Applied and Environmental Microbiology, Aug. 2006, p. 5190–5196 Vol. 72, No. 8
0099-2240/06/$08.00+0 doi:10.1128/AEM.02988-05

Measurement of Biocolloid Collision Efficiencies for Granular Activated Carbon by Use of a Two-Layer Filtration Model

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Received 19 December 2005/Accepted 18 May 2006

Point-of-use filters containing granular activated carbon (GAC) are an effective method for removing certain chemicals from water, but their ability to remove bacteria and viruses has been relatively untested. Collision efficiencies (α) were determined using clean-bed filtration theory for two bacteria (*Raoutella terrigena* 33257 and *Escherichia coli* 25922), a bacteriophage (MS2), and latex microspheres for four GAC samples. These GAC samples had particle size distributions that were bimodal, but only a single particle diameter can be used in the filtration equation. Therefore, consistent with previous reports, we used a particle diameter based on the smallest diameter of the particles (derived from the projected areas of 10% of the smallest particles). The bacterial collision efficiencies calculated using the filtration model were high ($0.8 \leq \alpha \leq 4.9$), indicating that GAC was an effective capture material. Collision efficiencies greater than unity reflect an underestimation of the collision frequency, likely as a result of particle roughness and wide GAC size distributions. The collision efficiencies for microspheres ($0.7 \leq \alpha \leq 3.5$) were similar to those obtained for bacteria, suggesting that the microspheres were a reasonable surrogate for the bacteria. The bacteriophage collision efficiencies ranged from ≥ 0.2 to ≤ 0.4. The predicted levels of removal for 1-cm-thick carbon beds ranged from 0.8 to 3 log for the bacteria and from 0.3 to 1.0 log for the phage. These tests demonstrated that GAC can be an effective material for removal of bacteria and phage and that GAC particle size is a more important factor than relative stickiness for effective particle removal.

Microbial contamination of drinking water is a major health problem in many areas around the world (8, 9). Because conventional water treatment technologies are often not available in places with the greatest water contamination problems, the World Health Organization has proposed that point-of-use (POU) water treatment devices are the most efficient solution to the problem (33). Many POU devices rely on adsorption onto granular activated carbon (GAC) as a mechanism for removal of contaminants. In order for POU water treatment to be successful, the GAC must be effective in removing both chemical and microbial contaminants from the water. While GAC is widely known to be effective for decolorization, dechlorination, and chemical removal (14, 15, 17, 31), the efficiency of this material for removal of bacteria and viruses is relatively unknown.

The methods used to characterize the relative adhesion of bacteria and viruses to GAC range from batch adsorption tests to column tests. Batch adsorption tests have confirmed that microorganisms adsorb exclusively to the exterior surface of GAC due to pore size exclusion (24, 25, 27), but they do not provide information that is sufficient to predict removal rates in columns. Mass transfer models can be used to characterize chemical removal with GAC in packed beds, but they do not provide detailed information concerning particle removal mechanisms. In previous studies of virus removal in packed beds, Cookson (6, 7) described adsorption of *Escherichia coli* bacteriophage T4 onto activated carbon as a diffusion-limited process. In more recent studies, filtration models have emerged as useful and descriptive models for particle removal rates as they include consideration of the removal mechanism (diffusion, interception, and gravitational sedimentation) and allow for scaling the effect of both particle and packing sizes (28, 29, 33). Unfortunately, filtration models do not adequately predict the effects of packing, surface chemistry, and roughness, factors which are known to be important for different types of GAC (22, 35, 36), and thus individual carbons must be compared on a case-by-case basis.

Using clean-bed filtration theory, particle collision frequencies can be predicted if it is assumed that the particles and the packing are perfectly spherical and the surfaces are smooth (18, 21, 23, 28). Differences in surface particle chemistry or collector surface roughness not directly included in the theoretical model are incorporated into the collision efficiency (α), which is defined as the probability that a particle attaches to a surface based on the frequency of collisions. Mini-column tests have proven to be an effective and rapid alternative to breakthrough column tests for measuring bacterial collision efficiencies (3, 5, 10, 19). Retention or total breakthrough can be measured by incorporating a radiolabel into the bacterium, which allows rapid calculation of the collision efficiency. To examine bacterial retention, a small slice of medium is removed from the top of the bed and analyzed by scintillation counting (10). However, a radiolabel-based method is not practical for GAC as the label sorption to the carbon does not permit subsequent measurement of bacterial retention (26). Total breakthrough methods are more effective for calculating particle retention, as long as the particle retention is sufficiently great to significantly reduce the effluent particle con-

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TABLE 1. Physical and chemical properties of granular activated carbons used in this study

Property	R_s	R_u	C_o	A
Description	Wood based, sieved	Wood based, unsieved	Coconut shell	Wood based, acid treated
Specific surface area (m^2/g)	1,745	1,745	1,480	1,309
Particle density (g/cm^3)	0.56	0.56	0.72	0.59
Bed porosity	0.59 ± 0.03	0.54 ± 0.01	0.54 ± 0.03	0.63 ± 0.03
Total pore vol (ml/g)	1.31	1.31	0.92	1.21
Vol of meso- and macropores (ml/g)	0.61	0.61	0.31	0.67
Point of zero charge (pH)	8.8	8.8	9.8	5
Isoelectric point (pH)	3.5	3.5	3.8	3.3

centration (11) and the packing support material itself does not appreciably contribute to particle removal.

Clean-bed filtration theory was used here to evaluate the relative adhesion of different biocolloids (two bacterial strains and a bacteriophage) and latex microspheres in packed beds of GAC. Preliminary experiments using radiolabeled bacteria and GAC established that the collision efficiencies for the surfaces were large, but the results were inconclusive due to label sorption to the medium (retention studies) and particle filtration by the support medium (total breakthrough tests) (26). Therefore, a two-layer filtration model approach was devised to calculate collision efficiencies for highly adhesive GAC that accounted for support medium filtration. Using this new method based on a two-step experimental procedure, it was possible to rapidly determine biocolloid collision efficiencies for several different types of GAC.

MATERIALS AND METHODS

Bacteria. The microorganisms used in this study were *Raoultella* (formerly *Klebsiella*) *terrigena* 33257 and *E. coli* 25922, both of which are gram-negative nonpathogenic bacteria. These two strains were provided by K. A. Metz (The Procter & Gamble Health Sciences Institute, Mason, Ohio). Cells were grown in Miller's Luria broth (25 g/liter; Sigma). Cells were stored at −80°C in a Luria broth-glycerol solution (50%, vol/vol), transferred to fresh medium (100 ml), and harvested in the mid-exponential growth phase (~10^8 cells/ml). Bacteria were washed by centrifugation three times (2,800 × *g*, 20°C, 10 min) in 1 mM phosphate-buffered saline (PBS) (0.026 g/liter KH_2PO_4, 0.047 g/liter K_2HPO_4; pH = 7.3) before they were used in the experiments. Projected surface areas of the cells were determined by light microscopy (BH2; Olympus) using an image analysis system (Image-Pro Plus 4.1; Media Cybernetics, United States). Equivalent diameters (d_p) were calculated from the projected surface areas (A) ($n = 20$) by using the following equation: $d_p = 2(A/\pi)^{1/2}$.

Bacteriophage. *E. coli* bacteriophage MS2 was used as a surrogate for a human enteric virus (2, 30). The bacteriophage and host bacterial strain *E. coli* 15597 were provided by K. A. Metz (The Procter & Gamble Health Sciences Institute, Mason, Ohio). Phage were grown and assayed by a double-layer agar technique (1). The agar medium contained (per liter) 10 g tryptone, 9 g NaCl, 5 g yeast extract, 1 g $MgCl_2 \cdot 6H_2O$, 1.11 g $CaCl_2$, and either 10 g technical agar (bottom agar) or 8 g technical agar (top agar). Bottom agar was poured into polystyrene plates (100 by 15 mm) and solidified. The host bacterial suspension (8 drops of a suspension of *E. coli* 15597 grown as described above but not washed) was added to the test solution containing phage (1 ml), combined with 3 ml of top agar (still warm), and poured onto a plate. The plates were incubated at 37°C (agar side up) for 14 to 24 h, and then the plaques were counted.

Microspheres. Carboxylated latex microspheres (Fluoresbrite YG; Poly-Sciences Inc., Warrington, PA) that were 0.97 μm in diameter were used as an inorganic surrogate for a biocolloid. Microspheres were washed by centrifugation (2,80 × *g*, 20°C, 10 min) once in deionized (DI) water and twice in buffer (1 mM PBS) and stored at 4°C in 2.5% aqueous suspensions.

Granular activated carbon. Four activated carbon samples (provided by D. Collias, Procter & Gamble, Cincinnati, Ohio) were used; these samples were sieved (R_s) and unsieved (R_u) basic wood-based carbon, an acidic sieved wood-based carbon (A), and a sieved coconut shell carbon (C_o). The sieved carbons were prepared by shaking (US Mesh no. 140 and no. 170 sieves; 80 by 120 μm), and the carbon was collected on the US Mesh no. 170 sieve. Carbons were washed by rinsing them with DI water and were degassed (with a vacuum) to avoid air bubble entrapment, and they were stored in DI water at 4°C. The Brunauer, Emmet, and Teller (BET) specific surface areas, the pore volume distributions, the dry particle densities, the point of zero charges, the isoelectric points, and the zeta potentials of the carbons (provided by D. Collias) are summarized in Table 1. The point of zero charge is the pH at which the total surface of the particles carries a net charge of zero; above this pH the particles are negatively charged. The isoelectric point is the pH at which the external surface charge of carbon particles is zero.

GAC particle size distributions were measured using a particle size analyzer (Galai CIS-100; Galai Production Ltd., Israel) set to a range of 0.1 to 200 μm. Particle size distributions were analyzed with an Ankersmid particle size analyzer (WCIS-100, version 1.45; Ankersmid Ltd., Israel), and the results were expressed in terms of discrete number, area, and volume distribution; in this analysis we assumed that the particles were spherical (16, 18, 20).

Some experiments were conducted using simulated "exhausted" carbons, which were prepared by preequilibrating carbon (1 g, dry weight) with humic acids (10 mg/liter; Sigma Aldrich) or bacteria (7×10^6 *E. coli* cells/ml) in 1 mM PBS. Humic acid concentrations were determined using a spectrophotometer (wavelength, 254 nm), and cell concentrations remaining in solution were determined by the acridine orange direct counting technique until no significant changes in the concentrations were observed (24 h). The carbon was separated from the solution by passing it through a no. 170 sieve.

Column tests. Mini-column tests were used to determine bacterial retention on GAC. Each column was a 3-ml syringe tube containing a glass fiber filter (Whatman GF/D; nominal pore size, 2.7 μm) cut to the diameter of the tube (0.8 cm) to hold the GAC. The columns were filled with degassed GAC to obtain a bed height of 0.5 to 1 cm. Experiments were conducted as previously described (10), except that particle breakthrough was measured instead of particle retention (3). Each column was first rinsed with 10 ml of the test solution (equivalent to 10 to 12 pore volumes); this was followed by 2 ml of the bacterial or viral solution and then rinsing with 7.5 ml of the test solution to remove slowly desorbing bacteria. The column was then extracted with a plunger, dried at 100°C, and weighed in order to determine the dry column mass. We were concerned that the glass fiber filters that were needed to retain the smallest GAC particles would also capture a significant number of bacteria based on previous measurements of bacterial retention in these large-pore filters (19). Therefore. the procedure used for the GAC was repeated using only the glass fiber filter in order to determine the bacterial retention by the filter.

Two-layer filtration model. In order to incorporate the potential effect of bacterial retention in the glass fiber filter, a two-layer filtration model was used, in which the GAC was the first layer and the glass fiber filter was the second layer. In this model the fraction of particles retained when only the glass fiber filter is used (F_F) is

$$F_F = \frac{N_o - N_F}{N_o} \quad (1)$$

where N_o is the suspended particle concentration applied to the filter and N_F is the concentration in the effluent (after accounting for dilution by the rinse solutions) when only the glass fiber filter is used. When both the GAC layer and the glass fiber filter are used, the concentration of bacteria leaving the GAC (N_G) can be calculated from the concentration of bacteria leaving the column in this test (N_{GF}) by assuming that the same fractional removal occurs in the glass fiber layer in both tests, using

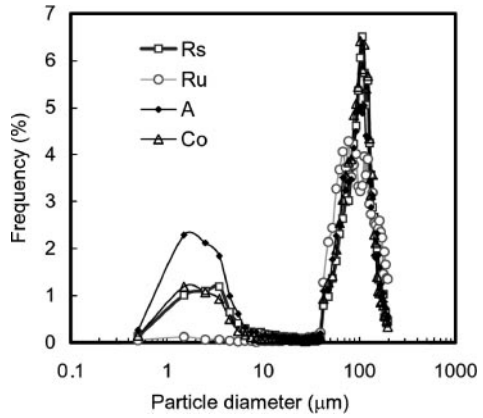

FIG. 1. Discrete distribution of particle sizes based on the area size distribution for carbons used in the study.

$$N_G = \frac{N_{GF}}{1 - F_F} \quad (2)$$

The fraction of particles retained only by the GAC (F_G) can therefore be calculated as follows:

$$F_G = \frac{N_o - N_G}{N_o} \quad (3)$$

The collision efficiency of the suspended particles within the GAC layer can be calculated using clean-bed filtration theory:

$$\alpha = -\frac{2\ln(1 - F_G)d_c}{3(1 - \theta)\eta L} \quad (4)$$

where d_c is the diameter of the packing or particle collector, θ is the bed porosity, η is the collision efficiency calculated using the model of Rajagopalan and Tien (18, 28), and L is the bed length. The single-collector efficiency (d_c) was selected from collector sizes based on measured GAC size distributions. Because the beds were short and the GAC packing was irregular, equation 4 was modified so that column mass could be used as a measured parameter instead of L. The filtration equation is derived from a mass balance on the number of isolated collectors in the bed that are length L. For a cylinder filled with spherical particles having diameter d_c, the number of collectors in a column that is length L and has cross-sectional area A is

$$N_c = \frac{6A(1 - \theta)L}{\pi d_c^3} \quad (5)$$

Rearranging and substituting into equation 4 results in

$$\alpha = -\frac{4A\ln(1 - F_G)}{\pi\eta N_c d_c^2} \quad (6)$$

The number of collectors in the column can also be calculated using $N_c = m_d/m_c$, where m_d is the total mass of the bed (dry) and m_c is the mass of a single collector ($m_c = \rho_d \pi {d_c}^3/6$, where ρ_d is the dry density of the carbon) (Table 1). With these definitions for N_c, the collision efficiency can be calculated as a function of the measured bed mass as follows:

$$\alpha = -\frac{2\ln(1 - F_G)A\rho_d d_c}{3m_d\eta} \quad (7)$$

Collision efficiencies for the glass fiber filter were calculated as described by Logan et al. (19), except that breakthrough concentrations were used, as noted above, and equation 4 was modified by replacing the geometrical factor for spheres (2/3 as written) with a factor appropriate for cylindrical glass fibers ($\pi/4$) (18). The GF/D filter characteristics were as follows: d_c, 2.7 μm; θ, 0.55; and L, 675 μm (19).

TABLE 2. Characteristic diameters for different particle size distributions, measured from 0.1 to 300 μm, showing a wide range of collector sizes for each carbon

Carbon	Size range (μm)	Distribution type	Characteristic diam (μm)			
			d_{10}	d_{50}	d_{90}	d_a
R_s	0.53–172	No.	0.53	1.35	3.25	2.65
		Area	55	104	152	
		Vol	74	115	172	
R_u	0.59–201	No.	0.59	1.56	33.73	10.24
		Area	52	106	180	
		Vol.	68	132	201	
C	0.60–175	No.	0.60	1.39	3.04	1.89
		Area	23	81	147	
		Vol	56	110	175	
C_o	0.49–165	No.	0.49	1.28	2.77	1.94
		Area	41	93	144	
		Vol	70	111	165	

RESULTS

Collector diameters for GAC. The four GAC exhibited bimodal size distributions, with a small particle size centered at 2 to 4 μm and a second peak at ca. 100 μm (Fig. 1). However, only a single collector size is used in the filtration equation. Previous work has shown that bimodal distributions should be classified on the basis of the smallest particle sizes because most of the removal occurs due to collisions with small particles (20). To determine the most reasonable method of characterizing the bimodal size distribution with a single particle size, collision efficiencies were evaluated using mini-column results for *R. terrigena* with carbon A and single-particle diameters calculated on the basis of the diameters of <10% of the particles (d_{10}) using number, area, and volume distributions (Table 2). A number distribution represents the fraction of particles based on the total number of particles, while area and volume distributions are determined on the basis of the total area or total solid volume. The collision efficiencies when d_{10} was used were 2.4×10^{-6} for a number distribution, 0.7 for an area distribution, and 6.4 for a volume distribution (Table 3). Values for other characteristic diameters based on 50% and 90% of the particles with a specified diameter (d_{50} and d_{90}) and average diameter (d_a) are also shown for comparison in Table

TABLE 3. Collision efficiencies for *R. terrigena* calculated using different characteristic collector diameters (carbon C, 1 mM PBS)

Distribution type	Diam	α
No.	d_{10}	2.4×10^{-6}
	d_{50}	2.7×10^{-4}
	d_{90}	2.5×10^{-3}
	d_a	6.5×10^{-4}
Area	d_{10}	0.7
	d_{50}	15
	d_{90}	53
Vol	d_{10}	6.4
	d_{50}	29
	d_{90}	74

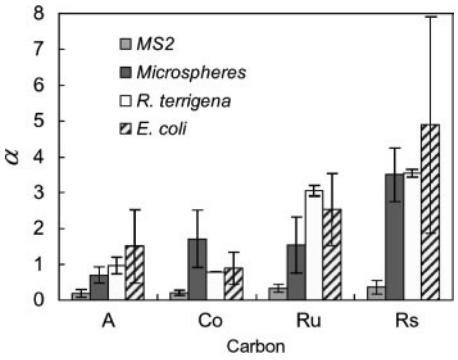

FIG. 2. Collision efficiencies of bacteria, bacteriophage, and microspheres calculated using a d_{10} based on the area distribution of the different GAC. The error bars indicate standard deviations based on aggregate data from multiple experiments using columns run in triplicate (n = 6 to 9).

3. Based on this comparison, the d_{10} based on an area distribution was used to classify the carbons for filtration calculations as this value provided the most reasonable values for the collision efficiency. The d_{10} volume distribution was not selected based on a high value of α under these conditions and substantially higher values in other preliminary tests (data not shown). The collision efficiency should theoretically be less than unity, although in practice it can be greater than unity for highly adhesive material (see Discussion). Calculated collision efficiencies based on a d_{10} number distribution were considered to be too low to be reasonable for this material.

Collision efficiencies for bacteria, microspheres, and viruses on GAC. The bacterial collision efficiencies on the four carbons ranged from 0.8 to 3.5 for *R. terrigena* and from 0.9 to 4.9 for *E. coli* when each carbon was classified by using a collector diameter of d_{10} based on an area distribution (Fig. 2). Possible reasons for collision efficiencies greater than unity are discussed below. In general, the collision efficiencies decreased in the order $R_s > R_u > C_o > A$. A pairwise comparison of collision efficiencies for each particle type on each carbon indicated that the collision efficiencies were significantly different ($P < 0.05$, as determined by a t test) for *R. terrigena* for comparisons of all carbons except for comparisons between R_u and A. For *E. coli*, there was no significant difference in collision efficiencies between R_s, R_u, and A, perhaps as a result of the large standard deviations for the data sets.

The collision efficiencies of the microspheres were generally similar to those obtained for the bacteria and ranged from 0.7 to 3.5 (Fig. 2). The ordering of the collision efficiencies with carbon type also followed a trend similar to that observed for bacteria, with R_s having the largest sticking coefficient of the different carbons ($\alpha = 3.5$) and A having the smallest sticking coefficient ($\alpha = 0.7$). The collision efficiencies for A, C_o, and R_u were not statistically different, suggesting the following order for microspheres: $R_s > R_u \geq C_o \geq A$.

The collision efficiencies for the bacteriophage were all significantly lower than those obtained for the bacteria, and the values ranged from 0.160 to 0.353. The trend in adhesion values was again consistent with the trend observed for the other types of particles, with carbon R_s having the highest collision efficiency and A having the lowest collision efficiency.

The calculated collision efficiencies described above were combined results based on two or three separate tests, with each test performed in triplicate. In general, the within-test variability was less than the variability between tests, in agreement with the previous finding by Johnson and Logan (12) that the day-to-day variability in bacterial transport tests is greater than the variability within a single test. A pairwise comparison of individual tests revealed no significant difference between tests ($P < 0.05$, as determined by a t test); therefore, data from all tests were pooled.

The α values calculated for the glass fiber filter supporting the GAC bed ranged from ≥0.01 to ≤0.04 for bacteria and microspheres. The fraction of cells retained by the filter ranged from 0.1 to 0.4, which was found to result in a significant reduction in the particle concentration. Thus, it was important that particle retention by the support filter be included in the calculation of α for bacteria and microspheres. However, the result was different for the bacteriophage. The average collision efficiency for the phage for the glass fiber filter was 0.23 ± 0.24, but neglecting the removal of the phage by the filter did not result in a statistically significant difference in the α values for the phage with the GAC (38). Thus, it should be possible in future tests to neglect the contribution of the supporting glass fiber filter in bacteriophage tests using GAC.

Predicted log removal rates for the different carbons. The comparison of the four carbons on the basis of the collision efficiency revealed which carbons are the "stickiest" for different types of particles, but other differences in the GAC, such as collector diameter, determine the overall removal efficiencies of the carbons. Fractional removal values determined in each test cannot be directly compared as the bed lengths were different in different tests. Therefore, collision efficiencies obtained in mini-column tests were used to predict removal in 1-cm-long GAC beds in order to compare the carbons on the basis of performance. The carbons were compared under two conditions, one where they had different measured diameters (d_{10}), and a second one where we predicted removals if the carbons all had the same d_{10}. We examine removal values under these two conditions in Fig. 3, where we show removal values for different diameters (Fig. 3A) and removal values predicated if the carbons all had the same diameters (Fig. 3B).

For the bacteria, 0.7- to 3.1-log removal was predicted for a 1-cm-long bed for three of the carbons (C_o, R_u, and R_s) based on measured particle size (Fig. 3). The highest removal values were predicted for the A carbon for all particle types, which was a direct result of the fact that the diameter of carbon A was smaller than the diameters of the other carbons. The predicted log removal values were lower for the microspheres than for the bacteria, ranging from 0.5 to 0.8. The bacteriophage removal values were predicted to be in a similar range, with log removal ranging from 0.31 to 0.95. When the results were compared on the basis of removal per cm, there was no significant difference in removal of the microspheres and *E. coli* between the sieved and unsieved R carbons (R_u and R_s). The effect of carbon size is shown in Fig. 3B, which shows the results when the calculations in Fig. 3A were repeated using the same carbon size (0.05 mm) for all GAC. We found that under these conditions, carbon A is less effective than the other carbons, consistent with measurements of the collision efficiency (Fig. 2).

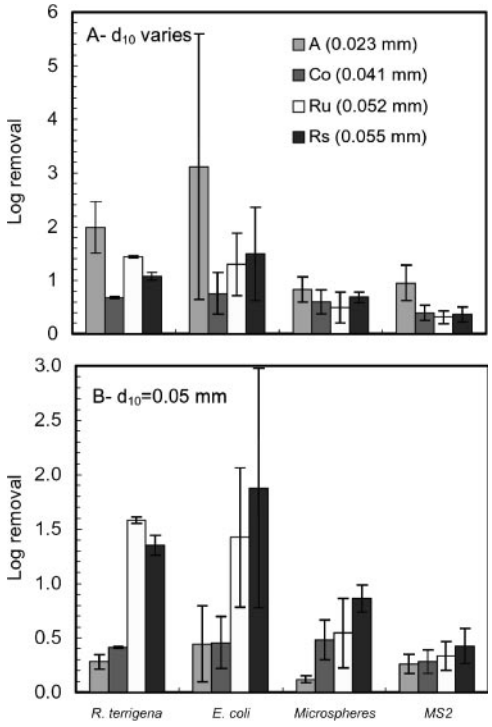

FIG. 3. Predicted log removal for GAC calculated for 1-cm-long columns on the basis of the measured α and the measured d_{10} (area distribution) for each GAC (A) or a d_{10} of 0.05 mm (B).

Effect of solution ionic strength. Decreasing the solution ionic strength (IS) by using deionized water (IS, <0.01 mM) significantly decreased the adhesion of *R. terrigena* to carbon C_o ($\alpha = 0.8 \pm 0.0$ and $\alpha = 0.4 \pm 0.1$; $P < 0.05$, as determined by a *t* test) (Fig. 4). However, the collision efficiency of *R. terrigena* with carbon R_s was not affected by the solution IS ($\alpha = 3.5 \pm 0.1$ and $\alpha = 3.3 \pm 0.80$; $P > 0.05$, as determined by a *t* test). This lack of an effect of IS could have been a result of the magnitude of α. For carbon R_s, α was greater than 1, indicating that the bacterium had a very high affinity for adhesion to this carbon. Changing the IS did not significantly affect this interaction. However, for carbon C_o, α was less than unity, and therefore an IS effect was observed.

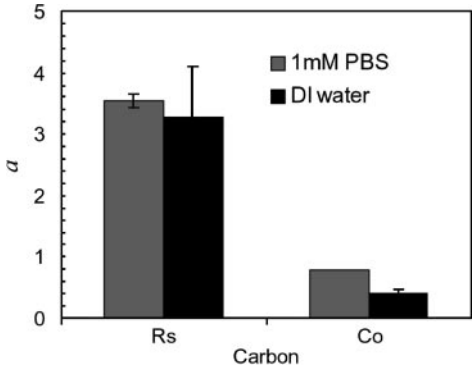

FIG. 4. Collision efficiencies of *R. terrigena* in unfavorable conditions (DI water) and in 1 mM PBS.

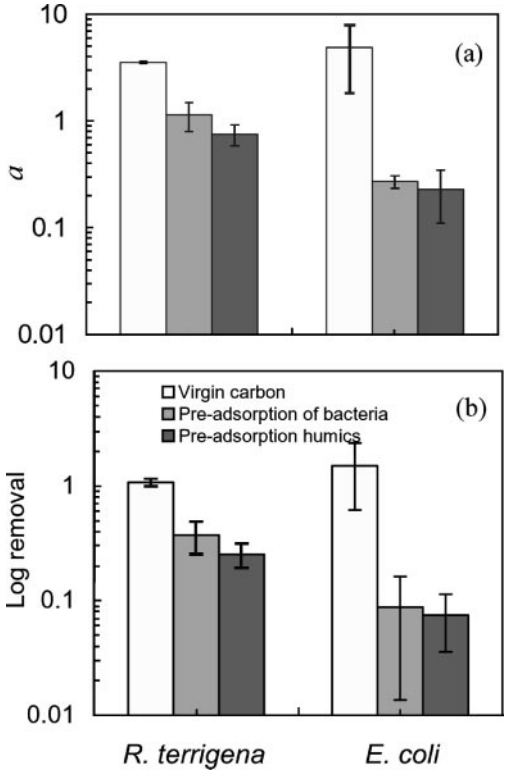

FIG. 5. Collision efficiencies (a) and removal (b) of *R. terrigena* and *E. coli* were reduced due to preadsorption of bacteria and humic acids on the surface of the carbon.

Effect of particle deposition on the surface of GAC on bacterial removal. In the tests described above, the carbons used were "clean" in that there were no preadsorbed particles or organic matter on the carbon. In practice, however, it is likely that particles and natural organic matter are present on GAC. Tests conducted with carbons preacclimated with the bacterial suspension and with humic acids resulted in collision efficiencies that were lower than those obtained with clean carbon (Fig. 5). When R_s was preacclimated with humic acids, for example, the collision efficiencies decreased ~3.5-fold for *R. terrigena* and 25-fold for *E. coli*. When R_s was preacclimated to adsorption of the same bacterium used in the filtration tests, the collision efficiencies decreased ~3.2-fold for *R. terrigena* and 16-fold for *E. coli*.

DISCUSSION

Filtration using GAC can be an effective method for removal of bacteria and viruses from water. Each 1-cm length of carbon resulted in a 0.7- to 3.1-log decrease in the bacterial concentration. The carbons therefore could reduce the bacterial concentration 6 logs, the requirement set by the U.S. Environmental Protection Agency for drinking water treatment systems, if the carbon bed lengths were 1.9 to 4.2 cm under the conditions tested. The bacteriophage removal values were lower, ranging from 0.31 to 0.95 log for a 1-cm slice, suggesting that carbon bed lengths need to be from 4.2 to 12.9 cm long to obtain the 4-log reduction required by the Environmental Protection Agency. These bed lengths are the lengths estimated for clean

carbon. As the carbon adsorbs organic matter and particles in the water, the removal efficiencies of the carbon decrease. For *E. coli*, preadsorption of humic acids reduced the removal efficiency of a 1-cm carbon bed 16-fold (carbon R_s).

The collision efficiencies calculated for the GAC samples ranged from near unity (0.8) to as much as 3.5 and 4.9 for the two bacteria. These values should be interpreted with caution, however, as there were large variations in the GAC size distributions. Therefore, the single collector diameter used here may not adequately characterize the relative stickiness of the carbon for the different types of colloids. The fact that collision efficiencies were greater than unity could be explained by assuming that the numbers of particle-GAC collisions were underestimated using the d_{10} values for the carbons. However, collision efficiencies of >1 are not unusual for highly irregular materials, such as quartz, compared with more spherical collectors, such as glass beads. Martin et al. (20) found that the collision efficiency was 1.9 when quartz was used, compared with 0.4 when glass beads were used under highly favorable interaction conditions. Similarly, Shellenberger and Logan (32) found that the collision efficiency was 2.3 for latex microspheres when mini-column tests were performed in a high-IS solution with glass beads having rough surfaces. However, the role of medium straining in producing collision efficiencies of >1 cannot be completely discounted as the size of the characteristic GAC particle is close to the size of a typical bacterium (34).

We concluded from the information described above that the highly nonspherical shape of the GAC was responsible for the high collision efficiencies. The acidic wood carbon (A) appeared to be the most efficient carbon (Fig. 3), but analysis on the basis of the collision efficiencies showed that this carbon was actually the least "sticky." On the basis of the single particle sizes used here, the filtration model indicates that the collision efficiencies were actually higher for the non-acid-treated samples (R_u and R_s). Thus, the better performance of carbon C in terms of particle removal was actually due to the smaller size of the GAC particles, not to a larger sticking coefficient.

Implications for water treatment. These results suggest that analysis of the GAC particle size distribution is more critical for assessing overall particle removal than the actual stickiness of the particles to the carbon. In general, the sticking coefficients of the bacteria were large enough, indicating that the bacteria stuck to the GAC when they collided with it. Therefore, few changes are needed in terms of the relative adhesive properties of bacteria. Some changes could perhaps increase virus particle removal, as the sticking coefficients for viruses were lower than those for bacteria. Sticking coefficients for viruses that are lower than sticking coefficients for bacteria have been reported previously (28). The extended performance of the system is therefore due to particle loading (i.e., the surface loading capacity of the GAC relative to the propensity of the material to clog with particles). The latter factors were not addressed here, but they can be ascertained in longer-term experiments based on breakthrough in longer columns using well-established methods (4, 13). Microspheres are suitable surrogate particles for bacteria in these collision tests and are easier to detect and prepare than bacteria or viruses, and thus their use may help in improving and controlling particle removal in GAC beds.

ACKNOWLEDGMENTS

This research was funded by Procter & Gamble.

We thank Don Langworthy, Dimitris Collias, and Kathy Metz for providing assistance with the experiments.

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