

# SCALING BACTERIAL FILTRATION RATES IN DIFFERENT SIZED POROUS MEDIA

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**ABSTRACT:** Aquifer sediments contain a wide distribution of particle sizes, but only a single collector diameter ( $d$ ) can be used in a filtration equation. To establish a method for selecting a characteristic  $d$  when media are composed of different sized particles, we measured bacterial retention in columns packed with either crushed quartz sand (separated into three different size ranges) or borosilicate glass beads. The best methods for choosing  $d$  were those that produced nearly constant collision efficiencies ( $\alpha$ 's). Characteristic diameters included:  $d_{10}$  (10% of all particles were smaller),  $d_{90}$  (90% of all particles were smaller),  $d_a$  (arithmetic mean), and  $d_g$  (geometric mean), where all diameters were calculated using number, area, and volume size distributions. Bacterial  $\alpha$ 's decreased in proportion to the distance traveled in the packed bed, and were scaled by the number of bacteria-sediment collisions using a dimensionless collision number ( $\xi$ ). These comparisons indicated that characteristic diameters based on the smaller particles ( $d_a$  and  $d_g$  using number distributions, and  $d_{10}$  using a volume distribution) most accurately described bacterial transport in the different-sized porous media.

## INTRODUCTION

The ability to predict bacterial transport is central to the study of a variety of subsurface processes, including microbial contamination of drinking water (Gerba 1985; Keswick 1984), and aquifer bioremediation (Wilson et al. 1986; Flathman et al. 1989). Although many aspects of bacterial transport are not completely understood, the use of clean-bed filtration theory (Yao et al. 1971; Rajagopalan and Tien 1976) has improved our understanding of factors that affect bacterial transport through ground-water aquifers (Harvey and Garabedian 1991).

Filtration models incorporate only a few soil and flow characteristics such as sediment size, porosity, and average fluid velocity to predict the collision frequency ( $\eta$ ) of particles such as bacteria with soil grains. Once particles collide with the soil grains, the probability of particles sticking to these grains is quantified in filtration models as the collision efficiency ( $\alpha$ ). The collision efficiency therefore incorporates all chemical interactions between the particle and media, and any inaccuracies assumed in the calculation of  $\eta$ .

Filtration theory has been verified in laboratory studies using uniform-sized, homogeneous porous media. Although filtration theory has been used to describe particle transport in soils and other media containing a distribution of particle sizes, there has been no methodological study performed to compare different methods for specifying the single collector diameter for the filtration equation when the media consists of many different collector sizes. Researchers have therefore used a va-

riety of different characteristic sizes, such as a weighted mean (McCaulou et al. 1995), median (Harvey et al. 1993; Harvey et al. 1995), or average grain size (Shonnard et al. 1994), or they have failed to state the method used to specify the collector size (Hornberger et al. 1992).

The purpose of the present study was to examine the effect of different choices of the characteristic collector size on predicting bacterial transport in media containing a distribution of grain sizes. The retention of radiolabeled bacteria was measured separately in two crushed quartz sediments having different size distributions, and in a third sediment prepared as an equal mixture (by weight) of the two quartz media. Several characteristic collector sizes were used in the filtration equation to represent the size distributions of the media including:  $d_{10}$  and  $d_{90}$ , the sizes for which 10% and 90% of all particles in the distribution are smaller;  $d_g$ , the geometric mean; and  $d_a$ , the arithmetic mean. All characteristic sizes of media were determined from length (average diameter), area, and volume distributions measured using an image analysis system. Values of  $\alpha$  calculated from bacterial retention were assumed to be equal since the media differed only in size and not surface chemistry. Thus, methods to characterize the collector size that produced similar values of  $\alpha$  for all four different sized media were defined as equally acceptable methods for characterizing the grain size distributions in terms of a single collector diameter.

Comparisons of  $\alpha$  were made for both low and high ionic strength solutions because in moderate to low ionic strength groundwater solutions  $\alpha$  is not constant even when the bacteria are derived from a monoclonal population (Albinger et al. 1994). In low ionic strength solutions, the distribution of bacteria-collector affinities produces mean values of  $\alpha$  that decrease with increasing transport distance even in well cleaned, homogeneous and uniform-sized media. It was hypothesized that by suspending cells in a high ionic strength solution that all cells would be completely destabilized. Therefore, bacterial transport in the different sized media was also examined using high ionic strength solutions (0.2 M CaCl<sub>2</sub>) in order to create conditions for constant  $\alpha$  values.

To compare the results from the different experiments on a similar basis,  $\alpha$  values were examined over the entire length of the packed bed based on a dimensionless collision number ( $\xi$ ) used to scale the transport length by the rate of bacteria-sediment collisions. This collision number was derived by normalizing the rate particles entered the column by the rate bacteria collided in the column at a concentration equal to the influent concentration.

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## Materials and Methods

### Bacterial Growth and Radiolabeling

The bacteria used for all experiments was *Pseudomonas fluorescens* strain P17 provided by C. P. Gerba (Department of Microbiology and Immunology, University of Arizona). The transport properties of P17 were established in previous studies (Kinoshita et al. 1993; Logan et al. 1993; Gross et al. 1995; Jewett et al. 1995). Cells were maintained by freezing 1 mL samples of P17 grown on glucose ( $0.25 \text{ g L}^{-1}$ ) in a morpholino-propane-sulfonate (MOPS)/mineral salts medium, consisting (per liter of deionized water) of the following: 4.624 g MOPS, 1.0 g  $\text{NH}_4\text{Cl}$ , 0.088 g  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ , 0.05 g  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.017 g  $\text{K}_2\text{HPO}_4$ , and 0.067 mg  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ . Frozen cultures were thawed, transferred to 5 mL of tryptic soy broth (Sigma Chemical Co.), and incubated to stationary growth at room temperature on a test-tube rotator. A sample from the liquid culture (100  $\mu\text{L}$ ) was transferred to MOPS/mineral salts medium (100 mL) in a 250 mL Erlenmeyer flask, and incubated at room temperature on a shaker table (150 rpm). Cells were harvested during late log growth ( $A_{600} \approx 0.100$ ) and diluted into 100 mL of artificial ground water (see below) to a final concentration of  $\sim 10^6$  cells/mL. Cells were radiolabeled by adding 40  $\mu\text{L}$  of  $^3\text{H}$ -Leucine (ICN, 79 Ci/mmol, 1 mCi/mL) to this suspension and incubating at room temperature for 8 h.

### Column Media

Column packings used to simulate aquifer sediments were either 40  $\mu\text{m}$  borosilicate glass beads (Whatman) or quartz sand (Unimin Corp.). The beads were cleaned by soaking in a 10%  $\text{H}_2\text{SO}_4$  solution, agitating on a shaker table (150 rpm) for 3 h, and rinsing with deionized water (Milli-Q, Millipore Corp.). Beads were dried overnight at  $105^\circ\text{C}$  and stored until use (Gross et al. 1995).

Different grain distributions of quartz media were prepared from stock quartz particles (Unimin Corp.) using a wet sedimentation technique (Litton and Olson 1993; Jewett 1995). Small particles were removed by flushing each quartz medium with tap water through the bottom of a 90 cm (length)  $\times$  7.5 cm (diameter) column at a constant flow that suspended all particles smaller than a chosen size based on the settling velocity. Overflow from the column was discarded. The desired size distribution was captured in the column effluent by re-fluidizing the bed at a higher flow rate, leaving larger particles in the column. Three different sizes of the quartz sand were separated in this manner, including: a large grain size ( $L$ ), a small grain size ( $S_1$ ), and a second small grain size distribution ( $S_2$ ) used only in larger column experiments (see the following). A fourth medium was produced using a 50:50 mixture by weight of  $L$  and  $S_1$ .

Quartz media were cleaned by soaking in 12 N HCl for 24 h (with periodic agitation), rinsing with deionized water, and cooking at  $810^\circ\text{C}$  for 8 h. Once cooled, the quartz was rehydrated by boiling in deionized water for 4 h, dried at  $105^\circ\text{C}$ , and stored until use (Litton and Olson 1993).

Projected areas of sediment were measured by light microscopy (Olympus BH-2) on an Image analysis system (Cue-2, Galai Inc.). The projected areas,  $A$ , were converted to equivalent diameters,  $d_{\text{eq}}$ , using  $A = \pi(d_{\text{eq}}/2)^2$ . Particle diameters were converted to size distributions based on 10  $\mu\text{m}$  increments [Fig. 1(a)]. Size distributions were converted to area and volume distributions, assuming spherical particles [Figs. 1(b) and 1(c), respectively]. These size distributions were used to calculate the four characteristic sizes of the sediment:  $d_{10}$ ,  $d_{90}$ ,  $d_g$ , and  $d_a$  (Table 1).

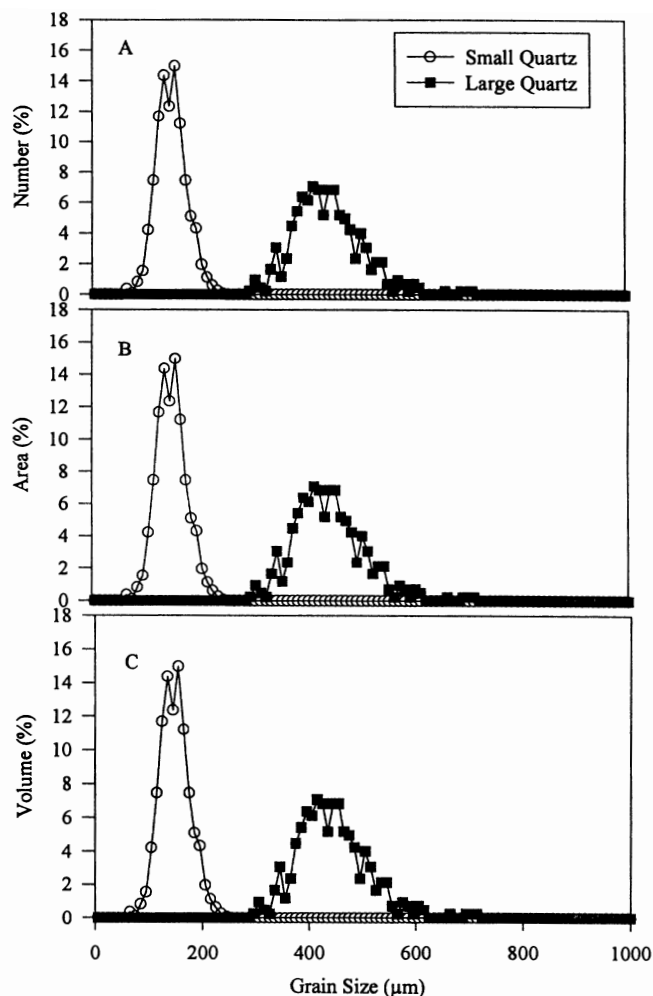


FIG. 1. Particle Size Distributions for Small Quartz ( $S_1$ ) and Large Quartz ( $L$ ) Based on Following: (a) Number of Particles; (b) Area of Particles; and (c) Volume of Particles

TABLE 1. Characteristic Sizes for Different Particle-Size Distributions

Column type (1)	Column media (2)	Media porosity $\epsilon$ (3)	Size range ( $\mu\text{m}$ ) (4)	Distribution type (5)	Characteristic Diameter ( $\mu\text{m}$ )			
					$d_{10}$ (6)	$d_{90}$ (7)	$d_g$ (8)	$d_a$ (9)
Mini	Small quartz	0.38	60-240	Number	109	182	149	146
				Area	118	192	159	157
				Volume	123	197	164	162
	Large quartz	0.37	290-715	Number	364	520	442	438
				Area	377	542	462	457
				Volume	384	567	473	468
Quartz mixture	0.33	60-715	Number	109	204	168	157	
			Area	126	476	267	232	
			Volume	140	508	343	306	
Large	Small quartz	0.36	60-280	Number	106	209	156	150
				Area	120	234	178	167
				Volume	128	250	189	184

### Column Experiments

Two types of column experiments were performed: mini-column experiments ( $3 \text{ cm}^3$  and  $10 \text{ cm}^3$  syringes) run according to the Microbe and Radiolabel Kinesis (MARK) method (Gross et al. 1995) as modified by Albinger et al. (1994), and large column experiments performed as described in Jewett (1995). Only the  $10 \text{ cm}^3$  minicolumns packed with quartz media were used to compare different methods for selecting a characteristic collector diameter. Both large columns filled with quartz media and minicolumns ( $3 \text{ cm}^3$  syringes) filled

with glass beads were used to examine the variability in  $\alpha$  over large ranges of collision numbers.

Bacteria were suspended in two different ionic strength solutions, an artificial ground water (low ionic strength) and a 0.2 M CaCl<sub>2</sub> solution (high ionic strength). Artificial ground water for the minicolumns consisted (per liter of deionized water) of the following: 0.069 g MgSO<sub>4</sub>·7H<sub>2</sub>O, 0.050 g NaHCO<sub>3</sub>, 0.00145 g CaCl<sub>2</sub>·2H<sub>2</sub>O, 0.064 g Ca(NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O, and 0.002 g KF. HCl (0.01N) was added to produce a final pH of 8 and ionic strength of  $3.6 \times 10^{-3}$  M. The pH and ionic strength of the 0.2 M CaCl<sub>2</sub> solution were 6.8 and  $6.0 \times 10^{-2}$  M, respectively. Although the final pH and ionic strength of the artificial ground water for the large column experiments were the same as the artificial ground water used in the minicolumns (pH = 8.1 and  $I = 4.4 \times 10^{-3}$  M, respectively), a slightly different ground water was used, consisting (per liter of deionized water) of the following: 0.006 g KNO<sub>3</sub>, 0.138 g MgSO<sub>4</sub>·7H<sub>2</sub>O, 0.048 g CaSO<sub>4</sub>·2H<sub>2</sub>O, 0.019 g NaCl, and 0.047 g NaHCO<sub>3</sub>.

Radiolabeled bacterial suspensions (50 mL) used in the large column experiments were filtered through a 0.2  $\mu$ m syringe filter (Supor Acrodisc, Gelman Scientific, Inc.) to remove unassimilated radiolabel. The filter was rinsed with 10 mL of artificial ground water, reversed, and the cells pushed off the filter using either the low or high ionic strength solutions. This procedure was performed twice to obtain 100 mL of the final radiolabeled bacterial suspension. This procedure was also used to resuspend the bacteria in the 0.2 M CaCl<sub>2</sub> solution for the minicolumn experiments.

The average projected area of cells was obtained using an epifluorescence microscope and the Image Analysis system. Cells were stained by an acridine-orange procedure (Hobbie et al. 1977). Projected areas were converted to equivalent diameter [ $A = \pi(d_{eq}/2)^2$ ]. Equivalent cell diameters decreased slightly from 1.0  $\mu$ m ( $\pm 0.23$   $\mu$ m,  $n = 472$ ) to 0.8  $\mu$ m ( $\pm 0.14$   $\mu$ m,  $n = 106$ ) as a result of the syringe filtration step used to remove unassimilated radiolabel.

**Minicolumn Experiments.** The larger (10 cm<sup>3</sup>) minicolumns [inside diameter (ID) 1.3 cm] were filled with quartz media (small quartz, large quartz, or the quartz mixture) supported by a GF/F filter (Whatman, 0.7  $\mu$ m nominal pore size). The smaller (3 cm<sup>3</sup>) minicolumns (ID 0.8 cm) were filled with glass beads supported by a GF/D filter (Whatman, 2.7  $\mu$ m nominal pore size). Preweighed amounts of media (1.5 g glass beads, 6 g small quartz, 12 g large quartz, or 10 g quartz mixture) were added to the minicolumns by mixing in deionized water, pouring the mixture into the column, and stirring the media with a Pasteur pipette to remove entrapped air. Columns were held on a vacuum manifold (Alltech) equipped with Luerlok connections.

To equilibrate the column media with groundwater solutions, 2 mL (3 cm<sup>3</sup> columns) or 6 mL (10 cm<sup>3</sup> columns) of test solution (either artificial groundwater or 0.2 M CaCl<sub>2</sub>) were pulled through the column and an approach velocity of  $0.9-1.4 \times 10^{-3}$  m/s selected by setting the vacuum pressure. The bacterial suspension (2 mL, 3 cm<sup>3</sup> columns; 6 mL, 10 cm<sup>3</sup> columns) was then pulled through the column, and the column was rinsed with either 4 mL (3 cm<sup>3</sup> columns) or 12 mL (10 cm<sup>3</sup> columns) of the appropriate solution, to flush out unattached cells.

The medium was removed from the column by cutting off the end of the column, and extruding it using the syringe plunger. Beginning at the top of the column, while the medium was extruded it was sliced in ~1 mm (small quartz and glass beads), ~6 mm (large quartz), or 2 mm (quartz mixture) increments. Slicing intervals were selected to produce slices with similar collision numbers. Slices were transferred to pre-

weighed scintillation vials and weighed to determine the exact length of each slice ( $L_i$ ).

**Large Column Experiments.** The experimental protocol for large column experiments was described in detail elsewhere (Jewett 1995). Briefly, columns (diameter, 2.6 cm; length, 12.5 cm) were packed with small quartz ( $S_2$ ) media to a depth of approximately 10.5 cm. Column inflow was controlled in a downward direction at  $4.5-4.8 \times 10^{-5}$  m/s with pediatric infusion pumps (AVI Micro 210A, 3M Health Care). A uniform water flow was established, and one pore volume (approximately 20 mL) of radiolabeled cells was pulled through the column, followed by three pore volumes of artificial ground water (low ionic strength experiments) or 0.2 M CaCl<sub>2</sub> (high ionic strength experiments). The column packing was removed, and the porous media was extruded in 1 cm sections, weighed, and transferred to scintillation vials as described previously.

**Cell Retention.** Scintillation vials containing quartz or glass beads were filled with 10 mL of cocktail (Ecolite, ICN Biomedicals, Inc.), agitated for 18 h, and analyzed on a Beckman LS 3801 scintillation counter with quench correction. The number of bacteria retained in each slice ( $N_i$ ) was determined from the mass of radiolabel retained in the slice, corrected for unassimilated radiolabel as previously described (Gross et al. 1995). The number of bacteria added to the column ( $N_0$ ) was estimated by radiolabel counts from total and filtered (0.2  $\mu$ m polycarbonate filters, Poretics Corp.) bacterial suspension (2 mL). The fraction of bacteria retained in each slice ( $R_i$ ) was calculated as

$$R_i = \frac{N_i}{\left(N_0 - \sum N_{i-1}\right)} \quad (1)$$

where  $\sum N_{i-1}$  = sum of mass of bacteria retained in previous slices. The fraction of bacteria that penetrated the column through the length of each slice ( $C_i/C_0$ ) is therefore

$$\frac{C_i}{C_0} = 1 - \frac{\sum N_{i-1} + N_i}{N_0} \quad (2)$$

where  $C_i$  = liquid phase concentration of bacteria at end of each slice; and  $C_0$  = influent concentration of bacteria.

## THEORY

### Filtration Equation

The fraction of bacteria remaining in the liquid phase after transport through a column of length ( $L$ ) of porous media can be calculated from the one-dimensional filtration equation (Yao et al. 1971) as

$$\frac{C}{C_0} = \exp(-\alpha\lambda L) \quad (3)$$

where  $C_0$  and  $C$  = concentration of particles entering and leaving column; and  $\lambda$  = filter coefficient, calculated as

$$\lambda = \frac{3(1-\epsilon)\eta}{2d} \quad (4)$$

where  $d$  = collector diameter;  $\epsilon$  = media porosity; and  $\eta$  = collector efficiency. Since  $C_i/C_{i-1} = (1 - R_i)$ , the collision efficiency in each slice of column of thickness  $L_i$  can be calculated by rearranging (3) in terms of  $\alpha_i$  as

$$\alpha_i = \frac{2}{3} \frac{d}{(1-\epsilon)\eta L_i} \ln(1 - R_i) \quad (5)$$

The collector efficiency was calculated using the Rajagopalan

and Tien (RT) (Rajagopalan and Tien 1976; Logan et al. 1995) model

$$\eta = 4A_s^{1/3}N_{pe}^{-2/3} + A_sN_{Lo}^{1/8}N_R^{15/8} + 0.00338A_sN_G^{1/2}N_R^{-0.4} \quad (6)$$

where  $A_s$ ,  $N_{pe}$ ,  $N_{Lo}$ ,  $N_R$ , and  $N_G$  = dimensionless numbers that account for effects of neighboring particles, diffusion, London-van der Waals forces, interception, and sedimentation on particle collisions, respectively. These quantities were calculated by

$$A_s = \frac{2(1 - \gamma^5)}{2 - 3\gamma + 3\gamma^5 - 2\gamma^6}; N_R = \frac{d_p}{d}; N_{pe} = \frac{3\mu\pi U d d_p}{kT} \quad (7-9)$$

$$N_G = \frac{g(\rho_p - \rho)d_p^2}{18\mu U}; N_{Lo} = \frac{4H}{9\pi\mu d_p^2 U} \quad (10, 11)$$

where  $\gamma = (1 - \epsilon)^{1/3}$ ;  $H$  = Hamaker constant (assumed here to be  $10^{-20}$  J);  $\mu$  = dynamic fluid viscosity ( $8.94 \times 10^{-4}$  Ns/m<sup>2</sup>);  $d_p$  = suspended particle diameter;  $U$  = superficial fluid velocity;  $d$  = collector diameter;  $\rho_p$  = suspended particle density (1,070 kg/m<sup>3</sup>);  $\rho_f$  = fluid density (997.1 kg/m<sup>3</sup>);  $k$  = Boltzmann's constant ( $1.38 \times 10^{-23}$  kg-m<sup>2</sup>/s<sup>2</sup>-K); and  $T$  = fluid temperature (298 K).

### Scaling Particle Removal Using Dimensionless Collision Number

Comparing bacterial filtration rates in media with different sized particles is difficult for several reasons. Even in two porous media that differ only in the collector size, removal rates of colloids varies in proportion to  $\eta/d$  and so removal rates will appear different when scaled by the distance traveled in the packed column. For the size range of typical soil particles, bacteria will collide more frequently with smaller than with larger soil particles, and will therefore be removed faster by smaller than larger soil grains. It was not known, however, if this generalization would be valid when media of different sizes were mixed together. Measuring bacterial filtration rates is further complicated by the fact that bacteria within a monoclonal population have a range of sticking coefficients (Albinger et al. 1994). The average  $\alpha$  of bacteria introduced into a column will therefore decrease with travel distance since the cells with higher  $\alpha$ 's are removed faster than cells with lower  $\alpha$ 's. As a result, measured  $\alpha$ 's decrease with travel distance in the column. It is therefore not possible to directly compare bacterial removal from columns on the basis of a characteristic collector size in columns filled with different sized media without placing travel distances on a common basis.

It is proposed here that bacterial removal rates measured in columns with different collector sizes can be directly compared on the basis of a dimensionless number, defined as the collision number,  $\xi$ , calculated as

$$\xi = \frac{\text{rate particles collide in a column}}{\text{rate particles enter a column}} \quad (12)$$

where both rates are evaluated on the common basis of the influent particle concentration. The rate particles enter a column of cross-sectional area  $A$  is  $UC_0A$ . The rate particles collide in a column can either be calculated from a mass balance (Logan et al. 1995) or can be calculated by recognizing that the removal rate of particles is equal to the absolute value of the product of the collision rate and  $\alpha$ . Particles are removed in the column at a rate,  $dc/dt$ , obtained from the chain rule as

$$\frac{dc}{dt} = \frac{dz}{dt} \frac{dc}{dz} \quad (13)$$

where  $dz/dt$  = fluid velocity,  $U$ . The instantaneous removal rate

at the column entrance where  $C = C_0$  is calculated by taking the derivative of (3), producing  $dc/dz = -\alpha\lambda C_0$ . The collision rate is calculated as the absolute value of the product of the fluid velocity and the removal rate,  $dc/dz$ , divided by  $\alpha$ , or

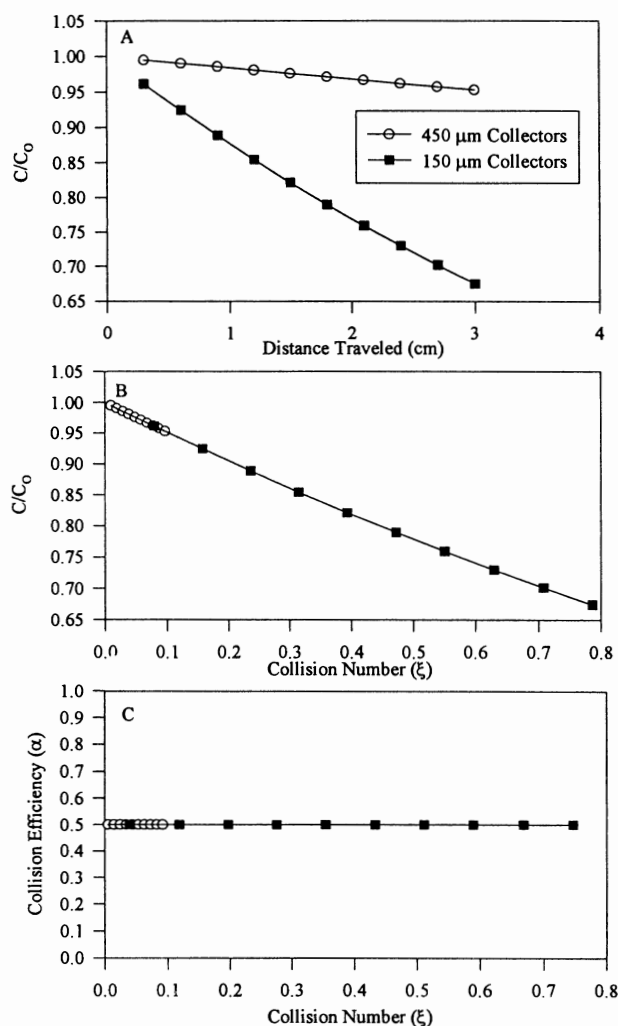
$$\frac{dc}{dt} = U \left| \frac{-\alpha\lambda C_0}{\alpha} \right| = U\lambda C_0 \quad (14)$$

Using these results in (12) for a column of volume  $V = AL$ , the collision number is

$$\xi = \frac{U\lambda C_0 AL}{UC_0 A} = \lambda L \quad (15)$$

Thus,  $\xi$  can be seen to be a ratio of the column length to a characteristic travel distance ( $1/\lambda$ ).

The utility of the collision number for scaling columns containing different sized media can be seen by comparing the fraction of bacteria penetrating two columns filled with 150  $\mu\text{m}$  and 450  $\mu\text{m}$  collectors. As shown in Fig. 2(a), assuming  $\alpha = 0.5$ ,  $U = 1.0 \times 10^{-3}$  m/s,  $d_p = 1 \mu\text{m}$ , and  $\epsilon = 0.37$ , only 5% of particles were removed within 3 cm in a column filled with 450  $\mu\text{m}$  collectors, versus 30% in a column filled with 150  $\mu\text{m}$  collectors. Because  $\alpha$  is constant, the difference in

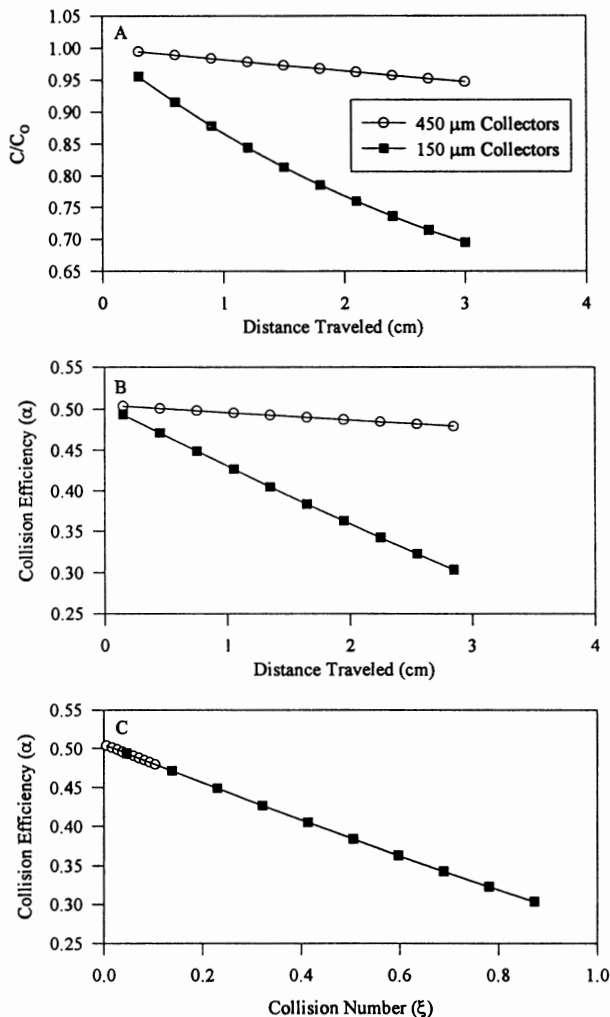


**FIG. 2. Theoretical Transport of Particles through Columns with 150  $\mu\text{m}$  Collectors Based on Filtration Model with All Particles Having  $\alpha = 0.5$ : (a) Fraction of Bacteria Penetrating Columns As Function of Transport Distance; (b) Fraction of Bacteria Penetrating Columns As Function of Collision Number ( $\xi$ ); (c) Collision Efficiency ( $\alpha$ ) As Function of Collision Number ( $\xi$ ) (Calculations Were Made Assuming  $U = 1 \times 10^{-3}$  m/s,  $\epsilon = 0.37$ , and  $d_p = 1 \mu\text{m}$ )**

the penetration is due to a higher rate of particle-collector collisions in the 150  $\mu\text{m}$  collector diameter column than in the 450  $\mu\text{m}$  collector diameter column. Utilization of the collision number to scale the column length results in identical penetration for the two different collector sizes [Fig. 2(b)]. Since  $\alpha$  was assumed to be constant, values of  $\alpha$  were also equivalent over the same number of collisions [Fig. 2(c)].

The collision number can also be used to scale removal rates in columns for particles with a distribution of collision efficiencies. For example, the fraction of particles penetrating columns with either 150  $\mu\text{m}$  or 450  $\mu\text{m}$  collectors can be calculated for a sample containing particles with two different collision efficiencies (50% of the influent particles have an  $\alpha$  equal to 1 and 50% have an  $\alpha$  equal to 0.01) [Fig. 3(a)]. Mean values of  $\alpha$  for each slice of the column were calculated using the filtration model [Fig. 3(b)]. Mean values of  $\alpha$  that would be measured in each slice decrease to 0.30 after 3 cm of transport distance in the column with the 150  $\mu\text{m}$  collectors, versus a decrease to 0.49 in the column with the 450  $\mu\text{m}$  collectors. The increase in the rate at which  $\alpha$  decreases is due to the increased collision rate with the 150  $\mu\text{m}$  collectors. However, by comparing  $\alpha$  on the basis of  $\xi$  instead of  $L$ , the results for both columns collapse onto the same line [Fig. 3(c)].

The magnitude of the collision number is also a useful pa-



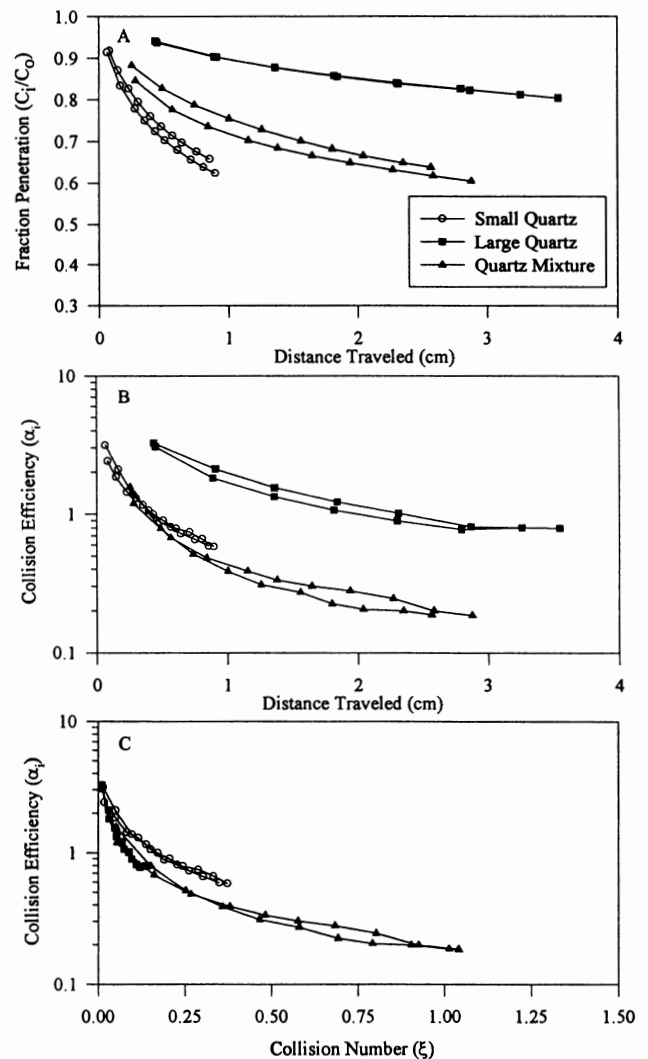
**FIG. 3. Theoretical Transport of Particles through Columns with 150  $\mu\text{m}$  and 450  $\mu\text{m}$  Collectors Based on Filtration Model with 50% of Influent Particles Having  $\alpha = 1.0$  and 50% Having  $\alpha = 0.01$ : (a) Fraction of Bacteria Penetrating Columns As Function of Transport Distance; (b) Mean  $\alpha$  per Slice of Column As Function of Transport Distance; (c) Mean  $\alpha$  per Slice of Column As Function of Collision Number ( $\xi$ ); (Calculations Were Made Assuming  $U = 1 \times 10^{-3}$  m/s,  $\epsilon = 0.37$ , and  $d_p = 1 \mu\text{m}$ )**

rameter that indicates the number of collisions a nonattaching ( $\alpha = 0$ ) particle must undergo to travel through a column of length  $L$ . A collision number of unity, for example, indicates that a distance,  $L = 1/\lambda$ , is the distance non-attaching cells will collide once, on average, with column media.

## RESULTS

### Bacterial Transport in Low Ionic Strength Ground Water

Bacterial transport in the low ionic strength ( $3.6 \times 10^{-3}$  M) groundwater increased with collector size as predicted by theory [Fig. 4(a)]. After 1 cm of transport distance approximately 90% of the bacteria penetrated columns with the large quartz collectors, versus 60% and 75% in columns with the small quartz and the quartz mixture, respectively. When the filtration model (5) was used to calculate collision efficiencies ( $\alpha_i$ ) for each slice of the columns, values of  $\alpha_i$  decreased with increasing transport distance [Fig. 4(b)]. The change in  $\alpha_i$  values for the different characteristic collector diameters is shown in Table 2. The dependence of  $\alpha_i$  on transport distance was similar in the quartz mixture and the small quartz columns. Values



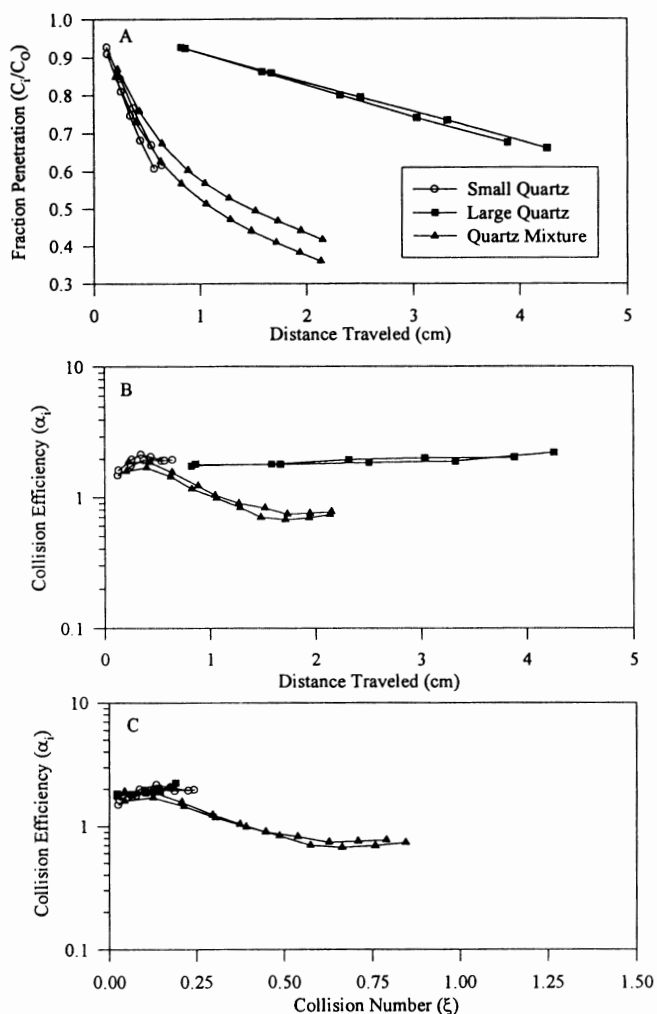
**FIG. 4. Results from Minicolumns ( $10 \text{ cm}^3$ ) with Low Ionic Strength Carrier Solution: (a) Fraction of Bacteria Penetrating Column As Function of Transport Distance; (b) Collision Efficiency ( $\alpha_i$ ) As Function of Transport Distance; (c) Collision Efficiency ( $\alpha_i$ ) As Function of Collision Number ( $\xi$ ) (All Calculations Were Made with  $d_{10}$  of Volume Distribution and Column Media Included Small Quartz (S), Large Quartz (L), and Quartz Mixture)**

**TABLE 2. Ranges of  $\alpha$  Calculated Using Different Characteristic Collector Sizes for Low Ionic Strength Experiments**

Distribution type (1)	Column media (2)	Range of Collision Efficiency ( $\alpha$ )			
		$d_{10}$ (3)	$d_{60}$ (4)	$d_s$ (5)	$d_g$ (6)
Number	Small quartz	2.4-0.45	7.1-1.3	4.7-0.88	4.5-0.84
	Large quartz	3.0-0.72	5.7-2.4	4.2-1.0	4.2-1.0
	Mix of quartz	0.88-0.11	3.5-0.94	2.3-0.63	2.0-0.24
Area	Small quartz	2.9-0.54	7.9-1.5	5.4-1.0	5.3-0.98
	Large quartz	3.2-0.77	6.1-1.5	4.6-1.1	4.5-1.1
	Mix of quartz	1.2-0.34	19-2.1	6.1-0.71	4.6-0.54
Volume	Small quartz	3.1-0.59	8.3-1.6	5.8-1.1	5.6-1.0
	Large quartz	3.3-0.79	6.6-1.6	4.8-1.2	4.7-1.1
	Mix of quartz	1.6-0.18	21-5.5	10-1.2	8.0-0.93

**TABLE 3. Ranges of  $\alpha$  Calculated Using Different Characteristic Collector Sizes for High Ionic Strength Experiments**

Distribution type (1)	Column media (2)	Range of Collision Efficiency ( $\alpha$ )			
		$d_{10}$ (3)	$d_{60}$ (4)	$d_s$ (5)	$d_g$ (6)
Number	Small quartz	1.2-1.7	3.2-4.2	2.2-3.2	2.1-3.0
	Large quartz	1.6-1.8	3.0-3.8	2.3-2.9	2.2-2.8
	Mix of quartz	1.1-0.26	4.0-1.6	2.8-1.1	2.4-0.93
Area	Small quartz	1.4-2.0	3.6-5.1	2.5-3.6	2.4-3.5
	Large quartz	1.7-2.2	3.2-4.1	2.5-3.1	2.4-3.1
	Mix of quartz	1.5-0.59	19-7.5	6.7-2.6	5.2-2.0
Volume	Small quartz	1.5-2.2	3.7-5.4	2.6-3.8	2.6-3.7
	Large quartz	1.8-2.2	3.5-4.5	2.6-3.2	2.5-3.2
	Mix of quartz	1.9-0.74	22-8.4	11-4.2	8.6-3.4



**FIG. 5. Results from Minicolumns (10 cm<sup>3</sup>) with High Ionic Strength Carrier Solution: (a) Fraction of Bacteria Penetrating Column As Function of Transport Distance; (b) Collision Efficiency ( $\alpha$ ) As Function of Transport Distance; (c) Collision Efficiency ( $\alpha$ ) As Function of Collision Number ( $\xi$ ) (All Calculations Were Made with  $d_{10}$  of Volume Distribution and Column Media Included Small Quartz (S), Large Quartz (L), and Quartz Mixture)**

of  $\alpha_i$  decreased from 3.0 to 0.6 over a distance of 1 cm in the columns filled with the small quartz media, and from 1.5 to 0.2 over a distance of 2.9 cm in the columns filled with the quartz mixture. Values of  $\alpha_i$  in the large quartz columns were substantially higher, decreasing from 3.3 to 0.8 over a transport distance of 3.5 cm.

When  $\alpha_i$  values were scaled by the rate of bacteria-sediment collisions using the collision number ( $\xi$ ), measured decreases in  $\alpha_i$  were similar for the three sizes of media [Fig. 4(c)].

Overall, values of  $\alpha_i$  decreased an order of magnitude ( $\sim 3$  to 0.2) after  $\sim 1\xi$ .

### Bacterial Transport in High Ionic Strength Ground Water

To achieve more constant bacterial  $\alpha$ 's, the column experiments were repeated using a high ionic strength ground water. As expected, the high ionic strength ( $6.0 \times 10^{-2}$  M) solution decreased total bacterial penetration relative to the low ionic strength groundwater [Fig. 5(a)]. Bacterial concentrations decreased by approximately 5% after 0.5 cm of transport distance in columns with the small quartz, 10% after 3.5 cm in columns with the large quartz, and 25% after 2 cm in columns with the quartz mixture. Collision efficiencies (5) calculated in large and small quartz columns were constant, with  $\alpha_i = 1.9$  [ $\pm 0.2$  SD, Fig. 5(b)] when  $d_{10}$  of the volume distribution was used as the characteristic collector diameter. Constant values of  $\alpha_i$  were always observed in the columns with the small and large quartz media regardless of choice of collector size (Table 3). In the columns with the quartz mixture,  $\alpha_i$  decreased by a factor of 0.35. However, this overall decrease is much less than the overall decrease of a factor of 0.07 observed in the low ionic strength experiments.

Scaling column length by the number of collisions did not affect the overall trends in  $\alpha_i$  [Fig. 6(c)]. Values of  $\alpha_i$  were still constant in the columns with the large and small quartz media, while  $\alpha_i$  decreased only slightly in the columns filled with the quartz mixture. However,  $\alpha$  values for different columns were similar when compared on the basis of  $\xi$ , if a more appropriate choice of a characteristic collector size was used (see the following).

### Collision Efficiencies Measured over Large Range of Collision Numbers

To see whether trends observed in the minicolumns would scale over larger transport distances,  $\alpha$ 's were measured over a larger range of collision numbers ( $\xi \leq 18$ ) using larger columns (instead of minicolumns) filled with the small quartz media and minicolumns filled with small diameter glass beads. When  $\alpha_i$  values were scaled by the collision number,  $\alpha_i$ 's for these different systems were nearly equal for  $\xi \leq 18$ , and indicated the same trend of a decreasing  $\alpha$  with  $\xi$  observed in the minicolumns with mixed quartz media at smaller collision numbers (Fig. 6).

Overall, values of  $\alpha_i$  decreased two orders of magnitude (1.5 to 0.01) for bacteria suspended in the low ionic strength artificial groundwater [Fig. 6(a)]. At lower collision numbers ( $\xi < 1$ ; quartz mixture),  $\alpha_i$  decreased an order of magnitude (from 1.5 to approximately 0.2). Similar order of magnitude reductions of  $\alpha$  were observed in the glass bead minicolumns ( $\alpha_i$  decreased from 0.4 to 0.03 for  $0.2 < \xi < 5$ ) and the large columns filled with smaller quartz media ( $\alpha_i$  decreased from 0.2 to 0.01 for  $1 < \xi < 18$ ).

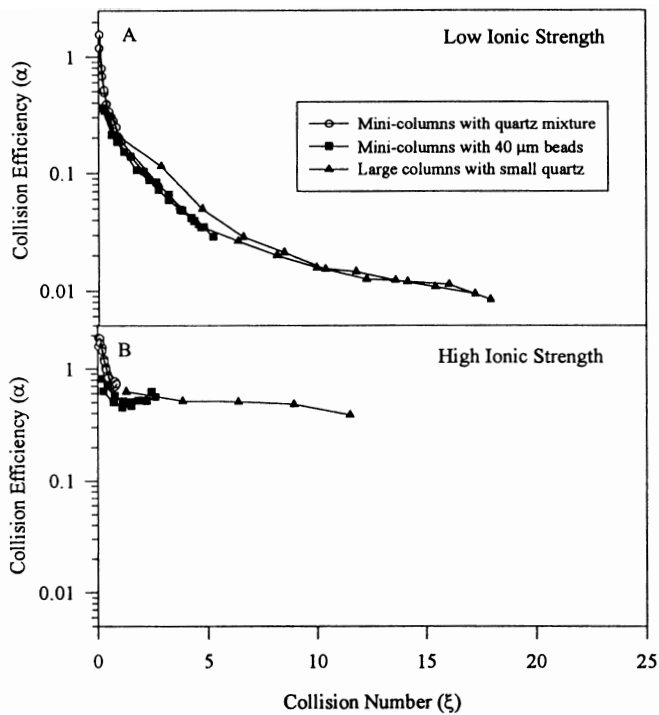


FIG. 6. Collision Efficiency ( $\alpha$ ) As Function of Collision Number ( $\xi$ ) for Following: (a) Low Ionic Strength ( $\sim 4 \times 10^{-3}$  M) Carrying Solution; (b) High Ionic Strength ( $6 \times 10^{-1}$  M) Carrying Solution [Experiments Were Performed in Minicolumns ( $10 \text{ cm}^3$ ) Filled with Quartz Mixture, Large Columns with Small Quartz ( $S_2$ ) and Minicolumns ( $3 \text{ cm}^3$ ) with  $40 \mu\text{m}$  Glass Beads and Calculations for Columns Filled with Quartz Mixture and Large Columns Were Made with  $d_{10}$  of Volume Distribution]

Higher values of  $\alpha_i$  were produced by increasing the ionic strength, as expected from previous research (Gross and Logan 1995; Jewett et al. 1995). The high ionic strength solution also produced more nearly constant values of  $\alpha$  over larger transport distances than observed in the low ionic strength experiments [Fig. 6(b)]. For  $\xi$  ranging from 1 to 10 in the large columns,  $\alpha_i$  for bacteria suspended in high ionic strength solutions was constant at approximately 0.6. Collision numbers are not shown in Fig. 6(b) for  $\xi > 10$  because the high ionic strength solution removed too many bacteria to permit accurate determination of  $\alpha_i$  at these larger travel distances. In the glass bead minicolumns  $\alpha_i$  only slightly decreased from 0.8 to 0.5 ( $0.2 < \xi < 3$ ). In the quartz mixture minicolumns, the decrease in  $\alpha_i$  was slightly larger, with  $\alpha_i$  decreasing from 2 to about 0.7 at low collision numbers ( $\xi < 1$ ) at high ionic strengths. However, this decrease was small compared to the order-of-magnitude decreases observed in low ionic strength experiments for this media over the same range of collision numbers.

### Choice of Characteristic Collector Size

Because of the large decrease in  $\alpha_i$  values with transport distance when bacteria were suspended in the low ionic strength solution, the characteristic collector size was determined from the experiments performed with the high ionic strength solution with nearly constant values of  $\alpha$  for small values of  $\xi$ . The constant portion of the  $\alpha$  versus  $\xi$  curves were used to visually compare the different methods of describing the distributions of collector size by a single value. Characterizing the sediment using  $d_a$  or  $d_g$  of the number distribution, or  $d_{10}$  of the volume distribution resulted in similar values of  $\alpha$  for the different media size distributions (Fig. 7). Therefore, these three characteristic diameters were considered

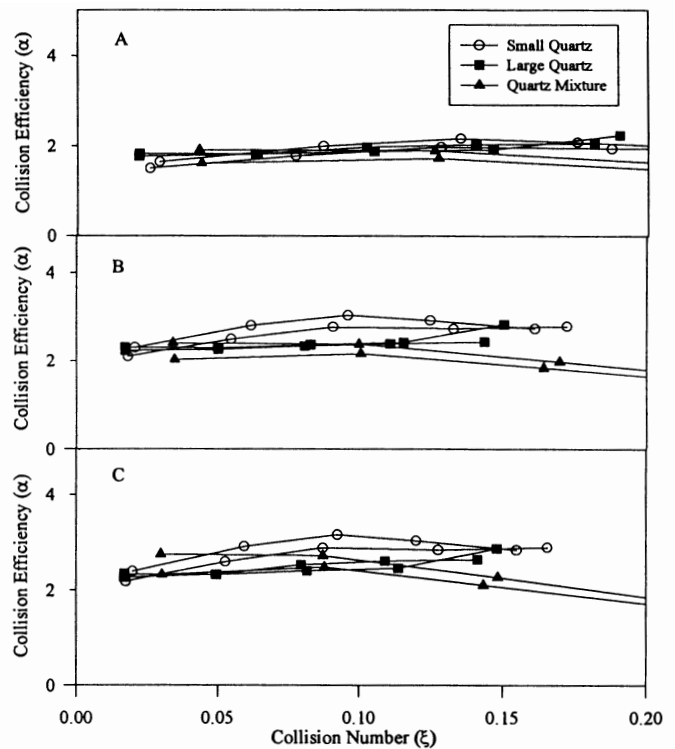


FIG. 7. Collision Efficiency ( $\alpha$ ) As Function of Collision Number ( $\xi$ ) Calculated with Following: (a)  $d_{10}$  of Volume Distribution; (b)  $d_g$  of Number Distribution; and (c)  $d_a$  of Number Distribution (Columns Were Performed with Bacteria Suspended in 0.2 M  $\text{CaCl}_2$  and Media Included Small Quartz ( $S_1$ ), Large Quartz ( $L$ ), and Quartz Mixture)

to be the best choice for representing different media size distributions.

### ANALYSIS

The results comparing collision efficiencies of bacteria using different characteristic diameters to represent media with a distribution of grain sizes indicated that the most consistent values of  $\alpha$  were obtained for methods that emphasized smaller particles in the size distribution. Porous media can therefore be characterized for use in a filtration equation using either  $d_a$  or  $d_g$  from a number distribution, or  $d_{10}$  from a volume size distribution. Since the media used in the present study had constant density, the volume distributions examined here would be equivalent to mass distributions produced by others to determine mean (McCaulou et al. 1995) and median grain sizes (Harvey et al. 1995) of the porous media using sieve analyses. Values of  $\alpha$  for  $d \equiv d_{10}$  were closer to (but in some cases still greater than) its theoretical maximum of unity suggesting that  $d_{10}$  (based on a volume size distribution) is the best characteristic diameter for filtration calculations for heterogeneous media.

When larger characteristic media diameters were used in our experiments to characterize a grain size distribution, for example  $d_a$  or  $d_g$  of the volume distribution, we found that bacterial collision efficiencies for the columns with the quartz mixture were at least two times as large as collision efficiencies for the columns with the small or large quartz media (Table 3). Choosing too large a collector size results in predicting too few collisions between bacteria and the media, and underestimates the potential for bacterial removal by filtration. The three acceptable methods for defining a single characteristic collector size, of the 12 different methods examined, were therefore those that emphasized the role of the smaller particles in the porous media size distribution.

Maximum collision efficiencies of  $\alpha$  in the range of 2 to 3, calculated at small values of  $\xi$ , were always larger than the theoretical maximum value of  $\alpha = 1$ . Decreases in  $\alpha$  were probably produced by removal of stickier microbes near the column entrance, resulting in a lower overall  $\alpha$  for bacteria that penetrated further into the column (Albinger et al. 1994). Calculated values of  $\alpha > 1$  may have been due to under prediction of  $\eta$  due to surface roughness (Harvey and Garabedian 1991) or a greater effective diameter of bacteria than measured here due to appendages or polymeric coatings (Kinoshita et al. 1993). Straining of bacteria can produce  $\alpha > 1$  for  $d_p/d > 0.2$  (Tien and Payatakes 1979), but straining was not considered to be a factor since  $d_p/d$  was always less than 0.2 in our experiments. Collision efficiencies greater than 1 have been observed by others even for inorganic colloids (Logan et al. 1995), suggesting that collision frequencies are underestimated by the RT filtration equation.

Other researchers have used more uniform sized collectors in experiments to study bacterial transport in porous media (Jewett et al. 1995; Kinoshita et al. 1993; Martin et al. 1992). For highly uniform sediment distributions, the method of calculating a characteristic collector size is relatively unimportant since mean, average and other characteristic diameters are essentially equal. In our experiments the choice of the characteristic collector size was therefore the most critical for the bimodal quartz distribution. The range in  $\alpha$  was much larger for the quartz mixture with a broad size distribution than it was for the small and large quartz media which had narrower size distributions (Table 3).

Because measured bacterial collision efficiencies varied by orders of magnitude in low ionic strength experiments, the comparisons of characteristic collector sizes made on the basis of bacterial  $\alpha$ 's different sized media were only possible when the travel distances were nondimensionalized using a collision number. This suggests that in future colloid and bacterial filtration studies it may be important to consider the number of collisions occurring in filtration tests used to determine average  $\alpha$ 's. From our studies it appears that columns with the potential for fewer collisions, or those columns with lower collision numbers based on total column length, will have higher measured  $\alpha$ 's than longer columns. If we extend such observations to interpret differences observed between laboratory studies (which typically have low collision numbers) and field tests (with high collision numbers) we would predict that lower  $\alpha$ 's would be measured in the field than in the laboratory even for identical bacteria and soils. Harvey and Garabedian (1991) calculated bacterial  $\alpha$ 's in the range of 0.005 to 0.01 based on field tests using fluorescently stained indigenous bacterial and the Yao filtration equation (Yao et al. 1971). As pointed out by Bouwer and Rittmann (1992), the use of more realistic (higher) cell densities and the RT model by Harvey and Garabedian in their calculations would have produced  $\alpha$ 's that were lower in their study by a factor of ~5.5. Such values were claimed by Bouwer and Rittmann to be close to lower limits of 0.001 observed in laboratory experiments. Thus, the observation of very low  $\alpha$  values in field tests is consistent with our results for experiments involving large collision numbers, suggesting that the low measured  $\alpha$ 's could be the result of the large number of collisions required for bacteria to be transported over long distances. Additional research would be necessary, however, to confirm this speculation.

In summary, our analysis indicates that  $d_a$  or  $d_s$  by a number distribution or  $d_{10}$  of the volume distribution should adequately represent the distribution of collector sizes in predicting bacterial transport, and that bacterial transport distances should be scaled by the dimensionless collision number ( $\xi$ ). The use of a collision number approach in future studies may help explain

variations in collision efficiency estimates under different laboratory and field conditions.

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