HYDRODYNAMIC CONTROL OF RETENTION IN HETEROGENEOUS AQUIFERS AND FRACTURED ROCK

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# Table of Content

**ACKNOWLEDGEMENTS** .......................................................................................................................... iii

**Table of Content** ........................................................................................................................................ v

**LIST OF APPENDED PAPPERS** ............................................................................................................. vii

**ABSTRACT** .................................................................................................................................................. 1

1. Introduction and background ...................................................................................................................... 1

   1.1. Modelling of flow and transport in heterogeneous aquifers and in fractured rock ...................... 2
   1.2. Tracer experiments in deep rock fractures ......................................................................................... 4
   1.3. Scope of thesis ..................................................................................................................................... 4

2. The LaSAR approach to solute transport .................................................................................................. 5

   2.1. The Lagrangian description of advection .......................................................................................... 5
   2.2. The LaSAR approach of solute transport .......................................................................................... 6
      2.2.1. Retention processes .................................................................................................................... 7
      2.2.2. Coupled mass transfer processes ............................................................................................... 9
      2.2.3. Two important parameters $\tau$ and $\beta$ ................................................................................. 9
   2.3. The Monte-Carlo simulation approach ............................................................................................ 10

3. Hydrodynamic control of retention in heterogeneous aquifers ............................................................... 11

   3.1. Preferential flow (Paper I) ................................................................................................................. 11
      3.1.1. First-order results ....................................................................................................................... 11
      3.1.2. Monte-Carlo simulations ............................................................................................................ 11
      3.1.3. Simulation results ....................................................................................................................... 12
   3.2. Coupled hydraulic and retention heterogeneity (Paper II) ................................................................. 12
      3.2.1. First-order solution of reaction flow path ................................................................................... 13
      3.2.2. Simulation results ....................................................................................................................... 14
      3.2.3. Solute discharge ......................................................................................................................... 15

4. Hydrodynamic control of retention in fractured rock .............................................................................. 16

   4.1. First-order results (Paper III) ............................................................................................................. 17
   4.2. Statistical properties of $\beta$ and $\tau$ (Papers III, IV and VI) ............................................................... 17
      4.2.1. Flow in a single fracture (Cubic law vs quadratic law) ............................................................... 17
      4.2.2. Transport in a single fracture ..................................................................................................... 18
      4.2.3. Internal and global heterogeneous fields (Paper VI) .................................................................. 18
      4.2.4. Flow and transport in a fracture series and impact of internal and global heterogeneities (Paper VI) ................................................................................................................... 19
   4.3. Statistical properties of $\beta$ and $Q$ and their correlations (Papers IV and VI) ................................. 20

5. Site characterization and performance assessment applications ............................................................ 22

   5.1. Evaluation of TRUE experiments in Site characterization scale (Papers VII to XI) ......................... 22
      5.1.1. Modelling of the TRUE-1 field experiments .............................................................................. 23
      5.1.2. TRUE Block Scale modelling (Papers VIII and X) ..................................................................... 26
      5.1.3. Block Scale Continuation modelling (Paper XI) ......................................................................... 26
   5.2. From site characterization to performance assessment modelling (Papers IX and X) ...................... 27
   5.3. Impact of temperature increase on retention of radionuclide (Paper V) ........................................ 28

6. Conclusions .................................................................................................................................................. 29

7. References ................................................................................................................................................... 32
LIST OF APPENDED PAPPERS

This thesis is based on the following papers, which are appended at the end of the thesis and referred to by their Roman numbers (I-VI). The papers are listed in their chronological order that may not be the same as when they are referred to in the thesis.


The following publications related to the research of this thesis are equally important, but not appended in the thesis; they will be referred to as "Papers" VII-XI.


Publications VII to XI will be available on the website of the Swedish Nuclear Fuel and Waste Management Company (SKB). (http://www.skb.se).
ABSTRACT

In this thesis, fluid flow and solute transport in heterogeneous aquifers and particularly in fractured rock have been investigated using Lagrangian Stochastic Adveotive-Reaction (LaSAR) framework. The heterogeneity of the aquifer structure or fracture configuration, as well as the various reaction/retention processes have been considered in the modelling approach. Advection and retention processes are considered to be the dominant transport processes.

Monte-Carlo simulation results for transport of nonreactive tracers in 2D generic heterogeneous aquifers indicate that the travel time $\tau$ can be well approximated by a lognormal distribution up to a relative high degree of heterogeneity of the aquifers. Comparison between the Monte-Carlo simulation results and the results of first-order approximation reveals that the analytical solutions of the statistical moments of $\tau$ are valid only when the variability of the aquifer properties is small. For reactive tracers, Monte-Carlo simulations have been conducted by accounting for spatial variability of both hydraulic conductivity and one sorption parameter simultaneously. The simulation results indicate that the reaction flow path $\mu$ is a nonlinear function of distance for shorter distance, linear function for longer distance, and also that $\mu$ and $\tau$ are well correlated over the considered parameter range. The parameter $\beta$, which is purely determined by the flow conditions, quantifies the hydrodynamic control of retention processes for transport of tracers in fractures. Numerical simulations have been performed to study the statistical properties of the parameter $\beta$, travel time $\tau$ and flow rate $Q$ in a single heterogeneous fracture and in a sequence of fractures. The results of Monte-Carlo simulations indicate that the parameter $\beta$ and $\tau$ are correlated with a power-law relationship $\beta \sim \tau^\alpha$. The correlation between $\beta$ and the flow rate $Q$ have also been studied and an inverse power-law relationship $\beta \sim Q^{-\gamma}$ is proposed. The establishment of these relationships provides a link between the parameter $\beta$ and measurable parameters $\tau$ (or $Q$).

The LaSAR approach has been applied for prediction, evaluation and interpretation of the results of a number of tracer tests (TRUE-1, TRUE Block Scale and TRUE Block Scale Continuation) conducted by SKB at the Äspö site for tracer transport in fractures. The breakthrough curves may be predicted reasonably well, provided that the retention parameters, boundary conditions and hydraulic properties of the domain are given. The evaluation of TRUE tests indicates that the retention occurs mainly in the rim zone on site characterization time scales, while on the performance assessment time scale, diffusion and sorption in the unaltered rock matrix are likely to become dominant retention mechanisms.

Key words: Aquifer; Fracture; groundwater; First-order solution; Monte-Carlo simulation; Nuclear waste; Heterogeneity; Retention processes; Matrix diffusion; Prediction; Evaluation; Travel time; Flow rate; Temporal moments; Site characterization; Performance assessment.

1. INTRODUCTION AND BACKGROUND

Groundwater contamination by various pollutants has become a worldwide problem. Mining activities, fertilizer and pesticide utilization in agriculture, landfill of municipal wastes, discharge of wastewater from a variety of industrial plants, acid rain caused by burning of fossil fuels all contribute to groundwater contamination. Nuclear waste disposal in geological formations, if not subject to stringent safety regulations, could also pose a potential threat to the quality of deep groundwater that will eventually come to the subsurface aquifers.

Protection of groundwater from pollution requires quantitative evaluation and prediction of transport of the contaminants. Due to the constraints of costs and accessibility, complete characterization through field measurements is seldom feasible. Different models of fluid flow and solute transport in
natural aquifers have to be used. These models have traditionally also been applied to areas like chemical engineering (transport in porous beds), petroleum reservoir engineering (multi-phase transport) and recently to nuclear waste management. As groundwater pollution becomes an ever-increasing threat to ecosystems and fresh water resources for human consumption, these studies have received more and more attention in the last two decades. The active participation of nuclear power industry for safe disposal of its nuclear wastes has contributed to the ever-increasing interests in such researches.

In Sweden as well as in many other countries, the spent nuclear fuel from nuclear power plants will be deposited in deep geological formations. Some suitable geological formations are the crystalline granitic rocks. The objective of this disposal option is to guarantee that no or only acceptably small amounts of radionuclides reach the biosphere over long time, up to hundreds of million years. In a repository of spent fuel, the metal canister and the engineered buffer (compact bentonite clay and backfill materials) form man-made barriers for the retention of radionuclides. The surrounding crystalline rocks will form the natural barrier since water access to the repository cannot be excluded and integrity of the canister and the buffer cannot be assumed over the long time period required. Studies of flow and transport of radionuclides in deep groundwater are therefore needed.

In subsurface geological formations, groundwater flows mainly in pores and fractures sufficiently large in size to conduct water. The heterogeneities of the structures of the pores and the fractures will strongly affect the patterns of the flow. The transport of solute will also be influenced by many other coupled processes. Some of the processes will enhance the retention of the pollutants thus decreasing the rate of transport and spreading of the pollutants. Other processes will enhance the transport and increase the polluted area. It is therefore of great importance to study the factors and processes that influence the flow patterns and the mechanisms of solute transport in the groundwater.

1.1. Modelling of flow and transport in heterogeneous aquifers and in fractured rock

Solute transport in heterogeneous aquifers is generally a result of complex interactions between advection of the moving groundwater and various physical, chemical and biological mechanisms that act to further immobilize and transform the solutes. The transport is also significantly influenced by the natural heterogeneity of aquifer properties, such as the spatially random distribution of the hydraulic conductivity (e.g., Dagan, 1989; Gelhar, 1993). In typical heterogeneous aquifers, hydraulic conductivity values can vary by orders of magnitude over a few meters (e.g., Freeze, 1975; Hoeksema and Kitanidis, 1984). Additionally, some solutes may be chemically reactive and react among themselves and with the rock minerals. Such reactions may have a significant impact on transport, resulting in either retention or spreading of the solutes. For this reason, much effort is being made to understand and quantify these effects (e.g., Dagan and Neuman, 1997). When dealing with reactive transport at field scale, both the hydraulic and the physico-chemical heterogeneities of the aquifers have to be considered.

The irregular character of observed hydraulic conductivity distributions has lead to the development of a stochastic framework for transport modelling. During the last two decades many stochastic theories have been developed for predicting the fate and transport of solute in heterogeneous aquifers, and are reviewed in several recent textbooks (Dagan, 1989; Gelhar, 1993; Rubin, 2003). In these models the hydraulic conductivity is regarded as a random space function (RSF) with some given statistical distribution. Consequently, the velocity and concentration are also random space function. The spatial statistical moments of solute concentration (e.g., Dagan, 1982, 1984) and temporal moments of solute discharge (e.g., Dagan et al., 1992, Paper I) have also been analyzed in the Lagrangian domain. Mass transfer
processes such as sorption may introduce additional spreading as compared to nonreactive transport, and in recent years the stochastic framework has been extended to reactive transport (Cvetkovic and Dagan, 1994; Paper II).

In fractured rock, the fracture structures observed in the field are extremely heterogeneous. Even though the configuration of a fracture may be essentially planar, the aperture will vary spatially. In some parts, the opening is larger while in some other parts the roughly parallel planes may be in direct contact and the aperture becomes zero. Fractures typically intersect with other fractures to form a fracture network. Moreover the trajectory of an indivisible tracer particle can pass through several different rock formations and can be viewed as a series of sequentially connected fractures, with structural heterogeneities within each of the fractures as well as among the different fractures. Groundwater flow and solute transport in a major fracture are influenced by the background fractures connected, by the networking of fractures and by the heterogeneities of the fracture structures. This view of fractured rock is supported either by direct observations or by indirect inference through borehole drilling or tunnel excavation in several large projects of field experiments, such as the Stripa project (Abellin et al., 1985; 1987) and the Aspö TRUE Project (Winberg et al., 2000; Andersson et al., 2002a).

In addition to the heterogeneities of fracture structures and the variabilities of the fracture configurations, there are a number of retention processes that also influence solute transport in fractures. Retention processes are the processes of mass exchange between mobile phases (groundwater flowing in a fracture) and immobile phases (e.g., the surrounding rock matrix). In the rock matrix surrounding the fractures there are micro pores and fissures. The porosity of the rock matrix acts as a sink for radionuclides and other dissolved species in the mobile water in the fractures (Neretnieks, 1980). Reactive solutes will sorb on the mineral surfaces of the pores and microfissures of the matrix.

The diffusion of the solutes into the rock matrix and sorption therein may considerably retard the transport of the solutes in the fractures. Other processes of physical and chemical interactions of the solutes with the minerals in the rock may also occur, such as sorption on the fracture surface and gouge material, dissolution and precipitation, redox reactions, microbiological reactions. For radionuclides with short half-lives, spontaneous decay plays a prominent role in reducing their concentrations.

Modeling flow and transport in fractured rock is complicated by strong heterogeneity of the fracture structure. Not all fractures play a relevant role on groundwater flow. In fact, most of the flow takes place through a limited number of major fractures or even planes located within fracture belts. The heterogeneity of the fracture structure, especially the random change of the fracture aperture, often causes the groundwater flowing in the fracture to seek the easiest ways inside the fractures and results in discrete preferential flow paths termed channeling or preferential flows (Neretnieks, 1993; 2002). The identification of those relevant features and their connectivity is essential to determine the hydrogeological behavior of the medium (e.g., Winberg et al., 2000; Andersson et al., 2002a). In order to simulate solute transport in fractured media, various models have been developed (e.g., Berkowitz, 2002; Bear et al., 1993; Sahimi, 1995; Paper III).

Flow and transport in a single fracture in low-permeability crystalline rocks are relevant for small scales, say around 5 m, and has been widely studied (e.g., Neretnieks, 1983; Moreno et al., 1988; Tsang and Tsang, 1989; Papers III-V). On larger scales, say > 10 m, the flow and transport occur through a series of fractures or fracture network. Various studies of flow and transport in fracture networks have been performed (e.g., Sudicky and McLaren, 1992; Nordqvist et al., 1992; Moreno and Neretnieks, 1993b; Dershowitz et al., 1998; Outters and Shuttle, 2000; Cvetkovic et al., 2004; Paper VI).

Numerical approaches, especially Monte-Carlo simulations which directly address the
heterogeneities of either a single fracture or a fracture network by simulating particle transport in fractures with, e.g., random aperture distribution, have been explored in the literatures (e.g., Sudicky and McLaren, 1992; Moreno and Neretnieks, 1993b; Tsang and Tsang, 2001; Tsang and Doughty, 2003; Papers III-VI).

1.2. Tracer experiments in deep rock fractures

To facilitate the understanding of the migration and retention properties of the crystalline rocks, injection-pumping tracer tests have been conducted by several nuclear waste management agencies in their underground laboratories (e.g., for SKB, see Winberg et al., 2000; and for Nagra, see Frick et al., 1992; Haderman and Heer, 1996; Heer and Smith, 1998). The SKB (Swedish Nuclear Fuel and Waste Management Company) tracer tests are conducted at the Åspö Hard Rock Laboratory (HRL) in southeastern Sweden. The underground facilities provide an opportunity for research, development and demonstration in a realistic and relatively undisturbed crystalline rock environment at depths comparable to that of a future repository.

Among other field experiments at the Åspö HRL, SKB has initiated a tracer test program referred to as Tracer Retention Understanding Experiments (TRUE) (Bäckblom and Olsson, 1994). The basic idea of the TRUE program is to perform a series of experiments with increasing complexity in terms of the involved retention processes and spatial scale, and to verify the capability of various modelling approaches in predicting radionuclide migration and retention. The TRUE program has progressed in different stages. The first stage (TRUE-1) was focused on a detailed scale (<10 m) in a single feature (e.g., Winberg et al., 2000; Papers VII and IX). The basic objective of TRUE-1 was to perform and analyze transport experiments with non-sorbing and sorbing tracers in a single fracture in crystalline rock. The second stage (TRUE Block Scale) was performed on a block scale (10 to 50 m) with possible multiple geological structures (e.g., Poteri et al., 2002; Papers VIII, and X). The general objective of the Block Scale tracer test was to provide the data from which we could increase the understanding and the capability to predict transport in a fracture network in a block scale (Andersson et al., 2002b; Andersson et al., 2004). To further address the questions of fracture structure complexities that had not been clearly answered in the TRUE Block Scale experiments, the TRUE Block Scale Continuation (BSC) experiments were also conducted. The TRUE BSC project has been conducted in two stages: the BS2A (e.g., Cvetkovic 2003) and the BS2B (Andersson et al., 2005, Paper XI). The BS2A studies are complementary to the BS2B studies. The BS2B studies aimed at performing sorbing tracer tests involving background fractures and subsequent predictions and evaluations.

Various models have been applied in evaluation, interpretation and "blind" prediction of the results of the different tracer tests. To bridge the site characterization (SC) modelling for the field tests and the performance assessment (PA) modelling for a spent fuel repository, Task 6 modelling has been performed. The Task 6 modelling is an international cooperation task established in the framework of the Åspö Task Force on groundwater flow and tracer transport (Papers IX and X).

1.3. Scope of thesis

In this thesis fluid flow and solute transport in heterogeneous aquifers and particularly in fractured rock have been investigated within a Lagrangian Stochastic Advection-Reaction (LaSAR) framework. The heterogeneities of the aquifer structure or fracture configuration, as well as the various reaction/retention processes have been considered in the modelling approach.

The LaSAR framework has first been developed and applied to fluid flow and solute transport in generic heterogeneous aquifers and fractured rocks (Papers I - VI). Decoupling of the flow and transport analyses in this approach makes detailed study of the statistical properties of some important model
parameters possible by Monte-Carlo numerical simulations of the flow fields. The solute travel time $\tau$ and the parameter $\beta$, which are purely determined by the flow conditions but quantify the hydrodynamic influences on the retention processes, are two such parameters. The influences of the structural heterogeneities of the aquifer and the fracture on the solute transport have been studied by using different flow models, different fracture configurations and fracture heterogeneities, as well as different input and boundary conditions. The influence of temperature increase on solute transport has been considered in Paper V.

The LaSAR approach has been applied to evaluation, interpretation and prediction of the results of a number of tracer tests (TRUE-1, TRUE Block Scale and TRUE Block Scale Continuation) conducted by SKB at the Åspö site (Papers VII - XI). Through inverse modelling by deconvolution of the breakthrough curves (BTCs) of conservative tracers measured in the field experiments and by calibrating on the BTCs of non-conservative tracers, in-situ values of the reaction/retention parameters are obtained and compared with those of laboratory or field measurements for the same parameters. Moreover, the LaSAR approach has also been extended from site characterization (SC) modelling to performance assessment (PA) modelling (Papers IX-X).

2. THE LASAR APPROACH TO SOLUTE TRANSPORT

In the Lagrangian Stochastic Advection-Reaction (LaSAR) framework of fluid flow and solute transport, the advective transport equations are coupled with related reactive equations and solved in the Lagrangian domain. Both flow parameters and parameters of chemical reactions and other retention processes like matrix diffusion may be assumed to be random space functions (RSFs) and treated with stochastic methods. The effects of hydrodynamic dispersion on solute transport are accounted for consistently by the statistical properties of the involved RSFs. The Lagrangian approach focuses on the displacements and travel times of the solutes, using the displacements and/or travel times along streamlines (trajectories) as fundamental variables to quantify the flow and transport. The plume of solute concentration and the expected solute discharge can be usually obtained from the joint distribution of the flow and reactive parameters and are characterized by the various statistical moments of the parameters. The statistical properties of the various parameters and their correlations can be studied by numerical analysis of Monte-Carlo simulations. The Lagrangian approaches are especially applicable to advection-dominated transport where the pore scale dispersion can be neglected. In this chapter, the LaSAR approach related to this thesis will be presented. For a complete presentation of the underlying theories of the Lagrangian approach for solute transport, readers are referred to Cvetkovic and Dagan (1994, 1996), Dagan and Cvetkovic, (1996), Paper II, and Paper III.

2.1. The Lagrangian description of advection

In the Lagrangian approach fluid flow and solute transport are described in a coordinate system that moves along a streamline in the flow field, i.e., the trajectory of a particle. An originally three-dimensional problem can thus be reduced to a one-dimensional transport problem of the mean flow direction (Cvetkovic and Dagan, 1994). Furthermore, when the effects of molecular diffusion and pore-scale (local) dispersion are neglected (for the effect of pore-scale dispersion, see e.g., Fiori, 1996), as is conventionally practiced in the study of field-scale transport problems (e.g., Simmons, 1995), there will be no interaction between adjacent trajectories. The associated mass transfer and retention processes take place along individual trajectories/streamlines. Field-scale hydrodynamic dispersion arises as a consequence of the velocity variation between streamlines due to macroscopic heterogeneity of the transport and retention properties of the flow system.

For the groundwater flow and solute transport in a heterogeneous aquifer, the hydra-
The hydraulic conductivity $K$ is regarded as a random space function (RSF). Alternatively if the aquifer is essentially horizontal (planar), the transmissivity $T$ will be used (as in Paper I). The transmissivity will be the hydraulic conductivity times the thickness of the aquifer. The heterogeneities of rock fractures can be characterized either by their aperture variability or by the local variation of the transmissivity. The transmissivity is often related to the fracture aperture by some empirical power laws, see Eq. (4.4) and the accompanying discussions later in this thesis.

The hydraulic conductivity, $K(x)$, or the transmissivity, $T(x)$, is a spatial function, with $x(x_1, x_2, x_3)$ being a Cartesian coordinate vector. The groundwater velocity, $V(x)$, will also be a RSF and satisfy the continuity equation $\nabla \cdot (\nabla T) = 0$ where the porosity $\theta$ is assumed to be a constant. In this thesis the flow is assumed to be laminar and in steady state. The groundwater is assumed to be a Newtonian fluid with constant density. The flow velocity $V(x)$, is then related to $K$, and the hydraulic head $h$ through Darcy’s law $V = -\frac{K}{\theta} \nabla h$.

In the following discussions, the mean flow direction is assumed to be in the direction of the $x$-coordinate. In the Lagrangian approach, the solute mass is viewed as a set of individual parcels or particles. Each particle is transported along its own trajectory, $X$. The trajectory is characterized by $x = X(t, a)$, where $x$ is the Eulerian displacement vector of the particle, $t$ is time and $a = X(0, a)$ is the injection place of the particle at a time $t = 0$. $X$ can be obtained by solving $dX(t, a)/dt = V[X(t, a)]$ (Taylor, 1921; Dagan, 1984). For steady-state flow and conservative solutes, the particle trajectories coincide with the streamlines of the velocity field, and the movement of the particles by the velocity field (advection) is the only transport mechanism for conservative solutes. For non-conservative (reactive) tracers (e.g., radioactive tracers) the transport will be influenced by retention processes that will be discussed later.

A key quantity related to the advective trajectory is the travel time $\tau$ of a solute particle obtained by solving $x = X(t, a)$ to yield $t = \tau(x, a)$. As $X(t, a)$ is the trajectory of the particle originating from $a$ and is assumed to be a monotonously increasing (unique) function of $t$, $\tau$ is then the travel time of the particle from the initial place $x = a$ to the place $x = x$ (see Figure 1 in Paper II). The advective travel time $\tau(x, a)$ can also be obtained through an integration along the trajectory (Cvetkovic et al., 1992):

$$\tau(x, a) = \int_{t}^{\tau} \frac{d\xi}{V[\xi, \eta(\xi), \zeta(\xi)]} = \int_{x}^{x} \frac{d\xi}{w(\xi)} \quad \text{(2.1)}$$

Where $\eta(x, a) = X_{\eta}(x, a)$ and $\zeta(x, a) = X_{\zeta}(x, a)$ are the transverse displacements of a particle and $V[\xi, \eta(x), \zeta(x)] = w(x)$ (Cvetkovic and Dagan, 1994). $w(x)$ is the Lagrangian velocity along the trajectory.

The travel time $\tau(x, a)$ is also a RSF. For conservative solutes, statistical properties of $\tau$ alone will characterize the spreading of the solute concentration and the distribution of the solute discharge.

### 2.2. The LaSAR approach of solute transport

In the Lagrangian Stochastic Advection-Reaction (LaSAR) approach, the advective transport and the solute retention are modelled by mass balance equations in the Lagrangian domain. The mass balance equations for the mobile concentration $C$ and for the immobile concentration $N$ are written as (Cvetkovic and Dagan, 1994; 1996):

$$\frac{\partial C}{\partial t} + \frac{\partial C}{\partial \tau} = \Psi_1 \quad \text{(2.2)}$$

$$\frac{\partial N}{\partial t} = \Psi_2 \quad \text{(2.3)}$$

where $C$ is the solute concentration in groundwater (mass of solute per unit volume of moving fluid), $N$ is the solute concentration sorbed or transferred to the immobile phase (mass of immobilized solute per unit volume of fluid), $t$ is the time, $\tau$ is the travel time defined in (2.1), $\Psi_1$ and $\Psi_2$ are source
Hydrodynamic control of retention in heterogeneous aquifers and fractured rock

terms for $C$ and $N$ respectively. The source terms are related to various retention processes that will be discussed in the next section. Note that the independent variables in (2.2) and (2.3) are only $t$ and $\tau$, and the problem has been reduced to be one-dimensional (in addition to the dependence on time). The $\tau$ is the advective travel time of a nonreactive solute particle along a three-dimensional streamline or trajectory. Therefore $\tau(x, a)$ characterizes the geometry of the trajectory along which the transport of reactive solutes can also take place. By tracking a particle along a trajectory and using the travel time $\tau$ as an independent variable, the original 3D system in the Eulerian coordinates becomes essentially 1D system in the Lagrangian coordinates.

For linear reactions represented by the source terms $\psi_1$ in (2.2) and $\psi_2$ in (2.3) the reactive solute transport along a trajectory can be characterized by a function $\gamma(t, \tau; \Omega)$ (Cvetkovic and Dagan, 1994). For a Dirac pulse input, the expected solute discharge $Q$ through the control plane at $x$ can thus be expressed as (Dagan and Cvetkovic, 1996; Papers II and III):

$$\langle Q(t,x) \rangle = M \int_{0}^{\tau} \int_{\Omega} \gamma(t, \tau; \Omega) g(t, \tau; \Omega) d\Omega \, d\tau$$  \hspace{1cm} (2.4)

where $g$ is probability density function (pdf) of the joint distribution of the travel time $\tau$ and the random parameter $\Omega$, which characterizes various reactive and retention processes described in the source terms. The function $\gamma$ is the solution to (2.2) and (2.3) for a Dirac pulse injection at $t = 0$ into a system initially free of the solute. For an arbitrary injection, the convolution of the input function and $\gamma$ should be used in (2.4) instead of $\gamma$ (see also Eq. (2.10) in the next subsection). The function $\gamma$ can have different forms depending on the nature of the source terms $\psi_1$ and $\psi_2$. The $\Omega$ may represent different reactive or retention processes. In Paper II, $\Omega$ represents reaction flow path $\mu$ for heterogeneous aquifers, while it represents parameter $\beta$ for a single fracture in Paper III. See also discussions in the following sections.

2.2.1. Retention processes

In (2.2) and (2.3), $\psi_1$ and $\psi_2$ are the source terms related to various reactive and retention processes representing the mass exchange between the mobile and immobile zones of the system. The immobile zone in fractured media may be the rock matrix in which the solute can diffuse and/or sorb. Alternatively it can be the solid part in heterogeneous aquifers where the solute may sorb on. Figure 2-1 illustrates some retention processes in an ideal single fracture. The retention processes can be generally classified into two categories: physical processes and chemical processes. For further discus-

Figure 2-1 Retention processes in a fracture
Sorption processes in a heterogeneous aquifer (the reaction flow path)

The sorption processes (here the term sorption is used in a broader sense to describe a variety of chemical and physical processes such as diffusion) can be either equilibrium or nonequilibrium. When the rate at which the mass is transferred between the mobile and immobile zones is faster relative to the rate at which the mass is advected in the system, the process is considered to be an equilibrium process. Otherwise the process will be considered nonequilibrium. The equilibrium processes are thus the extreme cases of the nonequilibrium process when the reaction rates become instantaneous. For linear nonequilibrium sorption of first order, the source term for the immobile concentration $N$ is given by $\gamma_2 = k_1 C - k_2 N$, where $k_1$ and $k_2$ are forward and reverse rate coefficients. The source term for $C$ is given by $\gamma_1 = -\frac{\partial N}{\partial t}$. When the values of $k_1$ and $k_2$ become sufficiently large (while ratio $k_1 / k_2$ still finite), the sorption process becomes an equilibrium process with a distribution coefficient $K_d = k_1 / k_2$.

Sorption parameters (e.g., $K_d$) generally vary in space as the mineralogy and the solid phase composition varies (e.g., Tompson et al., 1996). The incorporation of chemical heterogeneity into transport has been the subject of many studies (e.g., Huang and Hu, 2001; Allen-King et al., 1998; Paper III and its references). Knowledge about the actual spatial variability of reactive and retention parameters, e.g., $K_d$, as well as the correlation between them and the flow properties, such as the hydraulic conductivity $K$, is limited. A weak but significant negative correlation between $K$ and $K_d$ for strontium has been observed at the Borden site in Canada (Robin et al., 1991).

In the following, an example of the $\gamma$ expression for a special case of two-site sorption model is given. Here $K_d$ is considered to be a RSF, and $k = k_2$ denotes the backward rate coefficient. Assuming $k=\text{const.}$, then $\gamma$ in (2.4) will be (Paper II):

$$\gamma(t, \tau; \mu) = \exp(-k\mu)\delta(t - \tau) + k^2 \mu \exp\left(-k\mu + (t - \tau)\right)\tilde{I}_1(k^2 \mu(t - \tau))$$

(2.5)

where $\tilde{I}_1(Z) \equiv I_1(2\sqrt{Z})/\sqrt{Z}$, $I_1$ is the modified Bessel function of the first kind of order one, and $\mu$ is the so-called reaction flow path defined as (Paper II)

$$\mu(t; \tau) = \int_0^t K_d[X(\tau; \mu)] d\tau$$

(2.6)

$$\mu(x; \tau) = \int_0^x K_d[x, \eta(x, \tau), \zeta(x, \tau)] dx$$

(2.7)

In this example, $\mu$ in (2.6) stands specifically for the generic parameter $\Omega$ in (2.4). The reaction flow path $\mu$ has a dimension of time. The physical meaning of $\mu$ is the time retarded by the sorption process.

Matrix diffusion and sorption on fracture surface

For diffusion into the rock matrix and sorption on the fracture surface, the source term/sink terms for the mobile concentration $C$ in the open fracture are defined as (Selroos and Cvetkovic, 1996; Paper III):

$$\psi_1^{(M)} = \frac{D \theta}{b(t)} \frac{\partial N^{(M)}}{\partial z} - \frac{K_a \partial C}{b(t)} - \lambda C$$

where $z$ is the coordinate orthogonal to the fracture plane, $K_a$ is the partition/distribution coefficient for sorption on the fracture surface. The sorption is assumed to be at equilibrium. In $\psi_1^{(M)}$, 1-D diffusion into the rock matrix in the direction $z$ is assumed, $\theta$ is the matrix porosity and $D$ is the diffusivity in the rock matrix. Both of them are assumed to be spatially uniform effective values. $b(t)$ is the Lagrangian half-
Hydrodynamic control of retention in heterogeneous aquifers and fractured rock aperture, obtained through \( b(\tau) = b[X(\tau)] \), where \( X(t) \) is the advection trajectory (e.g., Dagan 1984); \( \lambda \) is the decay constant.

The source/sink term for the immobile concentration \( N \) in the matrix is defined as

\[
\psi_2^{(M)} = -K_d^m \frac{\partial N^{(M)}}{\partial t} + D \frac{\partial^2 N^{(M)}}{\partial x^2} - \lambda N^{(M)}
\]

where equilibrium sorption is assumed in the matrix with \( K_d^m \) being the distribution coefficient.

The source/sink term for the immobile concentration \( N \) in the matrix is defined as

\[
\frac{\partial N}{\partial t} = -\frac{H(t - \tau)K_d^m}{2\pi(t - \tau - \beta K_d^m)^{1/2}} \exp\left[\frac{-(\beta K_d^m)^2}{4(t - \tau - \beta K_d^m)} - \mu\right]
\]

where \( \kappa \equiv \theta \sqrt{DR_m} \) with \( R_m = 1 + \frac{K_d^m \rho}{\theta} \), and \( \beta \) is defined as:

\[
\beta = \int_0^\tau \frac{d\tau'}{b(\tau')} = \int_0^\mu \frac{dx}{V_1(x)b(x)}
\]

where \( L \) is the distance between the injection and the control plane. \( V_1(x) \) is Lagrangian quantity along a trajectory, e.g., \( V_1(x) = V_1[X(\tau(x))] \). It can be observed that the diffusion and the sorption in the matrix have been expressed in (2.8) by a parameter group \( \kappa \). The retention model (2.8) was first proposed by Neretnieks (1980).

In this case, \( \beta \) in (2.9) stands specifically for the generic parameter \( \Omega \) in (2.4).

### 2.2.2. Coupled mass transfer processes

We have obtained solutions of (2.2) and (2.3) for transport coupled with one of the mass transfer processes at a time in the above section. When more than one process exist concurrently, Equations (2.2) and (2.3) are coupled through the source/sink terms of the various mass transfer processes. The solution \( \gamma \) for the coupled processes can be obtained by calculating the convolution (with respect to \( \tau \)) of the solutions of each individual process.

For a continuous input \( \phi(t) \), the solution of (2.2) and (2.3) is the convolution of the input \( \phi(t) \) with the solution for a Dirac pulse input \( \gamma \):

\[
Q(t) = \phi(t) \ast \gamma(t) = \int_0^t \int_0^\tau \gamma(t', \tau; \Omega) g(t, \Omega) d\tau d\Omega
\]

where \( \Omega \) is a RSF representing the reaction/retention processes and can be \( \beta \) or \( \mu \) in special cases as have been discussed.

#### 2.2.3. Two important parameters \( \tau \) and \( \beta \)

The most generic solution for the transport equations (2.2) and (2.3) is given by (2.10). In addition to the input function and the probability density function (pdf) of the joint distribution between \( \tau \) and \( \Omega \), the solute discharge is determined by \( \gamma \) which is a function of the travel time \( \tau \) and the random spatial variable \( \Omega \) that characterizes the various reaction/retention processes. When the parameters involved in the reaction/retention processes are assumed to be constant over the entire flow region, i.e., they are assumed to be effective values, the parameter \( \beta \) (2.9) corresponds to \( \Omega \). Here \( \beta \) is also a quantity that depends only on the flow. Under this condition the spreading of the solute concentration and the distribution of the solute discharge will depend on the statistical properties of \( \tau \) and \( \beta \), and the correlation between them. For this reason, the parameters \( \tau \) and \( \beta \) are of great importance in the LaSAR approach for transport in fractured rock. We will investigate their statistical properties and correlation in a variety of flow systems.

Two lumped parameters in (2.8) defined as \( A = \beta K_d \) and \( B = \beta K_d \) are the critical parameters that characterize the solute retention due to sorption on the fracture surface and diffusion into the rock matrix. These two parameters relate the dynamics of flow to the mass transfer processes that control solute retention (Paper III). By definition

\[
\kappa = \theta \sqrt{D(1 + \frac{K_d \rho}{\theta})}
\]
is a material parameter group which describes the diffusion and sorption in rock matrix. The effect of aperture variation on matrix diffusion/sorption is described by the product $\beta \kappa$ and the effect of aperture variation on surface sorption is described by $\beta K_a$ in (2.8).

For flow in a rectangular planar fracture with a constant aperture, i.e., $b = \text{const}$, (2.9) can have a analytical form related to $\tau$ as

$$\beta = \frac{\tau}{b} = k \tau$$  \hspace{1cm} (2.12)

By using the flow rate $Q = 2VbW$, $\beta$ can also be related to $Q$ as

$$\beta = \frac{2LW}{Q}$$  \hspace{1cm} (2.13)

where $2LW$ is the area of the fracture surface that is in contact with the flowing water, $L$ is the length of the fracture, and $W$ is the width of the fracture.

Eq. (2.12) suggests a linear relationship between $\beta$ and $\tau$ for this case. Eq. (2.13) shows that $\beta$ can be expressed as the ratio of the flow-contacted area to the volumetric flow rate for the ideal case. This case is often considered in analytical models of diffusive mass transfer in fractures, where $2LW$ is referred to as the flow-wetted surface (e.g., Moreno and Neretnieks, 1993a), and $k$ is referred as "specific surface area" (Wels et al., 1996) and flow-wetted surface per unit volume of water (Andersson et al., 1998).

Eq. (2.12) simplifies the computation significantly, since the entire distribution of $\beta$ is replaced by the distribution of $\tau$ and a parameter $k$.

### 2.3. The Monte-Carlo simulation approach

Although $\gamma$ (Eq. (2.5) and Eq. (2.8)) is available in analytical form (or in closed-form) for linear retention processes, the Lagrangian random variables, $\tau$ and $\beta$ (or $\mu$, or $\Omega$) still depend on the random flow fields. To use $\gamma$ to calculate the solute discharge, the statistical properties of $\tau$ and $\beta$ are needed. On the other hand, their statistical properties usually cannot be determined by laboratory or field experiments. The most common approach is to solve the stochastic flow equations numerically by Monte Carlo simulations. With this approach, a large number of equally probable random realizations of the hydraulic properties are generated using geostatistical techniques such as Gaussian sequential simulation. The flow equations can be solved numerically by a conventional deterministic numerical flow simulator for each realization to obtain random flow fields. The particle tracking simulation can then be performed on each realization of the flow field. The statistical moments of random parameters $\tau$ and $\beta$ (or $\mu$) can be obtained by averaging the results over all realizations. This approach is conceptually straightforward, but it requires intensive computational efforts since the number of realizations needed to adequately describe the flow system is relatively high. Moreover, the computational task for each realization is also large in order to resolve the high space-time fluctuations in the random parameters using fine numerical discretizations.

In the papers included in this thesis, Monte-Carlo simulations have been conducted under various hydraulic conditions in heterogeneous aquifers and in fractures. The Monte-Carlo simulations in this thesis are performed essentially in three procedures: (1) to generate the random transmissivity (or hydraulic conductivity) fields; (2) to solve the flow equations to obtain the velocity fields; and (3) to perform particle tracking to obtain the various moments of $\tau$, $\beta$ (or $\mu$, or $Q$) and other parameters.

The random transmissivity fields are generated as unconditional random fields using the random field generator HYDROGEN (Bellin and Rubin, 1996). The number of the random fields generated, i.e., the number of realizations used, depends on the hydraulic conditions and the degree of heterogeneity, typically ranging from a few hundreds to a few thousands. A lognormal distribution was usually assumed for the transmissivity $T$, or the hydraulic conductivity $K$. An exponential correlation structure is also assumed, as will be discussed later in this thesis.
The numerical simulations for solving the flow fields are conducted using the either finite element algorithms (Mose et al., 1994; Papers I and II) or finite difference codes like MODFLOW 2000 (Harbaugh et al., 2000, Papers III-XI). The velocity fields are then obtained by Darcy’s law.

3. HYDRODYNAMIC CONTROL OF RETENTION IN HETEROGENEOUS AQUIFERS

In the previous sections, the basic concepts and theories related to the work of this thesis have been presented. It has been shown that the statistical properties of the travel time $\tau$ is a key parameter to quantify the advective flow and transport of conservative solutes. For non-conservative solutes, another important parameter is $\Omega \equiv \mu$, together with the reaction/retention coefficients, will quantify the effects of the various reaction/retention processes for the transport of the non-conservative solutes. In the following, we will present some of the results obtained using the LaSAR approach to different types of flow systems. In this chapter we consider the transport in heterogeneous aquifers. In the next chapter, the results obtained for fracture media (rocks) will be presented.

3.1. Preferential flow (Paper I)

Two main approaches have been widely used in analyzing the statistical properties of the travel time $\tau$. The first approach is based on the analytical method of first-order approximation and the second approach is based on Monte-Carlo simulations. In the first-order approximation it is assumed that the streamlines are essentially parallel and the transverse displacement is negligibly small. This thesis focuses on establishing a Monte-Carlo simulation scheme.

In Paper I, the Monte-Carlo numerical simulations have been used to study the flow and transport of conservative solutes in a two-dimensional aquifer with a spatially varying transmissivity field and to evaluate and verify the first-order analytical results for the statistical properties of travel time and transverse displacement. This approach of Monte-Carlo simulations has also been widely used in the literatures (e.g., Bellin et al., 1992; Chin and Wang, 1992; Selroos and Cvetkovic, 1994; Hassan et al., 1998).

For nonreactive particles, the mean and variance of the solute discharge can be evaluated by the travel time moments (e.g., Shapiro and Cvetkovic, 1988; Cvetkovic et al., 1992; Selroos and Cvetkovic, 1994). Numerical simulations have been used in several studies of nonreactive advective transport to test the applicability of first-order analysis (e.g., Bellin et al., 1992).

3.1.1. First-order results

In Paper I the travel time $\tau(x)$ (2.1) and the transverse displacement $\eta(x)$ are the two Lagrangian (random) variables used to quantify the solute advection along a trajectory. The advective solute discharge is proportional to the joint distribution of $\tau$ and $\eta$.

The main focus in Paper I is the first two moments of the travel time. The analytical results for the first two moments of $\tau$ and $\eta$ have been obtained from the first-order theory. The first moment (the mean) of $\tau$ is obtained as $\tau_A = x\theta / T_G J$ where $\theta$ is the porosity, $T_G$ is the geometric mean of transmissivity and $J$ is the hydraulic gradient. The second moment (variance) of $\tau$ and the transverse displacement $\eta$ have been obtained in Eq.(7) and Eq.(8) in Paper I.

3.1.2. Monte-Carlo simulations

Monte-Carlo simulations have been performed in a rectangular two-dimensional domain (Figure 1 in Paper I). The flow is driven by a hydraulic gradient between left and right boundaries, while no-flow boundary condition is assumed at the upper and lower boundaries. The flow field is solved by mixed hybrid finite element scheme (Mose, et al., 1994). The particle tracking is performed on a smaller inner domain of the flow field to minimize the boundary effects (e.g., Rubin and Dagan, 1989). The various statistical properties are computed along particle trajectories. The statistics of $\tau$ and $\eta$ are evaluated as a function of the normalized
distance $x/I_Y$. The simulation results are then compared with the first-order expressions.

The simulations are performed for variances of the transmissivity distribution $\sigma_Y^2$, ranging from 0.25 to 4.0. The number of realizations is between 500 and 1000 depending on the magnitude of $\sigma_Y^2$. A typical realization for $\sigma_Y^2=4.0$ is shown in Figure 3-1, where preferential flow paths are observed.

3.1.3. Simulation results

The statistical properties of the Lagrangian velocity $w$ have been analyzed in Paper I. Cumulative distribution of $\ln w$ is approximately normal over $x/I_Y$ (Figure 3a, Paper I). At the initial point of the particle trajectory, the Lagrangian velocity $w$ is the same as the Euclidian velocity $u$, the statistics of them are also the same. As the particle travels downstream, the statistics of $w$ and $u$ deviate considerably (Figures 3b and 3c in Paper I). This implies that the statistics of $w$ are nonstationary (Figures 4a and 4b in Paper I). The nonstationarity is more profound for larger $\sigma_Y^2$. The mean travel time $\tau$ is a nonlinear function of distance for shorter distance and is a linear function of distance for longer distance (Figure 7a in Paper I). Note that the results in Paper I were obtained for residence injection. Later work by Demmy et al., (1999) for flux injection found that $\tau$ is a linear function of distance, and variance of $\tau$ is a nonlinear function of distance.

The cumulative distribution of $\ln \tau$ is closely approximated by the normal distribution (Figure 3-2) for all simulated $\sigma_Y^2$ values. The Monte-Carlo simulation approach proposed in Paper I could be simply applied in investigating flow and transport in three-dimensional formations, possibly with more complex structure features under various boundary conditions (e.g., Paper X). The travel time statistics discussed for advective transport in this paper can be directly used for quantifying solute discharge of transport with reactions and other retention processes (e.g., Papers II-XI).

3.2. Coupled hydraulic and retention heterogeneity (Paper II)

Many contaminants (e.g., radionuclides) are not conservative. When these contaminants are transported through the aquifers, they move with the bulk water due to advection, are dispersed due to the velocity variation...
between flow trajectories, and are retarded due to different reaction/retention processes. The retention processes considered in the transport models in heterogeneous aquifers are often referred to as sorption processes. The sorption processes here may include a wide range of physical and chemical interactions, e.g., sorption into solid matrix, or diffusion into stagnant water. In the present analysis, however, we are not concerned with specific mechanisms of the retention processes, but will rather explore the consequences using the generic representation of the processes.

Like the flow parameters, the parameters that control the retention processes for reactive solutes can also be spatially variable. Several studies have been performed for evaluating reactive transport of linear and nonlinear sorption reactions using the Lagrangian framework (e.g., Cvetkovic and Dagan, 1994; 1996; Dagan and Cvetkovic 1996). This Lagrangian framework has later been extended to account for the spatial variability of the reaction parameters as well as for the variability of the flow parameters such as hydraulic conductivity (Paper II). In Paper II the concept of reaction flow path ($\mu$) has been proposed. This thesis focuses on the investigation of the statistical properties for $\mu$ and $\tau$ by using both the first-order analytical approach and Monte-Carlo simulation approach.

### 3.2.1. First-order solution of reaction flow path

The generic reaction parameter $P$ is assumed to be a RSF. Its variation consists of two parts: one is that caused by the variation of the flow parameter like the hydraulic conductivity, and the other part is the variation of the reaction parameter itself. Here the variation of $P$ is assumed to be related to variation of the flow parameter because it is plausible that there may exist a relation between the flow parameter and the reaction parameter. The random variable $P$ can then be expressed as:

\[
P(x) = P_G e^{\alpha X(x) + W(x)}
\]

(3.1)

where $P_G$ denotes the geometric mean of $P$, and $Y$ represents the variation of the flow parameter (e.g., hydraulic conductivity) and is normally distributed as $N(0, \sigma_Y^2)$ with $C_Y(x, x') = \sigma_Y^2 \exp[-(x - x') / I_Y] = \sigma_Y^2 \exp(-r / I_Y)$ and $I_Y$ being the integral scale of $Y(x)$. $W$ represents the variation of the reaction parameter itself and is a normally distributed
space function with \( N(0, \sigma_w^2) \), and the covariance function \( C_w(r) = \sigma_w^2 \exp(-r/I_w) \) and \( I_w \) being the integral scale of \( W(x) \). \( \alpha \) is a constant which determines the correlation between \( P \) and \( K \). \( Y(x) \) and \( W(x) \) are assumed to be statistically independent, i.e., \( \sigma_{yw}=0 \).

A first-order analytical result for \( \langle \mu \rangle \) is given as (Paper II):

\[
\langle \mu(x) \rangle = \langle P \rangle / \langle \tau \rangle \tag{3.2}
\]

The closed-form expressions with the first-order approximation for \( \sigma^2_\mu \) and \( \sigma_{\mu x}^2 \) for a two-dimensional statistically isotropic aquifer are derived as (Paper II):

\[
\sigma^2_\mu = \frac{I_y^2 \sigma_Y^2}{U^2} P^2_G e^{2\xi} \left\{ 2\chi - 3 \ln \chi + \frac{3}{2} - 3E + 3 \left[ Ei(-\chi) + \frac{(1+\chi)e^{-\chi}-1}{\chi^2} \right] \right\}
+ 2(e^{-\chi} - \chi - 1)(-2\beta + \beta^2 + \frac{\sigma^2_w}{\sigma^2_Y}) - 4\beta \left[ e^{-\chi} - \chi - Ei(-\chi) + 1 - E \right] \right\}
\]

\[
\sigma_{\mu x}^2 = \frac{I_y^2 \sigma_Y^2}{U^2} P^2_G e^{\xi} \left\{ 2\chi - 3 \ln \chi + \frac{3}{2} - 3E + 3 \left[ Ei(-\chi) + \frac{(1+\chi)e^{-\chi}-1}{\chi^2} \right] \right\}
- 2\beta \left[ e^{-\chi} - \chi - Ei(-\chi) - 1 \right] \right\} \tag{3.3}
\]

where \( \chi = x/I_y \) and \( E=0.577... \) is the Euler constant. For a three-dimensional statistically isotropic formation, closed-form expressions are also obtained in Paper II (Eq. 6.13). In (3.2)-(3.3) we have assumed for simplicity that \( I_w = I_y \).

### 3.2.2 Simulation results

The Monte-Carlo simulations are performed in a two-dimensional heterogeneous aquifer with a rectangular simulation domain similar to that in Paper I. The statistical properties of \( K(x) \) and \( P(x) \) are assumed in consistence with those in the analytical models: normal distribution with a negative exponential covariance function for \( \ln K \) and \( \ln P \). A constant head is assumed at the left and right boundaries and no-flow condition is assumed at the top and bottom boundaries of the simulation domain.

The procedures of the Monte-Carlo simulations in Paper II are: (1) to generate the random \( K \) field, (2) to solve the flow equations to obtain the heterogeneous velocity field, (3) to calculate the solute transport by monitoring the time, transverse location, \( x \)-velocity of each solute particle, and the \( K \) at different cross-sections along the trajectory, (4) to generate the random \( P \) field, (5) to sample particular \( P \) values along the same trajectory in (3), and (6) to calculate the statistics of \( \mu \).

The analytical first-order results (3.2) and (3.3) are compared with the Monte-Carlo simulation results for \( \alpha=0 \) and \( \alpha=-0.3 \) in Figure 2 in Paper II. With each value of \( \alpha \), the standard deviations of \( Y \) and \( W \) are selected as: \( \sigma_Y=1.0 \) and \( \sigma_W=0.5 \). The simulated \( \langle \mu \rangle \) is a nonlinear function of distance up to \( x/I_y = 4 \) (Figure 2a in Paper II) since \( \tau \) is also a nonlinear function of distance (Figure 7a in Paper I). For larger distance the simulated \( \langle \mu \rangle \) becomes a linear function of distance (Figure 2a in Paper II) as similar behavior has been observed for \( \tau \) (Figure 7a in Paper I). The first-order results are strictly valid only when \( \sigma_Y^2 \) is small due to the approximations made in the first-order analysis. Therefore for small values of \( \sigma_Y^2 \), the nonlinearity with distance for the simulated \( \langle \tau \rangle \) and \( \langle \mu \rangle \) values is diminishing, and the simulated results are close to the first-order analytical results. The simulated \( \sigma^2_\mu \) and the first-order analytical solution of Eq. (B3) in Paper II are compared in Figure 2b in Paper II. For small values of \( \sigma_Y \) and \( \alpha \) (i.e., \( K \) and
are essentially independent), the simulated results are close to the first-order results. As the \( \sigma_y \) increases, or the \( \alpha \) is equal to -0.3, the deviation becomes larger. The correlation coefficient between \( \mu \) and \( \tau \) computed from (3.3) using Eq. (D3) in Paper II is compared with the simulation results in Figure 2c in Paper II for the case of \( \sigma_y = 1.0 \) and \( \sigma_y = 0.5 \). The first-order solution closely matches the simulation results for \( \alpha = 0 \) as the distance increases. The deviation is apparent for \( \alpha = -0.3 \).

3.2.3. Solute discharge

A useful representation of transport in aquifers is by the solute discharge. The solute discharge defined as the flux integrated over a control plane was considered as a prime quantity of interest in a number of studies (e.g., Kreft and Zuber 1978; Cvetkovic and Shapiro 1990; Cvetkovic et al., 1992; Dagan et al., 1992; Destouni and Graham 1995). Current regulatory standards for the subsurface environment, especially those set in terms of travel time, make the solute discharge approach appealing for predicting subsurface contaminant transport. Physically the discharge can be perceived as a direct measurement of the rate of contaminant displacement.

For reactive solute, the transport velocity of the solute particles is not the same as the fluid velocity. The transport of the solute is usually retarded and delayed by the chemical reactions or other retention processes. The mass balance equations are (2.2) and (2.3) with the source terms of \( \psi_1^{(a)} \) and \( \psi_2^{(a)} \) discussed in previous section. The solution of the equations for a Dirac-pulse injection is given in (2.5) where \( \mu \) is a spatial random variable representing the chemical heterogeneity. The solute discharge is computed by (2.4) with \( \Omega \) standing for \( \mu \) with the notation of this section.

The two-site sorption model (2.5) is used for illustration. With (2.4) and (2.5), the expected discharge for nonequilibrium reversible sorption is obtained as:

\[
\langle Q(t,x) \rangle = \int_0^\infty e^{-k\mu} g(t,\mu;x)d\mu + \int_0^\infty k^2 \mu e^{-k(\mu+\tau)} \int_0^\infty \left[ k^2 \mu(t-\tau) \right] g(\tau,\mu;x)d\mu d\tau
\]

(3.4)

The solution \( \langle Q(t,x) \rangle \) in (3.4) will be determined by the five moments \( \langle \tau(x) \rangle \), \( \sigma_\tau^2 \), \( \langle \mu(x) \rangle \), \( \sigma_\mu^2(x) \), and \( \rho_\mu(\tau) \) derived from the joint lognormal distribution \( g(t,\mu;x) \) of \( \mu \) and \( \tau \) at a distance of \( x \). The five moments can be calculated using the first-order approximation, or obtained from the numerical simulations. In Figure 3-3, the mean discharges thus calculated are plotted against \( tU/I_y \). The results calculated from the first-order moments (solid lines) are compared with those obtained from the moments calculated by numerical simulations (the dashed lines for \( \sigma_y = 1.0 \), \( \sigma_y = 0.5 \) and \( \alpha = 0 \) and -0.3, the backward rate constant \( k \) is set to be 0.1. Deviations from the first-order results are observed in the initial parts as well as the parts around the peaks of the curves of the numerical simulations for both \( \alpha = 0 \) and \( \alpha = -0.3 \). However in the tail parts the deviations diminish. Larger discrepancies can also be expected for cases when \( \sigma_y \) or \( \sigma_y^2 \) is larger. The reason is that when \( \sigma_y^2 \) is larger, the first-order results of \( \sigma_\mu^2 \) are quite different from the numerical simulation results of \( \sigma_\mu^2 \). When \( \sigma_y \) is larger the first temporal moments of the simulation results also deviate from those of the first-order results.
4. HYDRODYNAMIC CONTROL OF RETENTION IN FRACUTRED ROCK

In the previous chapter we have presented modelling results of fluid flow and solute transport in heterogeneous aquifers using the LaSAR approach. Another important type of aquifer is fractured rock. As many countries including Sweden intend to build repositories for final disposal of the spent nuclear fuel in deep geological formations, understanding the flow and transport behavior of dissolved radionuclides in rock fractures becomes an important issue. While fractures in rocks are typically present as interconnected networks, the transport in a single fracture needs to be first understood before more complicated field-scale fracture networks can be addressed. In this thesis, we therefore first deal with the transport and retention in a single fracture and later on extend the work to address the transport in 3-dimensional fracture series, considering both internal and global heterogeneities of the fracture structure.

In numerical simulations for flow and transport in fractured rock, the Reynolds’ lubrication equation (Reynolds 1886; Zimmerman and Bodvarsson 1996; Eq. A1 in Paper III) will be used to calculate the hydraulic head field, and then the velocity field is obtained by assuming cubic law. It should be noted that both the cubic law and other generic power laws have been used in this thesis as has been discussed in the subsection 4.2.1. With both the lubrication equation and the power law, the primary flow parameter used is the fracture aperture instead of the hydraulic conductivity as has been used in the cases of flow in heterogeneous aquifers. Since local transmissivity can be explicitly related to fracture aperture, sometimes the transmissivity is also used as a primary flow parameter.

As presented in (2.8), the parameter $\beta$ defined in (2.9) is one of the key parameters quantifying the hydrodynamic control of retention processes in heterogeneous fractures. Similar to travel time $\tau$, $\beta$ is also a random variable dependent only on the flow conditions. As the statistical properties of the travel time $\tau$ have been widely studied in a variety of flow media in the literature (e.g., Dagan, 1989, Paper I) and $\beta$ has been relatively newly proposed in the transport models, it is desirable to establish a relationship between $\beta$ and $\tau$. When a relationship could be established between them, the calculation of solute discharge (2.4) would be signifi-
cantly simplified by elimination of one of the integration variables. Moreover it is difficult to evaluate the values of $\beta$ by field experiments. Its evaluation generally requires sorbing tracer tests which is costly and time consuming. In this thesis therefore much attention has been given to establishing an approximate relationship between $\beta$ and $\tau$ under various hydraulic conditions and fracture configurations (Papers III, IV and VI). With similar reasoning the establishment of an approximate relation between $\beta$ and $Q$ (volumetric flow rate) has also been attempted (Papers IV and VI). The approximate relation between $\beta$ and $\tau$ has also been established and applied in the prediction and evaluation of field measurement of breakthrough curves in the TRUE project at the Äspö site (Papers VII-XI).

4.1. First-order results (Paper III)

Using the first-order expansions, expressions for the first two moments of $\tau$ and $\beta$, as well as the joint moment between $\tau$ and $\beta$, are derived in Appendix C in Paper III which are summarized below. The means of $\tau$ and $\beta$ are:

\[
\langle \tau \rangle = \tau_0 \frac{x}{I_Y} \quad \text{(4.1)}
\]

\[
\langle \beta \rangle = \beta_0 \frac{x}{I_Y} = \frac{\langle \tau \rangle}{\beta_0}
\]

where the normalization constants $\tau_0$ and $\beta_0$ are given in (C6) of Appendix C in Paper III.

The variances and the joint moment of $\tau$ and $\beta$, with an exponential covariance function of $Y$ (i.e., with a finite integral scale) are given as (Paper III):

\[
\frac{\sigma^2_{\tau}}{\tau_0^2 \sigma^2_Y} = 8x' - 7 \ln x' + 2e^{-x'} + 7Ei(-x') + 3 \left[ \frac{(1 + x')e^{-x'} - 1}{x'^2} \right] - \frac{1}{2} - 7E
\]

\[
\frac{\sigma^2_{\beta}}{\beta_0^2 \sigma^2_Y} = 18x' - 11 \ln x' + 8e^{-x'} + 11Ei(-x') + 3 \left[ \frac{(1 + x')e^{-x'} - 1}{x'^2} \right] - \frac{13}{2} - 11E
\]

\[
\frac{\sigma_{\tau\beta}}{\tau_0 \beta_0 \sigma^2_Y} = 12x' - 9 \ln x' + 4e^{-x'} + 9Ei(-x') + 3 \left[ \frac{(1 + x')e^{-x'} - 1}{x'^2} \right] - \frac{5}{2} - 9E
\]

where $x' = x/I_Y$, and $E=0.577...$ is the Euler constant.

Monte-Carlo simulations are performed under the assumption of local cubic law for the variances of $\sigma_Y=0.1, 0.3$ and 0.5. The first and second moments of $\ln \tau$ and $\ln \beta$ obtained from the first-order analytical solutions (4.1) and (4.2) as well as from the Monte-Carlo simulations are compared in Figure 3 in Paper III. The first-order solutions (4.1) are independent of $\sigma_Y$. The simulation results indicate that the ensemble mean of $\beta$ increases with increasing $\sigma_Y$ (Figure 3a and 3b in Paper III), implying an enhanced retention in the rock matrix for increasing aperture heterogeneity (variability). The variances of $\ln \tau$ and $\ln \beta$ are strongly dependent on $\sigma_Y$, while the simulated variances of $\ln \tau$ and $\ln \beta$ are well approximated by first-order solution (4.2) (Figure 3c and 3d in Paper III). The simulation results indicate that $\ln \tau$ and $\ln \beta$ are strongly correlated, especially as $x$ increases (Figure 3e in Paper III). The strong correlation suggests a deterministic relation $\beta \sim \tau^m$ which will be discussed in next section.

4.2. Statistical properties of $\beta$ and $\tau$ (Papers III, IV and VI)

4.2.1. Flow in a single fracture (Cubic law vs quadratic law)

In the early models of fluid flow in rock fractures it was often assumed that the fracture has the simple geometry bounded by two flat parallel plates with a constant aperture between the plates. This constant aperture is equal to the mean aperture of the rough-walled fracture (e.g., de Marsily, 1986). The flow in this ideal fracture used to
be commonly described by a simple equation relating the flow rate with the cube of the aperture. This equation is known as the cubic law (4.3) where the volume flow rate can be expressed as:

\[ Q = \frac{\rho g}{12\mu} e^3 W h \]  

(4.3)

It is now well established in the literature that real fractures are rough-walled conduits with variable aperture and even points of contact (e.g., Novakowski and Lapcevic, 1994; Brown et al., 1998). The cubic law is often applied to rough-walled fractures by assuming that the cubic law holds locally for a given aperture, even though the aperture varies throughout the fracture (e.g., Neuzil and Tracy, 1981; Oron and Berkowitz, 1998). Such a representation simplifies significantly the mathematical representation of the flow. However, recent experimental evidence and field studies have indicated that the validity of the cubic law, even locally, is questionable. The cubic law tends to overestimate real flow rates (e.g., Brown et al., 1995; Konzuk and Kueper, 2004). The measured transmissivity is often lower than the predicted value and the velocity is not parabolic along the direction of the aperture, on which the cubic law is based, but rather skewed (e.g., Brown et al., 1995; Waite et al., 1998). In the discrete fracture network (DFN) simulations using the FracMan code, the empirical Doe's law has been assumed (e.g., Outter and Shuttle, 2000; Papers IV, VI, IX and X):

\[ T = 4e^2 \]  

(4.4)

where \( e \) is the fracture aperture and \( T \) is the transmissivity \([L^2/T]\). Here \( T \) is, and so is the flow rate \( Q \), proportional to \( e^2 \) rather than \( e^3 \) and \( Q \). Equation (4.4) is referred to as the quadratic law.

### 4.2.2. Transport in a single fracture

In Paper III, the \( \tau \) and \( \beta \) statistics have been studied using the Monte-Carlo simulation approach in a rectangular flow field. The flow is assumed to be driven by a horizontal hydraulic gradient. No flow condition is assumed on the other two boundaries. The cubic law is assumed to be applicable locally. Three different values of \( \sigma_y \) (0.1, 0.3 and 0.5) are used in the simulations. In the simulations for solute transport, a number of solute particles have been injected into the flow field in each realization. The values of \( \tau \) and \( \beta \) are computed by tracking the particles. A scattergram of the simulated logarithms of \( \tau \) and \( \beta \) is shown in Figure 4 in Paper III. A strong correlation between \( \tau \) and \( \beta \) with a power law relationship \( \beta \sim \tau^m \) has been found for all three values of \( \sigma_y \), from the simulations. The power \( m \) is approximately 1.5 for the simulation conditions considered in Paper III.

The statistical properties of \( \tau \) and \( \beta \) have further been investigated in Paper IV. The flow simulations in Paper IV include not only uniform gradient configurations, but also radially converging configurations. The "cubic law" is generalized to a "power law" with a power of \( n \). When \( n \) is 3, the power law becomes the cubic law. When \( n \) is 2, it becomes the quadratic law. This generation is supported by results of field observations that the conventional cubic law does not apply to all flow conditions (e.g., Outters and Shuttle, 2000). The Monte-Carlo flow simulations are performed for both the uniform flow and the radially converging flow, both the cubic law (\( n=3 \)) and the quadratic law (\( n=2 \)) have been tested in the simulations. In the transport simulations two sizes of the source sections \( W_s \), one being small \((W_s = 3I_y / 5)\) and the other being large \((W_s = 10I_y)\) for the uniform flow, and \( W_s = 1/16 \) and \( W_s = 1 \) of the circle's periphery respectively for the converging flow (see Figure 1 in Paper IV). The simulated \( \tau \) and \( \beta \) correlation is shown for the uniform flow in Figure 4-1a when the cubic law of flow is used (\( n=3 \)), and in Figure 4-1b when the quadratic law of flow is used (\( n=2 \)). The simulation results clearly suggest that \( \beta \) and \( \tau \) are related as \( \beta \sim \tau^m \) where \( m=1.4-1.8 \).

### 4.2.3. Internal and global heterogeneous fields (Paper VI)

In the stochastic approach, a lognormal distribution for hydraulic conductivity \( K \) (or
transmissivity) is often assumed (e.g., Dagan, 1989). With this probability distribution 
\[ K = K_G \exp[Y(x)] \], where \( K_G \) is the geometric mean of \( K \) and \( Y \) is a normally distributed random variable of \( N(0, \sigma^2_Y) \). The assumption of lognormal distribution of \( K \) is supported by available field data (e.g., Freeze, 1975; Sudicky, 1986). The covariance function of \( K \) is assumed to be exponential. The statistical properties of \( K \) are therefore determined by two parameter \( K_G \) and \( Y(x) \).

The studies of solute transport in heterogeneous aquifers and in a single fracture in this thesis focus only on a single field. In this case \( K_G \) or \( T_G \) can be assumed to be a constant. When studying the transport in fracture network, we consider in this thesis flow and transport also in a series of fractures. In this case, each fracture may have a transmissivity distribution, whereas different fractures may have different distributions. Thus \( T(x) \) in \( i^{th} \) fracture is expressed by (Paper VI)

\[
T_i(x) = T_G e^{Y_i(x)} = T_G e^{Z_i} e^{Y_i(x)}
\]

\[ C_r(t) = \sigma^2_Y e^{-r/\tau} \]  

(4.5)

where \( T_i(x) \) is the transmissivity field for the \( i^{th} \) fracture and \( Y_i(x) \) is a normally distributed RSF with the mean value and the variance described by \( N(0, \sigma^2_Y) \). \( T_G \) is a random number computed by \( T_G e^{Z_i} \), where \( T_G \) is the global geometric mean for the entire series of fractures and is assumed to be a constant, and \( Z_i \) is a random number characterized by \( N(0, \sigma^2_Z) \). It should be noted that, in (4.5), we have two levels of heterogeneity. The first level of heterogeneity is the geometric mean for the \( i^{th} \) fracture \( T_G \), which represents the variability among fractures. It characterizes the global heterogeneity. The second level of heterogeneity is the spatial variability \( Y_i(x) \) which represents the variability within a given fracture. It therefore characterizes the internal heterogeneity. \( T(x) \) will thus be determined by the heterogeneity on two levels: global \( \sigma^2_Z \) and internal \( \sigma^2_Y \).

4.2.4. Flow and transport in a fracture series and impact of internal and global heterogeneities (Paper VI)

In Papers III and IV, the flow and transport are considered in a single fracture. However, on larger scales flow and transport through fractured rock occur in multiple fractures. Each individual path may typically be long enough to pass through a number of fractures which can be characterized by several single fractures that are serially connected (Painter et al., 1998; Painter and Cvetkovic, 2001; Tsang, 1993; Paper X), as part of
fracture network. In a thread of serially-connected fractures each segment is part of a single fracture. The aperture of any single fracture in the series may vary from place to place, and the apertures of the different fractures in the series may vary even more profoundly. In Paper VI, we will consider the flow and transport in serially-connected fracture structures.

The impact of internal (within a single fracture) and global (among different fractures) heterogeneities of the fracture aperture on the $\beta$ and $\tau$ relation has been investigated. The internal and global heterogeneities are represented by the transmissivity given in Eq. (4.5) in which the random variables $Y$ and $Z$ characterize the internal and global heterogeneities respectively. Three degrees of global heterogeneity have been considered. They are (1) no global heterogeneity ($\sigma_Z^2=0$); (2) small global heterogeneity ($\sigma_Z^2=0.1$) and (3) large global heterogeneity ($\sigma_Z^2=2.0$). In each of the global heterogeneity considered, there are also two levels of internal heterogeneity $\sigma_Y^2=0.1$ and $\sigma_Y^2=2.0$, respectively.

The fracture structure considered is a quasi-3D fracture series (Figure 1 in Paper VI). The fracture aperture is assumed to be related to the transmissivity $T$ by the quadratic law (4.4). Monte-Carlo simulations have been performed for cases of different combinations of global and internal heterogeneities as discussed above. The $\beta$ and $\tau$ correlations for cases when the global heterogeneity is neglected ($\sigma_Z^2=0$) are presented in Figure 5a and Figure 5b in Paper VI. Figure 5c and Figure 5d in Paper VI show the $\beta$ and $\tau$ correlations for cases when the global heterogeneity is small ($\sigma_Z^2=0.1$), and Figure 5e and Figure 5f in Paper VI shows the correlations with large global heterogeneity ($\sigma_Z^2=2.0$). The results of these figures indicate that the $\beta$ and $\tau$ correlation is influenced more strongly by the global heterogeneity than the internal heterogeneity. The $\beta$ and $\tau$ become also less correlated as the internal and global heterogeneity increases. As has been found in the studies discussed earlier, the results here also suggest a power relation of $\beta \sim \tau^m$ in which $m=1.0-1.3$. The values of the power $m$ are, however, smaller than those obtained in Paper IV ($m=1.4-1.8$ in Paper IV). Moreover, the increase of the global heterogeneity will decrease the degree of correlation between $\beta$ and $\tau$ and will increase the mean values and the variances of both $\beta$ and $\tau$ significantly (Table 5 in Paper VI).

### 4.3. Statistical properties of $\beta$ and $Q$ and their correlations (Papers IV and VI)

We have investigated the statistical properties of $\tau$ and $\beta$ as well as their correlation in several papers of this thesis. In Papers IV and VI the statistical properties of $\beta$ and $Q$ are also studied. For variable apertures, there could exist some relation between the fracture aperture $b$ and the solute discharge $Q$, and consequently some correlation between $\beta$ and $Q$. From the application viewpoint, $Q$ is a directly measurable quantity and the establishment of some correlation between $\beta$ and $Q$ would link the parameter $\beta$, which characterizes the solute reaction/retention, and the experimentally measurable quantity $Q$.

The flow field configurations (including the size of the source section) and the flow model used for the study of the statistical properties and the correlations between $\beta$ and $Q$ in Paper IV are the same as those used for the studies of the $\beta$ and $\tau$ statistics. The simulated $\beta$ and $Q$ values are compared with the analytical solution (2.13) in Figure 4 of Paper IV. The results indicate that $\beta$ and $Q$ are more correlated for large sizes of the source section than for small ones for the uniform flow case. The analytical solution for a uniform aperture (2.13) captures the main trend of the simulated results even though some uncertainties exist.

In Paper VI, the studies are extended to a quasi-3D fracture series by considering both internal and global heterogeneities. The configuration of the fracture series is shown in Figure 1 in Paper VI. The local quadratic
law (4.4) has been assumed for flow field calculation. The Monte-Carlo simulations are performed for four cases with different combinations of the internal and global heterogeneities ($\sigma_z^2=0.1$ and 2.0; $\sigma_y^2=0.1$ and 2.0) as shown in Table 1 in Paper VI. For each case the size of the source section varied between 0.1 m and 5.0 m. In Figure 2 in Paper VI, the simulated $\beta$ and $Q$ data are compared with analytical solution for a uniform fracture (2.13) for a source section $W_s=5.0$ m. The results indicate that $\beta$ and $Q$ are strongly correlated, although $\beta$ and $Q$ have much small and large values in their respective cumulative distribution functions (CDFs) and complementary cumulative distribution functions (CCDFs) when the global heterogeneity increases (Figure 4 in Paper VI). The simulation results also shows that $\beta$ and $Q$ become less strongly correlated when the size of the source section becomes too small. The effect can be readily observed in Figures 3 in Paper VI when the source section is 0.1 m. The correlations between $\beta$ and $Q$ suggest an inverse power-law relationship of $\beta \sim Q^{-m}$ where $m=0.3-1.0$ for all cases studied in Paper VI.

In real fractured rock, the length of the fractures may also vary. By extending the previous studies in this thesis of the statistical properties of $\beta$ and $Q$, a fracture series of 22 fractures (compared with 3 in Paper VI) with a random distribution of the fracture length has been defined (Figure 4-2). The mean and the variance of the length distribution are 5 m and 1.6 m$^2$ respectively, which are typical for a fracture network in the real rock in Äspö region (Outters and Shuttle, 2000). The 22 fractures are connected in a similar way as in the previous studies in Paper VI. The fracture series is defined in a three dimensional domain as the fractures are layered (overlapped) in connection with the other fractures head to tail (Figure 4-2). The entire domain has a length $L=110$ m, a width $W=10$ m, and a height $H=0.24$ m. The domain is discretized into $1100 \times 100 \times 24$ elements. The 22 fractures are placed from layer 2 to layer 23 in the domain as shown in Figure 4-2. The Monte-Carlo flow and particle tracking simulations are performed for the 22 fracture series with 100 realizations. The simulated $\beta$ and $Q$ results for the four cases of different combinations of both the global and internal aperture heterogeneities ($\sigma_z^2=0.1$ and 2.0; $\sigma_y^2=0.1$ and 2.0) are presented in Figure 4-3. The results have the same trend as those for the fracture series of three fractures in Paper VI, but the spreading of the data of different realizations is larger. The $\beta$ and $Q$ are still correlated but to a lesser degree compared to the results in Paper VI. A power-relationship between the

![Configuration of a series of fractures with random length distribution](image)

Figure 4-2 Configuration of a series of fractures with random length distribution
simulated $\beta$ and $Q$ also exists as $\beta \sim Q^{-m}$ in which $m=0.6-0.9$.

5. Site Characterization and Performance Assessment Applications

5.1. Evaluation of TRUE experiments in Site characterization scale (Papers VII to XI)

The LaSAR modelling framework has been used to study the fluid flow and solute transport in different structures of heterogeneous aquifers and fractured media in the previous chapters of this thesis. The structures of the flow field studied are generic and there has been no intention to address any site-related specific issues. Using generic structures of the flow field has enabled us to explore with a broad perspective the statistical properties and correlations of several important quantities involved in the LaSAR approach, and to test a wide range of structure heterogeneity upon which the correlation is valid.

The LaSAR approach has also been directly applied to predict and evaluate the flow and transport of real field experiments using conservative and non-conservative tracers performed by the Swedish Nuclear Fuel and Waste Management Co. (SKB) at the Åspö site. These prediction and evaluation modelling are usually referred to as Site Characteri-
Hydrodynamic control of retention in heterogeneous aquifers and fractured rock 

zation (SC) modelling. In nuclear waste management, the performance assessment of the spent fuel repository is an important issue. The LaSAR modelling approach has also been applied to extend the SC model-ling to the performance assessment (PA) modelling of the radionuclide transport, and to bridge the gap between the SC and the PA. The approaches and results of the SC modelling will be discussed in this subsec-tion while the approaches and results related to the PA will be presented in the following subsection. Here we will only briefly present some of the typical modelling approaches and examples of the important results. More detailed information is too lengthy to be presented here and readers are referred to Papers VII - XI for it.

5.1.1. Modelling of the TRUE-1 field ex-periments

At the Åspö Hard Rock Laboratory (HRL) in southeastern Sweden, different stages of tracer tests have been conducted. The first stage of the Tracer Retention Understanding Experiments (TRUE-1) was performed at the TRUE-1 site in Åspö Hard Rock Laboratory (HRL) (Winberg et al., 2000; Paper VII). The sorbing tracer tests STT-1, STT-1B and STT-2 were performed in Feature A at the TRUE-1 site. The second stage of the TRUE Block Scale (BS) experiments were performed at the TRUE Block Scale rock volume. The Phase C sorbing tracer tests (C1, C2 and C3) were performed in the area around Structure #20 in the TRUE Block Scale tracer test volume (Poteri et al., 2002; Paper VIII). The third stage of the TRUE Block Scale Continuation (BSC) experiments (sorbing tracer tests BS2B) were performed in two flow paths: Path I in Structure #19 and Path II running from the background fracture BG1 to Structure #19 (Paper XI). Among these tests, all the three tests in the TRUE-1, the C1 test in the TRUE BS and the Path I test in the BS2B of the TRUE BSC were conducted basically in a single structure. The C2 and C3 tests in the TRUE BS and the Path II test in the TRUE BSC involved structures of multiple fractures. For more detailed information concerning the various tests, the reader is referred to Papers VII to XI and the references therein.

The main objective of the LaSAR modelling approach for SC scale presented in Papers VII - XI is to predict and evaluate the results of the TRUE experiments, and to identify possible and important retention processes of the tracers in the fractured rock. By considering the heterogeneity of the fracture structure and the variability of different retention parameters, and by calibrating the model results with the breakthrough curves (BTCs) of the field tests, the model approach will be able to improve our understanding of the importance of the various retention processes. Through "blind prediction" using "representative" data of some laboratory and field measurements, the modelling approach will provide insights to ranges of the variability of the parameter and to capture additional important retention mechanisms and processes that may not be readily revealed in the laboratory and field measurements.

In the modelling, the flow and transport in both a single fracture and in multiple fractures have been considered in consistency with the fracture structures in different field tests. The retention processes considered are sorption on the fracture surface, described by $K_a$, and diffusion/sorption in the adjacent rock matrix, characterized by $\kappa$ as defined in (2.11). When applied to modelling of BTCs, the LaSAR modelling approach usually proceeds in the following steps:

- The travel time distribution $g(\tau)$ is first determined by deconvoluting BTCs of conservative tracers, while accounting for the diffusion into the rock matrix. The actual form of $g(\tau)$ is usually assumed to be inverse-Gaussian or lognormal. The first two statistical moments of the water residence time are then calibrated for each flow path.
Alternatively the values of transmissivity ($T$) of a fracture (or a fracture network) measurement directly in the field, and boundary conditions are used in the Monte-Carlo numerical simulations (as have been described in the previous sections) to infer the first two statistical moments of $\tau$. It should be noted that the data points are usually extremely scarce (only a few for the entire flow path in most of the cases), the statistical inference may not be very reliable and the uncertainties involved are large.

- using $g(\tau)$, BTCs of sorbing tracers are modelled by accounting retention processes. The parameter groups $B=k\kappa$ and $A=kK_a$ are calibrated on the measured BTCs of sorbing tracers until a best fit is obtained. The two procedures are iterative until satisfactory fittings are obtained consistently for all tracers used in the same flow path. While the relation between $\beta$ (2.9) and the travel time $\tau$ (2.1) is assumed to be linear, i.e., $\beta=k\tau$.

The linear $\beta$ and $\tau$ relationship may be obtained by linear fitting the simulated data, one example is given for TRUE-1 STT-1 test in Figure 5-1. It should be noted that in the previous sections of this thesis, we usually consider a power-law relationship between $\beta$ and $\tau$ ($\beta=\tau^m$). The modelling results of the BTCs indicate that an approximation of linear relation seems to give good results under the conditions of these TRUE tests.

Figure 5-2 shows one set of the evaluation results based on the above procedures. The evaluated parameters are presented in Tables 5-1 and 5-2.

---

**Table 5-1 Calibrated parameters for TRUE-1 STT-1 tests**

<table>
<thead>
<tr>
<th>Tracer</th>
<th>$k\kappa$ ($h^{1/2}$)</th>
<th>$kK_a$ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HTO</td>
<td>0.1025</td>
<td>0</td>
</tr>
<tr>
<td>Sr-85</td>
<td>0.465</td>
<td>0.048</td>
</tr>
<tr>
<td>Rb-86</td>
<td>2.16</td>
<td>3</td>
</tr>
<tr>
<td>Cs-137</td>
<td>11.43</td>
<td>7.2</td>
</tr>
</tbody>
</table>

**Table 5-2 Calibrated first two moments of water residence time for TRUE-1 STT-1 test**

<table>
<thead>
<tr>
<th>Test</th>
<th>Mean (h)</th>
<th>Variance (h$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>STT1</td>
<td>8</td>
<td>40</td>
</tr>
</tbody>
</table>

One of the important findings made by the calibration procedures is that the tracer reaction/retention parameters of laboratory measurements for the TRUE-1 (MIDS data, Byegårds et al., 1998) are usually much smaller than the model calibrated values. For example in the TRUE-1 tracer tests in a single fracture, the $\kappa$ value from the MIDS data has to be multiplied by a factor 50-65 so that the modelled BTCs match the BTCs of the TRUE-1 field tests for strongly sorbing tracers like Ba, Rb and Cs (Paper VII). In terms of the constituent parameters involved in $\kappa$, the porosity ($\theta$) is estimated to be 5-10 times larger, the formation factor ($f$) up to 100 times larger, and the sorption coefficient ($K_d$ with units m$^3$/kg) roughly 4-5 times larger.

The rock samples used in the laboratory measurements were often obtained from the intact rock matrix. The retention properties of these samples represent the properties of the "true" rock matrix. In field tests, however, there always exist layers of rocks with various degrees of alteration adjacent to the fracture surface. This "rim zone" may have much larger retention capacity compared to the deeper intact rock matrix. The time scale involved in the tracer tests is relatively short (this is the time scale of site characterization) and the penetration depths (especially sorbing tracers) are relative short. The rim zone
Figure 5-1 Simulation data on $\beta$, $\tau$ from 89 realizations. One symbol represents simulated $\beta$, $\tau$ data for one realization obtained by averaging over 100 trajectories in that realization.

Figure 5-2 Evaluation of BTCs of STT-1 tests. Symbols are experimental data. Lines are obtained from evaluation.
therefore plays a dominant role in the retention of the tracers. This is considered to be the main reason that the laboratory measure data failed to predict the BTCs in the field tests. Other factors may also be responsible for the discrepancy, like sorption on the gouge materials.

5.1.2. TRUE Block Scale modelling (Papers VIII and X)

The objective of TRUE Block Scale project was to understand flow and transport in the block scale (10-100 m). The evaluation of sorbing tracer tests (C1, C2 and C3) has been conducted using similar procedures as for TRUE-1 (Paper VIII). The evaluation results indicate that the diffusion/sorption in the matrix in the rim zone are the dominant retention processes for the time scale in TRUE Block Scale. The estimated effective retention parameters are comparable to TRUE-1.

In connection with the TRUE program, and to bridge between site characterization (SC) and performance assessment (PA) approaches to solute transport and retention in fractured rock, an international cooperation has been established in the framework of the Åspö Task Force on groundwater flow and tracer transport. A series of tasks focusing on different aspects of the test and modelling within the program have been defined. Among these tasks, Task 6 aims at conducting performance assessment (PA) modelling using site characterization (SC) data.

Task 6D modelled the tracer test C2 in TRUE Block Scale project at the Åspö HRL and dealt with fracture networks of major conductive features that have been identified in the Åspö TRUE Block Scale experiments (Andersson et al., 2002a, Papers VIII and X). The works of the subtasks in Task 6 related to the PA will be discussed in the next section. The modelling of Task 6D is based on the Task 6C semi-synthetic hydro-structural model (Dershowitz et al., 2003). The simulations are different from the evaluation for TRUE-1 and TRUE Block Scale tests (Papers VII and VIII), where the calibration was performed both on conservative tracer BTCs to back up the parameter groups $k\kappa$ and $kK_a$.

Now a forward modelling for flow field based on the Task 6C structure model is performed to get the temporal moments based on the simulated $\tau$ values. The calibration is made only for the parameter groups $k\kappa$ and $kK_a$. The modelling results in Task 6D indicate that the mean travel time for the flow path including background fracture is longer than the path excluding background fracture (paper X).

5.1.3. Block Scale Continuation modelling (Paper XI)

The results of the Block Scale Continuation modelling (Paper XI) are summarized in the following. For detailed information the reader is referred to Paper XI of this thesis. The major difference of this modelling from the other site characterization modelings presented earlier is that step-wise heterogeneity of the retention properties of the rock matrix has been considered. In the Block Scale Continuation modelling (Paper XI), two flow paths have been selected for BS2B tests. The first flow path is in a single Type 1 (fault) structure (Structure #19) with a length of approximately 20 m. The second flow path starts in the background fracture BG1 and ends in Structure #19. The modelling of the BTCs for the BS2B test was conducted in two steps like the TRUE-1 and TRUE BS. The first step was "blind prediction", the second step was subsequent evaluation (Paper XI). The blind predictions of breakthrough curves (BTCs) in the BS2B test were conducted by calibrating on the data of the preparatory conservative tests CPT-4c and CPT-4b performed in the same flow paths. The results from blind prediction of the BTCs of BS2B test are shown in Figure 5-3 for Path I and Figure 5-4 for Path II. The prediction results generally match the measured BTCs well for the first flow path, but overestimate the retention for Rb-86. For the second flow path, we underestimate the retention for all tracers.

The evaluations are performed by assuming effective (uniform) retention parameters and by accounting for the depth-wise heteroge-
Hydrodynamic control of retention in heterogeneous aquifers and fractured rock

Figure 5-3 Comparison of predicted and evaluated BTCs with measured data for Path I in BS2B test. The symbols are measured data. The dashed lines are predicted BTCs. The solid lines are evaluated BTCs.

neity of the retention parameters. Comparison of the estimated \( \kappa \) (2.11) values for two flow paths indicate that the background fracture BG1 has lower \( \kappa \) compared to Structure #19, implying weaker retention in BG1 as quantified by the material parameter group \( \kappa \).

In accordance with the definition of \( \beta \) (2.12), the slope \( k \) is equivalent to the inverse of an effective "retention" half-aperture. The retention aperture \( 2b \) can also be estimated using the streamtube model (2.13) as

\[
2b = \frac{Q \tau}{LW} \quad (5.1)
\]

The estimated retention aperture \( 2/k \) used in the predictions and evaluation for the flow path I is consistent with a simplified estimate based on a streamtube model (5.1) with the effective width of the flow path of around 0.07 m. For flow path II the flow rate appears uncertain; a consistent estimate of retention aperture \( 2/k \) is obtained with a streamtube model, flow rate is on the order 2-4 ml/h.

5.2. From site characterization to performance assessment modelling (Papers IX and X)

It should be emphasized that the LaSAR modelling framework developed in generic flow fields and applied to SC modellings is readily extended to apply to the PA modelling. In the PA modelling, however, the water flow will be assumed to occur under natural gradient boundary conditions giving water travel times corresponding to the "PA time scale", in contrast to that in the SC modelling where the flow is subject to enhanced gradient of pumping.
In Task 6B the PA scale modelling is performed by reducing the flow rate 1000 times from STT-1b test for a single fracture, other conditions are unchanged (Paper IX). The PA modelling is also performed in a uniform flow condition driven by a hydraulic gradient of 0.1% in Task 6B2 (Paper IX).

Task 6E extends the Task 6D transport calculations to a reference set of PA time scales and boundary conditions (Paper X). Task 6E also deals with the modelling of solute transport over longer distances including several geological features. The basis for the modelling of Task 6E is also the block scale semi-synthetic hydro-structural model developed within Task 6C (Dershowitz et al., 2003) over 200m cubic block. The flow and transport occur in a number of deterministic features and many background fractures.

The modelling results for the PA modelling indicate that the retention in the unaltered rock is important process in contrast to the SC time scale where the retention in the rim zone adjacent to the fracture is dominated.

5.3. Impact of temperature increase on retention of radionuclide (Paper V)

In Sweden the KBS-3 (SKB, 1999) concept of design of a spent nuclear fuel repository is a multibarrier system. The temperature in the geosphere surrounding the canisters is about 15°C initially at the fuel canister deposition. The temperature will increase to about 60°C after tens of years to hundreds of years (SKB, 1999). The temperature condition in the Task 6 tests has been assumed to be similar to the initial temperature condition, i.e., about 15°C. The same temperature has been implied in the TRUE-1 tests and modelling.

The base cases considered in Paper V are the cases of Task 6A and Task 6B at T = 15°C (SKB, 1999). Here we investigate the effect of increased temperature, from about 15°C to 60°C, on the solute transport and retention in rock fractures. Two scenarios have been considered for comparison. In the first the temperature is at 15°C, corresponding to the initial stage of the repository. In the second the temperature is at 60°C, corresponding to the maximum temperature after the closure of the repository.

The simulation results for both Tasks 6A and 6B show enhanced retention for all tracers due to the increase of the temperature. The retention of the strongly sorbing radionuclides increases significantly.

Figure 5-4 Comparison of predicted and evaluated BTCs with measured data for Path II in BS2B test. The symbols are measured data. The dashed lines are predicted BTCs. The solid lines are evaluated BTCs.

In Task 6B the PA scale modelling is performed by reducing the flow rate 1000 times from STT-1b test for a single fracture, other conditions are unchanged (Paper IX). The PA modelling is also performed in a uniform flow condition driven by a hydraulic gradient of 0.1% in Task 6B2 (Paper IX).
tracers is more profoundly influenced by the temperature increase than the weakly to moderately sorbing tracers. In addition, the temperature effect seems to be stronger for the tracers in Task 6B, which has a lower pumping rate than in Task 6A. It can therefore be concluded from this study that the increased temperature may possibly provide additional safety margin for the retention of radionuclides, where strongly sorbing tracers acquire larger safety margins than weakly sorbing tracers.

6. CONCLUSIONS
Within the Lagrangian Stochastic Advection-Reaction (LaSAR) modelling framework, fluid flow and solute transport in heterogeneous aquifers and fractured rock have been studied. Based on the model concepts developed and modelling results obtained for the generic flow fields, the LaSAR modelling approach has been applied to site characterization (SC) modelling in "blind" prediction, evaluation, and interpretation of the results of various field tracer experiments conducted at the Äspö HRL in Sweden. In addition to the SC applications, the model approach has also been extended to be applied to the performance assessment (PA) modelling.

The conclusions are summarized in following aspects:

**Modelling approach**
With the adoption of the Lagrangian coordinate system, the flow and the transport modellings are decoupled in the LaSAR framework when the retention processes are linear. The fluid flow modelling can be performed by Monte-Carlo numerical simulations, and this is the part this thesis focuses on. The transport modelling involves analytically solving the transport and conservation equations in the Lagrangian coordinate system. Iterative and calibration procedures may be needed when there is a lack of truly representative data for the reaction/retention processes.

**2D aquifers**
For the transport of nonreactive solute transport in heterogeneous aquifers, the Monte-Carlo simulation results of the statistical properties of the travel time (or water residence time) \( \tau \) indicate that \( \tau \) can be well approximated by a lognormal distribution up to a log-variance of hydraulic conductivity of 4. The mean travel time is found to be a nonlinear function of distance for shorter distances, and becomes a linear function for longer distances for the conditions in this thesis for residence concentration. Later work by Demmy et al., (1999) found that the mean travel time \( \tau \) for flux injection is a linear function of distance. Comparison between the Monte-Carlo simulation results and the results of first-order approximation reveals that the analytical solutions of the statistical moments of \( \tau \) are valid only when the variability of the aquifer properties is small, e.g., the degree of heterogeneity is low.

Results of Monte-Carlo simulations for a two-dimensional statistically isotropic aquifers indicate that the mean of reaction flow path \( \mu \) is nonlinear function of distance for shorter distance, linear function for longer distance, the variance of \( \mu \) is a nonlinear function of distance. \( \mu \) and \( \tau \) are well correlated for the simulation cases in this thesis.

**Statistical properties of \( \beta \)**
In a variety of flow and transport conditions considered in this thesis, our modelling results show that the parameter \( \beta \) and \( \tau \) are generally strongly correlated. A deterministic power-law relation has been suggested for the correlation as \( \beta \sim \tau^m \). The degree of the \( \beta \) and \( \tau \) correlation may decrease with increasing fracture heterogeneity. For the case of flow in a fracture series, the ensemble means \( <\beta> \) and \( <\tau> \) generally increase with increasing fracture heterogeneities and are influenced more strongly by the global heterogeneity than by the internal heterogeneity. The increase of \( <\beta> \) implies that diffusion into the rock matrix is in a statistical sense enhanced by increasing fracture heterogeneity.

The statistical properties of \( \beta \) and \( Q \) studied both in a single fracture and in a quasi-3D fracture series have shown that there also
exists correlation between $\beta$ and $Q$ described by an inverse power-law relation $\beta \sim Q^{-m}$. The mean values of $Q$ change relatively small for the conditions in this thesis. Moreover, $\beta$ and $Q$ are more strongly correlated when the internal heterogeneity is small or when the global heterogeneity is large. For all levels of internal and global heterogeneities, $\beta$ and $Q$ are less strongly correlated when the size of the source section decreases.

**Predictability of tracer breakthrough**

The micro-structural information of the retention parameters (e.g., matrix sorption coefficient $K_d^m$, fracture surface sorption coefficient $K_a$ and porosity $\theta$) constitute the basis for predictions of reactive tracer breakthrough curves, provided that the water residence distribution $g(\tau)$ is known and the $\beta$ variability is accounted for. The $g(\tau)$ can be obtained by numerical simulations with known boundary conditions and hydraulic properties of the domain. However it is impractical or difficult to obtain complete hydraulic data in the field. The $g(\tau)$ was thus obtained by calibration on the conservative tracer test conducted in the same flow path for the blind predictions of TRUE tests. Given calibrated $g(\tau)$, predicted BTCs using laboratory retention parameters from the unaltered zone underestimated measured BTCs for TRUE-1. Predictability was significantly improved using the retention parameters for the rim zone in TRUE Block Scale and TRUE Block Scale Continuation tests.

**Retention properties**

Through inverse modelling by deconvolution of the breakthrough curves (BTCs) of conservative tracers measured in the field and by calibrating on the BTCs of non-conservative tracers, in-situ values of the reaction/retention parameters can be estimated. The data thus estimated can be compared with those of laboratory or field measurement for the same parameters. Agreement or discrepancy between them may provide insights into the reaction/retention processes actually involved in the field experiment. For example, in the TRUE-1 site characterization modelling, it has been revealed that through-diffusion tests on unaltered rock samples are likely to underestimate retention properties (such as porosity, sorption coefficient) of rim zone. In the SC time scales, the retention in the rim zone adjacent to the open fracture plays a dominant role and, when using the data of the unaltered rock may considerably underestimate (by a factor of a few tens) the retention capacity of the fractured rock system. The results of the PA modelling indicate that diffusion into and sorption in the unaltered rock matrix become important retention processes in contrast to the situations in the site characterization modelling, where the retention occurs mainly in the rim zone due to the short time scale (and thus small penetration depth) involved.

**Estimation of hydrodynamic control parameter $\beta$**

The hydrodynamic control parameter $\beta$ can be estimated in several ways:

- **Monte-Carlo simulations of particle tracking** for individual trajectories. An approximate linear relation could be established by linear fitting of the simulated $\beta$ and $\tau$ values as $\beta = k \tau$. Following the definition of $\beta$ (2.12), the slope $k$ is interpreted as the inverse of half "retention aperture". The numerical simulations can be used to determine $\beta$ statistically, provided that the fracture aperture statistics are known.

- **Assume a streamtube model** where $\beta$ is related to flow rate $Q$ by $\beta = 2LW/Q$. The width $W$ may be considered as the source size for uniform flow configuration or as the borehole diameter for radially converging flow. The length $L$ may be estimated from the average transport length, e.g., from the estimated distance between the injection and pumping borehole (or control plane). The flow rate $Q$ may be estimated as injection flow rate.

- **The variability of $\beta$** strongly influences the BTCs of sorbing tracers. Capture of
\( \beta \) variability is thus important for sorb- ing tracer transport in the field. However the distribution of \( \beta \) can not be measured directly. By using the linear relationship between \( \beta \) and \( \tau \), the entire distribution of \( \beta \) is replaced by the distribution \( g(\tau) \) and a slope \( k \). The problem is thus to find \( g(\tau) \) and estimate \( k \).

**Implications for PA modelling**

In the context of PA time scale, it is anticipated that the uncertainty in estimating \( \beta \) would increase. The modelling results in the TRUE tests (SC time scale) indicate that a linear approximation between \( \beta \) and \( \tau \) seems to give good modelling results. The question is if the linear relation still valid under PA time scale, if so under what conditions? More extensive simulations which combine full discrete fracture network modelling with internal fracture variability, are required for quantifying \( \beta \)-statistics on large scales.

The modelling results for a generic fracture (or sequence of fractures) indicate that \( \beta \) is to some extent correlated with the flow rate \( Q \), depending on the variability in hydraulic properties. If one can show that even under PA conditions \( \beta \) and \( Q \) are sufficiently correlated (using simulations), then it can provide an efficient means of estimating \( \beta \)-statistics in the field. The flow rate \( Q \) may be measured relatively easily along boreholes using Posiva flow-logs. If a distribution of \( Q \) can be inferred by conditioning on the measured values, the statistics of \( \beta \) can be estimated based on the established correlation between \( \beta \) and \( Q \). A similar proposal has already been put forward (Moreno and Neretnieks, 1993b).
7. References


