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A Method for Calculating Bacterial Deposition Coefficients Using the Fraction of Bacteria Recovered from Laboratory Columns

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To determine if a practical relationship exists between the easily determined fractional recovery of bacteria and the deposition coefficient, useful for predictive modeling, an analytical solution to a nondimensional transport equation incorporating bacterial deposition was integrated. A nonlinear relationship between the dimensionless deposition coefficient (κ_1) and fraction of bacteria recovered (fr) was observed, and this relationship was shown to be affected by dispersion. This theoretical relationship was tested for its utility using observed data from a suite of column experiments. The predicted relationship between κ_1 and fr was reflected quite well by the experimental data. The results indicate that a simple measure—fractional recovery of bacteria—can be used to obtain an accurate value for the dimensionless deposition coefficient.

Introduction

The ability to quantify bacterial transport is of importance in eliminating drinking water contamination by microbial pathogens (*1*), in preventing the facilitated transport of organic compounds (*2*) and radioactive pollutants by bacteria (*3*), and in determining the feasibility of using microorganisms with novel degrading abilities for in situ biorestoration of organically contaminated aquifers. The ability to obtain accurate deposition coefficients (where deposition represents the combined processes that result in removal of bacteria from suspension in porous media, e.g., straining, attachment, sorption, etc.) is essential for adequately predicting bacterial transport (*4*). Typically, deposition coefficients are estimated by fitting transport models [e.g., CFITIM (*5*), CXTFIT (*6*), CXTFIT2 (*7*)] to measured breakthrough curves (BTCs) of bacteria eluted from either packed columns or cores of intact aquifer material. Alternatively, sticking efficiencies have been calculated using the filtration model of Yao et al. (*8*) from the fraction of bacteria retained in the MARK assay (*9*). This calculation requires the estimation of porosity, size and density of bacteria, temperature, and column length and neglects the effects of dispersion. Measurement of complete BTCs or the dissection of a column and enumeration of the deposited bacteria is relatively time-consuming and is not a practical measurement to consider on large numbers of cores in an effort to characterize the spatial variability in bacterial deposition for a given aquifer. On the other hand, experiments that measure only the total recovery of bacteria eluted from a column over the course of an experiment are much easier to complete. Indeed, the fraction of bacteria recovered or retained, as measured from column experiments, has been used as an index to infer the factors affecting bacterial transport through porous media (*10*−*17*).

In the work reported here, we set out to determine if a relationship between the easily determined fractional recovery of bacteria and the deposition coefficient, useful for predictive modeling, could be found. Bacterial transport was described using a one-dimensional advection−dispersion equation that incorporates bacterial deposition. Calculations using an analytical solution to the nondimensional form of the transport equation showed that the fraction of bacteria recovered was a function of both the deposition coefficient and the Peclet number. The theoretical relationship was tested for its utility using observed data from a suite of experiments. The results indicate that a simple measure—fractional recovery of bacteria—can be used to obtain an accurate value for the dimensionless deposition coefficient.

Model Development

Assuming low surface coverage, the one-dimensional transport of bacteria can be described by (*4*, *18*−*20*)

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} - k_c c + R_d \tag{1}$$

$$\frac{\partial s}{\partial t} = k_c c - R_d \tag{2}$$

where c is the concentration of bacteria suspended in aqueous solution (cells mL^{-1}), s is the concentration of bacteria associated with the solid-phase expressed on a per volume of pore water basis (cells mL^{-1}), x is the distance from the inlet (L), t is elapsed time from the initial input of bacteria (T), D is the coefficient of hydrodynamic dispersion ($L^2\,T^{-1}$), v is the interstitial pore water velocity ($L\,T^{-1}$), k_c is the deposition coefficient (T^{-1}), and R_d is the rate at which bacteria are removed from the collector and has been defined in different ways (*4*, *19*). The deposition coefficient can be calculated based on colloid filtration theory by (*21*)

$$k_c = \frac{3(1-\theta)}{2}\frac{v}{d_c}\eta\alpha \tag{3}$$

where η is the single-collector efficiency that accounts for the physical effects on bacterial deposition (*22*, *23*), α is the sticking efficiency that accounts for the chemical effects on bacterial deposition, v is interstitial pore water velocity, d_c is the diameter of the collector (sand grain), and θ is porosity.

The dimensionless forms of eqs 1 and 2 are

$$\frac{\partial C}{\partial T} = \frac{1}{Pe}\frac{\partial^2 C}{\partial X^2} - \frac{\partial C}{\partial X} - \kappa_1 C + \kappa_2 \tag{4}$$

$$\frac{\partial S}{\partial T} = \kappa_1 C + \kappa_2 \tag{5}$$

where $Pe = vL/D$, $X = x/L$, $T = vt/L$, $\kappa_1 = k_c L/v$, $\kappa_2 = R_d L/v$,

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$C = c/c_0$, $S = s/c_0$, L is the length of the column, C is the normalized concentration of bacteria in the aqueous phase, S is the normalized concentration of bacteria on the solid phase, T is dimensionless time (pore volumes), X is dimensionless length, and κ_1 and κ_2 are Damkohler numbers representing time-scale ratios for deposition and entrainment to flow velocity, respectively.

Neglecting entrainment ($\kappa_2 = 0$); a solution to eq 4 for the normalized bacterial flux concentration, $C(X,T)$, at the end of a column ($X = 1$) for a step input can be obtained by modifying the solution of Parlange et al. (*24*):

$$C(1,T) = \frac{1}{2}\exp\left[\frac{Pe}{2}(1 - \sqrt{1 + 4\kappa_1 Pe^{-1}})\right] erfc\left[\frac{1 - T\sqrt{1 + 4\kappa_1 Pe^{-1}}}{\sqrt{4TPe^{-1}}}\right] + \frac{1}{2}\exp\left[\frac{Pe}{2}(1 + \sqrt{1 + 4\kappa_1 Pe^{-1}})\right] erfc\left[\frac{1 + T\sqrt{1 + 4\kappa_1 Pe^{-1}}}{\sqrt{4TPe^{-1}}}\right] \quad (6)$$

Because the above equation is linear with respect to C, the principle of superposition can be used for generating BTCs for pulse lengths of finite duration by (*25*)

$$C_\epsilon(X,T) = C(X,T) - C(X,T - \epsilon) \quad (7)$$

where $C_\epsilon(X,T)$ is the solution for a pulse input and ϵ is the length of the pulse input.

The cumulative recovery of bacteria is obtained by integrating eq 6 over time. As T approaches infinity

$$\int_0^\infty \text{erfc}\left[\frac{1 - T\sqrt{1 + 4\kappa_1 Pe^{-1}}}{\sqrt{4TPe^{-1}}}\right] dt \rightarrow 2T$$

and

$$\int_0^\infty \text{erfc}\left[\frac{1 + T\sqrt{1 + 4\kappa_1 Pe^{-1}}}{\sqrt{4TPe^{-1}}}\right] dt \rightarrow 0 \quad (8)$$

and the cumulative (normalized) mass recovery becomes

$$\exp\left[\frac{Pe}{2}(1 - \sqrt{1 + 4\kappa_1 Pe^{-1}})\right] T \quad (9)$$

For a pulse input of length ϵ, the mass recovery is given by eq 9 with T replaced with ϵ (by the principle of superposition). For an input concentration of $C/C_0 = 1$, the total normalized mass input becomes ϵ, so the fractional recovery is

$$\text{fr} = \exp\left[\frac{Pe}{2}(1 - \sqrt{1 + 4\kappa_1 Pe^{-1}})\right] \quad (10)$$

Rearranging eq 10 to solve for κ_1 yields

$$\kappa_1 = -\ln(\text{fr}) + \left[\frac{[\ln(\text{fr})]^2}{Pe}\right] \quad (11)$$

Equation 11 is independent of pulse length and incorporates the effects of dispersion in the calculation of the dimensionless deposition coefficient (κ_1).

Results and Discussion

The effects of entrainment were neglected in this study since rates of entrainment have been observed to be several orders of magnitude less than deposition rates (*4, 26*) and therefore will not greatly affect the fraction of bacteria recovered over an experiment that lasts only several (~1–10) pore volumes. Calculations for various values of the Peclet number (*Pe*) and dimensionless deposition coefficient (κ_1) using eq 10 showed that the recovery of bacteria was dependent on both *Pe* and κ_1 (Figure 1). Equation 11 (or Figure 1) shows that for relatively high fractional recoveries, the effect of dispersion on κ_1 is negligible (for Peclet numbers > 10) and therefore $\kappa_1 \approx -\ln(\text{fr})$; at steady-state, this relationship can be derived from the filtration model of Yao et al. (*8*). However, for low values of fractional recovery, dispersion is important; deposition is linearly dependent upon the pore fluid concentration of bacteria, and a decrease in the Peclet number represents greater mixing in the pores with a resulting decrease in the aqueous concentration of bacteria. Thus, for low fractional recoveries, sticking efficiencies obtained by rearrangement of the dimensionless form of eq 3 may be underestimated when calculated from measured steady-state values of C (c/c_0) using the filtration model of Yao et al. (*8*) where $\kappa_1 = -\ln(c/c_0)$ (Figure 2). The discrepancy between eq 11 and the Yao et al. (*8*) model is likely to be important in heterogeneous porous media where low bacterial recoveries and/or low Peclet numbers are likely.

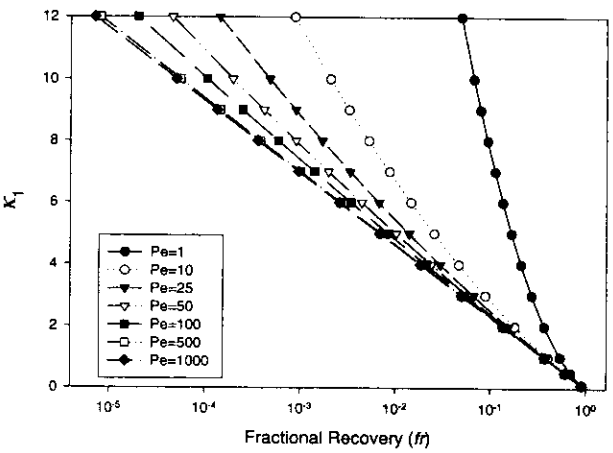

FIGURE 1. Effect of the Peclet number (*Pe*) on the relationship between the dimensionless deposition coefficient (κ_1) and the fraction of bacteria recovered (fr) as determined from calculations using eq 10. The effects of dispersion become important for low fractional recoveries (fr < 0.1).

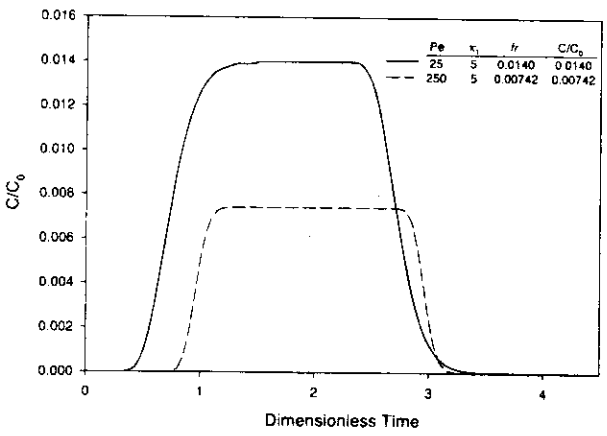

FIGURE 2. BTCs for Peclet numbers of 25 and 250 generated from eq 7 for $\kappa_1 = 5$. Calculating κ_1 from the fractional recovery (fr) using eq 11 yields a value of 5. Calculating κ_1 from the Yao (*8*) filtration model where $\kappa_1 = -\ln(c/c_0)$ yields a value of 4.27 and 4.9 for Peclet numbers of 25 and 250, respectively. The discrepancy between these two methods will yield different values for the sticking efficiency (α) where

$$\alpha = \frac{2d_c}{3(1 - \theta)} \frac{\kappa_1}{L} \frac{1}{\eta}$$

TABLE 1. Experimental Conditions for Column Experiments Depicted in Figure 3

	pulse length (*T*)	Peclet no.[a]	flow velocity, *m* (day^{-1})	ionic strength (m)	sand type (size in mm)	bacterial isolate[d]
Fontes et al. (*13*)	0.1	63–1016	3.4	0.00089–0.0089	clean quartz sand[b] 0.33–0.4	W6, W8
Morley et al. (*31*)	0.05	101–325	3.4	0.00089	clean quartz sand[b] 1.0–1.19 and 0.5–0.6	W8, S138
Winquist (*27*)	1.0	91–465	1.9–2.6	0.0036	aquifer sediments[c] 0.41–0.7	PL2W31, E2W3, E2W1, E1B5, E3T3
Bolster (unpublished data)	1.0	100	1.0	0.0036	aquifer sediments[c] 0.2–1.0	PL2W31
McCabe[e] (unpublished data)	1.0	100–150	3.4	0.0036	aquifer sediments[c] 0.2–1.0	PL2W31
Knapp et al. (*10*)	0.05	122–169	3.4	0.00089	Fe-coated quartz sand 0.5–0.6	W8

[a] Obtained by fitting CXTFIT2 to observed breakthrough curves of a conservative tracer. [b] Sand was acid washed to remove impurities. [c] Sediments contained iron, aluminum, and manganese oxyhydroxide coatings. Sorption isotherms have shown greater bacterial affinity to sediments with higher concentrations of these coatings (*32*). [d] Organisms W6, W8, S138, PL2W31, E2W3, and E2W1 are hydrophilic while E1B5 and E3T3 are hydrophobic, according to the scheme of Mozes et al. (*33*). [e] These experiments included live, dead, and DAPI-stained bacteria.

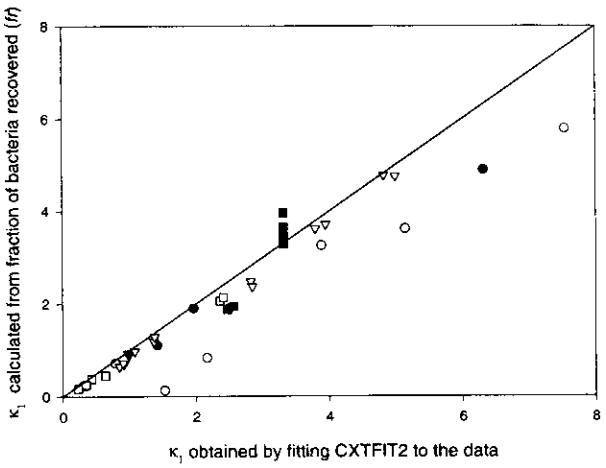

FIGURE 3. Relationship between calculated values (eq 11) and fitted values to breakthrough data (CXTFIT2) of the dimensionless deposition coefficient for a number of experiments in laboratory columns with a variety of organisms under a variety of conditions. The data represented are those from experiments summarized in Table 1. The line represents a 1:1 correspondence. The data presented are those from experiments summarized in Table 1: (○) Fontes et al. (*13*), (□) Morley et al. (*31*), (●) Winquist (*27*), (▼) Bolster (unpublished), (▽) McCabe (unpublished), and (■) Knapp et al. (*10*).

We tested the adequacy of the modeled relationship between κ_1 and fr using results for a suite of experiments that differed in a variety of experimental conditions (Table 1). In each study, κ_1 was obtained from both the fraction of bacteria recovered as determined by numerically integrating the measured BTC and then dividing the amount of bacteria recovered by the number of bacteria introduced into the column and from fitting eq 4 to the observed bacterial BTCs using CXTFIT2 (7). The two independently derived estimates of κ_1 could then be compared. Although the experiments differed in the organism used, the type of sand, the presence or absence of metal oxyhydroxide coatings on the sand, the Peclet number, flow velocity, pulse length, ionic strength of the carrier fluid, and entrainment rates (Table 1), the predicted relationship between κ_1 and fr was reflected reasonably well by the observed data from the various experiments (Figure 3). The values of κ_1 fitted to the data for Fontes et al. (*13*) show the greatest divergence from the calculated values of κ_1 obtained from eq 11 and may be due to the experimental procedures used in these experiments. In contrast to the other experiments used in this analysis where effluent samples were collected every 0.1 pore volume, Fontes et al. (*13*) collected their samples at every 0.25 pore volume. The loss of resolution due to sampling every 0.25 pore volume would not affect the calculation of the fraction of bacteria recovered but may underestimate the peak effluent concentrations that would in turn increase the fitted value of κ_1 in comparison to the value of κ_1 calculated based on the fraction of bacteria recovered. Indeed, the fitted values for κ_1 are greater than the calculated values for κ_1 using eq 11 for the data from Fontes et al. (*13*). For extremely low recovery of bacteria, entrainment may contribute to the fraction of bacteria recovered and therefore will result in under predictions of κ_1 and may explain the position of the data point representing the lowest recovery in the data of Winquist (*27*) (Figure 3).

We present a method to obtain bacterial deposition coefficients from column experiments by measuring only the total influent and effluent number of bacteria. The finding that eq 11 is independent of pulse length allows this method to be used with short pulse lengths, thereby reducing the likelihood that the clean bed assumption inherent in colloid filtration theory will be violated. Because of counting errors in the measurement of influent and effluent bacterial concentrations, Gross et al. (*9*) (using glass bead experiments) argued that the fraction retained is a more accurate way to measure bacterial deposition than the fraction recovered when bacterial recovery is high. However, the fraction of bacteria recovered from columns packed with sand (*13*) or aquifer sediments (*27*) is often low as compared with the fraction recovered from columns packed with glass beads (*9*, *28*). The approach we have suggested should provide an accurate representation of the deposition coefficient in environmentally realistic situations. Furthermore, this method greatly reduces the effort needed to obtain a deposition coefficient and allows the calculation of a deposition coefficient from experiments that are not suitable to analysis of breakthrough curves. It should be possible to develop a quick assay technique to estimate values for κ_1 using small core samples if values for the Peclet number can be either obtained directly from BTCs of a conservative tracer or estimated based on characteristics of the porous media (*29*). Mills and Hornberger (*30*) developed a method to describe the spatial variability in hydraulic conductivity by conducting falling head tests on syringe-sized samples of aquifer sediments. Because aquifer sediments are inherently heterogeneous, the potential ability to obtain bacterial deposition parameters on similar sized samples, comparable to the MARK assay of Gross et al. (*9*), may lead to better characterization of the spatial variability of bacterial deposition found at a given site. For example, Bolster et al. (unpublished data) observed variations of up to 2 orders of magnitude in concentrations of deposited bacteria on the

scale of only a few centimeters within intact aquifer sediments. The distribution of deposition parameters obtained from such an assay could then be used in stochastic or Monte Carlo type models to increase our predictive capabilities of bacterial transport in the subsurface.

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