

Enumerating and Disinfecting Bacteria Associated With Particles Released From GAC Filter-Adsorbers

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Granular activated carbon (GAC) in filter-adsorbers provides an excellent support surface for the proliferation of microorganisms. Therefore, GAC beds may release particles of carbon with attached bacteria that are protected from disinfection. In this pilot-plant study, particles were collected from the product waters of GAC filter-adsorbers, examined for bacterial colonization, and characterized by energy-dispersive X-ray analysis. Results showed that bacteria attached to GAC particles could be disinfected with chlorine if particle concentrations were in the range found in the product waters, i.e., $<20 \mu\text{g/L}$. Increasing particle concentration tenfold interfered with disinfection efficiency. Powdered activated carbon used in pretreatment contributed to the particles found in product water.

Granular activated carbon (GAC) in filter-adsorbers provides an excellent support surface for the proliferation of microorganisms.¹ In fact, biodegradation that results from this colonization provides a practical way to extend service time of filter-adsorbers and to reduce the amount of biodegradable total organic carbon (TOC) that enters the distribution system.²⁻⁴ Bacteria proliferate on GAC even with prechlorination because GAC catalyzes the reduction of chlorine⁵ and thus prevents bacterial deactivation. Some of the bacteria that colonize the surface of GAC are released into the product water.^{1,2,4} If these bacteria can-

not be easily disinfected, the risk of bacterial contamination of the drinking water is increased. Bacterial pathogens can colonize sterile GAC; however, they may not grow in competition with natural heterotrophic bacterial populations.⁶

Most of the concern over potential penetration of the disinfection barrier is focused on those bacteria attached to particles of GAC that escape in the product water of filter-adsorbers. It is generally well known that bacteria attached to any surface are more resistant to disinfection than unattached bacteria. LeChevallier et al,⁷ for example, showed that attachment to glass made *Klebsiella* more resistant to disinfection by chlorine and chloramine. Ridgway and Olson⁸ found that bacteria in water distribution systems were associated with particles, aggregates, and surfaces and suggested that attachment protected bacteria in chlorinated water.

In studies of filter-adsorbers, Camper et al^{9,10} and Stewart et al¹¹ found that GAC particles in the product water were colonized by bacteria (before disinfection in the plant). Camper et al⁹ found an extremely broad range of particle diameters ($1 \mu\text{m}$ – 3.5 mm) in a study of full-scale filter-adsorbers, whereas Stewart et al¹¹ reported a much narrower range of rela-

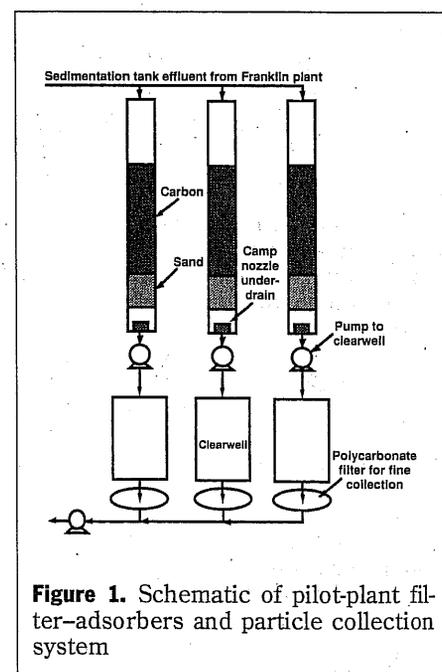


Figure 1. Schematic of pilot-plant filter-adsorbers and particle collection system

A full report of this project, "Microbial Activity on Filter-Adsorbers" (catalog # 90606), is available from AWWA Customer Service (1-800-926-7337). Reports are free to AWWA Research Foundation subscribers by calling 303-347-6121.

tively small particles (2–40 μm with a mean of 5.4 μm) in pilot-scale filter-adsorbers. The large particles from the full-scale filter-adsorber were on the order of the media size and may have been the result of a damaged underdrain system.

The resistance to disinfection of bacteria attached to particles has been studied by Camper et al.⁹ and Stewart et al.¹¹ Particles in the product water from filter-adsorbers were captured by gauze or membrane filtration and then resuspended in a small volume of water to yield a much higher concentration of particles than originally were in the product water. This solution was disinfected with free chlorine (2 mg/L) for 30 min, and it was inferred that the bacteria that were shown to survive disinfection in this experimental protocol would also survive under treatment plant conditions.

Bacteria attached to particles withdrawn from filter-adsorbers also resist disinfection. LeChevallier et al.¹² and Stewart et al.¹¹ suspended GAC media in a small volume of solution to study disinfection of attached bacteria.

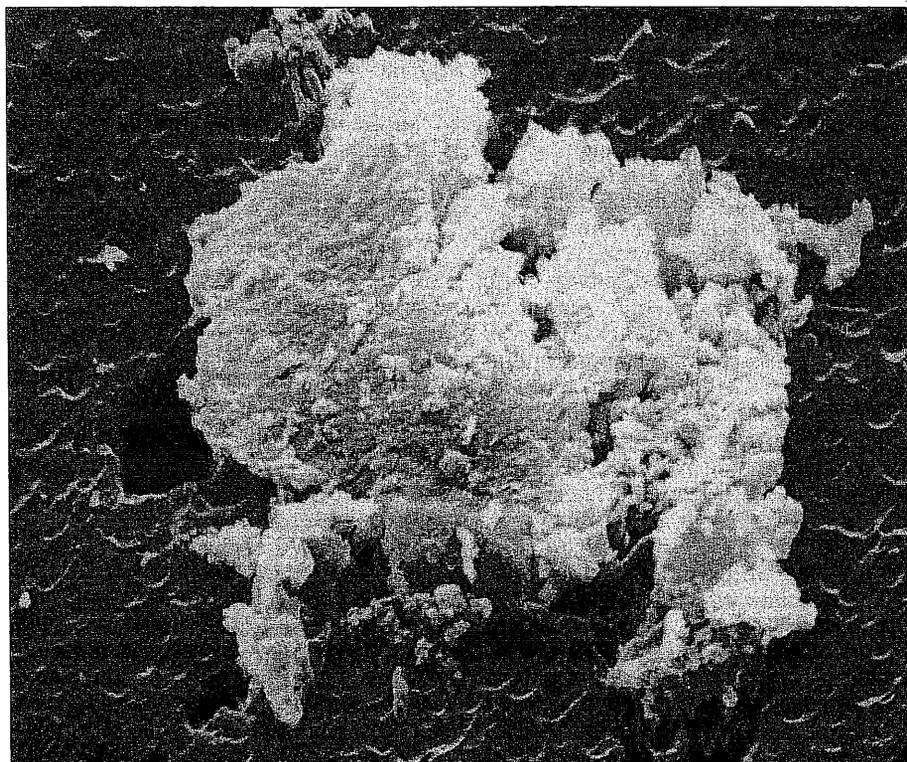
The problem with interpreting resistance of attached bacteria to disinfection from these laboratory experiments is that the procedure involves concentrating particles to the range of grams per litre. This concentration is much higher than would be present in the product water and could lessen disinfection because it is well known that bacteria are protected from disinfection in turbid solutions. Moreover, the disinfectant may be chemically reduced very rapidly by the large surface of activated carbon before it can react with attached bacteria.

Notwithstanding the factors that confound this interpretation, there is still concern that attached bacteria could escape the disinfection barrier following GAC filtration. This concern may be especially important if the underdrain system of the filter is damaged or is in generally poor condition such that GAC media can escape into the product water.

In this article the release of particles and attached bacteria from GAC filter-adsorbers is examined. Laboratory experiments were conducted to determine the dependence of disinfection efficiency on particle concentration. Particles released from GAC filter-adsorbers were characterized, and the number of attached bacteria on the particles was determined.

Materials and methods

Franklin water treatment plant. The pilot plant was located at the Franklin water treatment plant of the Charlotte-Mecklenburg Utilities Department (CMUD) in North Carolina. The Franklin plant processes 55 mgd ($2.08 \times 10^5 \text{ m}^3/\text{d}$) of water from Lake Norman, which is a high-quality source water (Table 1). The



This electron micrograph of a section of a 5- μm polycarbonate filter shows that some trapped particles from new GAC product water were at least partially coated with what appears to be biofilm or floc.

1989 annual mean values of raw-water turbidity and standard plate counts were 4.4 ntu and 412 cfu/mL, respectively. TOC is not usually measured at the plant. However, TOC was measured after sedimentation at the Franklin plant as part of this pilot-plant study from December 1988 to July 1990.¹³ The TOC in 86 grab samples ranged from 1.1 to 3.5 mg/L; the average and standard deviations were 2.3 and 0.7 mg/L, respectively. Free chlorine (0.8–1.2 mg/L) is added ahead of the rapid mix tank. Powdered activated carbon (PAC) (1–2 mg/L) and alum are added in the rapid mix tank, which is followed by coagulation, sedimentation and dual-media (anthracite-sand) filtration. The Franklin plant produces an excellent finished-water quality as indi-

cated in Table 1 by the 1989 annual mean value of 0.2 ntu of turbidity and <1 cfu/mL.

Pilot-plant operation. As illustrated in Figure 1, settled water from the Franklin plant passed by gravity through three pilot-scale filters (10.2-cm diameter) in parallel. Each clear acrylic column was fitted with a Camp underdrain nozzle (vertical slots). The columns were fitted with manometer ports at different depths of the media (not shown) for head loss measurements. Some of these ports were converted to withdraw water samples for measuring residual chlorine. Constant-rate filtration was provided by a positive displacement pump on the effluent side of each filter. The filter-adsorbers contained 3 ft (0.9 m) of GAC over 1 ft (0.3

TABLE 1
General water quality at Franklin plant and important treatment characteristics

Water Type	Turbidity* ntu	Heterotrophic Plate Count* cfu/mL	TOC† mg/L	Free Chlorine‡ mg/L	PAC‡ mg/L
Raw water	4.4	412		0.8–1.2	1–2
Settled water			2.3 (± 0.7)		
Finished water	0.2	<1		0.45	

*Average from Franklin plant records for 1989

†Average and standard deviation measured in this pilot-plant study during runs 1, 2, and 4 (86 measurements)

‡Range from Franklin plant records for 1989 (added before rapid mix tank); average not available

m) of sand. A control sand column contained sand alone to a depth of 2 ft (0.6 m). The difference in the depth between the control sand column and the columns containing GAC over sand was necessary to permit the same backwashing frequency for all filters. Each filter was backwashed at a rate of 12.6 gpm/sq ft (30.8 m/h) by reversing the flow through the pumps from the clearwell.

The product water was discharged into 55-gal (208-L) drums that served as clearwells. Attached to each clearwell was a stainless-steel, 293-mm-diameter filter holder.* Particles in the product water were captured using a 5- μ m-pore-size polycarbonate filter.† The procedures for collecting particles and gravimetric analysis are described later in this article.

Five filter runs, from two to four months each, were conducted during the pilot-plant study, which lasted from Dec. 14, 1989, to Nov. 15, 1990. The initial three runs are not discussed in this article, but the results are published elsewhere.¹³ These runs evaluated the effect of application rate and backwashing strategy on filter-adsorber performance.

Each of them began with virgin activated carbon.‡ Briefly, the data showed that the filter-adsorber performed as well as, if not better than, the full-scale plant's dual-media filters when measured by rate of head loss development and product-water turbidity, even when the application rate was as high as 6 gpm/sq ft (14.7 m/h). Further, these runs established that colonization of the filter-adsorber occurred despite the low concentration of TOC in the settled water and prechlorination in the plant. Backwashing with free chlorine (about 2 mg/L) every 48 h showed a significant reduction in heterotrophic plate count (HPC) levels in the product water over more frequent backwashing (every 24 h) without chlorine.

Various procedures for capturing and quantifying particles in the product water were attempted in runs 1-3. These relied on using a timer to remotely control a pump that operated intermittently over days and weeks to remove product water from the clearwell and pass it through the particle-collection filter (i.e., the 5- μ m polycarbonate filter). However, difficulties were encountered in maintaining

constant filtration rates (because of filter clogging) and thus the particle concentration could not be measured accurately. An acceptable method was finally established and is reported here for runs 4 and 5.

Run 4 was conducted from Apr. 11, 1990, to July 19, 1990, and run 5 from July 21, 1990, to Nov. 15, 1990. Backwashing was performed once a day, and no chlorine was added to the backwash water. Both of these runs included evaluation of particle release and bacterial attachment using GAC in the filter-adsorbers that had been in service for months and years at other water treatment plants. One of these GAC samples had been reactivated. The goal was to determine whether a weakening in physical structure caused by repeated backwashing or reactivation results in a greater number of particles, and possibly attached bacteria, to be released into the product water than would be released by a filter-adsorber containing new GAC.

As a result of particle abrasion during service, the two GAC samples taken from the water treatment plants had smaller average sizes than the new GAC. Therefore, all three GAC samples were rinsed, dried, and sieved, and only those particles between 8 and 30 US standard mesh size (corresponding to particle diameters between 0.59 and 2.38 mm) were used in the filter-adsorbers.

In run 4, one filter-adsorber contained sand and old GAC§ that had been in service for approximately two years at the Lynchburg, Va., water treatment plant. Another filter-adsorber contained sand and new GAC.¶ The third filter was filled entirely with sand taken from the Franklin treatment plant. In run 5, one filter-adsorber contained sand and reactivated GAC§§ obtained from the Buffalo Pound water treatment plant in Regina, Sask. This GAC had undergone approximately seven months of service after reactivation. The other two filter-adsorbers contained sand and either new or old GAC.

Measurements. TOC was measured by the persulfate oxidation method using a TOC analyzer.** Free and total chlorine concentrations were measured by amperometric titration.†† Turbidity was measured using an on-line turbidimeter.‡‡ HPC levels were measured using R₂A agar.§§ Samples were spread-plated on the agar and incubated for seven days at 25°C. Bacterial colony-forming units grown on this agar are reported as HPC/mL; those

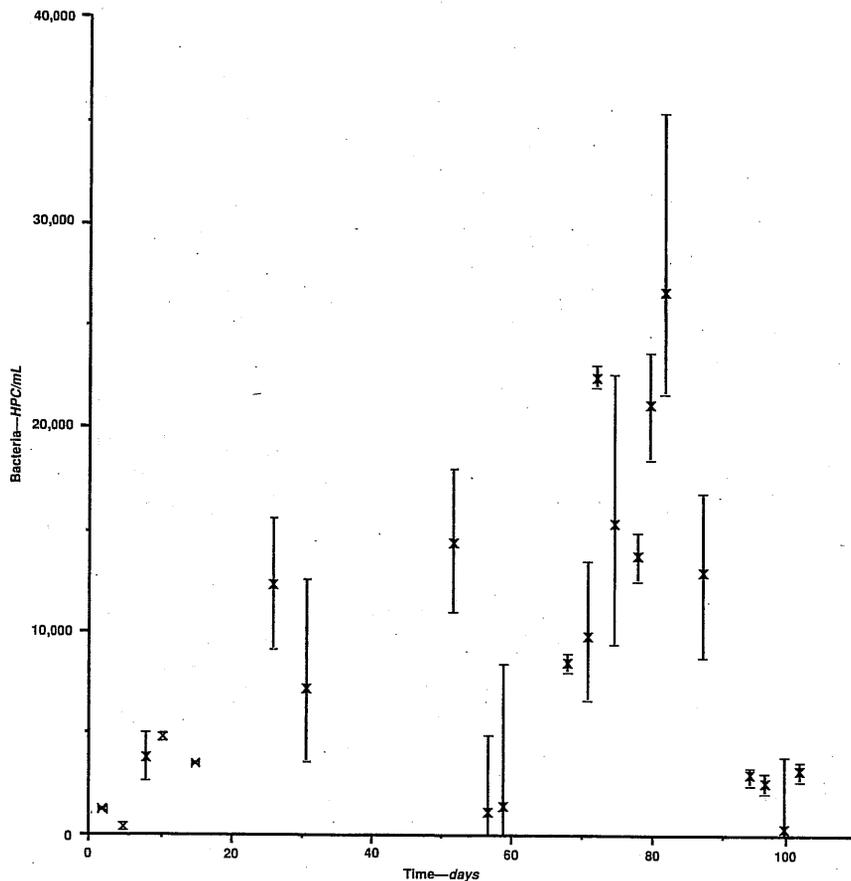


Figure 2. Heterotrophic plate counts (mean and standard deviation) in product water from filter-adsorber containing new GAC (run 4)

*Millipore Corp., Bedford, Mass.

†Nuclepore Corp., Pleasanton, Calif.

‡F-300 Filtrasorb, Calgon Corp., Pittsburgh, Pa.

§Mixture of F-300 Filtrasorb, Calgon Corp., Pittsburgh, Pa., and Cecacarbon, Atochem North America Inc., Pryor, Okla.

**Model 700, O.I. Corp., College Station, Texas.

††Fisher Scientific Co., Pittsburgh, Pa.

‡‡Hach Co., Loveland, Colo.

§§Difco Labs., Detroit, Mich.

grown on other media are reported as cfu/mL. Typically, the HPCs were conducted in triplicate on product water samples taken from the filter-adsorbers. Total coliforms were measured by membrane filtration and the presence-absence test using 100-mL sample volumes. All methods were conducted in accordance with the procedures described in *Standard Methods*.¹⁴

Collection of particles from product waters.

Particles were collected three times from the product water during run 4 (after 45, 90, and 99 days of service) and four times during run 5 (after 18, 33, 49, and 93 days of service). Clearwells (208 L in volume) in the pilot plant were drained and rinsed three times with chlorinated tap water before refilling with product water for sample collection. Approximately 1.5 h were required to refill the clearwell given a flow rate of about 0.35 gpm (1.3 L/min), which corresponds to an application rate of 4 gpm/sq ft (9.8 m/h). Some particles could have settled during the filling of the clearwells. However, a high-rate pump (10 gpm [15.1 L/min]) was used to remove the water from the bottom of the clearwell and deliver it to the polycarbonate filters. This pump was observed to create considerable mixing in the clearwell, thus assuring resuspension of any settled particles.

From one to four clearwell volumes (200–800 L) were filtered in this manner to measure particle concentration. Particles were washed from the polycarbonate filters with sterile water and resuspended in 100 mL of sterile distilled water. This produced a solution of particles that was 2,000 to 8,000 times more concentrated than the sampled product water. Microscopic examination of the polycarbonate filter confirmed that all of the particles had been dislodged. An aliquot was filtered through a 0.45- μ m glass-fiber filter, oven dried at 105°C, and weighed to determine particle concentration (i.e., from knowing the volume of sample collected from the clearwell and filtered by a 5- μ m polycarbonate filter). Immediately following the volume collection for gravimetric analysis of particles, another 200–800 L of product water was filtered to collect particles and attached bacteria for microbial analysis and disinfection studies. Particles were washed from the polycarbonate filters with sterile water and resuspended in 100 mL of sterile distilled water, and the filter was microscopically examined to confirm that all of the particles had been dislodged. A 5- or 10-mL sample of the solution was placed in a test tube containing sodium thiosulfate and was used for the determination of initial attached HPC bacteria.

Grab samples were collected for the quantitation of black particles in the column influent and product waters. Grab

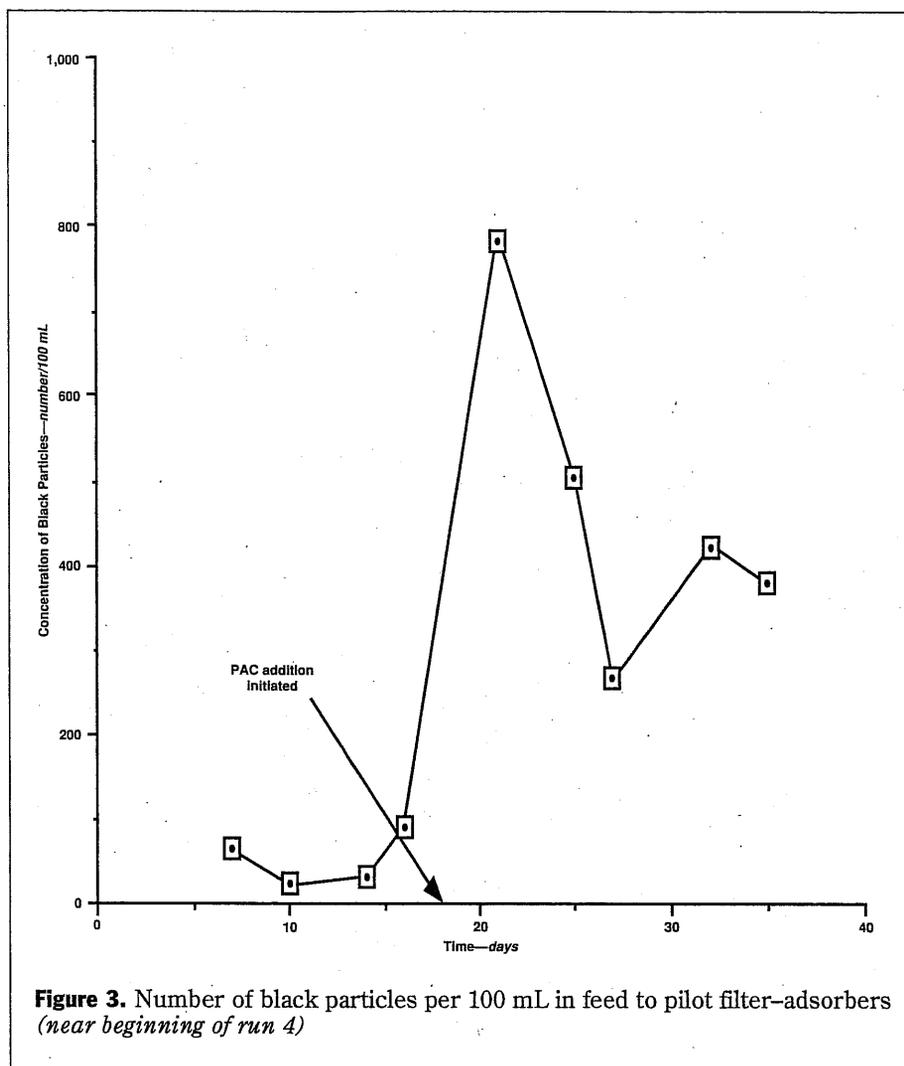


Figure 3. Number of black particles per 100 mL in feed to pilot filter-adsorbers (near beginning of run 4)

samples (100 mL) were filtered through a 0.45- μ m membrane filter,* and the black particles were counted using a 30 \times dissecting microscope. Black particles were used as a measure of GAC particles and PAC in water samples.

Disinfection studies. Disinfection experiments were conducted on particles collected on three different occasions during run 5. The mass of particles available from each collection did not permit replication of results. Various concentrations of particles were prepared with sterile, distilled water using particles collected from the product water of the filter-adsorbers containing new GAC. Chlorine-demand-free glassware and water were used. Reactions were run in sterile biochemical oxygen demand (BOD) bottles mixed with PTFE stir bars. HPC was measured in duplicate. A temperature of 10°C was selected to slow the kinetics sufficiently to follow the sharp decrease in HPC that typically occurred in a period of about 5 min. The disinfectant was hypochlorous acid at concentrations of 0.5, 1, and 2.0 mg/L. Experiments were conducted at a pH of 6.5, at which 95 percent of the hypochlorous acid was present as HOCl.

Samples (5 or 10 mL) were withdrawn from the BOD bottle over time and placed in test tubes containing sodium thiosulfate to reduce the chlorine. Vortex mixing was used to detach bacteria from the particles. Bacteria detached by vortex mixing were then enumerated on R₂A agar.† Vortex mixing was selected instead of homogenization because both methods gave nearly identical HPC recoveries for the samples.¹³ It should be noted that Camper et al¹⁵ found that homogenization greatly increased detachment. This difference may be due to the larger size of the particles collected in the Camper study.

Scanning electron microscopy. Particles captured from product water could originate from the release of GAC from the filter-adsorbers, penetration of alum floc, clumps of bacteria, and penetration of PAC (because of its introduction to the rapid mix at the Franklin plant). Scanning electron microscopy (SEM), therefore, was used in an attempt to distinguish these various origins. Particles were prepared for SEM analysis‡ by fil-

*Gelman Sciences, Ann Arbor, Mich.

†Difco Labs., Detroit, Mich.

‡S-200, Cambridge (Leica), Cambridge, England

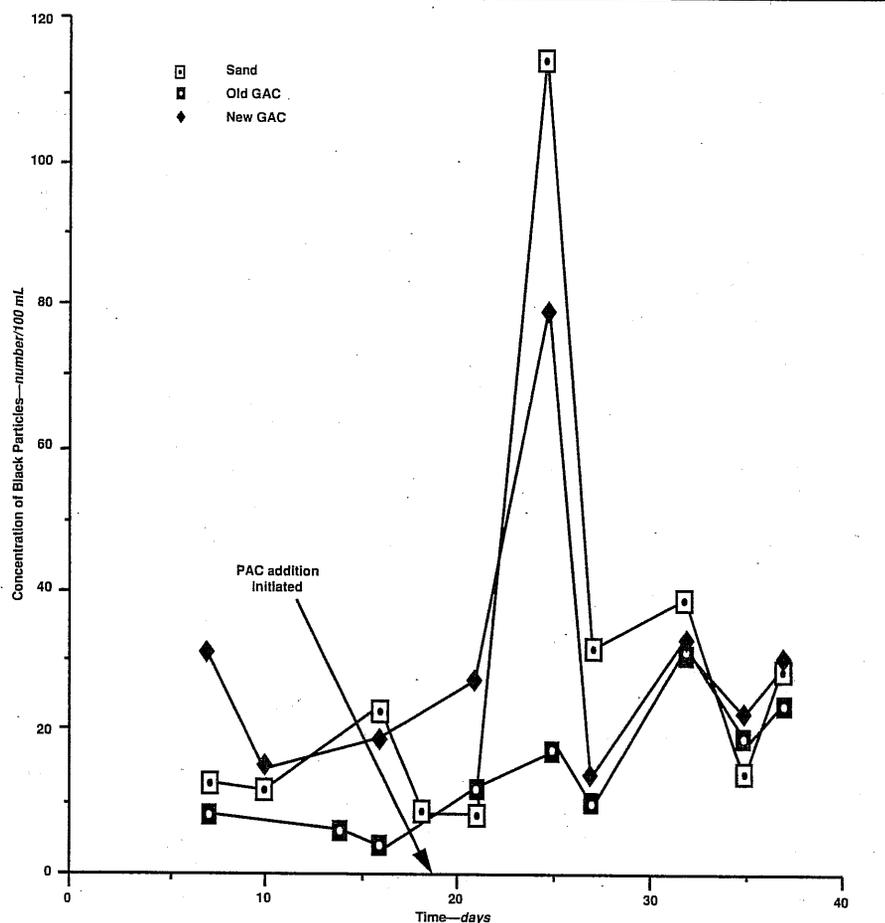


Figure 4. Number of black particles per 100 mL in product water of pilot filter-adsorbers (near beginning of run 4)

TABLE 2
Chlorine concentration in filter-adsorbers in run 5*

Column Depth ft	Chlorine Concentration†—mg/L		
	New Granular Activated Carbon	Two-Year-Old Granular Activated Carbon	Reactivated Granular Activated Carbon
0.5	0.65	0.64	0.65
1.5	0.10	0.09	0.03
2.5	0.00	0.00	0.00

*Average prechlorination concentration of 2 mg/L
†Average of four measurements taken once per week

TABLE 3
Total heterotrophic plate counts in product water in run 4

Filter Type	Total Heterotrophic Plate Counts—HPC/mL		
	Number of Samples	Mean	Standard Deviation
Settled water*	84	11	30
Sand	59	320	700
New GAC	56	11,780	8,080
Two-year-old GAC	52	6,310	6,670

*Influent to pilot filter-adsorbers

tering grab samples (100 mL) onto 0.2- μ m polycarbonate membrane filters. In addition, sections of the 293-mm-diameter, 5- μ m polycarbonate filters, which were used to collect particles from the clearwell, were cut and prepared for SEM. Each sample was rinsed three times with deionized water followed by dehydration by a series of ethanol-water mixtures (50, 75, and 95 percent each for 5 min and then 100 percent twice for 5 min each). The sample was then placed into a fluorocarbon solvent* (twice for 10 min) and transferred to a critical-point dryer† in the fluorocarbon for drying (the fluorocarbon is the intermediate solvent, and carbon dioxide is the transition or substitution fluid). After drying, the specimens were mounted onto aluminum stubs with colloidal silver paste and then coated with gold-palladium (Au-Pd) alloy (60:40) using a vacuum evaporator.‡

Energy dispersive X-ray analysis. To aid further in distinguishing the origin of particles, filtered samples were subjected to energy-dispersive X-ray analysis,§ which allows for the detection of specific metals present on captured particles. Known samples of alum floc, kaolinite, and activated carbon served as controls with which to compare results obtained with particles captured from the clearwell. The samples were dried in the same fashion as samples for SEM. After drying, the specimens were mounted on carbon planchettes with a colloidal graphite adhesive and coated with a thin film of carbon using a vacuum evaporator** prior to X-ray analysis.

Results and discussion

Release of bacteria into product waters. HPC data (triplicates) for the product water in run 4 from a filter-adsorber containing new GAC as a function of service time are given in Figure 2. After three days of operation, the product water contained an average of 2,120 HPC/mL. Given that prechlorination is practiced at the Franklin plant, routine collection of samples at the feed point to the pilot filter-adsorbers showed no measurable HPC. Thus, the HPC in the product water of the filter-adsorber after three days of operation is attributable to growth of bacteria within the bed and release of some fraction of this growth. Release of bacteria continued throughout the 101 days of data collection in run 4. The highest HPC/mL was 28,300 (standard deviation of 6,800 on three replicates), and many values exceeded 8,000/mL. The range of HPC/mL measured in the product water for run 4 is similar to that for all pilot-plant runs.¹³ Stewart et al¹¹ reported a

*Freon 113, Electron Microscopy, Fort Washington, Pa.
†CPD 020, Balzer, Hudson, N.H.
‡Denton Vacuum Inc., Cherry Hill, N.J.
§7000 EDX with quantx software, Keveex Corp., Foster City, Calif.
**Denton Vacuum Inc., Cherry Hill, N.J.

stable level of 10,000 HPC/mL, which is similar to these results. Wilcox et al.¹ typically found total bacterial counts in the range of 2,000–5,000 cfu/mL in product waters using soil extract agar instead of the agar used in this study and that of Stewart et al.¹¹

The GAC was colonized effectively despite the fact that the TOC concentration was very low (average of 2.3 mg/L). Also, measurements of TOC in the product water showed that only about 0.3 mg/L of this TOC was removed by the filter-adsorbers after long-term operation (several months), presumably by biodegradation.¹³

Colonization occurred even though the pilot filter-adsorbers received prechlorinated water (average of 2 mg/L). This result was expected given that chlorine is well known to be chemically reduced by the surface of activated carbon.⁵ The results shown in Table 2 provide evidence for rapid removal of chlorine in the filter-adsorbers, regardless of whether they contained new, old (two years in service), or reactivated (i.e., in service for seven months following reactivation) GAC. The measurements given in the table were the average of four taken once per week for four successive weeks at three bed depths during run 5.

The increases in bacterial numbers that occurred in the water passing through the GAC filter-adsorbers in runs 4 and 5 are summarized by the data in Table 3. The settled water introduced to the GAC filter-adsorbers still contained chlorine (0.5–1.0 mg/L), and bacterial numbers were quite low. After the water passed through GAC filter-adsorbers, chlorine was removed (Table 2), and countable bacteria significantly increased. No significant difference in the bacterial concentrations of product waters was observed ($\alpha = 0.05$) between the old GAC and new GAC. For comparison, data from a pilot sand filter are included. The sand filter also promoted some bacterial growth, but because the chlorine penetrated the sand column, bacterial growth was inhibited. Other studies have found that when the influent water is not chlorinated, the increase in bacterial load upon passage of the water through GAC columns is less significant.^{1,11}

No coliforms were found in the product water of any column. The high quality of the raw water, coupled with prechlorination, probably limited colonization of the filter-adsorber. However, the occurrence of coliforms in product waters of GAC columns can occur if the influent waters contain coliforms.^{1,9,11} Coliforms and bacterial pathogens appear to be at a competitive disadvantage on GAC, and proliferation of pathogens in the carbon bed appears unlikely, but not impossible.^{2,3,9}

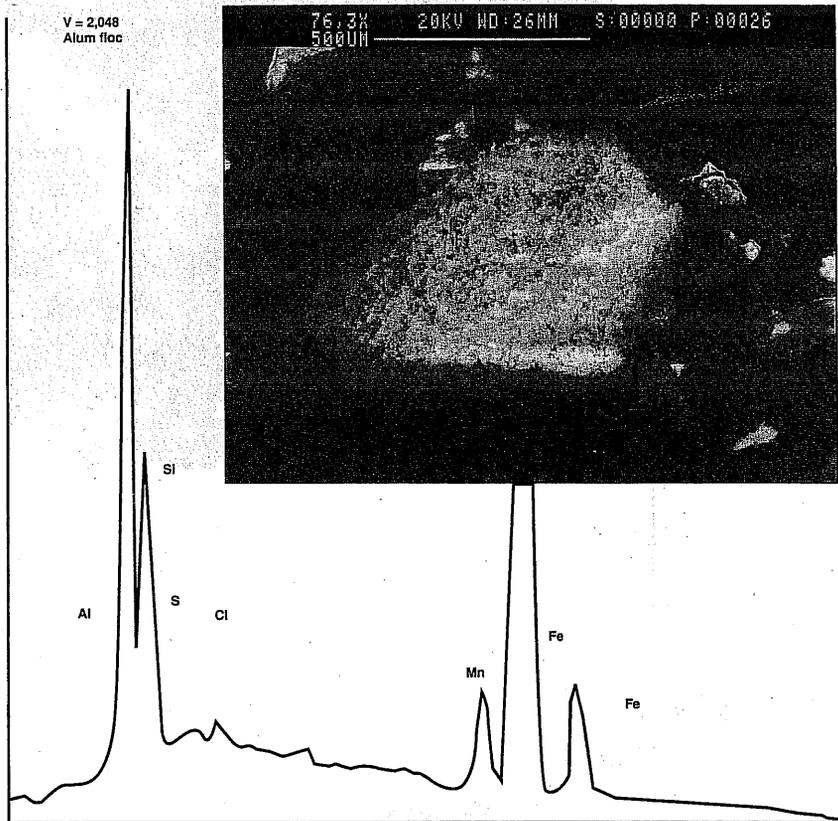


Figure 5. Energy-dispersive X-ray analysis of alum floc particle

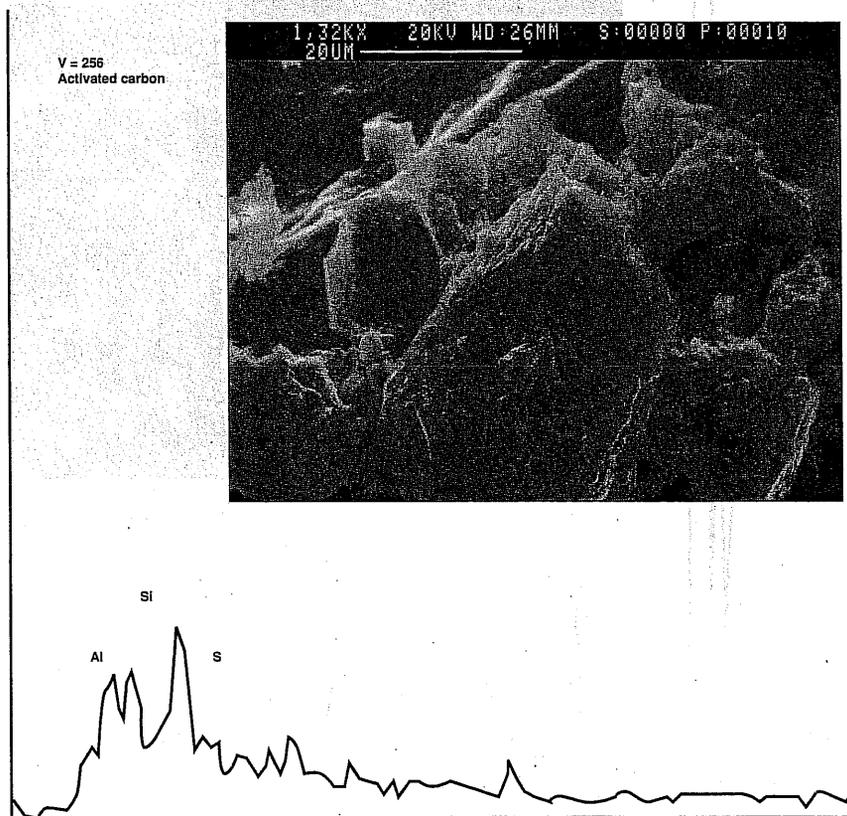


Figure 6. Energy-dispersive X-ray analysis of activated carbon particle

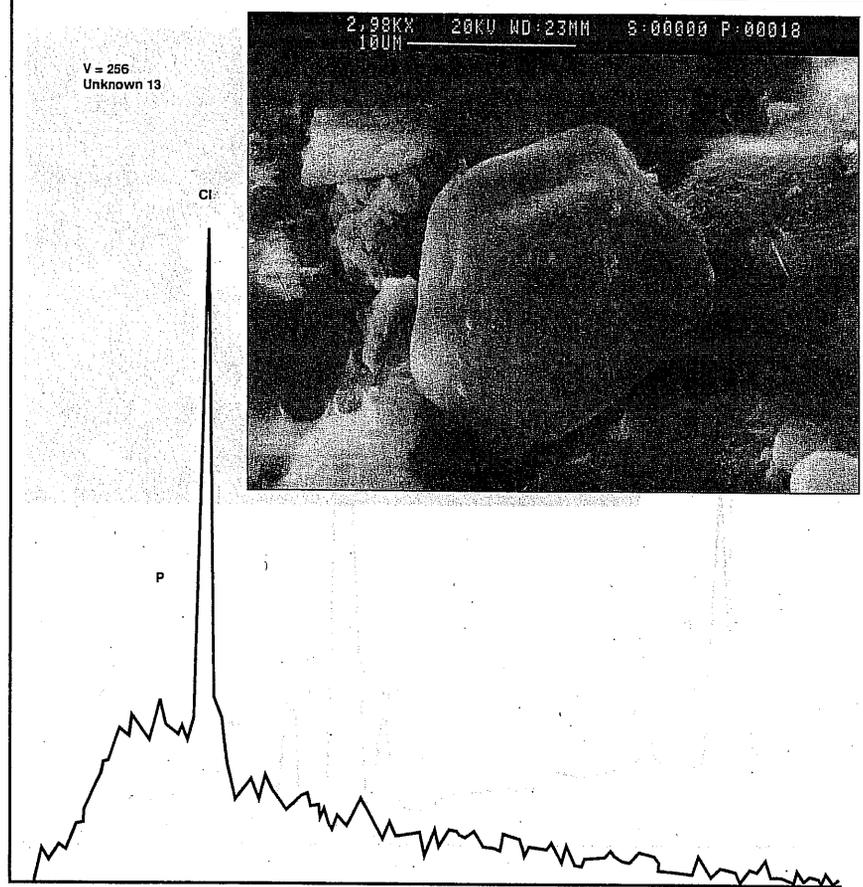


Figure 7. Energy-dispersive X-ray analysis of particle resembling activated carbon collected from product water of GAC filter-adsorber

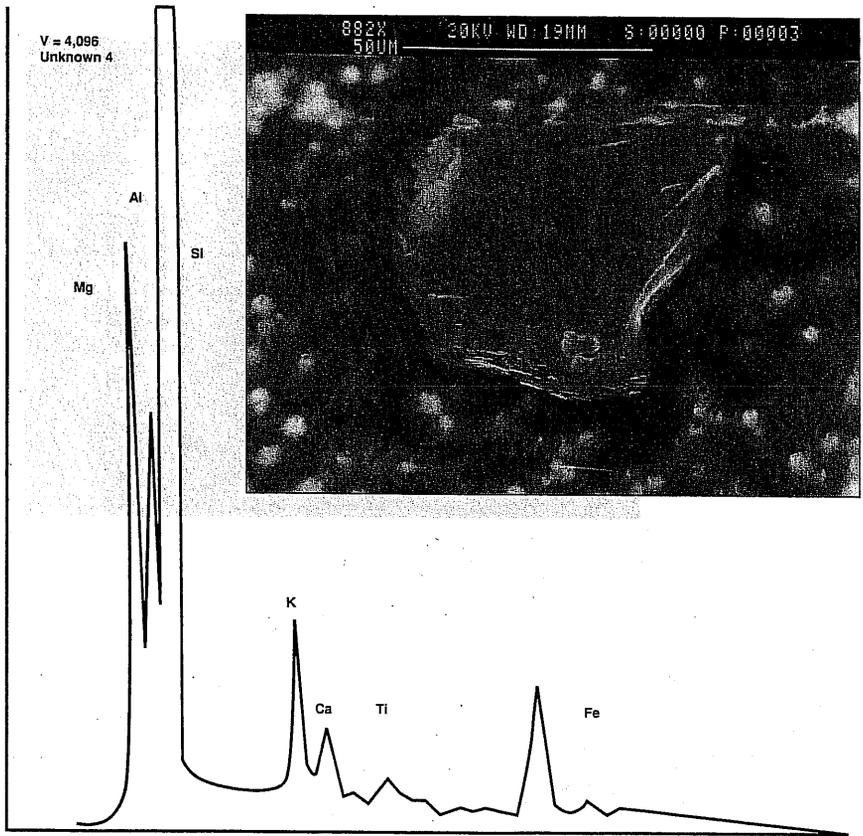


Figure 8. Energy-dispersive X-ray analysis of particle resembling clay collected from product water of GAC filter-adsorber

Disinfection of product waters. The product water from GAC filter-adsorbers was easily disinfected despite its having a high HPC. When 0.5 mg/L of free chlorine was used, no viable HPC could be detected after 5 min of contact. Effective disinfection in this study may have been expected given the very low turbidity levels in the product water for all three conditions of GAC (new, old, and reactivated) that were tested (Table 4); these data also show that the GAC filter-adsorbers are as effective as sand for filtration. A notable exception to these results was reported by Stewart et al.¹¹ They found that 1.0 mg/L of free chlorine and 1 h of contact with product water from a pilot GAC bed only achieved a one-log reduction of HPC (i.e., a reduction from 10,000 to 1,000 HPC/mL).

Release of particles from filters. The results of gravimetric analysis for particles captured on the 5- μ m polycarbonate filter in runs 4 and 5 are shown in Table 5. The concentration of particles averaged between 8 and 14 μ g/L. Sand filters and GAC filter-adsorbers produced similar particle concentrations in the product water. This implies that the amount of carbon fines generated and released by filter-adsorbers was not adding substantially to the total mass of particles. Moreover, reactivated GAC and old GAC did not produce significantly larger amounts of particles than new GAC. It should be noted that these GAC samples had all been sieved to give the same particle size fraction (8 \times 30 US standard mesh size) in each filter-adsorber. In practice, smaller particles may be found in filter-adsorbers, and thus the extent of particle break off may be different than measured in these tests. Nevertheless, previous studies have shown that the number of black particles found in filter-adsorber product water was not influenced by the age of the carbon being tested.¹⁰

Penetration of PAC particles. A potential source of carbon particles was the carryover of PAC from the sedimentation basin of the Franklin plant. Nominal operation at the plant included addition of 1 mg/L of PAC ahead of the rapid mix tank, but 2 mg/L were occasionally applied for taste and odor control. Microscopic examination revealed the presence of black particles in the settled water, which suggested the presence of PAC particles. Visual observation of the sand filter in the pilot plant showed that black particles were being captured in the topmost layer. Black particles were also observed among the particles collected in the product water of the sand filter.

At the beginning of run 4 in April 1990, the CMUD staff agreed to turn off the PAC feed for one month. The particle count data (measured in triplicate and averaged) for the settled water are given

in Figure 3. Black particle counts were very low while the PAC feed was turned off but increased significantly when the PAC feed was resumed.

The black particles found in the product water of the new GAC over sand, old GAC over sand, and sand filters during this same period are given in Figure 4. Although some increase in particles was observed after the PAC feed was resumed, the counts again returned to fairly low values (<40/100 mL) compared with the 400/100 mL entering the filter-adsorber (Figure 3). These data were consistent with the low values of product-water turbidity, which showed that the pilot-plant filters were very efficient. The number of black particles counted in the product water of the sand filter was similar to those counted in GAC filter product waters. It is apparent that some of the particles in the product water could have resulted from PAC penetration rather than in-situ generation of particles and that penetration of PAC may contribute more particles than in-situ generation of particles.

Najm et al¹⁶ reviewed previous studies on penetration of PAC through filters. PAC breakthrough was noted at a very large PAC dose (60 mg/L) and high application rate (>8–9 m/h), whereas breakthrough did not occur at a PAC dose of 30 mg/L and an application rate of 4.9 m/h. Both of these dosages are much higher than those used at the Franklin plant.

An experiment was performed to measure the residence time distribution of PAC particles in the filter-adsorber. At this time, the Franklin plant was once again using 1 mg/L of PAC. A slurry containing 5 g of PAC was added to the feedwater of the sand filter and the filter-adsorber containing new GAC. Particles were counted in grab samples over the next two days without backwashing the filters. None of these grab samples gave particle counts greater than measured in the product water just before adding the pulse of PAC. The average of five grab samples taken over about 1 h was 112 particles/L for the sand filter and 97 particles/L for the filter-adsorber. Visual inspection of the sand filter showed a thick layer of PAC at the top of the sand medium, and some PAC penetrating to a depth of 7 in. (17.8 cm). The ultimate fate of these particles could have been determined only by backwashings, which were not performed in this experiment.

PAC particles may become another source of attached bacteria in the product water, especially if their residence time within the filter-adsorber is long enough to establish a bacterial community. Most PAC particles have diameters >10 μm .¹⁶ Theoretical calculations of single-collector efficiency for particle capture in a filter suggest effective removal of these size particles,¹⁷ but the data from

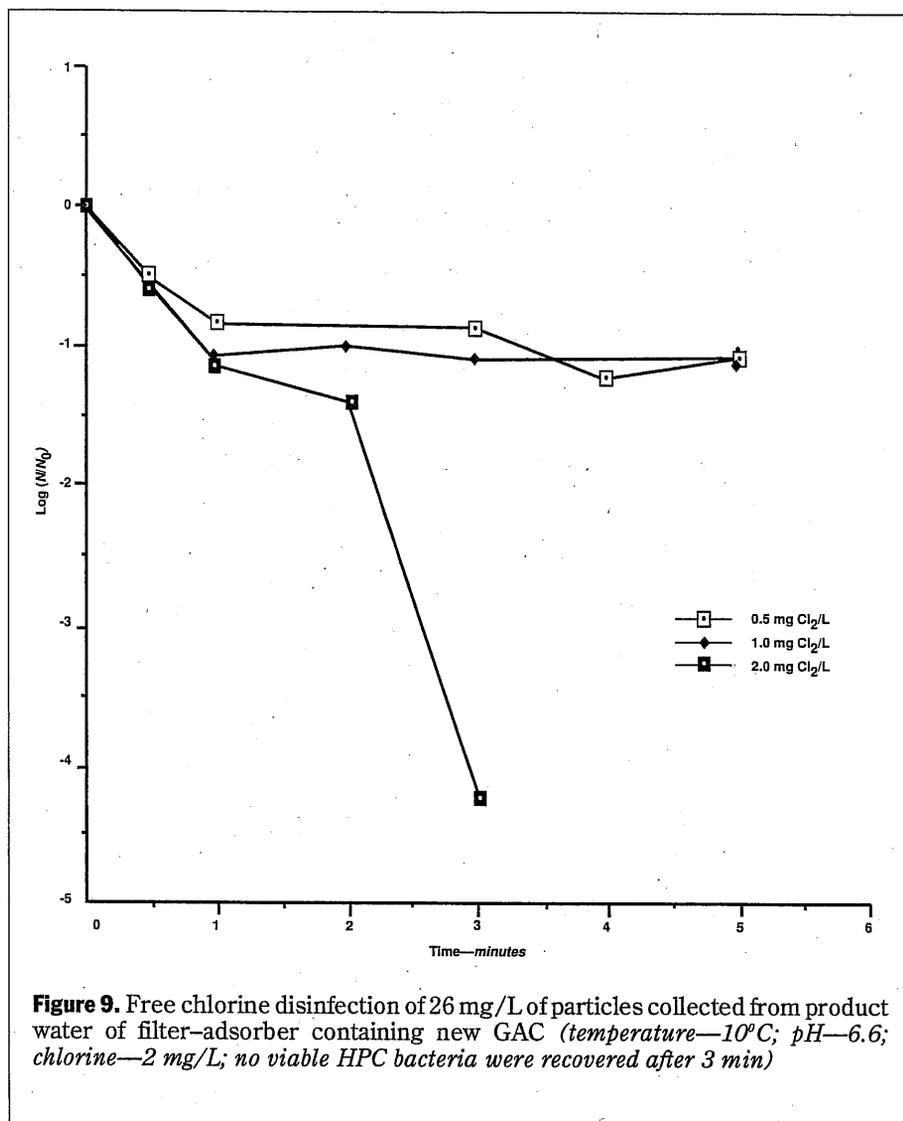


Figure 9. Free chlorine disinfection of 26 mg/L of particles collected from product water of filter-adsorber containing new GAC (temperature—10°C; pH—6.6; chlorine—2 mg/L; no viable HPC bacteria were recovered after 3 min)

this research showed their escape. Particle stability considerations may offer an explanation. That is, single-collector efficiency calculations describe only physical transport to the filter media and not the chemical factors affecting attachment to the media. Filtration efficiency must include collision efficiency, which in turn depends on particle stability. Chemically destabilized particles have a high collision efficiency because they are "sticky." However, it seems reasonable that PAC particles that have escaped sedimentation may be very stable. Thus, despite having large diameters that would favor capture, the particles are stable, which may allow escape from the filter-adsorber. More research is needed to determine the stability of PAC particles entering a filter.

Physical and chemical characterization of released particles. Energy-dispersive X-ray analysis was conducted to provide a partial elemental analysis of particles collected in the product water. Samples of known materials (e.g. clays, sand, alum flocs, activated carbon) were examined to determine whether the elemental pro-

files of these materials could be distinguished from each other. The spectra and SEM photograph of an alum floc particle and of an activated carbon particle (laboratory sample) are shown in Figures 5 and 6, respectively. The spectra are clearly different. The aluminum peak in Figure 5 is due to alum addition, whereas the silica, iron, and manganese peaks most likely originate from the water being treated. In contrast, the spectra of the activated carbon particle do not reveal any significant peaks except sulfur, which may be indicative of a bituminous carbon. The full-scale voltage response was also tenfold less than that of the floc particle. Because carbon does not have a characteristic spectral peak, it was only possible to conclusively prove a negative event, i.e., that a particle was not carbon.

Energy-dispersive X-ray analysis was then performed on samples of fines obtained from the clearwells of each of three filter media (new GAC over sand, old GAC over sand, and sand). The results for analysis of two particles obtained from the new-GAC-over-sand filter

are presented in Figures 7 and 8. The spectra of the particle in Figure 7 generally resemble that of activated carbon, although the sulfur peak was not found and a chlorine peak was noted. The chlorine peak of these spectra could very well be due to the presence of sorbed chlorinated natural organic matter, generated as a product of prechlorination, or sorbed chloride. The spectra of a particle that is clearly not carbon are presented in Figure 8. This particle is most likely a clay or alum floc. Examination of the partial

elemental profiles demonstrated that not all the particles collected on the 5- μm filters were carbon particles.

Particles in filtered (0.2- μm filter) grab samples of new GAC product water were examined by SEM. Of 181 particles examined, only nine were $>20\ \mu\text{m}$. Stewart et al¹¹ collected particles on a 10- μm filter from a pilot GAC filter without post-GAC sand filtration and found particles ranging in size from 2 to 40 μm in diameter.

Camper and co-workers^{9,10} collected samples of product water from filter-adsorbers in full-scale plants and found black particles. These particles were identified as activated carbon and ranged in diameter between 100 and 350 μm . The presence of such large particles could be due to nonoptimal underdrain conditions, but specific information as to underdrain condition was not given.^{9,10} Particle colonization can be affected by both particle size and surface character. Ridgway and Olson⁸ examined particles collected from a distribution system and found that significant colonization only took place on particles larger than about 10 μm . However, it should be noted that these particles were captured from the product water of a plant that did not use GAC filter-adsorbers; thus, their finding does not rule out attachment on smaller-size GAC particles. Disinfection of large, colonized particles may be of more concern than small particles because it is well known that the diffusion flux of a solute (in this instance, the disinfectant) is inversely related to the square of particle diameter.

Characterization of bacterial attachment to particles. Electron micrographs of sections of the 293-mm-diameter, 5- μm polycarbonate filters showed that several types of material had been collected. Many trapped particles were at least partially coated with what appears to be biofilm or floc. However, the majority of the particles did not have attached bacteria. In addition, clumps of material unassociated with defined particles were collected. These clumps may be sloughs of biofilm or alum floc that penetrated the filter-adsorber.

Not many of the particles examined by SEM had attached bacteria. Of 181 particles counted (captured by a 0.2- μm filter) from grab samples of new GAC product water, 13 were colonized by bacteria to some extent. Thus, it would appear that only 7 percent of the particles harbored attached bacteria.

The mean attached HPC, as measured by those collected on the 5- μm polycarbonate filter, is shown in Table 6 for runs 4 and 5. These attached bacteria are expressed in two ways: (1) attached HPC/L of product water and (2) attached HPC/ μg of particles. Approximately 2,000 to 3,000 attached bacteria/L were found in GAC filter-adsorber product waters in contrast to <100 attached bacteria/L in the product water of the sand filter. The particles in product water from new GAC, old GAC, and reactivated GAC contained about the same density of bacteria per mass of particles. The bacterial density found on the particles is within the upper range for that found in GAC beds.^{1,10} In contrast, particles collected from the product waters of the sand filter had a much lower bacterial density per particle than those of the filter-adsorbers (Table 6).

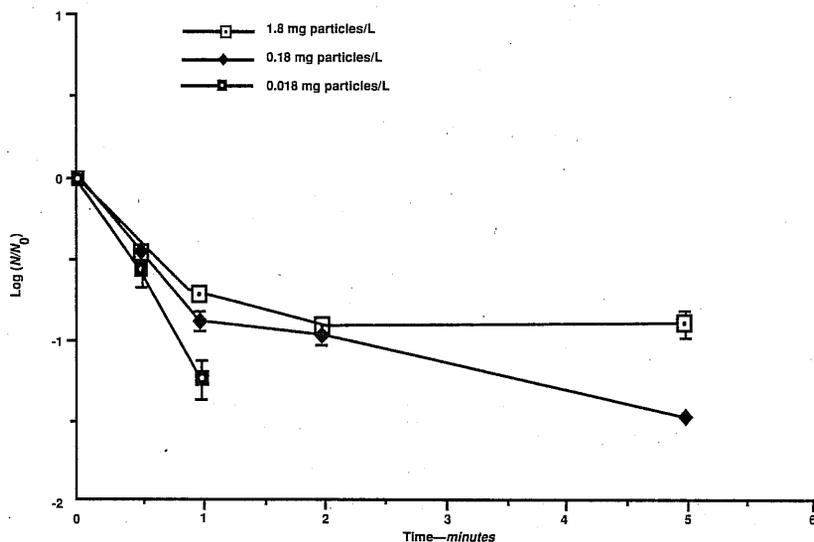


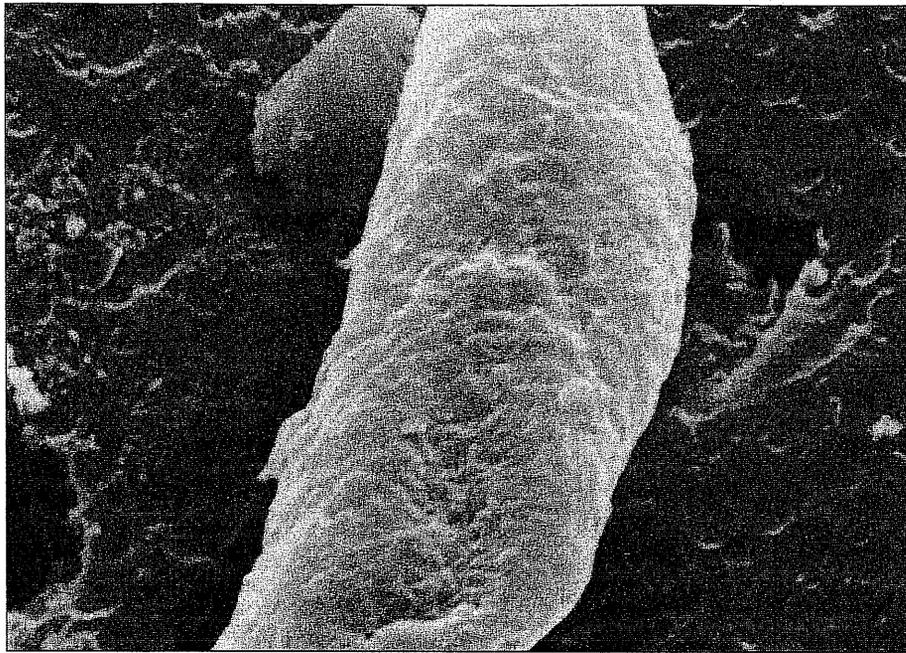
Figure 10. Free chlorine disinfection at three concentrations of particles collected from product water of filter-adsorber containing new GAC (temperature—10°C; pH—6.6; chlorine—2 mg/L; each of two replicate measurements [N] of HPC/mL taken at any time were normalized by the average of the two initial replicate values (N₀) of HPC/mL to plot the N/N₀ values shown by the bars)

TABLE 4
Product water turbidity for run 4

Filter Type	Turbidity—ntu		
	Number of Samples	Mean	Standard Deviation
New GAC	52	0.025	0.0024
Two-year-old GAC	52	0.027	0.0019
Reactivated GAC	52	0.046	0.0018
Sand	52	0.039	0.0023

TABLE 5
Particles in product water captured on 5- μm polycarbonate filter during runs 4 and 5

Filter Type	Particles Captured— $\mu\text{g/L}$		
	Number of Samples	Mean	Standard Deviation
New GAC	8	8.8	6.4
Two-year-old GAC	7	12.5	9.3
Reactivated GAC	2	8.6	(0.7)
Sand	2	14.0	(5.5)



This section of a 5-µm polycarbonate filter shows a particle trapped from new GAC product water without attached bacteria (the case for the majority of the particles).

As in other studies, not all the bacteria recovered by this technique were attached to particles, nor were they attached specifically to activated carbon. However, for this research, bacteria captured by the 5-µm filter were defined as attached bacteria, even if they were only attached to one another. The collection technique precludes distinguishing the difference in disinfection efficacy for clumped and truly attached bacteria.

Other studies that examined attached bacteria in GAC product waters did not quantify the number of attached bacteria per volume of product water.^{1,9,10,11} However, Stewart et al¹¹ reports finding between 10 and 62 particles/L, 77 percent of which were colonized with between one and 50 bacteria. Combining these figures would indicate the presence of 10–3,100 attached bacteria/L, about the same order of magnitude as found in this study. It should also be noted that Ridgway and Olsen⁸ found between 1,500 and 15,000 particle-bound bacteria/mL in typical drinking water distribution systems that were not receiving GAC-treated water.

The total number of bacteria in individual product water samples taken in run 5 are compared with the number of attached bacteria in Table 7. Attached bacteria were never found to be >0.1 percent of the total HPC in any one sample. Furthermore, the percentage of attached bacteria was similar for the GAC filter-adsorber and the sand filter product waters.

It has been shown that bacteria in aggregates or attached to surfaces are more resistant to disinfection.^{7,8} Therefore the most important question to be addressed was whether this small but

significant percentage of attached bacteria captured by a 5-µm filter could escape the chlorine disinfection barrier and enter the distribution system.

Disinfection of particles. Particles were collected from the product water of a filter-adsorber containing new GAC in run 5 and then diluted to a concentration of 26 mg/L. These solutions were treated with 0.5, 1.0, and 2.0 mg free chlorine/L to examine the disinfection of the at-

tached bacteria. The HPC remaining (N) were normalized by the initial HPC (N_0) and plotted as $\log N/N_0$ over time; these results are shown in Figure 9. Increasing the chlorine dose (>0.5 mg/L) increased the extent of disinfection despite the very high concentration of particles present (26 mg/L). No viable HPCs (plotted as 1 HPC/mL in Figure 9) were recovered after 3 min when 2 mg/L chlorine (1.25 mg/L free chlorine residual after 5 min) was added.

The particles collected from the product water of the filter-adsorber were concentrated 3,000 times in order to conduct the disinfection kinetic experiment at 26 mg/L. The resistance to disinfection is not surprising given that previous researchers^{9,11} found that bacteria attached to GAC media and to particles released from filter-adsorbers could not be disinfected after a particle concentration step (400 to 30,000 times) that produced concentrations on the order of grams per litre.

The influence of particle concentration on disinfection efficacy was investigated in a separate series of experiments using the highest of the three chlorine doses (2 mg/L). Particles were again collected from a GAC filter-adsorber containing new GAC. Solutions for disinfection were prepared by diluting these particles to yield concentrations of 0.018, 0.18, and 1.8 mg/L. The normalized HPCs remaining (N/N_0) are shown over the 5-min disinfection period in Figure 10 for each particle concentration. Although a chlorine demand of 0.75 mg/L was found

TABLE 6
Attached heterotrophic bacteria per litre of product water and per milligram of particles in runs 4 and 5

Filter Type	n^*	Mean Attached Bacteria HPC/L	Standard Deviation	Mean Attached Bacteria HPC/µg	Standard Deviation
New GAC	7	2,290	2,340	405	420
Two-year-old GAC	6	2,390	1,710	310	240
Reactivated GAC	1	3,540		430	
Sand	2	70	(60)	5	(2)

* n —number of samples

TABLE 7
Comparison of attached and unattached bacteria in product water in run 5

Filter Type	n^*	Attached Bacteria† HPC/L	Average Total Bacteria‡ HPC/L $\times 10^{-6}$	Attached Bacteria percent
New GAC	5	1,060	9.44	0.01
Two-year-old GAC	6	1,940	8.49	0.02
Reactivated GAC	1	2,810	12.5	0.02

* n —number of samples

†Calculated based on volume of water from which particles were collected by the 5-µm polycarbonate filter and bacteria subsequently detached

‡Calculated directly from samples of product water

using 26 mg/L of particles in the previously discussed experiment (Figure 9), the chlorine demand at the highest particle concentration used in this experimental series (1.8 mg/L) was insignificant; thus chlorine residual was nearly 2 mg/L after 5 min. However, the results in Figure 10 suggest that only a one-log reduction could be achieved at the highest concentration of particles. Disinfection efficiency improved when particle concentration was lowered to 0.18 mg/L; however, bacteria were still recovered (10 HPC/mL) after 5 min. Further improvement in disinfection was achieved at solids concentrations approaching the actual ambient concentration found in product waters. At a particle concentration of 0.018 mg/L, no culturable bacteria were found after 2 min at 2.0 mg chlorine/L. The difference between this result and those at higher particle concentrations is clearly significant.

Disinfection studies were conducted on another sample of product water from run 5, which contained 0.014 mg/L particles and was treated with 0.5 instead of 2 mg chlorine/L.¹³ No culturable bacteria were found at the end of 1 min. Again, increasing the particle concentration (to 4 and 40 mg/L in this experiment) produced less than one log reduction in HPC. However, these results are not easy to interpret because the combination of low chlorine dosage and relatively high particle concentration produced no measurable chlorine at the end of 2 min of contact.

Conclusions

The concentration of particles found in the product water from GAC filter-adsorbers was in the microgram-per-litre range. These concentrations were about the same for GAC filter-adsorbers and sand filters, which suggests that the mass of carbon fines entering the product water was small. The amount of particles released appeared to be independent of the condition of the GAC (new, old, or reactivated). Some, but not all, particles were populated with attached bacteria. Bacteria that were clumped together but did not appear to be attached to a particle were also collected.

Both SEM and energy-dispersive X-ray analysis were useful in examining the nature of particles found in product water. Both chemical floc and activated carbon were released from the GAC filter-adsorbers and captured on the 5- μ m polycarbonate filters. The majority of the particles were <20 μ m in diameter and were not highly populated with bacteria.

Attached bacteria were shown to represent a small fraction of the total bacteria released by GAC filter-adsorbers. When particles were collected on three occasions during one filter-adsorber run, results suggest that attached bacteria were not efficiently disinfected by

chlorine at high particle concentrations (orders of magnitude greater than actually observed in the product water). Increasing the chlorine dosage improved disinfection at high particle concentrations. Decreasing the particle concentration to within the range of particle concentrations found in GAC filter-adsorber product waters demonstrated that attached HPC bacteria could be disinfected by chlorine.

The results presented here suggest that bacteria attached or associated with particles in product water from filter-adsorbers will not penetrate the chlorine barrier if particle concentrations are low. Another factor that may have influenced the efficacy of disinfection is particle size. The majority of particles observed to be released were <20 μ m, but it is expected that larger particles may be more difficult to disinfect because of limitations in the diffusion rate of the disinfectant into the particle. The efficiency of disinfection may also be inhibited by poor filter performance or the release of high concentrations of GAC fragments. In this study, the concentration of carbon fines was very low, and product water quality was high. Thus disinfection of the product water was not difficult.

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