Effects of filtration temperature, humic acid level and alum dose on cryptosporidium sized particle breakthrough

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Abstract Recent *Cryptosporidium* outbreaks have highlighted concerns about filter efficiency and especially particle breakthrough. Understanding the causes of breakthrough is essential, as the parasite cannot be destroyed by conventional disinfection with chlorine. Particle breakthrough depends on many factors. This research aims to investigate the influence of temperature, humic acid (HA) level and chemical dosing on particle breakthrough in filtration. A series of temperatures were set at 5 °C, 15 °C and 25 °C; humic acid level was 5 mg L^{-1}. Each was combined with a series of Al doses. A laser particle counter was used to assess the particle breakthrough online. Turbidity, ζ potential, and UV_{254} absorption were measured before and after filtration. The results showed that particle breakthrough was influenced significantly by temperature, humic acid and dosing. Particle breakthrough occurred earlier at lower temperature, while at higher temperature it was reduced at the same coagulant dose. With coagulants, even at low dose, particle breakthrough was significantly reduced. With HA 5 mg L^{-1}, particle breakthrough was earlier and the amount was much larger than without HA even at high temperature. There was an optimal dose in filtration and it was well correlated with ζ potential.

Keywords Coagulation; *Cryptosporidium*; filtration; humic acid; particle; temperature

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Introduction

Cryptosporidium is a protozoan pathogen that is responsible for the gastrointestinal disease cryptosporidiosis. An outbreak may occur as a result of poor water treatment (Smith, 1995; Clancy, 2000). Research has shown that *Cryptosporidium* is physically classified as a biological particle, with an almost spherical shape and a diameter of 4.5–5.5 μm. It is only slightly denser than water with a density of 1.025–1.070 g cm^{-3} (Medema *et al.*, 1998). Surface charges measured by the ζ potential of the oocysts have been found to be neutral to slightly negative in most natural waters. Their electrophoretic mobility remained unchanged up to 121 days after collection (Drozd and Schwartzbrod, 1996; Brush *et al.*, 1998). However, this particular pathogen was resistant to standard chlorination (Korich *et al.*, 1990), so it must be effectively removed by physical separation methods, such as filtration.

It has been demonstrated that chemical inactivation can change oocyst zeta potential (Ongerth and Pecoraro, 1996). The zeta potential is indicative of the degree of particle destabilization and plays an important role in coagulation and subsequent filtration. A change in the original surface charge of colloidal particles like oocysts might affect their removal during granular media filtration (Amirtharajah and Mills, 1982; Lytle and Fox, 1994). Differences in oocyst zeta potential prior to coagulation should not impact overall coagulation chemistry because oocyst surface area is essentially insignificant relative to that of other particles present in water, even during investigations utilizing

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artificially high oocyst concentrations (Emelko, 2003). It has been demonstrated that formalin-inactivated *C. parvum* oocysts are reliable surrogates for viable oocysts during filtration studies (Emelko, 2003).

From the literature (Amirtharajah and Mills, 1982; Korich *et al.*, 1990; Lytle and Fox, 1994; Drozd and Schwartzbrod, 1996; Ongerth and Pecoraro, 1996; Brush *et al.*, 1998; Medema *et al.*, 1998; Emelko, 2003), it would be reasonable to think that in water treatment processes *Cryptosporidium* could be treated just as a typical particle and its breakthrough risk could be evaluated by testing the particle breakthrough after filtration, using surrogate particles with size similar to *Cryptosporidium*. This avoids the problem of using specific assay procedures for *Cryptosporidium*, which are difficult and time-consuming. So experimental work should focus on minimizing particle breakthrough in the light of the *Cryptosporidium* risk.

Traditional particle breakthrough has been monitored using turbidity meters; however, these readings could be misleading, as they do not give detailed information on particle size. Therefore, the *Cryptosporidium* breakthrough could not be assessed and predicted. Particle counters are better tools for such research as confirmed by Hall and Croll (1997), who concluded that they are far more sensitive for measuring effluent quality. Temperature and chemical dosing effects have been investigated in earlier particle breakthrough studies (Amirtharajah and Mills, 1982; Nieminski and Ongerth, 1995; Naranjo *et al.*, 1997; Emelko, 2003), but there are still many aspects that need further investigation.

Surface and ground waters may contain significant quantities of natural organic matter that are detrimental to water quality, i.e. color and odor of water. Also, natural organic matter (NOM) is known to form trihalomethanes (THM) upon chlorination. In addition, the presence of natural organic matter in a water distribution system favors bacterial regeneration in the network, which may cause sanitary problems. Consequently, as far as possible, NOM should be removed during water treatment. Some researchers reported that when NOM was present, oocyst removal by conventional filtration decreased from 51 ± 6% to 14 ± 1% (Dai and Hozalski, 2002). The effect of NOM should therefore be given more attention in the removal of oocyst-sized particles.

This research aimed to establish the relationship between particle breakthrough, presence of HA, temperature and chemical dose, and to analyze the mechanism of particle breakthrough, as affected by these parameters.

Methods

The laboratory filter was 1 m in height with an internal diameter of 100 mm, and contained a 0.5 m bed of 0.6–0.7 mm Leighton Buzzard sand. The filter was operated at $5\,m\,h^{-1}$. The influent suspension contained $10\,mg\,L^{-1}$ of kaolin. Some raw waters contained $5\,mg\,L^{-1}$ HA. Alum was dosed directly into the top of the column via a baffle mechanism. The runs were 3 hours in duration. The temperatures were controlled at 5 °C, 15 °C and 25 °C by the chiller and heater recycled in the raw water tank. Alum dosing ($Al\,mg\,L^{-1}$) was controlled in proportion to flow rate by changing the speed of the peristaltic pump. Alum dosages without HA were $0.0\,mg\,L^{-1}$, $0.3\,mg\,L^{-1}$, $0.6\,mg\,L^{-1}$ and $0.8\,mg\,L^{-1}$; with HA ($5\,mg\,L^{-1}$) alum dosages were $0.0\,mg\,L^{-1}$, $0.8\,mg\,L^{-1}$, and $1.6\,mg\,L^{-1}$. Particle counts were taken by using a Met One particle counter. It was able to record counts in six separate size channels from 2–15 μm. Turbidity, ζ potential and UV absorption values have been measured. After each run, the bed was backwashed using a set procedure of 5 minutes water and air scour, followed by 20 minutes of water wash at 50% bed expansion.

Although several size ranges have been recorded during the study, for the purpose of this paper, only the 2–7 μm range is considered here, as this represents the *Cryptosporidium* size range. The ripening time of initial filtration was about 40 minutes.

Results and discussion

Effect of alum dosing and ζ potential

Figures 1–3 illustrate the effects of alum dosing in the particle breakthrough during filtration at 5 °C, 15 °C and 25 °C with HA (5 mg L^{-1}) and without HA.

From Figure 1a, Figure 2a and Figure 3a, it can be seen particle breakthrough occurred very quickly and in large amount without coagulant: during 40–170 minutes the residual particles were 25,000–28,000 mL^{-1} (5 °C), 19,000–23,000 mL^{-1} (15 °C) and 20,000–24,000 mL^{-1} (25 °C). With coagulants, even at low dose (0.3 mg L^{-1}), particle breakthrough was significantly reduced: during 40–170 minutes the residual particles were 200–900 mL^{-1} (5 °C), 150–900 mL^{-1} (15 °C) and 200–550 mL^{-1} (25 °C).

There was an optimal alum dose range (0.6–0.8 mg L^{-1}) for filtration, which could significantly reduce particle breakthrough. With the optimal dose 0.6–0.8 mg L^{-1} during 40–170 minutes the residual particles were 90–400 mL^{-1} (5 °C), 150–500 mL^{-1} (15 °C) and 150–200 mL^{-1} (25 °C), respectively (Figure 1b, Figure 2b and Figure 3b). The optimal dose gave a ζ potential equal to about zero as shown in Figure 4. The results were not as good for doses away from the optimum, which caused the ζ potential to move away from zero. The reason is probably that, with the ζ potential of particles

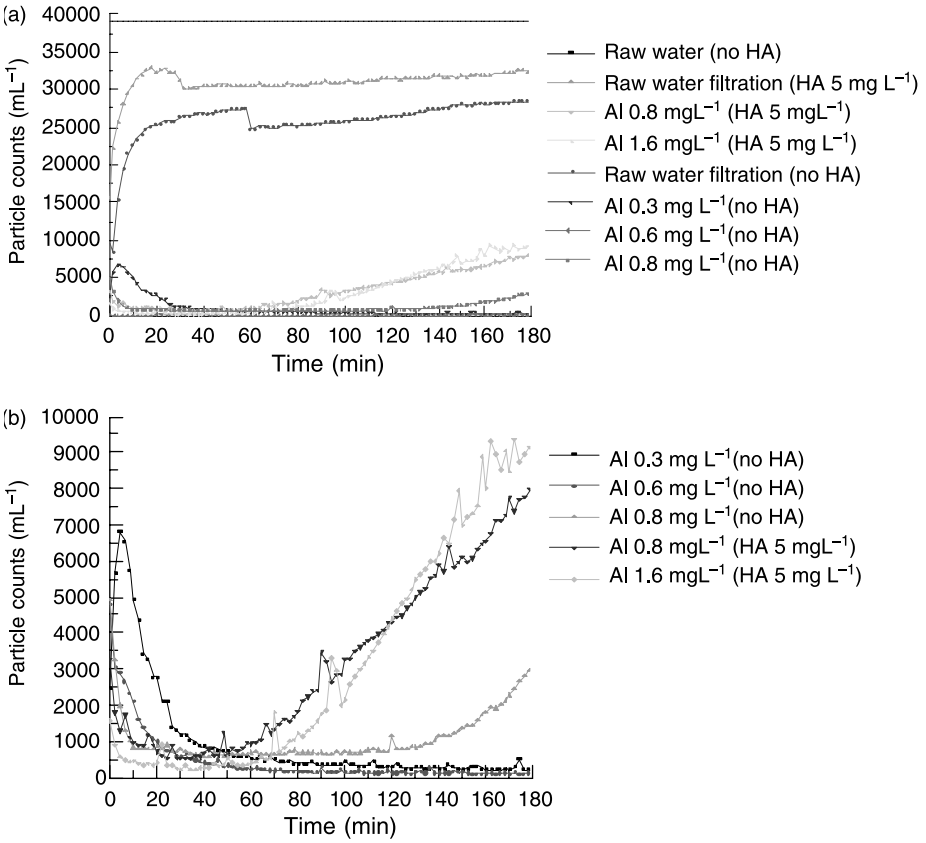

Figure 1 2–7 μm particle breakthrough with different doses at temperature 5 °C

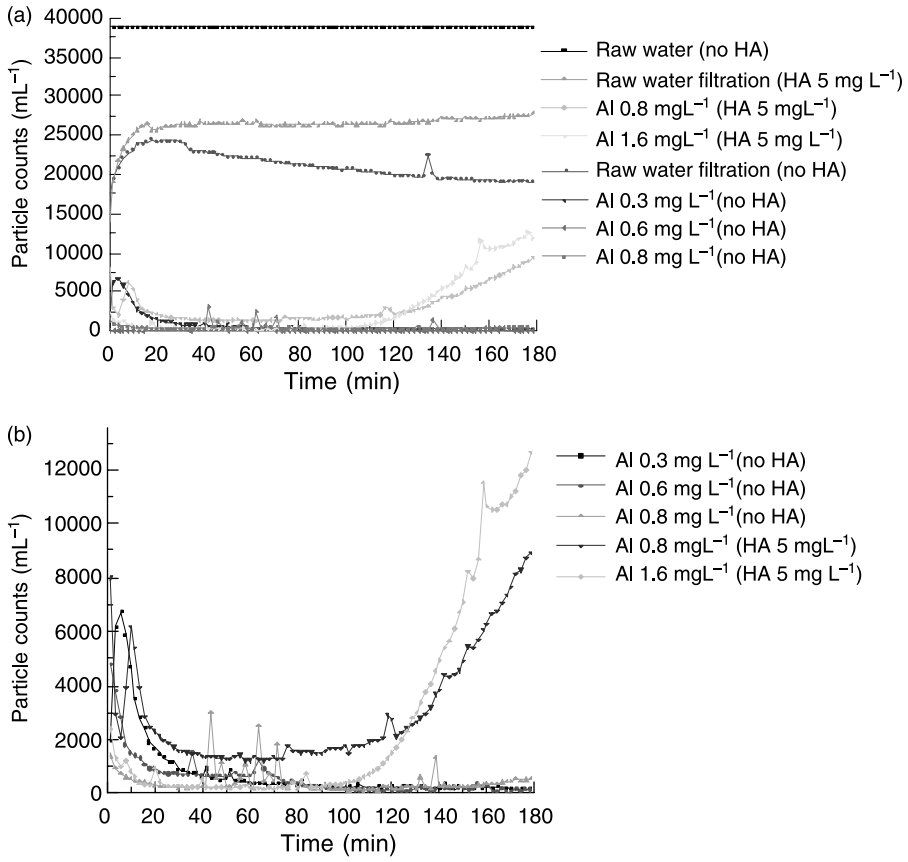

Figure 2 2–7 μm particle breakthrough with different doses at temperature 15 °C

around zero, enhanced coagulation among destabilized particles and adhesion to the filter media surface occurred.

Several studies (O'Melia, 1985; Amirtharajah and Tambo, 1991; Letterman *et al.*, 1999; Gregory and Carlson, 2003) have also demonstrated the importance of optimizing particle ζ potential (or electrophoretic mobility). The ζ potential range of properly destabilized particles was usually reported to be in the range of -4 to $+3$ mV. Some studies have used ζ potential for coagulation process control. The ζ potential is an indicator of the effective surface charge and therefore the degree of destabilization of a particle, which is perhaps one of the most important variables affecting attachment to filter media. It has also been shown that coagulation failure decreased *C. parvum* removal by >3 log relative to stable operation (Emelko, 2003). It can be assumed (O'Melia, 1985; Gregory and Carlson, 2003) that the primary collectors in the filters were actually the previously retained particles, not the filter medium itself.

It also can be seen from Figures 1–3 that the particle breakthrough with HA was much higher than those without HA especially towards the end of the filter run. For example, at 15 °C when the alum dosing was 0.8 mg L^{-1} during 140–170 minutes, the residual particles increased from 300–500 mL^{-1} without humic, but with humic the residual particles increased from 5,000–9,000 mL^{-1} (Figure 2b). Also the run time was much shorter than that without HA.

Without alum dosing the particle breakthrough with HA was much higher (30,500–32,000 mL^{-1} during 40–170 minutes at 5 °C; Figure 1a) than that without HA

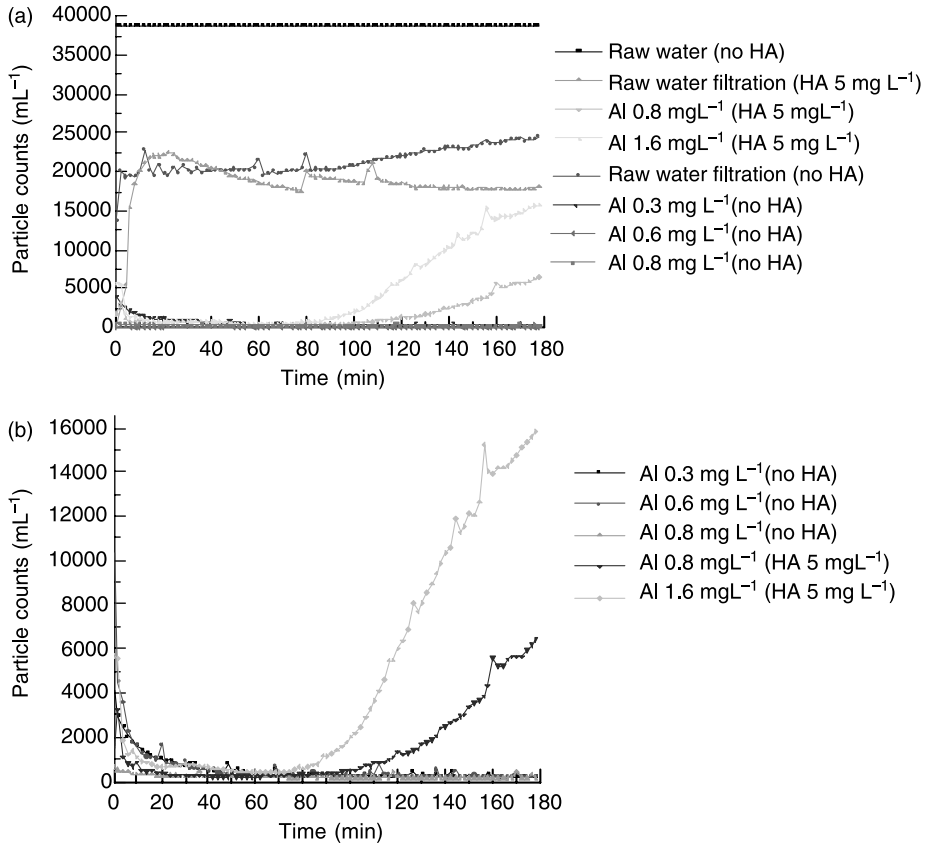

Figure 3 2–7 μm particle breakthrough with different doses at temperature 25 °C

(25,000–28,000 mL^{-1} during 40–170 minutes at 5 °C; Figure 1a), and at 15 °C (Figure 2a) the result was similar; while the result at 25 °C was opposite (during 40–170 minutes, 18,000–20,500 mL^{-1} with HA, but 20,000–24,000 mL^{-1} without HA; Figure 3a). The reason for this behavior is not clear.

With HA (Al: 1.6 mg L^{-1}) the particle breakthrough was nearly the same as that without HA (Al: 0.3 mg L^{-1}, 0.6 mg L^{-1} and 0.8 mg L^{-1}) at first 1 hour (5 °C) and first 100 minutes

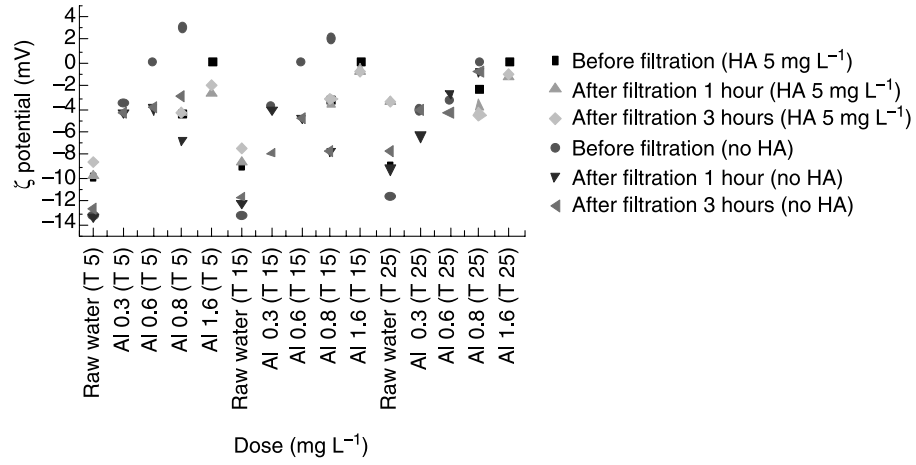

Figure 4 ζ potential with different temperatures and doses

(15 °C, 25 °C); at these stages the results of Al dose at 1.6 mg L^{-1} were better than those of Al at 0.8 mg L^{-1} except at 25 °C with HA. But after the first 1 hour (5 °C) and 100 min (15 °C, 25 °C) with HA the particle breakthrough went up very quickly and was many times higher than the particle breakthrough without HA at the end of the filter run; at these stages the results of Al dose at 1.6 mg L^{-1} went up much faster than those for Al at 0.8 mg L^{-1} with HA. For example, from Figure 1b, at 5 °C during 40–170 minutes with HA the residual particles went from 250–9,000 mL^{-1} (Al: 1.6 mg L^{-1}) and from 700–8,000 mL^{-1} (Al: 0.8 mg L^{-1}).

Effect of temperature

Figures 5a–c show the results of particle breakthrough in filtration at different temperatures (5 °C, 15 °C and 25 °C). The data for Figure 5 are the same as that in the previous section, but with a different scale.

It can be seen that even without alum high temperature could sharply reduce the particle breakthrough. The low temperature (5 °C) gave an increase in particle breakthrough and the breakthrough trend went up at the end of filtration (Figure 5a). Without HA when

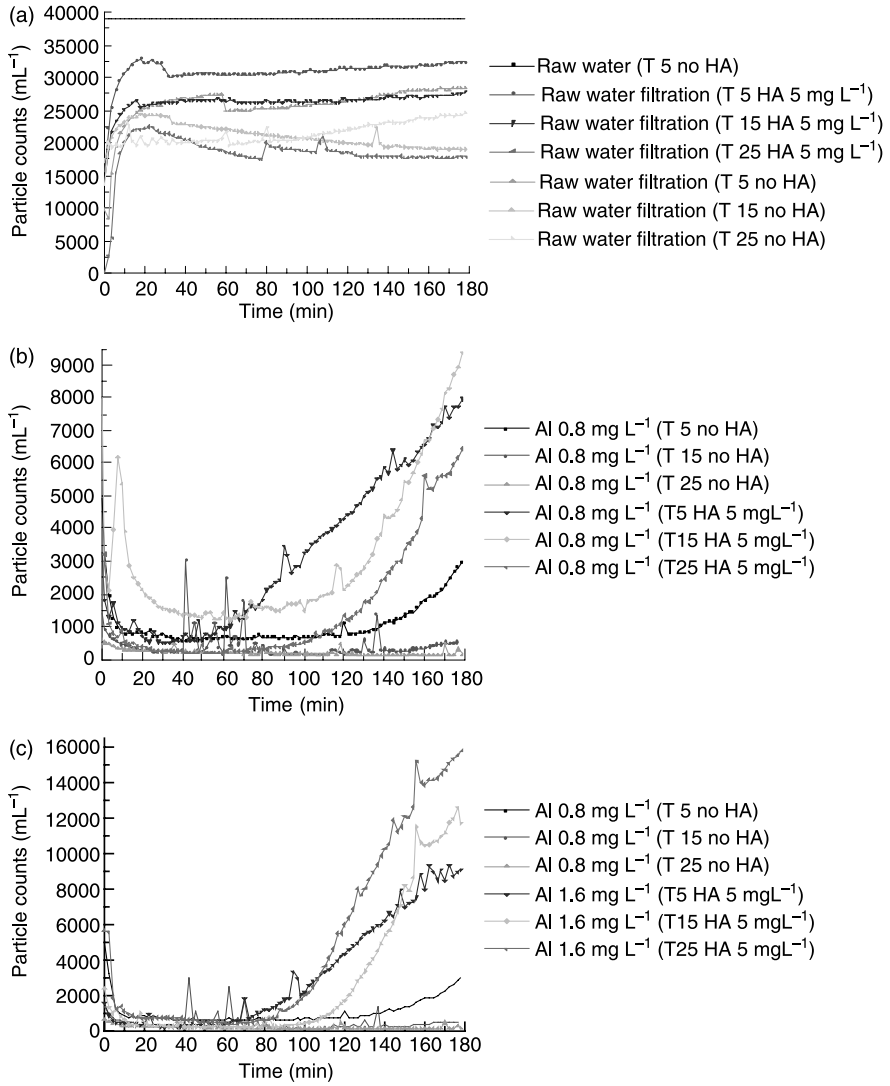

Figure 5 2–7 μm particle breakthrough in filtration with different temperatures.

alum dose was at 0.8 mg L^{-1} during 40–170 minutes, the residual particles went up from 150–200 mL^{-1} at 25 °C, but from 600–2,300 mL^{-1} at 5 °C (Figure 5b).

Lower temperatures brought rapid particle breakthrough (5 °C) while higher temperature (15 °C and 25 °C) reduced the particle breakthrough for the same coagulant dose without HA; the low temperature also made the breakthrough time shorter than the higher temperature (15 °C, 25 °C) at Al dose 0.8 mg L^{-1} without HA (Figure 5b).

However, higher temperature (25 °C) also made the particle breakthrough slightly higher than at moderate temperature. From Figure 5c, during 40–170 minutes when the alum dosing was 1.6 mg L^{-1}, with HA the residual particles were 200–11,000 mL^{-1}(15 °C), and 400–15,000 mL^{-1} (25 °C). This might be due to temperature influencing attachment, detachment and reattachment on the surface of media, giving greater penetration of the filter bed.

The results of turbidity

Turbidity also has been tested (Figure 6). The relationship of low concentration of particles was not directly linked to turbidity and almost could not be detected by turbidity meter in the sample test. In Figure 6, at 25 °C after 3 hours filtration without HA, the residual turbidity was 0.01NTU (Al 0.3 mg L^{-1}), 0.00NTU (Al 0.6 mg L^{-1}) and 0.01NTU (Al 0.8 mg L^{-1}) separately. That means the risk of *Cryptosporidium* outbreaks could not be assessed and predicted using turbidity meter. The results verified the reliability of the particle counter and it precedes the turbidity meter again.

The results of UV_{254} adsorption

Figure 7 shows the total UV_{254} adsorption (Figure 7a) and dissolved (Figure 7b) UV_{254} adsorption values. From the total UV adsorption diagram (Figure 7a) it can be seen that the UV adsorption values could be better removed after 1 hour filtration than those after 3 hours filtration. From Figure 7a, at 15 °C when the chemical dosing was 1.6 mg L^{-1}, with HA the UV_{254} adsorptions were 0.063 cm^{-1} (filtrating 1 hour) and 0.100 cm^{-1} (filtrating 3 hours). There was little difference of UV adsorption value removal during Al dose at 0.8 mg L^{-1} and 1.6 mg L^{-1}. But it was much better with dosing than those without dosing.

At 5 °C, with HA, after 1 hour filtration, the UV_{254} adsorption was 0.212 cm^{-1} (Al 0.0 mg L^{-1}) and 0.079 cm^{-1} (Al 0.8 mg L^{-1}) respectively (Figure 7a). A higher temperature could help to remove more UV adsorption value. When Al dose was

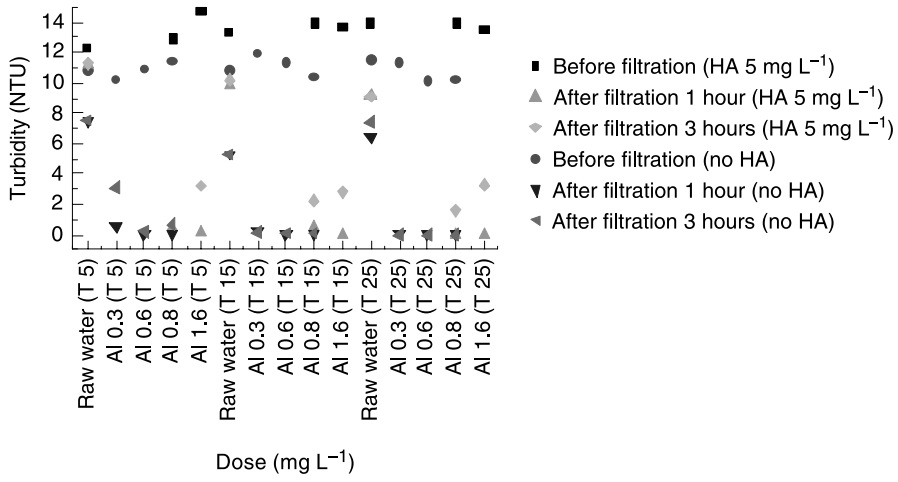

Figure 6 Turbidity with different temperatures and doses

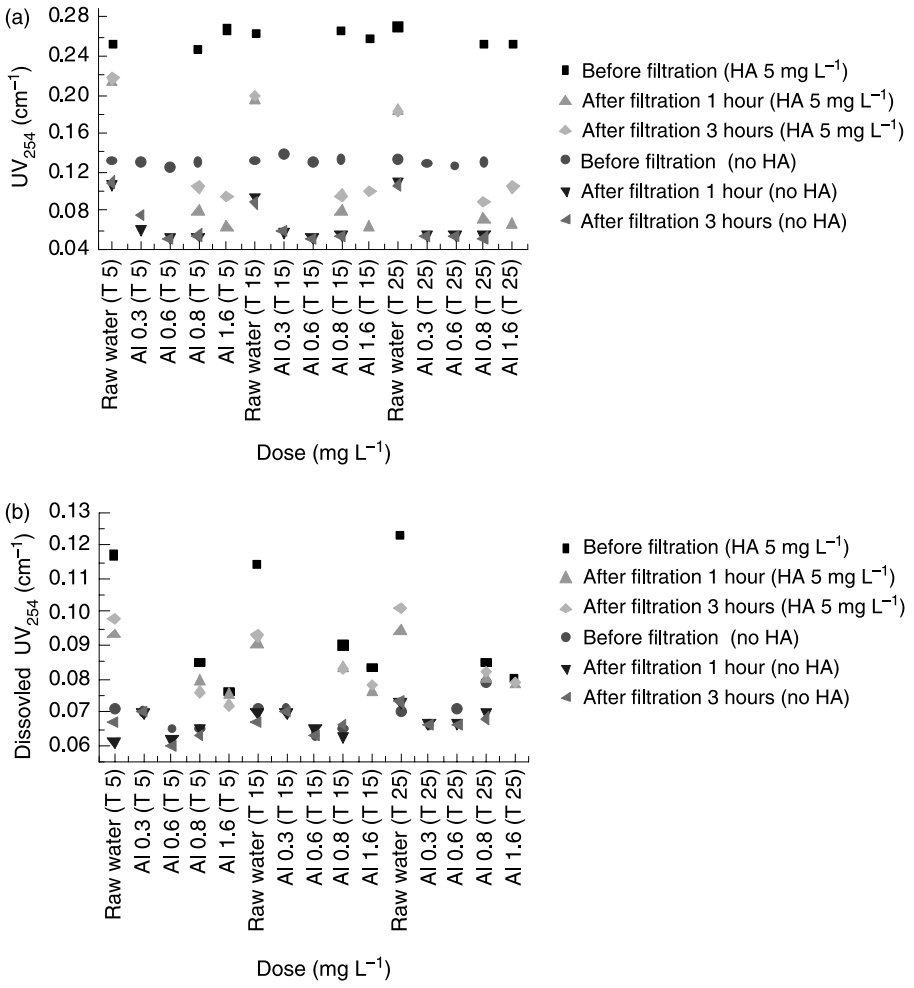

Figure 7 UV_{254} values with different temperatures and doses

0.8 mg L^{-1}, with HA, after 3 hours filtration, the UV_{254} adsorption was 0.105 cm^{-1} (5 °C), 0.096 cm^{-1} (15 °C) and 0.089 cm^{-1} (25 °C) respectively (Figure 7a).

The dissolved UV adsorption values had the same trend as the total UV adsorption values (Figure 7b). It should be pointed out that after dosing even before filtration the dissolved UV adsorption values could decrease and have little difference to the values after filtration. At 15 °C when the chemical dosing was 0.8 mg L^{-1}, with HA the dissolved UV of raw water was 0.114 cm^{-1}. The value before filtration was 0.090 cm^{-1}, after 1 hour was 0.083 cm^{-1} and after 3 hours was 0.083 cm^{-1}.

Conclusions

Temperature, humic acid and alum dose significantly influence particle breakthrough, and attention should be paid to these parameters in practice, especially at low temperature, higher humic acid level and low coagulant dose.

Lower temperature can bring rapid particle breakthrough and shorten the run time. Higher temperature can reduce the particle breakthrough trend at the same coagulant dose and prolong the run time.

There is an optimal dose in filtration, which can significantly reduce particle breakthrough; the dose is well correlated with ζ potential. A ζ potential of particles of zero enhances coagulation among particles and adhesion to the filter media surface.

There is no direct relationship between the particle amount and turbidity at low particle concentration. The results verify the reliability of the particle counter and it will precede the turbidity meter in practice.

With humic acid level at $5\,mg\,L^{-1}$, particle breakthrough is faster and the amount is much larger than those without humic acid even at high temperature. The required alum dose is higher and the filtration performance is poorer than that without humic acid.

In the water treatment processes *Cryptosporidium* can be treated just as a colloid and its breakthrough risk can be evaluated by testing the particle breakthrough after filtration. Particle size is similar to *Cryptosporidium* (or a little bigger) which can represent *Cryptosporidium* simply if the condition could not be permitted to detect the *Cryptosporidium* with equipment.

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