, although results obtained for two flow examples compare favorably with Monte Carlo results.

Cawlfield and Wu [1993] and *Wu and Cawlfield* [1992] use FORM to analyze, respectively, one- and two-dimensional analytical models of contaminant transport in homogeneous media. They assume the marginal distributions of various model parameters are known and construct the joint pdf using the Nataf model [*Der Kiureghian and Liu*, 1986]. Probabilistic sensitivity measures (e.g., $\partial\beta/\partial\mathbf{Z}$) are emphasized, and they conclude that variability in dispersivity is unimportant in comparison with variability in flow velocity and suggest dispersivity can therefore be treated as being deterministic. In comparison with Monte Carlo results (400,000 realizations), FORM is found to be less accurate when computing lower magnitude probabilities. *Piggott and Cawlfield* [1996] perform a similar sensitivity analysis for a one-dimensional vadose zone transport model.

Schanz and Salhotra [1992] apply a variation of FORM to both analytical and numerical transport models (homogeneous media) and compare the results with Monte Carlo simulation. In the method used by these authors, β is selected and then g is computed, whereas the opposite is true in the methodology described herein. *Schanz and Salhotra* [1992] treat various model parameters as being independent random variables and compute a few points on the concentration cdf. They observe that the method is generally more accurate estimating probabilities near the tail of the concentration cdf than in the middle of the cdf, although it is not clear if this is due to the fact that in the example they use the concentration is very small at the 50th percentile of the distribution.

Jang et al. [1994] consider a more complex problem and use FORM to simulate one- and two-dimensional transport in heterogeneous media. The spatial domain is discretized and the resulting elemental conductivities and dispersivities treated as correlated random variables with known marginal distributions. The flow boundary nodes are also treated as random, with the joint distribution of all variables constructed (presumably) using the method of *Der Kiureghian and Liu* [1986]. In the one-dimensional analysis, it is observed that FORM produces accurate results when the conductivity is homogeneous or mildly heterogeneous, but is less accurate under more heterogeneous conditions. Only a limited number of two-dimensional results are reported, with FORM being used in two simulations to compute a single point on the tail of a point concentration cdf. The FORM results differ from those obtained with Monte Carlo simulation by roughly a factor of 2. From these results it is hard to get a sense of FORM's ability to predict solute transport in two-dimensional heterogeneous formations because only a single point is computed on what is apparently the extreme tail of the concentration complementary cdf.

Hamed et al. [1995] use FORM to analyze three-dimensional transport in a uniform but uncertain velocity field. Various aquifer parameters are treated as random variables and a number of simulations are performed in which the concentration complimentary cdf is computed. FORM is found to be less accurate in computing the lower probabilities on the complimentary cdf. *Hamed et al.* [1996b] use FORM to simulate two-dimensional transport in mildly heterogeneous media and sensitivity results for the discretized hydraulic conductivity are presented. They also use FORM to analyze a plume containment scenario.

Discussion

From the previous studies and the description of the FORM methodology given above, we can draw some general conclusions regarding FORM and its applicability to subsurface transport problems.

Trade-offs associated with FORM. It is worthwhile to consider the trade-off involved when the approximate FORM methodology is used instead of the asymptotically exact Monte Carlo (MC) methodology, both in terms of the computational requirements and the information that is obtained. As noted by *Schanz and Salhotra* [1992], FORM typically reveals a narrower range of information than MC simulation because we must focus on computing the cdf a particular scalar function, whereas with MC methods we generally acquire the results needed to compute the cdfs of any number of functions. For example, with FORM we might compute the point concentration cdf at a particular location and time, whereas with MC simulation we compute realizations of the entire concentration field, from which the concentration cdf can be computed for any given location. On the other hand, FORM provides sensitivity information that is not easily obtained with Monte Carlo analysis, and for some applications this may be a consideration.

In terms of computational costs, it makes little sense to use FORM to compute distribution functions unless there is a computational advantage over MC simulation. In most of the works mentioned above, simple analytical models with only a few (<10) parameters were used, and the computational burden of both FORM and MC methods in such cases is minor. In contrast, the analysis of two-dimensional transport in heterogeneous media by *Jang et al.* [1994] was computationally expensive, and FORM fared poorly in comparison to MC simulation, requiring roughly the same amount of time to compute a single point on the (point) concentration cdf as would have been required to compute full cdfs for the entire spatial domain using MC methods. The high computational cost was due to the large (>100) number of parameters created by discretizing the spatial domain. As noted previously, the primary numerical difficulty is the repeated calculation of ∇G (where G is in this case a point concentration) and *Jang et al.* [1994] estimated ∇G using a divided difference approximation, requiring $n + 1$ calculations of G for each iteration of the optimization algorithm. So, for example, if there are 500 parameters ($n = 500$) and the algorithm takes five iterations to converge, the transport model will have to be run 2505 times for each point on the cdf that is calculated. That many simulation runs can easily serve as the basis for a very good MC calculation. Clearly, FORM is not a sensible methodology for problems with a large number of parameters if ∇G is computed using divided differences. *Skaggs and Barry* [1996a] and *Mok et al.* [1994] show that the computational costs of FORM are significantly reduced when sensitivity methods [e.g., *Ahlfeld et al.*, 1988; *Skaggs and Barry*, 1996b] are used to compute the gradient. The use of sensitivity methods in the FORM algorithm is further investigated in the simulations presented below.

Along these same lines, we note that *Jang et al.* [1994] and *Hamed et al.* [1996b] also use the second-order reliability method (SORM) and find that it is generally more accurate than FORM. SORM is similar to FORM with the difference being that a second-order approximation of the limit state surface is used. As described by *Der Kiureghian et al.* [1987], the first step in SORM is to locate the same design point used in FORM by solving (10). Constructing the second-order approximation through this point then requires roughly $8n$ additional evaluations of G. Again, when n is of order 10^2 or greater, it is likely that it would be possible to obtain an MC result with a similar amount of computational effort. Since there does not appear to be any way to avoid these $8n$ function evaluations, we conclude that SORM will not provide any advantage over MC methods when applied to problems with a large number of parameters.

When parameters are created from discretization of a random spatial domain, it may be possible to reduce the number of parameters in the FORM analysis. *Skaggs and Barry* [1997], *Hamed et al.* [1996a], and *Mok et al.* [1994] have investigated regionalizing subsets of the discretized parameters and performing the FORM analysis using a coarser grid than is required by the numerical scheme. *Skaggs and Barry* [1997] have also examined the possibility of defining an alternative design point that is "close" to the true design point but that can be located more easily. Either approach should make both FORM and SORM more efficient, and further investigations along these lines are warranted; however, many questions remain open, such as determining the optimal upscaling procedure and demonstrating that sufficiently accurate results can be obtained in a variety of problems.

Concentration versus mass flux formulation. Regardless of the trade-offs associated with FORM, we can make some observations about the suitability and expected accuracy of the methodology for different formulations of the transport problem. Recall we stipulated $G = g$ must define a surface that splits Z space into the two regions, $\mathcal{G}$ and $\mathcal{L}$. Although it has not been previously noted in the literature, a simple example illustrates that some formulations of the transport problem are not expected to satisfy this requirement.

Consider one-dimensional tracer transport in a homogeneous porous medium where pore water velocity is uniform but uncertain. Assume the velocity uncertainty is characterized by some distribution and all other parameters are known deterministic constants. Letting C be the tracer concentration at $x = \bar{x}$ and $t = \bar{t}$, the problem is to compute a point on the cdf of C, say $F_C(\bar{c})$. In this simple problem, we can characterize by inspection the regions delineated by $C = \bar{c}$ for two different transport scenarios.

First consider the case of a constant solute source at the inlet end of the transport domain. The solute profile computed with the mean velocity at time $\bar{t}$ might look something like the profile in Figure 4a. By physical reasoning we see that there is a particular velocity, $\bar{v}$, that will result in a concentration $\bar{c}$, and any velocity $v < \bar{v}$ produces $c < \bar{c}$ and any velocity $v > \bar{v}$ produces $c > \bar{c}$. As indicated in (1), F_C is the integral of f_V over region $\mathcal{L}$, which is illustrated in the inset of Figure 4a. The probability space is divided into the two regions $\mathcal{G}$ and $\mathcal{L}$ and thus the requirements of FORM are consistent with the problem formulation.

Next consider the case of solute being introduced as a short pulse at the inlet end. The solute profile at time $\bar{t}$ might look something like that shown in Figure 4b. In this scenario there are two velocities that can produce $\bar{c}$ at $\bar{t}$: a slower velocity in which $\bar{c}$ is realized as the leading edge of the plume passes $\bar{x}$ and a faster velocity in which $\bar{c}$ is realized as the trailing edge passes $\bar{x}$. The inset in Figure 4b illustrates f_V and the areas of integration. Clearly the parameter space is not divided into two regions. If FORM were applied to this problem, only one of the shaded areas would be accounted for and the results will be poor if the size of the second region is nonnegligible. In two- and three-dimensional heterogeneous transport problems where the random model parameters are the discretized conductivity elements, an analogous problem arises where a concentration can be realized at a particular location and time as a plume approaches a point, passes beyond a point, or passes

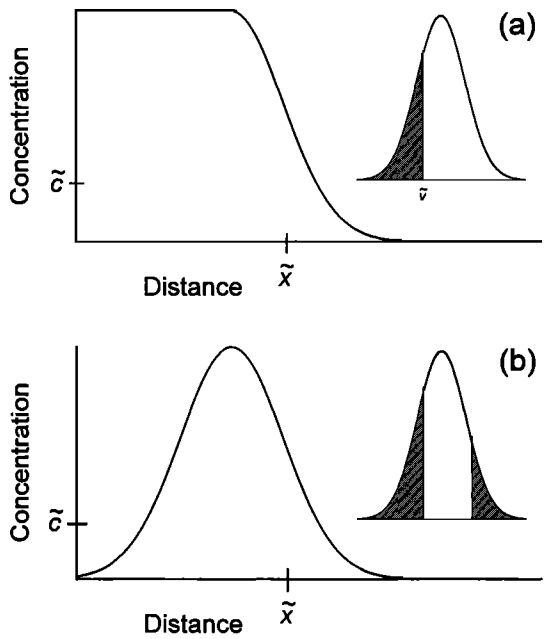

Figure 4. Illustration of the applicability of FORM to transport problems with (a) a constant source and (b) a transient source.

to the side of a point. Each possibility produces a local minima in (10), and experience has shown that gradient-based algorithms commonly fail in such cases, jumping endlessly back and forth between local minima. Of course, another optimization approach could be used to find the global minimum, but the point is that the probability content associated with these local minima may be significant and, consequently, a FORM approximation made at the global minimum will be a poor estimate of the cdf. In theory, FORM can be modified to handle such problems by defining multiple design points [see *Madsen et al.*, 1986], but as a practical matter it would be difficult to create a general computer code capable of identifying and solving these problems, and in the end would probably be computationally inefficient relative to MC simulation.

We conclude that if G is a point concentration, FORM can be used reliably only if the solute source is constant or monotonically increasing. If the solute source is transient, or if there is a finite solute body initially present in the system, FORM is likely to produce poor results. One way to avoid the limitation on source type is to formulate the transport problem in terms of the cumulative mass flux (M) across a control surface, defined as the mass of solute per unit time and unit area crossing a surface, integrated over time. In this case M is monotonic regardless of the source type and FORM can be used to compute f_M without the earlier difficulty. The description of transport in terms of solute flux has been studied previously using analytical methods [e.g., *Shapiro and Cvetkovic*, 1988; *Dagan*, 1989; *Dagan et al.*, 1992] and cumulative mass flux has been found to be a relatively robust quantity for describing field-scale solute transport [*Cvetkovic et al.*, 1992]. The emphasis of these previous works is on computing the mean and variance of M given a statistical description of the conductivity or velocity field. The only way to obtain full distribution information with such methods is to assume a priori a distribution for M. In the remainder of this paper we develop and test a FORM methodology for predicting M in heterogeneous formations.

Cumulative Mass Flux Prediction With FORM

Physical System and Governing Equations

We focus on two-dimensional (horizontal plane) tracer transport in a saturated heterogeneous porous formation under steady flow conditions. The governing transport equation is [*Bear*, 1972]

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_i}\left[D_{ij}\frac{\partial C}{\partial x_j}\right] - \frac{\partial}{\partial x_i}(V_i C) \tag{13}$$

where $C(x_1, x_2, t)$ is the solute concentration [$M\ L^{-3}$], $V_1(x_1, x_2)$ and $V_2(x_1, x_2)$ are the components of the depth-averaged linear groundwater velocity vector [$L\ T^{-1}$], and summation from 1 to 2 over terms with repeated indices is implied. The components of the hydrodynamic dispersion tensor are [*Bear*, 1972]

$$D_{ij} = \alpha_t|\mathbf{V}|\delta_{ij} + (\alpha_l - \alpha_t)V_iV_j/|\mathbf{V}|, \tag{14}$$

where α_l and α_t are, respectively, the longitudinal and transverse (local scale) dispersivities [L], $|\mathbf{V}|$ is the magnitude of the velocity vector, δ_{ij} is the Kronecker delta, and molecular diffusion is ignored. The solute velocity is obtained from Darcy's law and the steady flow equation,

$$V_i = -\frac{1}{\theta}K\frac{\partial H}{\partial x_i} \qquad i = 1, 2 \tag{15}$$

$$\frac{\partial}{\partial x_1}\left[lK\frac{\partial H}{\partial x_1}\right] + \frac{\partial}{\partial x_2}\left[lK\frac{\partial H}{\partial x_2}\right] = 0 \tag{16}$$

where K is the depth-averaged isotropic hydraulic conductivity [$L\ T^{-1}$], H is the hydraulic head [L], θ is the constant formation porosity [$L^3\ L^{-3}$], and l is the constant formation thickness [L]. The hydraulic conductivity, $K(x_1, x_2)$, is a random spatial function.

At time zero an initial body of solute is distributed over a finite volume and sits upstream from the control surface of interest, which for simplicity is assumed to be a planar surface that is normal to the x_1 axis (the direction of mean flow) and is referred to as the control plane (CP). For a given CP the transport problem can be formulated in either of two ways. We can compute the cumulative mass flux, M, at a fixed time, in which case M is random and time is a deterministic constant, or we can compute the time at which a fixed amount of the mass will have crossed the CP, in which case time is random and the mass amount is constant. We adopt the former conceptualization and seek the cdf of M, F_M. The cumulative mass flux across a CP that is an arbitrary distance downstream is

$$M(t) = \int_0^t j(\tau)\, d\tau \tag{17}$$

where

$$j(t) = \theta l \int_{-\infty}^{\infty} CV_1 - D_{11}\frac{\partial C}{\partial x_1} - D_{12}\frac{\partial C}{\partial x_2}\, dx_2. \tag{18}$$

As a particular example we consider a rectangular spatial domain measuring 10 m × 4 m, with a log-conductivity corre-

lation length scale of $\lambda = 1$ m (Figure 5). The top and bottom sides are no-flow boundaries and the left and right sides are fixed-head boundaries with a mean hydraulic gradient of 4×10^{-3} in the x_1 direction. The transport boundary conditions are $\partial C/\partial x_2 = 0$ on the top and bottom sides, meaning there is no mass flux across the no-flow boundaries, and $\partial C/\partial x_1 = 0$ on the right (downstream) side and $C = 0$ on the left side, meaning the mass flux across the exit boundary is due to advection alone and solute reaching the upstream boundary is immediately removed from the system [*Barry and Sposito*, 1988]. In our examples the upstream boundary condition is essentially irrelevant because the solute does not migrate there. Also, the control planes are located far enough away from the exit boundary so that the downstream boundary condition is not expected to influence transport across the CP.

Initially, the solute is uniformly distributed over a rectangular area. The conductivity is lognormally distributed with geometric mean $K_G = 8.0$ m d^{-1} and isotropic exponential correlation function $R = \exp(-h/\lambda)$, where h is the separation distance. Various values of $\sigma_{\ln K}$ will be considered. The local dispersivities are $\alpha_l = 0.1$ m and $\alpha_t = 0.05$ m, the effective porosity is $\theta = 0.3$, and the formation thickness is $l = 5$ m.

Numerical Solution

The flow equation is solved and the velocity field is obtained using Galerkin's finite element method with rectangular bilinear elements [e.g., *Pinder and Gray*, 1977; *Huyakorn and Pinder*, 1982]. The transport equation is solved using the Laplace-transform Galerkin technique [*Sudicky*, 1989]. The grid used in the spatial discretization is dimensioned 51×41 with uniform spacings $\Delta x_1 = 0.2$ m and $\Delta x_2 = 0.1$ m. These spacings allow five grid nodes per correlation length in the x_1 direction and 10 grid nodes per correlation length in the x_2 direction.

We apply FORM to compute the cdf of $M(\mathbf{K})$, F_M, where $\mathbf{K}$ is the vector containing the 2091 nodal conductivities. The finite element approximation of j can be written

$$j \approx \mathbf{a}'\mathbf{c}, \tag{19}$$

where $\mathbf{c}$ is the vector of nodal concentrations and $\mathbf{a}$ is the vector containing the basis function, velocity, dispersion, and quadrature terms necessary to approximate the operators in (18) [e.g., *Pinder and Gray*, 1977; *Huyakorn and Pinder*, 1982]. As part of the Laplace-transform Galerkin method, M is computed in the Laplace domain and numerically inverted to the time domain. Recall it is essential that we compute the gradient $\partial M/\partial \mathbf{K}$ efficiently. *Skaggs and Barry* [1996b] describe direct and adjoint methods that can be used in conjunction with the Laplace-transform Galerkin technique to compute the required sensitivities. Since M is a scalar function and is linear in $\mathbf{c}$, the adjoint method is the most efficient approach [*Skaggs and Barry*, 1996b]. Adjoint methods compute the sensitivities of functionals (i.e., M) without computing the Jacobian of state variables (i.e., $\partial \mathbf{c}/\partial \mathbf{K}$). Based on our previous results [*Skaggs and Barry*, 1996b], we expect that with 2091 parameters the adjoint method will require 2 orders-of-magnitude less CPU time than the perturbation method to compute a single gradient.

The parameter vector $\mathbf{K}$ follows the multivariate lognormal distribution with covariance matrix

$$\mathbf{C} = \sigma^2_{\ln K}\mathbf{R}, \tag{20}$$

where $\mathbf{R}$ is the matrix of correlation coefficients. The relevant parameter transformations are

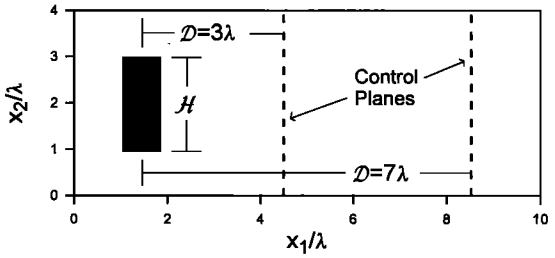

Figure 5. Diagram of the two-dimensional transport system; λ is the conductivity correlation length, $\mathcal{H}$ is the transverse dimension of the initial solute body, and $\mathcal{D}$ is the distance from the initial solute body to the control plane.

$$\mathbf{U} = T(\mathbf{K}) = \sigma^{-1}_{\ln K}\boldsymbol{\Gamma}^{-1} \ln(\mathbf{K}/K_G) \qquad \mathbf{R} = \boldsymbol{\Gamma}\boldsymbol{\Gamma}', \tag{21}$$

$$\mathbf{K} = T^{-1}(\mathbf{U}) = K_G \exp(\sigma_{\ln K}\boldsymbol{\Gamma}\mathbf{U}), \tag{22}$$

where $\boldsymbol{\Gamma}$ is lower triangular and $\exp([\ ,\ldots,\ ]') = [\exp(\ ),\ldots,\exp(\ )]'$. An interesting and important aspect of applying FORM with this covariance and parameter transformation is that the inverse transform of the design point, $\mathbf{K}^{DP} \equiv T^{-1}(\mathbf{U}^{DP})$, is independent of $\sigma_{\ln K}$. This means that once $F_M(m)$ is computed using a particular value of $\sigma_{\ln K}$, $F_M(m)$ can be obtained for any other value of $\sigma_{\ln K}$ by simply taking the new design point as $\mathbf{U}^{DP} = T(\mathbf{K}^{DP})$, where T is written with the new $\sigma_{\ln K}$.

To see the independence of $\mathbf{K}^{DP}$, recall that the design point $\mathbf{U}^{DP}$ is the solution to the constrained minimization problem (10), which was written in terms of the standard normal parameter space. The equivalent problem in the untransformed parameter space is found by substituting (21) into (10). Following some algebraic manipulations, we obtain

$$\min \sigma^{-2}_{\ln K}\mathbf{a}'\mathbf{R}^{-1}\mathbf{a} \tag{23a}$$

subject to

$$G(\mathbf{K}) = g \tag{23b}$$

where $\mathbf{a} = \ln(\mathbf{K}/K_G)$ and the solution is equal to $\mathbf{K}^{DP}$. Note that the vector $\mathbf{K}^{DP}$ that satisfies (23) is the same regardless of the value of the (nonzero) scalar $\sigma_{\ln K}$. Thus $\mathbf{K}^{DP}$ is independent of $\sigma_{\ln K}$, and $\mathbf{U}^{DP}$ can be obtained for any $\sigma_{\ln K}$ using $\mathbf{K}^{DP}$ and (21) as indicated above. The independence of the inverse transform of the design point also holds for multivariate normal parameter vectors with covariance matrices of the form (20).

To evaluate the accuracy of the FORM solutions, we compare the results with those obtained using Monte Carlo simulation. Realizations of the conductivity nodes are generated using the lower-upper triangular matrix technique [e.g., *Christakos*, 1992, pp. 326–328]. On the basis of visual inspection of the computed F_M, we found 1000 realizations were generally sufficient for the MC method to converge.

Results and Discussion

Previous studies [e.g., *Cvetkovic et al.*, 1992] indicate that F_M is influenced by the transverse extent of the initial solute body ($\mathcal{H}$), the travel distance to the control plane ($\mathcal{D}$), and the degree of conductivity heterogeneity (quantified by $\sigma_{\ln K}$). We assess the accuracy of FORM by performing a number of simulations in which these factors are varied. The results are

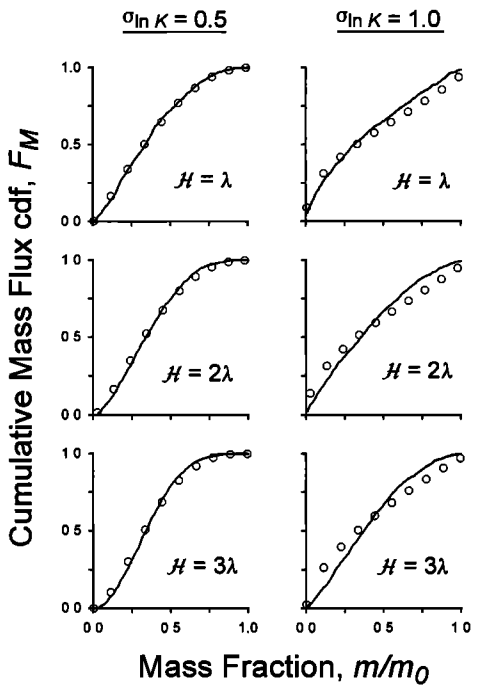

Figure 6. Computed cumulative mass flux cdfs for a control plane located a distance $\mathcal{D}$ = 3λ downstream from an initial solute body. Monte Carlo results are shown with solid lines, FORM results with open circles.

for the particular system under consideration and may not be readily transferable to other systems. For example, it is expected that the no-flow boundaries at the top and bottom are influencing the transport when the initial plume is large, so the results may not apply to unbounded spatial domains.

Let m_0 be the total mass in the initial solute body. Figure 6 shows plots of $F_M(m/m_0)$ computed at time t = 25 days for $\mathcal{D}$ = 3λ and various values of $\mathcal{H}$ and $\sigma_{\ln K}$. The Monte Carlo results are depicted with a solid line and the FORM results with open circles. The left column of plots is for $\sigma_{\ln K}$ = 0.5 and the right is for $\sigma_{\ln K}$ = 1.0. From top to bottom, the three rows of plots are for $\mathcal{H}$ = λ, 2λ, and 3λ. The longitudinal length of the initial solute body is of lesser importance in predicting F_M [*Cvetkovic et al.*, 1992] and is approximately equal to 0.5λ in all of our simulations.

The uncertainty in M should be less for larger $\mathcal{H}$ [*Cvetkovic et al.*, 1992]. This can be seen in our results by noting that F_M becomes steeper (i.e., has a smaller variance) as $\mathcal{H}$ is increased. The effect of $\mathcal{H}$ on FORM's accuracy is observed by comparing the three rows of plots. The accuracy is only slightly affected by $\mathcal{H}$, with the rows of plots showing a decrease in accuracy with increasing $\mathcal{H}$. However, over the range of source sizes considered here, the effect is very minor.

More important is the effect of $\sigma_{\ln K}$, which is observed by comparing the two columns of plots. FORM is significantly more accurate with the milder heterogeneity ($\sigma_{\ln K}$ = 0.5). The one-dimensional analysis of *Jang et al.* [1994] similarly showed a decrease in FORM's accuracy with increasing heterogeneity, although the pattern of misfit apparent in Figure 6 was not observable because they computed only two points on the concentration cdf.

Figure 7 shows results computed for the longer travel distance $\mathcal{D}$ = 7λ and time t = 65 days. At this larger travel distance, the nearly linear cdfs indicate that there is roughly an equal probability that any particular mass fraction will have passed the control plane. FORM is again significantly more accurate with the milder heterogeneity. The slight decrease in accuracy with increasing $\mathcal{H}$ is again observable. Overall, the results for the longer travel distance (Figure 7) appear to be slightly less accurate than those for the shorter distance (Figure 6).

Note that the pattern of misfit is consistent in Figures 6 and 7. That is, when FORM is inaccurate, it underestimates high values of F_M and overestimates low values. In some environmental applications it is the complimentary cdf, $1 - F_M$, that is of interest. For example, it may be required to compute the probability that a contaminant stored in the subsurface will migrate beyond the boundary of a storage facility at some time in the future. In this case the complimentary cdf defines exceedance probabilities for different amounts of mass. The pattern of misfit indicates that FORM tends to overestimate exceedance probabilities for high-mass, low-probability events (the compliment of underestimating high F_M values) and underestimate exceedance probabilities for low-mass, high-probability events. If one is interested in the former type of event, FORM may be seen as providing a "conservative" estimate of the exceedance probability. On the other hand, if the latter case is of interest, FORM tends to provide an overly optimistic estimate.

Computational Issues

In order to compare the computational demands of FORM and MC simulation, we consider the time required to compute the results shown in Figure 6. All calculations were done with a DEC ALPHA workstation. The computational requirements of MC simulation depend on the number of realizations used.

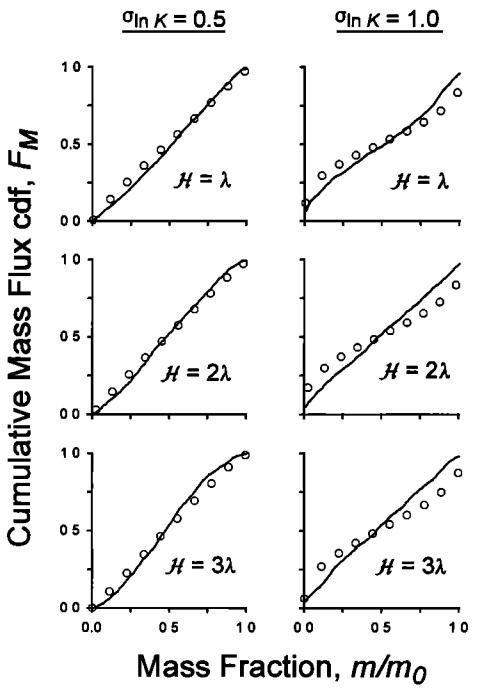

Figure 7. Computed cumulative mass flux cdfs for a control plane located a distance $\mathcal{D}$ = 7λ downstream from an initial solute body. Monte Carlo results are shown with solid lines, FORM results with open circles.

Each of the six MC results shown in Figure 6 required generating 1000 realizations of the conductivity field plus 1000 runs of the transport model. The approximate CPU time needed for the six MC results was 56 hours. The computational requirements of FORM depend on the number of points computed on F_M. We computed 10 points for each plot, and the approximate CPU time required for the three FORM results in the left column of Figure 6 was 6.5 hours. The FORM results in the right column for different $\sigma_{\ln K}$ were then computed by taking advantage of the invariance of $\mathbf{K}^{DP}$ as described above, and this required only seconds of additional CPU time, meaning the total CPU time needed to generate the FORM results in Figure 6 was 6.5 hours. Thus FORM required approximately 8.5 times less CPU time than MC simulation to obtain the results in Figure 6. The advantage of FORM would of course be greater if we had computed results for a whole series of $\sigma_{\ln K}$, because FORM would obtain those additional results with a negligible amount of computational effort whereas MC simulation would require 1000 simulations for each additional result. Conversely, if we had considered only one of the columns in Figure 6, the invariance of $\mathbf{U}^{DP}$ would not be any advantage, and FORM would require about 4.25 times less CPU time that MC simulation.

Conclusions

We have evaluated FORM as a method for predicting solute transport and applied the method to compute the cumulative mass flux cdf for solute crossing a control plane in a heterogeneous porous formation. The following conclusions can be drawn:

1. Except in the case of a constant or monotonically increasing solute source, FORM is not well suited for computing point concentration cdfs because the problem formulation is inconsistent with FORM's premise that the limit state surface divides the parameter space into two regions. FORM may be applied to transport simulation in a consistent manner by formulating the problem in terms of the cumulative mass flux across a control surface.

2. If FORM is not more efficient than Monte Carlo methods, the methodology is of little value for computing distribution functions. When a problem has a large number of random parameters, FORM will not have a computational advantage over MC methods if the perturbation method is used to compute the gradients used in the FORM algorithm. If special sensitivity methods are used to compute the gradients, FORM may prove computationally advantageous.

3. Similarly, the second-order reliability method will not provide a computational advantage over Monte Carlo methods when there is a large number of parameters because of the additional function evaluations that are necessary to construct the second-order approximation. In contrast to FORM, there does not appear to be any way to avoid these evaluations. SORM may become useful for predicting transport in heterogeneous media, however, if it is possible to reduce the number of discretized parameters [e.g., *Mok et al.*, 1994; *Skaggs and Barry*, 1997; *Hamed et al.* 1996a].

4. Based on our simulations, FORM provides accurate estimates of the cumulative mass flux cdf (F_M) in mildly heterogeneous porous media ($\sigma_{\ln K} = 0.5$). In more strongly heterogeneous media ($\sigma_{\ln K} = 1.0$), FORM is less accurate, overestimating low values of F_M and underestimating high values. The size of the initial solute body and the solute travel distance had only a minor effect on FORM's accuracy.

5. In our simulations FORM was more efficient than the Monte Carlo methodology, requiring approximately 8 times less CPU time to compute all of the presented results. FORM computes points on a cdf independently of one another, so FORM's advantage would have been greater if fewer points on the cdf were desired. FORM's efficiency was partly due to the fact that the design point in untransformed parameter space is independent of $\sigma_{\ln K}$ when the covariance of the discretized conductivity can be written $\mathbf{C} = \sigma^2_{\ln K}\mathbf{R}$; this allows one to obtain distribution functions for a series of $\sigma_{\ln K}$ values with only slightly more effort than is required to compute a single distribution function. More importantly, FORM's superior efficiency was dependent on using a transport code that employed an adjoint sensitivity method to compute the required gradients, and developing such computer programs requires significant effort.

Acknowledgments. This work was supported by the Australian Research Council.

References

Ahlfeld, D. P., J. M. Mulvey, G. F. Pinder, and E. F. Wood, Contaminated groundwater remediation design using simulation, optimization, and sensitivity theory, 1, Model development, *Water Resour. Res.*, *24*(3), 431–441, 1988.

Barry, D. A., and G. Sposito, Application of the convection-dispersion model to solute transport in finite soil columns, *Soil Sci. Soc. Am. J.*, *52*(1), 3–9, 1988.

Barry, D. A., J. Coves, and G. Sposito, On the Dagan model of solute transport in groundwater: Application to the Borden Site, *Water Resour. Res.*, *24*(10), 1805–1817, 1988.

Bear, J., *Dynamics of Fluids in Porous Media*, Elsevier, New York, 1972.

Cawlfield, J. D., and M.-C. Wu, Probabilistic sensitivity analysis for one-dimensional reactive transport in porous media, *Water Resour. Res.*, *29*(3), 661–672, 1993.

Christakos, G., *Random Field Models in Earth Sciences*, Academic, San Diego, Calif., 1992.

Cvetkovic, V., A. M. Shapiro, and G. Dagan, A solute flux approach to transport in heterogeneous formations, 2, Uncertainty analysis, *Water Resour. Res.*, *28*(5), 1377–1388, 1992.

Dagan, G., *Flow and Transport in Porous Formations*, Springer-Verlag, New York, 1989.

Dagan, G., V. Cvetkovic, and A. Shapiro, A solute flux approach to transport in heterogeneous formations, 1, The general framework, *Water Resour. Res.*, *28*(5), 1369–1376, 1992.

Der Kiureghian, A., and P.-L. Liu, Structural reliability under incomplete probability information, *J. Eng. Mech.*, *112*(1), 85–104, 1986.

Der Kiureghian, A., H.-Z. Lin, and S.-J. Hwang, Second-order reliability approximations, *J. Eng. Mech.*, *113*(8), 1208–1225, 1987.

Gelhar, L. W., *Stochastic Subsurface Hydrology*, Prentice-Hall, Englewood Cliffs, N. J., 1993.

Hamed, M. M., J. P. Conte, and P. B. Bedient, Probabilistic screening tool for ground-water contamination assessment, *J. Environ. Eng., N. Y.*, *121*(11), 767–775, 1995.

Hamed, M. M., P. B. Bedient, and C. N. Dawson, Probabilistic modeling of aquifer heterogeneity using reliability methods, *Adv. Water Resour.*, *19*(5), 277–295, 1996a.

Hamed, M. M., P. B. Bedient, and J. P. Conte, Numerical stochastic analysis of groundwater contaminant transport and plume containment, *J. Contam. Hydrol.*, *24*(1), 24, 1996b.

Huyakorn, P. S., and G. F. Pinder, *Computational Methods in Subsurface Flow*, Academic, San Diego, Calif., 1983.

Jang, Y.-S., N. Sitar, and A. Der Kiureghian, Reliability analysis of contaminant transport in saturated porous media, *Water Resour. Res.*, *30*(8), 2435–2448, 1994.

Liu, P.-L., and A. Der Kiureghian, Optimization algorithms for structural reliability, *Struct. Safety*, *9*, 161–177, 1991.

Madsen, H. O., S. Krenk, and N. C. Lind, *Methods of Structural Safety*, Prentice-Hall, Englewood Cliffs, N. J., 1986.

Melching, C. S., An improved first-order reliability approach for assessing uncertainties in hydrologic modeling, *J. Hydrol.*, *132*, 157–177, 1992.

Melching, C. S., B. C. Yen, and H. G. Wenzel Jr., A reliability estimation in modeling watershed runoff with uncertainties, *Water Resour. Res.*, *26*(10), 2275–2286, 1990.

Mok, C. M., N. Sitar, and A. Der Kiureghian, Numerically efficient reliability analysis of groundwater flow and subsurface contaminant transport, in *Computer Methods and Advances in Geomechanics, Proceedings of the Eighth International Conference on Computer Methods and Advances in Geomechanics*, vol. 2, edited by H. J. Siriwardane and M. M. Zaman, pp. 1153–1158, A. A. Balkema, Rotterdam, 1994.

Piggott, J. H., and J. D. Cawlfield, Probabilistic sensitivity analysis for one-dimensional contaminant transport in the vadose zone, *J. Contam. Hydrol.*, *24*(2), 97–115, 1996.

Pinder, G. F., and W. G. Gray, *Finite Element Simulation in Surface and Subsurface Hydrology*, Academic, San Diego, Calif., 1977.

Schanz, R. W., and A. Salhotra, Evaluation of the Rackwitz-Fiessler uncertainty analysis method for environmental fate and transport models, *Water Resour. Res.*, *28*(4), 1071–1079, 1992.

Shapiro, A. M., and V. D. Cvetkovic, Stochastic analysis of solute arrival time in heterogeneous porous media, *Water Resour. Res.*, *24*(10), 1711–1718, 1988.

Sitar, N., J. D. Cawlfield, and A. Der Kiureghian, First-order reliability approach to stochastic analysis of subsurface flow and contaminant transport, *Water Resour. Res.*, *23*(5), 794–804, 1987.

Skaggs, T. H., and D. A. Barry, Sensitivity and reliability analysis of groundwater contaminant transport, in *Computational Techniques and Applications: CTAC95*, edited by R. L. May and A. K. Easton, pp. 701–708, World Sci., Singapore, 1996a.

Skaggs, T. H., and D. A. Barry, Sensitivity methods for time-continuous, spatially discrete groundwater contaminant transport models, *Water Resour. Res.*, *32*(8), 2409–2420, 1996b.

Skaggs, T. H., and D. A. Barry, Assessing uncertainty in subsurface solute transport: Efficient first-order reliability methods, *Environ. Software*, in press, 1997.

Sudicky, E. A., The Laplace transform Galerkin technique: A time-continuous finite element theory and application to mass transport in groundwater, *Water Resour. Res.*, *25*(8), 1833–1846, 1989.

Wu, M. C., and J. D. Cawlfield, Probabilistic sensitivity and modelling of two-dimensional transport in porous media, *Stochastic Hydrol. Hydraul.*, *6*, 103–121, 1992.

D. A. Barry, Department of Environmental Engineering, University of Western Australia, Nedlands, WA 6907, Australia.

T. H. Skaggs, U.S. Salinity Laboratory, 450 W. Big Springs Rd., Riverside, CA 92507. (e-mail: tskaggs@ussl.ars.usdg.gov)

(Received June 28, 1996; revised December 31, 1996; accepted February 26, 1997.)