

Depressed filtration ripening enhances removal of *Cryptosporidium parvum*

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Abstract Pilot-scale experiments were conducted to investigate removal of *Cryptosporidium parvum* by contact granular filtration. The research demonstrated enhanced removal of *Cryptosporidium parvum* in the presence of kaolin particles. This is believed to be due electrostatic adhesion of *Cryptosporidium parvum* oocysts to the kaolin clay particles. The elementary physico-chemical interactions between filter granules and suspension particles will be discussed. This innovative concept was successfully implemented to reduce the ripening sequence of subsequent filtration experimental test runs by the addition of large surface area particles to slurry of kaolin and *Cryptosporidium parvum* in surface water.

Keywords Contact granular filtration; *Cryptosporidium parvum*; ripening; sand filter

Introduction

Drinking water contamination is one of the most important environmental concerns necessitating worldwide attention (US Safe Drinking Water Act; EU Council Directive 75/440/EEC; Japan Water Pollution Control Law 138). One of the greatest health risk management challenges for drinking water supplies is posed by disease-causing microbial contaminants, such as *Cryptosporidium parvum* (*C. parvum*). Penetration of oocysts of *C. parvum* through water treatment systems caused numerous outbreaks of cryptosporidiosis – a disease with no therapeutic care, potentially lethal for persons with depressed immune system and AIDS sufferers (CDC, 1995). Unlike other microorganisms, *C. parvum* oocysts are unusually resistant to traditional disinfectants like chlorine and chloramines (Finch *et al.*, 1993; Owens *et al.*, 1994). Thus, protection of municipal potable water source often relies on granular bed filtration as a robust, simple and inexpensive method to prevent penetration of disinfectant-resistant pathogenic microorganisms (US EPA, 2000).

Intensive research, initiated since the first *C. parvum* outbreaks, found safe level of more than 99% oocysts removal during the optimal operable stage of the filtration cycle (Swertfeger *et al.*, 1999; Lipp and Baldauf, 2000). Maintenance of proper coagulation/flocculation regime was recognized as the single most important step in *C. parvum* removal (Edzwald and Kelley, 1998; Dugan *et al.*, 1999). While keeping correct chemical regime, insufficient removal of pathogenic microorganisms can be attributed to the ripening stage with low performance of the pristine (or newly backwashed) filter. Despite alternative methods such as slow start (Monk, 1987) and flocculant addition during backwash (Francois and Van Haute, 1985), the common “solution” is to divert the ripening filtrate to sewage (also known as “filter to waste”). Alternatively, in places where water is hard to come by, this filtrate is pumped back to the beginning of the process, rendering the process both capital- and energy-intensive. Thus, a shorter ripening sequence would lower operational costs, minimize the presence of *C. parvum* in treated potable water (Rose, 1997), and depress outbreaks that still occur (Fricker, 1999).

Experiments conducted at the US EPA Test and Evaluation (T&E) Facility in Cincinnati, Ohio, indicated enhanced removal of *C. parvum* in the presence of kaolin particles. This led to the assumption that electrostatic adhesion of *C. parvum* oocysts to kaolin particles helps kaolin to act as an additional retaining agent in filtration columns. The proposed concept was successfully implemented to reduce the ripening sequence of filtration runs by addition of large-surface-area particles to the slurry of kaolin and *C. parvum* in the surface water.

Materials and methods

Figure 1 presents a schematic of the pilot-scale filtration system, designed and fabricated specifically for this type of research. The slurry, consisting of kaolin clay particles and dechlorinated city of Cincinnati tap water treated by reverse osmosis (RO), was pumped into a 1,000 L Cross-Linked Polyethylene (XLPE) feed water supply tank. The suspension in the tank was kept completely mixed by using a high-speed mixer. A centrifugal pump was used to lift the suspension into a 100 L head tank located 3.6 m above the filtration column. The water level in the feed tank was maintained at a constant height by an overflow line returning the suspension to the feed tank. To avoid remnant presence of oocysts in the feed tank, *C. parvum* was injected into the mainstream immediately before the static mixer. The major physical characteristics of suspension-forming kaolin clay particles and *C. parvum* oocysts are presented in Table 1.

The influent oocyst concentration of 8×10^6 oocysts/L was up to 7 logs higher than that found in surface water (Karanis *et al.*, 1998). Enumeration of *C. parvum* oocysts was performed using Immuno Fluorescent Assay (IFA) technique, following the procedure described by Kao and Ungar (1994). The high feed concentration was needed in order to ensure detectable effluent oocyst concentration. Even at the feed concentrations used in that work, oocysts only ranged from 0.007–0.06% of the total number of influent particles (greater than 0.5 μm). At these relative concentrations, oocysts were assumed to not significantly change expected accumulation dynamics.

A 17 cm diameter, 2.6 m high acrylic transparent filter column was used for the study. The column consists of 9 sampling and 9 pressure ports, located in pairs on opposite sides of the column. Sampling points were inserted 0.05 m inside the column to avoid wall effects that could lead to misinterpretation of filtrate quality. The pressure transmitters were constantly connected to the pressure ports. The column was packed with 1.6 m of uniform size sand having a geometric mean diameter of 1.05 mm and a uniformity coefficient of 1.55. The media porosity, determined by volumetric measurements, was 0.44. The column was operated at a constant flow rate of 5 and 10 m/h under contact (in line) filtration mode in a conventional downward direction. The filtration velocity was controlled and adjusted by a flowmeter connected to the column outlet. Immediately after each run, the filter was back-washed for 1 min. by air at 200 kN/m² pressure, followed by 10 min. upflow of Cincinnati tap water at 1.1 L/s. Both caused 50% bed expansion. To avoid risk of cross-contamination, the filter was chlorinated and all removable parts of the system (pumps, baths, pipes) were sterilized after each run. *C. parvum* sampling jars were autoclaved.

Table 1 Physical characteristics of suspension-forming entities

Particle	Size (μm)	Density (g/cm ³)	Zeta-potential in Cincinnati tap after RO system (mV)	Zeta-potential in Cincinnati tap after RO system with 20 mg/L alum (mV)
kaolin	0.778 ± 0.315	2.6	-10.33	-3.33
<i>C. parvum</i>	4–6	1.045	-16.0	-1.26

To imitate a filtration plant in the best possible manner, the suspension was destabilized using alum [$\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$] solution, injected into the suspension using a peristaltic pump through Tygon silicone pipes. The optimal dose of alum to be added to slurry of kaolin and *C. parvum* in water was established using a series of flocculation/sedimentation tests (Jar-tests). The following procedure was used in all tests: (i) rapid mixing at 100 rpm for 1 min.; (ii) flocculation at 30 rpm for 20 min.; and (iii) quiescent settling for 30 min. The tests were performed at a room temperature ($\sim 20^\circ\text{C}$). Turbidity samples were obtained using a 20 mL pipette from a point located 0.04 m below the clarified suspension level. *C. parvum* samples were filled into 0.25 L polypropylene jars by careful decantation from the upper part of 1 L test beakers.

Results and discussion

Figure 2 depicts the decrease in residual unsettled fraction of kaolin (measured as turbidity) and *C. parvum* (counted as oocysts), as a function of alum added. At doses higher than 20 mg/L, a sufficient removal efficiency improvement was observed. The optimal removal of the parasite was found at an alum dose of 40 mg/L. Surprisingly, the kaolin particles did not follow the *C. parvum* trend and remained as stable solution. The relatively high optimal alum concentration was explained by the water origin, downstream of the RO system, which

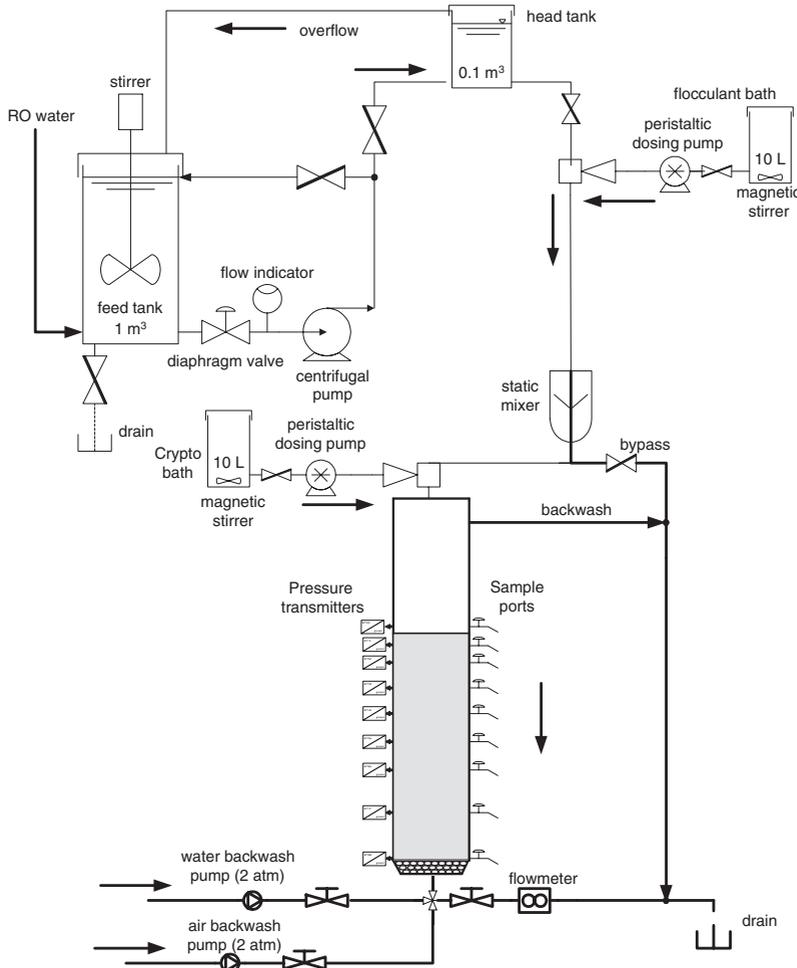


Figure 1 Deep-bed contact filtration scheme

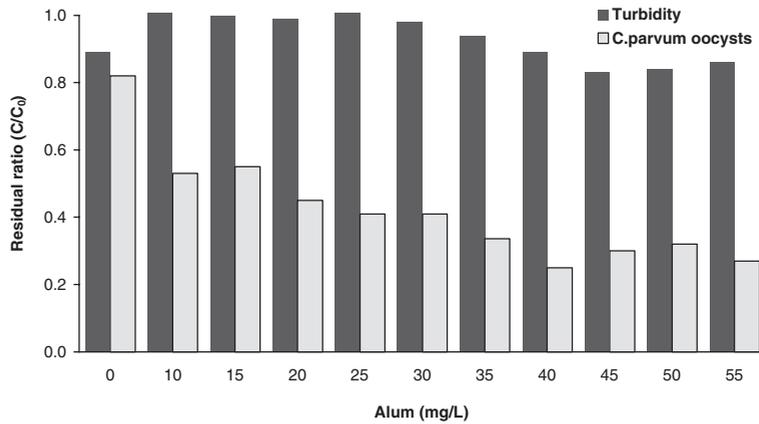


Figure 2 Flocculation efficiency as a function of alum addition in jar-test experiments (suspension consisted of 10 mg/L kaolin + 2×10^7 *C. parvum* oocysts)

not only reduced the number of background particles but also raised the average pH level to 8.1–8.2. A concentration of 20 mg/L alum was deemed as the optimal dose for filtration based on obtained data and previous recommendations (Adin *et al.*, 1979).

Figure 3 depicts the removal of *C. parvum* with and without kaolin particles, as a function of bed volumes from the beginning of the filtration run. Addition of kaolin particles to the initial suspension persistently enhanced removal of *C. parvum* oocysts. The increment was more pronounced for experiments performed with no flocculants at 5 m/h (Figure 3a), and 10 m/h (Figure 3b) resulted in approximately 60–75% oocyst removal. This compared to almost zero removal – 20% maximum, for experiments performed on the “uncovered bed” with no kaolin. However, it was more difficult to filter out *C. parvum* from “clean” water (i.e. water which contained less colloid material). Significant improvement in *C. parvum* removal was achieved with addition of alum (Figure 3c). Experiments performed with the addition of kaolin resulted in a filtrate of a stable quality not influenced by hydrodynamic conditions.

Table 2 presents experimental conditions and filtration coefficients λ_0 and K_r for filtration experiments depicted in Figures 3 and 5. The λ_0 coefficient was calculated using Iwasaki (1937) approach:

$$\lambda_0 = \frac{\ln(C_0 / C)}{L} \quad (1)$$

The ripening rate coefficient K_r was found by fitting theoretical curves to experimental data using best-fit value method. Parameter variation in curves fitting was used to attain the closest possible approximation of experimental data by theoretical curves. The employed curve was obtained from the local mass balance equation in its idealized form (with no axial dispersion) with irreversible attachment kinetics:

$$\frac{\partial}{\partial t} \left(\varepsilon_0 - \frac{C_s}{\rho} \right) C_1 + K_r u C_1 + u \frac{\partial C_1}{\partial z} = 0 \quad (2)$$

Here, C_1 (cell/cm³) = bacterial concentration in the liquid phase; C_s (cell/cm³) = bacterial concentration on the solid surface; t (min.) = time; u (cm/min.) = approach velocity; z (cm) = position in the column measured as the distance from the entrance in the direction of the flow; ε = porosity of filter bed; ε_0 = initial porosity of filter bed; K_r = attachment rate coefficient (cm⁻¹) and ρ = physical deposit layer density (cell/cm³).

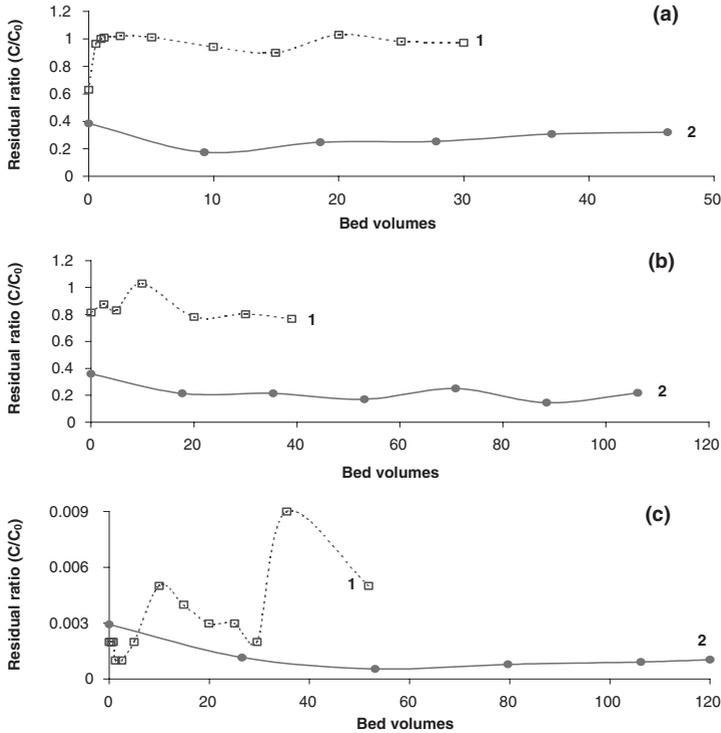


Figure 3 Comparative *C. parvum* oocyst residual ratio in filtration experiments performed without (curves 1) and with (curves 2) 10 mg/L kaolin. Experimental conditions: (a): $u = 5$ m/h, no flocculants; (b): $u = 10$ m/h, no flocculants; (c): $u = 5$ m/h, 20 mg/L alum

Table 2 Experimental conditions and values of filtration coefficient K_a and λ_0 obtained from the study. Filtration parameters: filter depth $L = 40$ cm, media size 1.06 mm; *C. parvum* initial concentration 8×10^6 oocysts/L; temperature varies from 24–26°C

kaolin clay concentration (mg/L)	Filtration velocity u (m/h)	alum concentration (mg/L)	λ_0 (m^{-1})	K_r (m^{-1})
–	5	–	0	0.1
10	5	–	2.39	2.95
–	10	–	0.51	0.5
10	10	–	2.56	4.15
–	5	20	15.54	19.5
10	5	20	16.91	24.9
–	10	20	12.58	24.5
10	10	20	14.58	27

Experiments performed with no kaolin constantly resulted in lower filtration coefficients that reflect lower oocyst attachment. Comparing the filtration efficiency on a more accurate K_r base, up to 30 times more efficient *C. parvum* removal was observed with kaolin addition. Addition of 20 mg/L substantially increased adhesion probability while keeping observed tendency of enhanced removal of *C. parvum* oocysts in kaolin presence. A 30% increase in *C. parvum* removal was observed with kaolin addition.

The observed tendency led to the hypothesis of the dual role that kaolin clay particles play in adsorption of *C. parvum* oocysts. In the absence of flocculant kaolin particles, structured as layers with partial positive charge on their rim (Figure 4), form an initial monolay-

er of deposit on media grains. After the layer is formed, kaolin acts as electrostatic adhesive bonding sand grain and oocyst, both negatively charged in surface water. Opposite to the electrostatic nature of interaction in absence of alum, in its presence the improvement is hydrodynamic. While addition of alum almost neglects the electrostatic effect by neutralizing the surface charge on oocyst side (Table 1), the presence of kaolin helps to create better hydrodynamic conditions by building denser smaller kaolin–alum–*C. parvum* flocs.

The performed analysis suggests that kaolin can act as an additional retaining agent of *C. parvum* in sand filters. Thus, addition of limited amount of kaolin to the filtration slurry can reduce the ripening sequence and improve initial filtrate quality. Figure 5 depicts turbidity (Figure 5a) and oocyst (Figure 5b) residual fractions as a function of time from the beginning of the run. Results of typical experiments, performed at 10 m/h approach velocity with addition of 20 mg/L alum (curves 1), were compared to the results of identical experiments carried out with addition of 2.5 mg/L flocculated kaolin particles during the first 10 minutes of the run. The test filter 2 evidenced a ripening period of 20 minutes, resulting in a final turbidity of 0.8 NTU. Conversely, in the control filter 1 the ripening lasted 90 minutes, and the final filtrate turbidity was 1.3 NTU. Comparing the experiments on oocyst basis, a prolonged ripening period is seen for the experiment performed with addition of kaolin. All other tendencies had no change.

Blank experiments showed that the addition of kaolin that was not pre-flocculated with alum resulted in increased filtrate turbidity. Moreover, too high a concentration of kaolin particles clogged the filter and led to early filter breakthrough. Conversely, too low amounts of kaolin had absolutely no effect on the filtration process.

Conclusions

Performed study suggests the following conclusions.

- Performed experiments indicate better removal of *C. parvum* oocysts in the presence of kaolin. Observed retaining function of the latter was successfully implemented for filtration studies. A short-time increase in initial kaolin concentration helped to reduce ripening period and to increase *C. parvum* removal efficiency.
- Small quantity of accumulated particles facilitates penetration of *C. parvum* oocysts through filter media. Increase in oocyst removal during months of initial low turbidity (usually summer) can be achieved by seasonal shift from conventional to the contact filtration scheme. Exploitation of the former causes sufficient reduction of turbidity (mainly contributed by clay particles) before the filtration, and filtration of low-turbid water.

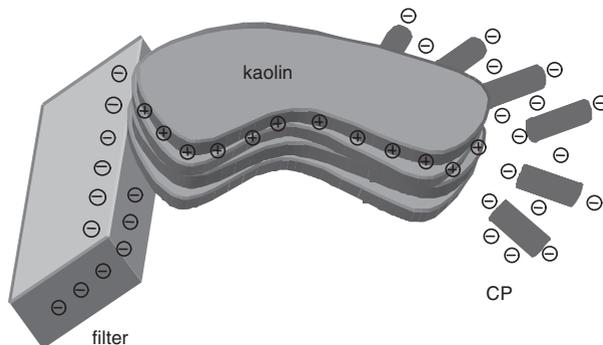


Figure 4 Cartoon showing how kaolin particles act as an electrostatic adhesive in the adsorption of *C. parvum* oocysts on sand granules

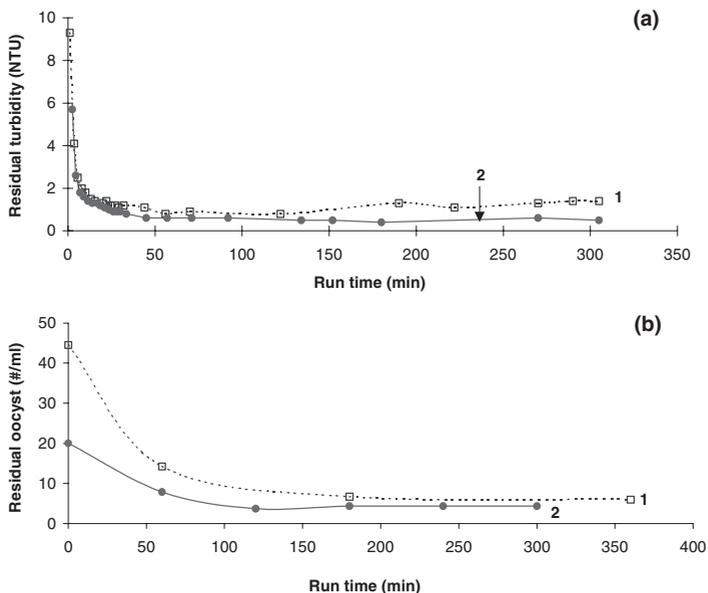


Figure 5 Comparative filtration experiments without (curves 1) and with (curves 2) aggregating particles. Experimental conditions: $u = 10$ m/h; $C_0 = 11.3$ NTU; flocculants = alum 20 mg/L; kaolin added – 2.5 mg/L during first 10 min. of the run

- The results from the pilot scale experiments, provided in current work, clearly show the advantages of the proposed method. However, the disadvantages including possibility of control and practical consideration of usage should be examined on full-scale filters before any final conclusions can be made. In case that the method shows similar results in different conditions, proposed minimization of ripening period may have crucial meaning in countries with limited water reserves.

Acknowledgements

This study was supported by the US Environmental Protection Agency, National Risk Management Research Laboratory, Water Supply and Water Resources Division through contract No. 68-C-99-211, Work Assignment No. 1-03, with IT Corporation. The authors thank James H. Owens of US EPA for providing purified *C. parvum* oocysts and Lee Heckman from IT Corporation for training in *C. parvum* sampling and analysis. Special thanks to James A. Goodrich and Herb Brass of US EPA, Cincinnati, OH and Pratim Biswas from Washington University in St. Louis, MO, for their cooperation in this study. The views expressed in this paper are those of the authors and do not necessarily reflect the views of the US EPA.

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