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Ecological Modelling 182 (2005) 355–370

ECOLOGICAL MODELLING

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A fractal investigation of solute travel time in a heterogeneous aquifer: transition probability/Markov chain representation

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Abstract

Solute transport behavior in heterogeneous type structures is generally assessed in terms of its basic statistical properties, such as mean, variance, and correlation. With increasing evidence that the solute transport process in subsurface aquifers exhibits some degree of order at multiple scales (i.e. fractal or scaling), it is crucial to investigate if random representations of aquifers could also explain fractal solute transport behavior. Here, we investigate transport processes that occur in first order Markov chain-type aquifer structures, defined by transition probabilities between constituent hydrofacies. Markov chain structures possess certain advantages with respect to data requirements when compared to traditional Gaussian approaches. The statistical moment scaling function method is employed to investigate solute travel times. The results indicate the potential presence of multi-fractal behavior in the solute transport process, revealing the ability of the transition probability/Markov chain (TP/MC) approach to represent aquifer structures that give rise to fractal solute transport. A sensitivity analysis of the solute transport behavior to the four principal hydrostratigraphic parameters in the TP/MC approach indicates that the degree of fractality increases with an increase in: (1) the number of facies; (2) the volume proportions of the coarse sediments; (3) the dip to vertical mean length ratio; and (4) the order of bedding sequences.

Keywords: Aquifer heterogeneity; Groundwater; Solute transport; Transition probability/Markov chain; Fractals; Sensitivity analysis

1. Introduction

As our reliance on groundwater resources increases, modeling and prediction of subsurface transport phenomena continue to be important topics for hydrogeological research. Flow and solute transport in natural subsurface formations are strongly affected by the hydraulic properties of the medium. Hence, reliable assessment of solute transport requires detailed aquifer characterization. Such characterization, however, is often difficult in subsurface formations, where multiple scales of heterogeneity may exist. As physical and economical constraints limit data collection, considerable uncertainty arises about aquifer heterogeneity patterns, which, in turn, leads to uncertain solute transport predictions.

Research over the years has resulted in the formulation of a variety of approaches and the development of numerous mathematical models to quantify both aquifer heterogeneity and solute transport prediction uncertainty. The existing approaches to aquifer heterogeneity characterization and uncertainty modeling may broadly be divided into three groups: (1) deterministic flow and transport models with heterogeneous aquifers characterized by geologic or hydraulic process-based models or by using descriptive methods (c.f. Koltermann and Gorelick, 1992); (2) stochastic flow and transport modeling based on Gaussian random fields (e.g. Dagan, 2000; Govindaraju, 2002); and

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doi:10.1016/j.ecolmodel.2004.04.010

(3) stochastic (mostly numerical) modeling of flow and transport in structure-imitating, non-Gaussian indicator or Boolean type random fields generated using (geo)statistical approaches (e.g. Desbarats, 1990; Scheibe and Freyberg, 1995).

Within the indicator geostatistical framework, Carle and Fogg (1996) proposed a significant variant of the traditional indicator geostatistical approaches in order to better handle typical hydrogeologic problems. This approach, known as transition probability/Markov chain (TP/MC) approach, readily incorporates "soft" geologic information into Markov-chain models of spatial variability to produce geologically plausible realizations of subsurface heterogeneity. The approach describes the aquifer hydrogeology in terms of its major hydrostratigraphic units (facies) rather than by extensive knowledge of the aquifer hydraulic conductivity distribution (the latter being the basis for Gaussian stochastic models). The hydrostratigraphy is characterized in a probabilistic manner by four geostatistical model parameters: (1) number of major textural categories (i.e. facies); (2) volume proportions of categories; (3) mean lengths (and thereby anisotropy ratio of mean length) of facies; and (4) juxtapositional tendencies (i.e. degree of entropy) among the facies. These parameters can be estimated either empirically through direct measurement (from well logs, drilling logs and soil survey maps) or through inference based on qualitative geologic interpretation (e.g. Carle, 1996; Weissmann et al., 1999). The ability of this approach to provide reliable representations of subsurface heterogeneity has been effectively demonstrated by Carle (1996) and Carle et al., 1998. The usefulness and appropriateness of the approach for natural subsurface aquifers have also been successfully tested by applying it to alluvial aquifer systems, such as the Lawrence Livermore National Laboratory (LLNL) site (e.g. Carle, 1996; Fogg et al., 2000) and the Kings River alluvial fan (e.g. Weissmann and Fogg, 1999; Weissmann et al., 1999) in California.

The present study investigates: (1) the degree to which solute transport in TP/MC type aquifers exhibits fractal behavior; and (2) the sensitivity of the fractal solute transport behavior to aquifer heterogeneity characterized by the above four hydrostratigraphic parameters. The solute transport process is simulated by integrating the TP/MC model with a groundwater flow model (MODFLOW) and a random walk particle transport model (RWHet). Time series of solute (particle) transport are analyzed to investigate the possible existence of fractal behavior. The statistical moment scaling function method is employed to investigate the possible presence of fractal behavior. The influence of the hydrostratigraphic parameters on the (fractal) behavior of solute transport is investigated by changing the number of facies, volume proportions of facies, anisotropy conditions in mean lengths, and entropy conditions (juxtapositional tendencies) in the TP/MC model. The western San Joaquin Valley aquifer system in California (e.g. Belitz and Phillips, 1995) is considered as a reference system to perform the analysis.

Fractal behavior in subsurface solute transport phenomena has been addressed in a number of studies (e.g. Hewett, 1986; Wheatcraft and Tyler, 1988; Benson et al., 2001; Berkowitz and Scher, 2001; Puente et al., 2001a,b). Differences in opinion exist as to the type of fractal behavior (mono- or multi-fractal) in transport phenomena, the underlying mechanisms involved, and the appropriate predictive methods. For instance, Benson et al. (2001) suggest that a mono-fractional derivative in the advective–dispersion equation may be adequate for solute transport predictions, while Puente et al. (2001a,b) use a (deterministic) multi-fractal approach to plume evolution using a model independent of the advection–dispersion equation. The present study neither participates in this continuing debate nor makes any a priori assumption regarding the presence or absence of fractal behavior (and its type) in underlying aquifer materials. Rather, it only employs the statistical moment scaling function method to investigate the possible presence of fractal behavior (and its type) in solute travel times. However, the novelty of this study lies in the investigative approach and the application to solute transport in heterogeneous aquifers characterized by the transition probability/Markov chain approach.

The organization of this paper is as follows. Section 2 reviews the transition probability/Markov chain geostatistical model, groundwater flow model, and particle transport model used in this study for simulating the solute transport process, and Section 3 presents a brief account of the statistical moment scaling function method. Details of the western San Joaquin Valley aquifer system, data generated, analyses performed, and results obtained are presented and discussed in Section 4. Important conclusions drawn from this study are reported in Section 5.

2. Models for aquifer medium representation, flow dynamics, and solute transport

For the study of solute transport in Markov chain random fields, we integrate a transition probabilility/Markov chain geostatistical model (for aquifer heterogeneity representation), a groundwater flow model (for modeling the groundwater flow process), and a random walk particle transport model (for solute transport modeling). We briefly review the basic mathematical concepts of these models.

2.1. Transition probability/Markov chain (TP/MC) geostatistical model

In the transition probability/Markov chain model (Carle, 1996, 1999; Carle and Fogg, 1996), readily observable geologic attributes (e.g. volumetric proportions, mean facies lengths, and juxtapositional tendencies) can be incorporated directly into development of a three-dimensional Markov chain model through a combination of fitting to transition probability measurements and inference from geologic concepts and principles. The Markov chain model is then used in a conditional sequential indicator simulation and simulated quenching (e.g. Deutsch and Journel, 1992) to generate "realizations" of subsurface facies distributions.

The transition probability, $t_{jk}(\boldsymbol{h})$, is defined as the conditional probability that a geologic facies of category k occurs at a spatial location $\boldsymbol{x}+\boldsymbol{h}$ given that a facies of category j occurs at a location $\boldsymbol{x}$:

$$t_{jk}(\boldsymbol{h}) = \Pr\{k \text{ occurs at } \boldsymbol{x}+\boldsymbol{h} \,|\, j \text{ occurs at } \boldsymbol{x}\}, \tag{1}$$

where $0 \le t_{jk}(\boldsymbol{h}) \le 1$, $\boldsymbol{x}$ is a spatial location vector, and $\boldsymbol{h}$ a separation or lag distance vector. Measurements of $t_{jk}(\boldsymbol{h}_\phi)$, where ϕ is the direction, reflect the spatial continuity and juxtapositional tendencies of the facies. Juxtapositional tendencies can be related to entropy (order or disorder) of transition probabilities of embedded occurrence. The entropy, E_j, of juxtapositional tendencies in a direction ϕ is expressed as:

$$E_{j,\phi} = -\sum_{k=1}^{K} r_{jk,\phi} \ln(r_{jk,\phi}) \tag{2}$$

where $r_{jk,\phi} = \Pr\{k \text{ is juxtaposed to } j \text{ in the direction } \phi \,|\, \text{an embedded occurrence of } j\}$ (Hattori, 1976; Carle and Fogg, 1997).

If abundant data are available, the Markov chain model may readily be developed by fitting to measured transition probability values. If the data availability is scarce, then the Markov chain model may be developed from semi-quantitative information on volumetric facies proportions, mean facies lengths, and estimates of juxtapositional tendencies. A Fortran program *T*ransition *PRO*bability *G*eostatistical *S*oftware (TPROGS) has been made available by Carle (1999) for implementing the above procedure, which is used in this study to generate realizations of subsurface heterogeneity.

2.2. Goundwater flow model

In the present study, groundwater flow in the heterogenous aquifer is assumed to be subject to steady-state three-dimensional saturated incompressible flow:

$$\frac{\partial}{\partial x}\left(K_{xx}\frac{\partial \Phi}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_{yy}\frac{\partial \Phi}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_{zz}\frac{\partial \Phi}{\partial z}\right) = 0 \quad \text{on } x,\ y,\ z\,\varepsilon\,\Omega \tag{3}$$

where K_{xx}, K_{yy}, and K_{zz} are hydraulic conductivities in x, y, and z directions, respectively, Φ is hydraulic or piezometric head, and Ω the domain of interest. Also, no flow (Neumann condition) or constant head (Dirichlet condition) are specified for the boundaries of the flow domain, which can be expressed as:

$$\frac{\partial[\Phi(x,y,z)]}{\partial n} = 0 \quad \text{on } x,\ y,\ z\,\varepsilon\,\Gamma_1 \tag{4}$$

$$\Phi(x,y,z) = \Phi_0 \quad \text{on } x,\ y,\ z\,\varepsilon\,\Gamma_2 \tag{5}$$

where Γ is boundary of the domain, $\Gamma = \Gamma_1 + \Gamma_2$, n is the unit vector normal to the boundary pointing outward, and Φ_0 the prescribed head. With these conditions, the flow process in the aquifer medium is simulated using the finite difference flow code MODFLOW (McDonald and Harbaugh, 1988; Harbaugh and McDonald, 1996; Harbaugh et al., 2000).

2.3. Particle transport model

For simulation of solute transport, we use a random-walk particle method (RWPM) to solve the standard advection–dispersion equation (ADE), an approach widely used for transport modeling in heterogeneous media (e.g. Uffink, 1985; Kinzelbach, 1988; Tompson et al., 1987, 1994; Tompson and Gelhar, 1990). The method used in this study is a variant of the standard RWPM that provides both local and global conservation of mass (standard RWPM preserves only global conservation of mass). The method retains the computational advantages of standard RWPMs, including the ability to efficiently simulate solute-mass distributions and arrival times while suppressing errors such as numerical dispersion (LaBolle et al., 1996, 1998, 2000).

The method applies a correction to the standard advection–dispersion equation by including an additional term (to take care of discontinuities), and is given by:

$$\begin{aligned}
&\frac{\partial}{\partial_t}[\Theta(X,t)\,c(X,t)] \\
&\quad = -\sum_i \frac{\partial}{\partial x_i}[v_i(X,t)\,\Theta(X,t)\,c(X,t)] \\
&\qquad + \sum_{i,j}\frac{\partial}{\partial x_i}\left[\Theta(X,t)\,D_{ij}(X,t)\frac{\partial c(X,t)}{\partial x_j}\right] \\
&\qquad + \sum q_k(x,t)\,c_k(x,t)\,\delta_k(x-x_k)
\end{aligned} \tag{6}$$

where X is sample path in space, t the time, c the concentration, v_i the pore water velocity, Θ the effective volumetric water content (or effective porosity), c_k the aqueous phase concentration in the flux q_k of water at x_k, D_{ij} a real symmetric dispersion tensor given as:

$$D_{ij} = (\alpha_T|v| + D'_d)\delta_{ij} + (\alpha_L - \alpha_T)v_i v_j/|v| \tag{7}$$

where α_T and α_L are transverse and longitudinal dispersivities, respectively, and D'_d is effective molecular diffusivity, δ_{ij} the Dirac delta function. We use the program *R*andom *W*alk particle model for simulating transport in *Het*erogeneous Permeable Media (RWHet) for implementing the modified RWPM (LaBolle, 2000).

2.4. Integration of models

The above three programs are linked through a meta-program called *S*ensivity *A*nalysis of *S*tochastic *H*ydrostratigraphy in an *A*quifer-*R*ectangular *B*ox (SASHA-RB) that automatically generates a rectangular aquifer flow domain and associated input files for TPROGS, MODFLOW and RWHet, sequentially executes the three programs, handles data processing and data transfer between the three modules, and processes the final solute transport results. The code and an executable version of this program are available from the authors upon request.

3. Statistical moment scaling function method

In the statistical moment scaling function method (e.g. Frisch and Parisi, 1985; Schertzer and Lovejoy, 1987; Over and Gupta, 1994), the data set is divided into non-overlapping intervals of a certain time/space scale. The ratio of the maximum scale of the field to this interval is termed the “scale ratio”, λ. Thus, λ is inversely proportional to the size of the scale examined. For different scale ratios, λ, the average threshold, $\varepsilon(\lambda, i)$, in each interval, i, is computed and raised to power q, and subsequently summed to obtain the statistical moment, $M(\lambda, q)$:

$$M(\lambda, q) = \sum_i \varepsilon(\lambda, i)^q \tag{8}$$

For a scaling field, the moment, $M(\lambda, q)$, relates to the scale ratio, λ, as

$$M(\lambda, q) = \lambda^{\tau(q)} \tag{9}$$

where $\tau(q)$ may be regarded as a characteristic function of the fractal behavior. If $\tau(q)$ versus q is a straight line, the series exhibits mono-fractal, whereas a convex function of $\tau(q)$ versus q is an indication of multi-fractal behavior (e.g. Frisch and Parisi, 1985; Svensson et al., 1996).

4. Application

4.1. Study area

We use the alluvial aquifer system of the western San Joaquin Valley as an example to study regional solute transport through an alluvial, heterogeneous aquifer system. Three major vertical zones are recognized: an upper semi-confined aquifer, a middle confining layer (referred to as the "Corcoran Clay"), and a lower confined aquifer. Our work focuses on the upper semi-confined aquifer. The geologic material in the upper aquifer consists of Coast Range alluvium in the western region and Sierran sand with shallow flood-basin overburden near the eastern edge of the region along the valley. It is bounded below by the Corcoran Clay, a laterally extensive lacustrine deposit with low hydraulic conductivity, ranging in thickness from about 20 to 120 ft. The drilling log data indicate that the upper semi-confined aquifer is composed of alternative layers of coarse and fine textured materials and the Corcoran Clay contains some amount of interbedded coarse-textured materials. Belitz and Phillips (1995) present an extensive account of the western San Joaquin Valley aquifer system.

This region is selected for analysis because non-reactive solute transport, specifically of salinity, is of particular importance in the region. Saline groundwater with dissolved solids concentrations from 983 to 35,000 mg/l dominates near the shallow water table within salt-enriched shallow alluvial deposits derived from marine sedimentary source rocks. Allochthone salinity generally decreases with depth (Dubrovsky et al., 1993). Historically, groundwater flow bypassed much of the shallow salinity as mountain front recharge was transferred laterally through the aquifer system to the Thalweg, where it discharged into the San Joaquin River. As extensive groundwater development occurred and large water projects were constructed in the middle of the 20th century, agricultural water use significantly altered the flow dynamics in the alluvial aquifer system. Diffuse recharge from irrigation water and regionally distributed pumping at depth transformed groundwater flow dynamics into a system that is regionally dominated by vertically downward net flux. The downward flux has mobilized shallow salinity and is transporting salts perpendicular to the dominant alluvial layering to lower portions of the aquifer. Belitz and Phillips (1995) estimate the regional downward flux to be on the order of 1 ft per year. The presence of interconnected coarse-textured sand and gravel facies within the semi-confined aquifer and the Corcoran Clay could significantly accelerate the migration of highly saline groundwater to the deep semi-confined and confined aquifer zones and potentially lead to the early degradation of water quality in the production zone.

4.2. TP/MC, MODFLOW and RWHet model specifications

For the present investigation, the upper semi-confined aquifer is conceptually represented as simple rectangular, three-dimensional $111 \times 111 \times 111$ grid structure. As numerical edge effects due to the TPROGS implementation are present along the top and bottom, left and right, and front and back of the simulation medium (Steve Carle, personal communications), five layers from each of these edges are eliminated after the random field generation to avoid edge effects. The final structure of the medium for flow and transport modeling contains $101 \times 101 \times 101$ (=1,030,301) cells.

For aquifer heterogeneity representation using the TP/MC model, realizations are generated for varying combinations of the four hydrostratigraphic parameters. Combinations include: (1) two combinations of facies: two facies (sand and clay) versus three facies (sand, clay and loam); (2) 30 combinations of proportions in two facies (i.e. sand from 15% to 60%) and one combination of proportions in three facies (i.e. sand 21.26%, clay 53.28%, and loam 25.46%); (3) three combinations of mean length anisotropy ratios (ratios of dip to strike and dip to vertical mean length are 2:1 and 300:1, 5:1 and 300:1, and 2:1 and 50:1); and (4) three combinations of juxtapositional tendencies or entropies (maximum, field, and low entropy conditions). All these combinations are arbitrarily chosen to investigate the effects of the parameters used in the sensitivity analysis on the solute transport behavior, but the chosen ranges may, to some extent, be considered reasonable representations of the San Joaquin Valley aquifer system.

For flow simulation, the following conditions are assumed: (1) constant head $H_1 = 420$ ft at the top; (2) constant head $H_2 = 385$ ft at the bottom; (3) no flow at the bottom; and (4) no flow at the left and right sides. The aquifer is of the same 404 ft depth in each realization, with a layer thickness of 4 ft. The lateral extent of the simulated aquifer varies with the mean length anisotropy ratios. Hydraulic conductivity (K) is assigned to each cell corresponding to the simulated hydrofacies from each geostatistical realization scenario. Values of hydraulic conductivities of (each) hydrofacies correspond to those for sand and fine-grained facies in the calibrated model by Belitz and Phillips (1995): 31 ft per day for sand, 0.004 ft per day for clay. We assume a low intermediate value of 0.04 ft per day for loam (Weissmann et al., 1999).

As salinity is of particular importance in the western San Joaquin Valley, in the present simulations, saline water is injected to study the solute transport behavior. A single pulse of saline groundwater is injected at the top. Ten particles are uniformly put in every cell in the second layer from the top (since a constant head is set for the top layer). The mass of each particle is weighted by the flux rate of the cell to achieve a flux-weighted unit-time application of solutes. The regional breakthrough curve (BTC) is monitored by recording the number and mass of particles arriving above the bottom layer (in the latter, by normalizing the mass of particles exiting the domain with respect to the total mass released from the source, we obtain travel time probability). It is found that the maximum travel time is bounded at approximately 2000 years. Local dispersion and molecular diffusion are ignored such that the BTCs represent only variations in travel time due to local variability in advection velocity. For simplicity, effective porosity is assumed uniform at 30%.

A total of 90 simulations are performed with two facies (30 proportions × three anisotropy conditions) and nine simulations are performed with three facies (one proportion × three anisotropy conditions × three entropy conditions).

The following points must be noted in regard to the assumptions made in the present theoretical simulations against the real western San Joaquin Valley aquifer system. In the real system, application of irrigation water at the top of the system has resulted in large vertical head gradients, whereas the horizontal gradients are typically much smaller (Belitz and Phillips, 1995). In reality, therefore, differences in density of saline water exist. However, the present simulations assume, for convenience, that salt particles are uniformly distributed in each cell in the second layer from the top of the model. Also, the assumption, in the simulations, of a no flow boundary in the immediate vicinity of the observation plane might influence the results, but no effort is made herein to study this issue.

4.3. Data, analyses and results

4.3.1. Number of facies

Two conceptual facies models are investigated for their effects on solute transport: a simple two-facies model that effectively corresponds to an indicator random field generated from a cutoff in a Gaussian random field (Carle and Fogg, 1996); and a three-facies model representing three major hydrofacies in alluvial systems: sand and gravel facies, muddy sand facies, and mud facies (Weissmann et al., 1999). A variety of proportions of facies (sand content varying from 15 to 60% and clay forming the remainder) are considered in the two-facies model [see below for further details], whereas only one combination of proportions (sand 21.26%, clay 53.28%, and loam 25.46%) is considered in the three-facies model. Fig. 1(a) shows an example breakthrough curve for salt transport in two facies (with sand content 20%), while the corresponding plot for the three facies model is shown in Fig. 2(a). Both cases have a mean length anisotropy condition of 2:1 and 300:1 and maximum entropy (low order or low juxtapositional tendency). Significant differences in the shape of the breakthrough curve can be observed between the two- and three-facies models, indicating the importance of using appropriate number of facies in the model for aquifer heterogeneity representations. However, here we focus our attention not on the statistical moments of the BTC, but on the fractal properties of the observed oscillations in the BTC.

Before presenting the results of fractal analysis, a brief description of the shape of the breakthrough curve, assessed in terms of skewness, may be useful to understand the manner in which the particles arrive. Right skewed (or negatively skewed) structure means that the curve extends far into the negative side of the Cartesian graph, which is an indication that most of the particles come out of the simulation domain very late in the simulation process. The reason for this could be that the hydraulic conductivities of materials are very low; for example, the proportion of

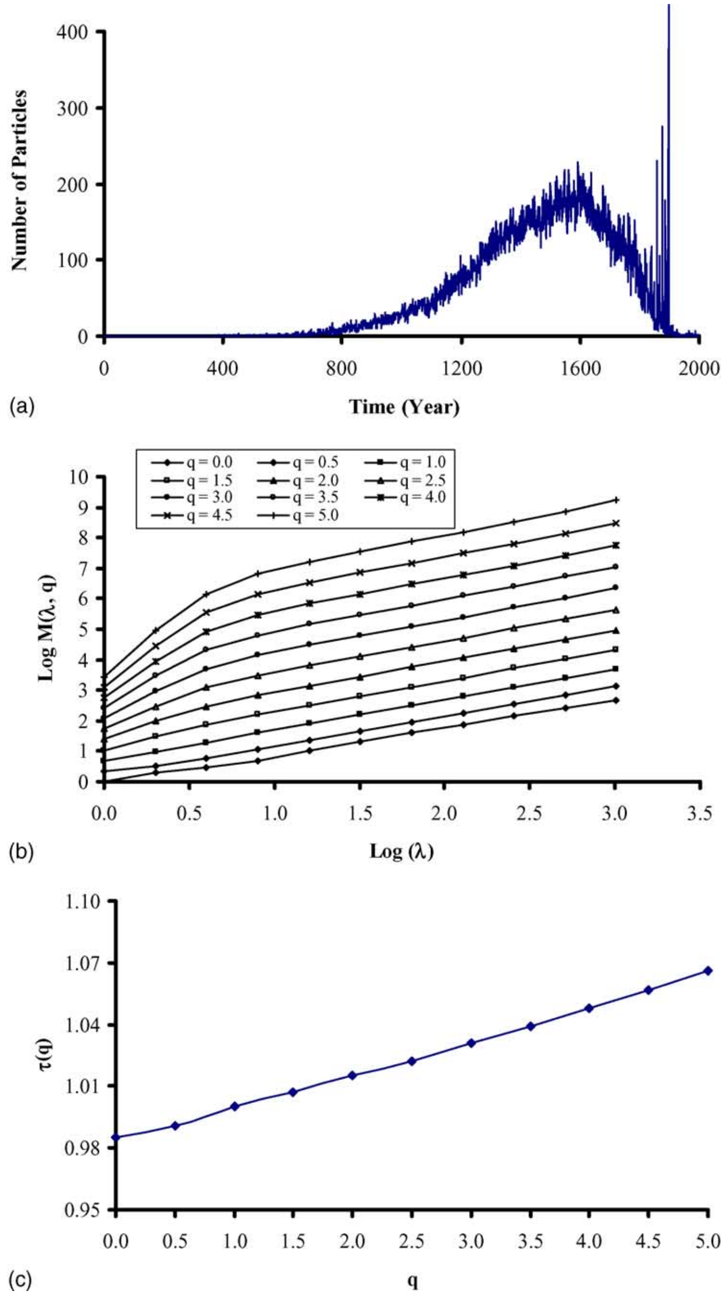

Fig. 1. Results of fractal analysis of solute transport phenomenon in two facies medium (sand 20% and clay 80%) with anisotropy condition 2:1 and 300:1: (a) time series plot of particle arrival; and (b) and (c) statistical moment scaling function.

clay is significantly larger than that of sand (as is the case in Fig. 1(a)). Furthermore, due to particle entrapment in clay early in the transport process, a second peak in the breakthrough curve may also occur, as is seen in Fig. 1(a). This situation is generally termed a "bimodal" breakthrough curve. On the other hand, a left skewed (or positively skewed) structure indicates that most particles come out of the domain early in

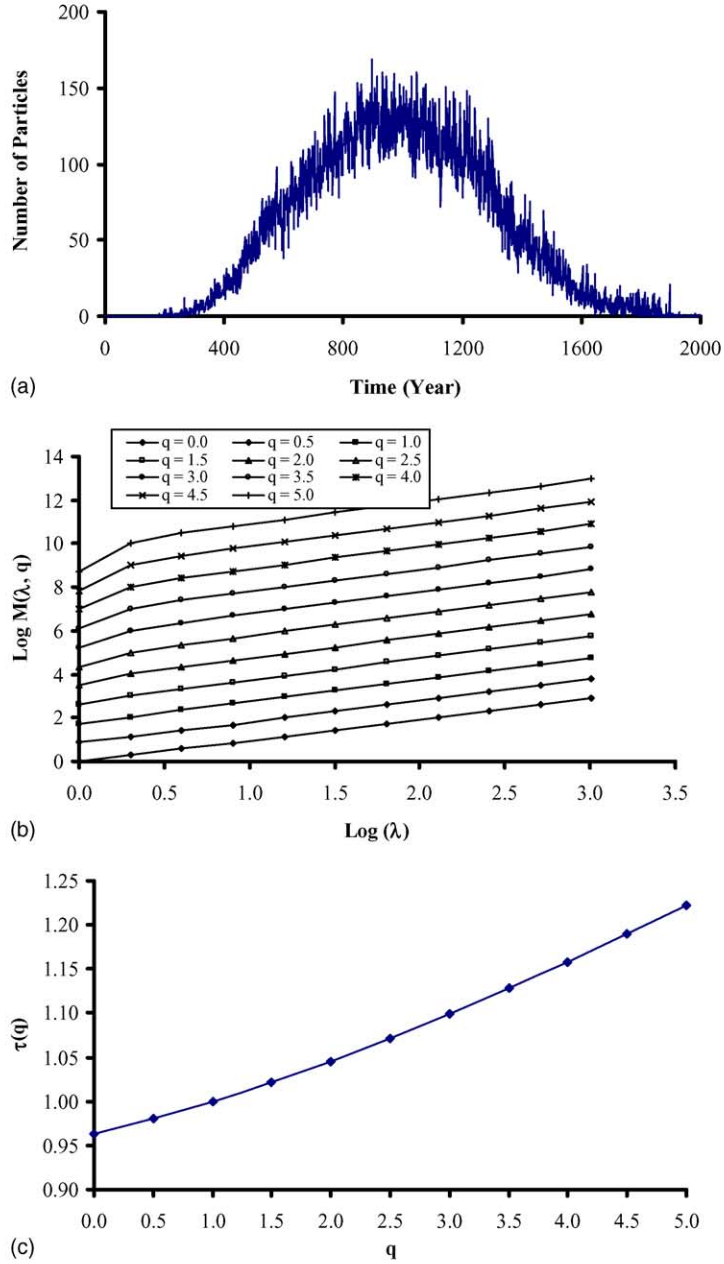

Fig. 2. Results of fractal analysis of solute transport phenomenon in three facies medium (sand 21.26%, clay 53.28%, and loam 25.46%) with anisotropy condition 2:1 and 300:1 and maximum entropy: (a) time series plot of particle arrival; and (b) and (c) statistical moment scaling function.

the simulation process. This may be the case when the sand proportion is significantly larger than the clay proportion [as for the case shown in Fig. 3(e); see below for details]. However, this need not necessarily be the case, since the particle movement and, hence, the breakthrough curve are also influenced by other properties of the medium, such as number of facies and mean length anisotropy ratios. Investigation of the influence of these properties is the purpose of this study.

In the statistical moment scaling function method, the particle transport rate is averaged over successively doubled time intervals (i.e. years) corresponding to successively halved values of the scale ratio λ. Then, for each λ, the q^{th} statistical moment is calculated according to Eq. (8). A log–log plot of the $M(\lambda, q)$

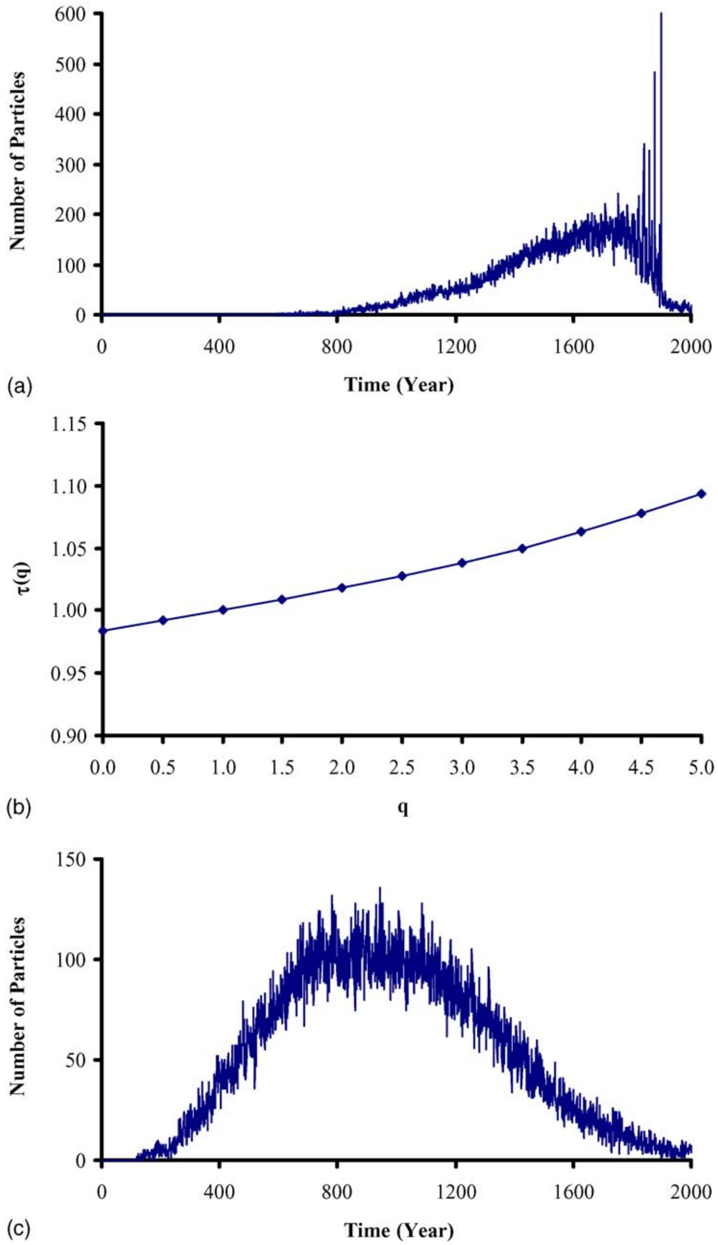

Fig. 3. Effect of volume proportions on solute transport behavior in two facies medium (time series and statistical moment scaling function plots) with anisotropy condition 5:1 and 300:1: (a) and (b) sand 15% and clay 85%; (c) and (d) sand 36% and clay 64%; and (e) and (f) sand 60% and clay 40%.

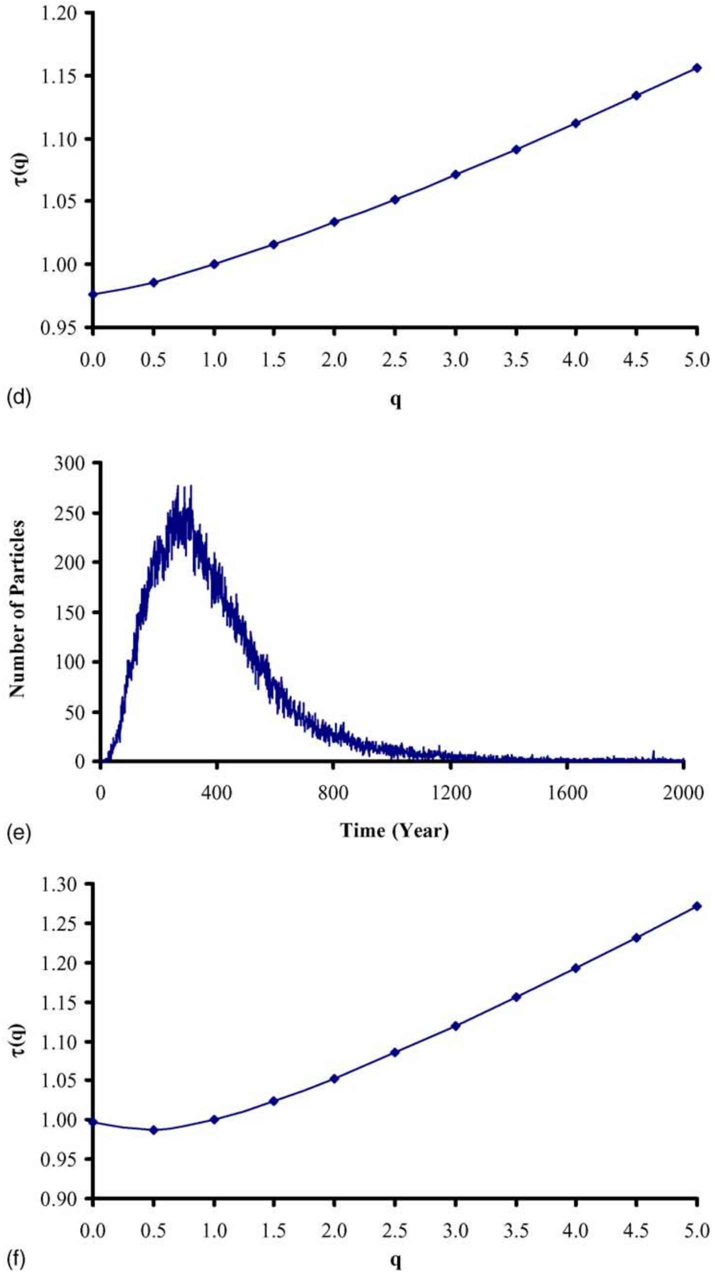

Fig. 3. (*Continued*).

as a function of λ, as expressed by Eq. (9), may be used to investigate the presence of fractal behavior in the particle transport series. If Eq. (9) is valid, the resulting curve would exhibit an approximately linear behavior with a slope that is an estimate of $\tau(q)$. By performing the procedure for different values of q, the entire $\tau(q)$ function can be estimated (Fig. 1(b) and 2(b)). The graphs exhibit large scaling-type regions, allowing reasonably accurate estimation of the slopes, $\tau(q)$, for all the values of q considered.

The $\tau(q)$ versus q functions (Fig. 1(c) and Fig. 2(c)) exhibit slightly convex curvatures (rather than straight lines) in both cases, consistent with the multi-fractal interpretation. Differences in the convex function between these cases are evident, revealing differences in the underlying structure. Greater convexity is observed in the three facies case than in the two facies case (where it is relatively flat), indicating that three facies transport is subject to a higher order dimension function (i.e. higher order multi-fractal model).

4.3.2. Volume proportions of facies

The sensitivity of the solute transport behavior to the volume proportions of facies may be explained from a "connectivity" perspective. Connectivity plays a crucial role in the transport process, since the presence of adjacent similar facies (or different facies) may either accelerate or decelerate the migration of solute particles, depending upon the type of facies. The presence of coarse grained sediments (e.g. sand) accelerates the transport process, whereas a deceleration occurs in the presence of fine grained sediments (e.g. clay). Larger proportions of sand facies (where the clay facies dominates) yield a larger degree of connectivity, resulting in faster downward transport through the aquifer system. Here, we vary sand content from as low as 15% (remaining 85% as clay) to as high as 60% (remaining 40% as clay). Three combinations, corresponding to 15, 36, and 60% sand, are selected to represent low, medium, and high proportions, all at an anisotropy ratio of 5:1 and 300:1 (Fig. 3(a), (c), and (e)).

Significant changes in particle transport behavior are observed as sand content increases. Starting from a right-skewed structure with 15% sand, the BTC becomes nearly symmetrical at 36% sand, and left-skewed at higher sand proportions. The results indicate, in general, the presence of fractal behavior (of multi-fractal type), as convex statistical moment functions are observed across the entire range of sand proportions (Fig. 3(b), (d), and (f)). However, the convexity apparently increases with sand facies proportion, suggesting that the transport process is subject to higher order multi-fractal behavior as the fraction of sand facies increases, at least to 60%. Multi-fractal behaviors are also observed for other anisotropy conditions (2:1 and 300:1, and 2:1 and 50:1). Note that, in the limit of 100% sand facies, the BTC collapses to a single deterministic pulse corresponding to the travel time in a homogeneous sand facies (due to the absence of local dispersion in the model).

4.3.3. Mean length anisotropy

The net downward flux in the aquifer system investigated here is a major departure from groundwater transport systems typically investigated, where groundwater flow is generally assumed to be approximately horizontal and parallel to the major sedimentary bedding planes. Most groundwater transport investigations are concerned with lateral solute displacement parallel to major bedding planes. In our system, regional solute transport is perpendicular to the major alluvial bedding planes with facies mean length significantly longer across the mean flow direction than along the mean flow direction. In the limit, as the horizontal to vertical mean length anisotropy ratios becomes infinitely large, the flux system represents flow across a perfectly layered system with the resulting travel time being identical for all particles. The finite mean length in the TP/MC simulations, however, results in a complex heterogeneous structure of the aquifer medium, with significant connectivity across layers. This forces groundwater flow to follow a tortuous path to reach the bottom of the aquifer. Mean length anisoptropy ratios directly influence the tortuosity of the flow path.

The fractal analyses are performed for all three anisotropy conditions for each of the 30 combinations (of sand content) in the two facies medium and each of the three combinations (of juxtapositional tendencies) in the three facies medium. For the purpose of brevity, the analyses and results of two of the combinations in the two facies medium (i.e. sand proportion equal to 20 and 50%) are presented here.

In the case of 20% sand, the time series of the solute transport phenomenon exhibits a right-skewed structure for anisotropy condition 2:1 and 300:1 and becomes gradually less right-skewed for lower anisotropy ratios 5:1 and 300:1, and 2:1 and 50:1, respectively [figures not presented]. On the other hand, the corresponding time series for the case of 50% sand exhibit, in order, a Gaussian-type structure followed by a partial left-skewed structure in the most anisotropic medium and a completely left-skewed one in the least anisotropic medium [figures not presented]. For both 20 and 50% sand proportions, multi-fractal behaviors in the solute transport phenomena are observed,

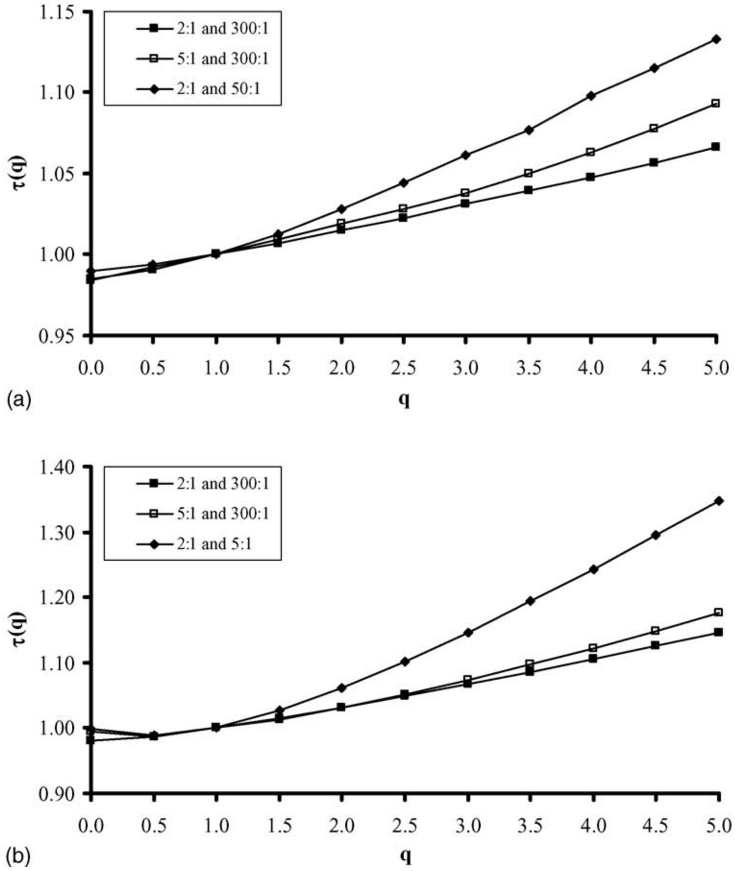

Fig. 4. Sensitivity of fractal behavior of solute transport phenomena to anisotropy conditions in two facies medium: (a) sand 20% and clay 80%; and (b) sand 50% and clay 50%.

as convex functions of $\tau(q)$ versus q are present (Fig. 4(a) and (b)). However, the multi-fractality of the phenomenon varies with the anisotropy condition. Also, the multi-fractality increases more with an increase in the ratio of dip to vertical mean length (i.e. from 2:1 and 300:1 to 2:1 and 50:1) than with an increase in the ratio of dip to strike mean length (i.e. from 2:1 and 300:1 to 5:1 and 300:1). Clearly, the dip to vertical mean length is critical, which is due to the vertical mean flow in the system. The multi-fractality of the solute transport phenomenon increases with an increase in the sand proportion. These results indicate that when an aquifer medium contains a high proportion of sand (i.e. coarse sediments) with high ratio of dip to vertical mean length, the fractality of the solute transport phenomenon may be more complex.

4.3.4. Juxtapositional preferences

From a connectivity point of view, another aquifer characteristic that plays an important role is the degree of order/disorder in the facies structure, i.e. the entropy of the facies assemblage. High entropy is synonymous with less order (or less probability that one particular facies is preferentially located adjacent to another particular facies). In classical stochastic theory, the aquifer medium is generally considered to exhibit high (or maximum) entropy, as a Gaussian random field is assumed for hydraulic conductivity. In the TP/MC approach, different levels of entropy may be considered to represent the medium, which are indicated by “juxtapositional tendencies” of the bedding sequences. Therefore, the TP/MC approach has the ability to represent different possibilities of order of facies, anywhere between determinism and randomness.

Here, we consider three different entropy conditions in the three facies medium: maximum entropy (low order, low juxtapositional tendency), field entropy (medium order), and low entropy (high order). For each of these entropy conditions, BTCs are simulated for all of the above three anisotropy conditions (i.e. 2:1 and 300:1, 5:1 and 300:1, and 2:1 and 50:1), and for volume proportions of sand 21.26%, clay 53.28%, and loam 25.46% [the same to the one presented in Section 4.3.1].

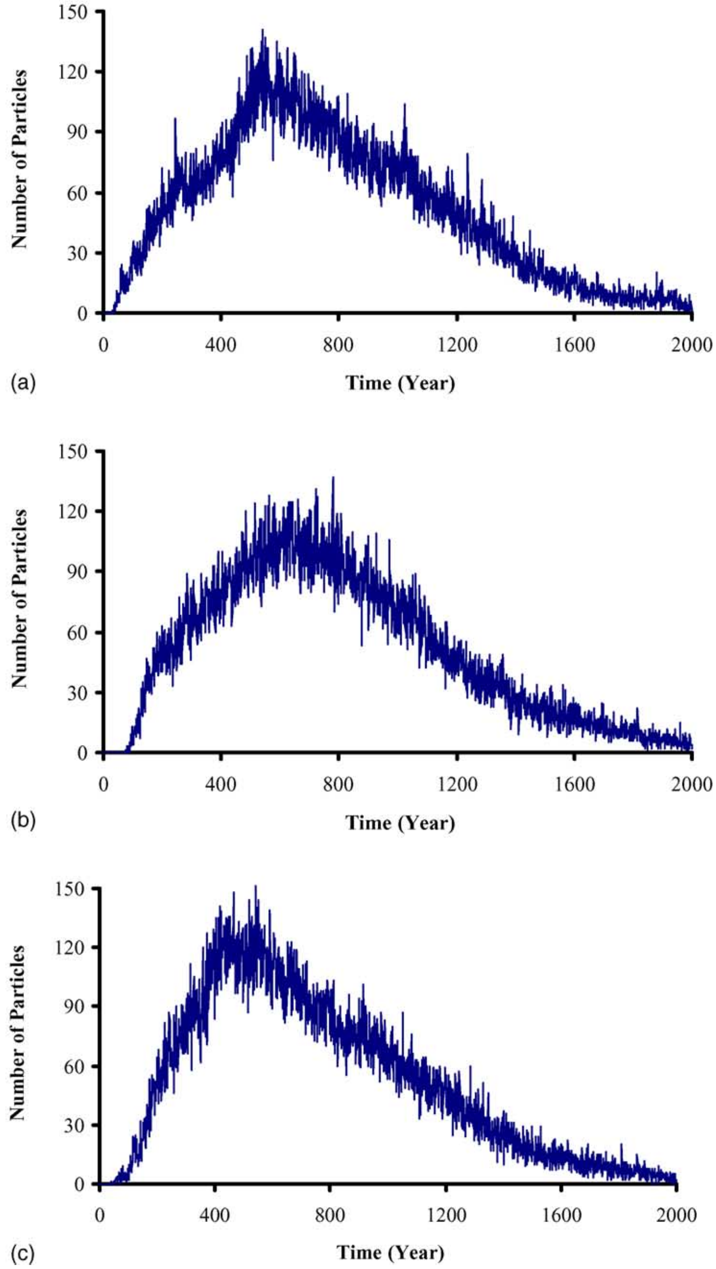

Fig. 5. Effect of entropy on solute transport behavior in three facies medium (sand 21.26%, clay 53.28%, and loam 25.46%) with anisotropy condition 2:1 and 50:1: time series plots: (a) maxium entropy, (b) field entropy, and (c) low entropy; and (d) moment scaling function.

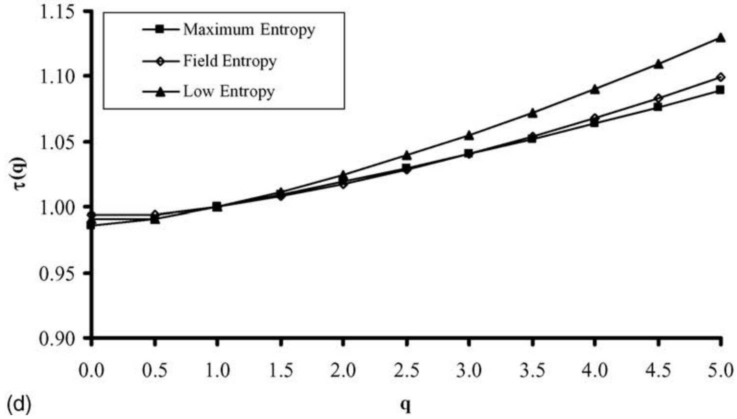

Fig. 5. (*Continued*).

Fig. 5(a)–(c) presents the time series of particle transport in the three facies medium with maximum, field, and low entropy conditions, respectively. The plots shown correspond to anisotropy condition 2:1 and 50:1. No significant difference is observed in the shape of the BTC. The results of statistical moment scaling function analysis [i.e. $\tau(q)$ versus q plots] for these three time series exhibit convex $\tau(q)$ versus q functions (Fig. 5(d)), indicating the potential presence of multi-fractal behaviors in the solute transport phenomena for all entropy conditions. However, the order of multi-fractality is found to increase slightly with a decrease in entropy, indicating that the solute transport phenomenon becomes more complex when there is higher order of facies in the medium (high juxtapositional tendencies of bedding sequences). Similar results (multi-fractal behavior and changes in fractality with entropy conditions) are also observed for solute transport phenomena simulated with the other two anisotropy conditions, i.e. 2:1 and 300:1, and 5:1 and 300:1 [figures not presented].

5. Conclusions and potential for further research

The present study investigated the possible existence of fractal behavior in the solute transport phenomenon in a heterogeneous aquifer medium, characterized by a transition probability/Markov chain (TP/MC) model. Time series of solute particle transport were analyzed for this purpose. Application of the statistical moment scaling function method indicated the presence of fractal behavior (of multi-fractal type) in the solute transport process.

A sensitivity analysis of the four hydrostratigraphic parameters involved in the TP/MC model (chosen with the western San Joaquin Valley aquifer system as a reference system) revealed the significance of these parameters on the (fractal) behavior of the solute transport phenomenon. For the cases studied, the complexity (i.e. degree of multi-fractality) of the solute transport dynamics was found to increase with an increase in: (1) the number of facies; (2) the volume proportions of the higher permeable facies (i.e. sand); (3) the ratio of dip to vertical mean length (than ratio of dip to strike mean length); and (4) the order of bedding sequences.

The present findings, by extension, apply also to (two-facies) indicator random fields, which is a special case of the TP/MC model (Carle and Fogg, 1996). As it is common to observe multiple scales in solute transport process (e.g. Wheatcraft and Tyler, 1988; Benson et al., 2001; Berkowitz and Scher, 2001; Puente et al., 2001a, b), the present results are a further confirmation of the usefulness and appropriateness of the TP/MC approach to represent complex aquifer systems that may also give rise to fractal behavior in the associated (flow and) solute transport processes.

Even though the present results are certainly encouraging, their usefulness for practical situations needs to be further assessed.

1. By analyzing only time series of particle arrival at one particular location in the aquifer, the study

limits itself to addressing only the temporal (fractal) dynamical behavior of the solute transport phenomenon. However (flow and), solute transport phenomenon in an aquifer system changes with respect to both time and space. Therefore, study of both temporal and spatial dynamics of the transport process is essential for obtaining realistic and reliable results.
2. The present study provides only inferential information on the fractal structure of the solute transport phenomenon (i.e. multi-fractal type), but quantitative information (e.g. order of multi-fractality and the associated equations) is crucial to understand the intricate details of the underlying scaling behavior.
3. The (multi-) fractal methods used in this study (and also others) have certain limitations when employed to short and noisy data (e.g. Harris et al., 1997). This problem has particular significance when one is dealing with real aquifer data, since such data are often short and always subject to measurement error.

Addressing these issues will be important future steps to determine the practical applicability of these methods.

Acknowledgements

Funding for this work was provided by the Collaborative UC/Los Alamos Research Program (University of California Directed Research Development) and by the University of California Salinity/Drainage Program. The development of the modeling tools and interpretation benefited greatly from our discussions with Dongxiao Zhang, Carlos Puente, and Nels Ruud. The authors thank the three anonymous reviewers for their suggestions, which helped improve the presentation of the work performed.

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