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A Brief Review of Bacterial Transport in Natural Porous Media

T. R. Ginn

December 1995

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest National Laboratory
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Summary

This report reviews advances in the descriptions of microbial transport processes. The advances can often be translated into technological advances for solute transport, with potential applicability to a number of subsurface concerns related to solutes. The processes involved in microbial transport include physically controlled processes, chemically controlled processes, and biologically controlled processes. The physical processes involved in the transport of microbes include advection, diffusion, dispersion, straining, filtration, and exclusion. Biomass removal by chemical reactions has received less attention, and included electrostatic attraction and hydrophobic sorption. In addition, microbiologic processes affecting the fate and transport of microbes in the subsurface include growth and decay; motility and chemotaxis; biological adhesion; and predation. Interdependencies among these processes arise through coupling, e.g., as multiscale mixing in heterogeneous environments affects nutrient availability (growth) and filtration velocities (attachment).

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Contents

Summary	v
1.0 Introduction	1
2.0 Processes Involved in Microbial Transport in Heterogeneous Media	1
2.1 Physically Controlled Processes	1
2.2 Chemically Controlled Processes	4
2.3 Biologically Controlled Processes	5
3.0 Modeling Microbial Transport Processes in Porous Media	5
4.0 Summary of Model Representations of Microbial Transport Processes	7
5.0 Effects of Heterogeneities on Reactive Transport	9
6.0 Upscaling	11
7.0 Roles for Modeling of Bacterial Transport	13
8.0 References	14

Figures

1 Multiple Scales of Heterogeneity Depicted Through the Effective Value of a Property Measured Over Increasing Sample Volume	10
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1.0 Introduction

Any treatment of the potential problems associated with the transport of pathogenic bacteria and viruses over long times or distances (> 100 m) requires a thoroughly tested ability to detect and predict the fate and migration of such pathogens (Martin and Noonan 1977; Keswick and Gerba 1980; Keswick et al. 1982; Matthess et al. 1988; Falcão et al. 1993). This detection and prediction ability is also required for effective bioremediation that involves the transport of microbes and/or microbial nutrients within a remediation site. Experimentation with and conceptualization of microbial transport and reaction processes involves tools (experimental and theoretical) that are typically generalizations of those used in the study of solute transport. Consequently, advances in the technology of microbial transport can often be translated into technological advances for solute transport, with potential applicability to a host of subsurface concerns related to solutes.

This report begins with the important physical, chemical, and biological processes controlling microbial transport in heterogeneous porous media. Past modeling of microbial transport processes in column-, intermediate-, and field-scale experiments is also summarized. Finally, the effects of heterogeneities on reactive transport and new upscaling approaches are discussed.

2.0 Processes Involved in Microbial Transport in Heterogeneous Media

2.1 Physically Controlled Processes

The physical processes controlling the transport of microbes in the saturated subsurface are relatively well established as being advection, diffusion, dispersion, straining, filtration, and exclusion (Harvey et al. 1993; Hornberger et al. 1992). Physical properties also can affect the processes of chemical deposition (attachment, sorption) and entrainment (detachment, desorption). Experimental confirmation of mathematical representations of these processes requires the unraveling of observations that represent the integrated effects of processes in the real environment. This unraveling is aided by an understanding of the physicochemical controls on these processes in terms of measurable aquifer properties.

Advection is defined as conveyance of a material (in this case, solute or microbes) at the field-scale average groundwater velocity, and it is controlled by the average hydraulic conductivity of the medium. Prediction of advection requires estimation of the regional groundwater velocity on the scale of the transport experiment; this is estimated based on measurements of the piezometric head gradient over the entire field, combined with an averaged (upscaled) hydraulic conductivity value representative of the field. When tracer data are available, the advective velocity can be computed directly at sampling wells from the mean travel time (the arrival time of the center of mass) of an instantaneously injected pulse. If the sampling well is at distance L from the injection point, and the observed mean travel time is T , then the field-scale average pore-water velocity is defined as $v = L/T$.

Diffusion of microbes follows a pattern of damped Brownian diffusion as applied to colloids, with the Fickian coefficient of diffusion inversely proportional to the radius of the colloid (de Marsily 1986), assuming that the colloid density is close to that of the solvent. It is generally agreed that diffusion is an insignificant transport process for microbes in a convective field. This fact becomes important when diffusion is the transport process by which solutes visit smaller scale pore spaces.

Dispersion in saturated flow is controlled by variations in hydraulic conductivity that create small-scale fluctuations in the velocity field. This important process is not well understood or well measured for microbial transport. When hydraulic conductivity varies on multiple-length scales (as is frequently the case in natural porous media), the resulting velocity fluctuations (which control dispersion) reflect multiple scales of randomness (in other words, multiple scales of correlation). The result is that dispersion, as understood and predicted by conventional transport theory, is not representative of the actual spreading process. Recent studies indicate that understanding the true spreading process requires characterization of the "scales of variability" of the hydraulic conductivity. That is, to understand the process, the frequencies and amplitudes of its spatial variability must be characterized (Dagan 1984; Cushman and Ginn 1993). Additionally, any large-scale heterogeneities (on the scale of the experimental domain) require deterministic characterization because they may dominate the flow pattern and do not admit a stochastic description. Predictive theories usually depend on stochastic representations of the velocity field fluctuations or of the hydraulic conductivity fluctuations that cause them, superposed on an interpolated image of large-scale heterogeneities determined from distributed measurements. Although the most valuable measurements for estimating the statistical properties of the fluctuations are tracer concentrations over time at multiple locations in the field (Deng et al. 1993a,b), it is also necessary to know something of the scales of heterogeneity a priori to designing a sampling scheme capable of resolving the dispersive process. Valuable observations for this purpose include visual inspection of aquifer material transections (to qualitatively identify heterogeneity scales), high-resolution dense sampling of hydraulic conductivity on small scales at such transections (Davis et al. 1993), and distributed small-scale measurements of hydraulic conductivity in the flow field.

Straining and physical filtration represent the removal of microbes from solution by physical (geometric and electrostatic) forces, as opposed to active biological adhesion. Straining is the trapping of microbes in pore throats that are too small to allow passage and is exclusively a result of pore geometry (Corapcioglu and Haridas 1984). Prediction of mass removal by straining, based on purely geometric relations between the effective diameter of colloids and the diameter and packing (coordination number) of grains forming the porous media, is detailed by Herzig et al. (1970) and experimentally by Sakthivadivel (1966, 1969). An important result of these analyses for microbial transport is the finding that straining is not significant in idealized packed beds (porous media made up of identical spherical grains) where the colloid diameter is less than 5% of the porous media grain diameter (Herzig et al. 1970; Corapcioglu and Haridas 1984; McDowell-Boyer et al. 1986; Harvey and Garabedian 1991). This rule has been generalized by Matthess and Pekdeger (1985) to porous media made up of a distribution of grain sizes.

Physical filtration is the removal of particle mass from solution via collision with and fixation (or adsorption) to the porous media. Physical forces involved are gravity, particle-particle and particle-solvent collisions (Brownian forces), electrostatic interaction potentials between the particle and the porous media, van der Waals attractive potentials, particle inertia, and pore-water hydrodynamic forces (Yao et al. 1971; McDowell-Boyer et al. 1986). Filtration due to gravity is termed sedimentation (McDowell-Boyer et al. 1986; Corapcioglu and Haridas 1984) and depends on particle buoyancy.

Many bacteria and viruses are neutrally buoyant (particle density is close to that of the solvent), in which case sedimentation is negligible. However, Gerba et al. (1975) report significant sedimentation for some bacteria; in these cases, the gravitational velocity expressed by Yao et al. (1971) can serve as a measure of sedimentation (Corapcioglu and Haridas 1984). Filtration due to the remaining physical forces (Brownian, electrostatic, van der Waals, and pore-water hydrodynamic) is the dominant mechanism for removal of inert particles and has received enormous attention, partly as a result of its quantitative tractability [cf. review by McDowell-Boyer et al. (1986)]. Physical filtration terminology is often used inconsistently between disciplines; while the term "physical filtration" is used in the engineering literature, "adsorption" is used in chemistry, and "attachment" in the microbiological literature.

Recent research has demonstrated that physical filtration is influenced by solute ionic strength (through its effect on the size of the diffuse double layer (Sharma et al. 1985; Scholl et al. 1990; McDowell-Boyer 1992; Tan et al. 1994), pH (ionization of mineral grains and coatings (McEldowney and Fletcher 1988), and mineralogy (Scholl et al. 1990). Shonnard et al. (1994) found significant increases in adsorption of a trichloroethylene- (TCE-) degrading bacterium as a result of increased ionic strength of solution in coarse sands. Scholl et al. (1990) found that iron hydroxide-coated sands filtered more bacteria than clean sand, as expected given the effect of iron hydroxide coatings on the interaction potential. Primarily quartzitic materials have negatively charged surfaces (surface charge being the electrostatic potential upscaled to the pore scale; Overbeek 1952), as do most bacteria; thus, the hydrodynamic and attractive (van der Waals) forces must overcome the repulsive electrostatic force for bacterial immobilization to occur. Iron hydroxide-coated sand grains have positive surface charges, thus reversing the electrostatic force from repulsive to attractive, increasing the likelihood of microbial attachment. The mechanism of hydrophobic sorption has also been investigated by measuring the hydrophobicity of the microbe on porous media with a homogeneous distribution of organic matter (McCaulou et al. 1994). Even small amounts of organic matter on porous media may result in microbial mass removal (Ryan and Gschwend 1990). [Despite the geochemical nature of controls on the electrostatic interaction potential, the potential is treated here as a physical filtration process, as done by Corapcioglu and Haridas (1984); the literature is inconsistent.] Subsequent study by Scholl and Harvey (1992) further identified effects of metal oxides, as well as organic material, on attachment reversibility.

The filtration effect resulting from these physical forces is quantified through upscaling the interaction potentials and hydrodynamic forces in the pore-scale behavior of a chemically inert spherical particle transported through a uniformly packed homogeneous bed of spherical grains (Herzig et al. 1970; Shaw 1976; Tien et al. 1979). The resulting mass removal is cast in terms of pore-water velocity, viscosity, and density; media grain size; and media porosity. The resulting "filtration theory" relations are well known [cf. textbook by de Marsily (1986)] and have been widely applied to microbial transport following the introduction of the idea by Harvey et al. (1989) and its application by Harvey and Garabedian (1991). The consequent reversibility of physical filtration mass removal (via reduction in solute ionic strength; Scholl et al. 1990; McDowell-Boyer 1992), as well as the reversibility incurred by changing the pore-water velocity direction (Sakthivadivel 1966), has generally been ignored in applications of filtration theory to microbial transport because the filtration models represent irreversible deposition only under conditions of uniform flow direction and fixed solution chemistry.

Exclusion denotes the phenomenon of transported particles moving faster than the pore water, or at least faster than the measured pore-water velocity (usually measured by the breakthrough of a salt

tracer). The process is hydrodynamic chromatography (Small 1974). Pore-water velocity within a capillary is generally parabolically distributed with the maximum velocity at the centerline being roughly 1.5 times the average pore velocity (de Marsily 1986). The velocity at the pore walls is zero. Conventional transport theory assumes that molecular-scale solutes thoroughly sample the full distribution of velocities in convecting pores of all significant sizes. The diffusion of solutes into microscale porosities (such as intragranular porosity occurring in sands) can be slow (Wood et al. 1990), requiring a two-porosity (otherwise known as mobile-immobile region) treatment of the transport (e.g., Coats and Smith 1964; DeSmedt and Wierenga 1979). Recently, nonuniform distribution of salt tracers has been observed even over large (convecting) porosity ranges [e.g., in media without microporous structure (Harvey 1993)]. Microbes and large colloids, by virtue of their size, necessarily incur a more nonuniform distribution over pore sizes, because the size of the microbes prevent their centers of mass from sampling points very close to pore walls. Consequently, the average pore velocity will be higher for microbes than for a molecular (salt) tracer, and the microbes will precede the tracers downgradient.

Results from hydrodynamic chromatography (Dodds 1982; quoted in de Marsily 1986) indicate that in idealized porous media, the ratio between colloid and water average velocities is quite small, usually between 1.0 and 1.1. Further, the occurrence of exclusion requires the colloid diameter be $< 1\%$ of the media grain diameter. The velocity ratio can be amplified in the presence of ionic forces (this is anion-exclusion, as opposed to size-exclusion). When the electrostatic forces between the media and colloid are repulsive, as is the case with negatively charged microbes in negatively charged quartzitic media, the force field tends to channel the microbes closer to the pore throat centerlines and away from the walls (and the slower velocities) (de Marsily 1986). Anion exclusion has been observed in unsaturated column experiments (Porro et al. 1993) and in field sampling of concentration/depth profiles of tritium, chloride, and sulfate in the irrigated coastal plains of Israel (Gvirtzman and Gorelick 1991). The resultant effect on the velocity ratio can be large; Gvirtzman and Gorelick (1991) report a ratio of 2.5 between anion (sulfate) and tritium average velocities. Anion exclusion, like filtration and hydrodynamic chromatography, is affected by the solution ionic strength. Thus, exclusion is expected in microbial transport experiments in such media, where the microbe size is roughly two orders of magnitude smaller than the media grain size. The effect is more pronounced at larger observation scales, as has been reported in many recent works [Wood and Ehrlich 1978 (although it was concluded that the aquifer was actually sorbing the bromide tracer); Pyle 1979; Engfield and Bengtsson 1988; Harvey et al. 1989, 1993; Powelson et al. 1993 (using viruses); Shonnard et al. 1994].

2.2 Chemically Controlled Processes

Mass removal by chemical (surface and ion-exchange) reactions involving microbes has received much less attention because early research dealt with porous media that were assumed to be inert (Weber 1972; Corapcioglu and Haridas 1984). Research on the effects of particulate surface chemistry on electrostatic attachment mechanisms is noted by McDowell-Boyer et al. (1986). More recent efforts that focused on microbial particulates have identified chemical processes and controlling geochemical properties that can strongly influence microbial mass removal through both electrostatic attraction and hydrophobic sorption. Hydrophobic sorption as conceptualized for organic solutes (de Marsily 1986) may incur microbial mass removal (Ryan and Gschwend 1990) when the porous media contain even small amounts of organic matter (Hornberger et al. 1992). This process can be investigated by

measuring the hydrophobicity of the microbe on porous media in which the distribution of organic matter is homogeneous (McCaulou et al. 1994).

2.3 Biologically Controlled Processes

Microbiologic processes affecting the fate and transport of microbes in the subsurface include growth and decay (in cell number and/or size or shape changes); motility and chemotaxis; biological adhesion (associated with changes in microbe surface coating changes and polysaccharide production); and predation. Growth is typically represented as a nonlinear function of limiting substrates according to the Monod (1949) kinetic equation (Wood et al. 1994). Microbial growth after changes in nutrient conditions often exhibits a lag time (Truex et al. 1992) as a result of metabolic changes undergone by the microbe. These changes are in response to the needs to reconstruct the microbe's enzymatic pathway for nutrient consumption; to repair cell damage; to remove inhibitory materials from the local environment; or to build up an extracellular pool of metabolites or enzymes necessary for growth (Bartford et al. 1982). Growth may also be associated with gas production, which immediately makes the transport problem a multiphase one. This behavior has been put to engineering use in microbially enhanced oil recovery, where microbially produced gas is used to move petroleum in tertiary recovery schemes (Jang et al. 1983). Experimental studies have found that gas production has a significant role in transport (Reynolds et al. 1989) and that it may be even more important than biomass growth in pore clogging (Soares et al. 1989). Motility is the active displacement of the microbe; independently, the resulting motion is usually assumed to be random and joined with diffusive processes. When associated with a gradient in solute nutrients, however, motile microbes will move toward higher concentrations (chemotaxis) (Dahlquist et al. 1972). The resulting flux has been quantified by Corapcioglu and Haridas (1984). Localization of substrate (e.g., isolation within high-conductivity or low-conductivity zones; Kitanidis 1994) may inhibit chemotactic behavior (Reynolds et al. 1989). A review and summary of measurement of chemotactic flux is reported by Ford et al. (1991).

Active biological adhesion can result from changes in microbe surface coat polymer changes and from polysaccharide production (Fletcher and Floodgate 1973; Allison and Sutherland 1987; Lappan and Fogler 1992). Predation (e.g., grazing by protozoa) is discussed by Harvey et al. (1989) and Harvey and Garabedian (1991). Strong discrimination in transport between native and nonnative microbe tracers has been observed and may indicate that strains are better adapted to transport in their home environments (Harvey and Garabedian 1991; Kinoshita et al. 1993).

3.0 Modeling Microbial Transport Processes in Porous Media

The basic representation of the biological phase is a matter without consensus as noted in the exchanges of Widdowson (1991), Baveye and Valocchi (1991), Baveye et al. (1992) and Jaffé and Taylor (1992). Typically biomass is represented as either a 1) continuous biofilm on the solid surfaces (Taylor and Jaffé 1990; Taylor et al. 1990) or 2) discontinuous patchy film (Vandevivere and Baveye 1992a,b; Rittmann 1993; Widdowson et al. 1988). Also, typically no assumptions are made on biomass structure (MacQuarrie et al. 1990; Sudicky et al. 1990; Zysset et al. 1994). Mathematically,

the continuous biofilm model is often associated with a diffusion limitation on the transport of solutes from aqueous phase to biomass phase where they can be degraded (Wood et al. 1994). The structured biofilm models accord to research focused on pore clogging and permeability changes and concerns about substrate degradation (Rittman 1993). The approaches involving no assumption on biomass structure treat the biomass as a dissolved, but kinetically sorbing/desorbing, species (MacQuarrie et al. 1990; Zysset et al. 1994), and this approach has been taken in recent column studies that focus on bacterial transport (Tan et al. 1994; Lindqvist et al. 1994).

Modeling subsurface microbial transport generally derives from colloid transport theory, which itself derives from the combination of convection-dispersion theory for solute transport with filtration theory (Herzig et al. 1970; Yao et al. 1971; Tien et al. 1979; O'Melia 1980), as discussed by de Marsily (1986) and McDowell-Boyer et al. (1986). Filtration-theory-based models provide only for irreversible first-order kinetic removal of particulates, where the removal mechanisms are independent of amount removed (i.e., there are no significant changes to the porous medium). In this approach, detachment is not accounted for because some early column studies indicated that attachment was not generally reversible (Wollum and Cassel 1978; Smith et al. 1985). Analytical solutions to this model have been provided by Dieulin (1982) as summarized by de Marsily (1986). The extended version of the model, which includes additional equilibrium sorption reactions, has been solved by Valocchi (1985). This model has been used by Matthess and Pekdeger (1985) and by Matthess et al. (1988).

Observations of column and field experiments quickly showed, however, that for many microbes the sorptive processes are reversible not only over column-scale transport (Scholl et al. 1990; Kinoshita et al. 1993; McCaulou et al. 1994), but also over larger time and space scales (Harvey and Garabedian 1991). This recognition has led to increased reliance on a kinetic detachment term in addition to the kinetic and equilibrium attachment terms espoused by Corapcioglu and Haridas (1984). The resulting model is the "two-site" reactive transport model, with irreversible kinetic reactions for attachment at one type of reaction site (in accordance with filtration theory), and with either equilibrium or kinetic reversible terms for adsorption and desorption at a second type of reaction site. Examples are given by Harvey and Garabedian (1991), Fontes et al. (1991), Bales et al. (1991), Lindqvist and Bengtsson (1991), Mills et al. (1991), Kinoshita et al. (1993), Hornberger et al. (1992), and McCaulou et al. (1994). Lindqvist et al. (1994) and identically Tan et al. (1994) extend this approach to incorporate attachment site saturation [termed "cell density" in Lindqvist et al. (1994), not to be confused with buoyant density] through a simple nonlinearity in the sorption term, while maintaining a linear detachment kinetic. The exact same model is recently proposed for colloid transport in Saiers et al. (1994). This recent reliance on kinetic detachment is not actually new; dual attachment/detachment mechanisms were modeled similarly in an early study on particulate "clogging/declogging" (Sakthivadivel and Irmay 1966; Herzig et al. 1970), as noted by Corapcioglu and Haridas (1984). Although the kinetic attachment terms used in many recent studies are still posed as results of filtration theory, the detachment kinetic term and equilibrium attachment term are usually taken from conventional transport modeling with surface reactions between solute and media (e.g., Ogata and Banks 1961; Ogata 1964; Cameron and Klute 1977). The resulting combined-term model has been used for some time to simulate other "two-site" reaction processes, in which one reaction is instantaneous (equilibrium) and the other is controlled by diffusion (e.g., Selim and Mansell 1976). As shown by Nkedi-Kizza et al. (1984) and Lassey (1988a,b), this model is mathematically equivalent to the two-domain (mobile-immobile region) model used to represent convective-dispersive transport through a porous medium with diffusion-limited transfer of solute to and from the immobile pore fraction ("dead-end" pores) (Coats and Smith 1964; DeSmedt and Wierenga 1979). Solutions to various

forms of the general model are given by Van Genuchten and Wierenga (1976) and Valocchi (1985). The simplification involved in lumping the many natural processes into two or three reaction terms is not yet avoidable, given limited understanding of the effects and interactions of these processes on large scales. For example, the resulting reaction model has as many degrees of freedom as it has parameters: without comparative tracer studies (by which one or more of the parameters can be fixed), modeling analysis can be nonunique (e.g., the data may be fit with multiple and different sets of parameter values), thus limiting its ability to discriminate even basic effects, such as attenuation (immobilized mass) versus retardation (slowed-down mass transport) (Jenneman et al. 1985, p. 390; Harvey et al. 1989, p. 56).

4.0 Summary of Model Representations of Microbial Transport Processes

The irreversible kinetic sorption terms represent physical filtration, with reaction rate coefficients estimated by filtration theory. This representation is the least empirical, because the reaction term comes analytically from the theory of the processes involved. Filtration theory has predicted the inverse relationship between particle/microbe size and removal rate (Harvey et al. 1989; Shonnard et al. 1994). As a quantitative approach to prediction of mass removal, however, it is of little use. This is because even in ostensibly similar homogeneous column-scale experiments, measured values can differ greatly from theoretical values (Harvey and Garabedian 1991; Bouwer and Rittman 1992) and may vary by as much as two orders of magnitude among researchers (Kinoshita et al. 1993). Chemical and other physical mechanisms of attachment, as well as all detachment mechanisms, are represented more empirically through various fitted kinetic and/or equilibrium reaction models. Chemotaxis is usually ignored [except by Corapcioglu and Haridas (1984)]. Earlier modeling emphasized attachment accounting as an equilibrium process (Gerba and Bitton 1984; Matthess et al. 1988). Matthess et al. (1988) concluded that attachment is an equilibrium process for flow velocities under 1 m/day (well within the realm of most subsurface convective fields). Subsequent observations indicate that this is not the case (Hornberger et al. 1992). Available methods for incorporating the effect of ionic strength of solution (as in Wnek et al. 1975) on the attachment/detachment process are rarely used; these methods are not robust, as pointed out by Tobiason (1989) and by Elimelich and O'Melia (1990), who found large discrepancies between theoretical and model predictions. Experimental studies, on the other hand, have been used to examine the effect of ionic strength on the effective attachment coefficient (Fontes et al. 1991; Hornberger et al. 1992), on the effective detachment coefficient (McDowell-Boyer 1992), and on both (McCaulou et al. 1994). Significant discrepancies are also usually found between the theoretically estimated values of the parameters and those fitted to obtain best model fits (Hornberger et al. 1992; McCaulou et al. 1994). The effects of heterogeneity, as reported in the few studies to date, are to make these discrepancies much worse (Harvey et al. 1993).

To the best of our knowledge, no attempts have been made to explicitly model exclusion as a microbial transport process in porous media, despite the demonstrated importance of this process in the hydrogeology literature [e.g., the textbook by de Marsily et al. (1986)]. In the relevant studies, Engfield and Bengtsson (1988) examined the effects of unaccounted exclusion on the effective value of partition coefficients for macromolecular transport, and Shonnard et al. (1994) analyzed early

breakthrough of microbes relative to phenol red in a capillary, based on Taylor tube-flow dispersion theory (Bear 1972; Taylor 1953). Shonnard et al. (1994) observed rapid breakthrough of microbes in flow cell and capillary experiments (i.e., rapid relative to breakthrough of phenol red) and offered an alternative to the exclusion hypothesis by way of (coarsely approximate) analysis via the Taylor theory of dispersion in a tube. However, the theory of exclusion is in fact based on Taylor's theory (Dodds 1982; de Marsily 1986). Anion exclusion of inert anionic solutes has been treated in the solute transport literature through application of the two-domain (mobile-immobile region) transport model (noted above for its use in representing reversible/irreversible attachment processes). In this application, used by Gvirtzman and Gorelick (1991) and in references cited therein, exclusion of anionic solutes is simulated by artificially eliminating an "excluded" porosity fraction that is unavailable to the anion. The remaining porosity is divided into mobile and immobile fractions. Gvirtzman and Gorelick (1991) set both the immobile porosity fraction and the mass exchange coefficient (governing transport to and from the immobile region) as calibrated parameters with demonstrated scale-dependent values. Thus, even if the calibrated parameters had instead been measured, as would be required for use of the model, they would likely be of little use to extend this empiricism to predictive modeling.

Microbial growth and decay (Monod 1949; Corapcioglu and Haridas 1984) are tractable through additional nonlinear (Michaelis-Menten kinetic model for growth) and linear (kinetic for decay) terms (Wood et al. 1994), but rigorous microbiological understanding of the interaction of growth with other processes is required. Growth and starvation are often associated with size or shape changes (Bakken and Olsen 1987; Cusack et al. 1992; Kjelleberg 1993), which in turn will affect physical filtration properties and possibly other properties. More importantly, alternative microbial production of polysaccharides has been shown to have a strong impact on attachment mechanisms (Lappan and Fogler 1992). However, most controlled experiments involve microorganisms in a resting state and/or short enough times of exposure to substrate that growth/decay can reasonably be ignored (Scholl et al. 1990; Hornberger et al. 1992). Chemotactic motility is usually assumed to be negligible, also as a result of the short time scales of experiments (Harvey et al. 1989), and it is partly controllable through strain selection and physiologic state.

The approach of recent microbial transport studies has been to use the models and experiments conjunctively to determine the relative dominance of processes and to measure the effects of nonmodeled processes on model parameters. Despite its limitations, the two-site modeling approach has been used with carefully controlled experiments to gain insight into the relative dominance of different processes, through examining the effects of unaccounted processes on effective parameters and through judicious use of selective tracers that allow pairwise discrimination of particular processes in a given medium. This approach has proven quite valuable in research by Hornberger et al. (1992), Harvey et al. (1993), and McCaulou et al. (1994). Thus, future modeling must rely on a knowledge of basic processes validated in controlled experiments and an awareness of model limitations.

5.0 Effects of Heterogeneities on Reactive Transport

Heterogeneities must be understood on their respective scales to accurately recognize processes that depend on spatially variable properties. The observations through which we sense these processes must account for the heterogeneities for the scale dependence of phenomena to be resolved (and thus to provide for the unraveling of the process dependencies). Because the processes governing the transport of materials in the saturated subsurface often are subject to significant natural heterogeneity, any attempt at characterizing the processes at the field scale must account for the effects of the heterogeneities. The most direct deterministic approach of full characterization is impossible because of the inaccessibility of the subsurface. The result is that simulation model components representing microbial dispersion, attachment and detachment, and exclusion that are developed and validated under laboratory conditions (with controlled single-scale heterogeneities at most) may not reflect field processes accurately. In fact, such models may not even serve for prioritizing governing processes in the field.

Figure 1 depicts a property that is spatially variable on multiple scales. In Figure 1, the horizontal axis represents the volume of porous media used to determine the property value; this axis is logarithmic. The vertical axis indicates the property value, as measured over the spherical sample at a fixed centroid. The plotted property value represents the effective value for the spherical sample volume, which becomes larger as one moves to the right on the horizontal axis. Notice how the property value undergoes wide variability when the sample scale (volume) intersects with a new heterogeneity that occurs on the same scale (e.g., clast, layer). As the sample size grows, variability attributable to that particular scale of heterogeneity is averaged out, and the property takes on a meaningful effective value. (This may never occur for media with continuously evolving heterogeneities.)

The plot in Figure 1 indicates how multiple-scale heterogeneities may affect a measured property such as porosity. Heterogeneities in physical/chemical properties may occur on scales at or below the observation scale. Generally speaking, the heterogeneities that occur on (or above) the observation scale must be characterized by field sampling. Heterogeneities at lower scales, which are unresolvable because of their inaccessibility or small scale or both, are assessed based on their effect on model components through upscaling. That is, controlled examination of the effect of the presence of such heterogeneities on particular processes is required to determine the average or upscaled effective process. Once upscaled, the small-scale process model, which was originally validated for the column scale, can represent the effective process on the observation scale. For certain processes, the upscaled model may simply be the same as the small-scale model with averaged or effective parameter values (e.g., upscaled flow in uniformly layered systems maintains Darcy's law with an effective conductivity proportional to the arithmetic average of the layer conductivities). Alternatively, an upscaled process such as dispersion, which is conventionally represented through the convection-dispersion equation with constant parameters, may require a variable parameter (e.g., preasymptotic dispersivity coefficient; Dagan 1984) or in certain cases a new form altogether (e.g., nonlocal dispersion; Cushman and Ginn 1993). When the transported solute is reactive with the porous media, the presence of physical heterogeneities can render the convection-dispersion equation approach inaccurate (Simmons 1981).

Figure 1. Multiple Scales of Heterogeneity Depicted Through the Effective Value of a Property Measured Over Increasing Sample Volume

Similar difficulties afflict the accuracy of microbial transport models in the presence of heterogeneities. Although the first-order kinetic models that are almost universally used to describe microbial deposition and entrainment processes appear to be relatively useful for transport in column experiments with homogeneous media, laboratory-determined reaction rate parameters may not be indicative of field conditions at all (Harvey et al. 1993, 1989). Physical mechanisms of bacterial transport differ from those for conservative solutes as a result of exclusion. In conventional modeling approaches, the only way to capture these effects is to empirically refit velocity retardation coefficients (in many cases to artificial values less than unity) or first-order reaction coefficients. Such parameter-fitting does not tell us whether the fitted models will work for longer times or distances or for other sites. The approach may provide simulations of the processes in a certain location on the specific scale of the observations, but even this goal may be out of reach without exhaustive observation and characterization. Harvey et al. (1993) report on an in situ multitracer experiment in which the relative order of breakthroughs of bacteria, microspheres, and salt differed for three sample locations within 1 meter of a single sampling well. In that study, significantly different retardation factors were required to explain the effects of depth within the well. The observed difference between microbial and salt breakthroughs is in accordance with previous small-scale studies in two-zone columns (Fontes et al. 1991) and in layered aquifer material (Harvey and Garabedian 1991), which suggested that "physical variability in aquifer structure increases dissimilarity between bacterial and conservative solute transport behavior in small-scale experiments" (Harvey et al. 1993). The ramifications of heterogeneities on larger scales are unknown.

The need for extensions beyond conventional kinetic models has been indicated, especially for use as empirical tools on larger scales (i.e., any scale of observation above a scale of heterogeneity). As

pointed out by Hornberger et al. (1992), "... the use of equilibrium adsorption models in conjunction with bacterial transport, such as the linear adsorption isotherm ... may not be fully adequate for describing transport of bacteria." Attention may also be given to application of such high-order (nonlinear) models at both the fine scale (high-resolution) and the larger scale but only as a preliminary investigation of the scaling of these models, an entirely different research area. There is mounting evidence that accurate nonlinear kinetics, when invoked as part of continuum-based transport models, may not upscale at all under conditions of chemical (Wise 1993) or physical (Ginn et al. 1993) heterogeneity. The implication is that for certain cases, the proper model for kinetics on the large scale has a different functional form than the conventional model. This notion has been mentioned by Hornberger et al. (1992), who states that the disagreement between observations and predicted behavior "suggests that further study is needed to better establish adequate mechanistic theory as the basis for appropriate description of bacterial transport." Similar disparities have been noted by Harvey et al. (1993).

6.0 Upscaling

Upscaling is the analysis of a process described on a given scale to determine the corresponding effective process on a larger scale. In subsurface transport, processes are modified principally as a result of heterogeneities in controlling properties. Thus, upscaling is the accounting of the influence of heterogeneities on intermediate scales, and thus links measurements on small scales to observations on large scales. Upscaling the dispersion (of a nonreactive tracer) in physically heterogeneous media has received significant attention in the literature (e.g., Dagan 1986). The upscaled dispersive process is known as "macrodispersion" and is represented either through modification of regular (small-scale) dispersion coefficients in time (Dagan 1984) or space (Bakr et al. 1978), or through introduction of memory into the spreading process (Cushman and Ginn 1993). Exploration of stochastic theories for representing dispersive mixing in the heterogeneous subsurface has brought substantial progress in characterizing macrodispersion in certain aquifer types (cf. Dagan 1989; Cushman 1990; Gelhar 1993). Continuum stochastic approaches (Cushman 1987) make use of the statistics of velocity (determined independently or through its dependence on pressure and conductivity), measured on a particular scale, to develop effective transport models that are applicable to larger scales of observation. Depending on the stringency of the statistical assumptions involved, the resulting models form various generalizations of the classical convection dispersion equation, ranging from conventional (local) models with asymptotic or time-dependent coefficients to nonlocal forms with time- and space-dependent coefficients (Cushman and Ginn 1993).

The more common picture of natural transport includes scaling interactions between the solute and the heterogeneous media (Nielsen et al. 1986). The introduction of chemical reactions into stochastic macrodispersive models has been addressed only recently. Application of scaling approaches to reactive transport has afforded limited upscaling of transport behavior for physically and chemically heterogeneous porous media that obey certain statistical constraints. Some recent applications include the extension of Dagan's method (cf. Dagan 1984; Dagan et al. 1992) by Bellin et al. (1993), Bosma

et al. (1993), Quinodoz and Valocchi (1993), Selroos and Cvetkovic (1994), and Cvetkovic and Dagan (1994); the related method of moments (cf. Kitanidis 1988; Dagan 1988; Dagan 1990), as used by Valocchi (1989), Chrysikopoulos et al. (1992), and Dagan and Cvetkovic (1993); and the first-order reliability approach (cf. Sitar et al. 1987) used by Cawfield and Wu (1993). Under these approaches, the chemical and physical heterogeneities have been represented as random fields (specifically, the constitutive model parameters have been represented as random fields with specified spatial correlation structures) and behavior has been predicted by averaging Monte Carlo simulations of the system (Bosma et al. 1993) or by deriving from this representation upscaled equations of reactive transport (Hu et al. 1995). The perturbation approach requires that nonlinearities in the reactions be of minimal importance and that the strength (or variability) of the heterogeneities is small. These approaches are based completely or in part on detailed descriptions of the statistical properties of the flow velocity field and/or the underlying physical heterogeneity. Modeling of average behavior of a kinetically reactive tracer for physically heterogeneous media is reported by Quinodoz and Valocchi (1993) and Dagan and Cvetkovic (1993), and chemically and physically heterogeneous media is reported by Bellin et al. (1993) and Bosma et al. (1993). Modeling the effects of pure chemical heterogeneity is relatively new; Wise (1993) briefly reviews relevant literature on scaling of nonlinear reactions in columns in which chemical heterogeneity occurs on lower scales. Some of these studies have indicated that the effective dispersion tensor for equivalent field-scale correction-dispersion-reaction (CDR) models depends on the presumed statistics of the reaction properties (e.g., Valocchi 1989; Bosma et al. 1993; Quinodoz and Valocchi 1993; Cvetkovic and Dagan 1994). There are no analogous results for the existence and behavior of effective CDR model properties for more general heterogeneities (e.g., when the necessary statistical assumptions fail) or for nonlinear reactions in general. Valocchi (1989) has pointed out that hidden correlations between velocities and reactive properties in field-scale reactive tracer experiments can lead to misinterpretation of effective dispersion.

In all of these studies, with the exception of the nonlocal model of Hu et al. (1995), the heterogeneities are idealized and simple: they occur on a single small scale relative to the model scale, and they are represented as second-order stationary random fields with known probability distribution and finite spatial correlation structure. Thus, these results may be limited in field application if these assumptions are violated, such as when multiple scales of heterogeneity occur, or when heterogeneity patterns are better represented by more complex probability distribution functions (pdfs) and/or correlation structures (Scheibe and Cole 1994). Perturbation approaches also require inclusion of the statistics and correlation structure of the chemical properties involved. Thus, it is crucial that sampling schemes used in upscaling reactive transport in heterogeneous media provide estimates of the statistics and correlations of the relevant flow and reaction properties. All of these approaches involve exclusively linear reactions [except for that of Wise (1993)] and assume first-order approximations that require restrictions on the magnitude of heterogeneity, as well as a priori specification of the statistical descriptors involved (including ergodicity). The nonlocal formalism of Hu et al. (1995) appears most promising from the perturbation approach and will be used here to address the effect of both chemical and physical heterogeneities on plume behavior. To test the assumptions behind the nonlocal form an alternative and independent scaling approach is required. A new approach to scaling reactive transport in physically and chemically heterogeneous media has been developed under the Intermediate-Scale Investigations of Microbial Heterogeneity in the Subsurface Science Program's Heterogeneity Subprogram. The Stochastic-Convective Reaction (SCR) perspective involves the representation of the reactive flow field as an ensemble of streamtubes, each of which acts as a convecting reactor. The convection travel time of each streamtube is viewed as a random variable. The pdf of this random variable is then automatically defined as the breakthrough curve resulting from an instantaneous

injection of solute at some point upstream. The power of the SCR in upscaling the effect of physical heterogeneities is that the hydraulics of transport are completely described by the travel-time pdf (obtained from the breakthrough curve of an inert solute). In this way, the hydraulics of transport are effectively separated from the reaction process. This approach makes full use of in situ breakthrough curves from tracer tests, unlike the conventional (CDR) approach, which utilizes only the first two moments of the breakthrough curve. A recent application to field experiments is described by Wise and Charbeneau (1994). The approach has been used in the design of experiments involving biodegradation of a substrate transported as a solute in binary inclusive heterogeneous media (Ginn et al. 1993). The SCR is shown in numerical Monte Carlo studies to upscale nonlinear reactions in binary inclusive (physically) heterogeneous media more accurately than the conventional upscaling by refitting effective reaction and transport parameters to the CDR equation, as done in Quinodoz and Valocchi (1993) and in Cvetkovic and Dagan (1994) (Ginn et al. submitted). Furthermore, the approach easily accounts for dynamic coupled changes in the reactive phase (e.g., microbial growth), which are ignored in many simulations of microbial reaction and transport [with the exception of that of Corapcioglu and Haridas (1984)].

7.0 Roles for Modeling of Bacterial Transport

Modeling analysis of laboratory and field data representing microbial transport in the natural subsurface must incorporate the catalogue of processes reviewed in this document. Process representations should be balanced as to their respective scientific maturity, accuracy in mathematical implementation, and degree of characterization. For instance, whereas in some cases microbial convection/dispersion models may be expressed with great confidence, the actual characterization of the controlling property (hydraulic conductivity) often severely limits the usefulness of any mathematical model.

Some recently published logical assessments of the role of modeling in the sciences form the proper philosophical context for this research approach. Limitations on process understanding and knowledge of subsurface properties (and their definitions) render representative models nonunique (Ginn and Cushman 1992) or, equivalently, not closed (Oreskes et al. 1994). Consequently, validation by modeling of subsurface processes is generally impossible (Ginn and Cushman 1990; Oreskes et al. 1994), as is particularly apparent in applications of modeling to microbial transport. Often model nonuniqueness (resulting from lumping of processes into simple reaction kinetics) prevents certain discrimination between general properties, such as attenuation versus retardation (e.g., Harvey and Garabedian 1991). Thus, the role of modeling is limited to conditional data analysis, including comparative testing of scaling theories, sensitivity analysis, and process hypothesis testing. As a result, experimental investigations into microbial transport can be productive only if hypotheses are carefully focused, certain experimental controls are exerted on the multiple processes involved, and data analysis/upscaling plans are integrated throughout the planning, execution, and interpretation of the experiments. In this way, the modeling component of the investigation serves to examine current process formulations and to prioritize processes for study relative to their implications for transport. This is in accordance with the recent logical assessment of modeling in earth sciences by Oreskes et al. (1994), who concluded that "models are most useful when they are used to challenge existing

formulations, rather than to validate or verify them." This approach avoids circular reliance on modeling validation and affords unencumbered investigation into process scale and property dependency through a stepwise modeling-measurement strategy.

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