A pilot-scale study of *Cryptosporidium*-sized microsphere removals from swimming pools via sand filtration

Ping Lu and James E. Amburgey

ABSTRACT

Cryptosporidium species are the most common cause of gastrointestinal illness in treated recreational water venues. In order to protect public health during swimming, *Cryptosporidium*-sized microsphere removals by high-rate sand filtration with six coagulants were evaluated with a 5.5 m^3 pilot-scale swimming pool. A sand filter without coagulation removed 20–63% of *Cryptosporidium*-sized microspheres. *Cryptosporidium*-sized microsphere removals exceeded 98% by sand filtration with five of the six tested coagulants. Continuously feeding coagulants A, B, and F (i.e., organic polymers) led to coagulant accumulation in the system and decreased removals over time (<2 days). Coagulant E (polyaluminum chloride) consistently removed more than 90% of microspheres at 30 m/h while the removals dropped to approximately 50% at a filtration rate of 37 m/h. Coagulant C was a chitosan-based product that removed fewer microspheres compared with other products, <75%, under the studied conditions. Results indicated aluminum-based coagulants (coagulants D and E) had an overall performance advantage over the organic polymer based coagulants primarily in terms of their tendency not to accumulate in the water and cease to be effective at improving filter efficiency.

Key words | coagulation, *Cryptosporidium*-sized microspheres, domestic waste, filtration, pilot-scale swimming pool, recreational water treatment

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INTRODUCTION

Cryptosporidium species are the most common cause of gastrointestinal illness associated with recreational water venues (Juranek 1995; Marion *et al.* 2010) and numerous waterborne outbreaks of cryptosporidiosis have been linked to swimming pools in the United States, United Kingdom, Australia, etc. (LeChevallier *et al.* 1991; Lisle & Rose 1995; Public Health Laboratory Service (PHLS) 2000; Puech *et al.* 2001; Briancesco & Bonadonna 2005; Karanis *et al.* 2006; Lu *et al.* 2013).

For filtered and disinfected aquatic venues, *Cryptosporidium* is a public health threat, because it is extremely resistant to halogen disinfection and its small size makes it a challenge for filtration systems (Korich *et al.* 1990; Carpenter *et al.* 1999). The drinking water industry has demonstrated that *Cryptosporidium* removal throughout all stages of the classical treatment process is largely influenced by the effectiveness of coagulation pretreatment (Dugan *et al.* 2001; Amburgey 2002; Amburgey *et al.* 2004; Hankins *et al.* 2006; Cummins *et al.* 2010; Karim *et al.* 2010; Lopez *et al.* 2010). Particles tend to repel each other, and there is no natural tendency for them to attach to other particles or surfaces unless effective coagulation to destabilize the particles is performed (Edwards 1997; Gao *et al.* 2002). Inorganic trivalent metal ions (such as aluminum and ferric iron) and water-soluble organic polymer coagulants are widely used for particle and natural organic matter (NOM) coagulation (Polasek & Mutl 2002; Bolto & Gregory 2007). Charge neutralization and sweep coagulation are the predominant mechanisms for *Cryptosporidium* removal via coagulation (Butkus *et al.* 2003; Xagoraraki & Harrington 2004; Okuda *et al.* 2006). The largest removals in drinking water studies have been shown to occur at high alum

doses where aluminum hydroxide precipitation was observed to be extensive (Xagoraraki & Harrington 2004). Since polymer addition generally does not impact the pH of the water being treated, pH adjustment is not necessarily required for optimal coagulation (Emelko & Huck 2003). Polymers acting as coagulants usually contain materials with high charge density (Bolto & Gregory 2007). Polydially-dimethyl ammonium chloride (polyDADMAC) is a commonly used water-soluble polymer. PolyDADMAC was found to be very effective for removing disinfection by-product (DBP) precursors and NOM when used as a primary coagulant or coagulation aid (Polasek & Mutl 2002; Chang *et al.* 2005; Hankins *et al.* 2006; Wei *et al.* 2010). Natural cationic polymers, such as chitosan, were reported to coagulate with particles and enhance the particle removals for drinking water applications (Guibal *et al.* 2006; Bolto & Gregory 2007; Fabris *et al.* 2010). Chitosan has a low charge density (Parsons *et al.* 2007). *Cryptosporidium parvum* oocyst removals by filtration following chitosan coagulation at optimal dosages were comparable to those achieved when filtration (at 10 m/h) was preceded by alum or iron coagulation during optimized operation in a drinking water pilot plant (Brown & Emelko 2009). The effectiveness of coagulation depends on dosage (Divakaran & Pillai 2001). Investigation indicated chitosan coagulation at a dosage less than 1.0 mg/L did not result in appreciable improvements in *Cryptosporidium parvum* oocyst removal (Brown & Emelko 2009).

Direct filtration is similar to conventional treatment in that a coagulant is used to form larger particles, but coagulated water is applied directly to the filters without a gravity separation step. Direct filtration is generally used for low and consistent turbidity water. The removal of *Cryptosporidium* in direct filtration is usually lower than that in conventional drinking water treatment (Nieminski *et al.* 1995; Nieminski & Ongerth 1995; Amburgey 2002). A two-year evaluation of *Cryptosporidium* was conducted at a full-scale treatment plant and a pilot plant operating with coagulation and direct filtration (Nieminski & Ongerth 1995). Results showed 99.87% (2.9 log) of *Cryptosporidium* was removed when the treatment plant produced water of consistently low turbidity (i.e., 0.1–0.2 NTU). There is minimal published research on *Cryptosporidium* removals from swimming pools by filtration with coagulation.

The primary objective of this study was to determine the approximate level of *Cryptosporidium* oocyst-sized microsphere removals that can be achieved in a swimming pool system through high-rate sand filtration with six coagulants. Removals of microspheres, particles, turbidity, and organic matter (indicated by UV_{254}) were evaluated for each coagulant in addition to pressure loss across the filter.

MATERIALS AND METHODS

Experimental setup

A 5.5 m^3 swimming pool was built with a filtration system and a chemical control system. Pool water was pumped through the sand filter as shown in Figure 1. The smaller sand filter was made from transparent polyvinyl chloride (PVC) pipe, which utilized an integral media support cap (Leopold, ITT) as support for filter media as well as backwash flow distribution. The filter had a surface area of 0.02 m^2 and a sand depth of 0.3 m. The effective size of the sand was 0.49 mm. The hydraulic loading rate (HLR) for the sand filter was 37 m/h, which is a typical high-rate filter loading rate used in US swimming pools. All chemicals and microspheres were fed using peristaltic or metering pumps. The pool's pH and oxidation reduction potential (ORP) sensors were mounted in a bypass line as shown in Figure 1. These sensors were connected to a controller (CAT 5000, Poolcomm, Rockville, MD, USA) for monitoring and automatic chemical feed control. Coagulant and microspheres were fed into the pipe ahead of the pump, and a streaming current meter (Micrometrix, Suwanee, Georgia, USA) was installed on the sample influent line to approximate the surface charge of the particles entering the filter. Turbidimeters (HF scientific, Fort Myers, Florida, USA), particle counters (Chemtrac, Norcross, Georgia, USA), and UV transmission monitors (HF scientific, Fort Myers, Florida, USA, and Real Tech Inc., Canada) were installed both on filter influent and effluent lines. The reading of the turbidimeter is more strongly influenced by the number of submicron particles (<1 μm) present in the sample (Hunt 1993; Gregory 1994). Particle counters collected the particle size data in the range of 2 μm to 100 μm.
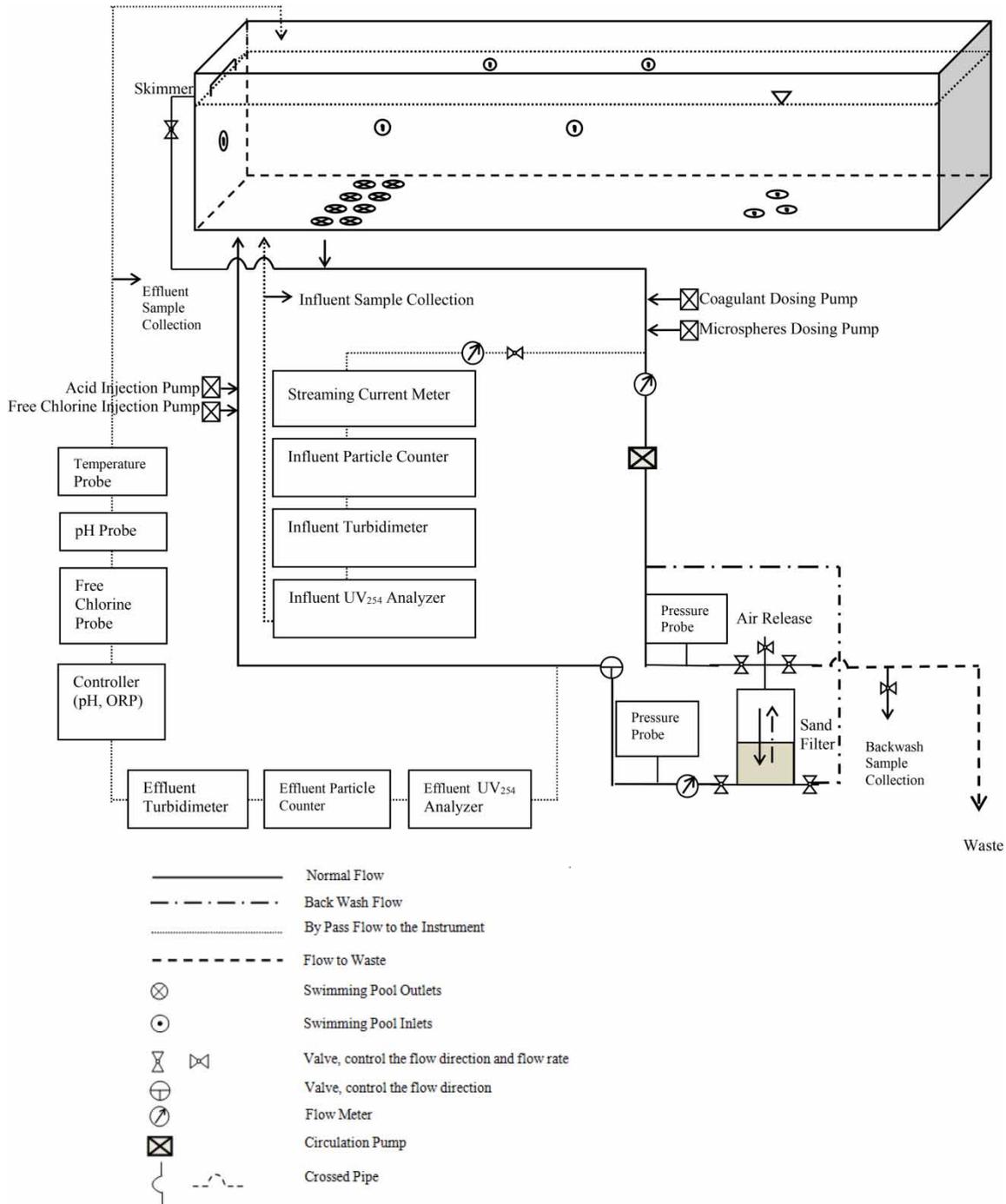
Figure 1 | Experimental set-up.

Synthetic pool water

The recipe for the synthetic pool water was based on a previous swimming pool water quality investigation (Lu 2012).

Chemical characteristics of the simulated swimming pool water samples were pH of 7.5, hardness of 200 mg/L, as $CaCO_3$, alkalinity of 100 mg/L as $CaCO_3$, and free chlorine of 2 mg/L. Approximately 5,500 L of Charlotte, NC tap

water with dissolved organic carbon (DOC) of approximately 1 mg/L was supplemented with $NaHSO_4$, $CaCl_2$, $NaHCO_3$, and $Ca(OCl)_2$ to create the synthetic pool water. A synthetic bather load based on the major constituents of sweat and urine was shown to have no impact on the surface charge or particle removal and was not used in these experiments (Lu 2012).

Cryptosporidium-sized polystyrene microspheres and coagulants

The use of polystyrene microspheres as an oocyst surrogate has been done by multiple researchers and was also done in this study (Li *et al.* 1997; Amburgey 2002, 2011; Dai & Hozalski 2003; Amburgey *et al.* 2004, 2005, 2012). Microspheres with a diameter of 4.5 μm (Fluoresbrite™ Carboxylate YG microspheres, Cat. #16592, Polysciences, Inc., Warrington, Pennsylvania, USA) were used as the surrogate since these microspheres are virtually identical to *Cryptosporidium* oocysts in size, shape, density, and surface charge in pool water (Amburgey 2010). The concentration of the stock suspensions of microspheres was 4.37×10^{11} #/L. A diluted suspension of microspheres with a concentration of 4.37×10^8 #/L was prepared by 1:1,000 dilutions of the stock solution. Microsphere samples were mixed by vortexing and hand shaking for at least two minutes each before analyzing. Samples were passed through 3.0 μm pore size polycarbonate track-etched filters. Each polycarbonate filter was mounted on a glass microscope slide with a polyvinyl alcohol-DABCO solution, covered with a glass cover slip and counted under an epi-fluorescent microscope at 100× total magnification (Freer 1984; Harlow & Lane 1988; Arrowood 2002). For ease of counting and obtaining statistically valid data, microscope slides needed to contain between 10 and 150 microspheres. Removal efficiency was calculated by comparing the concentrations between influent and effluent samples. Six coagulants used in pool water treatment were selected. Coagulants A and B were polyDADMAC products, coagulant C was chitosan, coagulant D was an aluminum-based coagulant, and coagulant E was polyaluminum chloride (Robarb 2000; BioGuard 2001; SeaKlear 2008; Vantage 2009; Kemira 2012). The formulation of coagulant F was proprietary.

General experimental approach

Experiments were performed over multiple theoretical hydraulic detention times (or turnovers) of the swimming pool, which were 8 hours each. Coagulants were fed at one vendor-recommended dose every turnover. Extended feeding of coagulants was evaluated. Samples were taken during each turnover. High-rate sand filtration control experiments without coagulant addition were also conducted. A backwash was performed after each experiment along with replacing the water in the pool and the sand in the filter.

The concentration of polyDADMAC was analyzed by the UV-Visible (UV) spectrophotometry method (Parazak *et al.* 1987) to identify the existing polyDADMAC concentration in the bulk water, and to find out if polyDADMAC accumulation impacted microsphere removals. The UV spectrophotometry method was proven to be simple, economical, and effective. The method involves the formation of an insoluble complex between the cationic polymer and the anionic dye Ponceau S. The polymer/dye complex precipitates out of solution and is collected at the interface between the aqueous layer and a solvent. The aqueous layer is collected, and the concentration of dye remaining in solution is measured by a UV Spectrophotometer at 520 nm (Agilent Technologies, Varian Cary 100 BIO UV, Santa Clara, California, USA). Ponceau S (200 mg/L) (Fisher BioReagents), 0.5 M sulfuric acid (Fisher BioReagents), and dichloromethane (Fisher BioReagents) were used in this assay. The detailed experimental description can be found elsewhere (Fielding *et al.* 1999). The absorbance of the standards versus polyDADMAC concentration calibration plot is shown in Figure 2. Standards with known polyDADMAC
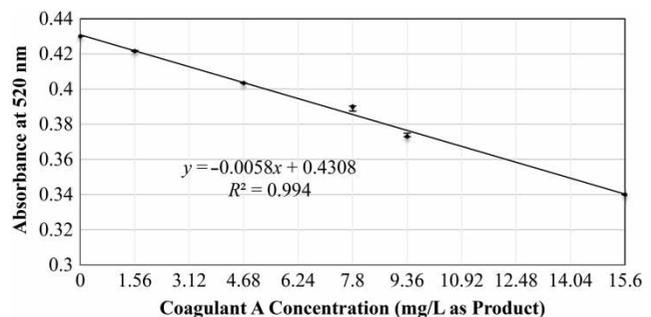

$$y = -0.0058x + 0.4308$$
$$R^2 = 0.994$$

Figure 2 | Absorbance versus coagulant A concentration calibration curve (10 mm cell).

concentrations were made of tap water and polyDADMAC polymer (coagulant A). A calibration plot was created and the regression equation is shown in Equation (1).

$$y = -0.0058x + 0.4308 \tag{1}$$

where y is the absorbance at 520 nm for the standard and x is the concentration of coagulant A in the standard.

Data analysis

Box plots were used to represent the results for each coagulant. A box plot is a convenient way of graphically depicting groups of numerical data through their five-number summaries: the smallest observation (sample minimum), lower quartile, median, upper quartile, and largest observation (sample maximum).

Quality assurance and quality control

A control experiment was conducted without filter media to test if there were microsphere losses in the system. An average of 1% removal (approximately zero) was obtained and demonstrated no significant system losses. Duplicate experiments were conducted and triplicate samples were taken. The swimming pool was drained, rinsed, refilled, and recirculated with tap water at least three times between experiments to limit any cross-contamination between experiments. Fresh sand was used for each experiment and was backwashed (at 55 m/h and 29 °C to obtain a bed expansion of approximately 30–40%) with simulated pool water for 5 minutes before experiments to ensure the sand was clean and the sand grains were restratified.

RESULTS AND ANALYSIS

The zeta potential of simulated pool water with 10^6 microspheres/mL versus coagulant concentration is plotted in Figure 3 for all six coagulants. Titration experiments suggested the zeta potential of suspensions increased in the positive direction as coagulant dosage increased. The manufacturers' recommended dosages were 1.56 mg/L for coagulants A, B, C, and F, 305 g/m² of filter surface area for coagulant D, and
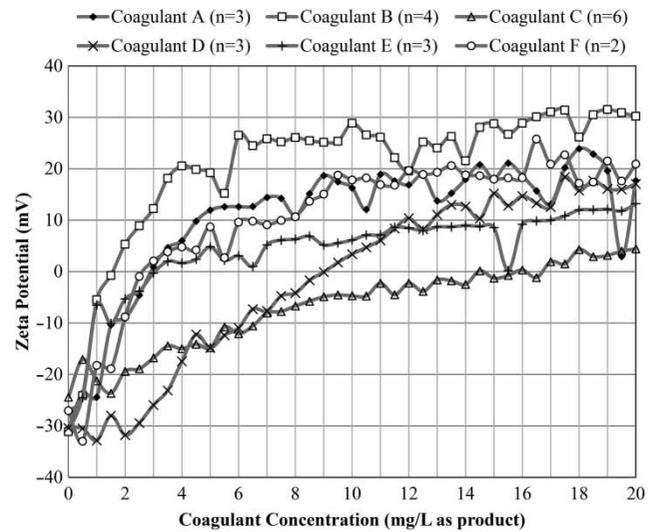

Figure 3 | Zeta titration for six coagulants into simulated swimming pool water (10^6 #/mL microspheres) (for coagulant D: 1 mg/L = 160.5 g/m²; for coagulant E: 1 mg/L as product = 0.1 mg/L as Al).

1 mg/L as product (0.1 mg/L as Al) for coagulant E (Goodman 20π). A previous study of drinking water revealed that achieving a zeta potential of between −10 mV and 10 mV for the suspension being treated was an effectively destabilized system (Tseng et al. 2000). Coagulants A, B, and F had dosages between 0.5 mg/L and 3 mg/L as the product, coagulant D had a dosage between 6 mg/L (963 g/m²) and 12 mg/L (1,926 g/m²) as a product, and coagulant E had a dosage between 0.1 and 1.6 mg/L as Al (1 and 16 mg/L as product), resulting in microsphere zeta potentials of −10 mV to 10 mV for simulated pool water. Coagulant C could not achieve a microsphere zeta potential of −10 mV at the recommended dosage (1.56 mg/L). Zeta potential values greater than 10 mV were observed for all coagulants except C, as shown in Figure 3.

A suspension of microspheres was used as a model colloid and coagulated by these cationic coagulants. The most likely mechanism of coagulation by these cationic coagulants is charge neutralization (Singley 1970; Bratby 2008; Letterman & Yiacoumi 2010).

Cryptosporidium-sized microsphere removals

Control experiments without coagulant were conducted over 24 hours and followed by a backwash. Microsphere removals were consistently in the 20%-63% range during these experiments, as shown in Figure 4.
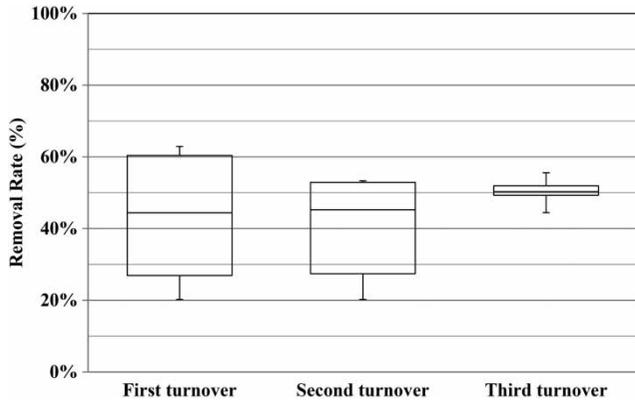
Figure 4 │ Microspheres removal by sand filter during 24 hrs *(1.8 #/mL microspheres, 30 cm sand, 37 m/h filtration rate).*

Coagulants were continuously fed into the system at the rate of one recommended dose per turnover (e.g., 1.56 mg/L every 8 hours). Figure 5 shows *Cryptosporidium* oocyst-sized microsphere removals by high-rate sand filtration preceded by continuous feeding of coagulant at one recommended dosage every 8 hours (i.e., one turnover). Up to 99% microsphere removals were obtained by filtration with coagulant A as shown in Figure 5(a). Microsphere removals decreased to less than 90% with continuous feeding of coagulant A after three turnovers (or 24 hours). Table 1 shows coagulant A concentration in each turnover. The concentration of coagulant A in the pool water after 24 hours of feeding was approximately three times the recommended dose.
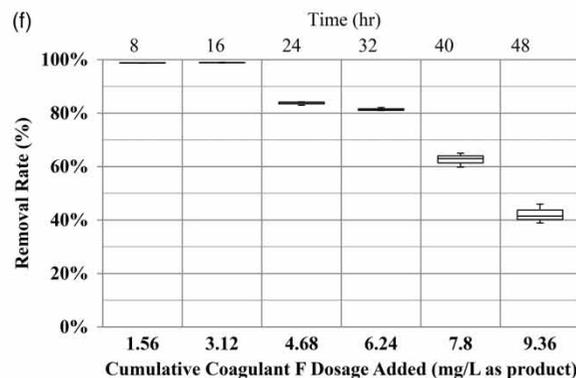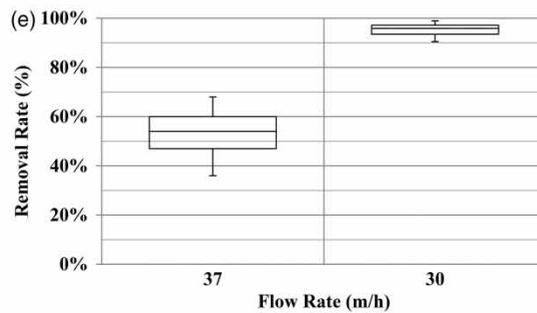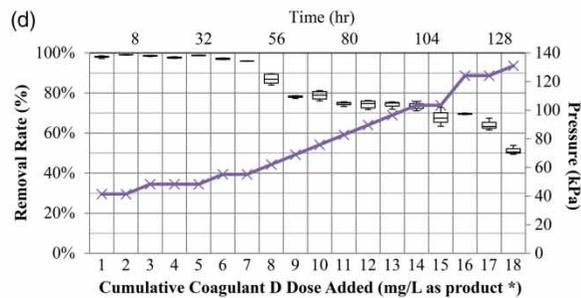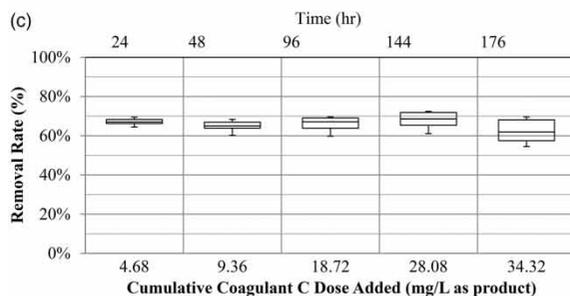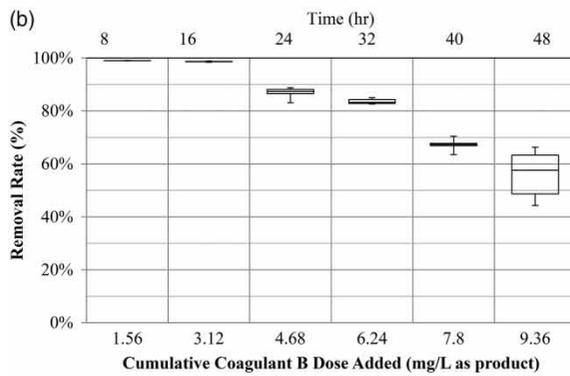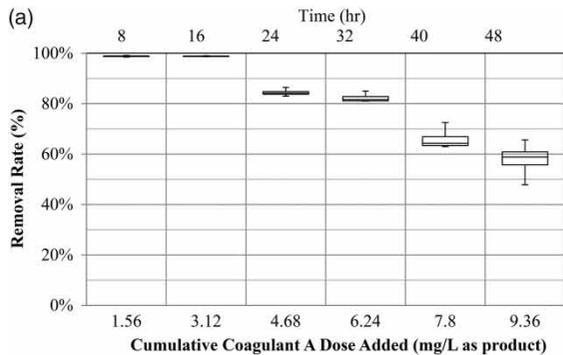

Figure 5 │ Microsphere removals by high-rate sand filtration with coagulants, A, B, C, D* (for coagulant D: 1 mg/L = 160.5 g/m²), E, and F (duplicate experiments with triplicate samples).

Table 1 │ Measured and estimated coagulant a concentration in each turnover

Time (hr)	Influent concentration (mg/L)[a]	Effluent concentration (mg/L)[b]	Average influent and effluent (mg/L)[c]	Estimated concentration (mg/L)[d]	Differences between 'c' and 'd'[e]
8	1.40	1.38	1.39	1.56	12%
16	2.76	3.03	2.90	3.12	7%
24	4.36	4.41	4.39	4.68	6%
32	5.86	6.07	5.97	6.24	4%
40	7.36	7.72	7.54	7.8	3%
48	9.16	9.09	9.12	9.36	3%

c: average of column a and b, which meant $c = (a+b)/2$.
e: difference between c and d, which meant $e = (c-d)/[(c+d)/2]*100\%$.

Based on the data in Table 1, it appears that polyDADMAC rapidly accumulated in the bulk water, which could help explain the rapid decline in removals over time as the effective coagulant dosage was exceeded and charge reversal occurred within 48 hours as witnessed by the removals dropping below 60%. Coagulant B performed similarly to coagulant A as shown in Figure 5(b). Microsphere removals by coagulant C were always less than 80% as shown in Figure 5(c). Figure 5(d) shows microsphere removals in each turnover by coagulant D. Up to 99% removals were achieved by coagulant D in the first 56 hours by continuously feeding coagulant D, but the removals dropped as the pressure drop across the filter increased. Microsphere removals were in the range of 35% to 70% with coagulant E at 37 m/h as shown in Figure 5(e). A similar result for coagulant E was also found previously (Goodman 2011). Greater than 90% of the microspheres were removed by coagulant E at a filtration rate of 30 m/h (0.1 mg/L as Al). Figure 5(f) shows microsphere removals by coagulant F. Coagulant F performed similarly to coagulants A and B (with removals up to 99%), but the removals all decreased over time. Decreased removals for coagulants D and E were correlated with filter pressure build-up/filter pore clogging (as will be discussed subsequently).

Particle counts, turbidity, UV$_{254}$, and pressure variation

The filter influent particle counts for coagulant A are shown in Figure 6(a). Total particle counts from 2–100 μm are shown. Intermittent peaks in influent particle counts were caused by seeding events of the 4.5 μm microspheres. No backwash was conducted until the end of the experiment.

A similar trend in particle counts was obtained by coagulants B, C, D, E, and F. Influent particles decreased over time in each microsphere seeding trial (as shown in Figure 6(a)). Similar trends of influent particle counts were observed for other coagulants and can be found elsewhere (Lu 2012).

Turbidity was another parameter used to evaluate the particle removal in this system. Turbidity variation for coagulant A is illustrated in Figure 6(b). Initial simulated pool water turbidity was approximately 0.2 NTU. Effluent turbidity for coagulant A was less than 0.2 NTU.

The approximate removal of organic matter was monitored by continuously measuring the UV transmittance at 254 nm. Influent and effluent UV$_{254}$ transmittance are shown in Figure 6(c). Both filter influent and effluent water UV$_{254}$ transmittance was in the range of 96% to 98% and showed no obvious trends. Similar UV$_{254}$ variations were observed for the rest of the five coagulants (data not shown). There was no significant removal of UV$_{254}$ in the filter system for these experiments. A previous study showed polymeric coagulants were not effective in reducing UV$_{254}$ absorption (Freese et al. 2001).

Figure 6(d) shows pressure variation for coagulant A. The initial filter influent pressure was 41.3 kPa, and effluent pressure was 20.7 kPa. Both influent pressure and effluent pressure for each coagulant increased during the experiments. There were no significant differential pressure increases for coagulants A, B, C, and F. However, the differential pressure increased significantly for coagulants D and E. The influent pressure increased 70 kPa over the original starting pressure, which could have been caused by the aluminum hydroxide precipitate from coagulants D and E being retained by the filter. Pressures decreased after
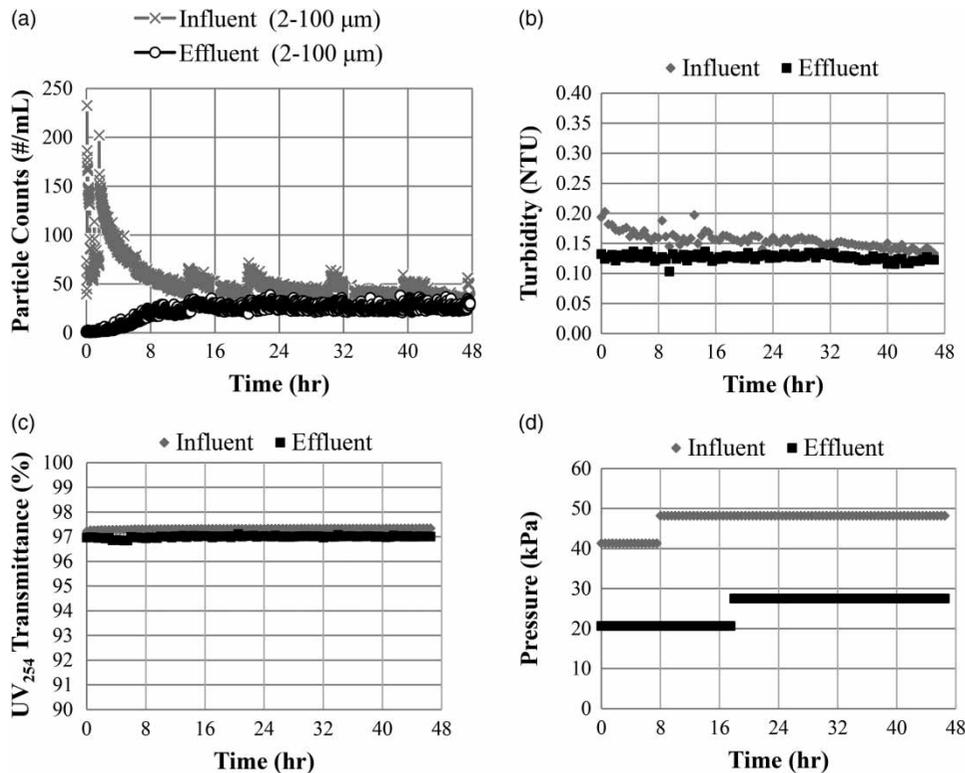
Figure 6 │ (a) Particle counts, (b) turbidity, (c) UV$_{254}$ transmittance, and (d) pressure variations over treatment time (*1.56 mg/L coagulant A, 1.8 #/mL microspheres, 30 cm sand, 37 m/h filtration rate*).

backwash for coagulants D and E. Pressure buildup (or more specifically pore clogging) for coagulants D and E could help to explain the decreased microsphere removals with time as shown in Figure 5(d). Figure 7 compares the pressure differentials for all six coagulants.

Coagulation evaluation

A streaming current meter (SCM) was installed on the filter influent sample line. Initially, the streaming current was negative. The streaming current increased when cationic coagulants were added, since negative particles can be charge neutralized by coagulants. Figure 8 displays the streaming current values for each coagulant over time. The streaming current typically increased to a certain value with the addition of coagulants A, B, and F from −200 streaming current units (SCU) to 400 SCU or 500 SCU, and the SCM detector became saturated at high polymer concentrations. Coagulants A, B, and F appeared to overdose, which was confirmed by decreased microsphere
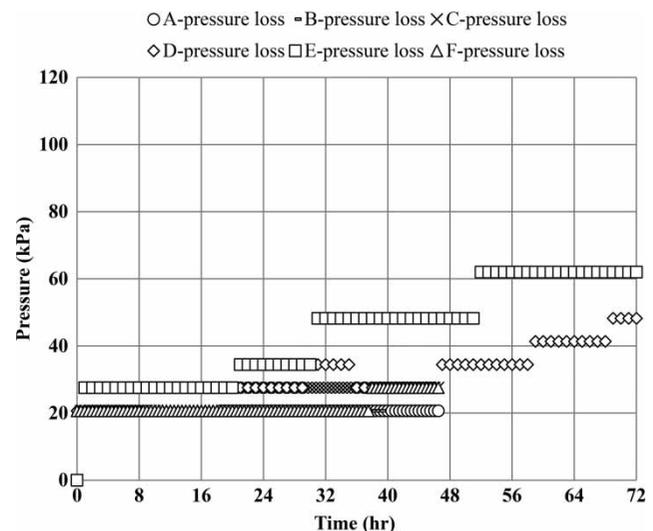

Figure 7 │ Pressure losses over treatment time for six coagulants (*1 recommended dosage of coagulant, 1.8 #/mL microspheres, 30 cm sand, 37 m/h filtration rate*).

removals, positive zeta potential measurements, and increasing polyDADMAC concentration in the pool with time. The streaming current did not change with the
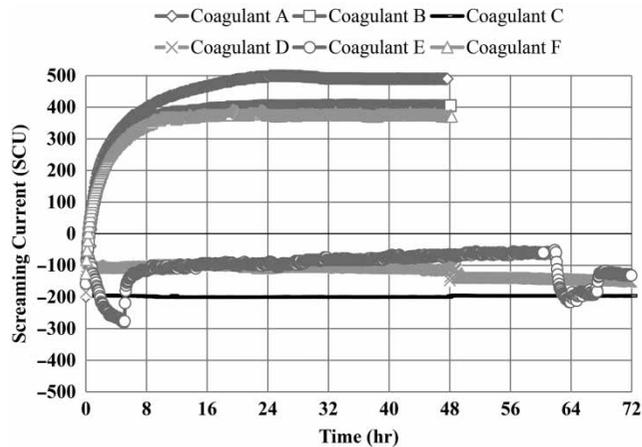
Figure 8 │ Streaming current variation over treatment time (1.56 mg/L coagulant A, 1.56 mg/L coagulant B, 1.56 mg/L coagulant C, 305 g/m² coagulant D, 0.1 mg·Al/L coagulant E, 1.56 mg/L coagulant F, 1.8 #/mL microspheres, 30 cm sand, 37 m/h filtration rate).

addition of coagulant C, which corresponded with the zeta titrations (as shown in Figure 2) and helped explain the poor microsphere removals obtained by coagulant C in Figure 5(c). Coagulants D and E did not show a tendency to overdose or accumulate in Figure 8.

DISCUSSION

PolyDADMAC based coagulant

Greater than 99% removal was achieved by polyDADMAC products, which indicated that polyDADMAC was a promising coagulant in *Cryptosporidium* removal from swimming pools. Similar performance for polyDADMAC coagulation of drinking water was reported previously in drinking water treatments (Bolto & Gregory 2007; Wei *et al.* 2009, 2010). However, the polyDADMAC product accumulated in the recirculating pool system after multiple turnovers, which would not occur in a drinking water system, due to the unique circumstance of swimming pool water treatment, where continuous circulation of pool water can lead to charge reversal. Charge reversal was also observed in the zeta potential titration results. The reversal of the zeta potential of the microspheres also indicated that the coagulant adsorbed onto the surface of the microspheres. A similar conclusion was made in the research on the

interaction between *Cryptosporidium* and coagulants (Bustamante *et al.* 2001).

Chitosan

Chitosan is a polymer with a low charge density (Parsons *et al.* 2007). In acidic solutions, chitosan becomes an extended chain and charged, while in neutral solutions, chitosan is a more coiled structure and only slightly charged (Pan *et al.* 1999; Huang *et al.* 2000). When pH values shift from 4 to 7, the positive charge on the chitosan surface significantly decreases, and the contribution toward charge neutralization by chitosan to destabilize particles is less significant in neutral pH conditions (Huang & Yin 1996; Parsons *et al.* 2007). However, swimming pool water pH must be maintained around 7.5 for human comfort and equipment safety concerns. The optimum dosage for chitosan coagulation would be expected to be smaller in acidic solutions (pH <7) due to the increased number of protonated amine groups on chitosan at lower pH, and the destabilization of particles has been enhanced by the increased number of charged groups (Pan *et al.* 1999; Huang *et al.* 2000). In addition, a non-charge coagulation mechanism has been proposed for chitosan (Parsons *et al.* 2007), but the non-charge mechanism did not appear to result in significantly increased microsphere removals by chitosan coagulation under the conditions examined in the present study.

Coagulant C (chitosan) could only remove up to 80% of *Cryptosporidium*-sized microspheres. This can be related back to surface zeta potential, in that chitosan could not achieve the minimum target zeta potential, −10 mV, at the recommended dosage of 1.56 mg/L as product under the experimental condition. The microsphere removals by coagulant C (chitosan) were contradictory to the research conducted on in-line filtration (in a non-swimming pool water treatment system) with chitosan at pH 7.3 to 7.4, which showed approximately 99% *Cryptosporidium* and microsphere removal with 1.5 mg/L chitosan (Brown & Emelko 2009). Possible reasons for the differences in removal for chitosan between the two studies include: chitosan source, physical and chemical processing of the chitosan, chitosan concentration, coagulant demand, filtration rates, flocculation, and media size and depth. In

particular, the filtration rate was 10.4 m/h compared with 37 m/h in this study.

Aluminum based coagulants

Alum has previously performed better than the polyDAD-MAC and chitosan on NOM removal from drinking water (Bolto 2001). However, filter pressure loss increased significantly compared to the other coagulants used in this study. With aluminum-based coagulants in the present study (i.e., coagulants D and E), there did not appear to be a tendency toward overdosing/charge reversal over time under the conditions studied, which was not the case for cationic polymer coagulants. However, removals by aluminum-based coagulants were sensitive to both the filtration rate and the degree of clogging of the filter. Previous investigations have also shown that sand filter removal efficiencies with aluminum based coagulants are also dose dependent (Croll *et al.* 2007; Goodman 2011).

CONCLUSIONS

Cryptosporidium-sized microsphere removals from a pilot-scale swimming pool via high-rate sand filtration with six coagulants were studied separately. *Cryptosporidium*-sized microsphere removals were 20–63% by filtration with high-rate sand filtration without coagulation. Up to 99% of *Cryptosporidium*-sized microspheres were removed through high-rate sand filtration with coagulants A, B, D, and F at 37 m/h. Continuously feeding a cationic organic coagulant (coagulants A, B, and F) led to coagulant accumulation and a positive surface charge of particles in the system, and consequent decreases in the average filter removals to less than 60% within 48 hours. Coagulant D was an aluminum-based coagulant and did not appear to cause charge reversal, but the removals declined over time as the filter clogged and the pressured drop across the filter increased. Stable microsphere removals of greater than 90% were achieved with coagulant E (polyaluminum chloride) only after decreasing the filtration rate to 30 m/h.

Coagulant C was a chitosan-based product that removed fewer microspheres compared with the other products (less than 75% under the studied conditions). Overall,

aluminum-based coagulants (coagulants D and E) had a performance advantage over the organic polymer based coagulants primarily in terms of their tendency not to accumulate in the water after multiple rounds of coagulation/filtration and cease to be effective at improving filtration efficiency.

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