



Significance of straining in colloid deposition: Evidence and implications

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[1] Considerable research suggests that colloid deposition in porous media is frequently not consistent with filtration theory predictions under unfavorable attachment conditions. Filtration theory was developed from an analysis of colloid attachment to the solid-water interface of a single spherical grain collector and therefore does not include the potential influence of pore structure, grain-grain junctions, and surface roughness on straining deposition. This work highlights recent experimental evidence that indicates that straining can play an important role in colloid deposition under unfavorable attachment conditions and may explain many of the reported limitations of filtration theory. This conclusion is based upon pore size distribution information, size exclusion, time- and concentration-dependent deposition behavior, colloid size distribution information, hyperexponential deposition profiles, the dependence of deposition on colloid and porous medium size, batch release rates, micromodel observations, and deposition at textural interfaces. The implications of straining in unsaturated and heterogeneous systems are also discussed, as well as the potential influence of system solution chemistry and hydrodynamics. The inability of attachment theory predictions to describe experimental colloid transport data under unfavorable conditions is demonstrated. Specific tests to identify the occurrence and/or absence of straining and attachment are proposed.

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1. Introduction

[2] Accurate knowledge of the processes that control the transport and deposition of colloids in subsurface environments is needed to protect water resources from a wide variety of contaminants. For example, pathogenic microorganisms pose a serious threat to water quality [Craun and Calderon, 1996; U.S. Environmental Protection Agency, 2000] in many regions of the world due to their low infectious dose [Loge *et al.*, 2002]. Many treatment techniques for surface water or effluent from sewage treatment plants rely on soil passage to remove pathogens (e.g., riverbank filtration, infiltration basins and trenches, and sand filters). An understanding of microbe transport is also essential for engineered bioaugmentation and bioremediation strategies [Wilson *et al.*, 1986; Rockhold *et al.*, 2004] and microbially enhanced oil recovery systems [MacLeod *et al.*, 1988]. Furthermore, the environmental fate of many organic and inorganic contaminants can be influenced by colloid-facilitated transport due to partitioning onto high surface area colloids (i.e., clays and dissolved organic

matter) that are mobile [Kretzschmar *et al.*, 1999; Ouyang *et al.*, 1996; de Jonge *et al.*, 2004].

[3] Clean bed filtration theory (CFT) has been developed and is now often used to characterize colloid attachment in porous media [e.g., Logan *et al.*, 1995; Tufenkji and Elimelech, 2004a]. This theory invokes a first-order attachment term to describe a spatially constant rate of deposition, which leads to an exponential spatial distribution of retained colloids in porous media. In the absence of an energy barrier to deposition (favorable attachment conditions), attachment is undoubtedly an important mechanism of colloid deposition and profiles are well-described by filtration theory [e.g., Li and Johnson, 2005]. In this case, filtration theory provides a very useful tool for predicting attachment behavior over a wide range of conditions.

[4] Many environmentally relevant colloids (microorganisms and clays) and porous media (quartz) possess a net negative charge at a neutral pH, and groundwater and surface water resources typically have a low ionic strength. Under these conditions an energy barrier to attachment may occur (unfavorable conditions), and colloids that collide with collector surfaces (soil grains) are not expected to attach to the solid-water interface. In practice, however, a finite number of colloids are retained in porous media even under unfavorable attachment conditions. In contrast to CFT predictions, colloid deposition rates have also frequently been found to vary spatially with depth under unfavorable attachment conditions [Camesano and Logan, 1998; Bolster *et al.*, 1999; Redman *et al.*, 2001; Bradford *et al.*, 2002; Tufenkji *et al.*, 2003; Li *et al.*, 2004; Tufenkji and Elimelech, 2005a,

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2005b]. Deviations between observed and CFT predicted deposition profiles have been reported to increase for larger colloids and relatively fine textured sand [Bradford *et al.*, 2002; Tufenkji and Elimelech, 2005a]. Bradford *et al.* [2003] reported that CFT predictions did not adequately describe measured deposition profiles when the ratio of the colloid diameter to the median grain diameter was greater than around 0.005. This ratio corresponds to bacterial and protozoan sized microbes in fine sand.

[5] A variety of explanations for attachment under unfavorable conditions and for spatial variations in the deposition rate coefficient have been proposed in the literature. Explanations can be broadly classified as chemically or physically based. Proposed chemical explanations include porous media charge variability [Johnson and Elimelech, 1995], heterogeneity in surface charge characteristics of colloids [Bolster *et al.*, 1999; Li *et al.*, 2004], deposition of colloids in a secondary energy minimum [Redman *et al.*, 2004; Hahn *et al.*, 2004; Tufenkji and Elimelech, 2005a], and colloid detachment [Tufenkji *et al.*, 2003]. Sometimes several of these explanations are invoked simultaneously to explain observed deposition behavior. For example, heterogeneity of surface charge characteristics and deposition of colloids in a secondary energy minimum have been used to explain nonexponential deposition profiles [Tufenkji and Elimelech, 2005a].

[6] Colloid filtration theory was developed from an analysis of deposition behavior on a single spherical grain collector. The Happel sphere-in-cell model is used in filtration theory to preserve overall porosity by representing the liquid as a continuous sheath completely surrounding the grains of a porous medium. Hence filtration theory neglects pore structure and grain-grain junctions. Other researchers have therefore suggested that deposition may occur as a result of physical factors that are not included in filtration theory, such as straining [Cushing and Lawler, 1998; Bradford *et al.*, 2002, 2003, 2004, 2005, 2006a; Li *et al.*, 2004; Tufenkji *et al.*, 2004; Bradford and Bettahar, 2005, 2006; Foppen *et al.*, 2005], soil surface roughness [Kretzschmar *et al.*, 1997; Redman *et al.*, 2001], and hydrodynamic drag [Li *et al.*, 2005].

[7] Review of the literature indicates that the extent of colloid attachment and the range of applicability of clean bed filtration theory is much less certain in the presence of repulsive interactions between colloids and grain surfaces than in the presence of attractive interactions. The objective of this work is to highlight reported discrepancies in experimental observations and filtration theory predictions, and to present experimental evidence that straining can play a significant role in colloid deposition under unfavorable attachment conditions. Implications for colloid straining on transport in saturated systems, the vadose zone, and heterogeneous formations will also be discussed. For the remainder of this manuscript we will limit our discussion to unfavorable attachment conditions.

2. Mechanisms of Colloid Deposition

[8] Three mechanisms of colloid deposition in saturated porous media have commonly been identified [e.g., McDowell-Boyer *et al.*, 1986], namely, (1) attachment, (2) mechanical filtration, and (3) straining. Each of these deposition mechanisms will be briefly discussed below

to provide a common reference point for subsequent discussion.

2.1. Attachment

[9] Attachment is a well-known mechanism of colloid deposition that involves collision with and subsequent retention of colloids at the solid-water interface. Mass transfer of colloids to the solid-water interface occurs via diffusion, interception, and sedimentation [Yao *et al.*, 1971]. Once a colloid collides with the solid-water interface, attachment depends on a force and/or torque balance that acts on the colloids [Cushing and Lawler, 1998; Li *et al.*, 2005] at this location. Cushing and Lawler [1998] indicated that as colloids approach the solid-water interface they experience forces due to gravity and buoyancy, fluid drag, electrical double-layer repulsion (or attraction), and London-van der Waals interaction. Variations in the fluid drag force with distance from the solid-water interface creates a torque on colloids near the solid surface.

[10] Colloid transport and fate models commonly employ a modified form of the advective-dispersive equation which incorporates colloid attachment theory [Harvey and Garabedian, 1991; Corapcioglu and Choi, 1996; Bolster *et al.*, 1999; Schijven and Hassanizadeh, 2000]. In this case, the aqueous phase mass balance equation for colloids may be written as

$$\frac{\partial(\theta_w C)}{\partial t} = -\nabla \cdot J_T - E_{sw} \quad (1)$$

where C [$N_c \text{ L}^{-3}$; N_c and L denote the number of colloids and length, respectively] is the colloid concentration in the aqueous phase, t [T ; T denotes units of time] is time, θ_w (dimensionless) is the volumetric water content, J_T [$N_c \text{ L}^{-2} \text{ T}^{-1}$] is the total colloid flux (sum of advective, dispersive, and diffusive fluxes), and E_{sw} [$N_c \text{ L}^{-3} \text{ T}^{-1}$] is the colloid mass transfer terms between the aqueous and solid phases.

[11] When attachment between colloids and grain surfaces control deposition the E_{sw} term can be written as

$$E_{sw} = \rho_b \frac{\partial(S_{att})}{\partial t} = \theta_w \psi_{att} k_{att} C - \rho_b k_{det} S_{att} \quad (2)$$

where ρ_b [$M \text{ L}^{-3}$; M denotes mass] is the soil bulk density, S_{att} [$N_c \text{ M}^{-1}$] is the solid phase concentration of attached colloids, ψ_{att} (dimensionless) is a dimensionless colloid attachment function, and k_{att} [T^{-1}] and k_{det} [T^{-1}] are the first-order colloid attachment and detachment coefficients, respectively. Clean bed attachment is assumed and filtration theory is incorporated into the k_{att} term of equation (2) when $\psi_{att} = 1$ and $k_{det} = 0$ [e.g., Yao *et al.*, 1971; Logan *et al.*, 1995] as

$$k_{att} = \frac{3(1 - \theta_w)}{2d_{50}} \eta \alpha v \quad (3)$$

where η (dimensionless) is the collector (porous medium) efficiency, α (dimensionless) is the colloid sticking efficiency, d_{50} [L] is the median porous medium grain diameter, and v [$L \text{ T}^{-1}$] is the pore water velocity. The collector efficiency is related to the frequency of colloid collisions on the grain surfaces and accounts for colloid removal due to diffusion, interception, and gravitational sedimentation [Logan *et al.*, 1995]. The value of η is frequently calculated using correlations written in terms of

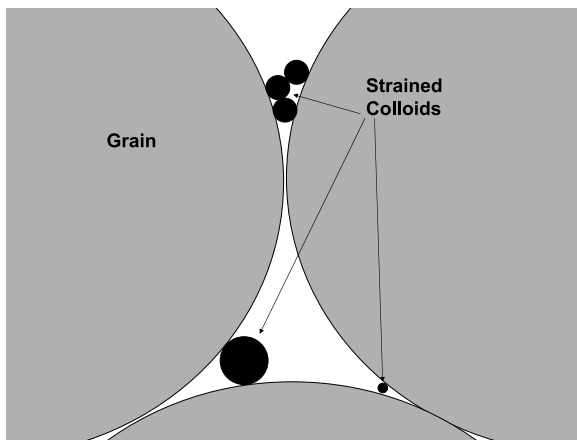


Figure 1. Illustration of strained colloids in the smallest regions of the soil pore space formed adjacent to points of grain-grain contact.

dimensionless variables [e.g., *Rajagopalan and Tien, 1976; Logan et al., 1995; Tufenkji and Elimelech, 2004a*]. The sticking efficiency is defined as the ratio of the deposition flux under unfavorable relative to favorable conditions, and it depends on the surface chemistries of the colloids and the grain surfaces [*Ryan and Elimelech, 1996*]. The value of α is typically assumed to be independent of the size of the colloid and the grain surface, which is accounted for by η in equation (3). In practice α is usually derived from experimental breakthrough curves, or from fitted values of k_{att} and calculated values of η , although theoretical approaches have also been proposed to predict α [*Ryan et al., 1999*].

[12] Time dependency in the colloid attachment rate as a result of blocking or ripening can be accounted for using the ψ_{att} term in equation (2). During blocking the value of ψ_{att} decreases with increasing S_{att} as a result of filling of favorable attachment sites such as metal oxide surfaces or clay edges. According to the Langmuirian approach [*Adamczyk, 1994*], $\psi_{att} = 1 - S_{att}/S_{att}^{max}$, where S_{att}^{max} [$N_c M^{-1}$] is the maximum solid phase concentration of attached colloids. In contrast, the random sequential adsorption (RSA) approach assumes that ψ_{att} is a nonlinear function of S_{att} [*Johnson and Elimelech, 1995*]. Less research has examined colloid ripening. During ripening ψ_{att} increases with increasing S_{att} due to favorable colloid-colloid interactions (interactions between colloids in solution and those attached to grain surfaces). *Tien* [1989] proposed a functional form that could be included in the ψ_{att} term to account for ripening.

[13] More recently it has been proposed that colloids and/or grain surfaces exhibit distributions of surface charge characteristics that produce a corresponding distribution of attachment coefficients and/or sticking efficiencies [*Bolster et al., 1999; Tufenkji et al., 2003; Li et al., 2004*]. Various probability density functions (pdf) for the attachment coefficient distribution have been hypothesized, including normal, lognormal, power law, and bimodal formulations [*Tufenkji et al., 2003*].

2.2. Mechanical Filtration

[14] Mechanical filtration refers to the complete retention of colloids, particles, and/or aggregates at the soil surface

that are larger than all of the soil pores [*McDowell-Boyer et al., 1986*]. Hence mechanical filtration implies that particle transport will not occur through a given porous medium when the diameter of the particle is greater than the largest pore size (network). Deposited particles at the soil surface form a filter cake that decreases the permeability of the filter cake–porous medium system over time as the cake thickens and the porosity decreases. Mechanical filtration is commonly used to remove colloids and particles from suspension during membrane filtration, but it is avoided in water treatment operations that are based on passage through porous media (infiltration basin and trenches, riverbank filtration, and sand filters) because of clogging problems.

2.3. Straining

[15] Figure 1 provides an illustration of strained colloids in the smallest regions of the soil pore space formed adjacent to points of grain-grain contact [*Cushing and Lawler, 1998*]. To better understand and define straining we will compare and contrast it to mechanical filtration and attachment. Similar to mechanical filtration, straining of colloids occurs in soil pores and depends on the ratio of the colloid and pore size. In contrast to mechanical filtration, straining only occurs in a fraction of the soil pore space, and colloid transport can still occur in the larger portions of the continuous pore networks. Similar to attachment, strained colloids experience forces due to gravity and buoyancy, fluid drag, electrical double-layer repulsion (or attraction), and London–van der Waals interaction. In contrast to colloid attachment, straining occurs in pores that are defined by the intersection of at least two solid–water interfaces (surfaces); i.e., grain–grain contacts or surface roughness. Hence straining deposition exhibits many similarities to both mechanical filtration and attachment.

[16] *Sakhivadivel* [1966, 1969] and *Herzig et al.* [1970] developed geometric relations between the effective diameter of colloids and soil grain size distribution characteristics to predict mass removal by straining. *Matthess and Pekdeger* [1985] generalized this rule to porous media made up of a distribution of grain sizes and presented theoretical criteria suggesting that the ratio of the colloid to median grain diameter needs to be greater than 18% for straining to occur in uniform sand, although increasing the sand gradation would lower this threshold. Previous reviews on microbe transport in saturated and unsaturated systems have therefore mostly neglected straining as a mechanism for colloid retention [e.g., *Schijven and Hassanizadeh, 2000; Harvey and Harms, 2002; Jin and Flury, 2002; Ginn et al., 2002*]. Recent experimental observations, however, suggests that straining occurs for much lower ratios of the colloid to median grain diameters. Below we summarize available evidence for the significance of straining deposition under unfavorable attachment conditions, and discuss the implications of these findings on colloid transport in porous media.

3. Evidence and Implications for Straining

3.1. Pore Size Distribution Information

[17] Attachment is commonly assumed a priori to be the controlling deposition process, and little attention has been paid to the colloid and porous media parameter ranges that may limit filtration theory applicability. The following

Table 1. Average Parameter Values for the Capillary Pressure–Saturation Model of *van Genuchten* [1980] for 12 Major Soil Textural Groups According to *Carsel and Parrish* [1988], as Well as the Calculated Percentage of the Pore Space Smaller Than Critical Straining Pore Diameter for Virus, Bacteria, and Protozoa

Soil Texture	S_{rw}	α_p , cm ⁻¹	n ($m = 1 - 1/n$)	Percent Excluded Pore Space		
				Virus	Bacteria	Protozoa
Sand	0.10	0.15	2.68	10.5	10.5	10.5
Loamy sand	0.14	0.12	2.28	13.9	14.0	14.4
Sandy loam	0.16	0.08	1.89	15.9	17.1	19.3
Loam	0.18	0.04	1.56	19.8	27.0	34.5
Silt	0.07	0.02	1.37	16.9	36.1	50.0
Silt loam	0.15	0.02	1.41	21.1	36.1	47.9
Sandy clay loam	0.26	0.06	1.48	27.7	34.4	40.4
Clay loam	0.23	0.02	1.31	34.0	50.5	61.2
Silty clay loam	0.21	0.01	1.23	42.2	63.2	74.4
Sandy clay	0.26	0.03	1.23	42.2	57.9	66.7
Silty clay	0.19	0.01	1.09	70.8	86.2	91.8
Clay	0.18	0.01	1.09	68.1	83.3	89.3

simple example illustrates that straining has significant implications for colloid transport in field soils such as those found in the vadose zone, at contaminated waste sites, and during riverbank filtration. Assume that the critical straining pore diameter for viruses, bacteria, and protozoa are 0.1, 2, and 6 μm , respectively. These pore diameters can be related to a critical capillary pressure (P_c) for straining according to Laplace's equation by assuming that soil pores can be approximated as a bundle of cylindrical capillaries that are tortuous [e.g., *Dane and Hopmans*, 2002; *Flint and Flint*, 2002]. The fraction of pore space where straining will occur (γ) can then be inferred from measured capillary pressure curves and residual saturations. If soil water retention data are parameterized using the model of *van Genuchten* [1980], then γ is given as:

$$\gamma = (1 - S_{rw})^* \left[1 + \left(\frac{2\sigma\alpha_p}{\rho g R} \right)^n \right]^{-m} + S_{rw} \quad (4)$$

where S_{rw} (dimensionless) is residual water saturation, α_p [L^{-1}] is the reciprocal of the air entry pressure head, ρ [M L^{-3}] is the density of the liquid phase, g [L T^{-2}] is the acceleration due to gravity, σ [M T^{-2}] is the surface tension, R [L] is the pore radius that corresponds to a given colloid size, and m (dimensionless) and n (dimensionless) are fitting parameters.

[18] Table 1 presents the average capillary pressure-saturation parameters of the 12 major soil textural groups according to *Carsel and Parrish* [1988], as well as the percentage of the pore space where straining would occur according to equation (4). Depending on soil texture, straining is predicted in 10.5 to 70.8, 10.5 to 86.2, and 10.5 to 91.8% of the soil pore space for virus, bacteria, and protozoa transport, respectively. It should be noted, however, that the actual pore space of soils is expected to be more complex than that predicted by the bundle of capillary tubes model. Furthermore, the above analysis does not consider the potential influence of surface roughness. *Tufenkji et al.* [2004] reported that straining increases with increasing surface roughness. Despite these acknowledged

limitations, Table 1 is believed to provide useful information to help identify the relative importance of straining in the various soil textural groups.

[19] Alternatively, *Herzig et al.* [1970] calculated the volume of spherical colloids that could be retained in soil pores based on geometric considerations. Geometric estimates of the straining fraction of the pore space such as those of *Herzig et al.* [1970] underestimate this value because the approach assumes a uniform grain size equal to d_{50} . In reality, the smaller soil pore spaces will control straining processes.

3.2. Size Exclusion

[20] Although size exclusion of colloids is widely acknowledged in the colloid transport literature [*Ryan and Elimelech*, 1996; *Ginn*, 2002], the complementary nature of straining and size exclusion has generally not been recognized. Size exclusion affects the mobility of colloids by constraining them to more conductive flow domains and larger pore networks that are physically accessible. As a result, colloids can be transported faster than a conservative solute tracer [*Reimus*, 1995; *Cumbie and McKay*, 1999; *Harter et al.*, 2000; *Bradford et al.*, 2003]. Conversely, size exclusion also implies that some pores of the medium are smaller than the colloids. If colloids encounter these smaller pores they can become physically entrapped as a result of straining. Hence straining and size exclusion are complementary deposition and transport processes, respectively. The minimum part of the pore space where straining occurs can be estimated by γ from Table 1. Mobile colloids may move in at most the remaining fraction of the water filled pore space (i.e., $S_w - \gamma$; where S_w is the water saturation). When $\gamma > S_w$ no colloids will be mobile.

[21] We note that size exclusion also has important implications for attachment because it will lead to an increase in the colloid transport velocity [*Ryan and Elimelech*, 1996; *Ginn*, 2002] and a decrease in the solid surface area that is accessible for attachment; i.e., mobile colloids are transported in only a fraction of the pore space. Filtration theory predicts a velocity dependence for the attachment coefficient [e.g., *Logan et al.*, 1995]. Several methods have recently been proposed to simulate the enhanced velocity of colloids as a result of size exclusion [*Ginn*, 2002; *Bradford et al.*, 2003] and the interested reader is referred to these publications for details on this topic. Conversely, little is known about the potential influence that size exclusion has on the accessibility of the solid surface area for attachment. We propose herein that the fraction of the solid surface area (f_{SA}) that is available for attachment can be estimated from:

$$f_{SA} = \frac{\int_0^{S_w} P_c(x) dx}{\int_0^1 P_c(x) dx} \quad S_w > \gamma \quad (5)$$

where P_c is the capillary pressure, and x is a dummy variable of integration. Equation (5) is developed by integrating the area contributed by colloid accessible pores (the numerator) and then normalizing this quantity by the total area of the soil pore space (denominator). This approach predicts an increasing γ or decreasing S_w will lead to a lower f_{SA} .

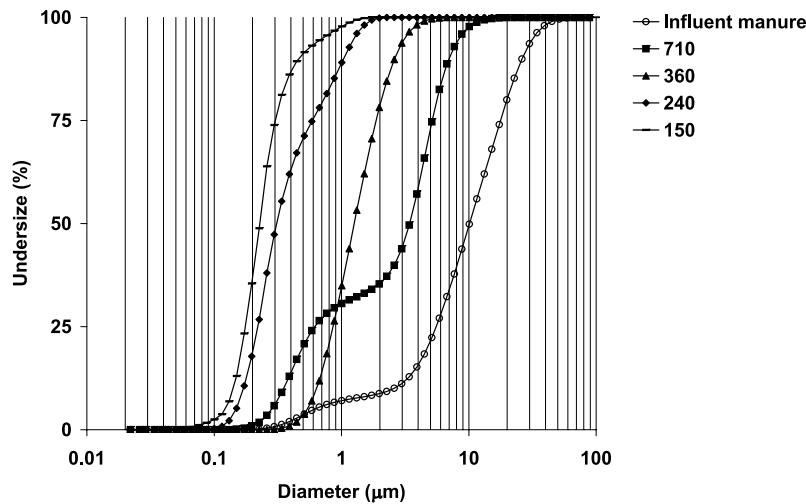


Figure 2. Cumulative size distribution (CSD) for manure effluent in 710, 360, 240, and 150 μm Ottawa sands after 95 min (around 2.5 pore volumes). The CSD of the influent manure suspension is also shown in the figure for reference. Reprinted with permission from *Bradford et al.* [2006a].

3.3. Colloid Size Distribution Information

[22] Information on the mechanical filtration and/or straining potential of soils can be deduced by considering changes in the colloid size distribution characteristics before and after passage through different sized porous media. *Bradford et al.* [2006a] recently presented the cumulative size distribution (CSD) of manure particles in effluent samples after 95 minutes of passage (approximately 2.5 PV) through several sized sands, as well as the CSD for the influent manure suspension. This same information is presented in Figure 2. Manure particles larger than about 12, 5, 0.8, and 0.5 μm were completely removed after passage through the 710, 360, 240, and 150 μm sands, respectively, due to mechanical filtration and/or straining. This corresponds to ratios of manure particle to median grain size of 0.003–0.017. These ratios are significantly smaller than the straining criterion of 0.18 proposed by *Matthess and Pekdeger* [1985] but are similar to the 0.005 guideline proposed by *Bradford et al.* [2003].

3.4. Time- and Concentration-Dependent Deposition

[23] Time- and concentration-dependent deposition coefficients have been reported in the literature [*Tan et al.*, 1994; *Lindqvist et al.*, 1994; *Johnson and Elimelech*, 1995; *Liu et al.*, 1995; *Rijnaarts et al.*, 1996; *Comesano and Logan*, 1998; *Comesano et al.*, 1999; *Brown et al.*, 2002; *Foppen et al.*, 2005; *Bradford and Bettahar*, 2006]. Although blocking of favorable attachment sites (metal oxide surfaces or clay edges) has typically been used to explain this behavior, filling of straining sites by colloids provides a plausible alternative explanation [*Foppen et al.*, 2005; *Bradford and Bettahar*, 2006]. In this case, the number of colloids required to fill γ (Table 1) for a given colloid and porous medium can be calculated [*Foppen et al.*, 2005]. The rate of filling of these sites is theoretically dependent on the concentration of the colloids in suspension (e.g., higher colloid concentrations fill straining sites more rapidly than low concentrations). Large numbers of colloids will be required to fill even small straining fractions of the pore space. As accessible straining sites become filled, water and

colloids are diverted from these regions. This leads to less deposition and increasing effluent concentrations of larger particles that would have previously been retained in these (unfilled) straining sites. *Bradford et al.* [2006a] observed this time- and size-dependent deposition behavior for *Giardia* cysts and for manure particles in several sized sands.

3.5. Hyperexponential Deposition Profiles

[24] Under unfavorable attachment conditions deposition profiles for many types of colloids (latex microspheres, bacteria, viruses, protists) have been reported to follow a hyperexponential profile distribution [*Albinger et al.*, 1994; *Baygens et al.*, 1998; *Simoni et al.*, 1998; *Bolster et al.*, 2000; *DeFlaun et al.*, 1997; *Zhang et al.*, 2001; *Redman et al.*, 2001; *Bradford et al.*, 2002; *Li et al.*, 2004; *Tufenkji and Elimelech*, 2005a], implying a decreasing rate of deposition with increasing transport distance. As an example, Figures 3a and 3b present observed and simulated effluent concentration curves (Figure 3a) and deposition profiles (Figure 3b) for 1 μm carboxyl latex microspheres in 150 μm Ottawa (quartz) sand. Additional information about the experimental conditions is provided by *Bradford and Bettahar* [2006]. The simulations in Figure 3 considered (1) clean bed attachment, (2) attachment and Langmuirian blocking, or (3) attachment and RSA blocking. Model parameters were fitted to the effluent concentration curves and deposition profiles. Figures 3a and 3b indicate that the various attachment models provided a reasonable characterization of the effluent concentration, but a poor description of the deposition profiles. Proper characterization of both effluent curves and deposition profiles is essential for predicting colloid transport at larger spatial and temporal scales [*Bolster et al.*, 2000].

[25] As discussed earlier in the manuscript, straining and mechanical filtration are closely related processes of deposition. It is therefore logical to anticipate that deposition profiles for mechanical filtration and straining will share some similarities. Similar to mechanical filtration, *Bradford et al.* [2002, 2003, 2004, 2005, 2006a] observed that the bulk of colloid straining occurred near the column inlet (see

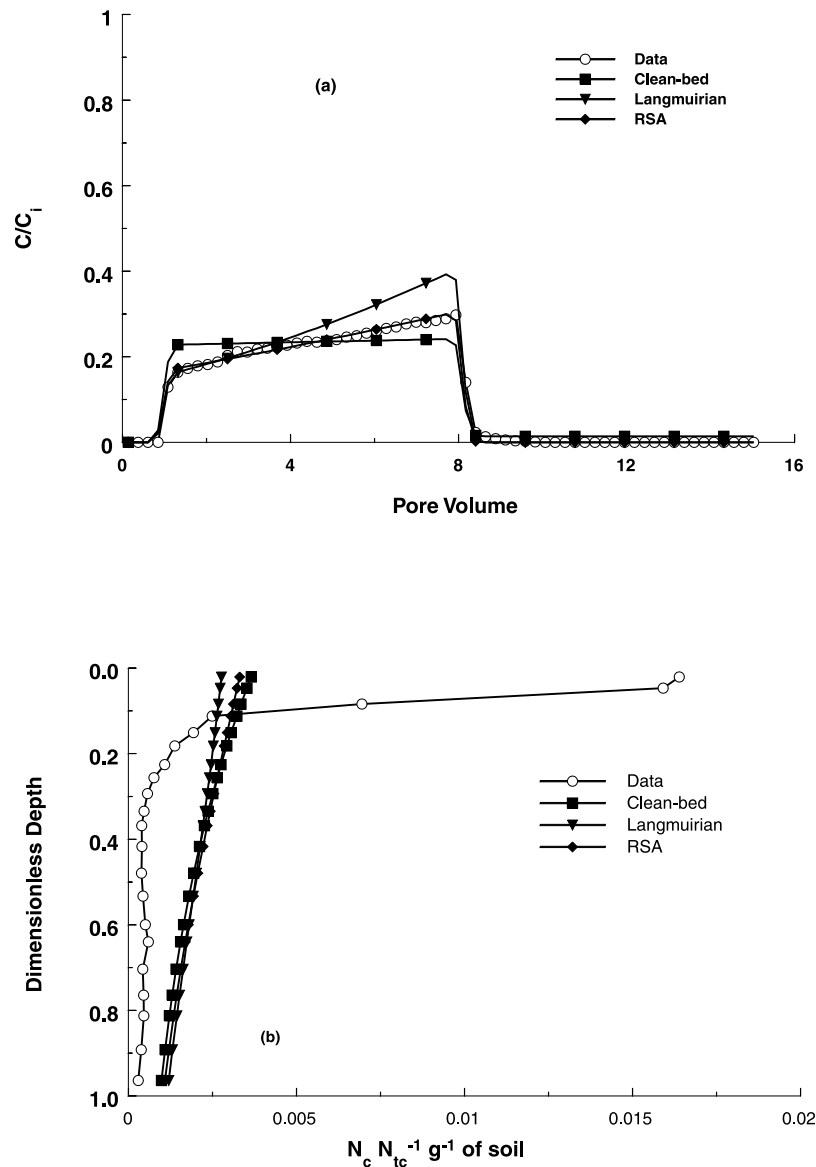


Figure 3. Observed and simulated (a) effluent concentration curves and (b) deposition profiles for $1 \mu\text{m}$ carboxyl latex microspheres in $150 \mu\text{m}$ Ottawa (quartz) sand. In Figure 3a, relative effluent concentrations (C/C_i) are plotted as a function of pore volumes, whereas in Figure 3b the normalized concentration (number of colloids, N_c , divided by the total number added to the column, N_{tc}) per gram of dry sand is plotted as a function of dimensionless distance (distance from the column inlet divided by the column length). The simulations in Figure 3 considered clean bed attachment, attachment and Langmuirian blocking, or attachment and RSA blocking.

Figure 3b for dimensionless depth <0.2). *Bradford et al.* [2002] hypothesized that small pores that can cause straining were more accessible to colloids at the sand surface. One potential explanation can be obtained if we view the porous medium as a bundle of capillary tubes. Straining occurs primarily near the sand surface at the entrance of the smallest capillaries in the region of the pore space defined by γ , whereas colloid transport may still occur in the larger capillaries in the region of the pore space defined by $S_w - \gamma$. Deposition rapidly decreased with increasing distance (see Figure 3b for dimensionless depths >0.2) because colloids that initially enter the pore space defined by γ have been removed upgradient. Advection, dispersion, and size exclusion tend to constrain the remaining mobile colloids to the

larger more conductive pore networks ($S_w - \gamma$) and thus bypass much of the smaller pore spaces ($<\gamma$). Hence attachment becomes relatively more important at greater depths [*Bradford et al.*, 2003].

[26] The potential roles of size exclusion and dispersion on colloid transport to small pores are relatively easy to understand. Advective transport processes can also play an important role. The ratio of the water flux that is confined to the region of the pore space defined by γ ($q_{ws} - LT^{-1}$) to the total water flux ($q_w - LT^{-1}$) can be calculated as:

$$\frac{q_{ws}}{q_w} = \frac{k_{rs}}{k_{rw}} \quad (6)$$

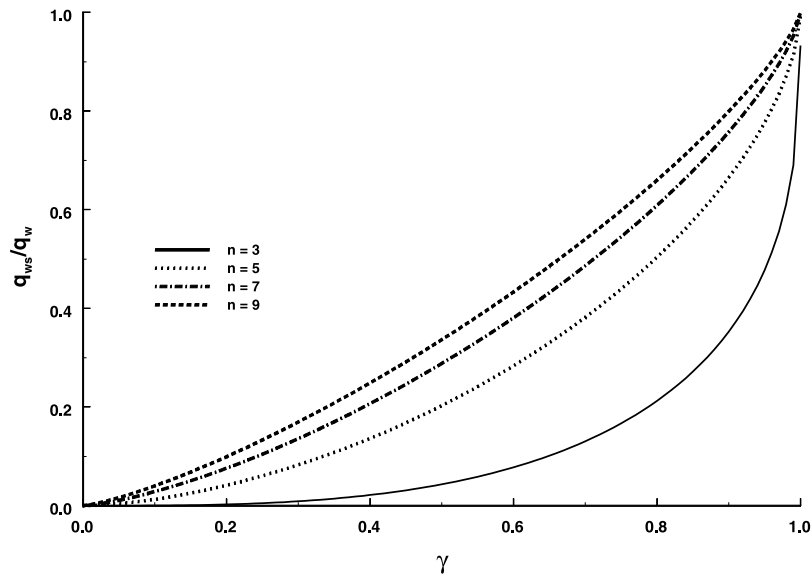


Figure 4. Relative water flux associated with straining sites (q_{ws}/q_w) as a function of γ for various hypothetical (saturated) porous media. The value of n in the legend corresponds to the *van Genuchten* [1980] capillary pressure curve parameter ($m = 1 - 2/n$).

where k_{rw} (dimensionless) is the water relative permeability and k_{rs} (dimensionless) is the water relative permeability to straining sites. The value of k_{rw} is typically determined from capillary pressure data using a pore size distribution model. In analogy to this approach, the value of k_{rs} can be determined using the pore size distribution model of *Burdine* [1953] by limiting the integration to only the straining areas of the porous medium as follows:

$$k_{rs}(S_w) = S_w^2 \frac{\int_0^\gamma R(x)^2 dx}{\int_0^1 R(x)^2 dx} \quad (7)$$

Equation (7) assumes that the value of γ is independent of the water saturation, see equation (4), and that the influence of tortuosity of the water flow path can be quantified as S_w^2 [Burdine, 1953]. The predicted value of q_{ws}/q_w for a particular porous medium as a function of γ can be calculated from equation (6) and the analytical solution for equation (7) [e.g., *van Genuchten et al.*, 1991]. The values of q_{ws}/q_w as a function of γ for various hypothetical (saturated) porous media are shown in Figure 4. The value of n in the legend corresponds to the *van Genuchten* [1980] capillary pressure curve parameter ($m = 1 - 2/n$), which is related to the slope of the capillary pressure curve at approximately the inflection point. A higher value of n corresponds to a more uniform pore size distribution. Hence Figure 4 indicates that for small values of γ only a small fraction of the water flux is associated with straining sites. Larger fractions of the water flux occur in straining sites with increased values of γ and in more uniform soils.

[27] Alternative explanations for hyperexponential deposition profiles have also appeared in the literature. Colloid surface charge heterogeneity has frequently been invoked as a cause for observed deviations between filtration theory predictions and measured hyperexponential deposition pro-

files [Bolster *et al.*, 1999; Tufenkji *et al.*, 2003; Li *et al.*, 2004; Tufenkji and Elimelech, 2005b]. In addition, a second hypothesis of attachment in the secondary minimum of the Derjaguin–Landau–Verwey–Overbeek (DLVO) potential energy distribution is needed for unfavorable attachment conditions. Two relatively simple experiments could be used to test the surface charge heterogeneity hypothesis. First, the measured zeta potential distribution for a colloid suspension should be consistent with the assumed attachment coefficient pdf. Second, a measurable change in the zeta potential distribution must exist for this colloid suspension before and after passage through the porous medium. Additional research is needed to fully test the surface charge heterogeneity hypothesis outlined above.

[28] It should be noted that Li *et al.* [2004] suggested that small variations in the measured colloid surface charge (<0.5% of a measured zeta potential) could yield sufficient variations in the attachment coefficient to produce hyperexponential deposition profiles. We believe that this analysis is only valid in the absence of an energy barrier to attachment in the primary minima or under conditions that are favorable to attachment in the secondary minimum of the DLVO potential energy distribution. To illustrate this point, the total interaction energy (sum of London–van der Waals attraction and electrostatic double-layer repulsion) upon approach of variously charged (zeta potentials ranging from -40 mV to -5 mV) $1.1 \mu\text{m}$ spherical colloids to a glass bead surface (sphere–plate interaction) was calculated using DLVO theory [Derjaguin and Landau, 1941; Verwey and Overbeek, 1948]. In these calculations, constant potential electrostatic double layer interactions were quantified using the expression of Hogg *et al.* [1966] (zeta potentials were used in place of surface potentials), and retarded London–van der Waals attractive interaction was determined from the expression of Gregory [1981]. A value of 3.8×10^{-21} J was chosen for the Hamaker constant [Li *et al.*, 2004], and the zeta potential of the glass bead in 10 mM solution was taken to be -50.2 mV [Tufenkji and

Table 2. Summary of Attachment Model Parameters for Column Experiments With Various Sized Carboxyl Latex Microspheres and Ottawa Quartz Sands (710, 360, 240, and 150 μm)^a

d_{50} , μm	d_c , μm	Column k_{att} , min^{-1}	Column k_{det} , min^{-1}	η	α	Observed Batch C/C _i	Predicted Batch C/C _i
710	0.45	3.75E-03	3.02E-03	2.09E-02	4.92E-02	0.66	0.56
710	1.00	4.01E-03	7.50E-05	1.25E-02	9.12E-02	0.78	0.39
710	2.00	2.01E-02	1.06E-03	1.34E-02	4.03E-01	0.65	0.06
710	3.20	1.93E-02	4.10E-04	2.23E-02	2.27E-01	0.59	0.03
360	0.45	5.46E-03	2.15E-03	3.44E-02	2.00E-02	0.68	0.40
360	1.00	3.57E-03	6.65E-04	2.03E-02	2.19E-02	0.84	0.46
360	2.00	3.69E-02	8.61E-04	1.86E-02	2.43E-01	0.67	0.02
360	3.20	2.97E-02	2.87E-04	2.70E-02	1.34E-01	0.63	0.01
240	0.45	3.06E-03	4.54E-03	4.45E-02	5.22E-03	0.66	0.66
240	1.00	5.68E-02	1.00E-03	2.64E-02	1.62E-01	0.81	0.02
240	2.00	6.30E-02	6.00E-04	2.32E-02	2.12E-01	0.60	0.01
240	3.20	6.30E-02	3.75E-04	3.29E-02	1.44E-01	0.61	0.01
150	0.45	1.28E-02	1.44E-03	5.62E-02	1.07E-02	0.61	0.13
150	1.00	2.22E-02	1.24E-03	3.55E-02	3.07E-02	0.80	0.06
150	2.00	7.70E-02	7.09E-04	3.28E-02	1.16E-01	0.59	0.01
150	3.20	1.32E-01	2.40E-03	4.49E-02	1.48E-01	0.61	0.02

^aSee Bradford *et al.* [2002]. Here d_c is the colloid diameter equal to 0.45, 1, 2, and 3.2 μm . Observed and predicted (equation (8)) relative concentrations for corresponding batch experiments are also presented. Read 3.75E-03 as 3.75×10^{-3} .

Elimelech, 2005a]. As the zeta potential of the colloid is varied from -40 to -5 mV the height of the repulsive energy barrier changed from 1243 to 20 $k_b T_k$ (where k_b is the Boltzman constant and T_k is the temperature in degrees Kelvin), respectively, and attachment in the primary minima is therefore not expected for these conditions. Attachment in the secondary energy minima is also not believed to play a significant role in deposition over this range in zeta potentials, because colloid diffusion and hydrodynamic drag are sufficient to overcome this weak association (depth of the secondary minima ranged from -0.4 to $-0.7 k_b T_k$ when the zeta potential of the colloid ranged from -40 to -5 mV).

3.6. Dependence of Deposition on Colloid and Porous Medium Size

[29] Filtration theory predicts that the colloid sticking efficiency, α in equation (3), depends primarily on the surface chemistry of the colloids and the soil grains. In contrast, the collector efficiency, η , includes the dependence

of the size of the colloid and the porous medium, as well as other factors. The value of α or a pdf of α is therefore expected to be independent of the size of the colloid and soil grain for a given set of chemical properties. Evidence exists that this condition will be violated when significant straining occurs. For example, Bradford *et al.* [2002] measured values of k_{att} and k_{det} for 0.45, 1, 2, and 3.2 μm carboxyl latex microspheres in column experiments using various sized Ottawa sands (710, 360, 240, and 150 μm) and glass beads (260 μm). The Darcy velocity and porosity in these experiments ranged from 0.10–0.11 cm min^{-1} and 0.32–0.38, respectively. Variability in the surface charge characteristics for the latex microspheres (the standard deviation of measured zeta potentials was less than 5.9 mV) and quartz sands (99.9% quartz) was thought to be minimal. Table 2 presents values of η (from a correlation presented by Tufenkji and Elimelech [2004a]) and α calculated from some of this experimental data. Notice that a systematic variability exists in the calculated values of α , which tend

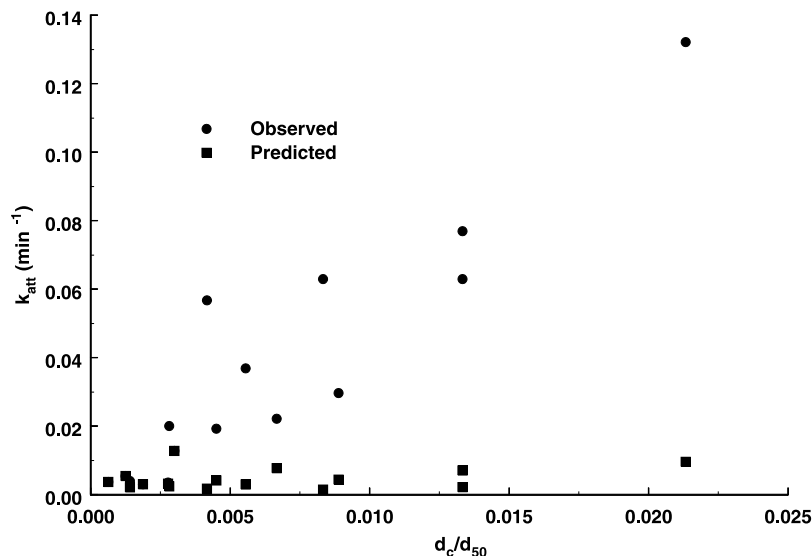


Figure 5. Measured and predicted values of k_{att} as a function of d_c/d_{50} . Predicted values of k_{att} were obtained using equation (3), η from Table 2, and values of α determined from the smallest 0.45 μm colloid (system with minimal straining) in each porous medium.

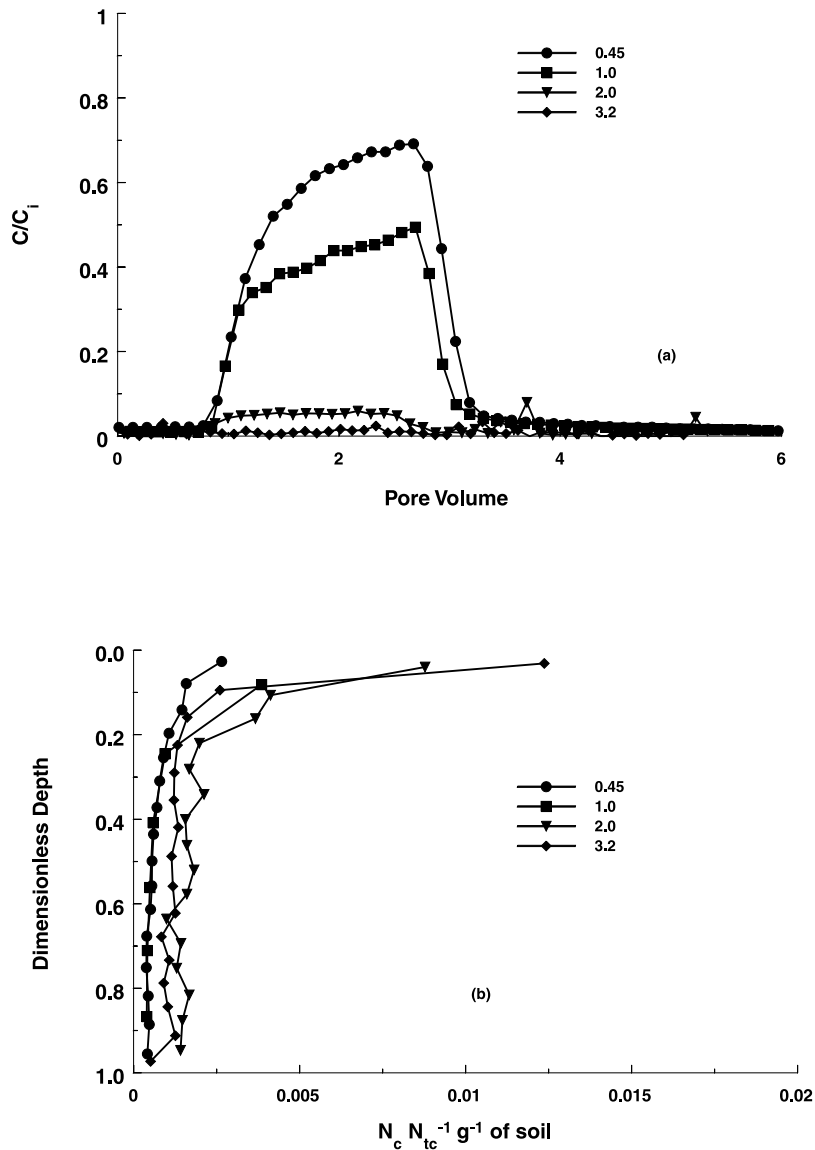


Figure 6. (a) Effluent concentration curves and (b) deposition profiles for 0.45, 1.0, 2.0, and 3.2 μm carboxyl latex microspheres in 150 μm Ottawa sands.

to increase with increasing size of the sand and colloid. Figure 5 summarizes the data by means of a plot of measured and predicted values of k_{att} as a function of the ratio of the colloid diameter (d_c) to the median grain size. Predicted values of k_{att} were in this case obtained using equation (3) and η (Table 2), with values of α determined from the smallest 0.45 μm colloid (system with minimal straining) in each porous medium. We believe that this assumption is reasonable because DLVO calculations (following a similar protocol as discussed above) for these colloids (0.45, 1, 2, and 3.2 μm colloids with zeta potentials of approximately -57 , -77 , -71 , and -57 mV, respectively) and experimental conditions (ionic strength of 1 mM and zeta potential of quartz equal to -38 mV) indicate the presence of a large energy barrier to deposition in the primary minima ($>576 k_b T_k$), and an insignificant secondary energy minima ($> -0.09 k_b T_k$). Predicted values of k_{att} significantly underestimated deposition with increasing size of the sand and colloid, thus suggesting that η is not a

complete descriptor of attachment under unfavorable conditions or that another mechanism may be involved in the colloid deposition.

[30] The shape of the deposition profile is also expected to be independent of the colloid and soil size according to filtration theory (e.g., only the magnitude of deposition changes with η). The same result is anticipated when using a given distribution of k_{att} . Figure 6 presents effluent concentration curves (Figure 6a) and deposition profiles (Figure 6b) for 0.45, 1.0, 2.0, and 3.2 μm carboxyl latex microspheres in 150 μm Ottawa sand. Bradford *et al.* [2002] provides details on the experimental conditions for these data. Notice that the shape of the deposition profile changes with colloid size. As colloid size increases, the deposition profile exhibits a sharper initial decrease with increasing depth (i.e., becomes more hyperexponential). Similar behavior can be observed for a given colloid in variously sized Ottawa sands; i.e., increasing hyperexponential deposition profiles for smaller sized sands [Bradford

et al., 2002]. Hence deviations between filtration theory and experimental deposition profiles tend to increase with increasing colloid size [Bradford *et al.*, 2002; Tufenkji and Elimelech, 2005a] as well as decreasing sand size [Bradford *et al.*, 2002]. All of these observations suggest that attachment (in the primary or secondary minima of the DLVO potential energy distribution) is not the only factor controlling colloid deposition.

3.7. Batch Release Rates

[31] Differences in the deposition behavior of colloids in batch and column displacement studies have frequently been reported [Schijven and Hassanizadeh, 2000], and have been attributed to differences in the accessibility of colloids to the solid grains (greater accessibility in batch than column), system hydrodynamics, and time of equilibration. Despite these acknowledged limitations, comparison of batch and column values of colloid deposition provides a useful way to infer the potential role of pore structure (largely absent in continuously mixed batch systems) on straining deposition. The analytic solution of equation (1) and equation (2) for batch conditions ($J_T = 0$ and $\psi_{att} = 1$) is given as [Schijven and Hassanizadeh, 2000]:

$$\frac{C}{C_i} = \frac{k_{det} + k_{att} \exp[-(k_{att} + k_{det})t]}{k_{att} + k_{det}} \quad (8)$$

Comparison of batch and column derived values of k_{att} and k_{det} therefore provides one means to examine mechanisms of colloid deposition. As an example, Bradford *et al.* [2002] measured values of k_{att} and k_{det} for 0.45, 1, 2, and 3.2 μm carboxyl latex microspheres in column experiments using various sized Ottawa sands (710, 360, 240, and 150 μm) and glass beads (260 μm). These authors also determined relative concentrations for these same colloids after 4 hours of batch equilibration (10 gram of sand in 10 ml of suspension) with the various porous media. Table 2 provides values of k_{att} and k_{det} determined from some of these column experiments, as well as the measured and predicted (equation (8)) relative concentrations for colloids in the batch studies. The predicted relative concentrations in the batch system were much lower than the measured values (as low as 1% of the measured value), especially for the finer textured sand and larger colloids. The large differences in measured and predicted relative concentrations in the batch systems strongly suggest that attachment is not the dominant mechanism of deposition in these systems.

[32] A second test to determine whether column-derived values of k_{att} and k_{det} are consistent with batch systems is outlined below. After completion of a colloid transport experiment, the porous medium can be excavated into vials containing excess eluant. The vials can then be slowly shaken for several minutes, and the concentration of the colloids in the excess solution can be measured. In the absence of straining, equilibrium colloid concentrations in batch systems should be consistent with the measured detachment rates from the column experiments and therefore given as

$$C = \frac{k_{det}\rho_b S}{k_{att}\theta_w} \quad (9)$$

Here parameters on the right hand side of equation (9) can be determined from the column data. Hence low detachment rates in the column experiments should produce low colloid concentrations in the batch system. Conversely, if high concentrations of colloids are recovered in the batch system and low values of k_{det} were measured in the column experiments, then this implies that deposition was not controlled by attachment. It should be noted that protocols similar to those outlined above were employed by Bradford *et al.* [2002, 2003, 2004, 2005, 2006a] to determine deposition profiles for various colloids (0.45–3.2 μm carboxyl and sulfate latex, *Cryptosporidium* oocysts, and *Giardia* cysts). Reasonable mass balances were obtained in the experiments, while column derived values for k_{det} were typically low. This suggests that attachment was not the controlling mechanism for deposition in these systems. Other researchers have altered the batch solution composition (i.e., high pH and lower ionic strength) [Tufenkji and Elimelech, 2004b] or sonicated [Li *et al.*, 2004] the excavated sand from column experiments when determining deposition profiles. In these cases, direct comparison of batch and column values of k_{att} and k_{det} is not possible.

3.8. Solution Chemistry

[33] Previous research on straining deposition has focused on the influence of physical factors such as the size of the colloid and the porous medium. Little is known about the role of solution chemistry on straining deposition, and we are unaware of any published studies on this topic. It is, however, logical to anticipate that solution chemistry will play an important function in straining because it influences the force balance associated with colloids in pores.

[34] Deposition under unfavorable conditions is known to be highly dependent on solution chemistry [e.g., Li *et al.*, 2004; Tufenkji and Elimelech, 2004b, 2005a]. For a given solution composition, increasing the solution ionic strength tends to produce an increase in the colloid deposition. This has been attributed to changes in the DLVO total interaction energy between colloids and collectors. Specifically, at higher ionic strengths the energy barrier to deposition in the primary minima decreases and the depth of the secondary minima increases. Enhanced attachment with increasing ionic strength has therefore been attributed to surface charge variability of the collector and colloids, and interactions associated with the secondary minima.

[35] We believe that small soil pores (straining sites) provide optimum locations for colloids that are weakly associated with the solid phase (secondary energy minima) to be retained because of reduced fluid drag, pore size limitations, and enhanced DLVO interactions. Recent experimental evidence supports this hypothesis. For example, X-ray microtomography observations by Li *et al.* [2006a, 2006b] indicate that deposition at grain-grain contacts is highly dependent on the solution chemistry. Deposition rates in impinging jet systems and in packed sand columns under similar hydrodynamic and chemistry conditions also demonstrate a significant role of pore structure [Redman *et al.*, 2004; Walker *et al.*, 2004; Brow *et al.*, 2005]. Hoek and Agarwal [2006] reported that colloids in small pores can experience an interaction energy that is up to five times larger than the sphere-plate interaction energy profile. As an example of the coupled influence of solution chemistry on straining deposition consider the effluent concentration

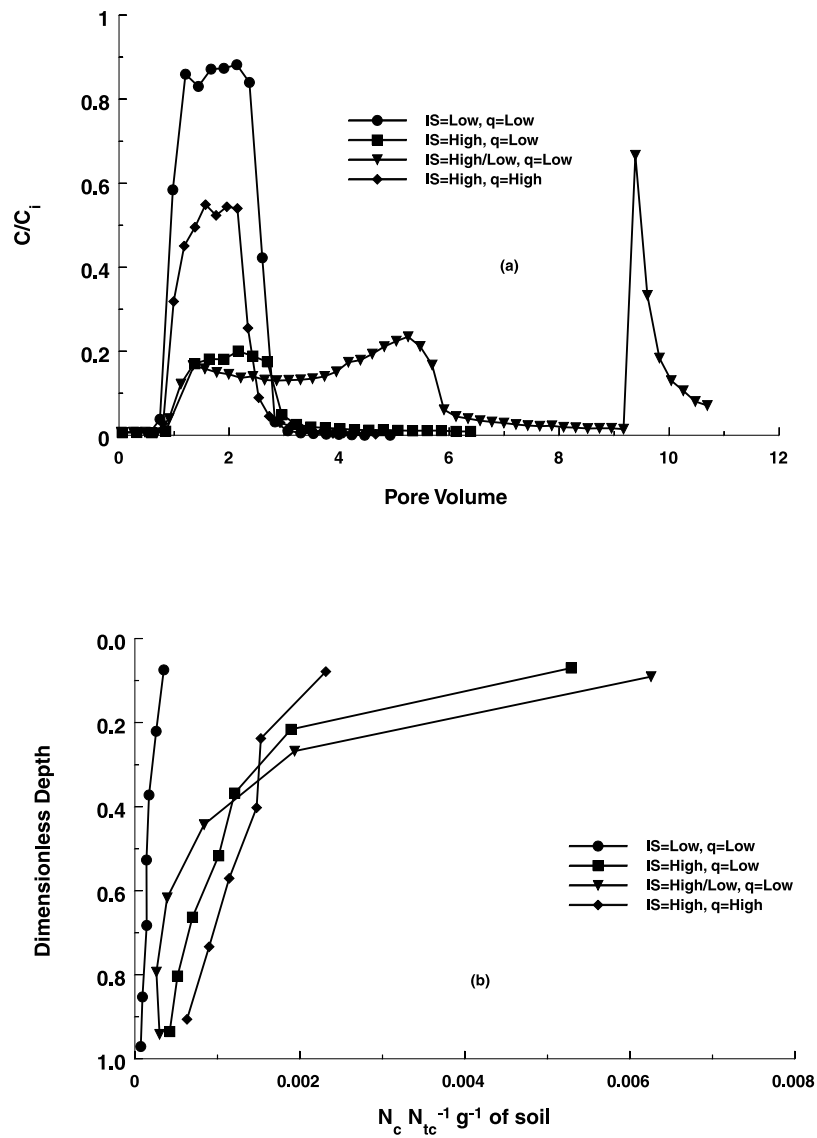


Figure 7. (a) Effluent concentration curves and (b) deposition profiles for 1.0 μm modified carboxyl latex microspheres in 150 μm Ottawa sands. All of the experiments were conducted at a solution pH of 10 (buffered with 1.7 mM NaHCO_3 +1.7 mM Na_2CO_3) but using various Darcy velocities and/or ionic strengths. In the legend, IS denotes ionic strength with Low = 6 mM, and High = 56 mM. IS = High/Low indicates that the solution ionic strength was switched from 56 to 6 mM after around 8.5 pore volumes. The Darcy velocity is denoted by q in the legend, and q = Low is 0.21 cm min^{-1} and q = High is 0.45 cm min^{-1} .

curves and deposition profiles for 1 μm carboxyl modified latex microspheres (zeta potential was -110 to -70 mV at ionic strengths ranging from 6 to 56 mM, respectively) in 150 μm Ottawa sand shown in Figures 7a and 7b, respectively. These transport experiments were conducted using deionized water buffered to a pH of 10 (1.7 mM NaHCO_3 + 1.7 mM Na_2CO_3) and employed different ionic strengths (adjusted using sodium bromide). As the ionic strength was increased from 6 mM (IS = Low, q = Low) to 56 mM (IS = High, q = Low), increased colloid deposition occurred and the retention profiles became more hyperexponential. An additional 56 mM experiment was conducted (IS = High/Low, q = Low), but this time the input colloid pulse duration was three times longer and the column was flushed with an additional 2.2 pore volumes of 6 mM solution before

excavation. Because of the lower ionic strength of this solution, a second peak in the breakthrough curve occurred as a result of released colloids. This behavior has typically been ascribed to colloids associated with the secondary minima [Franchi and O'Melia, 2003; Hahn and O'Melia, 2004; Hahn et al., 2004]. We believe that secondary minima association alone cannot explain the observed behavior for the following reasons. First, the corresponding deposition profile for this experiment was hyperexponential, even after flushing with an additional 2.2 pore volumes of 6 mM solution. Second, mass balance information indicates that the recovered colloids in the effluent only accounted for 25% of the injected mass (IS = High/Low, q = Low), whereas in the experiment conducted using 6 mM solution (IS = Low, q = Low) 83% of the injected colloids was

recovered in the effluent. Hence temporal changes in solution ionic strength increased the total deposition in the sand and produced more hyperexponential profiles. All of these observations strongly indicate that the pore structure (straining) played a coupled role with solution chemistry in retaining these colloids. This is a research topic of ongoing investigation.

[36] It should also be mentioned that factors that influence the particle and/or pore size distribution in a given system are expected to influence straining. For example, solution chemistry determines colloid stability and hence the particle size distribution in suspension [Elimelech *et al.*, 1998]. Furthermore, the pore size distribution of porous media is reported to depend on solution chemistry as a result of dispersion and flocculation processes in fine-textured soils [Ayers and Westcot, 1989]. Solution pH can also alter the surface charge characteristics of colloids and porous media [e.g., Goldberg *et al.*, 1991] and is therefore anticipated to play a role in straining.

3.9. Hydrodynamics

[37] Mechanical filtration and straining are also known to be strong functions of the flow direction. The flow direction is commonly reversed in sand filters to unclog pores containing colloids and particles retained by mechanical filtration and straining. Silliman [1995] measured the transport behavior of a distribution of latex microspheres in various configurations of three coarse textured sands. Reversing the direction of water flow liberated large numbers of colloids in these systems. The detachment rate of attached colloids is not expected to depend on the direction of water flow. Hence all of these observations suggest that flow reversals can be used to deduce the occurrence and/or absence of straining. It should be noted, however, that the recovery efficiency upon flow reversal may be low due to reentrainment of colloids, shielding of straining sites by soil grains (flow stagnation zones), and/or the coupled influence of solution chemistry on straining.

[38] The deposition rate of colloids in porous media has frequently been reported to depend on the system hydrodynamics [Wang *et al.*, 1981; Tan *et al.*, 1994; Kretzschmar *et al.*, 1997; Compere *et al.*, 2001; Li *et al.*, 2005]. Filtration theory predicts that the attachment coefficient will increase with increasing pore water velocity. In contrast, several studies have shown that the value of the deposition coefficient decreased with increasing flow rate under unfavorable attachment conditions [Compere *et al.*, 2001; Li *et al.*, 2005]. These studies were presumably conducted on systems that were dominated by attachment, while hydrodynamic drag was postulated as a mechanism to release secondary-minimum associated colloids [Li *et al.*, 2005].

[39] It is also likely that straining will be a function of system hydrodynamics. Cushing and Lawler [1998] studied particle deposition using trajectory analysis on a porous media model composed of a face-centered cubic packing of spheres. Their analysis indicated that straining of particles at grain contact points can be an important mechanism of deposition due to hydrodynamic forces that funnel particle trajectories to these locations. Straining was predicted to decrease with increasing approach velocity. As an additional example, consider the influence of hydrodynamics on the effluent concentration curves and depositions profiles for 1 μm carboxyl modified latex microspheres (zeta potential

was -70 mV) in 150 μm Ottawa sand shown in Figures 7a and 7b, respectively. In particular, the transport experiments that were conducted using deionized water buffered to a pH of 10 (1.7 mM NaHCO_3 + 1.7 mM Na_2CO_3) and an ionic strength of 56 mM. As the Darcy velocity increases (IS = High, q = High) higher effluent concentrations occurred, and the deposition profiles became less hyperexponential than the experiment conducted at a lower velocity (IS = High, q = Low). As mentioned in the section above, deposition in these systems was strongly influenced by pore structure. Hence it is likely that the shape and magnitude of the deposition profiles for strained colloids is very sensitive to hydrodynamics. The degree of sensitivity is expected to depend on the size of the colloid and porous medium, and on the solution chemistry (a force balance in a pore). This is a topic of current research investigation.

[40] It is also possible that strained colloids could be slowly mobilized by hydrodynamic shearing forces. This could produce deposit profiles that are not hyperexponential, but rather nonmonotonic (exhibiting a peak at some distance downgradient from the source). Nonmonotonic deposition profiles have been reported to be a function of pore water velocity [Tong *et al.*, 2005; Bradford *et al.*, 2006b]. Additional research is warranted to more fully understand the role of hydrodynamics on straining and the development of deposition profiles.

3.10. Micromodel Observations

[41] Several studies have recently examined colloid transport and deposition processes at the pore scale using various microscopic techniques. This research has provided valuable insight on colloid retention at the air-water interface and soil-air-water contact line [Wan and Wilson, 1994a; Sirivithayapakorn and Keller, 2003b; Crist *et al.*, 2004; Wan and Tokunaga, 2005; Steenhuis *et al.*, 2005; Crist *et al.*, 2005], colloid size exclusion [Sirivithayapakorn and Keller, 2003a], and colloid dispersion [Auset and Keller, 2004]. Transport experiments were also recently conducted in a specially designed micromodel to examine the deposition behavior of the colloids and microorganisms in porous media under unfavorable attachment conditions. The micromodel consisted of a 0.2 cm thick by 2.0 cm wide by 7 cm long glass chamber (inside dimensions), with a glass tube (0.5 cm inside diameter) and septum assembly joined at both ends of the chamber. Details of the experimental setup are found in Bradford *et al.* [2005]. After completion of a transport experiment, the final deposition behavior of the fluorescent colloids and microorganisms was microscopically examined at several locations using a Leica DM IRB epifluorescent microscope (Leica Microsystems Inc., Bannockburn, IL 60015).

[42] Figure 8 presents several representative photos of *E. coli* O157:H7/pGFP strain 72 deposition in 150 μm quartz sand. These photos demonstrate that the fluorescent bacteria can be deposited at grain junctions due to straining. Additional photos were presented by Bradford *et al.* [2005] for 1 and 3 μm sulfate latex colloids in Ottawa sands. In Figure 8 photos, as well as those presented by Bradford *et al.* [2005], large numbers of colloids were deposited at straining sites. Aggregation of colloids in suspension cannot explain this behavior since the uniformity of the colloid size distribution was experimentally verified with a laser scattering particle size and distribution analyzer. These

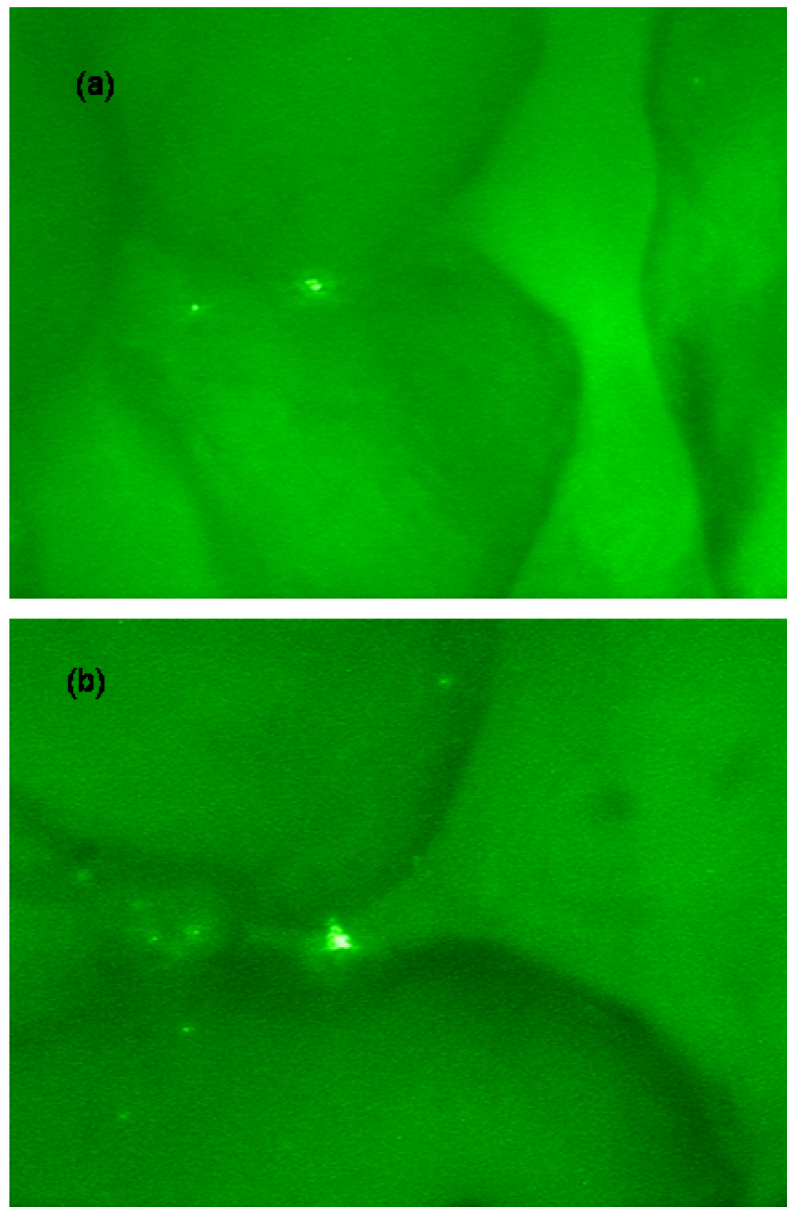


Figure 8. (a and b) Micromodel photos (magnified 600 times) of *E. coli* O157:H7 deposition in 150 μm sand.

observations suggest that colloid-colloid interactions may play an important role in straining behavior. We postulate that experimental conditions favoring attractive colloid-colloid interactions (colloids that are hydrophobic and/or have neutral surface charges) will enhance straining. In contrast, repulsive colloid-colloid interactions are anticipated to diminish straining.

3.11. Unsaturated Systems

[43] In unsaturated systems, the water and air distribution within a porous medium is controlled by capillary forces. If the porous medium is water wet, air will occupy the larger pores and water will be found in the smaller portions of the pore space, as well as in the form of films around solid grains adjacent to drained pores. As the water content of the porous medium decreases, the average size of water filled pores decreases, while the air-water interfacial area tends to increase [Bradford and Leij, 1997]. No published studies

have yet examined pore straining behavior in unsaturated systems. Straining is anticipated to be even more important in unsaturated than saturated systems because water flow is restricted to increasingly smaller pores at lower flow rates (less hydrodynamic shear), and hence a greater fraction of the water flows through smaller portions of the pore space. Conversely, an infiltrating water pulse causes water to flow through larger spaces, thereby potentially liberating strained colloids.

[44] Changes in water content can significantly influence colloid transport [Wan and Wilson, 1994b; Schafer et al., 1998; Sayers and Lenhart, 2003]. Decreases in the water content have been reported to lead to increasing colloid deposition in a porous medium. This behavior has been attributed to increased interfacial sorption of colloids onto the air-water interface [Wan and Wilson, 1994a, 1994b; Schafer et al., 1998; Sayers and Lenhart, 2003] or physical

restrictions imposed by thin water films (film straining) [Wan and Tokunaga, 1997]. Unsaturated colloid deposition profiles observed by Wan and Tokunaga [1997] tended to become more hyperexponential and greater in magnitude as the water content decreased, the sand size decreased, and the colloid size increased. These trends were similar to those observed in saturated systems for strained colloids [Bradford et al., 2002], and are consistent with our hypothesis of increased straining at lower water contents.

[45] Attachment to the air-water interface depends on the colloid's affinity for and accessibility to this interface, as well as on the magnitude of the interfacial area. Increased partitioning of colloids at the air-water interface is expected for greater interfacial areas. The air-water interfacial area is a function of the particle size distribution, water content, and wetting film continuity. For continuous water films, the air-water interfacial area tends to increase with decreasing water content [Bradford and Leij, 1997]. Although finer-textured soils are expected to have greater interfacial areas at a given water saturation, larger colloids may also be physically excluded from some of the interfaces due to film and/or pore straining. Similarly, the accessible solid surface area for attachment is expected to decrease with decreasing water content (equation (5)). Clearly, additional research is needed to quantify accessible air-water interfacial areas and surface areas for variously sized colloids in unsaturated systems.

3.12. Heterogeneous Systems

[46] Natural soil and aquifer systems contain layers and lenses of contrasting soil texture. Colloid transport across such textural interfaces has received relatively little research attention [Saiers et al., 1994; Silliman, 1995; Bradford et al., 2004, 2005]. Bradford et al. [2005] recently observed in saturated column experiments that the transport and deposition of colloids were highly dependent upon the textural interface. For a given sand and pore water velocity, deposition of colloids was always most pronounced at the sand (inlet) surface. Here colloids enter a new pore network and are more likely to encounter smaller pores or dead-end regions of the pore space that contribute to straining. Less deposition occurred at textural interfaces within the column than at the sand surface, presumably due to the fact that advection, dispersion and size exclusion tends to confine colloid transport to the larger pore networks and thus limit accessibility to straining sites (flow focusing). Increasing the textural contrast at an interface produced greater colloid deposition when water flowed from coarser to finer textured sands. Conversely, when water flowed from finer to coarser textured sands, little deposition occurred. We note that a pdf of colloid attachment coefficients cannot explain this observed deposition behavior. In this case, mobile colloids should possess a more uniform distribution of attachment coefficients as transport distance increases and therefore should not depend on the ordering of the sand layers.

[47] Physically heterogeneous systems will also exhibit a spatially and temporally dependent water flow field. In saturated systems water flow is controlled by the permeability distribution, with high flow rates occurring in more permeable zones. Conversely, the topology and geometry of the hydraulically active network will greatly depend on the degree of saturation in unsaturated systems (i.e., at low

water contents flow is restricted to finer pores). Hence a clear understanding of the role of hydrodynamics, the flow direction, flow focusing, and the water content on straining is needed to accurately characterize and predict colloid transport in heterogeneous systems. Further research is needed to adequately address these issues.

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