Wastewater Disinfection with Peracetic Acid (PAA), UV Irradiation and Combined PAA-UV Treatments

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Abstract
Both UV irradiation and peracetic acid (PAA) are strong and proven disinfectants. In this study, we have investigated the potential synergistic effect of combined PAA and UV irradiation treatments for wastewater disinfection. Initial benchtop studies were followed by a side-stream pilot study to investigate the effect of both PAA and UV irradiation individually or together in combination. In the benchtop studies, the final effluent samples were split into three parts. The first part was treated with various doses of PAA ranging from 1.0 to 5 ppm. The second part was exposed to 10 mJ/cm^2 dose of UV and the final or third part of effluent was first treated with 1-5 ppm doses of PAA followed by 10 mJ/cm^2 of UV irradiation. Both benchtop and side-stream pilot studies found that pre-treatment of effluent with PAA significantly increased the disinfection efficiency of UV. Without PAA pre-treatment, 10 mJ/cm^2 UV dose achieved a 2.0 log reduction in fecal coliform and E. coli in the benchtop studies. With PAA pre-treatment, and 10 mJ/cm^2 UV irradiation, a 3.5 log reduction was achieved with 5 ppm and 5 minute contact time. Similar pattern was observed in the pilot study in which UV's disinfection efficiency significantly increased after pretreatment of effluent with low dose of PAA. These data suggest that the PAA/UV sequential treatment is more effective than the UV or PAA treatments separately. This strategy can be used to significantly lower the disinfection dose of UV irradiation which in turn can save the utility financial resources in the form of reduced energy needs.

Introduction
Disinfecting wastewater effluent is a final yet critically important step in the treatment of wastewater. It protects the public's health and the environment by inactivating or destroying disease-causing organisms such as bacteria, viruses, and parasites. Various methods and technologies are used to accomplish the goal of effluent disinfection. These include ultraviolet (UV) irradiation (Carmineo et al., 1994; Lazarova et al., 1998; Kolch, 2000), ozone treatment (Lazarova et al., 1998; Andreottola et al., 1996), and the use of various chlorine derivatives (Hajenian & Butler, 1980; Zanetti et al., 1996; Legnani et al., 1996). In the USA, wastewater effluent is mainly disinfected by chlorine derivatives because of their wide spectrum disinfection efficiency and low treatment cost. Recent research, however, has evoked concerns about effluent chlorination promoting the formation of toxic, mutagenic, and carcinogenic properties in its disinfection by-products (DBPs). These harmful DBPs increase the toxicity of the effluent that is discharged into water bodies with potential to cause harm to the water quality and the environment (Dell'Erba et al. 2007; Kauppinen et al. 2012; Veschetti et al. 2003). These concerns have led to the search of a new disinfection like peracetic acid (PAA).

Peracetic acid (PAA) is a clear, colorless liquid available at a concentration of 12% to 15% in an equilibrium mixture of acetic acid, hydrogen peroxide and water (Block 1991; Alsari et al., 1992; Gehr et. Al., 2002):

$$\underset{\text{Acetic Acid}}{CH_3COOH} + \underset{\text{Hydrogen Peroxide}}{H_2O_2} \leftrightarrow \underset{\text{Peracetic Acid}}{CH_3COOOH} + \underset{\text{Water}}{H_2O}$$

PAA is a strong oxidizing organic compound with a wide spectrum of antimicrobial/biocidal properties similar to liquid chlorine or sodium hypochlorite (NaOCl). It has been widely used in the food, beverage, medical, and pharmaceutical industries for over 20 years (Kitis, 2004). Because of its strong antimicrobial properties, PAA has been getting a lot of attention as a wastewater disinfectant to replace chlorine in recent years (Lefevre et al., 1992; Baldry et al., 1995; Sanchez-Ruiz et al., 1996; Stampi et al., 2001, 2002; Wagner et al., 2002). It has been reported that PAA and sodium hypochlorite have similar antimicrobial activities against E. coli, fecal coliform, and total coliform (Veschetti et al., 2003); however, PAA holds multiple advantages over sodium hypochlorite as disinfectant for wastewater effluent. These advantages include: need for lower doses, lower residuals, faster disintegration, and absence of disinfection byproducts (DPBs) in the treated effluent (Booth and Lester, 1995; Liberti and Notarnicola, 1999; Monarca et al., 2000; Kitis 2004; Vaschetti et al., 2003; Crebelli et al., 2005; Koivunen & Heinonen-Tanski, 2005; Antonelli et al., 2013).

When oxidizing agents, such as PAA, H_2O_2, and ozone (O_3) are used in combination with UV irradiation, a highly reactive hydroxyl radical (°OH) is formed which reacts vigorously with biological, organic and inorganic matters and act as disinfectants. PAA goes through a photolysis process in the presence of UV interrupting the O-O bond in the PAA molecule. This reaction leads to the formation of hydroxyl (°OH) radical (Ceretti & Lubello, 2003). Formation of hydroxyl radicals during the UV treatment is considered the key for the synergistic effect of PAA/UV combination treatment.

The purpose of this study was to investigate if pre-treatment with low dose PAA can synergistically increase the efficiency of UV disinfection of wastewater effluent. The study presents data on wastewater disinfection achieved separately with PAA and UV, and with the PAA-UV combination treatment.

Materials and Methods

Materials: The effluent samples were collected from the Little Miami wastewater treatment plant owned by the Metropolitan Sewer District of Greater Cincinnati (MSD), Ohio, USA. Peracetic acid (15%), marketed under the commercial name of VigorOX WWT II, was supplied free of charge by PeroxyChem, Philadelphia, USA. Sodium hypochlorite (NaOCl) (12%) was supplied by PVS Chemical Solutions, Chicago, USA. E. coli and fecal coliform broth were obtained from Hach. Buffered water with magnesium, micro filters, sampling bottles, and microbiological petri dishes were from Thermo Fisher Scientific, Pittsburg, USA.

Methods:

Bench Study Experimental Set-Up

Grab samples of non-chlorinated secondary effluents were collected in sterile 100 milliliter plastic bottles at the Mill Creek Wastewater Treatment plant, Cincinnati, Ohio. The samples were divided into three parts for treatment: (1) PAA only (2) UV only, and (3) sequential treatment with PAA and UV combination. For PAA only, samples were treated with 1, 3, or 5 ppm of PAA concentrations for 1 to 5 minute contact time. The second portion of the sample was irradiated with a single dose of 10 mJ/cm^2. The third and final portion of the sample was first treated with PAA (1-5 ppm for 1-5 min contact time) followed by a single exposure of 10 mJ/cm^2 UV irradiation. Treated samples were analyzed for fecal coliform and E. coli to measure the microbial inactivation after each treatment using the membrane filtration method (Standard Methods, 22nd Ed., American Public Health Association).

Pilot Study Experimental Design

Pilot Study Experimental Set-Up

For the pilot study, non-chlorinated secondary effluent was continuously pumped from Mill Creek wastewater treatment plant, Cincinnati, Ohio, into the US Environmental Protection Agency's Testing & Evaluation facility. The secondary effluent was then blended with PAA to

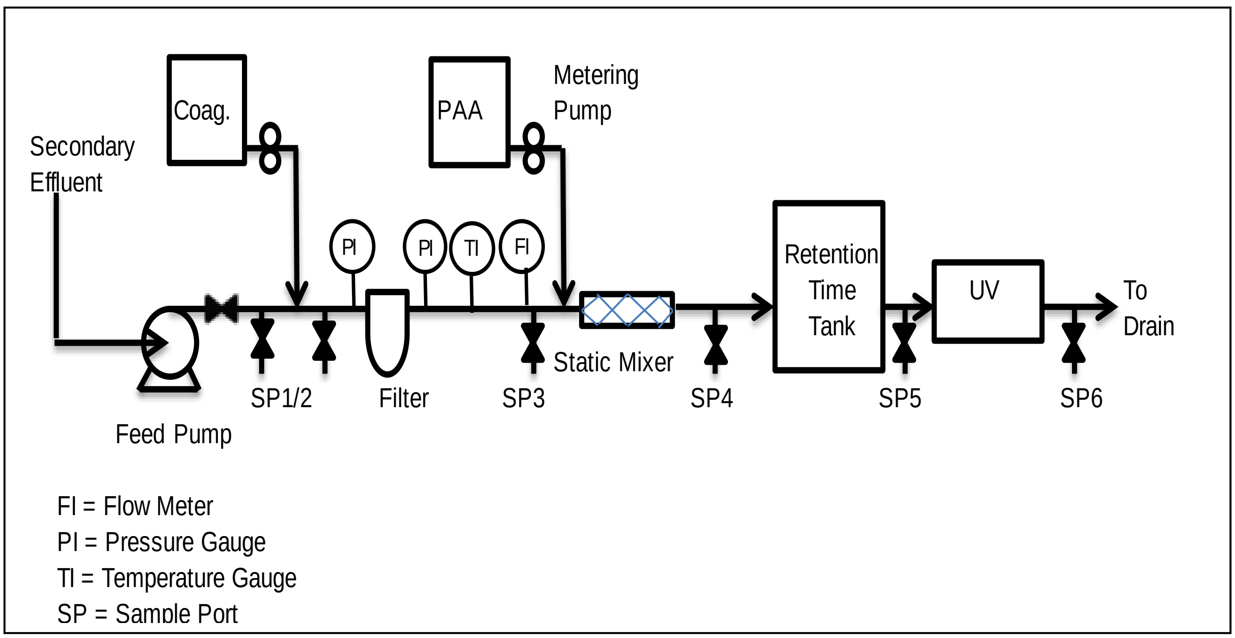

Fig. 1: Schematic of the wastewater effluent treatment with UV and PAA combination

achieve the final concentration between 0.5 and 2.5 ppm and pumped through a Sanitron Model S50B UV Water Purifier for the UV disinfection studies (Fig. 1). The Sanitron unit is rated to provide a UV dose of 30 mJ/cm^2 at a water flow rate of 20 gallons per minute (gpm). This UV dose is proportional to the water flow rate; therefore, a flow rate of 40 gpm would result in a UV dose of 15 mJ/cm^2 and a flow rate of 120 gpm in a UV dose of 5 mJ/cm^2.

A 10-micron bag filter was installed prior to the Sanitron unit to remove suspended solids fine particulates from the effluent. It has been shown that suspended solids interfere with operation of UV disinfection systems and increase the PAA demand of wastewater. Figure 1 shows a flow diagram for the pilot-scale PAA/UV disinfection tests.

Microbiological Analysis during Pilot Study:
The effluent samples for microbial analysis were collected in 100 ml sterile plastic bottles containing 10 mg sodium thiosulfate (Thermo Fisher Scientific, Cat# 05-719-361) for neutralizing any residual PAA and H_2O_2 instantaneously. Each analysis was carried out on fecal coliform and E. coli using a membrane filter. The fecal coliform colonies were counted after incubation for 24 ± 2 hours in a 44.5 ± 0.2ºC water bath. *E. coli* plates were incubated for the same time in a 35 ± 0.5ºC water bath (Standard Methods, 22nd Ed.).

UV only treatment: Non chlorinated secondary effluent from the Mill Creek WWTP was pumped into the UV treatment system at different flow rates to obtain final UV dosages of 6, 10, 20 and 40 mJ/cm^2 to study the effectiveness of UV only treatment. The samples were analyzed for fecal coliform and *E. coli*.

Combined PAA and UV treatment: A total of five experiments were conducted by applying a low PAA + low UV intensity, low PAA + high UV intensity, moderate PAA + high UV intensity, moderate PAA + low UV intensity and finally high PAA + low UV intensity. This experimental design allowed an understanding of the relative contribution of each of the treatment components in the combined disinfection system. The PAA concentrations evaluated were 0.5 mg/L (low); 1 mg/L (moderate) and 2.5 mg/L (high), applied for a maximum of 30 min contact time. The flow in the UV system were adjusted to produce UV intensities of 7 – 20 mJ/cm^2 (Low); 40 – 60 mJ/cm^2 (moderate) and 120 mJ/cm^2 (high). The effluent from the UV system was analyzed for *E. coli* and fecal coliforms.

Results:

Comparing disinfection efficiencies of UV with PAA and PAA/UV Sequential Treatments in the lab:

The disinfection efficiency of various doses of PAA and contact periods were compared in the lab with UV (10 mJ/cm^2) and sequential PAA/UV combination treatments. The disinfection rate was measured by estimating the inactivation of E. coli and fecal coliform microbes. As shown in figures 2 and 3, a dose- and time-dependent response was observed on E. coli and fecal coliforms with PAA. No such time-dependent response was observed with UV irradiation which was a single onetime exposure. A 2-log reduction was observed with the single UV dose of 10 mJ/cm^2 which remain unchanged from 1 minute to 5 minutes. However, with 1 ppm PAA, initially about 0.5 log reduction was observed at 1 minute which increased to a 1.6 log reduction after 5 min contact time. With 3 ppm PAA, fecal coliform counts were reduced by a 1.6 log at 1 min and 2.5 log reduction at 5 min. A 5 ppm dose of PAA caused a 3 log reduction in the fecal counts at 1 min and 3.5 log reduction after 5 minutes. (Fig 2). Similar 2.5 to 3 log reduction was observed in the E. coli concentration after treatment with 3 ppm and 5 ppm of PAA respectively.

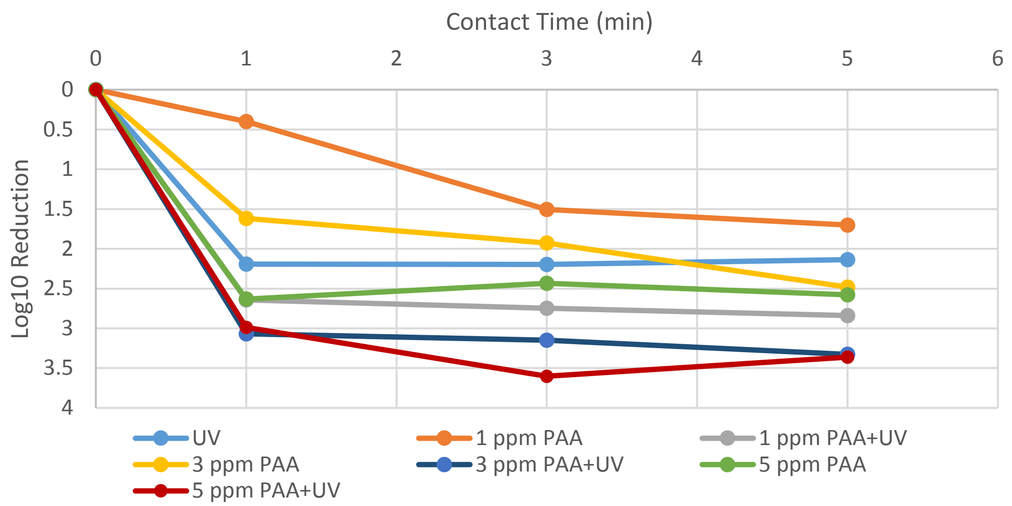

Fig. 2 Fecal Coliform Log^{10} Reduction vs Contact Time with 10 mJ/cm^2 UV

Sequential PAA/UV treatment was found to be significantly more effective than PAA or UV treatments alone. Pre-treatment of effluent with 1 ppm of PAA followed by 10 mJ/cm^2 UV

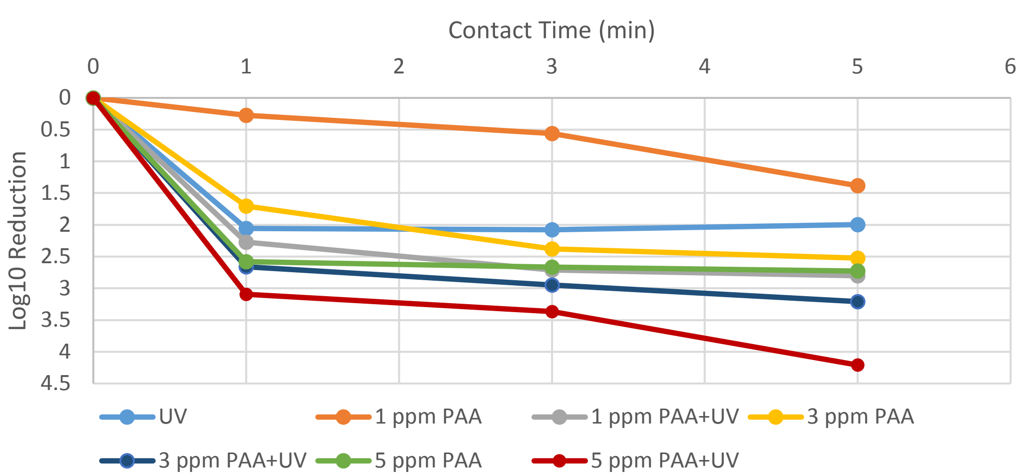

Fig. 3 E. coli Log^{10} Reduction vs Contact Time with 10 mJ/cm^2 UV

treatment increased the disinfection from a 0.5 log to 2.5 log reduction for both fecal coliform and E. coli at 1 minute and about 3 log reduction if the effluent was treated with 1ppm PAA for 5 minutes prior to UV exposure. Similar significant increase was observed both with 3 ppm and 5 ppm doses. The sequential treatment with 3 ppm of PAA and 1 minute contact time and 10 mJ/cm^2 UV exposure doubled the inactivation rate from 1.5 log reduction to 3 log reduction. A similar increase was observed when 5 ppm PAA was combined with 10 mJ/cm^2 UV treatment (Figs 2 and 3).

Side-stream Pilot Study:

UV-only treatment

The initial E. coli counts and the log removal achieved by UV only treatment are plotted in Figure 4. Log removal of the pathogen improved with increasing UV intensity with very low effluent counts at 40 mJ/cm^2. The removal of *E. coli* increased steadily with increase in UV intensity (Figure 4). This implies that in the tested wastewater matrix, *E. coli* was very sensitive to UV disinfection.

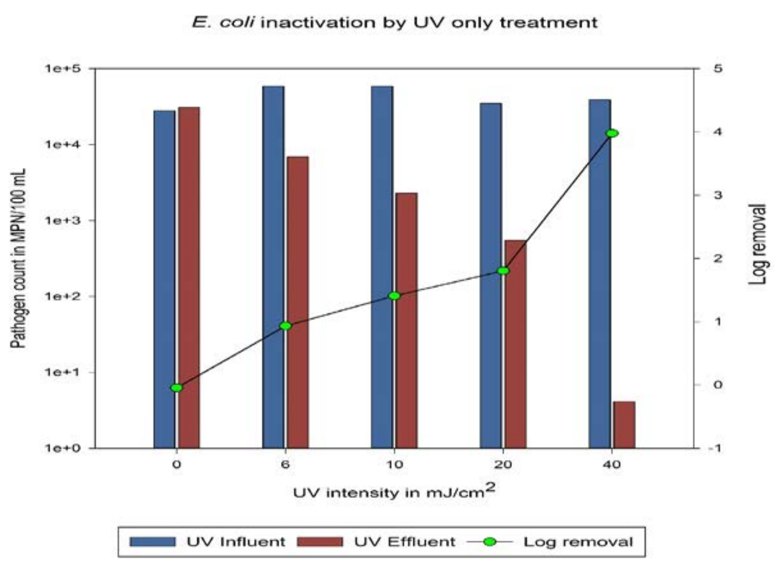

Figure 4 – Disinfection efficiency of UV only treatment in secondary wastewater effluent.

Combined PAA + UV treatment

The effectiveness of the sequential PAA and UV treatment was tested under different PAA and UV dose combinations. Since the secondary effluent was filtered before any treatment, the total solids concentration were always less than 5 mg/L. The pathogen counts measured after PAA treatment and PAA+UV treatment were plotted along with the corresponding log removals. Pathogen count or log removal after PAA + UV refers to the overall process efficiency and the difference between the treatment efficiencies of PAA and the combined treatment provides a measure of the impact of the UV treatment in a given experiment.

Low PAA + Low UV intensity

Low PAA dosage (0.5 mg/L) and low UV intensity (7 mJ/cm^2) showed poor disinfection performance (Fig. 5). PAA alone did not kill any of the pathogens at this dose level, showing negligible log removals even after 30 min contact time. But the low UV intensity resulted in less than one log removal of *E. coli* and fecal coliforms. Consistent with the trend seen from the UV only experiment, both were inactivated at the similar rate.

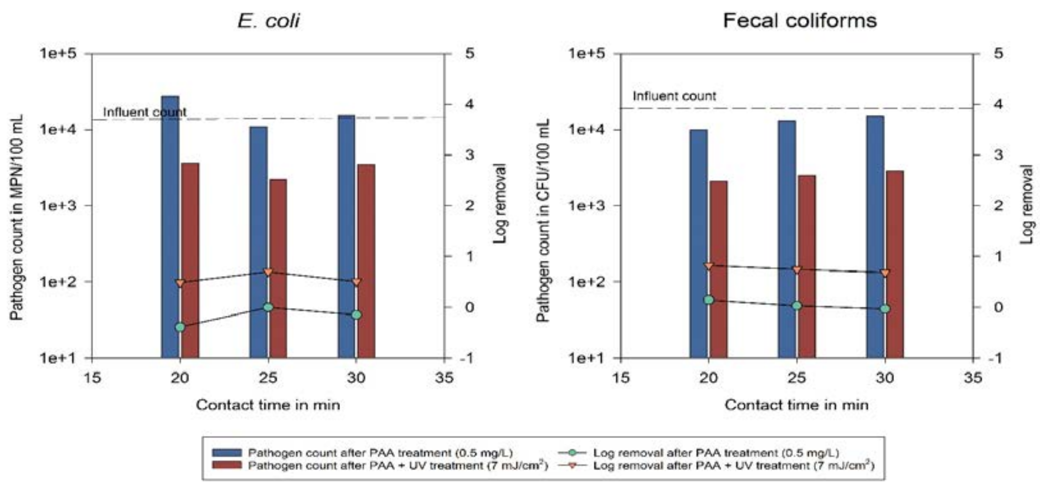

Figure 5 a, b – Disinfection efficiency of combined treatment with low PAA (0.5 mg/L) and low UV (7 mJ/cm^2) dosage.

Moderate PAA + High UV intensity

The disinfection trends at a moderate PAA level of 1 mg/L was similar to the previous low PAA dosage experiment with respect to removals of enterococci and fecal coliforms. Over two log removal was observed for fecal coliforms. *E. coli* inactivation improved at 1 mg/L PAA dosage, with counts decreasing steadily with time and a reported 1.2 log removal at 30 mins (Fig. 6). As expected, applied UV doses (120, 60 and 40 mJ/cm^2) achieved virtually complete inactivation of the pathogens studied, with microbial counts below detection.

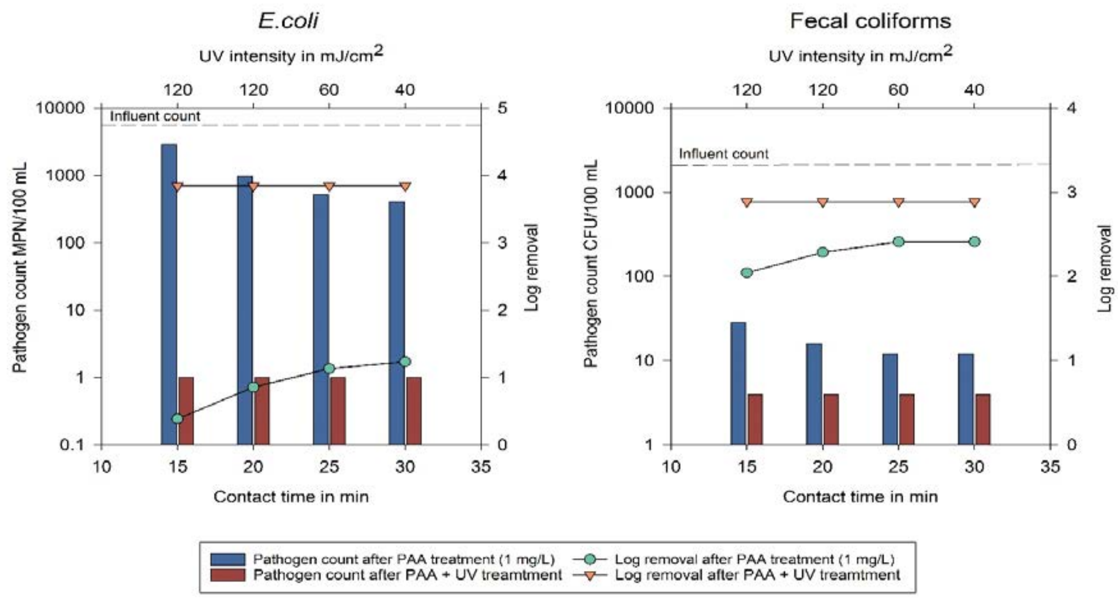

Figure 6 a, b – Disinfection efficiency of combined moderate PAA (1 mg/L) and high to moderate UV (120 to 40 mJ/cm²) treatment. Contact time of the PAA treatment shown in bottom x-axis. Top x-axis shows the UV intensity applied at the corresponding time point.

High PAA dosage + High UV intensity

A high PAA dosage of 2.5 mg/L resulted in excellent disinfection performance even at high influent concentrations for both E. coli and fecal coliforms (Figure 7 a, b). The influent *E. coli* concentration of 64,000 CFU/100 mL decreased to 740 CFU/100 mL in 15 min (Figure 7 a) and the subsequent moderate UV intensity treatment was sufficient to bring the counts to below detection (< 1 CFU/100 mL). For fecal coliforms, a PAA concentration of 2.5 mg/L completely inactivated them to levels below detection (< 10 CFU/100 mL) even at 15 min time. Since there were not any fecal organisms available for disinfection in the UV system, the overall log removal remained constant around 3 (Figure 7 b).

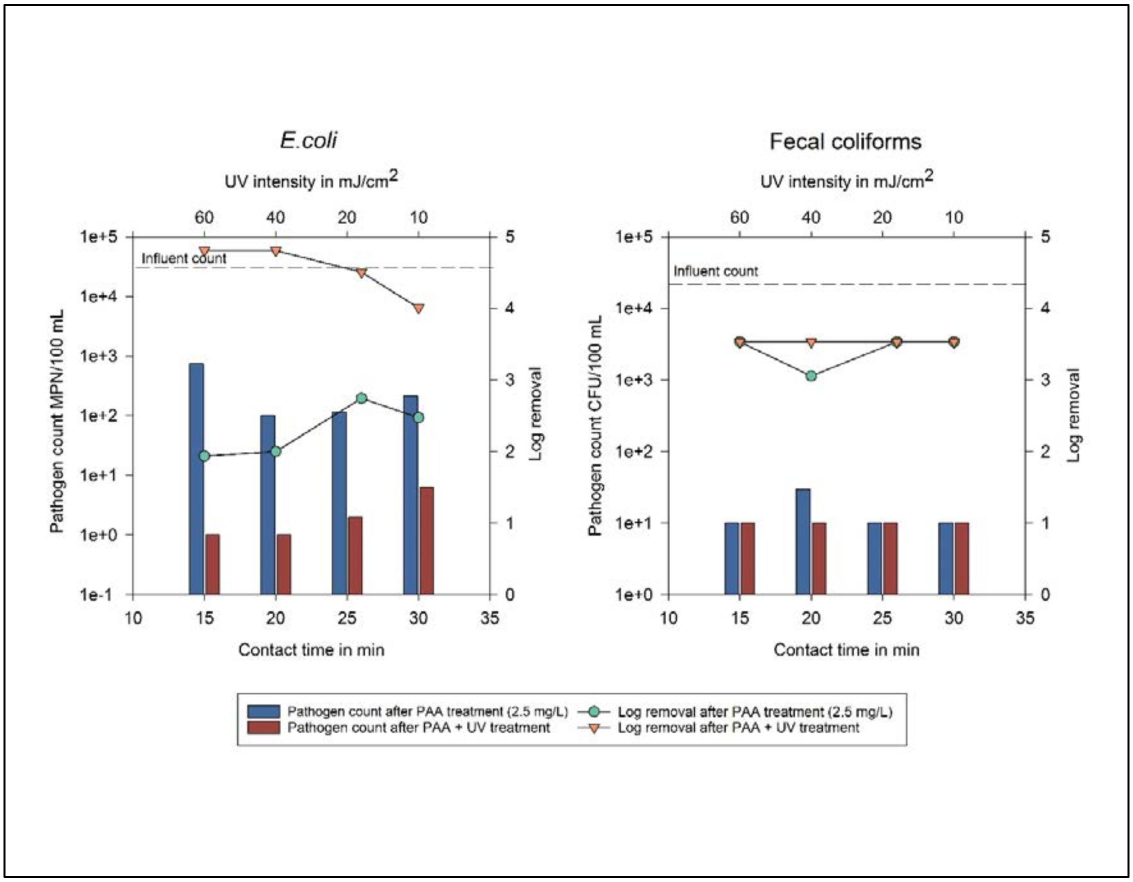

Figure 7 a, b – Disinfection efficiency of combined high PAA (2.5 mg/L) and moderate to low UV treatment. Contact time of the PAA treatment shown in bottom x-axis. Top x-axis shows the UV intensity applied at the corresponding time point.

Discussion:

This study was conducted in two phases. The first phase was conducted in the lab to investigate the disinfection effectiveness of PAA and UV irradiation separately and then by using a combination of PAA/UV for the treatment of wastewater. In the second phase, the lab studies were repeated in a side-stream pilot study. Again, PAA and UV efficiencies was investigated separately and in a PAA/UV combined treatment. In our benchtop studies, the PAA and UV efficiencies appeared to be directly proportional to both concentration and contact time (C.t; where "C" being concentration and "t" being contact time), suggesting a low dose of the disinfectant with prolonged contact time can achieve the same degree of disinfection as the higher dose with shorter contact time. This observation is consistent with previously published reports (De Luca et al., 2008; Koivunen & Heinonen-Tansski, 2005; Wagner et al., 2002). We achieved about 0.5 log reduction both in E. coli and fecal coliform with 1 ppm of PAA at 1 min contact time which increased to about 1.6 logs after 5-minute contact time. At 5 ppm dose of PAA, about 3 log reduction was achieved in E. coli and fecal coliform after 1 minute contact time which was 6 time greater than the 1 ppm dose at the same contact time. Thus, the reduction in the microbial concentration was directly proportional to the PAA dose. Stampi et al. (2001) achieved a 5 log reduction in total coliform and E. coli with 1.5 to 2 mg/L of PAA after

20 minute contact time; Madoni et al., (1998) reported a similar reduction in total coliform and enterococci density at 3 mg/L and 15 minute contact time, while Lazorva et al., (1998) used much higher doses of up to 10 mg/L for 10 minute to get 3 log reduction in total and fecal coliform.However. When UV and PAA were compared to one another, UV was found to be a more effective than low dose PAA alone. At higher doses (5 ppm) though PAA worked better than UV. Because the quality of the wastewater and its secondary effluent are important factors in determining the efficiency of PAA, the PAA disinfection dose cannot be generalized. Each wastewater plant must determine the optimal dose and contact time of PAA to disinfect effluent after characterizing the quality of its effluent.

Disinfection was proportional to contact time and significantly increased with the increase in contact time. Initial inactivation of fecal coliform and E. coli up to 3 minutes was fast and quick which followed by inactivation at a slower rate between 3-5 minute. This phenomenon has been reported previously by several labs (Koivumen & Heinonen-Tanski, 2005; Luukkinen et al., 2014)**;** Antonelli et al., 2006; Falsanisi et al., 2006. It is believed that free-floating microbes are inactivated promptly by PAA in the first phase; in the second phase, there is a slower inactivation of microbes, showing a tailing of the inactivation curve phase (Tyrrell et al., 1995; Liberti et al., 2000). Although the precise mechanism of microbial inactivation in the second or slow phase is not clearly understood, it is proposed that the presence of suspended solids in the effluent appear to provide shelter to microbes from PAA thus requiring either a higher PAA dose or longer contact time (Koivumen & Heinonen-Tanski, 2005). This further suggests microbial inactivation is a direct function of both PAA concentration and contact time.

Both PAA and UV are strong wastewater disinfectants. Broad spectrum of disinfection properties, high rates of disinfectant efficiency at lower doses and shorter contact times, little to no impact on the presence of organic matter present in wastewater, lack of disinfection byproduct (DBP) production, and ease of use are some of the benefits associated with PAA and UV (Koivunen & Heinonen-Tanski , 2005; Nurizzo et al., 2001). However, combining PAA with UV in a sequential treatment has shown significant improvement in the microbial disinfection efficiency over individual UV or PAA treatments (Koivunen & Heinonen-Tanski, 2005).

UV and PAA inactivate microbes by different mechanisms. UV irradiation attack the genetic material, i.e., DNA, of the bacteria while PAA kills the bacteria by causing damage to its cell wall and its critical enzymes by removing electrons from susceptible chemical groups or the cellular components (Melly et al., 2002; Linley et al., 2012; Leggett et al., 2015; Finnegan et al., 2010). Thus, when the bacteria are exposed to a non-lethal low dose of PAA, it may cripple the enzyme system that repairs the cellular damage. The inability of bacteria to repair cellular and DNA damage leads to its destruction when the bacteria is irradiated subsequently with UV.

There are two-fold advantages of using the combined PAA/UV disinfection treatment. One, it can potentially reduce the disinfection cost by requiring lower dose of UV and thus saving the energy cost. Second, it can improve the bacteriostatic effect of UV disinfection by broadening the range of microbial disinfection. Generation of highly reactive oxidative and hydroxyl (°OH) radicals are considered responsible for the increased disinfection ability in PAA/UV combined treatment. Oxidative radicals are produced by PAA and the hydroxyl radicals are generated by the reaction between UV and PAA or H_2O_2. For PAA/UV combination treatment to be successful, it is important that the non-lethal dose of PAA is introduced prior to UV treatment so the reactive oxidative and hydroxyl radicals can be formed during the UV irradiation. By combining the disinfection powers of PAA and UV, an extensive and permanent damage can be caused to a much wider range of bacteria potentially at a lower cost. Additional studies are currently in progress to investigate the impact of other wastewater characteristics such as suspended solids and other water components on the PAA/UV disinfection efficiency.

Conclusions: Peracetic acid is a potent oxidizing agent with a wide spectrum antimicrobial activity. UV is also a very effective wastewater disinfectant. When used individually, both PAA and UV need high doses to achieve full inactivation of fecal coliform and E. coli bacteria. However, pretreating secondary effluent with a low dose PAA prior to UV irradiation can significantly reduce the UV dose needed for disinfection. Our initial benchtop studies in the lab were confirmed by a side-stream pilot study on secondary effluent. Our studies suggest that a combined PAA/UV treatment is much more effective in achieving the disinfection of the secondary effluent. The combine PAA/UV treatment can reduce the cost of disinfection treatment and can be effective against a much wider range of microbial.

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