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Modeling bacterial detachment during transport through porous media as a residence-time-dependent process

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Abstract. Bacterial transport through porous media was modeled using detachment functions that incorporate the dependence of detachment rate on bacterial residence time on the collector. Model parameters and the relative merit of alternative forms for the detachment function were evaluated on the basis of comparisons between model simulations and experimentally derived bacterial breakthrough and elution curves. Only detachment functions that provided an initial period in which bacteria were rapidly released, followed by slow bacterial detachment, were able to reproduce the elution portion of the breakthrough curves. In optimal simulations, 90% of the bacteria that were captured by the porous medium detached within 1 min of attachment. Experiments involving saturated flow through columns packed with sand indicated that the time to achieve complete breakthrough was inversely related to the influent bacterial concentration. On this basis and because of the relatively slow approach to breakthrough that was typically observed in transport experiments, it was hypothesized that the experimental medium contained a number of preferred attachment sites that must be essentially filled before breakthrough is achieved. Only when such (irreversible) sorption sites were included in the model formulations was it possible to produce transport simulations that matched both the breakthrough and elution portions of the empirically derived curves. It is concluded that both a time-dependent detachment function and a degree of sorption site heterogeneity are required to describe bacterial attachment and detachment during transport as observed in our laboratory.

1. Introduction

Interest in the transport of microorganisms in porous media is motivated by a variety of factors. These include public health concerns over microbes as pollutants, as in the contamination of drinking water by sewage or septic waste [*Fontes et al.*, 1991; *Corapcioglu and Haridas*, 1985]; microbes as disseminators of pollutants, as in the case of mobility enhancement of radionuclides by association of the nuclides with microbial cells [*Champ*, 1986]; the fate of genetically manipulated microbes intentionally or inadvertantly released to groundwater aquifers [*Harvey and Garabedian*, 1991; *Marlow et al.*, 1991]; and the discovery of abundant, diverse microbial communities in the deep subsurface [*Fliermans and Balkwill*, 1989].

Because bacteria have low diffusivities relative to most dissolved constituents, colloid filtration theory [*Yao et al.*, 1971; *McDowell-Boyer et al.*, 1986] provides a conceptual basis for bacterial transport modeling. The retention of colloids, including biocolloids, during transport through porous media has been successfully modeled using filtration theory to simulate the steady performance of laboratory columns [*Martin et al.*, 1991; *Kinoshita et al.*, 1993] and to model bacterial transport in field experiments [*Harvey et al.*, 1989; *Harvey and Garabedian*, 1991; *Bouwer and Rittmann*, 1992]. Despite the variety of models that have been used to describe unsteady bacterial transport through porous media, none provides an entirely successful description of even the qualitative aspects of the breakthrough-elution curve obtained from pulse-injection experiments [*McCaulou et al.*, 1995]. The generalized breakthrough-elution curve has at least five sections of interest (Figure 1a). The time or flow volume to the point at which bacteria are observed in the column effluent is a function of dispersive transport and the practical limit of detection (section A). Section B represents breakthrough; section C, the steady state plateau or slow climb; section D, the elution of aqueous-phase bacteria at the end of the injection pulse; and section E, a persistent tailing, presumably due to slow release of attached bacteria from the porous media. Most models successfully predict the height (steady state) of the breakthrough curve [*Bales et al.*, 1991; *Hornberger et al.*, 1992], although they fail to simultaneously provide the shape and timing of the breakthrough and elution portions of the overall curve. As a result, both portions of the curve have not been successfully simulated with a single set of model parameters.

The use of linear axes by some investigators [*Harvey et al.*, 1989; *Harvey and Garabedian*, 1991; *Lindqvist et al.*, 1994; *Tan et al.*, 1994] gives an incomplete picture of bacterial break-

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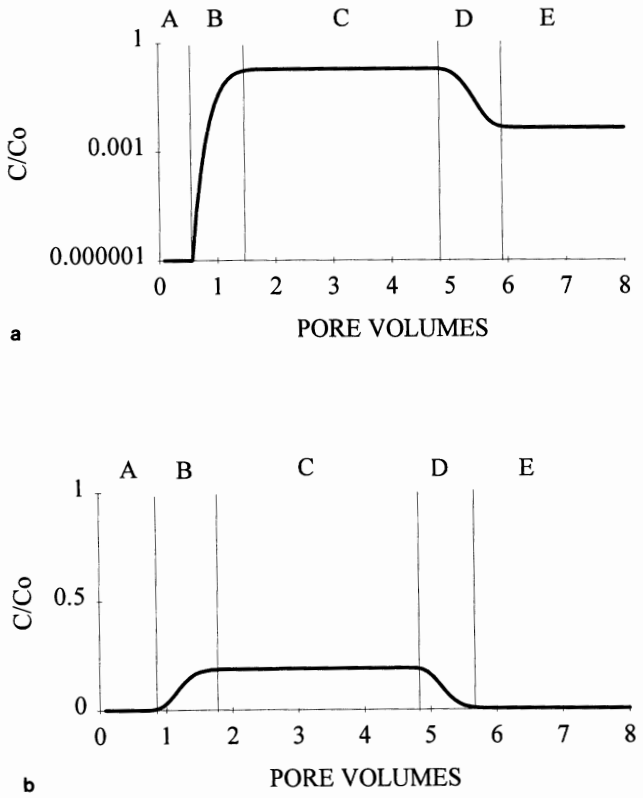

Figure 1. Illustration of sections of bacterial breakthrough-elution curve using (a) log ordinate axis and (b) linear ordinate axis. See text for explanations of sections A–E.

through and elution. When the curve shown in Figure 1a is plotted on a linear ordinate axis (Figure 1b), section A becomes wider, in effect delaying breakthrough (section B), and the elution "plateau" (section E) is simply not observed.

A property of bacterial transport that has only recently been incorporated into models is the observed inverse dependence of bacterial breakthrough time on influent concentration [*Lindqvist et al.*, 1994; *Tan et al.*, 1994]. Dependence of breakthrough time on influent concentration has been attributed to a blocking effect by attached particles and subsequently encountered particles [*Song and Elimelech*, 1993], or alternatively, to filling of a number of electrostatically attractive attachment sites that exist due to surface charge heterogeneity [*Song and Elimelech*, 1994]. A characteristic of bacterial transport that has not been previously incorporated into models is the inverse dependence of bacterial detachment on bacterial residence time on the media surface. Bacterial detachment is commonly modeled as a kinetic process being solely related to the sorbed concentration of bacteria [*Bales et al.*, 1991; *Harvey and Garabedian*, 1991; *Kinoshita et al.*, 1993; *Lindqvist et al.*, 1994; *Tan et al.*, 1994]. Experimental observations however, have demonstrated that detachment is a function of bacterial residence time and is not solely dependent upon attached concentration (*Escher* [1986], described by *Escher and Characklis* [1990]).

The purpose of this study was to investigate the relative importance of residence-time-dependent detachment and concentration-dependent breakthrough on bacterial transport. A variety of functions relating the specific desorption rate to residence time on the collector were evaluated in terms of their respective abilities to reproduce specific features that are generally observed in empirical breakthrough data. The effect of variation in the influent bacterial concentration was predicted, incorporated into the model, and tested by comparison to column transport experiments. Our goal was to use the resulting model to accurately describe bacterial breakthrough and elution.

2. Methods

2.1. Experimental Procedure

The bacterial transport models were calibrated using experimental data from *McCaulou et al.* [1995] and verified in additional experiments performed in our laboratory. Experimental conditions were identical to those of *McCaulou et al.* [1995] except as noted. Briefly, Savannah River strain A0500, derived from a depth of 180 m in the Middendorf geologic formation, was used in column studies because of its relatively low attachment efficiency and lack of aggregation. Bacteria were grown to stationary phase, centrifuged, and resuspended in nutrient-free artificial groundwater [*McCaulou et al.*, 1995] to the desired concentration. All experiments were performed in 30-cm-long glass columns at $p\mathrm{H} = 8$ and an ionic strength of 0.0028 M. Columns were packed with sediment from the Ringold Formation near Hanford, Washington, a medium sand with a weighted average grain diameter of 224 μm. Flow rate was maintained at 1.0 mL min^{-1}, producing a pore volume of 60 min. Temperature was maintained at 4°C to minimize cell growth and death during the experiment. Collected effluent was serially diluted with 0.001 M NaCl solution and plated on agar for enumeration of colonies prior to observed growth of indigenous strains (48 hours). Bacterial growth and survival was monitored in a parallel flask throughout the duration of the experiment. Loss of bacteria became measurable after 25 hours and was accounted for in subsequently detected column effluent concentrations. Elution of bacteria by artificial groundwater immediately followed the cessation of influent bacteria.

The experiments performed in this study were performed at an influent concentration of 10^8 cells mL^{-1}, an order of magnitude lower than the influent concentration used in the experiments of *McCaulou et al.* [1995]. This allowed comparison between experimental data sets to indicate the effect of influent bacterial concentration on breakthrough.

2.2. Transport

2.2.1. Governing equation. The model we develop in this paper used retention-time-dependent detachment along with limited irreversible attachment, interpreted as being due to blocking by previously attached bacteria, to simulate the bacterial breakthrough and elution curve.

To investigate the effects of retention-time-dependent desorption and blocking by attached bacteria, we examined two functions (single- and two-term exponential functions) relating bacterial detachment with residence time. Each of these two models could also be modified to include a blocking effect by attached bacteria, thus resulting in four possible variations. Before describing these models in detail it is appropriate to describe the overall model, including a brief summary of filtration theory, which is used to simulate capture of bacteria by the porous media.

The governing equation describing advection, dispersion, attachment, and detachment of bacteria during transport through sediments is

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} - k_a C + R_d \qquad (1)$$

where k_a is the adsorption rate coefficient derived from filtration theory, R_d is the rate at which bacteria desorb from the collector, C is the aqueous-phase concentration of bacteria, D is the dispersion coefficient for the bacteria, and v is the pore water velocity.

Equation (1) was solved numerically using a Crank-Nicolson finite difference method. Time and spatial steps were set to a ratio which eliminated numerical dispersion. Bacteria were removed from the fluid stream by filtration ($k_a C$) and placed in a storage array. Bacterial release R_d was modeled using a retention-time dependent desorption function described below.

For a constant step function input of bacteria C_0, the following boundary conditions apply:

$$C(x, 0) = 0 \qquad x \geq 0$$

$$C(0, t) = C_0 \qquad 0 \leq t \leq t_p$$

$$C(0, t) = 0 \qquad t \geq t_p$$

$$\frac{\partial C(x, t)}{\partial x} = 0 \qquad x > L$$

where t_p is the time of particle pulse.

2.2.2. Filtration model. The filtration coefficient is determined as shown below:

$$k_a = \tfrac{3}{2}(1 - \theta)\eta\, v(L/d_c) \qquad (2)$$

where d_c is the collector diameter, L is the column length, θ the porosity, and η the single collector efficiency [*Rajagopalan and Tien*, 1976; *Logan et al.*, 1995].

The single-collector efficiency (η) is based on hydrodynamic models, and represents the ratio of the number of particles which strike a collector to the number of particles which approach the collector. The single-collector efficiency is the sum of collector efficiencies for transport of a particle of diameter d_p, to collector of diameter d_c, by diffusion, interception and van der Waals forces, and interception and gravity, and is

$$\eta = \gamma^2[4A_s^{1/3} Pe^{-2/3} + A_s Lo^{1/8} R^{15/8} + 0.00338 A_s G^{1.2} R^{-0.4}] \qquad (3)$$

Dimensionless parameters within these terms are $Lo = H/(9\pi\mu d_p^2 U_0)$ and $A_s = 2(1 - \gamma^5)/(2 - 3\gamma + 3\gamma^5 - 2\gamma^6)$, where H is the Hamaker constant (10^{-13} ergs), $\gamma = (1 - \theta)^{1/3}$, θ is the void volume in the porous medium, U_0 is the characteristic velocity, and D is the particle diffusivity. These dimensionless collector efficiencies were developed from correlations with the Peclet number $Pe = U_0 d_c/D$, the interception number $R = d_p/d_c$, and the gravitation number $G = U_p/U_0$. The particle-settling velocity is $U_p = g(\rho_p - \rho_f)d_p^2/18\mu$, where g is the gravitational constant, ρ_p is the particle density, ρ_f is the fluid density, and μ is the fluid viscosity.

Equation (3) was derived in part from numerical simulations of particle trajectories calculated during flow through porous media and particle removal by sedimentation and interception, and by using Happel's model for removal by Brownian diffusion [*Rajagopalan and Tien*, 1976]. Under the conditions of our experiments the diffusion term dominates and the contributions of the interception and gravity terms are negligible. However, we chose to retain all three terms in order to make the model applicable to other experimental conditions. The value of k_a is 2.8655×10^{-12} s as calculated in the algorithm based upon (2) and (3), and the parameter values are given in Table 1. The dispersion coefficient for bacteria used in the simulations were determined by *McCaulou et al.* [1995] during model simulations of their experimental data.

Table 1. Values for Parameters Used in Governing Equations (1)–(3)

Parameter	Symbol	Value
Bacterial dispersion coefficient, $cm^2\ s^{-1}$	D	2.0E-5
Characteristic (superficial) velocity, $cm\ s^{-1}$	U_0	0.0033
Pore water velocity, $cm\ s^{-1}$	v	0.0083
Porosity	θ	0.40
Column length, cm	L	30.0
Collector diameter, cm	d_c	2.20E-2
Particle diameter, cm	d_p	1.26E-4
Fluid density, $g\ cm^{-3}$	ρ_f	1.00
Particle density, $g\ cm^{-3}$	ρ_p	1.03
Fluid viscosity, $cm^2\ s^{-1}$	μ	0.012

Read 2.0E-5 as 2.0×10^{-5}.

2.3. Detachment Functions

Subroutines within the Crank-Nicolson finite difference algorithm determined bacterial detachment R_d for each time step. R_d was determined from detachment functions which described the change in fraction retained, N/N_0, with time, where N_0 was the number of bacteria collected in one time step at one column segment and N was the number of that collection of bacteria remaining at times thereafter. Upon removal from the fluid stream by filtration ($k_a C$), bacteria were placed in a storage array. This array was used to determine the timing of detachment of different fractions of the collected bacteria as directed by the detachment function describing N/N_0. There were 600 cells in each storage array; the last cell represented a residence time of the bacteria on the collector of 480 min. After a 480-min residence time, bacteria were considered to be irreversibly sorbed. In simulations it was found that the model was insensitive to the time to irreversible desorption after about 100 min for all models tested because N/N_0 decreased very slowly after 100 min. The first cell in the array represented the fraction of collected bacteria ($k_a C$) that would detach in the next time step. The second and subsequent cells in the array represented detachment in the third and subsequent time steps following attachment. This process occurred at each length segment of the column and for each time step as well. Thus each time step superimposed a new bacterial detachment array upon previous arrays at each node of the column. Because the detachment function itself was constant during the simulation, the shorthand computation method for this process involved shifting the contents of each cell "forward" after each time step, so that the "foremost" cell represented bacteria to be released during the next time step, R_d. The "foremost" cell thus consisted of cumulative populations of bacteria attached during the previous time steps, of which some had a residence time of only one time step and others had longer residence times of up to 480 min. The detachment functions therefore allowed for a variety of bacterial residence times ranging from essentially zero to infinity. Additional description of the mechanics of this process is given by *Blue* [1994].

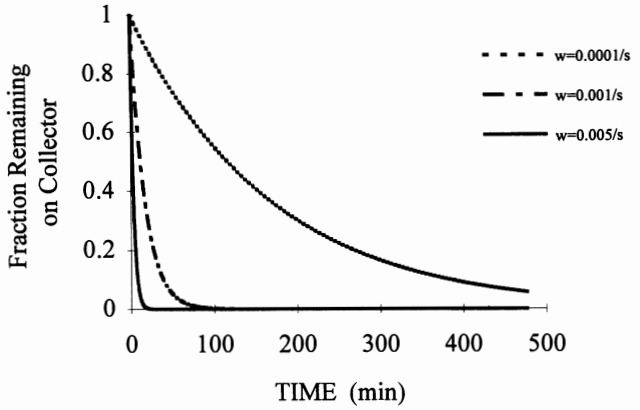

Figure 2. Effect of w on the shape of the single-term exponential desorption function.

2.3.1. Exponential model. The first approach used to describe bacterial detachment as a function of residence time [*Escher*, 1986] was made by inclusion of a single-term exponential decay function, given as

$$N/N_0 = e^{-wt} \qquad t < t_i$$
$$N = \text{const.} \qquad t \geq t_i \tag{4}$$

where N is the number of bacteria remaining on the collector at time t, N_0 is the initial number of bacteria on the collector at the time of initial adsorption, and w is an exponential desorption rate constant. N was set to a constant after a specified time, t_i, in order to provide a fraction of cells which were irreversibly attached after the specified residence time. The effect of w on the desorption rate is illustrated in Figure 2; higher values of w increased the rate of detachment.

2.3.2. Two-rate model. In order to allow greater control over the shape of the detachment curve, the second approach used was to model detachment as being governed by two different rate coefficients. This two-rate desorption model provides for a large number of bacteria to be quickly released initially, followed by a very slow release of attached bacteria. The model is given as

$$N/N_0 = Ae^{-k_1 t} + (1 - A)e^{-k_2 t} \tag{5}$$

where k_1 and k_2 are the fast and slow desorption rate coefficients and A is a weighting factor. The effects of A, k_1, and k_2 on the desorption function are shown in Figure 3. As A increases (Figure 3a), the value of N/N_0 at which slow desorption dominates becomes lower. For high values of A, the crossover of domination from the fast desorption coefficient k_1 to the slow desorption coefficient k_2, occurs at a high value of N/N_0. For lower values of A, this crossover occurs at a lower values of N/N_0. As k_2 increases, the rate of slow desorption increases (Figure 3b).

2.3.3. Two-site model. The two-site model was developed to test the hypothesis that a limited number of irreversibly sorbing attachment sites could account for the observed relation of breakthrough to influent concentration. To achieve this effect, a limited number of sites were defined as sorbing bacteria irreversibly. Irreversible attachment of a portion of a bacterial population transported through porous media has been observed by *Scholl and Harvey* [1992]. The remaining sites utilized the two-rate exponential function to describe release of bacteria. This model requires definition of several additional parameters, among them the number of bacteria that defines saturation of the irreversibly sorbing sites (N_s). This parameter is calculated from the physical attributes of the collectors using the following equations:

$$N_s = f_s f_a f_v n_c S_b \tag{6}$$

where n_c is the number of collectors in the column,

$$n_c = \frac{V_{\text{col}}(1 - \theta)}{V_c} \tag{7}$$

and S_b is the maximum number of bacteria that can accumulate on one collector, or

$$S_b = \frac{S_c}{P_b} = \frac{4\pi r_c^2}{\pi r_b^2} \tag{8}$$

where f_s is the fraction of sites that are saturable, f_a is the fraction of the collector that is available for bacterial attachment where the other fraction of the collector is spatially constrained by contact with other collectors, f_v is the fraction of bacterial coverage on the area available for attachment calculated from a packing factor with four spherical bacteria contained in a square, V_{col} is the volume of the column, V_c is the volume of the collector, S_c is the surface area of the collector, and P_b is the projected area of a bacterium.

Equation (6) represents a calculated theoretical maximum number of bacteria that attach irreversibly upon first contact.

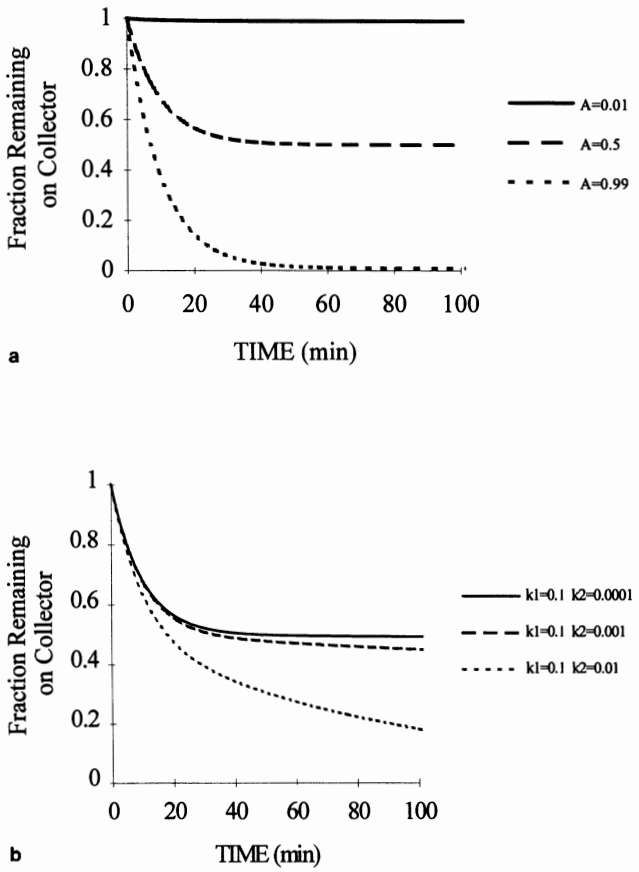

Figure 3. Effects of k_1, k_2, and A, on the two-rate exponential desorption function: (a) $k_1 = 0.1$, $k_2 = 0.001$; (b) $A = 0.5$.

Table 2. Equations and Calculated Parameters for Saturable Two-Site Model Using Experimental Parameters From *McCaulou et al.* [1995]

Equation	Calculated Value
$f_v = 4(\pi r^2)/(4r)^2 = \pi/4$	0.7854
$n_c = [V_{col}(1 - \theta)]/V_c$	8.071E12
$V_{col} = 2\pi rh$	75 cm^3
$V_c = \frac{4}{3}\pi r^3$	5.575E-12 cm^3
$S_b = S_c/P_b$	1.220E5
$S_c = 4\pi r^2$	1.521E-07 cm^2
$P_b = \pi r^2$	1.247E-12 cm^2

In the two-site model, upon removal from the fluid stream by filtration ($k_a C$), a fraction of the collected bacteria, f_s, were attached irreversibly. The remaining fraction were placed in the storage array according to the two-rate detachment function. The cumulative number of bacteria immediately irreversibly attached was monitored until N_s such bacteria were sorbed, after which time no collected bacteria were immediately irreversibly attached, and all collected bacteria were placed into two-rate storage arrays. Equations and values of parameters used in the two-site model (equations (6)–(8)) are summarized in Table 2.

The two-site model was applied to an algorithm, XTRACTR, which uses the Marquardt method of nonlinear least squares optimization [*Press et al.*, 1986] to produce the best fit parameters. During optimization the parameter k_1 was found to be redundant with respect to the parameters A and k_2. The value of k_1 was therefore set arbitrarily at 0.1, and A and k_2 were allowed to vary. Likewise, the parameter f_a, used with f_s to determine the maximum amount of immediately irreversible attachment, was directly correlated with f_s. The parameter f_a was therefore arbitrarily set at 0.1. The optimized values of A and k_2 therefore depended upon the set value of k_1, and the optimized value of f_s depended upon the set value of f_a.

3. Results

Incorporation of residence-time-dependent desorption into the advection-dispersion-filtration model was performed to test the hypothesis that both the breakthrough and elution portions of the curve would be reproduced with a single set of model parameters. Evaluation was made by comparison with the experimental results of *McCaulou et al.* [1995], which conform to the generalized shape of the bacterial breakthrough-elution curve shown in Figure 1a.

3.1. Exponential Model

The exponential model was able to reproduce the breakthrough portion of the experimental curve. However, it could not reproduce the elution portion of the curve (Figure 4). This was due to the fact that this single-term exponential function decays to zero. The fast initial release of bacteria resulted in near-zero attachment at later times and subsequent inability to reproduce the elution plateau observed in section E. Likewise, modification of w to allow higher attachment at later times was necessarily preceded by slow initial release and an inability to reproduce the initial drop of the elution portion of the curve (Figure 1, section D).

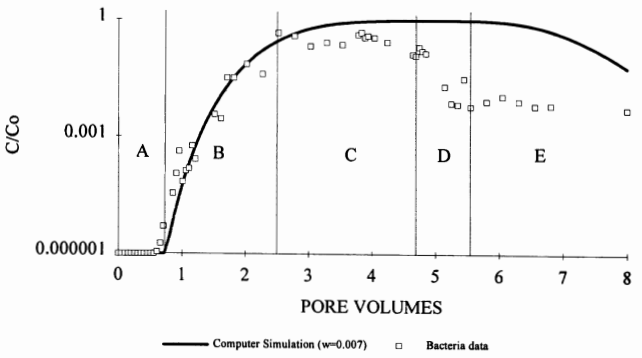

Figure 4. Best fit for *McCaulou et al.* [1995] bacteria transport data using the exponential model. Vertical lines show generalized sections of interest described in text.

3.2. Two-Rate Model

The two-rate model was able to reproduce the breakthrough portion and, using a different set of model parameters, elution/tailing portion of the experimental curve (Figures 5a and 5b). Inclusion of two-rate terms in the detachment function did not allow simultaneous simulation of the breakthrough and elution portions of the curve.

3.3. Two-Site Model

Only inclusion of a limited amount of irreversible attachment allowed model results to simultaneously match both the breakthrough and elution portions of the curve (Figure 6a). Two simulations of the data are shown in Figure 6a; one based on numerically optimized parameter values (XTRACTR) and

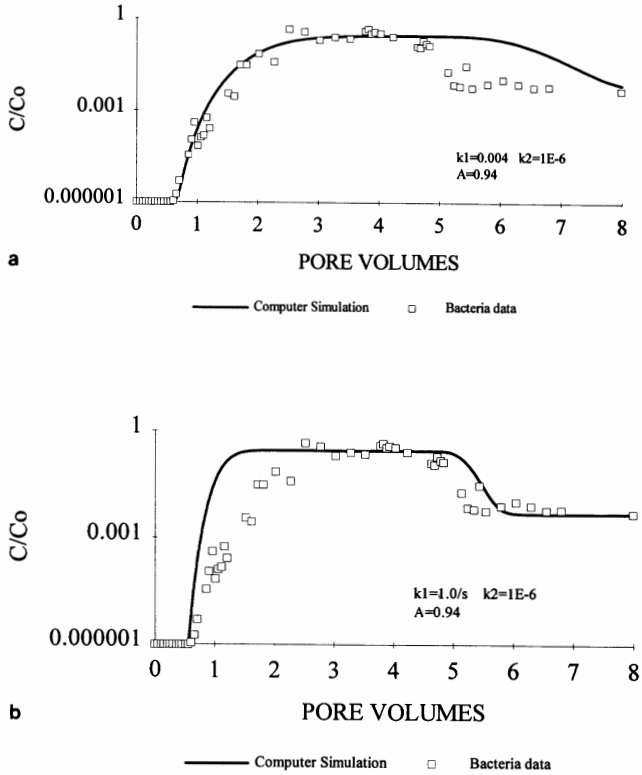

Figure 5. Best fit for (a) breakthrough portion and (b) elution portion of bacterial transport data from *McCaulou et al.* [1995] using the two-rate model.

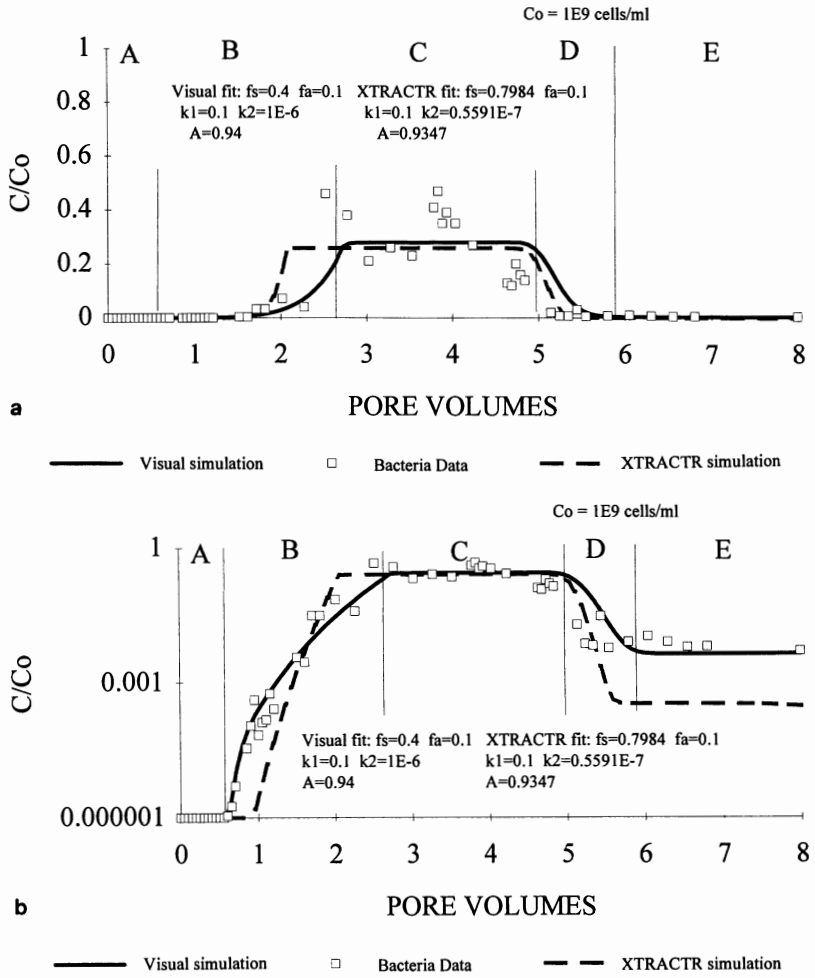

Figure 6. Numerically optimized (XTRACTR) and visually determined (visual) best fits on (a) log scale and (b) linear scale for *McCaulou et al.* [1995] bacteria transport data using the two-site model.

the other based on visually determined parameter values (visual). The numerically optimized simulation appears to be a poorer fit because of the log scale used in Figure 6a and because of the emphasis on larger numbers in numerical optimization. On a linear scale (Figure 6b), the optimized fit was only slightly better than the visually determined best fit. Optimization performed using XTRACTR for log values of the data resulted in a poor match due to loss of sensitivity in data after log conversion (results not shown). The parameters that produced the best log-scale fit were visually determined and were most appropriate for describing bacterial transport. The values of A and k_2 that best simulated the data were 0.94 and 1×10^{-6}, respectively, based on k_1 equal to 0.1. The best fit value of f_s was 0.4, based on a value for f_a of 0.1. Different values of A and k_2 would have resulted if the value of k_1 had been held at a value other than 0.1; however, the form of the detachment function would have been unchanged. Likewise, a different best fit value of f_s would have resulted from a set value of f_a other than 0.1, but the number of immediately irreversibly attached bacteria would have remained the same. Both portions of the log-scale curve were matched using a single set of parameters, three of which were obtained by fitting.

Sensitivity of the two-site model to parameters f_a and f_s, the fraction of surface area available for attachment, and the fraction of total sites that are irreversible, respectively, is shown in Figure 7. High values of f_s apportioned a higher fraction of collected bacteria to irreversible type sites, resulting in greater delay of breakthrough for all bacteria in the column (Figure 7a). In contrast, high values of f_a increased the number of irreversible sites without changing the relative fraction of collected bacteria apportioned to those sites, resulting in delayed breakthrough of the irreversibly sorbed fraction of bacteria with no effect on the two-rate fraction (Figure 7b).

3.4. Test of Two-Site Model

If the two-site model correctly describes bacterial transport, then it should predict the delaying effect of lowered influent concentrations on bacterial breakthrough. In order to develop a means of testing of the validity of the two-site model, experiments were performed to observe the breakthrough of A0500 under identical conditions for which the model was developed but at an order of magnitude lower influent concentration. Complete breakthrough of A0500 with an influent concentration of 10^8 cells mL^{-1} was delayed significantly relative to breakthrough with an influent concentration of 10^9 cells mL^{-1} (compare Figures 6b and 8). This was consistent with our hypothesis that a limited number of irreversibly sorbing sites required filling prior to breakthrough, since a significant delay in breakthrough accompanied a factor of 10 decrease in influ-

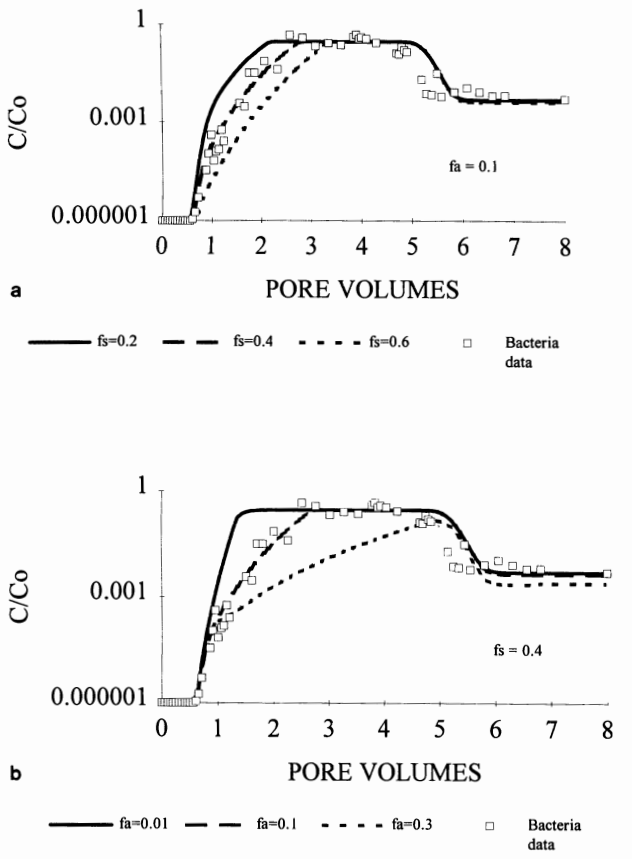

Figure 7. Sensitivity of saturable two-site model to f_s (fraction of irreversible sites), and f_a (fraction site surface available for attachment): (a) $f_s = 0.1$, (b) $f_a = 0.4$.

ent concentration. In order to reach steady state, bacterial injection required ~15 pore volumes at 10^8 cells mL^{-1}, as opposed to only 3 pore volumes at 10^9 cells mL^{-1}.

The prediction of the two-site model was compared against our own experimental data at an influent concentration of 10^8 cells mL^{-1} (an order of magnitude below that of *McCaulou et al.* [1995]) in order to examine whether this model could predict the timing of breakthrough at the lower influent concentration. The same parameters used in the previously discussed best fit of the two-site model to the data of *McCaulou et al.* [1995] were used in this test. The model predicted an increased delay of breakthrough of the order of that observed in the experiments (Figure 8). Although the earliest breakthrough of A0500 was overestimated by the model, the time to achieve steady state breakthrough was adequately described by the model.

4. Discussion

The three models examined here utilized time-dependent desorption functions to reflect the inverse dependence of bacterial desorption rate on residence time. However, only models that allowed for quick initial release followed by very slow release were successful in reproducing the elution portion of the experimental results. The possibility that the experimental curve could be described adequately by residence-time-dependent detachment or, more specifically, by the lag between attachment and detachment (*Escher* [1986], described by *Escher and Characklis* [1990]), has been considered only briefly by other researchers [*Lindqvist et al.*, 1994]. Our results show that time-dependent desorption alone cannot account for both the delayed bacterial breakthrough and the extended tailing observed in our experiments. While time-dependent detachment is responsible for the extended bacterial tailing, a saturation mechanism (i.e., limited immediately irreversible attachment) is responsible for delayed bacterial breakthrough. Without both time-dependent desorption and a saturation mechanism for immediately irreversible sorption, the complete range of breakthrough data (over orders of magnitude) could not be matched.

The form of the detachment function that best simulated the data indicated that 90% of an attached population of bacteria undergo detachment within 1 min of attachment (Figures 9a and 9b). Similar timescales for bacterial detachment have been observed by *Lawrence et al.* [1987]. Visual observations showed that *Pseudomonas fluorescens* cells initially attached to a surface reversibly in a loose manner and, if they had not detached

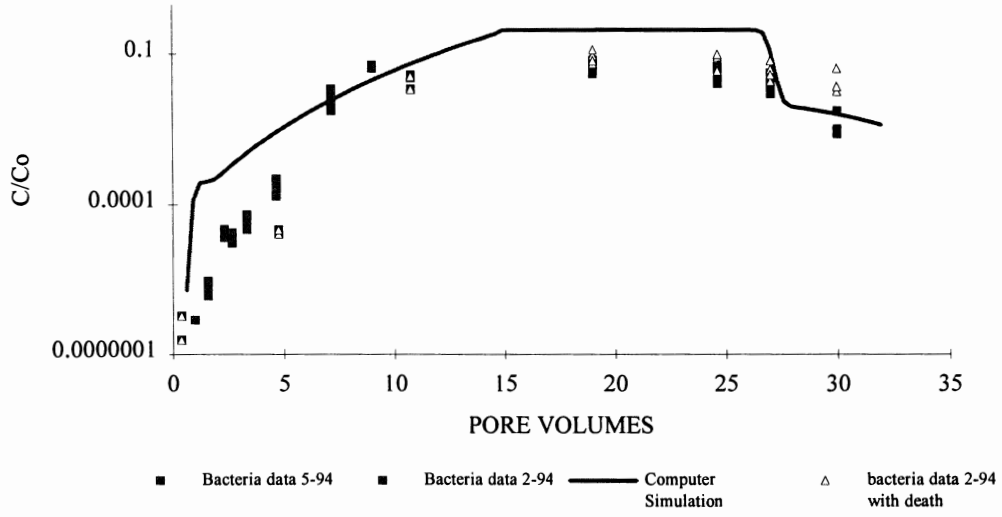

Figure 8. Comparison of model simulation to our own experimental results for an influent concentration of 10^8 cells mL^{-1}, 1 order of magnitude below that of *McCaulou et al.* [1995].

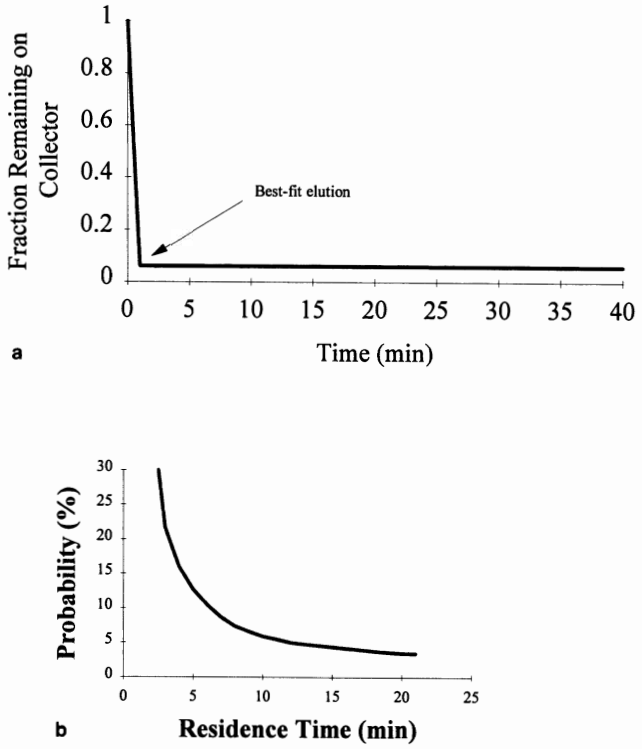

Figure 9. (a) Best fit two-rate desorption function showing that 90% of attached bacteria are detached within 2 min. (b) Probability of bacteria detaching from a surface versus retention time [*Escher*, 1986], shown for comparison.

after 1–2 min, assumed a longitudinal position and attached irreversibly. The necessity of a finite period for reversible attachment is also consistent with the hypothesis that time required for the production of exocellular polysaccharides by bacteria is necessary for irreversible attachment [*Vandevivere and Baveye*, 1992; *Gannon et al.*, 1991; *MacLeod et al.*, 1988].

The use of a packing factor f_v of 0.78 in (6) to describe the maximum number of bacteria attached to the grain surfaces is merely representative of a surface saturation effect and is not meant to stipulate an actual fractional surface coverage by bacteria. Statistical jamming limits of spheres onto a surface in two dimensions have been investigated and found to be about 55% [*Hinrichsen et al.*, 1986]. However, this jamming limit, estimated assuming random interactions without reference to possible attraction or repulsion, may not be directly applicable to our system. In our experiments the overall interaction between bacterial and collector surfaces was electrostatically repulsive; however, the complexity of sediment and bacterial surfaces results in charge heterogeneities that may cause local favorable interactions between bacteria and grain surfaces [*Song and Elimelech*, 1994]. Bacteria may also interact with each other and so may not attach strictly in a monolayer fashion. Resolution of these heterogeneities and the resulting grain surface coverage is beyond the scope of our paper. Instead, we have chosen to enforce a limitation on the amount of immediate irreversible attachment by using simple physical representations. A similar limiting mechanism was introduced in the recent articles by *Lindqvist et al.* [1994] and *Tan et al.* [1994] in which a saturation expression was used to limit the number of sorption sites. In the first article a theoretical maximum number of sorbed bacteria was estimated by dividing total collector surface area by the projected surface area of a bacterium, resulting in a 100% theoretical surface coverage. This theoretical maximum was certainly in violation of the statistically determined jamming limit but is useful as a number to compare to model-estimated counts of sorbed bacteria.

The value of parameter f_s that best simulated the data was 0.4. Thus 40% of the possible number of attachment sites (based on a monolayer coverage, a packing factor of 0.78, and a surface availability factor of 0.1 for grain-grain boundaries) were sites that attached bacteria irreversibly upon contact. These sites were filled more quickly at the higher influent concentration (10^9 cells mL^{-1}) than at the lower influent concentration (10^8 cells mL^{-1}). The remainder of the sites allowed for detachment, up to a 480-min residence time.

Because 90% of an attached population of bacteria detached within a minute of attachment, the great majority of bacteria were held or, more accurately, delayed in the column similarly to pinballs, bouncing from one site to the next. Each time a bacterium struck a site, it initially became attached. However, all attached bacteria were released according to the detachment function, except for those that were immediately irreversibly attached. Therefore attached bacteria experienced residence times ranging from zero to infinity, with individual attachment efficiencies α ranging from zero to unity. The heterogeneity with respect to α may be attributed to heterogeneity in either the bacteria or the porous media surfaces. Heterogeneity in α has been ascribed to differences in bacterial surfaces even for a monoclonal population [*Albinger et al.*, 1994].

An apparent overall value of the attachment efficiency can be estimated from the value of C/C_0 in the steady state ("plateau") portion of the bacterial breakthrough curve, using the following equation [*Yao et al.*, 1971]:

$$C/C_0 = \exp\left[-\frac{3}{2}\frac{(1-\theta)}{d_c}\alpha\eta L\right] \tag{9}$$

Equation (9) is derived from equation (1) assuming steady state conditions. The apparent value of α was estimated to be 0.065 [*McCaulou et al.*, 1995]. This value is relatively low compared with values of α for "sticky" strains of bacteria, which can approach unity [*Gross and Logan*, 1995], although it is well within the range of α values measured for other bacteria-soil combinations studied in our laboratory (unpublished data).

The two-site model overpredicted the first pore volume breakthrough of bacteria in the low-influent-concentration experiment. Several factors, such as the value of the dispersion coefficient or the presence of numerical dispersion, could have caused this overprediction to occur. Computer simulations showed that this overprediction was not affected by changes in the dispersion coefficient, nor was it due to numerical dispersion. An alternative explanation is that the number of collisions were underestimated by the filtration theory component of the model. This could be produced by not accurately describing the effect of grain size heterogeneity in the sediment, since an average grain diameter is used in calculating the collision efficiency. However, model simulations showed that the overprediction was not remedied by reasonable changes in the average grain diameter. Lowering the fast desorption parameter k_1 while holding A and k_2 constant did eliminate the overprediction of breakthrough, indicating that the best fit detachment function insufficiently described the earliest breakthrough portion of the low-concentration experiment. Further work will be necessary to increase the ability of the desorption functions to better describe all experiments.

The results of this experimental and modeling investigation provide an alternative method for quantifying bacterial transport through porous media. Modeling detachment of sorbed bacteria as a function of residence time on the sediment describes bacterial transport as adequately as do existing models that relate detachment solely to attached bacterial concentrations. Bacterial breakthrough was shown to depend on saturation of a number of irreversible sites which control the rate of approach to steady state breakthrough. The practical implication of this phenomenon is that a pulse injection of bacteria at a field site will result in a breakthrough concentration that is a function of the influent concentration and the number of irreversibly sorbing sites in the porous media.

The usefulness of present bacterial transport models is presently limited by the inability to establish the values of coefficients for attachment and detachment for a particular bacteria-media system a priori. In our own model, further work is required to determine to what extent model parameters (A, k_2, and f_s) can be applied to natural sediments and soils. Different sediments and soils tend to display different surface charges and hydrophobicities due to differences in mineralogies and organic matter content [*Murphy et al.*, 1990, 1994]. Because of the variations in bacterial retention for different sediments and soils, the shape of the detachment functions, as well as the value of f_s, may differ between sediments. Likewise, different bacterial strains display different hydrophobicities and surface charges under a given solution condition [*van Loosdrecht et al.*, 1989, 1990]. The nature of the bacterial surfaces would also exert an effect on the parameter values. Solution conditions (i.e., ionic strength, pH, and the presence of chemical entities that modify the cell surfaces) also effect bacterial retention on a particular soil or sediment [*Gross and Logan*, 1995]. Thus the values of model parameters would be affected by solution chemistry as well as bacterial strain and sediment-soil characteristics. Despite these variations, we suspect that the tendency of bacteria to detach quickly or not at all is prevalent and that surface saturation mechanisms apply to many porous medium surfaces.

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