COMBINED SEWER OVERFLOW PRETREATMENT WITH CHEMICAL COAGULATION AND A PARTICLE SETTLER FOR IMPROVED PERACETIC ACID DISINFECTION

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ABSTRACT

Many old cities are drained by combined sewer systems in which wastewater mix with rainwater for transport to the wastewater treatment plant. When intense rainfalls occurs the design capacity of combined sewer systems exceeds resulting in discharge of combined sewer overflows (CSO) to nearby surface water resulting in contamination with various pathogenic organisms, suspended solids and chemicals.

The European Union has stated that good microbial quality for bathing water should not exceed 500 MPN/100 mL for *E. coli* and 200 MPN/100 mL for *Enterococcus*. Bathing water quality can be maintained by disinfecting the CSOs.

This study was conducted to characterize the disinfection by peracetic acid in combination with chemical coagulation in a HydroSeparator® system for CSO. HydroSeparator® CSO system is a patented and specialized system consisting lamella settler and mess filter (20 microns). The entire system was installed in Middelfart to treat the CSO from the towns of Båring and Asperup (Denmark) and contains a traditional CSO structure before the HydroSeparator and a constructed wetland to polish the disinfected effluent.

Samples for experiment were collected from inlet and outlet of HydroSeparator at different flow to optimize the PAA dose without coagulation. In experiment II, samples were collected from the inlet of the HydroSeparator to optimize the coagulation dose in a Jar test using PAX-XL 100. Experiment III was performed in full scale by applying PAX-XL 100 as flocculent (5 mg-Al/L) followed by disinfection with 10 mg/L PAA in the HydroSeparator. In order to confirm the PAA dose delivered in the field, comparable PAA treatments were made in the laboratory on samples collected after coagulation.

Turbidity and phosphorus was reduced by applying increasing flocculent doses, but higher than 5 mg-Al/L achieved insignificant improvements. In experiment III the removal of turbidity was 92%, COD 28%, total nitrogen 61% and phosphorus 27% with 5 mg-Al/L.

The stability of PAA increased after the HydroSeperator treatment, but was markedly further improved by the coagulation. Consistently with this, PAA disinfection was more efficient after the HydroSeparator, and further improved by the coagulation. In experiment III, removal of *Enterococcus* was 2.2 log for onsite disinfection and 2.4 log for the laboratory disinfection, which confirms the field dosing considering the analytical uncertainty.

Overall, it is evident that disinfection efficiency of PAA was more effective in the flocculated and HydroSeparator treated water, but as long as the HydroSeparator was applied efficient disinfection could be achieved by PAA dosing towards preserving bathing water quality.

Keywords: Combined sewer overflow, Chemical coagulation, HydroSeparator, Disinfection, Peracetic acid

1. Introduction

In combined sewer systems wastewater is mixed with rain water and transported to wastewater treatment plant for treatment. When intense rainfalls occurs, the design capacity of combined sewer systems exceeds resulting in the discharge of untreated combined sewer overflow (CSO) to nearby surface water. Discharge of untreated CSO affects the quality of receiving water since it contains a variable mixture of rain water, raw sewage, watershed run-off pollutants, variable pathogenic organisms, suspended solids, and chemicals (USEPA, 1999). European Union has regulated the bathing water quality by issuing bathing water directives and to qualify to microbial good bathing water number of *E. coli* should not exceed 500 MPN and *Enterococcus* number should not exceed 200 MPN per 100 mL water(Directive 2006/7/EC, 2006).

Microbial safe bathing water quality can be maintained by disinfecting the CSO water. According to Tchobanoglous *et al.* (2003), an ideal disinfectant should guarantee the maximum efficiency in pathogenic microorganism removal, without generating toxic and undesirable by-products. In addition, it should be inexpensive and technologically compatible (Tchobanoglous *et al.*, 2003). There are various well known disinfectants used in the water industries such as hypochlorite and chlorine dioxide (White, 2010), which could be used to reduce contamination by microorganisms from CSO events, but the by-products of these are of environmental concern (Hrudey and Charrois, 2012; Watson *et al.*, 2012). The organic peroxide, PAA is a strong disinfectant with a wide spectrum of antimicrobial activity that which was introduced to wastewater treatment around 15 years ago (Falsanisi *et al.*, 2006; Kitis, 2004). Commercial PAA is available as an acidic quaternary equilibrium mixture of peracetic acid (PAA), hydrogen peroxide, acetic acid, and water:

$$CH_3COOH + H_2O_2 \rightleftharpoons CH_3CO_3H + H_2O \qquad (1)$$

The degradation products of PAA are acetic acid, hydrogen peroxide, and water. Acetic acid is biodegraded further to carbon dioxide and hydrogen peroxide degrades to oxygen and water; neither of which is considered toxic to aquatic life (Liberti and Notarnicola, 1999). Recently we investigated in laboratory scale the possibility to use peracetic acid and performic acids for disinfection of CSO in terms of doses required for disinfection, kinetic of reaction in water and residual disinfectants (Chhetri *et al.*, 2014). The first full scale demonstration of performic acid disinfection of CSO was just described using a sea outfall pipe north of Copenhagen, Denmark as reaction tank (Chhetri *et al.*, 2015).

The disinfection efficiency can be influenced by inlet water quality since disinfectant will react with other contaminant in untreated water. Therefore, pre-treatment of CSO water with physical chemical process may be considered to improve the disinfection efficiency. The primary purpose of physical chemical treatment with chemical coagulation is to reduce the suspended solids (SS) and the contaminants associated with it. A lamella (series of inclined-plate) clarifier was designed to remove solid particulates from CSO water. When CSO water is passing lamella clarifiers, solid particles begin to settle on the inclined plates and begin to accumulate in the bottom of collection hoppers resulting in effective removal of suspended solids.

Study on chemical coagulation and lamella clarification of CSO had shown efficient removal of total suspended solids, COD, total phosphorus (Jolis and Ahmad, 2004) and heavy metals (El Samrani *et al.*, 2008). A priority pollutants from CSO water was treated by lamella clarification (Gasperi *et al.*, 2010). To our knowledge, there are no studies in literature on the disinfection of CSO water treated by chemical coagulation followed by lamella clarification.

Thus, the application of physical chemical pre-treatment will likely lead to improved disinfection efficiency and alter the degradation kinetics of PAA, thus it will be important to quantify in order to operate a full scale CSO treatment facility. Therefore, the aim of this study was to test the full scale design and predicted performance of a disinfection system for combined sewer overflow (CSO) using Peracetic acid based on our previous work in combination with chemical coagulation in real CSO water. Moreover, degradation kinetics of peracetic acid in CSO water before and after chemical coagulation water will be studied.

2. Methods
2.1. Chemical analysis
Conductivity, COD, turbidity, pH, phosphorus and NH_4^+ were determined according to standard methods (APHA, 2012). PAA concentration was analyzed using the colorimetric method described by Chhetri *et al.* (2014) based on selective oxidation of ABTS by PAA without interference from hydrogen peroxide.

2.2. Site description
HydroSeparator® CSO system was installed in Kærby, a small city in Middelfart, Denmark to treat the CSO from the towns of Båring and Asperup in extreme weather which occurs several times a year and when the treatment capacity of Nørre Aaby wastewater treatment plant exceeds. Before installation of HydroSeparator system, untreated CSO used to discharged to Pavebækken stream which further lead to Storeåen river and finally to the sea near to Varbjerg and Bro. HydroSeparator system was equipped with lamella followed by filter with the sieve size of 20 microns (Figure 1). The HydroSeparator has a capacity to treat CSO water with 5-25 $l·s^{-1}$ flow. The retention time of CSO water in the HydroSeparator was 33 min. To optimize the SS removal from HydroSeparator, chemical coagulation was established in the inlet of HydroSeparator. The treated CSO water from HydroSeparator was disinfected with PAA in reaction chamber which lead to constructed wetlands.
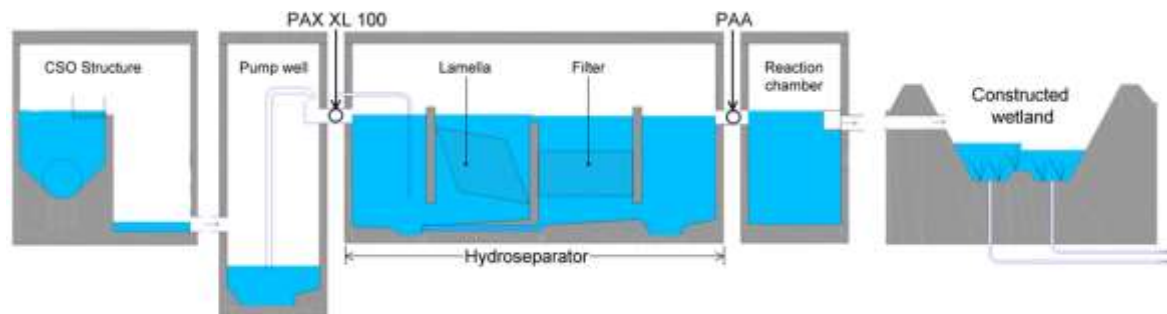

Figure 1: Schematic diagram of Kærby plant including the HydroSeparator.

2.3. Experiment performed
The experiment was divided into two segments: (1) pre-experiment with chemical coagulation and disinfection in lab and, (2) full scale chemical coagulation in the HydroSeparator and disinfection. Pre-experiments were performed in the lab to design the ideal dose of coagulant and disinfectant, which was used for full scale experiment.

Samples were collected from HydroSeparator, one from the inlet and three from the outlets of HydroSeparator with different flow. PAA was added at a dose of 2, 6 and 10 $mg·L^{-1}$ and concentration profiles over time were measured until PAA concentration was completely degraded. In parallel after 60 min of contact time residual PAA from sample was quenched by adding 100 $mg·L^{-1}$ sodium thiosulphate followed by 50 $mg·L^{-1}$ catalase as described by Chhetri *et al.*, (2014) from sample to enumerate *Enterococcus*. Our previous study on disinfection of *E. coli* and *Enterococcus* from CSO water showed that *Enterococcus* was more difficult to disinfect from CSO sample with respect to *E. coli* (Chhetri *et al.*, 2015, 2014). Therefore, disinfection of *Enterococcus* was only considered in this study. *Enterococcus* were enumerated using the Enterolert methods from IDEXX (IDEXX laboratories, Maine, United States) as described by Chhetri *et al.* (2014).

In another pre-experiment, chemical coagulation experiment was performed on the samples from the inlet of HydroSeparator by jar test using different concentration of PAX- XL100 to determine the ideal dose for full scale experiment. Chemical coagulation was done by adding 2.5, 3.5, 5, 7.5, 10 and 15 $mg-Al·L^{-1}$ to influent sample. Turbidity and Ortho-phosphorus was measured from each sample before and after chemical coagulation to measure the removal. The ideal dose obtained from jar test was used for full scale chemical coagulation experiment.

Full scale experiment was performed by applying 5 mg-Al·L^{-1} PAX-XL 100 as coagulant followed by disinfection with 10 mg·L^{-1} PAA in HydroSeparator. Samples from inlet, outlet after chemical coagulation and outlet after disinfection chamber in Hydroseparator were taken and transported to the laboratory for analysis. In order to confirm the PAA dose delivered in the field, comparable PAA treatments were made on field collected samples after chemical coagulation in the laboratory. After 10 min and 60 min of contact time, a fraction of each sample was processed for *Enterococcus* enumeration and in parallel concentration profiles of PAA was followed until 60 min in the remaining sample. In field, sample was collected from outlet of disinfection unit after 10 min and 60 min of contact time and residual PAA was quenched by adding sodium thiosulphate and catalase as described above and *Enterococcus* was enumerated from the samples.

3.   Results

In full scale chemical coagulation experiment, 92% of turbidity, 28% COD, 18% total nitrogen and 27% total phosphorus was removed. Initial consumption of PAA increased with increase in nominal PAA dose. Initial consumption of PAA was 2.7 mg·L^{-1} in average when 10 mg·L^{-1} PAA was used in experiment I whereas initial consumption was 1.67 mg·L^{-1} by applying the same concentration of PAA to disinfect the CSO water pretreated with HydroSeparator and chemical coagulation. While initial consumption of PAA was 0.36 mg·L^{-1} in average when 2 mg·L^{-1} PAA was used to disinfect the CSO water in both experiment I and III.

The area under curve (AOC), a plot of PAA concentration against time (C·t), were calculated for 60 min for all graphs from experiment I and 10 min and 60 min for the graph from experiment III. AOC of inlet samples treated with 2, 6 and 10 mg·L^{-1} PAA were 54, 120 and 152 mg·L^{-1}·min respectively for experiment I which was less than 75, 228 and 400 mg·L^{-1}·min for experiment III where same PAA concentration was applied to treat coagulated CSO water. It was observed that AOC of the inlet samples was less than the samples from HydroSeparator treated with PAA. The AOC of samples after HydroSeparator treatment (flow 5 l·s^{-1}) treated with PAA was 64, 163 and 297 mg·L^{-1}·min whereas AOC of sample with 15 l·s^{-1} and 25 l·s^{-1} flow in HydroSeparator treated with PAA was 63, 159 and 330 mg·L-1·min and 56, 155 and 256 mg·L-1·min respectively. The data for AOC revealed that degradation kinetics of PAA was affected by HydroSeparator treatment and by applying chemical coagulation leads for slower degradation. This is explained by the increased removal of particles and dissolved matter that would otherwise react with PAA. Similarity in removal of *Enterococcus* was observed between laboratory and field-scale disinfection by PAA. The removal of *Enterococcus* increased when treatment time of PAA increased. When the treatment time was increased from 10 min to 60 min, the removal of *Enterococcus* also increased from 0.3 log to 1.0 log in the samples treated with 2 mg·L^{-1} PAA and by applying 6 mg·L^{-1} PAA, removal of *Enterococcus* increased from 1.4 log to 2.2 log with increased in contact time.

The effect of treatment of PAA disinfection efficiency can generally be explained by the difference in the AOC that vary as the degradation of PAA is influenced by reaction with particles and organic matter that is differently removed with coagulation and HydroSeparator. Overall, the disinfection efficiency of PAA was more effective in the coagulated sample where suspended solids and other parameters were removed than non-coagulated samples.

4.   Conclusions

Pretreatment of CSO water had a clear effect on disinfection treatment efficiency and degradation of PAA. Degradation kinetics of PAA and disinfection efficiency both increased when CSO water was pre-treated in HydroSeparator in combination with coagulation. The effect of pre-treatment method on parameters such as total nitrogen and phosphorus might in many cases require to maintain environmental water quality and the decrease in disinfectant dose should be seen as an added benefit.

Overall a combined treatment of coagulation, particle separation and disinfection by PAA seems feasible and able to significantly mitigate negative effects of CSO on surface waters both for aquatic organisms and recreational use.

REFERENCES

1. Antonelli, M., Rossi, S., Mezzanotte, V., Nurizzo, C. (2006), Secondary effluent disinfection: PAA long term efficiency, Environmental Science and Technology. doi:10.1021/es060273f
2. APHA, (2012) Standard Methods for the Examination of water and wastewater (22nd Edition). American Public Health Association /American Water Works Association/Water Environment Federation, Washington DC, USA.
3. Bayo, J., Angosto, J.M., Gómez-López, M.D. (2009), Ecotoxicological screening of reclaimed disinfected wastewater by Vibrio fischeri bioassay after a chlorination-dechlorination process. J. Hazard. Mater. 172, 166–171. doi:10.1016/j.jhazmat.2009.06.157
4. Chhetri, R.K., Flagstad, R., Munch, E.S., Hørning, C., Berner, J., Kolte-Olsen, A., Thornberg, D., Andersen, H.R., 2015. Full scale evaluation of combined sewer overflows disinfection using performic acid in a sea-outfall pipe. Chem. Eng. J. 270, 133–139. doi:10.1016/j.cej.2015.01.136
5. Chhetri, R.K., Thornberg, D., Berner, J., Gramstad, R., Ojstedt, U., Sharma, A.K., Andersen, H.R., (2014), Chemical disinfection of combined sewer overflow waters using performic acid or peracetic acids. Sci. Total Environ. 490, 1065–1072. doi:10.1016/j.scitotenv.2014.05.079
6. Delporte, C., Pujol, R., Vion, P. (1995), Optimized lamellae settling for urban stormwater waste. Water Sci. Technol. 32, 127–136.
7. Directive 2006/7/EC, 2006, 2006. European bathing water directive. Official Journal of the European Union L64, 37-51.
8. El Samrani, A.G., Lartiges, B.S., Villiéras, F. (2008), Chemical coagulation of combined sewer overflow: heavy metal removal and treatment optimization. Water Res. 42, 951–60. doi:10.1016/j.watres.2007.09.009
9. Falsanisi, D., Gehr, R., Santoro, D., Dell'Erba, A., Notarnicola, M., Liberti, L. (2006), Kinetics of PAA demand and its implications on disinfection of wastewaters. Water Qual. Res. J. Canada 41, 398–409.
10. Gasperi, J., Rocher, V., Gilbert, S., Azimi, S., Chebbo, G. (2010), Occurrence and removal of priority pollutants by lamella clarification and biofiltration. Water Res. 44, 3065–76. doi:10.1016/j.watres.2010.02.035
11. Hrudey, S.E., Charrois, W.J. (2012), Disinfection By-products and human health. IWA Publishing, London United Kingdom.
12. Jolis, D., Ahmad, M.-L. (2004), Evaluation of High-Rate Clarification for Wet-Weather-Only Treatment Facilities. Water Environ. Res. 76, 474–480. doi:10.2175/106143004X151563
13. Kitis, M., 2004. Disinfection of wastewater with peracetic acid: A review. Environ. Int. 30, 47–55. doi:10.1016/S0160-4120(03)00147-8
14. Koivunen, J., Heinonen-Tanski, H. (2005), Peracetic acid (PAA) disinfection of primary, secondary and tertiary treated municipal wastewaters. Water Res. 39, 4445–4453. doi:10.1016/j.watres.2005.08.016
15. Liberti, L., Notarnicola, M. (1999), Advanced treatment and disinfection for municipal wastewater reuse in agriculture. Water Sci. Technol. 40, 235–245. doi:10.1016/S0273-1223(99)00505-3
16. Nurizzo, C., Antonelli, M., Profaizer, M., Romele, L. (2005), By-products in surface and reclaimed water disinfected with various agents. Desalination 176, 241–253. doi:10.1016/j.desal.2004.11.012
17. Svecevicius, G., Šyvokiene, J., Stasiunaite, P., Mickeniene, L., Syvokiene, J., Stasiŭnaite, P. (2005), Acute and chronic toxicity of chlorine dioxide (ClO2) and chlorite (ClO2-) to rainbow trout (Oncorhynchus mykiss). Environ. Sci. Pollut. Res. 12, 302–305. doi:10.1065/espr2005.04.248
18. Tchobanoglous, G., Burton, F.L., Stensel, H.D. (2003), Wastewater Engineering, treatment and Reuse Metcalf & Eddy Inc., 4th ed. McGraw Hill, New York.
19. USEPA (1999), Combined Sewer Overflow Technology Fact Sheet alternative Disinfection Methods. United States Environmental Protection Agency, Washington DC, USA.
20. Watson, K., Shaw, G., Leusch, F.D.L., Knight, N.L. (2012), Chlorine disinfection by-products in wastewater effluent: Bioassay-based assessment of toxicological impact. Water Res. 46, 6069–6083. doi:10.1016/j.watres.2012.08.026
21. White, G.C. (2010), Handbook of chlorination and alternative disinfectants, 5th ed. John Wiley & Sons, Inc., New York.