OZONE-ENHANCED PARTICLE REMOVAL IN DRINKING WATER TREATMENT

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ABSTRACT

High residual particle counts in filtered water have implications for potential breakthrough of protozoan particles such as *Cryptosporidium* and *Giardia*. A pilot plant study utilising dual media filters was conducted to investigate a number of techniques for enhancing particle removal from settled water through filtration. These included use of filter aid polymer, increased coagulant dose and ozonation. While applying a filter aid polymer and/or increasing the coagulant dose was found to result in significant reductions in the residual particle counts, the most dramatic effect was achieved when the settled water was dosed with a combination of ozone and filter aid polymer.

INTRODUCTION

While the *Cryptosporidium* and *Giardia* events of 1998 in Sydney (Stein, 2000) and 1993 in Milwaukee, Wisconsin (MacKenzie et al., 1994) seem to be in the distant past, the need for reducing residual particle counts in filtered water has not been forgotten. In fact, the latest issue of the Australian Drinking Water Guidelines (ADWG, 2011) has just introduced new guideline values for filtered water turbidity (a surrogate measure of particle counts).

ADWG (2011) states that "where filtration alone is used as the water treatment process to address identified risks from *Cryptosporidium* and *Giardia*, it is essential that filtration is optimised and consequently the target for the turbidity of water leaving individual filters should be less than 0.2 NTU, and should not exceed 0.5 NTU at any time". Note that while these turbidity targets are significantly tighter than the previous target of 1 NTU for effective disinfection with chlorine, they are still not as tight as the USEPA limits for claiming of an additional 0.5-log *Cryptosporidium* treatment credit (USEPA, 2012).

Apart from proper considerations for filter design, there are a number of operational parameters that may be optimised for improving filtered water turbidity and particle counts in conventional water treatment plants. Well-known factors include coagulation chemistry and use of filter aid polymer. While it has been reported in the literature that oxidants (e.g., chlorine, chlorine dioxide, potassium permanganate, ozone or ozone/H_2O_2) can enhance filter performance (Becker et al., 2004; Jasim et al., 2008; Rahman et al., 2010), they are not widely used *per se* and often applied as a result of other treatment requirements such as for soluble metals removal.

This paper presents particle removal results from a recent pilot plant investigation treating settled water using dual media filters. The objective is to compare the relative effect of employing filter aid polymer, increasing coagulant dose and ozonating settled water on improving particle counts.

METHODOLOGY

The study was conducted using a pilot plant housed in a 20' container. The pilot plant was equipped with an ozone generation and contacting facility. A contact time of 14.5 minutes was used in this study with a target residual ozone concentration on the contactor outlet of ≤0.1 mg/L.

A schematic of the pilot plant process is shown in Figure 1. Three identical filter columns were used with an internal diameter of 100 mm and constructed from clear PVC. Each filter column assembly consisted of two 1.5-metre flanged sections, filter nozzle plate, nozzle and a plenum section. Equally spaced tappings along the length of the filters enabled measurement of headloss through the filter bed. Online monitoring of the filtered water turbidity was also provided.

The filter media configuration in each filter was identical (except media size) and consisted of a granular activated carbon (GAC) bed on top of a layer of filter sand (as the polishing layer). Three effective sizes of GAC were used as shown in Table 1. The GACs were sourced from Activated Carbon Technologies (VIC, Australia) and were coal-based, steam activated (Acticarb BAC GA1000N). Note that GAC media was used in this study because the ultimate aim of the project was to study ozone and biological activated carbon (BAC) filtration. For the results presented in this paper, the GAC media was relatively fresh (only up to six weeks old), except for the 'dirty' feed water trials for which the media had already been used intermittently for approximately 14 weeks. A constant filtration rate of 10 m/h was used in each filter.
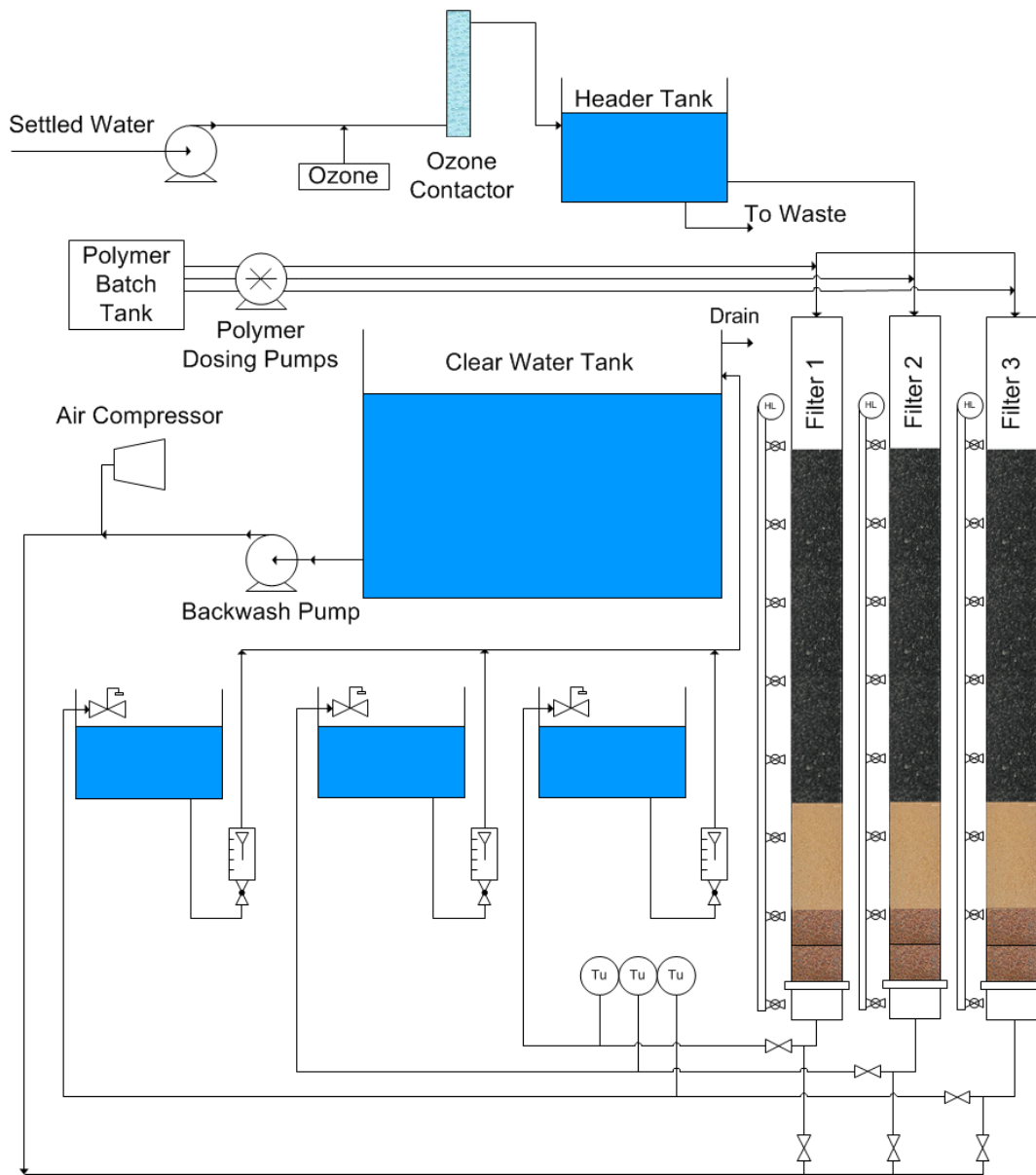
Figure 1: Pilot Plant Process Schematic

Table 1: Filter Media Configuration

Material	Criteria	Units	Filter 1	Filter 2	Filter 3
GAC 1,600 mm depth	Effective Size	mm	0.9 – 1.1	1.1 – 1.3	1.3 – 1.5
	Uniformity Coefficient	-	< 1.4	< 1.4	< 1.4
Polishing Sand 300 mm depth	Effective Size	mm	0.55 – 0.65		
	Uniformity Coefficient	-	< 1.3		
	L/d	-	2,100	1,833	1,643
Coarse Sand 100 mm depth	Effective Size	mm	0.8 – 1.8		
	Uniformity Coefficient	-	< 1.5		
Garnet Layer #1 100 mm depth	Nominal Size	mm	1.2 – 2.4		
Garnet Layer #2 100 mm depth	Nominal Size	mm	2.4 – 4.8		

Feed water for the pilot plant was drawn from the settled water of a conventional full-scale water treatment plant (WTP) utilising alum as the coagulant and a non-ionic polymer (FA920PWG, SNF Australia) as coagulant aid. Typical alum dose was 47 mg/L (as 46% solution). The feed water quality under typical conditions was of consistently good quality. Elevated turbidity (or 'dirty') settled water was also trialled by drawing water from a lower take off point from the WTP clarifier. This was to see how the pilot plant process performed in un-optimised conditions or with a process upset. Feed water quality during the 'typical' and 'dirty' water trials is outlined in Table 2.

Table 2: Quality of Feed Water for the Pilot Plant

Parameter	Units	'Typical' Feed Water	'Dirty' Feed Water
Turbidity	NTU	0.6 – 0.8	1.7 – 2.3
Colour (Apparent)	HU	5 – 7	11 – 18
Particle Counts	>2µm/mL	1,700 – 2,350	7,380 – 11,794

Grab samples from the pilot plant filters were analysed using bench-top instruments including particle counter.

RESULTS AND DISCUSSION

A number of methods for reducing filtrate particle counts were compared and their relative contributions examined. Target water turbidity from the individual filters during the trial was <0.15 NTU 95% of the time and always less than 0.3 NTU. No particle count target was set. When the WTP was operated at the typical alum dose of 47 mg/L (as 46% solution), the pilot plant filters were consistently achieving the turbidity requirement without dosing the filter aid polymer or ozone. However, the filtered water particle counts remained consistently high (~400 particles/mL for >2µm particles).

All trial runs were conducted using freshly backwashed filters, except for the alum dose trial.

Effect of Filter Aid Polymer

For this trial a non-ionic polymer (FA920PWG, SNF Australia) was dosed directly into the feed water of each individual filter. A small static mixer was installed directly after the polymer dosing point to increase mixing. Ozone was turned off in the pilot plant and the full-scale plant was operated using a typical alum dose of 47 mg/L (as 46% solution). The effect of filter aid polymer dosing on residual particle counts was examined using both 'typical' and 'dirty' settled water. As expected, a small dose of filter aid polymer resulted in significant improvements in the

residual particle counts in the filtered water, both for 'typical' and 'dirty' settled water. Note that only data from Filter 2 are shown here since near identical results were observed from all three filters.

The results from the 'typical' feed water are shown in Figure 2. It shows that a small filter aid polymer dose resulted in significant improvements in the residual particle counts in the filtered water. The particle counts were reduced from approximately 330 to around 240 particles/mL for >2µm particles, a reduction of approximately 30%. It is interesting to note that increasing the polymer dose to 0.05 mg/L did not provide further noticeable improvement in particle removal performance.
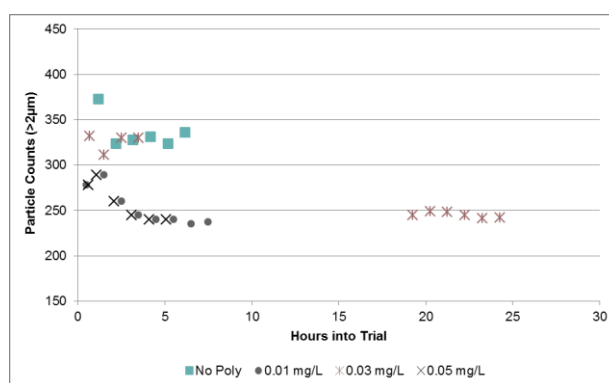

Figure 2: Effect of Filter Aid Polymer Dose on Filtered Water Particle Counts (per mL) ('Typical' Feed Water: 0.6-0.8 NTU)

The results tested on 'dirty' feed water are shown in Figure 3. It was identified that in this case a higher polymer dose of >0.03 and up to 0.05 mg/L was required to achieve meaningful reductions in particle counts. The particle reduction achieved was from approximately 330 to around 200 particles/mL for >2µm particles, a reduction of approximately 40%.
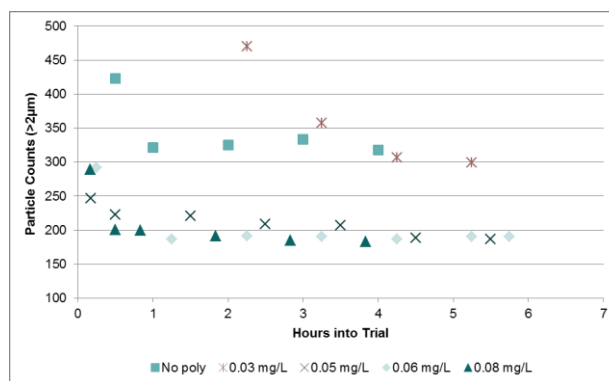

Figure 3: Effect of Filter Aid Polymer Dose on Filtered Water Particle Counts (per mL) ('Dirty' Feed Water: 1.7-2.3 NTU)

The use of filter aid polymer was also found to effectively shorten the filter ripening time on the 'typical' feed water from approximately 45 minutes

(without filter aid polymer) to approximately 20 minutes (with filter aid polymer) at polymer doses of 0.01 mg/L or higher. For the 'dirty' feed water the ripening time was reduced from approximately 45 minutes to 20 - 30 minutes at polymer doses of >0.03 mg/L.

Effect of Higher Coagulant Dose

The coagulation and flocculation process plays a significant role in the filter performance due to the characteristics of the flocs formed. For this study the coagulation process in the WTP was optimised to provide acceptable water quality whilst reducing operating costs through lower chemical usages and hence sludge volumes. Despite optimising the pilot plant, it was considered that perhaps the WTP's coagulant dose was limiting the performance of the pilot plant filters.

A coagulant dose change was effected on the full-scale plant, taking into account delays in settled water quality changes due to the long residence time in the WTP clarifiers. The test was undertaken by starting with a typical WTP alum dose of 47 mg/L (as 46% solution), increasing it to 65 mg/L (selected based on plant experience) and then reducing it back to 47 mg/L. No ozonation was used for this trial but varying filter aid polymer doses were applied on the pilot plant filters.

The results are presented in Figure 4. Note that Filters 1 and 2 had a filter aid polymer dose of 0.01 mg/L and Filter 3 had no filter aid polymer dose. In addition, Filter 1 had already been running at the normal plant alum dose of 47 mg/L for a number of hours prior to increasing the alum dose on the full-scale plant, whereas Filter 2 and Filter 3 were left offline after the previous backwash and started when the high alum dose influence was expected to reach the pilot plant. Due to the residence time in the WTP clarifiers, a delay of approximately two hours was expected for the onset of the alum dose effect.

Figure 4 shows that increasing the alum dose in the coagulation process resulted in a significant improvement in the filtered water particle counts, as well as turbidity and filter ripening performance. The reductions in >2µm particle counts were by over 100 particles/mL in the absence of the filter aid polymer (Filter 3) or 150 particles/mL in the presence of the filter aid polymer (Filter 1 and Filter 2). Overall, improved filter performance from the use of a higher alum dose appeared to be similar to that from the combination of normal alum dose and use of filter aid polymer.

Figure 4 also shows that after the 'Expected High Alum Influence End' was reached, the filtrate particle counts did not shot up to the levels that would have been expected based on the data presented in Figure 2. It is not clear why there was a much longer than expected high alum dose influence period. But it is possible that coagulation condition changes on the full-scale plant may take much longer than the estimated clarifiers' hydraulic residence time to fully impact on the settled water quality.
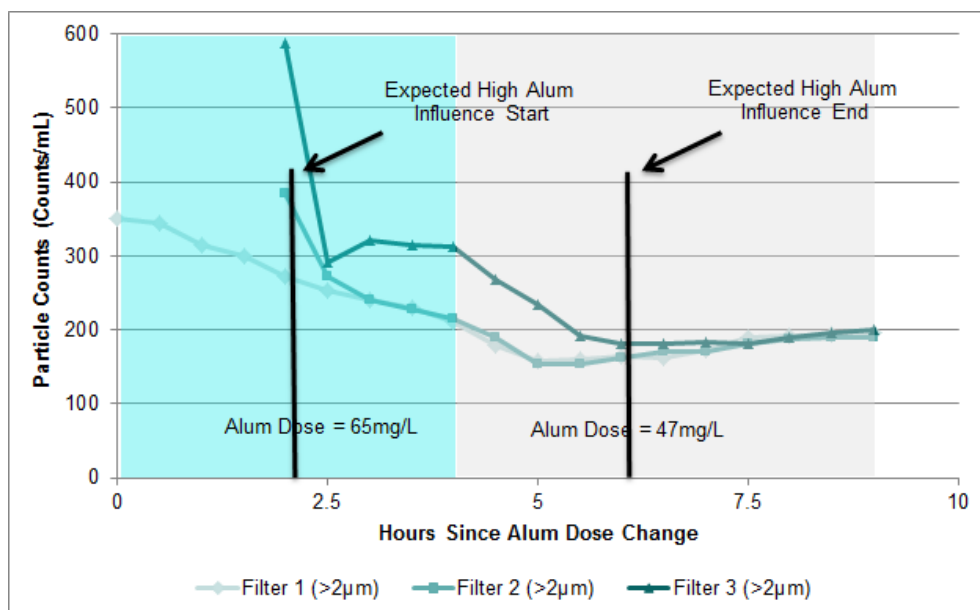

Figure 4: Effect of Alum Dose Change on Filtered Water Particle Counts (Feed Water Turbidity: 0.3-0.6 NTU). Polymer Dose: Filter 1 and Filter 2 = 0.01 mg/L; Filter 3 = 0 mg/L

Effect of Ozonation

In this trial the dual media filters were tested with a filter aid polymer dose of 0.01 mg/L on all filters. The full-scale plant was operated using a typical alum dose of 47 mg/L (as 46% solution). Ozone was injected into the pilot plant feed water and a ≤0.1 mg/L ozone residual after approximately 14.5 minutes ozone contact period was maintained at the outlet of the ozone contactor. The ozonated water was then fed to the pilot plant filters into which the filter aid polymer was dosed.

The results are presented in Figures 5 and 6 for 'typical' and 'dirty' settled water, respectively. The particle count reductions in Figures 5 and 6 are shown for both >2μm particles and >5μm particles since these particle size bands are of particular concern for the removal of *Cryptosporidium* oocysts (3-5μm) and *Giardia* cysts (8-10μm) (Hargesheimer et al., 1992).

Note that prior to the ozone trial, each filter had recently been backwashed. That is, each filter started the run 'fresh'. Therefore, the high particle counts seen at the beginning of the trial were due to filter ripening effect.

As can be seen in Figures 5 and 6, the combination of ozonation and a small filter aid polymer dose (0.01 mg/L for 'typical" feed water, or 0.03 mg/L for 'dirty' feed water) reduced the residual particle counts dramatically in comparison to the trials described earlier which used filter aid polymer and/or increased alum dose. Particle counts were reduced to below 100 particles/mL for >2μm particles, both for 'typical' and 'dirty' feed water, representing reductions of approximately 60% and 85%, respectively, when comparing the steady-state data between the 'Ozone on' and 'Ozone off' periods.

Figures 5 and 6 also show that when ozone was switched off, the filtered water particle counts rose almost immediately to levels typically seen in earlier polymer trials without ozonation. When the ozone was switched back on (trial only done for the 'dirty' feed water), the particle counts were reduced down to similar levels to those prior to ozone being switched off.

Becker et al. (2004) and Bourgine et al. (1998) reported similar results to what was found during this trial but without the use of a filter aid polymer. Unfortunately, ozone effects in the absence of a filter aid polymer were not investigated in this study. Therefore, the results cannot be compared.

Bourgine et al. (1998) also suggests that an ozone residual of 0.15 mg/L should be maintained to improve particle reduction. This is consistent with our pilot plant investigation, which was set to maintain an ozone residual of 0.1 mg/L after the required contact time.

Interestingly, Wu et al. (2008) found in a tertiary Ozone/BAC pilot plant study that increases in post-ozone dosage resulted in a decrease in particle removal by the activated carbon layer, but this influence was alleviated by improvements in particle removal by the sand bed. However, there was still an overall increase in particle removal across the dual media filter.
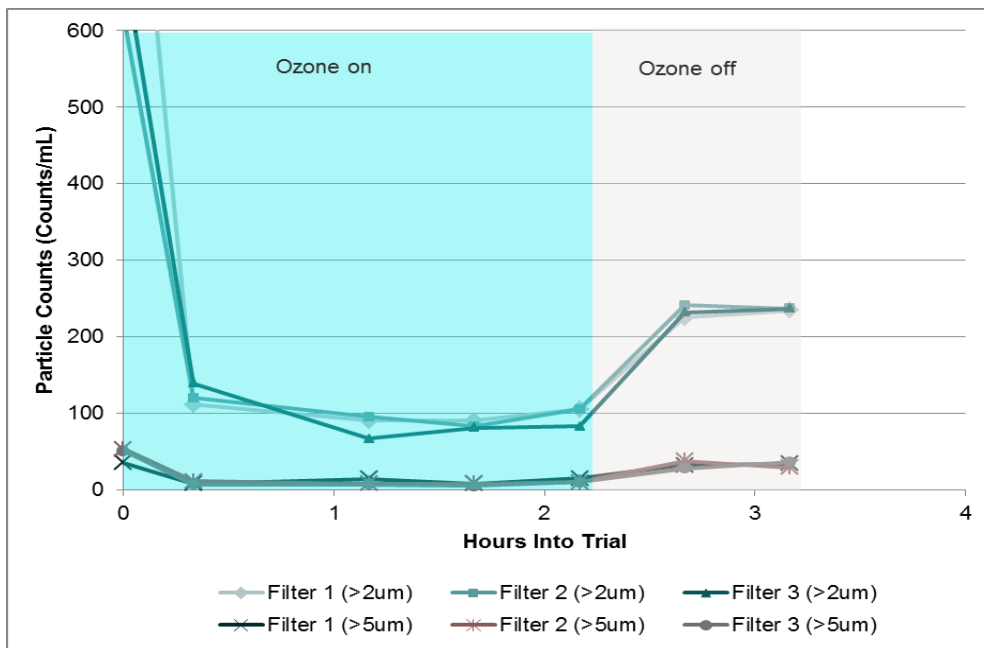

Figure 5: Impact of Intermediate Ozonation on Residual Particle Counts ('Typical' Feed Water: 0.6-0.8 NTU). Polymer Dose = 0.01 mg/L on All Filters
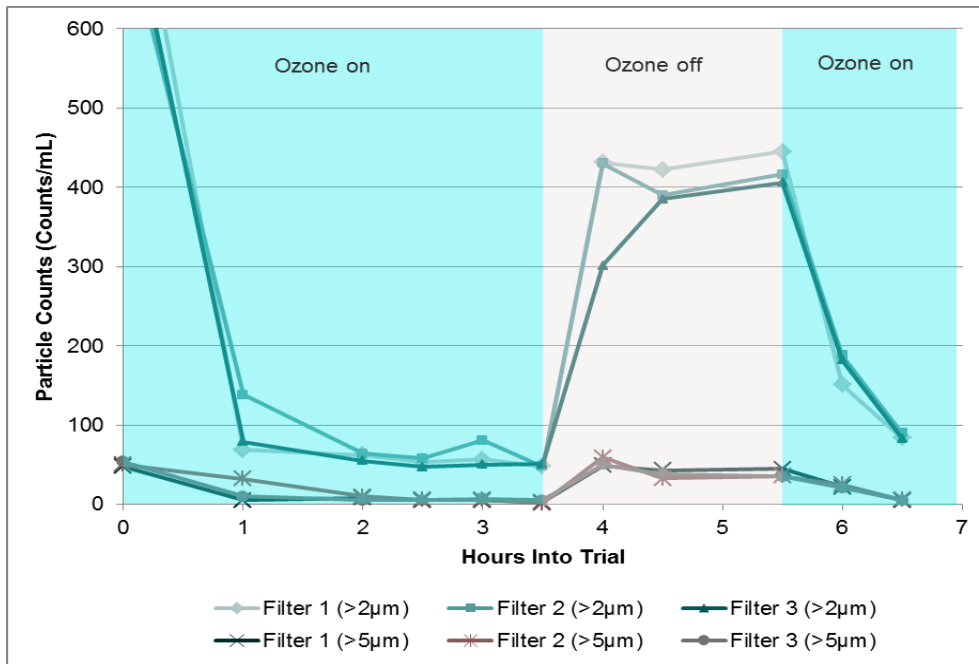
Figure 6: Impact of Intermediate Ozonation on Residual Particle Counts ('Dirty' Feed Water: 1.7-2.3 NTU). Polymer Dose = 0.03 mg/L on All Filters

Underlying Mechanisms

Filter Aid Polymer

It is widely accepted that dosing a filter aid polymer prior to filtration is to condition the filtration media and make it more "sticky" to the floc and hence better floc binding to the media. This does not exclude the possibility that floc growth as a result of the added polymer also takes place, through the so-called bridging flocculation mechanism, prior to water entering the filter media. The resultant larger and stronger flocs can also lead to better particle removal.

Coagulant

Metal salt coagulants such as alum provide cations including cationic metal complexes from hydrolysis that can neutralise the surface charges of particles in the water which are typically negatively charged. This destabilisation mechanism is known as charge neutralisation and takes place rapidly and hence requires flash mixing. At higher doses metal salt coagulants themselves hydrolyse to form hydroxide flocs. These flocs enmesh and "sweep" the colloidal particles from the water. This enmeshment of particles is known as sweep flocculation.

Sweep flocculation generally results in larger and stronger flocs than charge neutralisation (Gregory, 2006). Such flocs have better settleability and filterability.

Ozone

How ozone impacts on particles in the water and in particular enhances particle removal is not well understood. A number of theories have been proposed, most of which relate to pre-ozonation (i.e., prior to adding coagulant). These include the so-called ozone-induced microflocculation (or the destabilisation or coagulation of the constituents in the raw water as a result of the use of ozone). Pre-ozonation has sometimes been found to increase the efficiency of the coagulation, flocculation and clarification processes, and reduce coagulant doses as well as improve filtered water particle counts (Jekel, 1994; Mazloum et al., 2004; Park et al., 2001).

Ozone-induced microflocculation must be related to the effect that ozone has on dissolved and particulate matter in the raw water. Some researchers (e.g., Singer et al., 2003) have attributed the enhanced particle removal through pre-ozonation to the breakup (oxidation) of large natural organic molecules that are adsorbed onto the surface of particles. The adsorbed organic layer is usually thick to overcome the attractive van der Waals forces and hold colloidal particles in suspension. Due to ozonation, the organic layer decreases in thickness and the smaller, more polar molecules (as a result of ozonation) become less of a steric hindrance. This allows the particles to come together and aggregate more easily.

A number of other postulated mechanisms have also been cited by Bablon et al. (1991) and Rahman et al. (2010). One of these is *polymerisation of organic matter*. It is believed that ozonation of natural organic matter (NOM) present in the water leads to the formation of metastable organics which might encounter other stable or metastable organics and undergo condensation or polymerisation reactions. Such reactions may lead to the formation of a polymer that can cause a number of secondary effects such as acting as a "bridging" polyelectrolyte. Although as mentioned above, most of these mechanisms were proposed to explain pre-ozonation effects, ozonation of settled water as performed in this study might also result in the formation of "bridging" polymers which in turn acted as a filter aid. Nevertheless, the dramatic reductions in the filtered water particle counts after ozonation as seen in Figures 5 and 6 suggest that in addition to the polymerisation effect, there may be other complex factors in play, such as decreased particle stability and hence better particle attachment efficiency (Becker et al., 2004).

Implications

Particle counts in filtered water can act as a surrogate for filter breakthrough and potential contamination of specific contaminants of concern such as *Cryptosporidium* and *Giardia*.

Water treatment plants having properly designed filters should be able to improve filtered water particle counts (and turbidity) through optimisation of the coagulant and/or filter aid polymer dose. Use of ozone may be of particular interest for plants where it is undesirable to use either a high coagulant dose (e.g., in direct filtration process) or filter aid polymer (e.g., in coated media process or for filters without air scour facility).

For treatment plants facing disinfection by-product (DBP) issues, ozone can be a better alternative oxidant to chlorine because ozone has less potential to produce DBPs. For waters with high background bromide concentrations, the risk of forming bromate upon ozonation can be mitigated through a range of measures including lowering the ozonation pH. Improved particle removal using ozone renders another level of protection against *Cryptosporidium* and *Giardia* risks, regardless of whether or not the ozonation process is designed for disinfection purpose.

It is worth pointing out that ozonation effects are greatly influenced by the characteristics of raw water (for pre-ozonation) or clarified water (for intermediate ozonation) such as NOM type and concentration, pH and hardness (particularly the calcium ion concentration). The effects may not be observed with all waters. Therefore, when considering the use of ozone for enhancing particle removal, the effects of ozonation should be critically evaluated in pilot studies.

CONCLUSION

This investigation has shown that filtered water particle counts can be dramatically reduced through a combination of conventional (coagulant and/or filter aid polymer) and oxidation approaches. It has also argued that for some applications ozone can be a better alternative to other oxidants.

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