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A Multifunctional Treatment Process for Combined Sewer Overflow Pollutants and Nutrient Control

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Abstract

Combined sewer overflows (CSOs) contain a highly variable, wide range of contaminants, both in particulate and soluble form, making conventional water treatment processes unable to offer adequate public health protection. Moreover, the disinfection of combined sewer overflow discharges is necessary to reduce the amount of microorganisms discharged into urban waters. To overcome CSO impacts, new and adaptable multifunctional treatment schemes need to be developed. To date, to the best of our knowledge, no study proposed an efficient and cost-competitive treatment able to remove a broad spectrum of CSO pollutants. This research demonstrated that a chemical pre-treatment, followed by micro-sieve filtration and UV disinfection is an efficient and cost-competitive treatment process able to simultaneously remove typical combined sewer overflow pollutants (suspended solids, chemical oxygen depends, turbidity, and fecal bacteria) in conjunction with nutrient (nitrogen and phosphorus). The removal of particulates, as well as dissolved nitrogen and phosphorus, was achieved by first adsorbing soluble pollutants on zeolite and powdered activated carbon, and subsequently applying filtration carried out by polymer-enhanced microsieving. An optimal treatment condition, consisting of 1.1 mg/L of the cationic polymer, 250 mg/L of zeolite, and 5 mg/L of powdered activated carbon, was identified by Pareto analysis. Under this condition, expected performance would be reductions of 72%, 56%, 35%, and 75% for turbidity, total Kjeldahl nitrogen, total chemical oxygen demand, and total phosphorous, respectively. Moreover, the efficiency of UV disinfection with and without chemical pre-treatment was investigated and a microbial inactivation model able to predicts the inactivation of fecal coliform (FC) bacteria was developed. Experimental results reported that 4-log removal of FC was achieved at fluence 10 mJ/cm^2 when the UV disinfection was enhanced by chemical pre-treatment and microsieving filtration using a 32 µm mesh size. Under these conditions, the TSS removal achieved was 73% and the UVT increase of 32%.

The findings presented in this thesis demonstrate the possibility to quickly and effectively treat a large amount of wastewater flow by reducing equipment and operating costs, providing municipalities with viable and low footprint treatment options where the issues of combined sewer overflow and nutrient management are simultaneously tackled.

Summary for Lay Audience

Combined sewer overflows (CSOs) contain a highly variable, wide range of contaminants, making conventional water treatment processes unable to offer adequate public health protection. Treating combined sewer overflow discharges is necessary to reduce the amount of pollutants discharged into rivers, lakes, or seas. To overcome CSO impacts, new and adaptable multifunctional treatment schemes need to be developed. To date, to the best of our knowledge, no study proposed an efficient and cost-competitive treatment able to remove a broad spectrum of CSO pollutants. This research demonstrated that a chemical pre-treatment, followed by micro-sieve filtration and UV disinfection is an efficient and cost-competitive treatment process able to simultaneously remove combined sewer overflow pollutants (i.e. suspended solids, chemical oxygen depends, turbidity, and fecal bacteria) in conjunction with nutrient (nitrogen and phosphorus). The removal of particulates, as well as dissolved nitrogen and phosphorus, was achieved by first adsorbing soluble pollutants on zeolite and powdered activated carbon, and subsequently applying filtration carried out by polymer-enhanced microsieving. An optimal treatment condition, consisting of 1.1 mg/L of the cationic polymer, 250 mg/L of zeolite, and 5 mg/L of powdered activated carbon, was identified by Pareto analysis. Under this condition, expected performance would be reductions of 72%, 56%, 35%, and 75% for turbidity, total Kjeldahl nitrogen, total chemical oxygen demand, and total phosphorous, respectively. Moreover, the efficiency of UV disinfection with and without chemical pre-treatment was investigated and a microbial inactivation model able to predicts the inactivation of fecal coliform (FC) bacteria was developed. Experimental results reported that 4-log removal of FC was achieved at fluence 10 mJ/cm2 when the UV disinfection was enhanced by chemical pre-treatment and microsieving filtration using a 32 µm mesh size. Under these conditions, the TSS removal achieved was 73% and the UVT increase of 32%.

The findings presented in this thesis demonstrate the possibility to quickly and effectively treat a large amount of wastewater flow by reducing equipment and operating costs, providing municipalities with viable and low footprint treatment options where the issues of combined sewer overflow and nutrient management are simultaneously tackled.

Keywords

Coagulation-flocculation; Combined sewer overflow; Micro-sieving; Pareto analysis; Response surface methodology; Wastewater treatment; UV disinfection, inactivation kinetic model, stormwater management model.

Co-Authorship Statement

This Ph.D. thesis contains materials that are published or in preparation for submission in peer-reviewed journals as listed below.

Chapter 3: A microsieve-based filtration process for combined sewer overflow treatment with nutrient control: Modeling and experimental studies.

Tiziana Venditto, Michele Ponzelli, Siva Sarathy, Ajay K. Ray, Domenico Santoro (2020)

Water Research. DOI 10.1016/j.watres.2019.115328

The primary author of this chapter was Tiziana Venditto under the supervision of Dr. Ray and Dr. Santoro. The experimental plan was developed by Venditto with guidance from Dr. Ray and Dr. Santoro while the execution of experiments and data collection was predominantly completed by Venditto and Ponzelli under the supervision of Dr. Santoro and Dr. Ray. Data analysis and drafting of the manuscript were predominantly completed by Venditto under the supervision of Dr. Sarathy, Dr. Santoro, and Dr. Ray. The work was conducted at Trojan Technologies. Feedback on the manuscript was received from the co-authors.

Chapter 4, Chapter 5, and Chapter 6: Low-fluence UV disinfection for combined sewer overflow treatment: environmental impacts and costs comparison

Tiziana Venditto, Kyriakos Manoli, Siva Sarathy, Ajay K. Ray (2020)

In preparation for submission to Science of the Total Environment.

The primary author of this chapter was Tiziana Venditto under the supervision of Dr. Ray and Dr. Sarathy. The experimental plan was developed by Venditto with guidance from Dr.

Ray and Dr. Sarathy while the execution of experiments, data collection and analysis, modeling, cost-analysis, and drafting were predominantly completed by Venditto under the supervision of Dr. Sarathy and Dr. Ray. The work was conducted at Trojan Technologies. Feedback on the manuscript was received from the co-authors.

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List of Abbreviations and Symbols

Avg	Average
BB-DOE	Box Behnken Design of Experiments
BWF	Base Wastewater Flow
BOD_5	Biochemical Oxygen Demand (5-day)
CCD	Central Composite Design
CSOs	Combined Sewer Overflows
CSSs	Combined Sewer Systems
DWF	Dry Weather Flow
E.coli	Escherichia Coli
EPA	Environmental Protection Agency
FC	Fecal Coliform
GWI	Ground Wastewater Infiltration
LTCP	Long-Term Control Plan
MOE	Ministry of the Environment
NMC	Nine Minimum Controls
NPDES	National Pollutant Discharge Elimination System

NTU	Nephelometric Turbidity Units
PAA	Peracetic Acid
PAC	Powdered Activated Carbon
p-COD	particulate Chemical Oxygen Demand
PFA	Performic Acid
POTW	Publicly Owned Treatment Works
PPCP	Pollution Prevent and Control Plant
RDII	Rainfall Derived Infiltration/Inflow
RSM	Response Surface Methodology
s-COD	soluble Chemical Oxygen Demand
SWMM	Stormwater Management Model
t-COD	total Chemical Oxygen Demand
TKN	Total Kjeldahl Nitrogen
TP	Total Phosphorous
TSS	Total Suspended Solids
UVT	Ultra-Violet Transmittance
WWTP	Wastewater Treatment Plant

μm	microns (micrometer)

Chapter 1

1 Introduction

Due to urbanization and population growth, municipalities began to install sewer systems in the middle of 1800, having as a main goal to improve the urban sanitary condition and public health. Today, sewers are a fundamental part of the urban water infrastructure.

Two types of sewer systems are commonly used, such as:

- Combined sewer systems (CSSs), i.e. a sewage collection system characterized by only one pipe designed to collect wastewater and stormwater.
- Separate sewer systems (SSSs), i.e. a sewage collection system characterized by two separate systems of pipes: one for wastewater management and one for stormwater management.

At the end of the 19th century, CSSs appeared to be the most efficient way for the collection and conveyance of stormwater and wastewater (EPA, 2004). However, management of Combined Sewer Overflow (CSO) has become a main concern in the last years because of combined sewer network limitation and/or to the overcoming of maximum Wastewater Treatment Plants (WWTPs) capacity during wet weather periods. Specifically, in the case of CSSs, the additional flow associated with strong rainfall events may lead to situations where sewer capacity is exceeded, and an overflow occurs (Zukovs and Marsalek, 2004). When this happens, water bypasses the WWTP and untreated water is discharged directly into the receiving body (e.g. lakes, rivers, sea). Visible matter, infectious (pathogenic) bacteria and viruses, oxygen-demanding substances, nutrients, toxicants (e.g., heavy metals, pesticides, and petroleum hydrocarbons), and fecal bacteria are discharged into the urban waters (Metcalf & Eddy, 2014). As such, sewage discharges coming from CSSs are considered a source of pathogens, organic and inorganic pollutants, and solids. As a result, several public health risks could be induced by sewage overflow discharges; the Environmental Protection Agency (EPA) has estimated that, per year, between 3.448 and 5.576 illnesses are associated with untreated wastewater discharge (EPA, 2004).

1.1 Rationale

To date, integrated treatment strategies to effectively remediate a wide spectrum of CSO contaminants are limited. In several cities (Maruejouls et al., 2011; Nascimento et al., 1999), the most common approach to reduce pollutant loads from CSO is to develop storage facilities or retention treatment basins to reduce the hydraulic peak flow (Gasperi et al., 2012a; Li et al., 2003). Nevertheless, CSOs cannot be entirely controlled only by reducing the overflow loadings by storage facilities or retention treatment basins but also by developing remedial measures to achieve improvement of water quality in streams (EPA, 2004). Also, since CSOs are caused by wet weather conditions and occur while storm water and other nonpoint source pollutant loads are delivered to surface water, it is hard to directly identify specific CSO pollutant sources. Then, there is a need to effectively treat CSOs by new treatment approaches able to remove different types of pollutants in the same process.

This research aims to develop an integrated treatment process able to simultaneously remove solids, organic matters, nutrients, and microorganism loads and to assess its performance at the urban scale.

Following are the questions which were attempted in this study:

- To what extent an improvement of water quality attainable via technology deployment at catchment scale would translate into environmental and public health benefits?
- What is the return of investment for a municipality that has invested in CSO treatment?

This study could likely evolve into a supporting tool for municipalities that need to invest in CSO treatment strategies and management.

1.2 Thesis Organization

This Ph.D. thesis is written in the article-integrated format specified by the School of Graduate and Postdoctoral Studies of The University of Western Ontario. The contents of the six chapters included in this thesis are presented below.

Chapter 1 provides a brief introduction related to the background and motivation for developing combined sewer overflow treatment.

Chapter 2 provides a comprehensive literature review of different treatment techniques used to treat combined sewer overflow. The review also highlights the advantages and disadvantages of existing combined sewer overflow treatments. The research objectives of the thesis are also included in Chapter 2.

Chapter 3 is a research article entitled *"A microsieve-based filtration process for combined sewer overflow treatment with nutrient control: Modeling and experimental studies"*. The objective of this work was to assess the effects of the various treatment variables, i.e. cationic polymer, zeolite, powder activated carbon, and microsieve size, on the removal of typical combined sewer overflow pollutants (suspended solids, chemical oxygen depends, turbidity) in conjunction with nutrient (nitrogen and phosphorus). Additionally, an optimization study was carried out to identify the optimal process conditions.

Chapter 4, Chapter 5, and Chapter 6 are part of a research article entitled *"Low-fluence UV disinfection for combined sewer overflow treatment"*. In Chapter 4, the treatment developed in chapter 3 was implemented by adding the UV disinfection process as the final step of the treatment train. In this study, the efficiency of UV disinfection with and without pre-treatment was assessed on the removal of TSS, UVT, and fecal coliform inactivation. The inactivation kinetic model was to estimate the six kinetic parameters controlling the Fecal coliform inactivation process. In Chapter 5, the CSO treatment developed was compared with existing CSO treatments reported in the literature. In Chapter 6, the developed CSO treatment was simulated by using the stormwater management model (SWMM) to assess, at the urban scale, the performance of the treatment in restoring the water balance of the receiving water. Cavendish area in London (ON) was used as a study area.

Chapter 7 includes the main conclusions of the thesis along with study limitations and recommendations for future work.

1.3 References

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Chapter 2

2 Literature Review

2.1 Combined sewer overflows (CSOs)

A combined sewer system (CSS) collects rainwater runoff, domestic sewage, and industrial wastewater in a single pipe (Diaz-Fierros et al., 2002; EPA, 1994; Scherrenberg, 2006). Under normal conditions, CSS transports wastewater to a wastewater treatment plant (WWTP) for treatment. During a heavy rainfall events, the volume of wastewater into the sewer network can increase to five to ten times (Field, 1990). Under these conditions, the capacity of a sewer system can be reached, and an overflow occurs (Madoux-Humery et al., 2016). When this occurs, untreated stormwater and wastewater are discharged directly to nearby streams, rivers, and other water bodies.

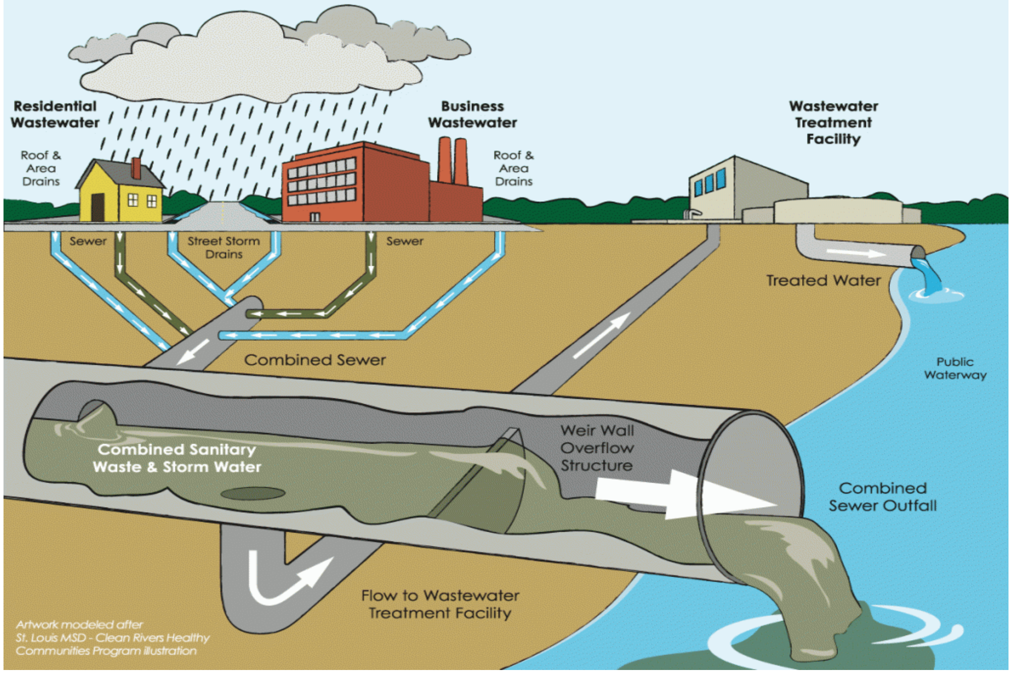

Figure 2-1: Combined sewer overflow. Image courtesy of www.CivicGardenCenter.org

2.2 Impacts of CSOs

2.2.1 Physical-chemical and Microbial CSO characteristics

Combined sewer overflows (CSOs) carries a mixture of stormwater, untreated human and industrial waste, toxic materials, and debris. When an overflow occurs, visible matter, infectious (pathogenic) bacteria, and viruses, oxygen-demanding substances, nutrients, toxicants (e.g., heavy metals, pesticides, and petroleum hydrocarbons), and fecal bacteria are discharged into the urban waters (Becouze-Lareure et al., 2016; EPA, 1994; Gasperi et al., 2008; Iannuzzi et al., 1997; Launay et al., 2016; Weyrauch et al., 2010). The EPA Report to Congress on the Impacts and Control of CSOs and SSOs (EPA, 2004) reported that pollutants from CSOs can potentially impact four designated water uses, i.e. aquatic life support, drinking water supply, fish consumption, and recreational water.

Total Suspended Solids (TSS), Chemical oxygen demand (COD), 5-day Biochemical Oxygen Demand (BOD_5), Ammonium (NH_4), total Phosphorus (P_{tot}), and fecal bacteria (*E.coli* and Enterococci) concentrations were considered as basic parameters of CSOs test to assess the overflow impacts on the receiving water (Gasperi et al., 2008; Hajj-Mohamad et al., 2014; Launay et al., 2016; Servais et al., 1999). Sewer deposit resuspension, mainly in a particulate form (>70%), is the primary contributor for TSS, COD, and BOD_5. Eroded particles are highly organic and biodegradable with TSS values around 50-80% (Ahyerre et al., 2000; Gasperi et al., 2010, 2008; Gupta and Saul, 1996; Madoux-Humery et al., 2013; Riechel et al., 2016; Soonthornnonda and Christensen, 2008). Ammonium occurs in higher concentrations because it mainly comes from urine and feces in the wastewater portion of CSO, as well as from residues of food production or slaughterhouse effluents (Degree, 2013; Tondera et al., 2018). Contamination from fecal bacteria occurs along with solid matter (attached) and wastewater (Jalliffier-Verne et al., 2016). CSOs showed to increase fecal bacteria in the receiving waters by four orders-of-magnitude (Smith and Perdek, 2004) and it remains high during the whole rainfall event (Pongmala et al., 2015; Sztruhár et al., 2002).

2.2.2 Factors affecting the CSOs discharges

In-sewer flow is characterized by Dry Weather Flow (DWF) and Wet Weather Flow (WWF). DWF includes Base Wastewater Flow (BWF) and Ground Wastewater Infiltration (GWI). BWF is the residential, commercial, institutional, and industrial flow, collected from the sanitary sewer system and treated to the wastewater treatment plant (WWTP). GWI is the groundwater infiltration that enters the collection system through cracked pipes or deteriorated manholes when the ground surface is extremely saturated (EPA, 2017). Rainfall Derived Infiltration/Inflow (RDII) is added to GWI and BWF during WWF. Rainfall inflow refers to the water that enters the sanitary sewer system through direct connections (e.g., roof and stormwater cross-connection); rainfall infiltration refers to the water that filters through the soil before entering the sanitary sewer system through damaged pipe sections, deteriorated manholes, or connected foundation drain. RDII is the major component of peak wastewater flows during wet weather and it is typically responsible for overflows (Muleta and Boulos, 2008). CSO impact management requires a comprehensive knowledge of natural and anthropogenic factors: rainfall variability, land use, implementation of human infrastructures, different agricultural practices, and agroforestry species or deforestation (Im et al., 2009; Nasrin et al., 2013). Each one of these factors can affect the CSO discharge resulting in different CSO impacts in terms of pollutant loads and flowrate, which are ultimately dependent on time-variability and spatial-variability. Several studies (Chebbo et al., 2001; Gasperi et al., 2010; Gromaire et al., 2001) showed that the peak concentration of pollutants is observed during the first 30 minutes of overflow. During this time, solid matter concentration is higher because of in-sewer phenomenon processes, i.e. resuspension/erosion of the sewer deposits. The spatial-variability impacts are associated with the urban growth, and the consequent rapidly increasing of impervious areas. Impervious areas lead to an increase of hardly identified non-point sources that carry various pollutants from urban runoff into the sewer network (Acharyaa et al., 2010; Kim et al., 2014; Tsihrintzis and Hamid, 1998). Kim et al. investigated CSO pollutant loads concerning different land-uses (Kim et al., 2014). The results of this analysis showed that runoff characteristics of non-point pollutants are different and site-specific; therefore, each CSO treatment should meet specific watershed characteristics. Additionally, considering the overflow impact variability, an effective water quality investigation is necessary to characterize the CSO discharges in terms of pollutant loads.

2.3 Regulatory contest for CSOs

Under the Clean Water Act's National Pollutant Discharge Elimination System (NPDES), The U.S. Environmental Protection Agency (EPA) regulates discharges of pollutants from municipal and industrial collection systems. All municipalities and facilities must obtain a permit to discharge a pollutant into waterways. Without a permit, the discharge is considered unlawful.

To address CSOs under the NPDES permitting program, EPA developed a CSO control policy. The policy contains the following fundamental principles to ensure that CSO controls are cost-effective and meet local environmental objectives (EPA, 1994):

- Clear levels of control to meet health and environmental objectives.
- Flexibility to consider the site-specific nature of CSOs and find the most cost-effective way to control them.
- Phased implementation of CSO controls to accommodate a community's financial capability.
- Review and revision of water quality standards during the development of CSO control plans to reflect the site-specific wet weather impacts of CSOs.

In order to facilitate the implementation of CSO control policy, EPA published nine minimum controls (NMC) (EPA, 2004). The NMCs are as follows:

1. Proper operation and regular maintenance programs for the sewer system and CSO outfalls
2. Maximum use of the collection system for storage
3. Review and modification of pretreatment requirements to ensure that CSO impacts are minimized
4. Maximization of flow to the POTW for treatment
5. Elimination of CSOs during dry weather
6. Control of solid and floatable materials in CSOs
7. Pollution prevention programs to reduce containments in CSOs
8. Public notification to ensure that the public receives adequate notification of CSO occurrences and CSO impacts
9. Monitoring to effectively characterize CSO impacts and the efficacy of CSO controls.

In addition to implementing the NMC, municipalities are required to develop a Long-Term Control Plan (LTCP) which should include:

- Characterization, monitoring, and modeling of the CSS
- Public participation
- Consideration of sensitive areas
- Evaluation of alternatives
- Cost/performance considerations
- Operational plan
- Maximization of treatment at the POTW treatment plant
- Implementation schedule
- Post-construction compliance monitoring

In Canada, the Ontario Ministry of Environment (MOE) published specific guidelines about CSO control requirements, the most relevant being the Procedures F-5-5. The main goals of the guidelines are 1) to eliminate the occurrence of dry weather overflows; 2) to minimize the impacts on human health, environment, and aquatic life resulting from CSOs; and 3) to achieve as a minimum compliance with body contact recreational water quality objectives. Moreover, the Procedure F-5-5 requires a minimum of 50 % of total suspended solids (TSS) removal and 30 % of five-day Biochemical Oxygen Demand (BOD_5) removal prior to discharge. To meet the Procedure F-5-5 goals for CSO control, municipalities are required to adopt a Pollution Prevention and Control Plan (PPCP) in order to define the nature and cause of pollution problems and establishes remedial measures to overcome them.

2.4 Best available processes and technologies

Based on the literature, the main pollutants found in CSO are classified according to their effects on the following categories of contaminant loads: (a) oxygen demand (BOD and COD), (b) nutrients (N and P), (c) toxic substances (NH_3, heavy metals, microcontaminants), (d) hygiene (fecal bacteria), and (e) physical factors (suspended solids) (Diaz-Fierros et al., 2002; Gasperi et al., 2008; Hajj-Mohamad et al., 2014; Launay et al., 2016; Servais et al., 1999). However, most pollutants have a strong affinity for suspended solids (Ahyerre et al., 2000), therefore, the removal of particle matters will very often remove many of the other pollutants found in urban stormwater (Li et al., 2003). In the last decade, different treatment practices, i.e. primary treatment, chemically enhanced primary treatment, advanced treatment, and disinfection treatment have been considered and investigated to reduce the CSO impacts. Gasperi et al. tested a coagulation/flocculation/clarification processes to treat CSO water achieving about 80% of TSS removal, between 40-70% of BOD_5, but no removal for ammonium (Gasperi et al., 2012a). Averill et al. compared the efficiency of solid/liquid separation process, with and without chemical coagulants, followed by UV disinfection (Averill et al., 1997). The treatment involving chemical coagulants showed a better TSS removal (50% of TSS removal) which allowed to obtain a fecal coliform concentration of fewer than 100 counts per 100 mL by UV disinfection. However, to date, no study proposed an efficient and cost-competitive treatment able to simultaneously remove solids, organic matters, nutrients, and microorganism loads.

2.4.1 Primary treatment techniques

Solids and organic matters are removed by primary treatment (Metcalf & Eddy, 2014). Particulate matters are removed by physical treatments involving solid-liquid separation and the typical physical treatment removal efficiency is around 70% for suspended solids (Bridoux et al., 1998; Delporte et al., 1995; Plum et al., 1998). The main primary treatment techniques applied for CSO treatment are 1) wetlands, 2) storage tanks, 3) settling tanks, and 4) coarse screen.

2.4.1.1 Wetlands

Wetlands are engineered systems that use natural processes involving wetland vegetation, soils, and organism to treat wastewater (Kadlec and Wallace, 2008; Scherrenberg, 2006). For CSO treatment, the general and most used wetlands configuration is characterized by vertical flow soil filters with a detention basin on top of the filter layer. A throttle in the outlet structure is installed to control filtration rate and detention time and the filter is completely drained and emptied after every CSO event to guarantee aeration for aerobic degradation within the filter layer (Uhl and Dittmer, 2005). Typical treatment operational performance for solids and organic matter removal is 85–99% (Masi et al., 2017; Scholz and Xu, 2002; Tao et al., 2014; Uhl and Dittmer, 2005).

The main advantages of using wetlands treatment are related to its cost/efficiency and its low maintenance (Tao et al., 2014). On the other hand, treatment of CSO using wetlands carry numerous disadvantages, for example, it requires a large area, cannot support long dry weather periods, and it is not able to remove soluble organic matters (Scherrenberg, 2006).

2.4.1.2 Storage and settling tanks

Storage tanks are built to provide extra storage in the sewer system (EPA, 1999a). Flow is stored during the rainfall event and it is emptied at the end of the event by pumping water into the sewer system (Scherrenberg, 2006). Storage tanks are mainly used as quantity control in order to reduce the flow into the sewer system (EPA, 2007a). Settling tanks are used to remove solids by settling, usually enhanced by coagulation and flocculation process (De Cock et al., 1999). Typical TSS and BOD_5 removal are between 50-70% and 25-40%, respectively (Metcalf & Eddy, 2003).

Storage and settling tanks can survive for a long period without feed water and have low maintenance. However, CSO is usually characterized by a large amount of flow discharged which usually requires bigger tanks to avoid tanks overflow. Large tanks require a large area to be installed which leads to high construction and material costs.

2.4.1.3 Coarse screens

Coarse screens are bares located at the overflow pipe to prevent solids from entering the overflow pipe (EPA, 2007a, 1993). Coarse screens can get clogged due to large amount of solids and floating material transported during the overflow. For this reason, bares need to be cleaned after each overflow resulting in high maintenance costs (Scherrenberg, 2006).

2.4.2 Chemically enhanced primary treatment

In chemically enhanced primary treatment, pollutants are removed by primary treatment techniques enhanced by the use of coagulants and flocculants. Common coagulants used in the chemically enhanced primary treatment for CSO include ferric chloride, alum, and polymers (Chhetri et al., 2016; El Samrani et al., 2008; He et al., 2016; Li et al., 2004;

Zahrim et al., 2011). By chemically enhanced primary treatment, the removal efficiency achieved is ranged between 80-90% for suspended solids, and 35-75% for COD removal depending on the water quality and type of coagulants and/or flocculants used (Bourke, 2000; Bridoux et al., 1998; Delporte et al., 1995; Plum et al., 1998; Shewa and Dagnew, 2020). Despite the high pollutant removal achievable, chemically enhanced primary treatment has its disadvantages, for example, the high amount of coagulant doses needed, and the large quantity of sludge produced.

2.4.3 Advanced treatment techniques

Advanced treatments are used to enhance the removal of suspended solids and organic matter when suspended solids and organic matter are not effectively removed by primary treatment techniques (Metcalf & Eddy, 2014). Most of the time, these treatment techniques need an additional process, the adsorption, to remove nutrients and micro-pollutants (Liao et al., 2015; Scherrenberg, 2006). For CSO treatment, the main advanced treatment techniques identified are 1) Hydrodynamic vortex separation, 2) lamella clarification, 3) Chemically enhanced high rate sedimentation, and 4) membrane filtrations.

2.4.3.1 Hydrodynamic vortex separation

The hydrodynamic vortex separation systems are self-inducing high rate rotary flow devices designed for the removal of solid materials. The flow containing solids enters in tangential direction into a cylindrical vessel; the velocity of water moves the solids towards the vortex creating a swirling motion and they will settle down by gravity. The base of the vortex separator is characterized by a slope to sweep the solids in a central drain (Andoh et al., 2002; Faram et al., 2004; Scherrenberg, 2006). It is impossible to estimates the efficiency of the hydrodynamic vortex separation since it depends on several variables such as, rainfall characteristics, the particle size of solids, and settling velocities (Andoh and Saul, 2003; Boner et al., 1995).

2.4.3.2 Lamella clarification

Lamella clarification systems are advanced settling apparatus with storage usually enhanced by the use of coagulant to increase particle removal. By lamella settler, solids will settle on the lamella plate and will fall forming sludge. Sludge will be collected into the sludge hopper and subsequently pumped out (Scherrenberg, 2006). The main advantage of lamella clarification is the system footprint; indeed, lamella clarification requires only one-third of the area used by storage and settling tanks (Fuchs et al., 2014; Takayanagi et al., 1997). Previous investigations on the lamella clarification efficiency reported a suspended solids removal ranged between 50%-90% (Daligault et al., 1999).

2.4.3.3 Chemically enhanced high rate sedimentation

Chemically enhanced high rate sedimentation is a Physico-chemical treatment where coagulation/flocculation process is employed to form dense flocs with high settling velocity (Scherrenberg, 2006). Actiflo and DensaDeg are the two commercial technologies used for CSO treatment. Actiflo is a compact process that operates with microsand. It combines chemical precipitation and lamella settling involving weighted settling. A coagulant is injected to untreated CSO water before entering the coagulation tank. In the coagulation tank, solids are destabilized and moved into the injection tank where polymer and microsand are added allowing the formation of large and heavy flocs. Because of their weight, the flocs can be easily removed by sedimentation. After this stage the water enters the settling zone with lamella while microsand containing sludge is treated with a hydrocyclone; this step will allow microsand to separate from sludge and the microsand will be reintroduced into the injection tank (EPA, 2003; Plum et al., 1998; Scherrenberg, 2006). The typical removal efficiency of Actiflo system was ranged between 70-95%, 60-85%, and 50-75% for TSS, COD, and BOD_5, respectively (EPA, 2003; Jolis and Ahmad, 2004)

DensaDeg is a three-components system characterized by coagulation, flocculation, and clarification unit (EPA, 2003). The CSO water enters the first tank where grit removal takes place, water is aerated, and a coagulant is injected. Then, water will flow to the second tank. In the second tank, a flocculant and sludge from the clarifier tank are added and the flocculation process is promoted by turbines installed in the chamber for mixing. This process will increase contact between the solids and recycled sludge forming denser flocs. The flocculated stream enters the third chamber, the clarifier. Here, solids will settle, and sludge is thickened and recirculated into the flocculation unit tank (EPA, 2003; Scherrenberg, 2006). Typical removal efficiency of DensaDeg system was ranged between 80-90%, 45-60% and 40-63% for TSS, COD, and BOD_5, respectively (EPA, 2003; Frank et al., 2006)

Both Actiflo and DensaDeg have a start-up time ranged between 10 and 30 minutes. Despite the high pollutant removal achievable, the long start-up time makes Actiflo and DensaDeg unsuitable to treat the first flush of an overflow (EPA, 2003; Jolis and Ahmad, 2004).

2.4.4 Disinfection treatment techniques

Disinfection is required to reduce the amount of bacteria and pathogenic microorganisms, which can be dangerous for public health (Metcalf & Eddy, 2014). CSOs are recognized as a primary source of fecal bacteria. The main disinfection treatment techniques for CSO treatment reported by the literature are 1) chlorination, 2) ozonation, 3) peracetic and performic acid and 4) UV irradiation.

2.4.4.1 Chlorination

Chlorine is the most used disinfectant techniques. In CSO water with a small amount of solids, a low amount of chlorine is enough to achieve a high level of pathogen inactivation (EPA, 1999b). However, because of the high flowrates and the high solids contents associated with a CSO discharge, an effective CSO disinfection by chlorine requires high chlorine doses resulting in a high level of toxicity by-products (EPA, 1999b; Watson et al., 2012; Wojtenko et al., 2002).

2.4.4.2 Ozonation

With Ozonation, we are referring to the inactivation of pathogenic through the infusion of ozone. Ozone is one of the most powerful oxidizers able to inactivate bacteria, viruses, and organic material (EPA, 1999c; Shammas and Wang, 2005; Tondera et al., 2015; Xu et al., 2002). One of the main disadvantages related to the ozonation is that, due to reactions of ozone with organic and inorganic compounds and suspended solids, wastewater requires a high dosage of Ozone. Moreover, due to the quantity and quality flow variability during CSO events, the dosage of ozone needs to change simultaneously with the water quality; this will increase the complexity and costs of the disinfection process (Gehr et al., 2003).

2.4.4.3 Peracetic and Performic acid

Peracetic and Performic acid are emerging chemical disinfectants that have demonstrated the potential to inactivate microorganisms (Maffettone et al., 2020; Manoli et al., 2019).

Peracetic acid is produced by a combination of water, acetic acid, and hydrogen peroxide. Sulfuric acid is added as a catalyst to increasing the chemical reaction rate (Coyle et al., 2014). Performic acid is an organic chemical that belongs to the family of aliphatic peracids (Swern, 1949). Chhetri et al. reported that peracetic acid can be used to treat CSO discharge only where treatment facilities have a long retention contact time, while disinfection by performic acid is more efficient at low fluences and can be used where the overflow structures have a short retention time (Chhetri et al., 2014). The aforementioned limitations make peracetic acid and performic acid unsuitable to treat CSO discharges since the ideal CSO disinfectant has to be adaptable to the rapid change of quality and quantity of CSO discharges during CSO events.

2.4.4.4 UV irradiation

UV irradiation is a physical process able to neutralizes microorganisms by ultraviolet lamps submerged in the effluent (EPA, 1999d; Gibson et al., 2017). The process adds nothing to the water but UV light, and therefore, has no impact on the chemical composition of the water (Averill et al., 1997). Wojtenko et al. highlighted that UV disinfection is an effective alternative to chlorination for CSO treatments (Wojtenko et al., 2001). Gehr et al. emphasized that UV disinfection is the most suitable treatment process for UV disinfection against higher costs related to the peracetic acid and higher dosage of ozone (Gehr et al., 2003). Additionally, UV disinfection does not need large and expensive contact tanks due to short contact time. Anyway, pre-treatment of CSO water plays a fundamental role in disinfection efficiency since the presence of solids is a concern for several CSO technologies (Boner et al., 1995; Jolis et al., 2001; Madge and Jensen, 1999; Wojtenko et al., 2001).

2.5 Synopsis of the literature

CSOs are very frequent events in CSSs. In 2004, EPA released an official position to Congress reporting on health and environmental impacts caused by CSOs (EPA, 2004). In this document, it has been estimated that each year, in the U.S.A., 850 billion gallons of untreated water is discharged, impacting aquatic life, drinking water, fish consumption, shellfish harvesting, and water recreation activities, as well as causing diseases, i.e. gastroenteritis, dysentery, cholera, and hepatitis. For this reason, CSO is today one of the major sources of environmental pollution that cause severe damage to human health. To develop effective stormwater management strategies, municipalities need to have a detailed understanding of CSO characterization both qualitatively and quantitatively. That said, the impacts of wet weather events on the performance and reliability of combined sewer systems for flood control, pollutant loads, and environmental protection are extremely challenging, despite this being an essential and inevitable task. Integrated treatment strategies to effectively remediate a wide spectrum of CSO contaminants are limited and not well researched. Nowadays, existing CSO treatments are mainly focused on the removal of conventional pollutants such as total suspended solids and oxygen-depleting substances. Treatment targets are also set to a minimum level required, as in Procedure F-5-5 (50% of TSS and 30% of BOD) with no efficiency regarding the removal of soluble oxygen matters. Such removal objectives are likely to be insufficient; an optimal and effective CSO treatment should be aimed to achieve multiple treatment goals, thus maximizing the removal of suspended solids, oxygen demand substances, nutrients, and fecal bacteria. To achieve such a goal, advanced treatment techniques involving solid-liquid separation and adsorption process seem to be the best option. The UV disinfection appears to be the best option for fecal bacteria removal due to the fast start-up time and small footprint. However, pre-treatment with high particle removal is needed to improve the UV disinfection process. Additionally, the reliability of CSO treatment processes and technologies, as well as their cost-effectiveness and value-added features, have not been considered during their development; all these factors must be considered since locations associated with CSO events are typically not easily accessible and space-constrained. Therefore, to ensure high results, integration of large scale (urban scale) and small scale (sewer network) is necessary while developing a CSO treatment.

2.6 Discussion

Based on the literature review, the following observation could be made to guide the development of a novel treatment process and assess its efficacy at catchment scale:

1. an optimal and effective CSO treatment should be aimed at maximizing particle removal, thus having a high solids removal efficiency as well as sufficient oxygen demand and nutrients removal.
2. a physical-chemical pre-treatment is likely required to enhance the subsequent disinfection process; however, the minimization of chemicals used in CSO treatment is a highly desirable goal for public health protection.
3. a disinfection process is required to remove fecal contamination from CSO water discharges.
4. a good characterization of CSO discharge is necessary to size the treatment process as the water quality characteristics of CSOs are highly site-specific dependent.

5. a complete understanding of the CSO impacts at the urban scale is required; specifically, a stormwater management model should be employed to allow the identification of site-specific dynamics associated with CSO events.

2.7 Research Objectives

This research was conducted to develop a novel, multifunctional CSO treatment process able to cost-effectively achieve multiple treatment objectives to simultaneously remove visible matter, oxygen-demanding substances, nutrients, and fecal bacteria. To achieve such an ambitious goal, six main objectives have been identified and outlined as follows:

I. To assess and validate the effectiveness of coagulation/flocculation process using polymer as coagulant on the removal of visible matter, and oxygen-demanding substances.

II. To investigate the effectiveness of zeolite and power activated carbon on the removal of nutrients and soluble oxygen-demanding substances.

III. To investigate the effectiveness of UV disinfection on the removal of fecal bacteria and its interaction with coagulation/flocculation and adsorption processes.

IV. To develop regression equations able to quantitively describe the synergies between the aforementioned treatment processes and the simultaneous removal of visible matter, oxygen-demanding substances, nutrients, and fecal bacteria.

V. To develop model-based simulations at the urban scale with the intent to assess the sewer network response during wet weather events and to assess the environmental impacts of the proposed treatment.

VI. To determine and compare the proposed CSO treatment costs with existing CSO treatment strategies.

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Chapter 3

3 A Microsieve-Based Filtration Process for Combined Sewer Overflow Treatment with Nutrient Control: Modeling and Experimental Studies

3.1 Introduction

Pollution from urban stormwater discharges and combined sewer overflows (CSOs) are reported as one of the main factors affecting the water quality of receiving bodies (Anne-Sophie et al., 2015; Bryan Ellis and Yu, 1995; Eganhouse and Sherblom, 2001; Gasperi et al., 2008; Passerat et al., 2011a; Riechel et al., 2016; USEPA, 2004). Consequently, developing strategies for CSO management has become central in the environmental agenda of municipalities around the world, exacerbated by the limitations of combined sewer system (CSS) infrastructure and/or the limited capacity of municipal wastewater treatment plants (WWTPs). Furthermore, the additional flow generated by extreme wet-weather events could lead to a bypass of wastewater treatment plants (WWTPs) and untreated wastewater being discharged directly into the environment. As a result, oxygen-depleting matter and pathogens, are discharged into the environment together with solids, nutrients, and other micropollutants including heavy metals and chemicals of emerging concern (USEPA, 2004).

For the last two decades, stormwater management strategies have been centered around mitigating the CSO impacts by reducing runoff volume or peak flow by employing storage facilities and retention treatment basins (Li et al., 2004). However, since locations associated with CSO discharges are typically not easily accessible and often space-limited, the design, operation and management of these facilities may be complex. As a result, there is a need to develop new, space-efficient treatment schemes able to remove a broad spectrum of pollutants in a single and multifunctional train (Gasperi et al., 2010, 2008; Iannuzzi et al., 1997; Launay et al., 2016; Soonthornnonda and Christensen, 2008). Passerat et al. (Passerat et al., 2011b) highlighted that sewer sediments were estimated to contribute to about 75% of the solid matter, 10-70% of the E. coli (about 77% attached to the solid matters), and 40-80% of the intestinal enterococci that were discharged by overflows. Therefore, effective removal of particulate matters from CSO water could immediately lead to improve the performance of disinfection processes by which the inactivation of microorganism occurs (Chhetri et al., 2014; Gehr et al., 2003; Kitis, 2004; Wojtenko et al., 2001). To date, a number of studies (Bridoux et al., 1998; Delporte et al., 1995; Ebeling et al., 2003; El-Gendy et al., 2008; Gasperi et al., 2012a; Plum et al., 1998) have examined the performance of physico-chemical treatment, such as coagulation-flocculation, on the removal of particulate matter by using polymer as primary coagulant, a process entailing the neutralization of negative charge and allows small particles to react with the polymer to form insoluble precipitates before flocculation of the solids commences (Bolto et al., 2001; Scherrenberg, 2006). To achieve advanced nitrogen control, the removal of particulate nitrogen is not sufficient as ammonium is mostly present in dissolved form. A review paper on the application of zeolite for wastewater treatment (Wang and Peng, 2010) reported that natural zeolite is a promising technique for the removal of ammonium due to the low costs and its physico-chemical proprieties such as the high cation exchange and sorption capacity (Liao et al., 2015). However, its adsorption efficiency may be reduced by the presence of organic matters. On the other hand, powdered activated carbon (PAC) is the most widely applied adsorption material for the removal of dissolved organics (Gai and Kim, 2008; Ma et al., 2013; Scherrenberg, 2006; Seo et al., 1997). Moreover, a synergistic effect of zeolite and activated carbon on the removal of nutrients and organic contaminants has been reported by Malekmohammadi et al. and Liao et al. (Liao et al., 2015; Malekmohammadi, 2016). In these works, the authors stated that a mixture of carbon and zeolite increases the adsorption efficiency against nutrients and organic pollutants while neither could remove the pollutants if used alone. Table 3.1 summarizes information available in the literature on the effectiveness of treatment by cationic polymer, PAC, zeolite, and microsieving process, when taken individually, to remove specific CSO pollutants.

The goal of this study was to develop and assess the performance of an integrated treatment process starting from an idea of possible treatment technologies to deploy at the urban scale (Fig.3-1).

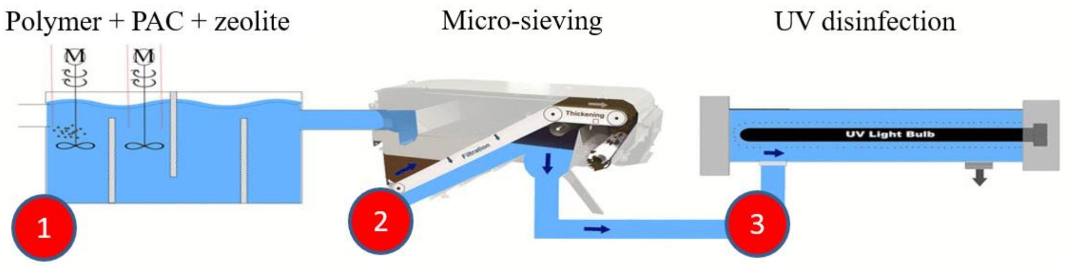

Figure 3-1: Multifunctional CSO treatment process

At this stage, only the first two steps of the treatment represented in Fig. 3-1 will be tested, while further studies on the UV disinfection process will be presented in Chapter 4. The proposed treatment has to be able to simultaneously remove nutrients and CSO pollutants in a multifunctional reactor. The main advantage of using a multifunctional reactor is to carry multiple functions at the same time (i.e. coagulation, adsorption, and filtration) and in a single unit. This new approach opens the possibility for municipalities to address CSO and nutrient pollution with a single capital upgrade. More specifically, the proposed integrated treatment process relies on multiple treatment agents combined in a single multifunctional process where fine particles, such as zeolite and powdered activated carbon, first adsorb soluble nutrient and are subsequently removed by polymer-enhanced microsieving allowing the removal of both soluble and particulate pollutants in a single treatment step. Furthermore, this work describes an innovative method for the removal of ammonia via a dual mechanism of ammonia capture by zeolite absorption, followed by zeolites removal by polymer coagulation and microsieving filtration. By polymer coagulation, smaller particles of zeolite were incorporated into bigger particles and easily removed by microsieving

Table 3-1: Individual and combined effects of polymer, PAC, zeolite and microsieving on CSO pollutants removal

Treatment agents	**Pollutants**					**References**
	Turbidity	*Particulate COD*	*Soluble COD*	*TKN*	*TP*	
Polymer	X	X				(Bolto et al., 2001) (Liao et al., 2015)
PAC		X	X			(Gai and Kim, 2008) (Scherrenberg, 2006) (Ma et al., 2013) (Seo et al., 1997)
Zeolite				X	X	(Wang and Peng, 2010)

						(Liao et al., 2015)
Microsieving	X	X	X			(Evren Ersahin et al., 2012) (Scherrenberg, 2006)
Integrated Treatment	X	X	X	X	X	This study

.

In order to do so, we first investigated the efficiency of the individual treatment agents, and then explored the synergies achieved when the chemicals were dosed simultaneously in an integrated treatment process targeting the removal of particulate CSO pollutants (such as turbidity and chemical oxygen demand) as well as dissolved nutrients (ammonium and nitrogen).

3.2 Materials and Methods

3.2.1 Source of wastewater and analytical measurements

Primary influent (PI) was used as a surrogate to establish CSO treatment efficiency during bench-scale experiments. Samples were collected manually from the Greenway WWTP, located in London, Ontario, Canada. The city of London is characterized by approximately 2,750 kilometers (km) of the sanitary, storm, and combined sewers. Greenway WWTP is one of the six wastewater treatment plants with a combined rated capacity of 152 million liters per day and an average daily flow of 117 million liters per day in 2018 (City of London Corporation, 2016). The plant receives wastewater from approximately 9,100 ha and services a population of 180,000 equivalent inhabitants with a combination of industrial wastewater, residential sewage, including the combined sewers from the older parts of the City. For each sample, the level of turbidity was measured by a HACH 2100 turbidimeter following the Nephelometric method (Standard Method 2130B). Turbidity was used as an indicator of removal efficiency for particulate matter since a linear correlation between turbidity and TSS exists (Hannouche et al., 2012; Ru et al., 2013).

COD tests were carried out by Standard Method 5220-D. The soluble COD (s-COD) was measured by filtering samples through a 0.45 μm pore size filters and the particulate COD (p-COD) measurement was obtained by subtracting s-COD from the total COD (t-COD). TKN was used to quantify the amount of nitrogen contained in organic form and it was determined by digestion and distillation (Standard Method 4500-N_{org}C). TP was measured following Standard Method 4500-P.

3.2.2 Chemically-enhanced pre-treatment

The coagulation-flocculation process was performed on the collected samples using 1 L of raw wastewater. Experiments were carried out using the jar test method in 1-L beakers where polymer, PAC, and zeolite were mixed simultaneously. Natural zeolite NV-Na (surface area 40m^2/g, pore volume 15%, particle size of 0.42 mm; bulk density: 45-50 lbs ft^{-3}) used in this study was obtained from St. Cloud Mining Company, Winston, New Mexico. Zeolite nv-na was selected from previous studies where different type of zeolite with different surface area were compared. Among all the zeolite tested, zeolite NV-Na provided the best results in terms of nutrients removal. PAC (grain size of 10-220 μm; total surface area: 650 m^2 g^{-1}; bulk density: 0,51 g ml^{-1}) was purchased from Cabot Norit Americas Company, Marshall, USA. Cationic Acrylamide polymer (PG-906) was used as coagulant with 10% mole charge purchased from ChemTreat Company, Virginia, USA.

The jar test employed the following steps: (1) rapid mix at a constant speed of 200 rpm for 1 min to maximize the destabilization of colloidal particles and initiate coagulation, (2) slow mix at a constant speed of 20 rpm for 2 min to increase the number of contact events among treating agents and particles, and to facilitate the development of large flocs and (3) the last step was the settling stage. After the coagulation-flocculation process, water was filtered through meshes of three different pore sizes: 158, 350, and 500 µm. Table 3.2 summarizes the employed ranges of mesh size, the dosage of polymer, PAC and zeolite. At the end of each treatment, turbidity, s-COD, p-COD, TKN, and TP were analyzed. Results were compared with the concentration of pollutants in the collected samples to assess the treatment efficiency in terms of percentage removal.

Table 3-2: List of independent variables and the levels tested

Independent Variable	Symbol	Coded variable level		
		Low	Center	High
		-1	0	+1
Polymer (mg/L)	x_1	1	2	3
Zeolite (mg/L)	x_2	0	2500	5000
PAC (mg/L)	x_3	0	250	500
Mesh (µm)	x_4	158	350	500

3.2.3 Design of experiments and response surface analysis

The Box-Behnken (BB-DOE) scheme with four-factor, and three-levels for each factor, was selected as experimental design for this study. The BB-DOE is an independent quadratic design in which the combinations of experimental plans are located at the midpoints of edges and at the center of the process space. The number of experimental points (N) is defined by the expression N = 2k (k-1) + C_0, where k is the number of factors and C_0 is the number of center points (Ferreira et al., 2007). As reported by Zolgharnein et al. (2013), the BB-DOE requires fewer combinations of the independent variables (i.e., the treatment agents) to estimate a potentially complex response function when compared with the central composite design (CCD). This is in line with the findings by Ferreira et al. (2007) who demonstrated that the BB-DOE is an adequate scheme for response surface modelling (RSM), and subsequent optimization studies, in case of non-linear relationships among independent and dependent variables. As a matter of fact, based on previous studies (Ghafari et al., 2009; Liao et al., 2015; Trinh and Kang, 2010; Wang et al., 2011), the relationship between the treatment agents and removal is expected to be non-linear in the case of physico-chemical processes applied to water treatment. Therefore, a second-order model must be used as a surface response to fit the data and identify the optimal treatment conditions.

As shown in Table 2, polymer, PAC, zeolite and mesh size were placed at one of three equally spaced values, coded as −1, 0, +1. The responses were expressed as a second-order polynomial equation and a mathematical model was developed according to Eq. 1:

$$\begin{aligned} Y = \beta_0 + \ \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_4 + \beta_5 x_1^2 + \beta_6 x_2^2 + \beta_7 x_3^2 + \beta_8 x_4^2 + \beta_9 x_1 x_2 \\ + \beta_{10} x_1 x_3 + \beta_{11} x_1 x_4 + \beta_{12} x_2 x_3 + \beta_{13} x_2 x_4 + \beta_{14} x_3 x_4 \end{aligned} \tag{1}$$

where Y is the predicted response in terms of pollutant removal; β_0 is the constant coefficient; x_1, x_2, and x_3 are the independent variables which influence the predicted response Y; $\beta_1, \beta_2, \beta_3$, and β_4 are the linear coefficients; $\beta_5, \beta_6, \beta_7$ and β_8 are the quadratic coefficient, and β_9 , $\beta_{10}, \beta_{11}, \beta_{12}, \beta_{13}$, and β_{14} are the cross-product coefficients. The model equation easily clarifies the interaction effect such as synergism, antagonism, and addition of the independent parameters. The statistical significance of each variable was analyzed by observing the p-value; p-values less than 5% ($p < 0.05$), indicates that the variable is considered to be statistically significant. The validity of each model was expressed by the coefficient of determination R^2, ranged between 0 to 1. A R^2 value close to 1 is desirable to ensure a good fit of the quadratic model to the actual data, as well as to assure that the RSM correctly explained the interactions between dependent and independent variables based on the experimental results.

The RSM obtained with the aforementioned procedure were used to optimize the treatment process. In the optimization study, treatment factors were calculated for feasible combinations of polymer, PAC and zeolite (ranged between -1 and 1 in coded units), and then evaluated for the relative dominance status of each pollutant removal objectives. The Pareto analysis was used to evaluate the entire feasible space of treatment combinations for each model response (pollutants removal). The Pareto frontier was identified by setting two treatment objectives: 1) minimization of the amount of chemical used and 2) maximization of the extent of removal for each pollutant (%). The desirable goal was to identify a combination of treatment agents able to maximize pollutant removal while minimizing the amount of chemicals. By inspecting the Pareto frontier, non-dominated designs could be easily identified for each of the pollutants considered in this study and compared.

The experimental results were processed and interpreted by employing Minitab Statistical Software (version 17, State College, Pennsylvania, USA).

3.3 Results and discussion

3.3.1 Wastewater characteristics

The physico-chemical characteristics of the wastewater used in this study are shown in Table 3.3 and compared with data obtained from the literature. TSS data were estimated by correlation with turbidity based on 14 samples analyzed with both methods. No general trends was identified from the information available in the literature thus confirming that CSO water quality is region-specific, and largely dictated by the catchment and rainfall characteristics of the geography of concern (Kafi et al., 2008; Madoux-Humery et al., 2013; Suárez and Puertas, 2005). Since this work focused on the simultaneous control of CSO pollutants and nutrient runoffs (P and N), and because CSOs water quality is originated from rainwater-diluted sewage, we considered raw wastewater suitable CSO surrogate for our research purposes.

Table 3-3: Sample characteristics and comparison with CSO characteristics reported by literature

	This study				**From literature**							
	Samples			Average ± Standard Deviation	(Metcalf & Eddy, 2014)		(Suárez and Puertas, 2005)		(Diaz-Fierros et al., 2002)		(Gasperi et al., 2012b)	
	1	2	3		Min	Max	Min	Max	Min	Max	Min	Max
Turbidity (NTU)	269	278	276	274.33±4.7	-	-	-	-	-	-	-	-
*TSS * (mg/L)*	619	640	636	632±9.31	270	550	561	1722	160	411	204	393
t-COD (mg/L)	759	744	780	761±18	260	480	569	1717	134	540	270	560

TKN (mg/L)	74	67	68	69.6±3.8	4	17	-	-	13.2	33	29	46
TP (mg/L)	34	33	33.5	33.5±0.5	1.2	2.8	-	-	0.5	4.6	4.3	6.5

** Data converted from turbidity to TSS using the correlation included in Appendix A (Figure S3.3)*

3.3.2 Integrated treatment performance

Table S3.1 in Appendix A summarizes results obtained during the experimental runs. It should be noted that the effect on treatment performance associated with the influent particulate, is secondary as the overall solids content of the treatment is dominated by the externally added carbon/zeolite particles. For the soluble components, the removal is mostly associated with absorption mechanisms by carbon and zeolite. A statistical test was conducted on the experimental data collected with the BB-DOE scheme, at a significance level of p-values < 0.05. The latter indicated whether the removal of a given pollutant (listed as a column in Table 3.4) was affected by the treatment agent utilized in the study (listed as a row in Table 3.4). The full statistical analysis, reported in Table S3.2 of Appendix A, confirmed the hypothesis that an integrated treatment process able to cope with a wide spectrum of pollutants requires the simultaneous use of all treatment agents, and justifies a modeling study to optimize the process while achieving multiple treatment objectives.

Table 3-4: Statistical significance ($p < 0.05$) for each treatment agent on the removal of each pollutant

	BB-DOE main factors analysis			
Parameters	*Polymer*	*PAC*	*Zeolite*	*Mesh size*
Turbidity	Yes	No	No	No
s-COD	No	Yes	No	No

p-COD	Yes	Yes	No	No
TKN	Yes*	No	Yes	Yes*
TP	Yes	No	No	No

**Statistically significant factor via a two-way interaction*

Figure 3.2 shows the tri-dimensional plots, in the form of the response surface, highlighting the trend in performance manifested by the combined process for the case of turbidity and total phosphorus removal. The experimental results revealed that, by using a cationic polymer, >75% of both pollutants could be simultaneously removed leading to a final concentration of <50 NTU and <7 mg/L TP, respectively, in the CSO-simulated treatment. Such considerable extent of particulate removal is consistent with findings from previous studies (Li et al., 2003; Scherrenberg, 2006; Zahrim et al., 2011) and confirms the effectiveness of coagulation and microsieve filtration when used in combination. Indeed, the positive charge of a cationic polymer effectively neutralizes the negative charge of particles allowing floc formation, therefore facilitating particle separation by fine sieve microfiltration. Also, during the microsieving process, it is possible that polymer facilitated the formation of a thin cake layer on the filter surface, which in turn further enhanced particle removal.

In the case of total phosphorus, high level of removal is mainly associated with the combined effect of direct sieving (for particulate phosphorus) and the sequestration, by adsorption, of phosphate by zeolite. This mechanism is confirmed by the statistically significant effect associated with the product of polymer and zeolite at p-value < 0.05.

The regression analysis returned the following expressions by statistical analysis for the two pollutants considered in this section (in coded units):

$$\begin{aligned} Turbidity = {} & 78.22 + 6.737\,Polymer - 1.359\,Zeolite + 0.072\,PAC - 0.340\,Mesh \\ & - 3.98\,Polymer^2 - 1.05\,Zeolite^2 + 0.39\,PAC^2 + 0.30\,Mesh^2 + 1.97\,Polymer \\ & * Zeolite - 0.17\,Polymer * PAC + 0.31\,Polymer * Mesh + 0.32\,Zeolite * PAC \\ & - 0.17\,Zeolite * Mesh + 0.18\,PAC * Mesh \end{aligned}$$

$$\begin{aligned} TP = {} & 75.629 + 2.334\,Polymer - 0.355\,Zeolite + 0.293\,PAC + 0.031\,Mesh \\ & - 0.597\,Polymer^2 - 0.280\,Zeolite^2 - 0.373\,PAC^2 + 0.672\,Mesh^2 \\ & + 1.382\,Polymer * Zeolite + 0.429\,Polymer * PAC - 0.075\,Polymer * Mesh \\ & + 0.691\,Zeolite * PAC - 0.075\,Zeolite * Mesh - 0.131\,PAC * Mesh \end{aligned}$$

The model equations showed an $R^2 = 0.89$ and 0.90 for turbidity and TP removal, respectively.

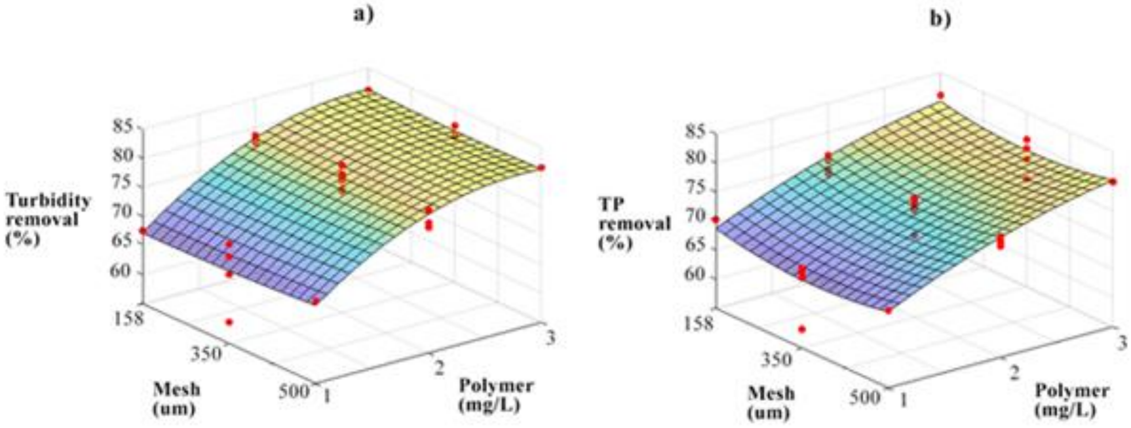

Figure 3-2: 3D surface plot for (a) turbidity removal, and (b) TP removal.

The same analysis was repeated in the case of COD and TKN. The statistical analysis confirmed that also for this case, all the treatment agents were statistically significant in achieving up to 75% of p-COD removal. Moreover, response surface analysis revealed that the two most important factors were polymer and PAC. The regression equation for p-COD removal (%) is proposed in coded unit as follows:

$$
\begin{aligned}
p\text{-}COD = {} & 59.30 + 8.29\,Polymer - 1.68\,Zeolite - 5.33\,PAC - 0.72\,Mesh - 0.14\,Polymer^2 \\
& - 0.32\,Zeolite^2 + 2.02\,PAC^2 + 8.69\,Mesh^2 + 2.40\,Polymer * Zeolite \\
& + 1.74\,Polymer * PAC - 6.31\,Polymer * Mesh - 1.62\,Zeolite * PAC \\
& - 1.99\,Zeolite * Mesh - 3.84\,PAC * Mesh
\end{aligned}
$$

It is interesting to note how the surface plot for p-COD removal (Fig.2a) reports an inverse relationship between polymer and PAC: the p-COD removal efficiency increases by adding the highest concentration of polymer and the lowest concentration of PAC. This could be due to the fact that PAC was a facilitating agent for coagulation by providing external coagulation nuclei, but only until a critical upper concentration of carbon particle was reached. On the other hand, the p-values analysis highlighted that PAC played a decisive role in the removal of s-COD. The regression equation (coded units) developed by RSM for s-COD removal (%) is proposed as follows:

$$
\begin{aligned}
s\text{-}COD = {} & 12.65 + 0.991\,Polymer + 1.086\,Zeolite + 3.211\,PAC - 0.026\,Mesh - 1.88\,Polymer^2 \\
& - 3.26\,Zeolite^2 - 1.88\,PAC^2 - 3.04\,Mesh^2 - 1.30\,Polymer * Zeolite \\
& - 0.94\,Polymer * PAC + 1.08\,Zeolite * PAC + 1.45\,Zeolite * Mesh + 1.37\,PAC \\
& * Mesh
\end{aligned}
$$

The curve plot in Figure 3.3 confirms that the level of s-COD removal increased by increasing the concentration of PAC.

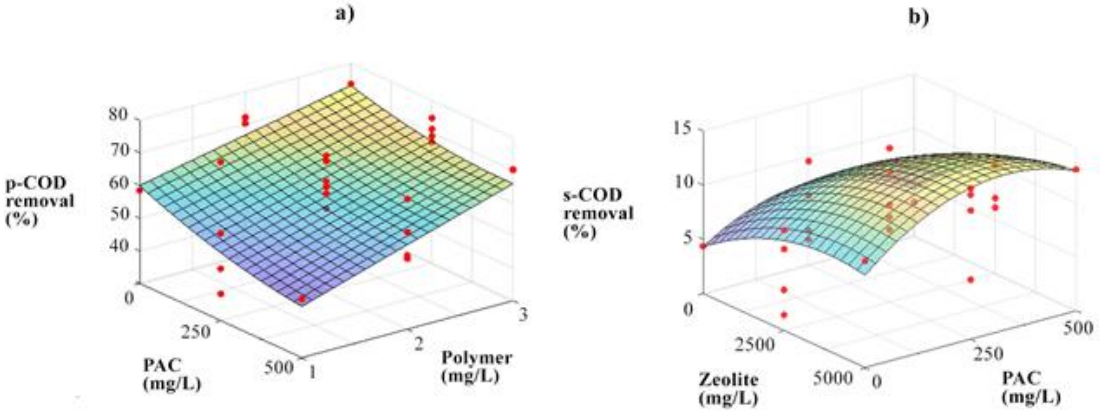

Figure 3-3: 3D surface plot for (a) p-COD removal, and (b) s-COD removal.

By observing the surface curvature, a maximum in the removal of s-COD could be observed for an optimal combination of PAC and zeolite (250 mg/L and 2,500 mg/L, respectively). This is in agreement with previous studies (Liao et al., 2015; Malekmohammadi, 2016) who emphasized that PAC and zeolite if used in combination, lead to an increase in adsorption efficiency. The regression equation for TKN removal (%) is proposed in coded unit as follows:

$$\begin{aligned} TKN = {} & 68.759 + 0.027\,Polymer + 7.308\,Zeolite + 0.106\,PAC - 0.638\,Mesh - 1.794\,Polymer^2 \\ & - 4.249\,Zeolite^2 - 2.436\,PAC^2 - 2.658\,Mesh^2 - 0.399\,Polymer * Zeolite \\ & + 0.053\,Polymer * PAC - 1.472\,Polymer * Mesh + 1.135\,Zeolite * PAC \\ & + 0.301\,Zeolite * Mesh - 0.195\,PAC * Mesh \end{aligned}$$

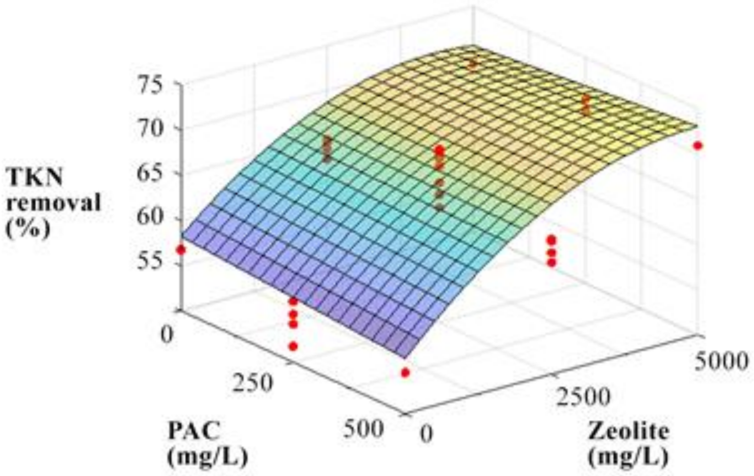

Figure 3-4: 3D surface plot for TKN removal.

The surface plot reported in Figure 3.4 shows that the highest level of TKN removal (> 65% and with ammonia removal up to 40%) is achieved at high zeolite concentration, regardless of the concentrations of the other agents including PAC. Moreover, experimental results indicated that no TKN could be removed without the addition of zeolite, thus confirming the importance of this treatment agent for nitrogen control and in line with findings from previous studies (Wang and Peng, 2010). In Table 4, a linear interaction of zeolite and a two-way interaction of polymer and mesh size for the removal of TKN are reported. This result confirm that ammonia removal occurs via a dual mechanism of ammonia capture by ion exchange on zeolite, and a subsequent step of polymer coagulation and microsieving filtration.

Figure 3.5 shows the predicted and the observed values for the removal of the five pollutants considered in this study, indicating an excellent agreement between model and experimental values. The model has also been successfully tested by the “leave one out” cross-validation method, with results are reported in supporting information file (Figure

S3.2 of Appendix A). As expected, each variable has its importance in the developed model.

The chart in Figure.3.5 has been divided into three arbitrary regions aimed to classify the various pollutants removal: poorly removed (<30% removal), moderately removed (between 30% and 60%) and efficiently removed (>60%). Among the considered pollutants, only s-COD was poorly removed, while turbidity, TP, p-COD and TKN all displayed removal in the range of 45% to 80%.

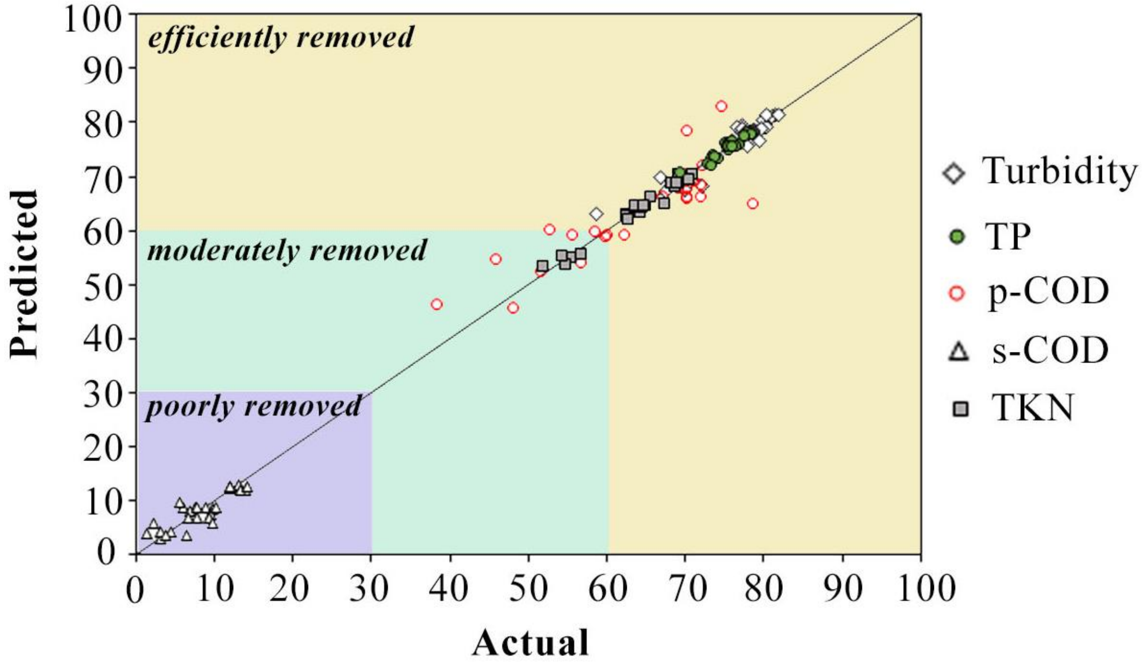

Figure 3-5: Predicted vs. actual values for turbidity, TP, p-COD, s-COD, and TKN

3.3.3 Pareto frontier and scenario analysis

Figures 3.6, 3.7, 3.8 and 3.9 report the assessed combinations of treatment agents as well as those falling onto the Pareto frontiers when the maximization of removal and the minimization of treatment agents used in the process are simultaneously specified as the treatment objectives. Moreover, these plots provide useful information on the trade-offs between the two treatment objectives (i.e., simultaneous minimization of chemical cost and maximization of treatment performance).

In the following figures, the Pareto designs (non-dominated solutions), each design consisting of a unique combination of treatment agents, are reported in red, yellow, green and blue for turbidity, t-COD, TKN and TP, respectively. The dominated solutions (sub-optimal designs) are reported in grey. From Figures 3.6 and 3.7, it can be seen that all the frontier designs for turbidity, t-COD, TP and TKN converge towards the minimum amount of chemicals used to detach themselves when the amount of chemicals increase. Turbidity, t-COD and TP follow the same trend suggesting that a combination of designs can be easily selected to effectively remove all these pollutants.

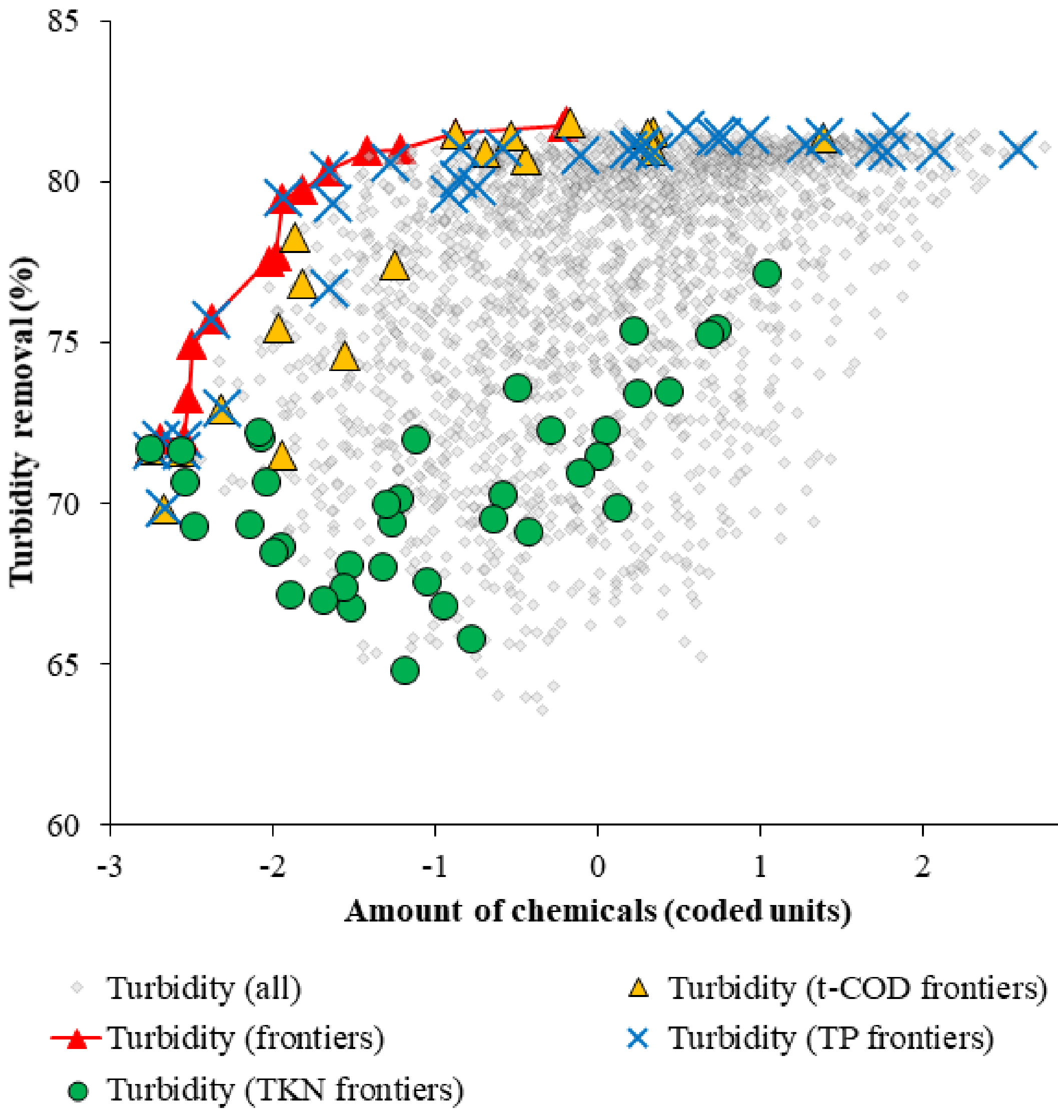

Figure 3-6: Pareto frontier (non-dominated designs) plot for turbidity removal (red triangles). In grey, the dominated solutions for turbidity.

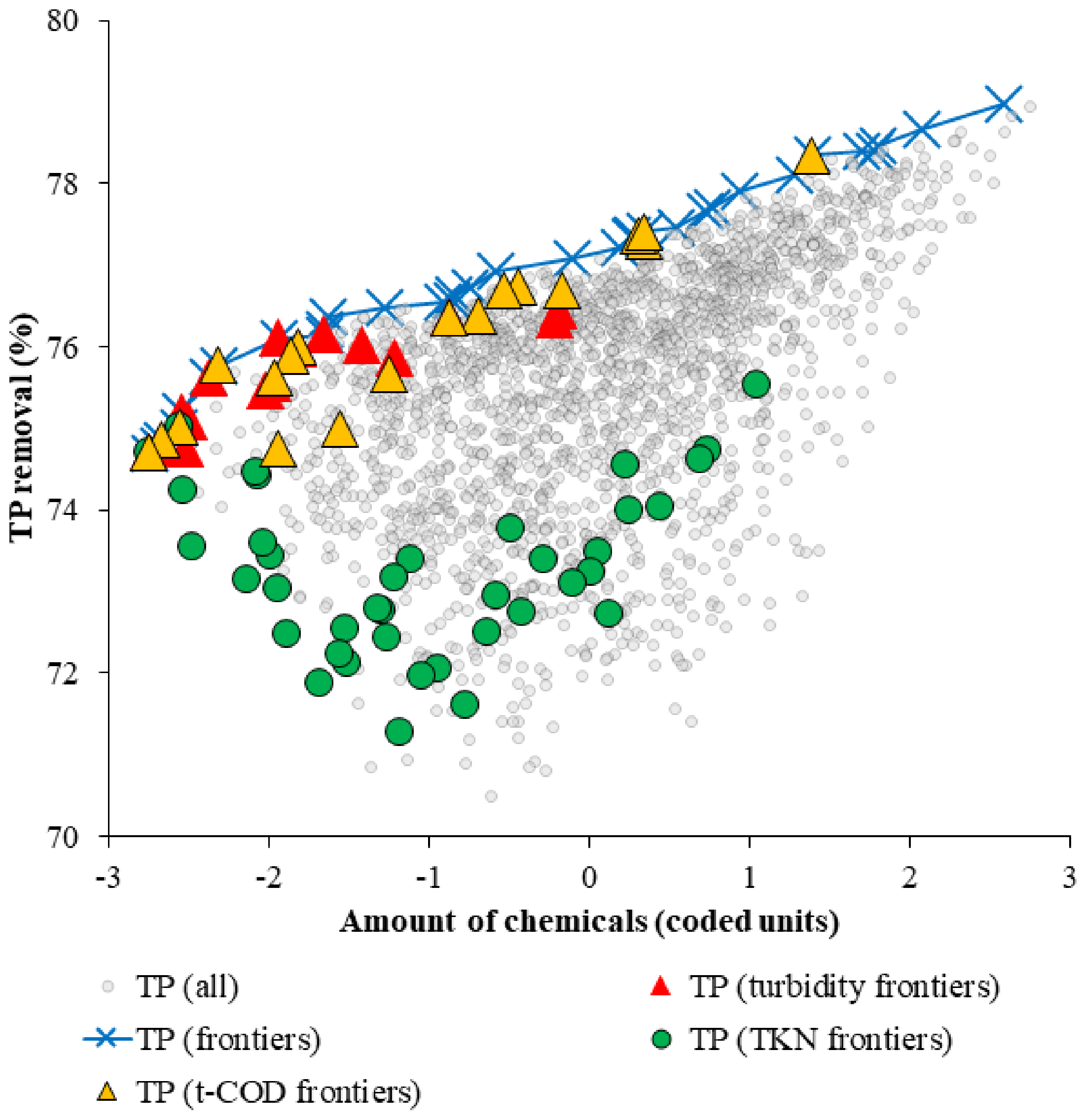

Figure 3-7: Pareto frontier designs (non-dominated solutions) plot for TP removal (blue crosses). In grey, the dominated solutions for TP.

However, by plotting the TP-optimal designs on the t-COD plot (Figure 3.8), it is possible to observe that two sub-curves are defined: the first one follows the t-COD optimal design (Pareto frontiers), while the second one departs from the t-COD-optimal designs. Interestingly, it was seen that the distance between TP and t-COD frontier designs tends to increase when zeolite and mesh size range between 3000-5000 mg/L and 300-500 µm while it tends to decrease when zeolite concentration is kept between 2500 mg/L to 3000 mg/L and the mesh size at 200 µm.

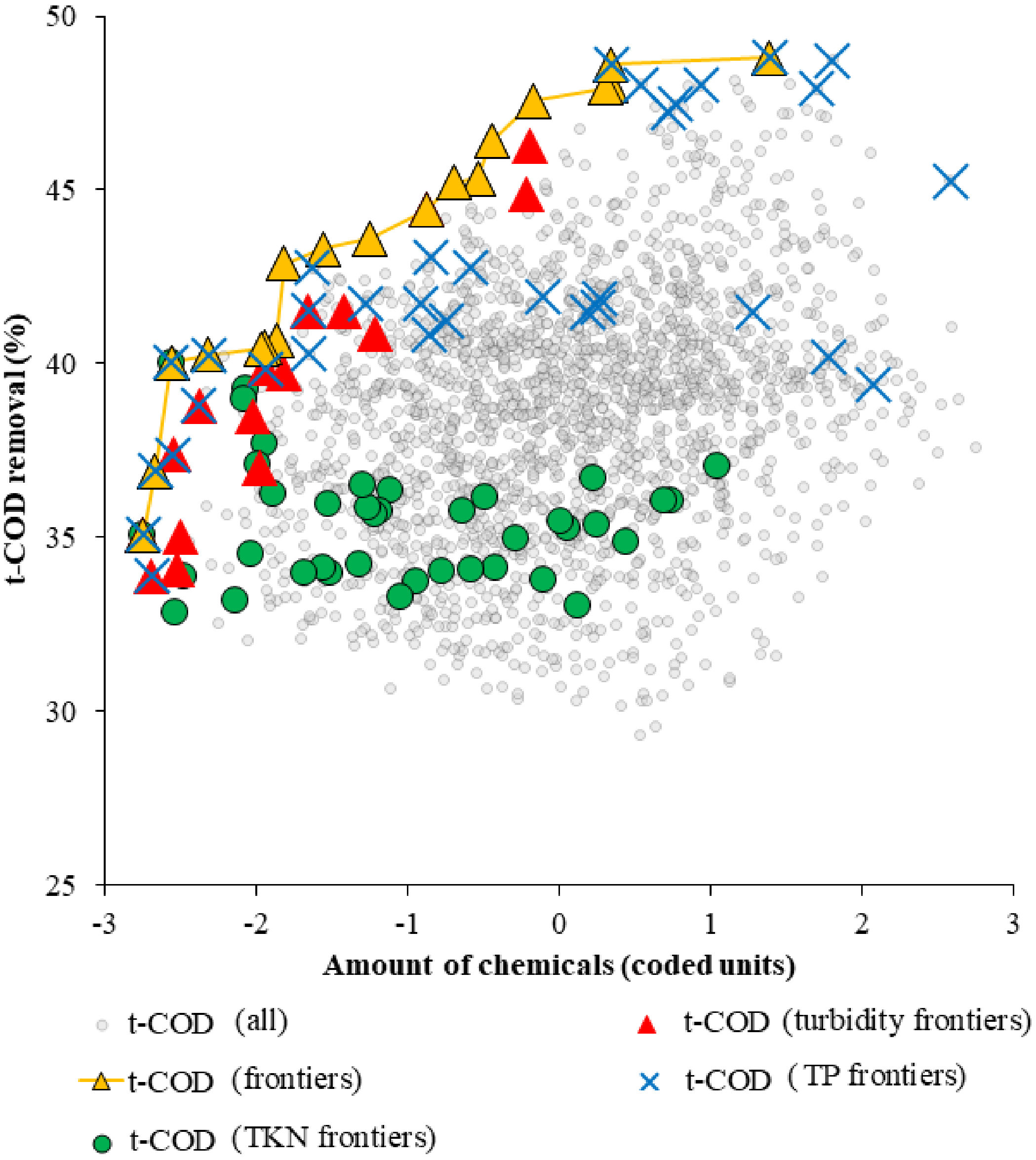

Figure 3-8: Pareto frontier designs (non-dominated solutions) plot for t-COD removal (yellow triangles). In grey, the dominated solutions for t-COD.

On the other hand, as the shape of the TKN Pareto frontiers indicates, TKN removal is very sensitive to the zeolite concentration, with the highest level of removal (72%) reported by using the highest concentrations of zeolite (4500 mg/L). When the zeolite concentration is reduced to 250 mg/L, the TKN removal drops to 53%. Observing Figure 3.9, it is also interesting to note that the designs that are optimal for nitrogen removal are sub-optimal for all the remaining pollutants. As such, a universal treatment able to optimally remove all the considered pollutants cannot be advanced. However, all the designs belonging to the four Pareto frontiers tend to converge when the minimum amounts of chemicals are used. Such point is characterized by the following combination of treatment agents: 1.1 mg/L of the cationic polymer, 250 mg/L of zeolite, 5 mg/L of PAC, and a 370 μm mesh size. Under these conditions, excellent performance as high as 71.6% of removal in turbidity, 55.7% removal in TKN, 35% for t-COD and 75% for TP are observed.

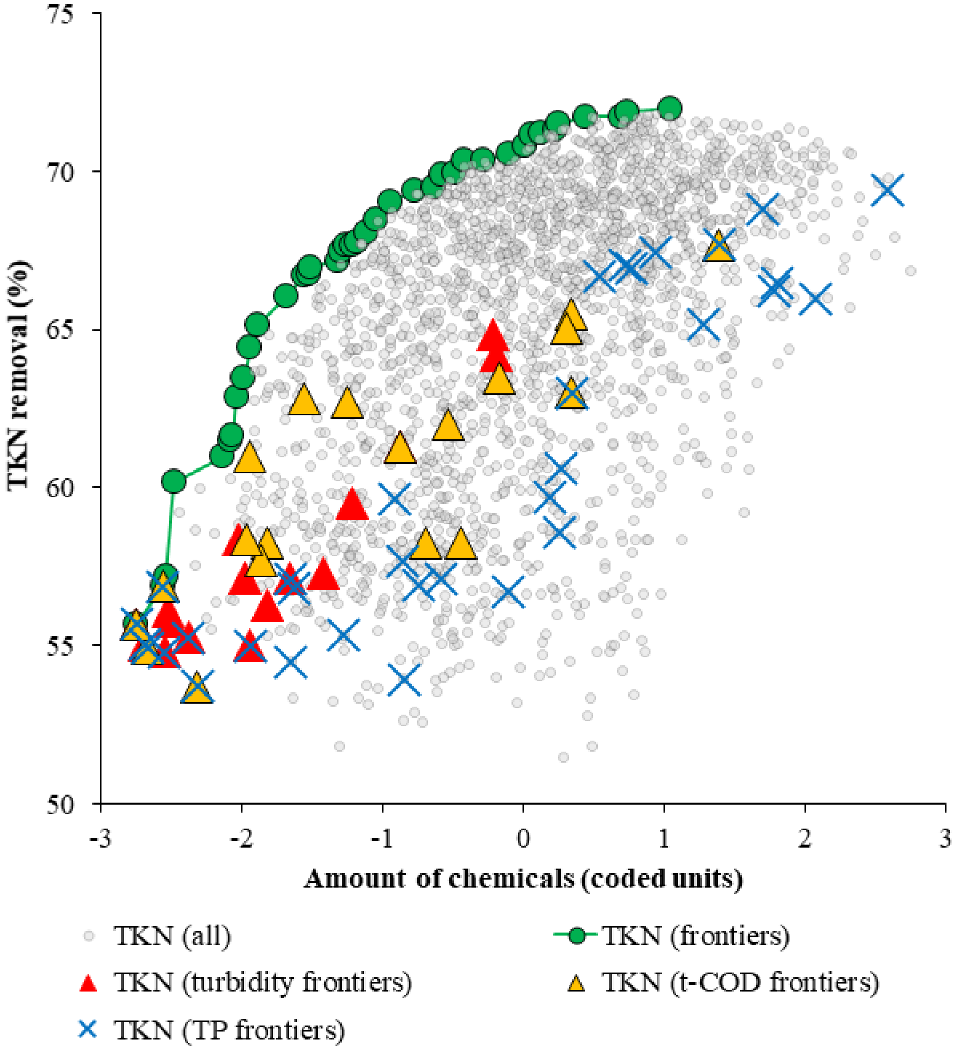

Figure 3-9: Pareto frontier designs (non-dominated solutions) plot for TKN removal (green circles). In grey, the dominated solutions for TKN.

Since the minimization of chemicals used in CSO treatment is a highly desirable goal for public health protection, we explored the performance of optimal designs that were simultaneously able to perform with low-chemicals usage while also achieving the advanced treatment goals such as a) simultaneous maximum removal of turbidity, t-COD, TKN, and TP achieving the best performance on each removal, b) simultaneous maximum removal of t-COD, TKN, and TP, and c) simultaneous maximum removal of turbidity and

TP. Table 3.5 shows the three combinations of treatment agents extracted from the Pareto frontier designs and in line with the three scenarios indicated above. In Table S3.3 of Appendix A, the treatment agents have been normalized by the influent pollutant loads to provide useful sizing information for process scale-up as a function of the different treatment goals pursued in this study.

The design O-1 was identified as the best combination of treatment agents for the first scenario (case a). Remarkably, O-1 belongs to all the four frontiers associated with the removal of each pollutant taken individually. When the treatment objective was set to maximize the removal of nutrient and organic pollutants by using the smallest amount of chemicals (case b), an increase of zeolite dosage is required, and the O-2 combination resulted in being optimal. Such design appears to be common for three frontiers, i.e. t-COD, TKN, and TP frontiers. Finally, for the third scenario (case c), the design O-3 appeared to be optimal: an increase of polymer dosage and a reduction of mesh size is required to pursue the simultaneous removal of turbidity and TP. It should be noted that O-3 allows achieving one of the highest removals in turbidity and TP, even with a relatively small amount of chemicals.

Table 3-5: Treatment alternatives based on different treatment objectives

	Removal (%)				**Design**				**N. of frontiers per design**
	Turbidity	*t-COD*	*TKN*	*TP*	*Polymer (mg/L)*	*Zeolite (mg/L)*	*PAC (mg/L)*	*Mesh (µm)*	
O-1	72	35	56	75	1.13	250	5	370	4
O-2	72	40	57	75	1.17	650	2.5	475	3

O-3	80	42	57	76	2.1	550	5	180	2

3.4 Conclusions

One of the main goals of this study was to investigate the performance of a novel, integrated process based on the simultaneous treatment by cationic polymer, zeolite, and powdered activated carbon followed by microsieving filtration. Results suggested that:

- The novel integrated treatment process proposed in this study could be exploited to deal with multiple contaminants and the associated impacts in the receiving bodies caused by CSO pollution and nutrients discharge in the environment.
- All treatment agents tested in this study, i.e. cationic polymer, powdered activated carbon, and zeolite, have shown synergistic effects when simultaneously dosed prior to microsieving for treating the major CSO pollutants.
- Cationic polymer played a fundamental role in coagulating zeolites, on which ammonia was initially absorbed, thus indirectly enabling the removal by a soluble constituent by microsieving filtration.
- As highlighted by response surface analysis, while zeolite played a central role in achieving satisfactory removal (>50%) of dissolved nitrogen in the form of ammonium. At the same time, low removal of soluble COD by powder activated carbon was observed (<15%).
- A regression model able to describe the relationship between treatment agents and CSO pollutants removal was developed. The model was employed to perform a multi-objective optimization of the treatment method, and to identify Pareto frontiers, demonstrating the possibility of pursuing, with a single treatment method, multiple treatment objectives.

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Chapter 4

4 Low-fluence UV disinfection for Combined Sewer Overflow

4.1 Introduction

During wet weather events, the flow in a combined sewer system usually exceeds the maximum capacity and a combined sewer overflow (CSO) occurs (Scherrenberg, 2006). When a CSO event occurs, a mixture of raw sanitary wastewater, raw industrial wastewater, and rainwater is discharged to surface waters without receiving any treatment. According to EPA (EPA, 2004), it was estimated that, in the USA, the number of CSO discharge events in 2002 was more than 9,000, corresponding to approximately 850 billion gals of untreated wastewater being discharged to surface waters nationwide. While the cost estimate to address CSO was $50.6 billion, only $6.0 billion had been spent through 2002, highlighting the discrepancy between the cost to address CSO and the amount of available funds to bring CSO into compliance with water quality standards (EPA, 2004). As a result, the receiving waters will get polluted by dissolved (soluble) and insoluble pollutants impacting aquatic life, drinking water resources, fish health and consumption, shellfish harvesting, water recreation, and human health with a risk of causing diseases as gastroenteritis, dysentery, cholera, and hepatitis (Anne-Sophie et al., 2015; Barco et al., 2008; Diaz-Fierros et al., 2002; Eganhouse and Sherblom, 2001; EPA, 2004; Kafi et al., 2008; Venditto et al., 2020; Weyrauch et al., 2010). Several studies reported data on the microbiological CSO quality characteristics and their impact on the receiving waters (Donovan et al., 2008; Ham et al., 2008; Mclellan et al., 2007; Passerat et al., 2011), concluding that CSO is one of the primary sources of microbial pollution in surface waters such as lakes and rivers. Mclellan et al. (2007), (Mclellan et al., 2007) reported that *E. coli* levels in the Milwaukee River, following CSO events, ranged from 10^4 to nearly 10^5 CFU/100 mL, highlighting that CSO has a considerable impact on the microbiological quality of the river. In another study on pathogen-related disease risk for users of the Lower Passaic River in New Jersey, it was stated that the release of pathogens into the river via CSO will continue to be a significant human health risk until CSO discharges are adequately controlled (Donovan et al., 2008),.

To reduce the impact of discharges in surface waters, the EPA published a guidance document, ‘‘Combined Sewer Overflow Control’’ (EPA, 1993), discussing methods to achieve high-rate disinfection of wet weather flows. The guidance document indicates that an effective CSO disinfection would achieve a reduction in bacteria concentration of at least 4-log (99.99% removal) at a contact time of 15-30 minutes. To combat waterborne diseases, different disinfection methods have been used to inactivate pathogens coming from CSO discharges (Corporation, 2010). Among them, chlorine-based disinfecting agents, ozone, peracetic acid (PAA), performic acid (PFA), and ultraviolet (UV) light are the most common disinfection processes. Chlorine is the most widely used disinfectant, but its use is decreasing in the water industry because of the formation of potentially toxic and carcinogenic chlorinated by-products (Bayo et al., 2009; Nurizzo et al., 2005; Watson et al., 2012). Ozone is a very efficient disinfectant for drinking water, but its application in wastewater treatment is limited due to operation and maintenance limitations (Chhetri et al., 2014; EPA, 1999; Gehr et al., 2003; Xu et al., 2002). PAA and PFA are emerging chemical disinfectants that have demonstrated the potential to inactivate microorganisms

(Maffettone et al., 2020; Manoli et al., 2019). This study deals with a physical disinfection process, UV light, to inactivate bacteria and address CSO challenges.

To date, UV irradiation is the most attractive disinfection process for CSO events due to its short contact time requirements and the lack of any toxic by-products (Botturi et al., 2020; Gehr et al., 2003; Gibson et al., 2017; Muller and Lem, 2011; Scherrenberg, 2006; Tondera et al., 2015). However, the high suspended solids content of CSO is a major challenge for UV disinfection (Muller and Lem, 2011; Wojtenko et al., 2001). With particles present in the influent, UV transmittance is significantly decreased, reducing the efficiency of the UV light for microorganism inactivation. Studies investigated the effects of solids on the efficiency of UV disinfection revealing that a relationship exists between the concentration of total suspended solids (TSS) and the removal rate of fecal bacteria (Air et al., 2013; Darby et al., 1993; Jolis et al., 2001). Air et al. (2013) (Air et al., 2013) reported that as the TSS level increase, the UV inactivation rate constants decrease. Madge et al. (1999) (Madge and Jensen, 1999) stated that not only the concentration of solids but also the particle size affect the UV disinfection efficiency. Indeed, the authors underlined that a slower disinfection rate of fecal coliforms (FC) was associated with particles over 20 µm.

To overcome CSO impacts, new and adaptable multifunctional treatment schemes need to be developed. To date, to the best of our knowledge, no study proposed an efficient and cost-competitive treatment able to remove a broad spectrum of CSO pollutants. In our previous study (Venditto et al., 2020), we contributed to fill this gap by developing a microsieve-based filtration pre-treatment process where the effectiveness of chemical pre-treatment followed by micro-sieve filtration was assessed on the removal of multiple contaminants, i.e., chemical oxygen demand (COD), turbidity, as well as nutrients such as nitrogen and phosphorus. The removal of contaminants was achieved by first adsorbing soluble pollutants on zeolite and powdered activated carbon (PAC), and subsequently applying filtration carried out by polymer-enhanced microsieving. An optimal treatment condition consisting of 1.1 mg/L of cationic polymer, 250 mg/L of zeolite, 5 mg/L of powdered activated carbon was identified. Under these conditions, excellent performance as high as 71.6% removal of turbidity, 55.7% removal of total Kjeldahl nitrogen (TKN), 35% for total COD (t-COD), and 75% for total phosphorous (TP) were observed (Venditto et al., 2020).

In this paper, the aforementioned treatment process is implemented by adding the UV disinfection process as the final step of the treatment train. The study aims at contributing towards the identified knowledge gaps by developing an efficient, adaptable, and cost-competitive disinfection treatment process able to improve the quality of surface waters by simultaneously removing microbial and chemical pollutants coming from CSO discharges.

4.2 Material and methods

4.2.1 Source of wastewater

Raw wastewaters (primary influents) were grab sampled after the screening process of a wastewater treatment plant (WWTP) located in London, Ontario, Canada. Four sampling campaigns were carried out (campaigns 1, 2, 3, and 4) under different weather conditions. Campaigns 1 (December, 13th) and 3 (December, 17th) corresponded to the dry weather campaigns while campaigns 2 (December, 14th) and 4 (December, 18th) corresponded to the rain campaigns (precipitation height H = 3.6 mm and 4.6 mm, respectively). For each wastewater sample, TSS, UV transmittance (UVT), and viable fecal coliforms (FC) were measured in triplicates, and averages with standard deviations were reported.

4.2.2 Analytical methods

Wastewaters were analyzed for TSS following Standard Methods 2540 (APHA, 1998), and UVT at 254 nm was measured using a REALUVT meter (REALTECH, Whitby, Ontario, Canada). The standard membrane filtration method (9222D) (APHA, 1998) has been used to measure the concentration (CFU/100 mL) of FC.

4.3 CSO disinfection treatment train and experimental procedure

To identify the best CSO disinfection treatment process, four scenarios were developedand the following experiments were carried out at bench scale:

Scenario 1. Microsieve-based filtration using 350 µm mesh followed by UV irradiation,

Scenario 2. Microsieve-based filtration using 32 µm mesh followed by UV irradiation,

Scenario 3. Chemical pre-treatment followed by microsieve-based filtration using 350 µm and UV irradiation,

Scenario 4. Chemical pre-treatment process followed by microsieve-based filtration using 32 µm mesh and UV irradiation.

While the overall objective of this research was to develop an efficient, adaptable, and cost-competitive disinfection treatment process, the above-mentioned scenarios were used (i) to select the best mesh size for microsieving by evaluating the TSS removal efficiency and UVT improvement, and (ii) to investigate the performance of UV disinfection based on the pathogens inactivation. Thereupon, all the scenarios have been compared to find the best

CSO disinfection process. Ultimately, the best CSO disinfection process was simulated and compared with the no-treatment scenario on a SWMM-simulated CSO to assess its environmental impacts at the urban scale.

4.3.1 Chemical pre-treatment and microsieve-based filtration

The chemical pre-treatment included two main processes, i.e., a coagulation/flocculation process using cationic polymer as coagulant (1.1 mg/L), and an adsorption process using powdered activated carbon (5 mg/L) and zeolite (250 mg/L). The treatment was performed by following the procedures described elsewhere (Venditto et al., 2020), using 1 L of raw wastewater. The pre-treatment process was followed by microsieve-based filtration where two different pore size meshes were tested, i.e., 32 µm and 350 µm. At the end of each treatment, UVT and TSS were measured in triplicates, and averages were reported. Results were compared with UVT values and TSS concentration in the raw (untreated) wastewaters to assess the treatment efficiency in terms of percentage removal.

4.3.2 UV disinfection and microbial inactivation kinetic model

The UV disinfection process was the final step of the proposed CSO treatment train. The UV fluence inactivation response curve was determined in a bench-scale apparatus, known as collimated beam, in which part of the output of a UV lamp is directed onto a horizontal surface through a non-reflective inner surface (Bolton et al., 2003). Fifty milliliters of wastewater were poured into a 60 mm (diameter) x 35 mm (height) crystallization dish containing a magnetic stirring bar, and then placed on a magnetic stirrer under the collimated beam lamp. A low-pressure mercury amalgam UV lamp emitting 253.7 nm has been utilized. The intensity of the incident UV light was measured by placing the IL1700 radiometer detector (International Light Technologies, Peabody, USA) at the same height as the surface of the wastewater. The exposure time was controlled manually by a shutter. The wastewaters were exposed to 4 UV irradiation fluences: 10, 20, 40, and 80 mJ/cm^2. The UV fluence was calculated as the product of the average UV intensity (mW/cm^2) and the average exposure time (s) (Kuo et al., 2003). For each sample, the concentration of FC was measured before and after irradiation, and the microbial inactivation was investigated. All experiments were performed in triplicates, and averages with standard deviations were reported.

For several disinfecting agents such as peracetic acid, performic acid, ferrate and UV, microbial inactivation has been reported to exhibit an initial fast inactivation of dispersed microbes followed by a slower inactivation of particle-associated microbes (Campo et al., 2020; Maffettone et al., 2020; Manoli et al., 2020; Santoro et al., 2015). In this study, an inactivation kinetic model able to describe the aforementioned biphasic behavior was applied to estimate the kinetic parameters controlling the FC inactivation (Santoro et al., 2015):

$$N = N_0 * (1-\beta) * e^{-k_d * UV_{Fluence}} + N_0 * (\beta) * e^{-k_p * UV_{Fluence}} \qquad \textbf{(Eq. 1)}$$

where N is the FC concentration (CFU/100 mL), N_0 is the initial FC concentration (CFU/100 mL), β is the fraction of particle-associated FC (dimensionless), and k_d and k_p are the UV fluence-based microbial inactivation kinetic rate constants for dispersed and particle-associated FC (cm^2/mJ), respectively. To minimize the sum of square errors (SSE) between experimental data and model prediction, β, $k_{d,}$ and k_p were fitted for each experiment individually using Excel Solver. The inactivation kinetic model was evaluated by the coefficient of determination R^2 ranged between 0 and 1. An R^2 value close to 1 is desirable to ensure a good fit of the model to the observed data.

4.4 Results and discussion

4.4.1 Wastewater characteristics

Table 4.1 summarizes the measured wastewater quality parameters for the collected campaigns. Microbial and physical properties measured were consistent with literature-reported CSO values. In particular, a low UVT ranged between 13.0% and 14.0% was measured. The TSS concentration fluctuated between 143 mg/L and 159 mg/L. Importantly, these TSS concentrations are in agreement with previous studies reported on characteristics of CSO (Arnone and Walling, 2006; Gasperi et al., 2012a). The suspended solids content in CSO is a major source of inhibition to disinfection due to its ability to absorb, or scatter, a large amount of UV irradiation, thereby decreasing the amount of UV light available for disinfection. The concentration of FC ranged from 0.55×10^6 CFU/100 mL to 1.25×10^6 CFU/100 mL (Table 1). These values are consistent with data reported for concentrations of FC in combined wastewater or CSO (Louisville, KY and Atlanta, GA). (Arnone and Walling, 2006; Metcalf & Eddy, 2014). The fact that the TSS and FC concentrations of the wastewater used in this study are in agreement with reported CSO quality characteristics is important in terms of CSO relevance of the present paper.

Table 4.1- Microbial and physical sample properties and comparison with CSO characteristics reported by literature Wastewater characteristics

			UVT (%)	TSS (mg/L)	FC (10^6 CFU/100 mL)
This study	**Campaigns** [a]	*1st*	14 ± 0.5	143 ± 11.7	0.60 ± 0.5
		2nd	13 ± 0.5	158 ± 10.4	0.55 ± 1.0

		3rd	13 ± 0.5	151 ± 10.8	1.05 ± 0.5
		4th	14 ± 0.0	159 ± 10.6	1.25 ± 1.1
	Average		***14***	***153***	***0.86***
From literature	(Metcalf & Eddy, 2014)	*Range*	-	270 - 550	0.1 - 1
	(Arnone and Walling, 2006)	*Range*	-	14 - 227	0.03 - 0.43
	(Gasperi et al., 2012a)	*Range*	-	121 - 394	-

[a] All measurements were performed in triplicates and averages with standard deviations were reported.

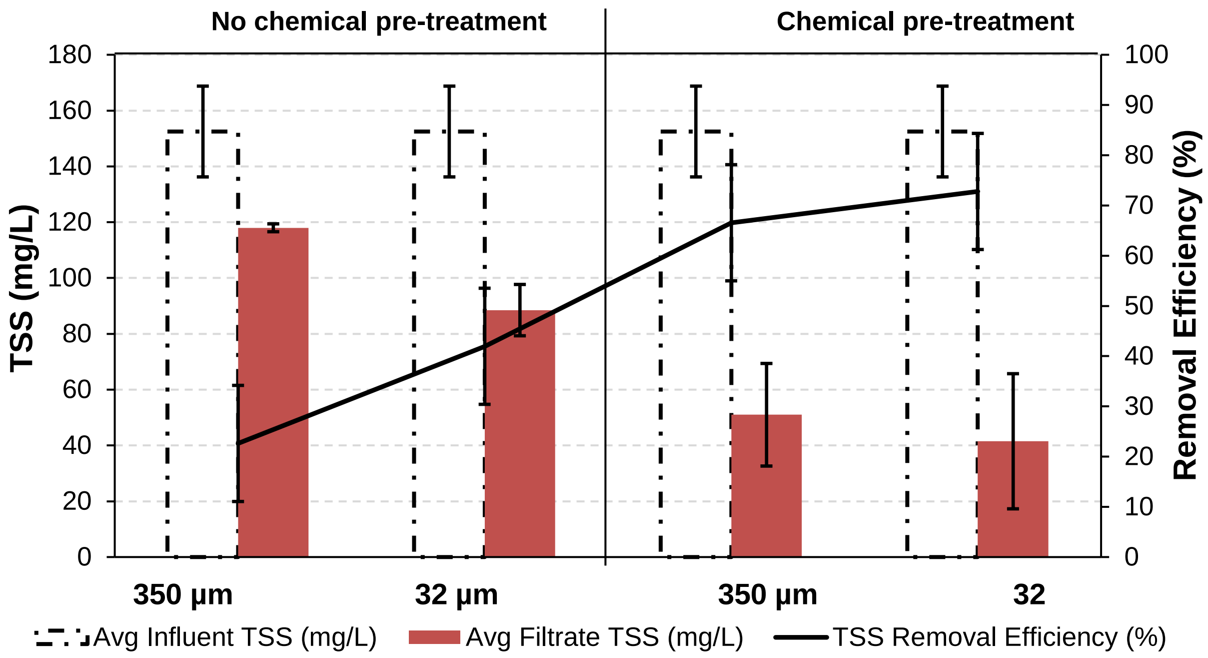
4.4.2 Treatment efficacy on TSS removal and UVT

Figs. 4.1a) and 4.1b) compare the performance of microieve-based filtration in terms of TSS and UVT removal, with and without chemical pre-treatment. As expected, the microsieved-based filtration enhanced by chemical pre-treatment achieved higher removal of TSS compared to the TSS removal observed in the absence of chemical pre-treatment. The TSS concentration ranged between 90-120 mg/L without chemicals and 41-51 mg/L with chemical pre-treatment. At 350 μm mesh, the TSS removal efficiency was 20% by filtration alone (no chemical pre-treatment), while TSS removal increased to 68% with the chemical pre-treatment. At 32 μm mesh, the TSS removal efficiency achieved was 73% and 40% with and without chemical pre-treatment respectively (Fig. 4.1a).

Filtration alone slightly increased UVT from 14.0% to 14.4% and 16.3% by 350 μm mesh and 32 μm mesh, respectively. When filtration was preceded by chemical pre-treatment, UVT increased from 19.6 to ~30.5% by 350 μm mesh, and to ~32% by 32 μm mesh (Fig. 4.1b). Moreover, Fig. 4.1a) shows that, while without chemicals a smaller mesh size significantly increased the level of solids removal, the mesh size slightly affected the removal of particles when chemicals were used. These results are in agreement with previous studies where the performance of coagulation-flocculation using polymer as primary coagulant was investigated in terms of particulate matter removal, e.g., reported typical particle removal efficiencies of around 70-90% using polymer (Chhetri et al., 2016; Delporte et al., 1995; EPA, 2003; Jolis and Ahmad, 2004; Li et al., 2003), and around 39% with filtration alone (Botturi et al., 2020).

The use of polymer as a primary coagulant is considered to be the main contributor to the enhanced particle removal. Indeed, the micorsieve-based filtration increased the removal of particulate matter (up to 73%) when polymer was used as a primary coagulant. The high level of removal is mainly associated with the combined effect of coagulation-flocculation and adsorption. The likely mechanism of the enhanced particulates removal was charge neutralization of negatively charged particles, through reaction with the cationic polymer, followed by adsorption on zeolite and powdered activated carbon. This would result in large floc formation allowing for better exclusion during subsequent filtration. As a result, the negative charge of particles was neutralized through the positive charge of cationic polymer and then adsorbed on zeolite and powdered activated carbon allowing large floc formation. This mechanism facilitated the sieving process regardless of the mesh size used and confirmed the effectiveness of the chemical pre-treatment.

a)


No chemical pre-treatment
Chemical pre-treatment
TSS (mg/L)
0
20
40
60
80
100
120
140
160
180
Removal Efficiency (%)
0
10
20
30
40
50
60
70
80
90
100
350 µm
32 µm
350 µm
32
Avg Influent TSS (mg/L)
Avg Filtrate TSS (mg/L)
TSS Removal Efficiency (%)

b)

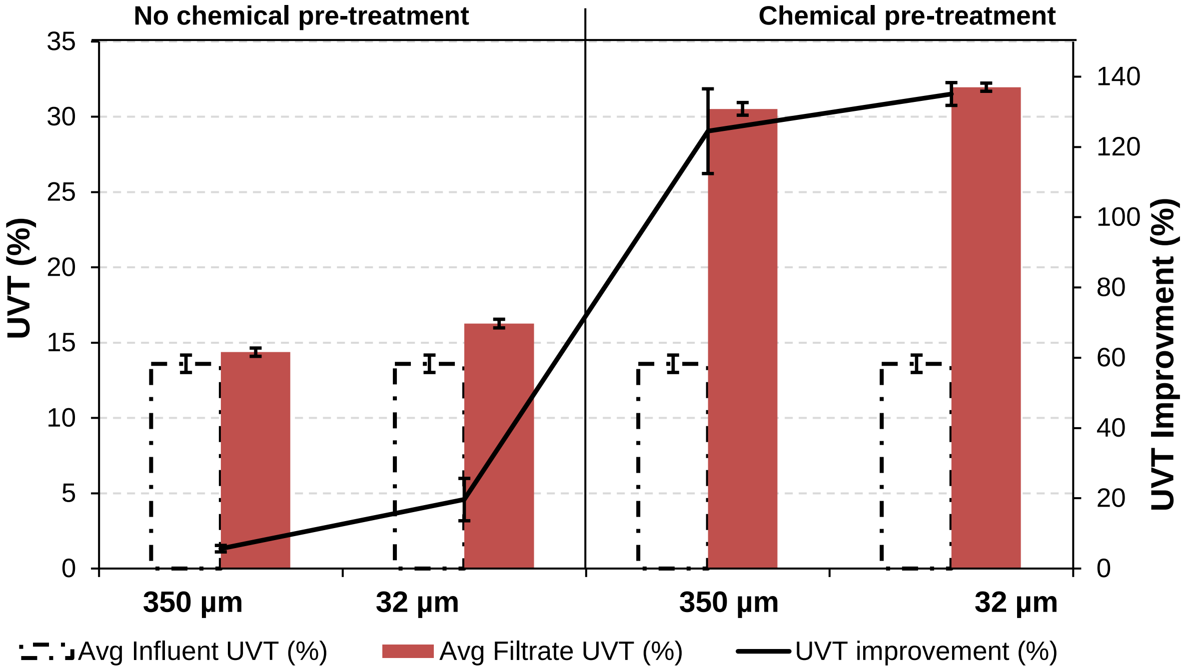

No chemical pre-treatment
Chemical pre-treatment
UVT (%)
0
5
10
15
20
25
30
35
UVT Improvment (%)
0
20
40
60
80
100
120
140
350 µm
32 µm
350 µm
32 µm
Avg Influent UVT (%)
Avg Filtrate UVT (%)
UVT improvement (%)

Figure 4-1: Comparison between microsieve-based filtration alone (no chemical pre-treatment) and chemical pre-treatment: a) TSS concentration (mg/L) and removal efficiency, and b) UVT (%) and UVT improvement with respect to the raw water. Error bars represent standard deviations of reported data.

4.4.3 Treatment efficacy on fecal bacteria

4.4.3.1 UV disinfection enhanced by microsieve-based filtration

Fig. 4.2 shows the UV fluence response curves for the inactivation of FC with and without microsieve-based filtration by 350 μm and 32 μm mesh size. The curve obtained filtering with 350 μm mesh shows a steep decline in numbers with an approximate 2-log reduction at fluences up to 10 mJ/cm^2 followed by an asymptote beyond that fluence. The same trend was observed for the inactivation of FC by UV alone and UV after filtration by 32 μm mesh size (Fig. 4.2). Indeed, the curves obtained using UV alone and UV with 32 μm mesh show a steep decline with an approximate 2.2-log and 2.7-log reductions, respectively, at fluences up to 10 mJ/cm^2. While the dispersed microorganisms were inactivated rapidly in all cases (~2.5-log reduction at UV fluence of 10 mJ/cm^2), much higher UV fluences are needed to further increase the FC inactivation efficiency. For example, to increase the FC reduction from 2.5-log to 3.5-log, a UV fluence higher than 40 mJ/cm^2 was required. This behavior may be related to shielding embedded bacteria from UV irradiation which affects the disinfection process (Darby et al., 1993). It is also observed that filtration by 32 μm mesh did not affect the disinfection process significantly, despite the higher particle removal efficiency than 350 μm mesh (Fig. 4.1a). This result point to the probability that particles smaller than 32 μm are still present in the wastewater, decreasing the disinfection efficiency. Qualls et al. (Qualls et al., 1985) showed that complete inactivation of FC was achieved only in wastewaters where particles bigger than 8 µm were removed. Likewise, Jolis et al. (Jolis et al., 1996) studied the effect of particles on UV inactivation of coliform bacteria reporting that suspended particles smaller than 7 µm have little impact on the bacteria inactivation.

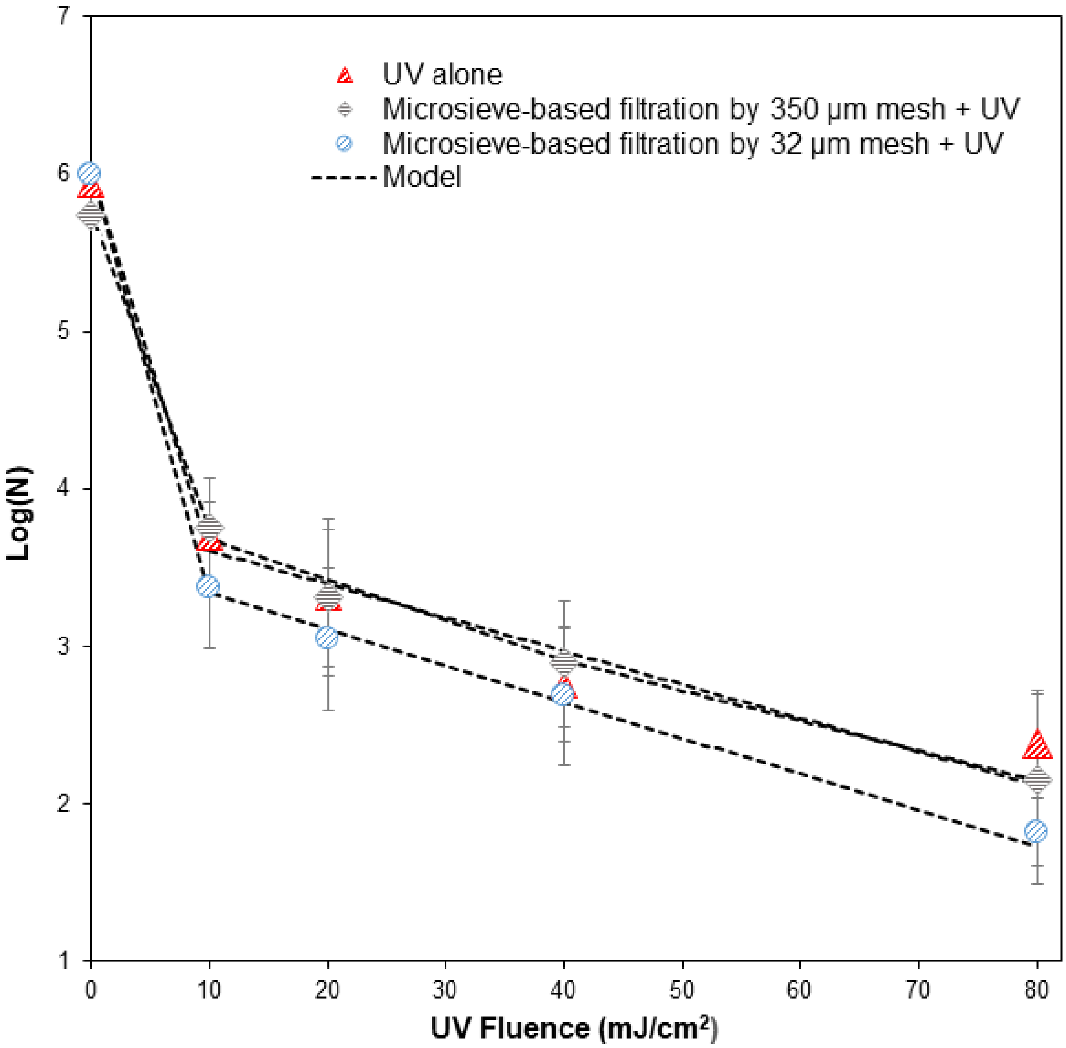


Figure 4-2: UV fluence response curves for the inactivation of FC after microsieve-based filtration by 350 µm mesh and 32 µm mesh. Error bars represent standard deviations of reported data.

4.4.3.2 UV disinfection enhanced by chemical pre-treatment and microsieve-based filtration

Fig. 4.3 shows the FC inactivation curve by UV enhanced by chemical pre-treatment and filtration. During filtration, 350 μm mesh and 32 μm mesh were tested to investigate their impact on the disinfection process. Results show a higher FC inactivation using the 32 μm mesh compared with filtration by 350 μm mesh. The most remarkable result emerges comparing the FC inactivation curve with and without chemical pre-treatment at 32 μm mesh (Figs. 4.2 and 4.3). Significantly, a 4-log reduction at a UV fluence of 10 mJ/cm^2 was achieved with chemical pre-treatment (Fig. 4.3), while without pre-treatment, a 4-log reduction could be achieved at a higher UV fluence of 80 mJ/cm^2 (Fig. 4.2). As it was expected, the lower TSS concentration obtained after the chemical pre-treatment (Fig. 4.1a)) improved the efficiency of the disinfection process, i.e., higher FC inactivation was achieved (Fig. 4.3) (Friedler et al., 2021; Liang et al., 2013). Previous studies of Gehr et al., where the performance of UV disinfection enhanced by physico-chemical processes using ferric and/or alum coagulation was investigated, the FC inactivation curve showed an asymptote zone at UV fluences >20mJ/cm2 achieving approximately 3-log reduction (Gehr et al., 2003). In our study, with chemical pre-treatment, a 4-log reduction of FC was achieved at lower UV fluence of 10 mJ/cm2 (Fig. 4.3). Importantly, the upshot of this result is the possibility to use less UV equipment when a chemical pre-treatment and microsieve-based filtration are applied before the UV disinfection process, thereby reducing the UV treatment cost. This may result in a quick and effective treatment of a large amount of wastewater flow, which is of utmost importance to address CSO challenges. Moreover, since locations associated with CSO discharges are typically not easily accessible and often space-limited, the use of low footprint equipment has the potential to provide municipalities with a compact treatment unit for CSO.

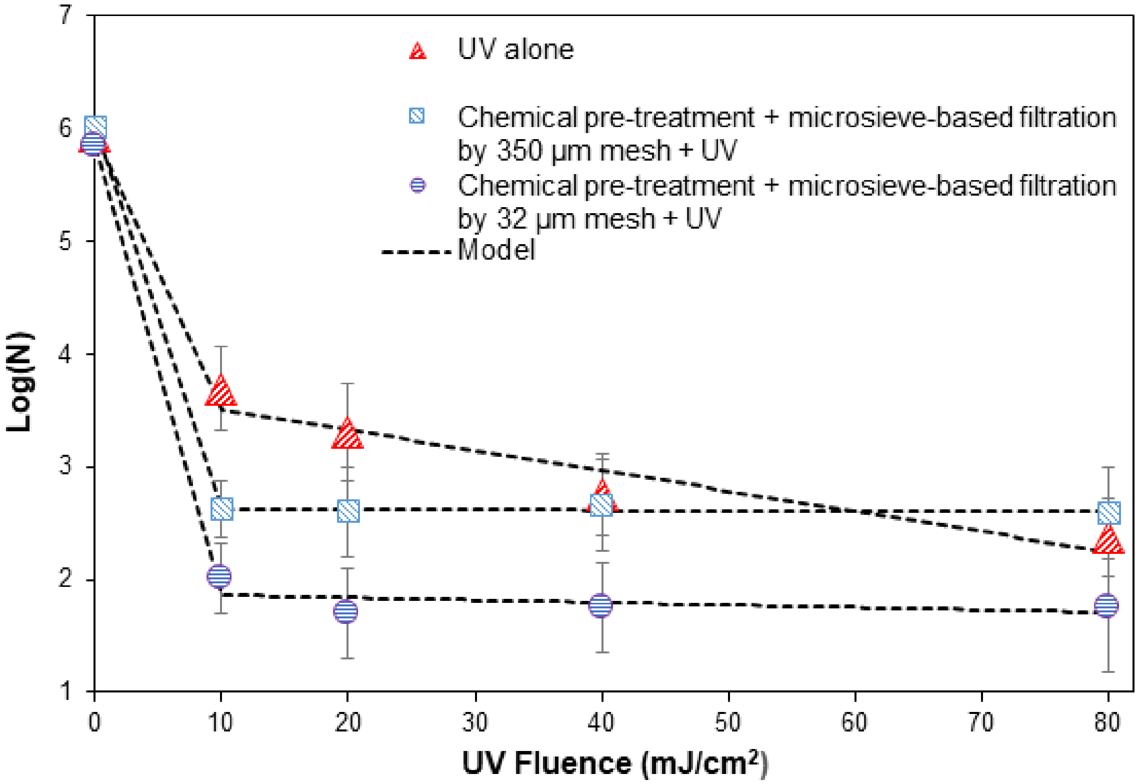


Figure 4-3: UV fluence response curves for the inactivation of FC after chemical pre-treatment and microsieve-based filtration by 350 μm mesh and 32 μm mesh. Error bars represent standard deviations of reported data.

4.4.4 Evaluation of microbial inactivation kinetic model

Table 4.1 reports the microbial inactivation kinetic model fitted parameters. The fraction of particle-associated FC, β, varied from 0.0001 to 0.0085, with the highest β value determined for the UV disinfection after 350 μm mesh filtration with no chemical pre-treatment. This is consistent with the result of low TSS removal (< 25%) obtained after

350 μm filtration (Fig. 4.1a). The lowest β of 0.0001 was determined for the UV disinfection enhanced by chemical pre-treatment followed by 32 μm filtration, demonstrating the efficacy of this treatment train to remove particle-associated microbes. Interestingly, the β values determined by the model fitting are consistent with the results of TSS removal (Fig. 4.1a)). The kd varied from 2.220 to 3.215 cm2/mJ (Table 4.1) with lowest kd determined for the UV alone. The highest kd (~3.2 cm2/mJ) was determined for the UV disinfection enhanced by chemical pre-treatment, indicating the efficiency of the treatment to inactivate dispersed microorganisms. In all cases, a lower k_p (0.005-0.053 cm^2/mJ) than k_d was determined. This result is consistent with the FC inactivation curves presented in Fig. 4.3, where a marked tailing effect is observed after fluence 10 mJ/cm^2 for both the chemical pre-treatment followed by 32 μm and 350 μm.

Table 4-1: Microbial inactivation kinetic model fitted parameters

Treatment	**β**	**k_d cm^2/mJ**	**k_p cm^2/mJ**
UV alone	0.0066	2.220	0.049
Microsieve-based filtration using 32 μm mesh followed by UV irradiation	0.0037	2.626	0.053
Microsieve-based filtration using 350 μm mesh followed by UV irradiation,	0.0085	2.626	0.050
Chemical pre-treatment process followed by microsieve-based filtration using 32 μm mesh and UV irradiation	0.0001	3.203	0.005
Chemical pre-treatment followed by microsieve-based filtration using 350 μm and UV irradiation	0.0004	3.215	0.009

Figure 4.4 shows the model predicted and the observed values for different log reductions of FC for each treatment process. The chart has been divided into three arbitrary regions aiming at classifying the efficiency of each treatment process as low removal (<2-log FC reduction), medium removal (between 2-log and 4-log FC reduction), and high removal (>4-log FC reduction). Among the tested treatments, only the UV irradiation enhanced by chemical pre-treatment followed by microsieve-based filtration using 32 μm mesh achieved a high removal (>4-log reduction of FC) at all the UV fluences applied (10-80 mJ/cm^2). The high R^2 of 0.99 for all the treatments tested, and 0.98 for the UV disinfection alone, indicates an excellent agreement between model-predicted and experimental values. In our previous work (Venditto et al., 2020), we developed a regression model for each one of the main CSO pollutants, and we demonstrated how the regression models were able to achieve different treatment objectives. The inactivation kinetic model (Eq. 1), combined with the regression models developed in our previous work, can be used to pursue multiple treatment objectives making the treatment adaptable to different CSO water quality and quantity.

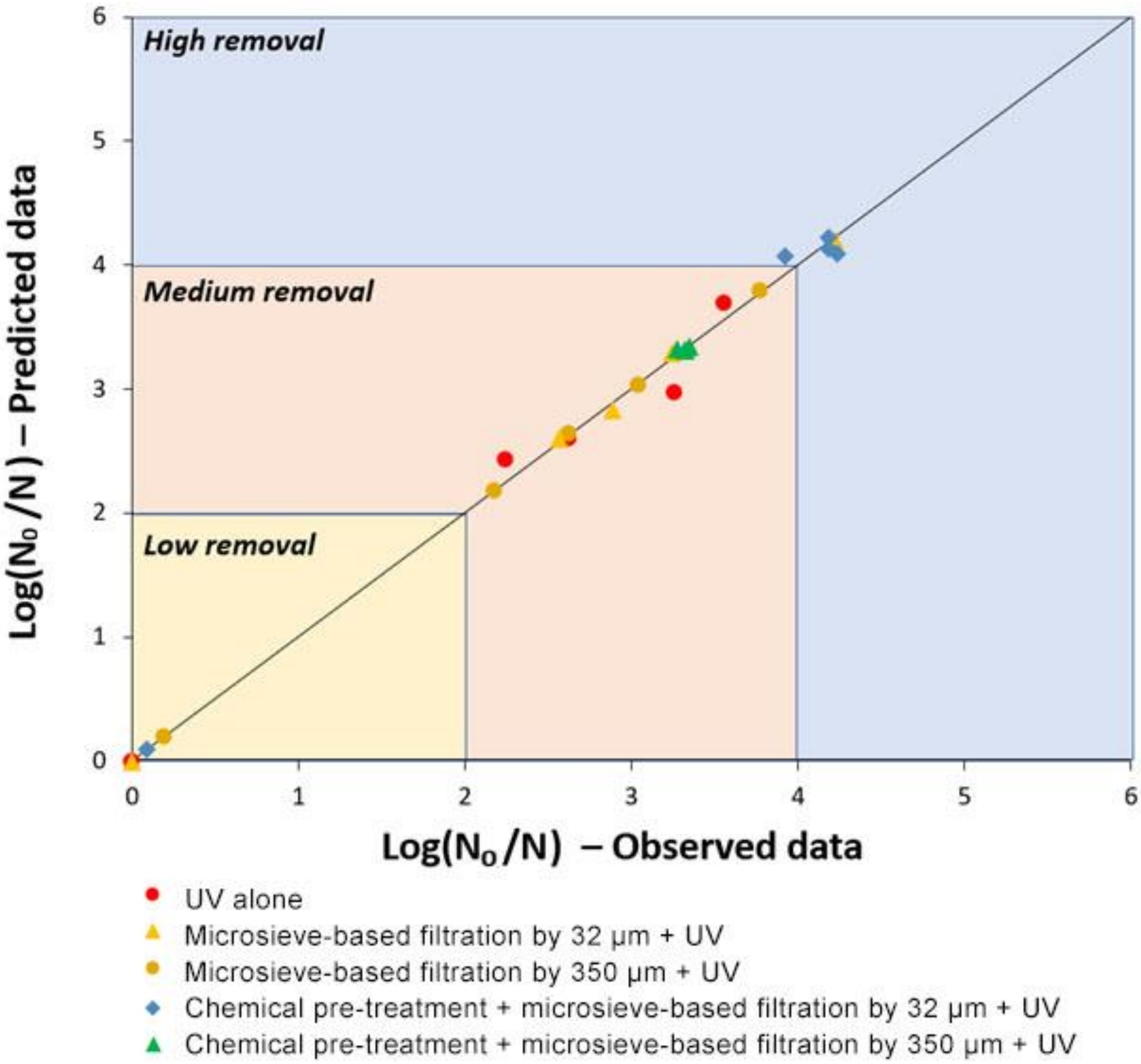


Figure 4-4: Microbial inactivation model assessment: predicted vs. observed

4.5 Conclusion

The main goal of this study was to develop an efficient, adaptable, and cost-competitive disinfection treatment process for low-quality wastewaters. The main conclusions are:

- The UV disinfection enhanced by chemical pre-treatment and microsieving filtration showed better performance at lower UV fluence than without chemical pre-treatment. The highest FC inactivation was obtained by the UV disinfection enhanced by chemical pre-treatment and filtration by 32 μm mesh size, i.e., 4-log reduction of FC at a UV fluence of 10 mJ/cm^2.

- The low UV fluence requirements of the proposed treatment train may result in a reduced cost for the treatment of a high wastewater flow, with reduced UV equipment and operating costs, providing municipalities with a smaller and compact treatment unit for CSO.
- The double exponential microbial inactivation model applied in this study, well predicted the FC inactivation kinetics with an R^2 of 0.98, for all the treatments tested. This model, used in combination with the regression models developed in our previous work (Venditto et al., 2020), makes the treatment adaptable to different CSO water quality and quantity, to pursue multiple treatment objectives with a single treatment process.
- The SWMM simulations showed a considerable environmental efficacy of the UV disinfection enhanced by chemical pre-treatment and filtration by 32 μm mesh, i.e., TSS removal of 73% and 4-log reduction of FC.
- The cost analysis performed herein suggests that the proposed treatment train is competitive to current CSO treatment technologies and strategies in terms of cost-effectiveness.

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Chapter 5

5 Environmental impacts of CSO treatment: Stormwater management modeling study

5.1 Introduction

Since the extent and the frequency of CSO discharges is also function of land-use, rainfall and sewer network characteristics, the integration of the large (catchment scale) and the small scale (sewer network scale) in a comprehensive hydraulic model is required in order to predict and control CSO impacts (Field and Jr, 1972). Thus, the application of a dynamic rainfall-runoff-routing simulation model is necessary to simulate the sewer network response during a rainfall event in terms of pollutant loads and discharge volume (Freni et al., 2010; Lucas and Sample, 2015). A The Storm Water Management Model (SWMM) is one of the most effective models to simulate the response of sewer networks under several weather conditions and to assess overflow pollution (Warwick and Tadepalli, 1991) merging different scales. The SWMM has been successfully used in several projects to simulate short and long-term hydraulic sewer network response to the rainfall events. These projects showed that SWMM application is very useful on urban drainage flooding analysis (Akdoğan and Güven, 2016; Tsihrintzis and Hamid, 1998), water quality and transport of contaminants (Liu et al., 2010; Temprano et al., 2007). The SWMM allows users to manage, simultaneously, hydraulic and hydrological modules identifying pollutant sources and their impacts on the water quality. The hydrology module operates on the catchment areas that receive precipitation and generate runoff and pollutant loads (Tsihrintzis and Hamid, 1998). The model requires several input data such as the sub-catchment area, pervious and impervious areas with and without depression storage, width, slope and

Manning's roughness overland (Niaizi et al., 2017). The hydraulic module works on the sewer network tracking the quantity and quality of runoff generated within the catchment area, the flow rate into the system, flow depth, and quality of water. The input data used to describe pipes, manholes and outfalls are diameter, length, material, slope, Manning coefficient, and type of flow (stormwater or sanitary flow) (EPA, 2017).

5.2 Study area

In this study, the SWMM was applied to simulate the receiving body water quality impacts when a CSO occurs. Cavendish area, located to the north-west of London Ontario (CA), was used as a study area (Figure 6.1).

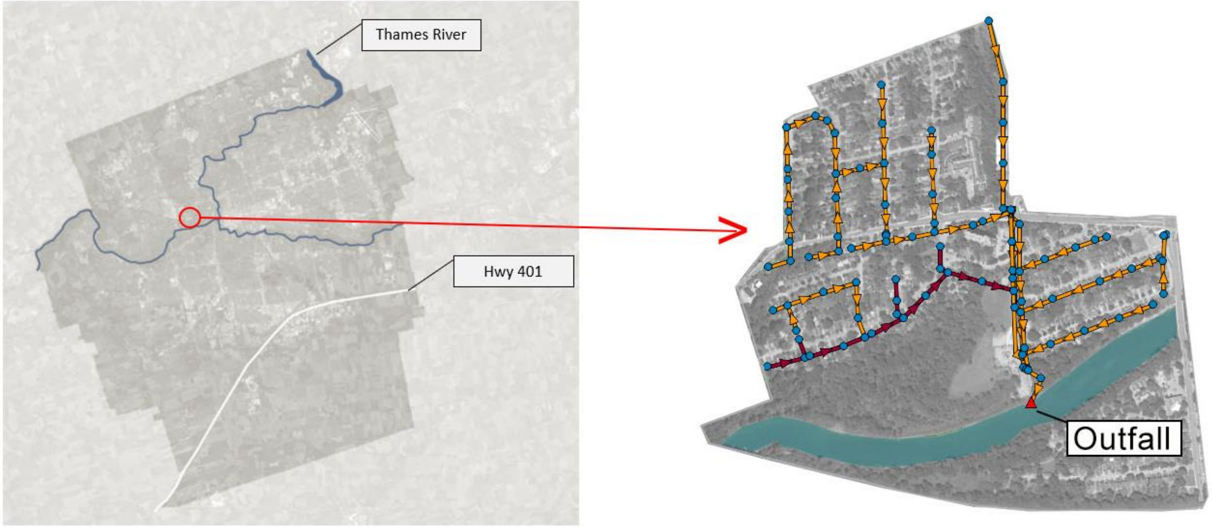

Figure 5-1: Location (left side) and discretization (right side) of the catchment area

The total catchment studied has an area of 41 ha and the land use is predominantly low-density residential area with remaining land use (about 30%) as open space. The studied area is characterized by a mixture of combined sewer drainage system and sanitary sewer system with a separate stormwater drainage network. The sewers consist mostly of circular concrete pipes the total length of the main sewers is 6.2 km, measured from the upstream to the overflow outfall point. The studied sewer network discharges through the outfall located in the south of the area directly into the Thames River.

5.3 Calibration and Validation

Table 6.1 summarizes the input data used for model calibration. An initial representation of the area was constructed using shapefiles from the municipality of the City of London and sanitary flows and infiltration flows were modeled based upon design data provided by the municipality. Details about land use and estimated population of the study area are reported in Figure S6.1 and Table S6.1 of Appendix B.

Table 5-1: Hydrologic and hydraulic input data added to the SWMM model

Hydrologic input data	***Hydraulic input data***
Slope and width	Pipe physical characteristics
Depression storage	Manhole physical characteristics
Pervious and impervious area (with and w/o depression storage)	Outfall physical characteristics
Manning’s roughness coefficient	Pump station characteristics
Soil infiltration capacity	

Raw rainfall data from October 2006 to December 2010 were acquired from local weather stations operated by the municipality (A.J. Taylor Operations Centre). Depth and flow velocities registered by three monitoring stations of Cavendish area were also provided by the City of London from October 2006 to December 2010. Rainfall data, depth and flow velocities data were used to generate 5-min intervals time-series input files for the modeling software and analyze the sewer network response.

To calibrate the SWMM, the aforementioned monitoring data were added into the model in tabular form. Quality Assurance/ Quality Control (QA/QC) analysis (Lai et al., 2007; Vallabhaneni et al., 2012) were carried out by sanitary sewer overflow analysis and planning (SSOAP) toolbox to ensure accuracy and reliability of observed data. Thus, the relationship between depth and velocity, and depth and flow rate of flow monitoring data was investigated by scatter plot. Data were considered consistent if a positive correlation among depth, flow and velocity data was identified. Specifically, an increase of the water-depth into the sewer network must correspond to an increase of flow and velocity; no inconsistencies were found at the end of this analysis, confirming the accuracy and reliability of the observed data (Figure S6.2 and S6.3 of Appendix B). Additionally, by analyzing observed flow and rainfall data, dry weather days and wet weather days were identified. To assess the reliability of flow and rainfall data, days were divided in weekday and weekend days. The average of weekday and weekend flow for the period recorded was calculated, expecting a different amount of flow (higher) during the weekend day (Nasrin et al., 2013). The average flow for weekdays and weekend-days was graphically compared by line chart; the weekend day flow path is greater and a little bit shift on the right than weekday flow path, validating the previous assumption and in agreement with the literature (Figure S6.4 of Appendix B).

To carefully investigate the sewer network response during rainfall events, the sewer network flow was analyzed in order to identify the amount of the base wastewater flow (BWF), groundwater flow (GWF) and rainfall derived infiltration and inflow (RDII) into the sewer network. As already reported in Chapter 2, Base Wastewater Flow (BWF) and Ground Wastewater Infiltration (GWI) are a consequence of dry weather flow. BWF is the residential, commercial, institutional, and industrial flow, collected from the sanitary sewer system and treated to the wastewater treatment plant (WWTP). GWI is the groundwater infiltration that enters the collection system through cracked pipes or deteriorated manholes when the ground surface is extremely saturated (EPA, 2017). During Wet Weather Flow (WWF), Rainfall Derived Infiltration/Inflow (RDII) is added to GWI and BWI. Rainfall inflow refers to the water that enters the sanitary sewer system through direct connections (e.g., roof and stormwater cross-connection); rainfall infiltration refers to the water that filters through the soil before entering the sanitary sewer system through damaged pipe sections, deteriorated manholes or connected foundation drain. RDII is the major component of peak wastewater flows during wet weather and it is typically responsible for overflows (Muleta and Boulos, 2008). The SSOAP toolbox provides automated identification of BWF, GWF, and RDII generating a simulated RTK hydrograph. The RTK hydrograph contains three types of hydrographs 1) short, 2) medium and 3) long responses, and it is based on three values: R, T and K. R is the fraction of rainfall that enters into the system, T is the time of onset of rainfall to the peak in hours, and K is the time to recession to the time to peak (Vallabhaneni et al., 2012). The SWMM requires the R, T, and K values as input data to calculate the amount of RDII flow that comes into the sewer network during a rainfall event. SSOAP toolbox helps to identify the best combination of R, T, and K values by visual curve fitting. The visual curve fitting is an interactive and visual approach by which users can define manually R, T, and K getting short, medium and long response curves (Gheith, 2010; Lai et al., 2007). Initial values of R, T, and K were selected based on pre-defined guidelines identified during the literature review and they were adjusted to reach a good fit between observed RDII flow and simulated RDII flow. Observed RDII flow and simulated RDII flow achieved a good visual comparison with the following R, T, and K values (Figure S6.5 of Appendix B):

- For the short response, R-value was 0.169, T-value was 0.5, and K-value was 15.
- For the medium response, R-value was 0.20, T-value was 3, and K-value was 3.
- For the long response, R-value was 0.30, T-value was 10, and K-value was 9.

From observed dry weather and wet weather days, the strongest rainfall event was identified and used for model calibration. The observed depth and the observed flow velocity from each monitoring station were compared with simulated depth and simulated velocity data for the monitoring station n.1, monitoring station n.2, and monitoring station n.3. To find the best fit between observed and modelled data, a sensitivity analysis was performed using pervious and impervious area, depression storage, width, slope, and Manning's roughness coefficient for each sub-catchment were used as sensitive parameters (EPA, 2016a). According to the sensitivity analysis, pervious and impervious area showed a greater impact on calibration results. Calibration results are reported in Appendix B. To assess the calibration reliability, the model was validated by using a different rainfall event and the model response was compared with the observed data. Validation results are also reported in Appendix B.

5.4 Environmental impacts of CSO treatment

The SWMM was used to simulate an overflow and analyze the benefits of treating CSO discharges on the receiving body water quality by the proposed treatment. For this purpose, the no-treatment scenario was compared with the best treatment scenario identified in chapter 4, such as UV disinfection (fluence 10 mJ/cm^2) enhanced by chemical pre-treatment and filtration by 32 μm mesh. The overflow impacts on the water quality of the Thames River were investigated in terms of TSS and FC concentrations. The deposit of TSS on the catchment (pollutants build-up) during dry weather and their movement from a catchment surface (pollutants wash-off) during dry wet weather was simulated using SWMM build-up and wash-off equations (EPA, 2016b). Since no TSS data were available from the municipality about pollutants build-up and pollutants wash-off, these parameters were estimated according to land use and obtained from the literature (Tu and Smith, 2018). FC was assumed to be co-pollutants of TSS in overland flow. The attachment fraction of FC with TSS was assumed to be 50% (Characklis et al., 2005; Wu et al., 2009). The concentration of TSS and FC coming from the sewer network was assumed to be the same as the collected samples.

Under wet weather conditions, an overflow occurred during a 3-hours rainfall event with maximum rainfall intensity of 50 mm/hr. The CSO lasted 6 hours (from 4:10 a.m. to 10:50 a.m.) and resulted in the discharge of 1213 m^3 of untreated water into the Thames River with an average flow rate of 44.42 L/s (Table 6.3).

Table 5-2: Overflow characteristics and pollutant load before and after treatment

	No treatment	**Microsieve-based filtration (32 µm) + UV dose 10 (mJ/cm^2)**
Duration Overflow (min)	440	
Overflow discharged volume (mc)	1213	
Flow rate average (L/s)	44.42	
Cumulative precipitations during overflow (mm) (mean intensity (mm/hr))	46.77 (0.61)	
Cumulative TSS discharged (g)	184955	50332
Cumulative FC discharged (counts)	1.05 x 10^{13}	1.24 x 10^{9}

The flow peaked after 1 hour from the beginning of the overflow reaching 102 L/s and the water discharged during the first 2 hours of overflow carried the highest amount of TSS and FC (Figure 6.2). As regards the solid matters, the high concentration of TSS at the beginning of the overflow may be due to the particles washed by the stormwater runoff on urban surfaces and the resuspended sewer sediments. On the other hand, the high concentration of FC during the first 2 hours of overflow may be related to the fraction of microorganisms attached to solid matters (Passerat et al., 2011b). These results suggest that a CSO can have considerable impacts on the water quality of the Thames River. Indeed, it is interesting to note that for a total of 1213 mc of water overflowed, a total of about 180000 g of TSS and 1.05 x 10^{13} FC were discharged into the river. However, Figure 6.3 clearly shows that 73% of TSS were removed by the treatment while the number of FC was at least 4 orders of magnitude higher than the number of FC observed in the treated effluents.

These results validate the effectiveness of the treatment as a key point for the improvement of the water quality of the Thames River.

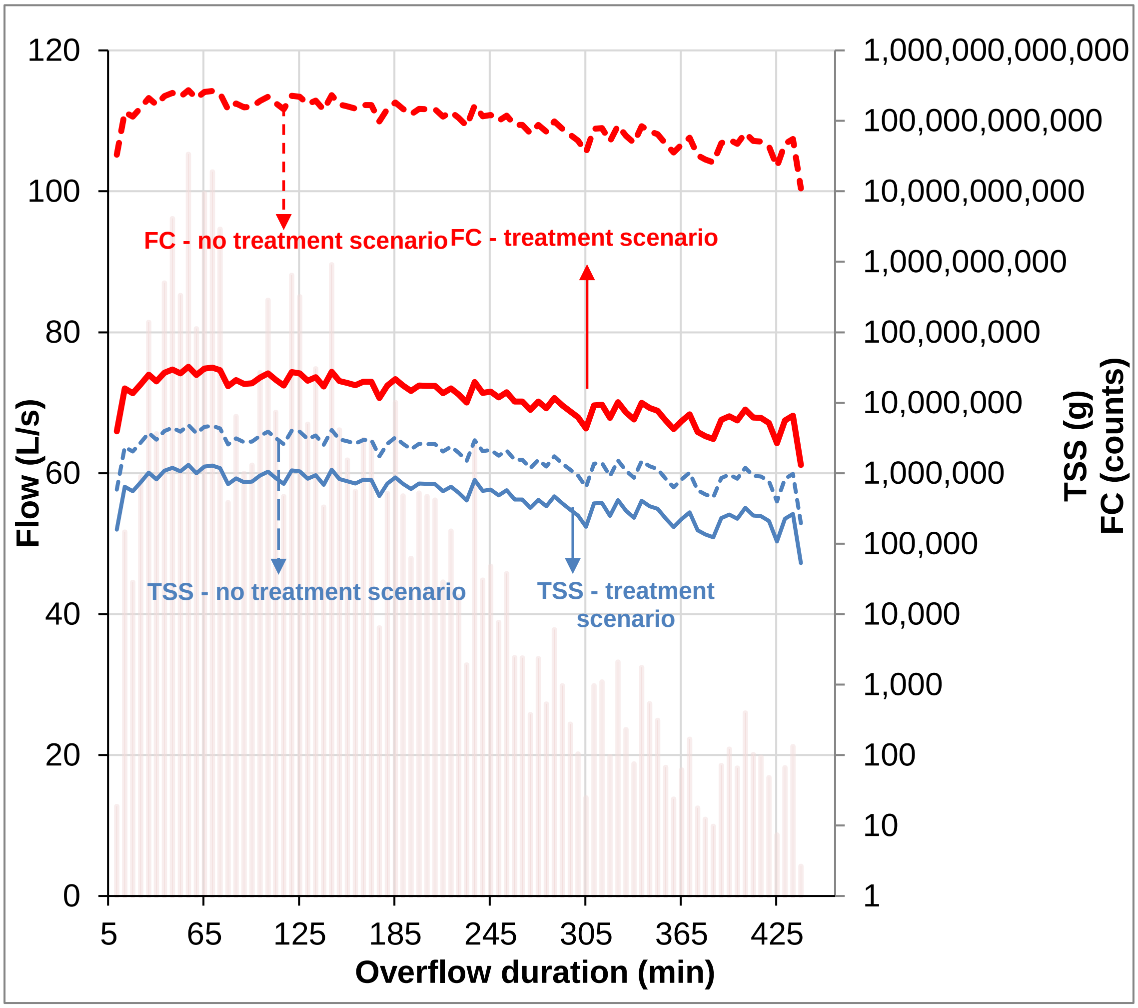

Figure 5-2: Variation of CSO parameters over time - Flow rate (orange bars); TSS (moving 5-min average, line) discharged into the Thames River during the overflow (blue); FC (moving 5-min average, line) discharged into the Thames River during the overflow (red).

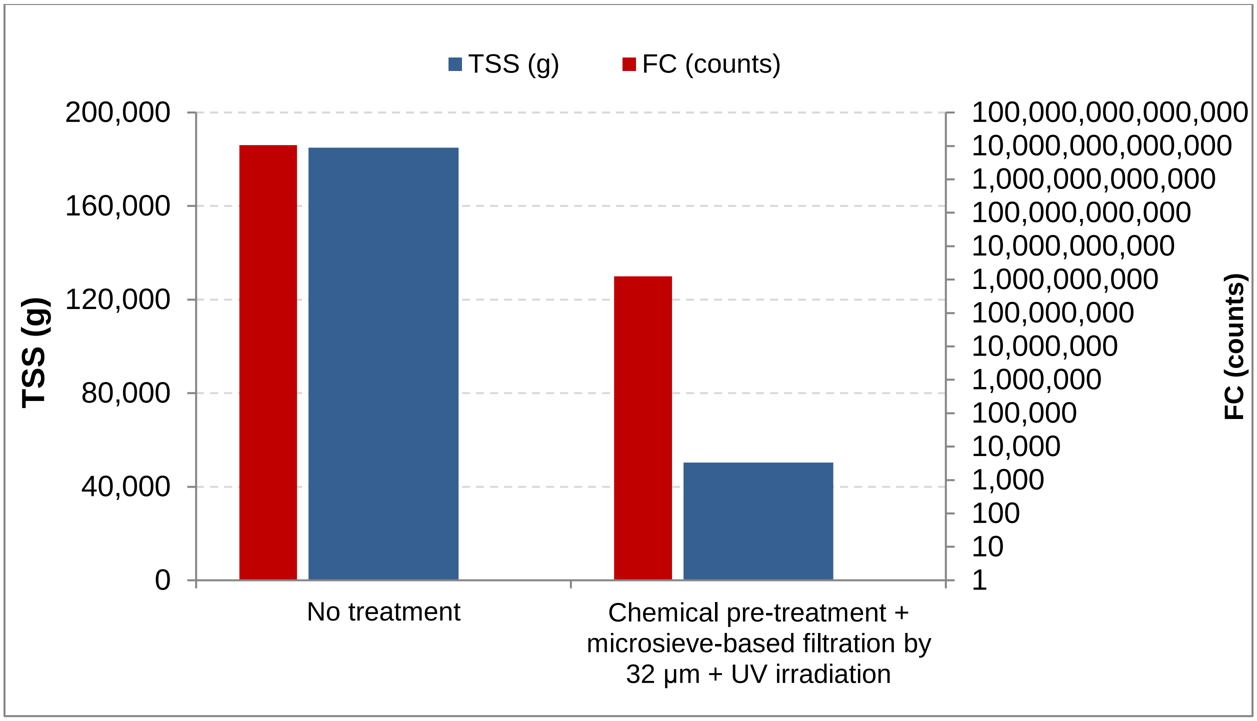

Figure 5-3: Comparison of the cumulative mass of TSS discharged into the Thames River before (red) and after (blue) treatment, and cumulative count of FC discharged into the Thames River before (red) and after (blue) treatment.

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Chapter 6

6 Conclusion and Recommendations

6.1 Conclusions

The detailed summary of the major findings of the various subprojects have been included in chapters 3-6. The principal findings of this study were:

- The developed microsieve-based filtration enhanced by low-dose chemical pre-treatment and followed by UV disinfection, still not explored in other studies, was able to deal with multiple contaminants and the associated impacts in the receiving bodies caused by CSO pollution and nutrients discharge in the environment.
- The developed treatment relies on multiple treatment agents combined in a single multifunctional process where fine particles, such as zeolite and powdered activated carbon, first adsorb soluble nutrient and are subsequently removed by polymer-enhanced microsieving allowing the removal of both soluble and particulate pollutants in a single treatment step. At the optimum dosage of treatment agents, about 72% of Turbidity removal, 65% of TSS removal, 56% of TKN removal, 35% of t-COD removal and 75% of TP were observed. Furthermore, this work describes an innovative method for the removal of ammonia via a dual mechanism of ammonia capture by zeolite adsorption, followed by zeolites removal by polymer coagulation and microsieving filtration.
- The UV disinfection showed better performance at lower UV fluence when enhanced by low-dose chemical pre-treatment and microsieve-based filtration. Indeed, 4-log inactivation of fecal coliform was obtained at fluence 10 mJ/cm^2. This result opens the possibility for municipalities to deploy smaller and compact

treatment units for CSO treatment able to treat large amount of flow quickly and effectively, reducing operating costs.

- The proposed treatment showed its advantages in terms of cost-effectiveness if compared with existing CSO treatments, sewer separation or storage tank applications.

6.2 Limitations

The developed microsieve-based filtration enhanced by low-dose chemical pre-treatment and followed by UV disinfection achieved high removal efficiencies. Thus, this treatment is promising for treating CSO discharges. However, one of the major limitations of this treatment is the low removal of soluble COD (<15%). Although the literature suggest that soluble COD removal increase with the increase of carbon dose due to an increase of active site available on the activated carbon for the adsorption of soluble COD, we preferred to keep a low-dosage of treatment agents to develop a cost-competitive CSO treatment. Moreover, in Chapter 4, the rate constants of particle-associated microbes K_d for all the treatment scenarios, may present a marginal error due to only two data points corresponded to UV Fluence 0 mJ/cm^2 and 10 mJ/cm^2.

6.3 Recommendations

The cost-effective microsieve-based filtration enhanced by low-dose chemical pre-treatment and followed by UV disinfection require further investigations. The following recommendations for future work are made:

- Experiments on a pilot scale are needed to quantify the performance and overall treatment cost of the proposed treatment.

- The amount of chemical sludge produced by the physico-chemical treatment has to be analyzed. Remedial measures to deal with chemical sludge must be investigated.
- Large-scale scenarios associated with centralized and decentralized CSO treatment strategies should be investigated in order to quantify the performance of the developed treatment in terms of environmental and economic sustainability.
- Zeolite regeneration methods should be investigated, and the efficiency of regenerate zeolite should be tested. The economical advantages of the regeneration of the zeolite should be assessed.

Appendices

Appendix A: Supplementary material of Chapter 3

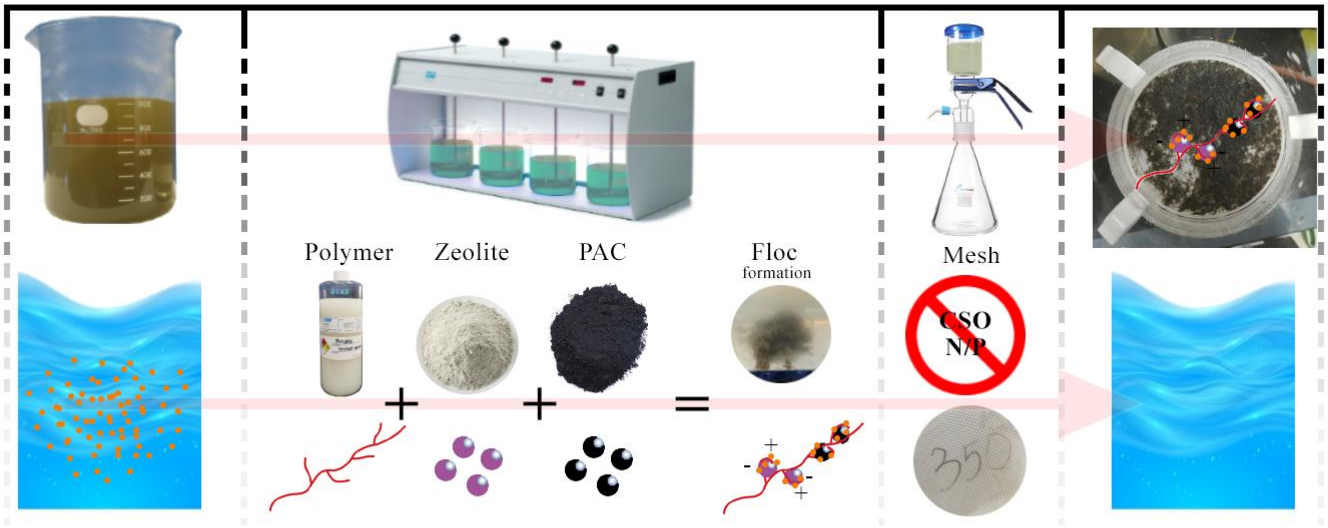

Figure S3. 1: Graphical abstract of work presented in Chapter 3.

Table S3. 1: Treatment results obtained by testing different combinations of mesh size, Polymer, Zeolite, and PAC dosages.

Treatment agents					**Pollutants (after treatment)**				
Run	**Polymer (mg/L)**	**Zeolite (mg/L)**	**PAC (mg/L)**	**Mesh µm**	**Turbidity (NTU)**	**t-COD (mg/L)**	**s-COD (mg/L)**	**TKN (mg/L)**	**TP (mg/L)**
1	1	0	250	350	91.4	565	340	31.5	8.8
2	3	0	250	350	54.5	456	319	30.5	7.7
3	1	5000	250	350	114	581	324	21.7	10.2
4	3	5000	250	350	55.3	450	321	21.8	7.3
5	2	2500	0	158	62.4	453	337	25.2	8.1
6	2	2500	500	158	64.6	451	326	24.7	8
7	2	2500	0	500	62.4	458	334	26.5	8.2
8	2	2500	500	500	62.6	501	304	26.5	8.2
9	1	2500	250	158	84.9	498	318	25.2	8.9
10	3	2500	250	158	50.8	420	314	24.3	7.3
11	1	2500	250	500	89.6	411	322	23	8.6
12	3	2500	250	500	52.15	438	318	26.2	7.2
13	2	0	0	350	54.1	447	330	30.5	7.9
14	2	5000	0	350	65.4	458	334	22.1	8.8
15	2	0	500	350	58.5	478	311	32	8.2
16	2	5000	500	350	66.3	501	300	20.4	8.1
17	2	0	250	158	56.2	437	313	32.2	8.1
18	2	5000	250	158	56.5	428	311	20.8	7.9
19	2	0	250	500	58.9	456	332	34	8
20	2	5000	250	500	61.1	460	310	21.7	7.9
21	1	2500	0	350	83	496	323	25.7	9.1
22	3	2500	0	350	54.1	422	312	25	8.2
23	1	2500	500	350	77	513	297	25.7	8.9
24	3	2500	500	350	50	423	299	24.8	7.5
25	2	2500	250	350	61.5	461	304	22.4	8.2
26	2	2500	250	350	59.2	462	296	21.7	8.1
27	2	2500	250	350	59.5	489	304	21.9	8

Table S3. 2: Summary of the analysis of variance: linear and two-way interactions of treatment agents on turbidity, p-COD, s-COD, t-COD, TKN and TP removal. The table shown p-value, standard error of the coefficients (SE coeff) and variance inflation factor

	Turbidity			**p-COD**			**s-COD**			**t-COD**			**TKN**			**TP**		
Source	*p-Value*	*SE Coef*	*VIF*	*p-Value*	*SE Coef*	*VIF*	*p-Value*	*SE Coef*	*VIF*	*p-Value*	*SE Coef*	*VIF*	*p-Value*	*SE Coef*	*VIF*	*p-Value*	*SE Coef*	*VIF*
Polymer	0	0.756	1	0.002	2.13	1	0.246	0.813	1	0.002	1.31	1	0.945	0.381	1	0	0.248	1
Zeolite	0.097	0.756	1	0.444	2.13	1	0.206	0.813	1	0.75	1.31	1	0	0.381	1	0.178	0.248	1
PAC	0.925	0.756	1	0.028	2.13	1	0.002	0.813	1	0.288	1.31	1	0.785	0.381	1	0.261	0.248	1
Mesh	0.661	0.756	1	0.74	2.13	1	0.975	0.813	1	0.762	1.31	1	0.119	0.381	1	0.902	0.248	1
Polymer*Zeolite	0.158	1.31	1	0.526	3.68	1	0.372	1.41	1	0.755	2.27	1	0.556	0.659	1	0.007	0.43	1
Polymer*PAC	0.9	1.31	1	0.645	3.68	1	0.516	1.41	1	0.821	2.27	1	0.937	0.659	1	0.338	0.43	1
Polymer*Mesh	0.818	1.31	1	0.112	3.68	1	1	1.41	1	0.154	2.27	1	0.045	0.659	1	0.865	0.43	1
Zeolite*PAC	0.811	1.31	1	0.667	3.68	1	0.456	1.41	1	0.865	2.27	1	0.111	0.659	1	0.134	0.43	1
Zeolite*Mesh	0.897	1.31	1	0.6	3.68	1	0.324	1.41	1	0.854	2.27	1	0.656	0.659	1	0.865	0.43	1
PAC*Mesh	0.89	1.31	1	0.317	3.68	1	0.348	1.41	1	0.526	2.27	1	0.772	0.659	1	0.766	0.43	1

Table S3. 3: Treatment alternatives based on different treatment objectives. Treatment agents have been normalized by the influent pollutant loads

	Removal (%)				**Design**				**N. of frontiers per design**
	Turbidity	*t-COD*	*TKN*	*TP*	*Polymer (mg/g of TSS)*	*Zeolite (mg/g of TKN)*	*PAC (mg/g of COD)*	*Mesh (μm)*	
O-1	72	35	36	75	1.79	3592	149	370	4
O-2	72	40	57	75	1.85	9339	75	475	3
O-3	80	42	57	76	3.32	7902	149	180	2

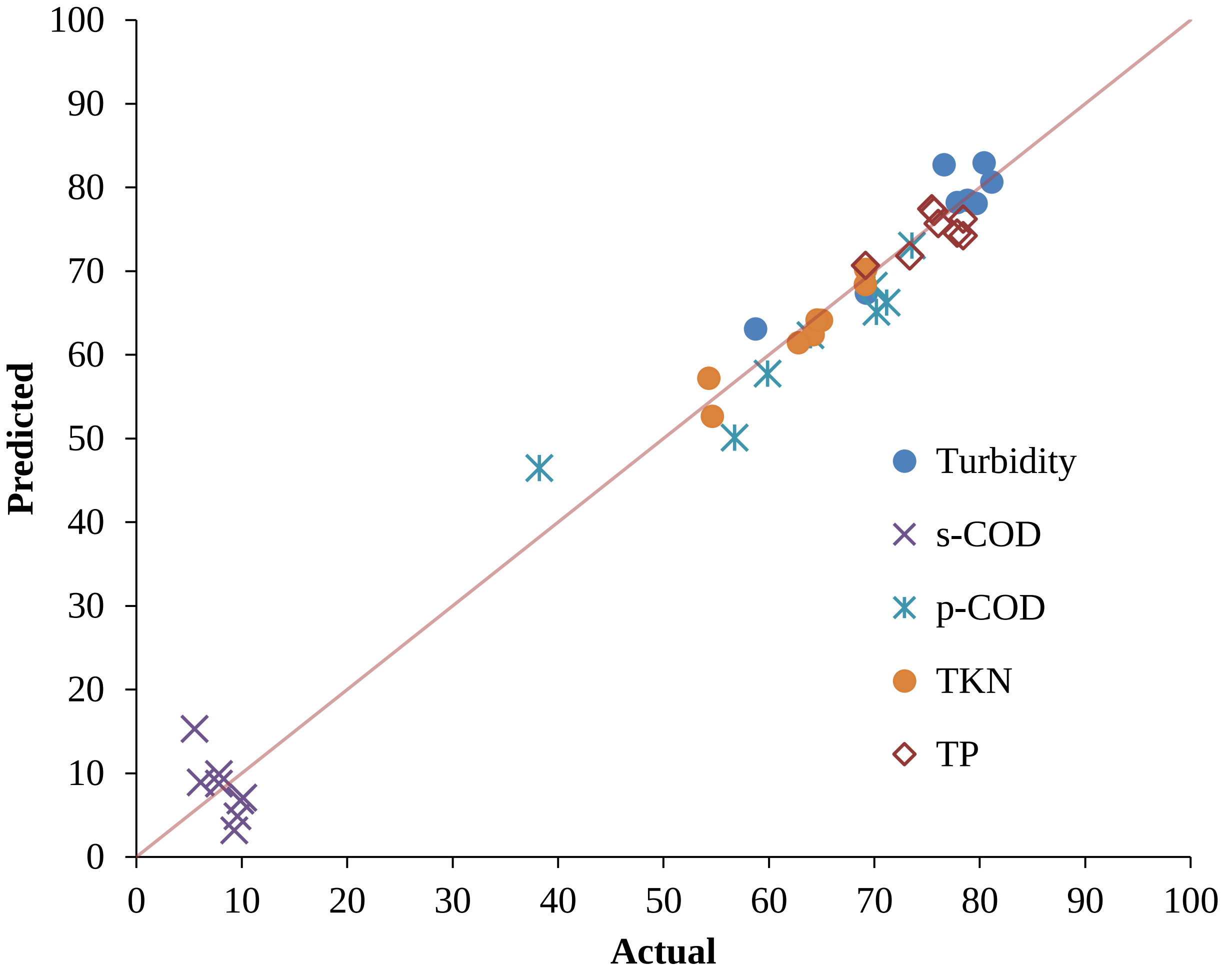

Figure S3. 2: Predicted (% removal) vs. actual values (% removal) plot for turbidity, TP, p-COD, s-COD, and TKN. Results obtained from cross-validation model.

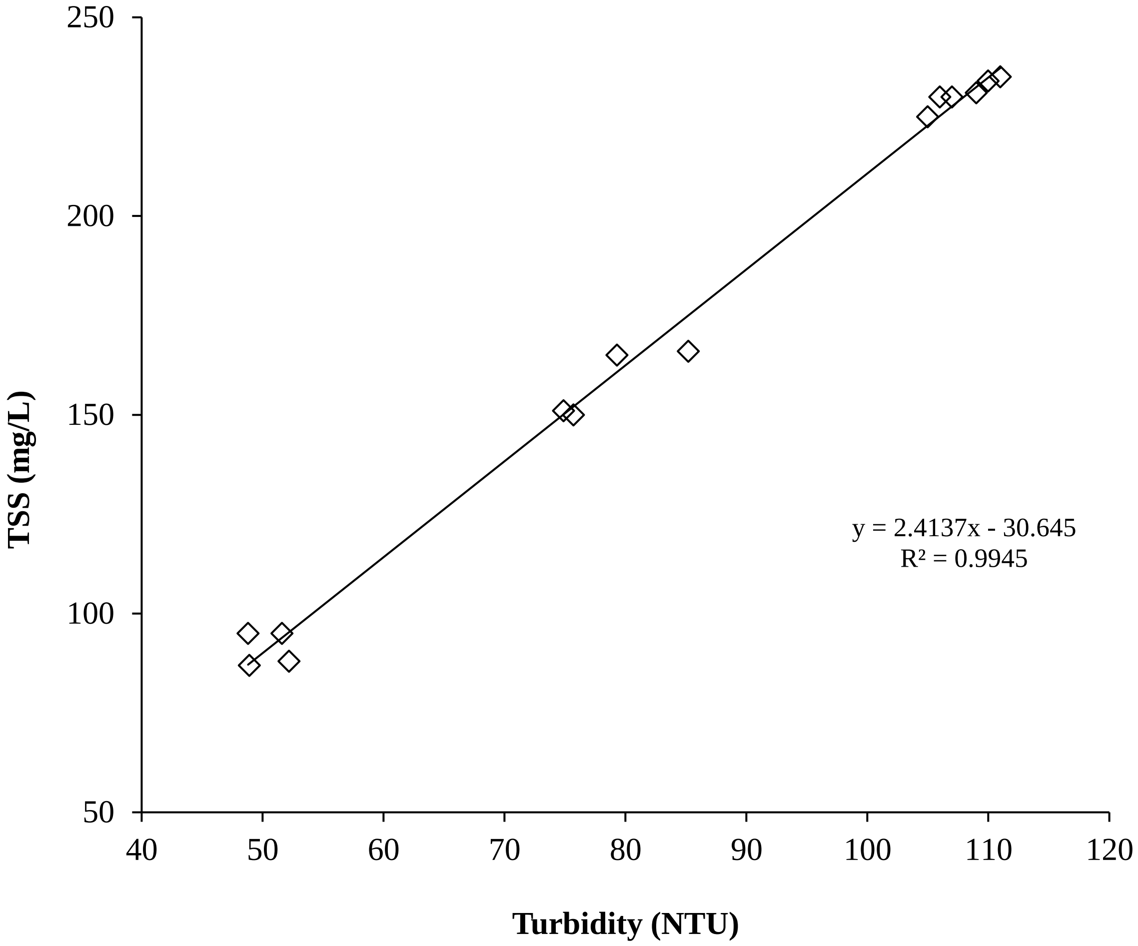

Figure S3. 3: Correlation plot between Turbidity and TSS.

Appendix B: Supplementary material of Chapter 4

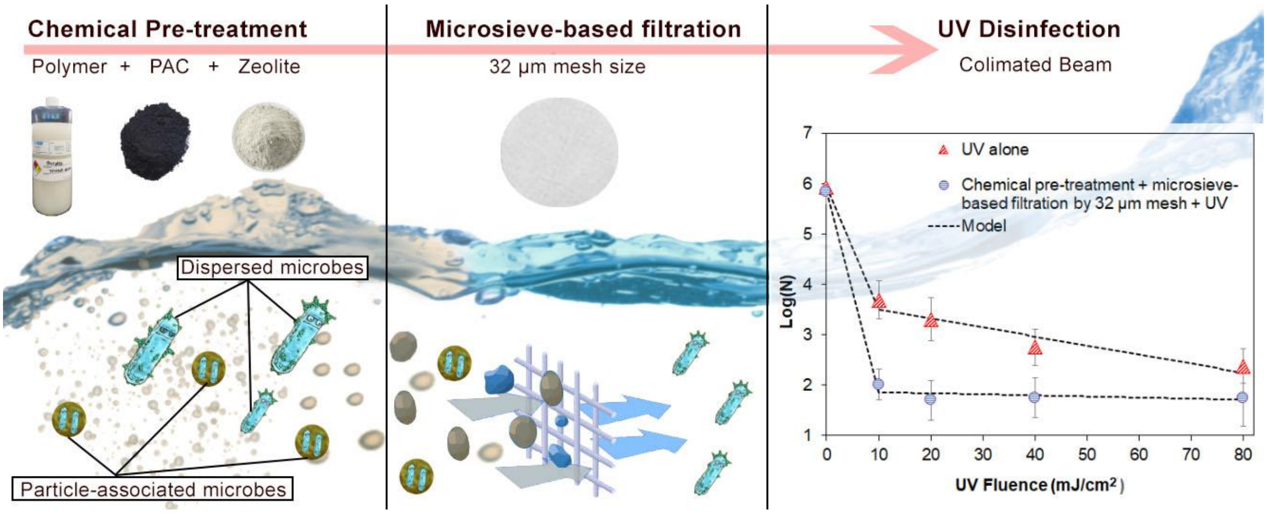

Figure S4. 1: Graphical abstract of work presented in Chapter 4.

Appendix C: Supplementary material of Chapter 6

Figure S6. 1: Land Use of the study area

Table S6. 1: Land Use and estimated population based on "Design Specifications & Requirements Manual" of the City of London

Land Use Type	**Area (ha)**	**Population Density**	**Estimated Population**
Residential – Low Density	86.5	3 persons/unit	2.500
Green area	13.5	26.4 persons/ha	357
Total	*100*	-	*2.857*

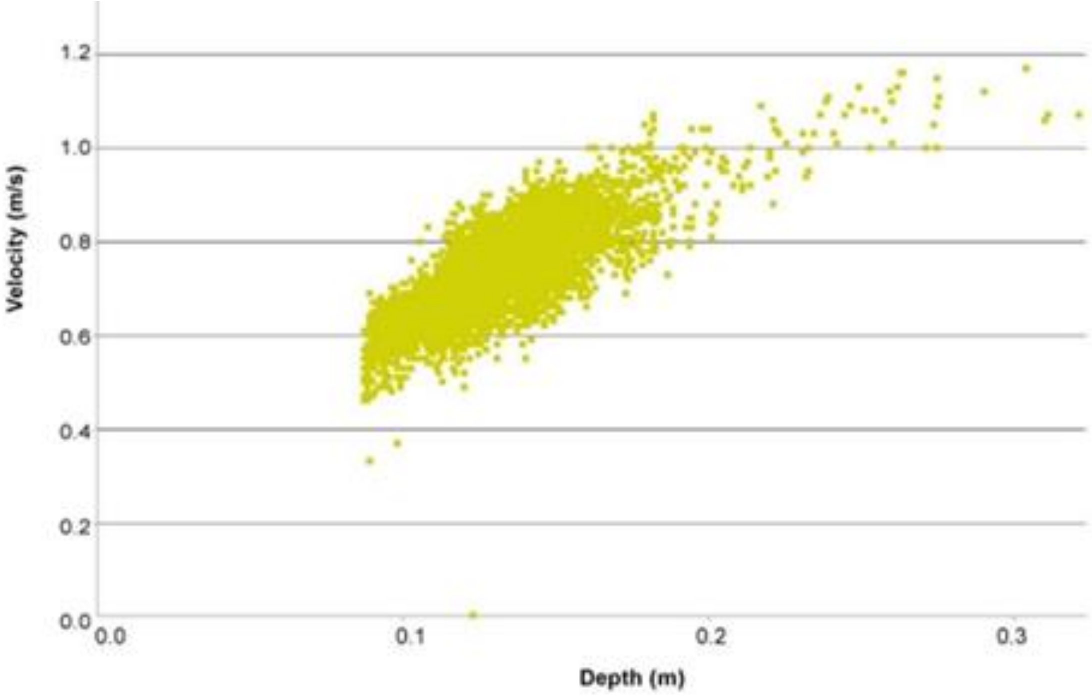

Figure S6. 2: Scatter graph showing the relationship between velocity and depth into the sewer network during the rainfall event

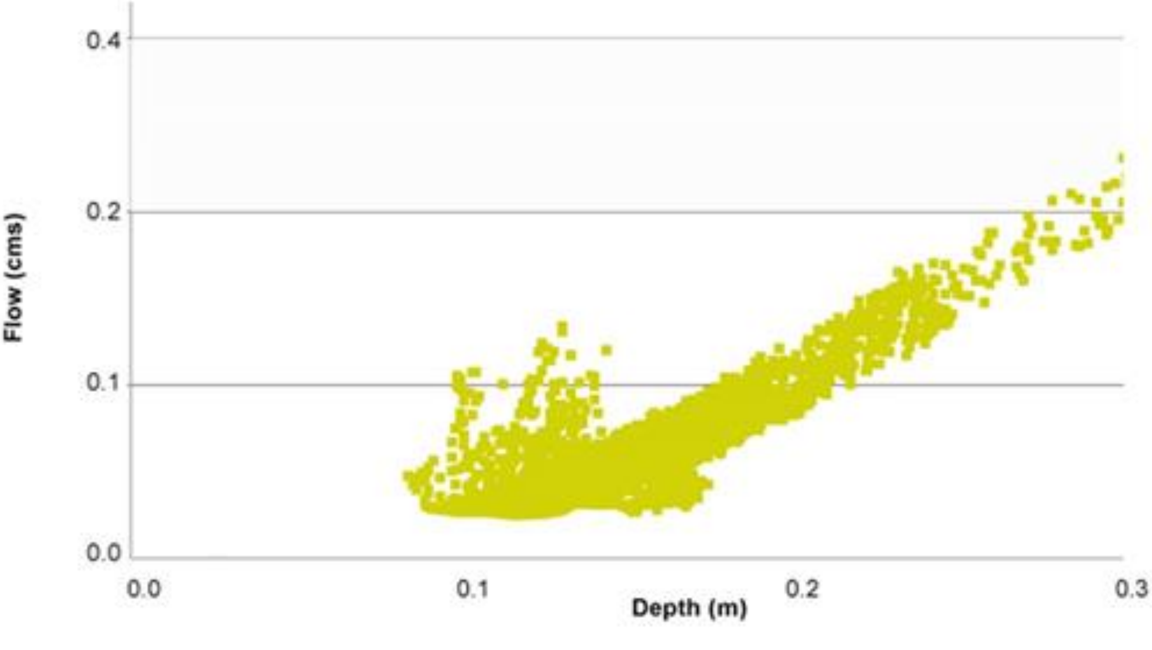

Figure S6. 3: Scatter graph showing the relationship between flow and depth into the sewer network during the rainfall event

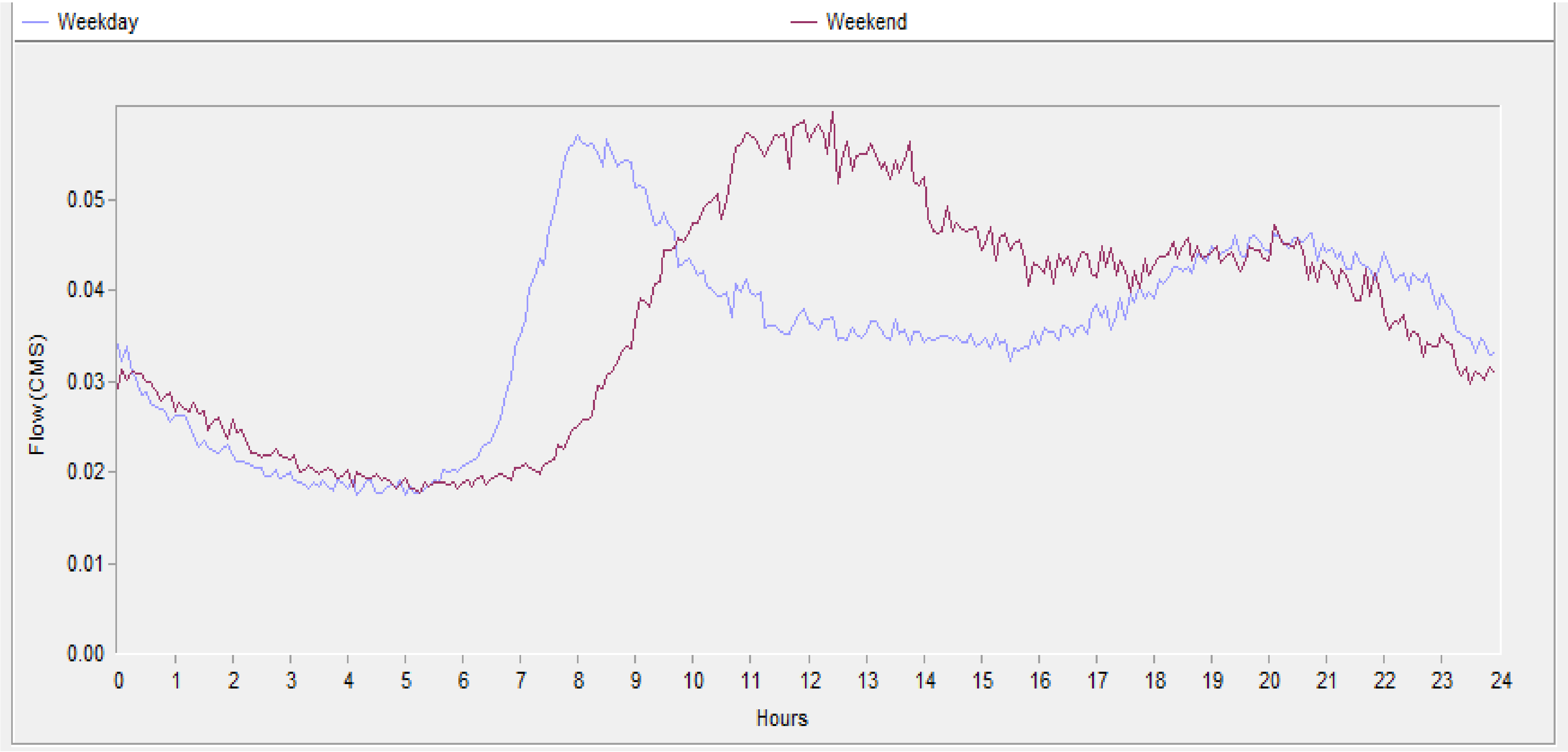

Figure S6. 4: Dry weather hydrographs showing the flow path of the weekday and weekend day average flow

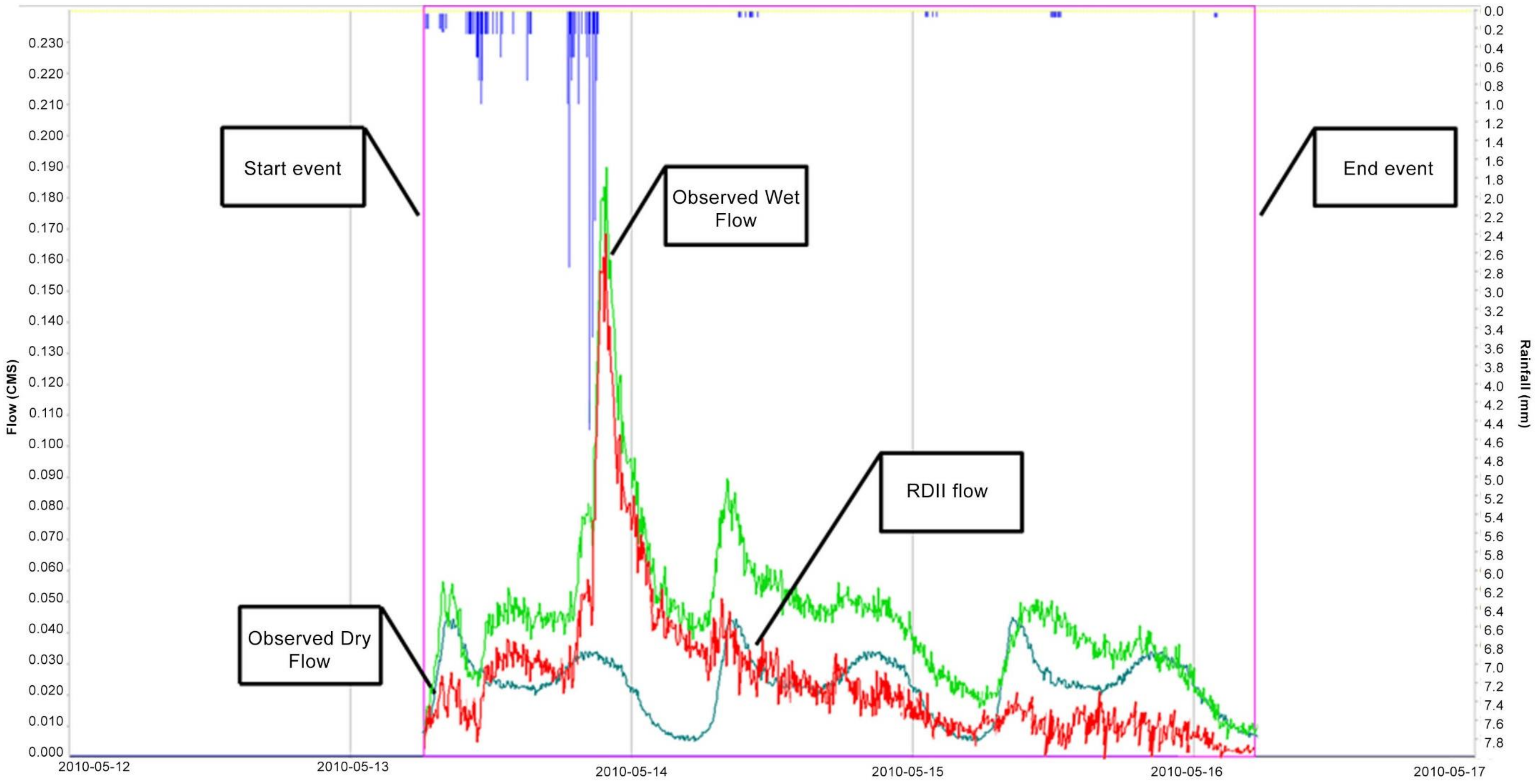

Figure S6. 5: RDII graph showing the flow characterization into the sewer network during wet weather period: Observed wet flow (light-green line), Observed dry flow (dark-green line), Simulated RDII flow (red line).The histogram (purple columns) represents the height of rainfall (mm). The beginning and end of the rainfall event is marked by the pink square shape.

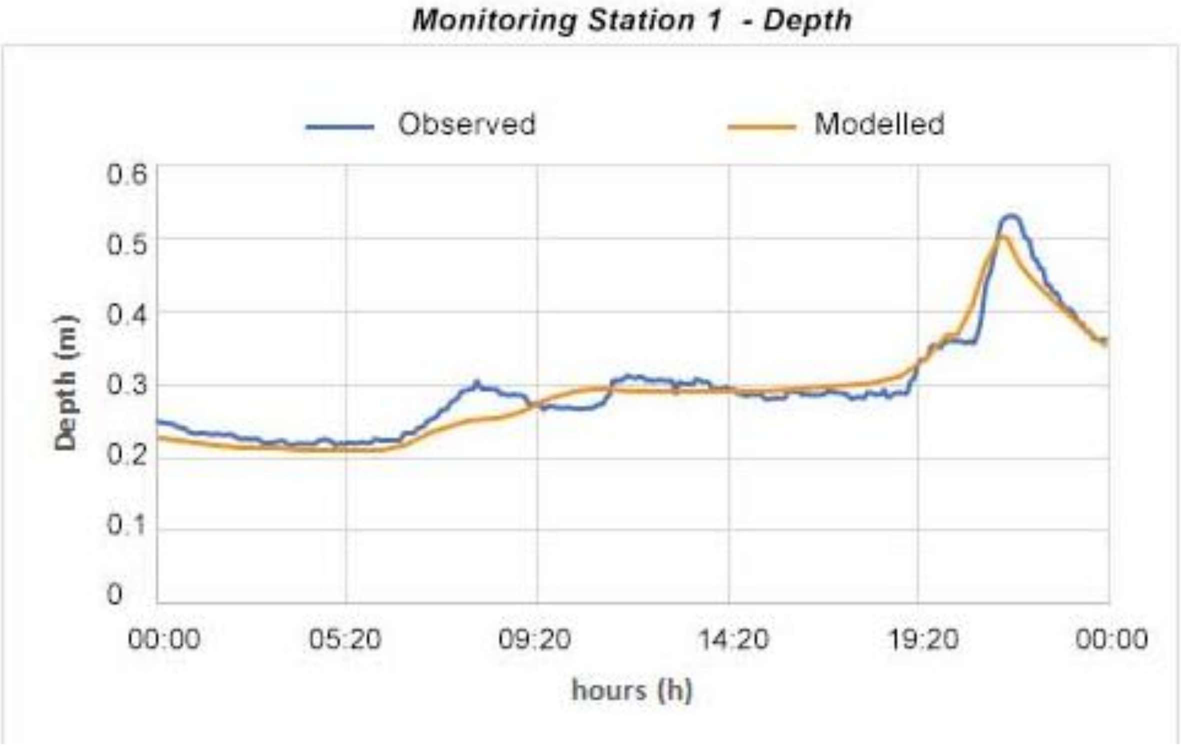

Figure S6. 6: Calibration results in terms of depth for monitoring station n.1

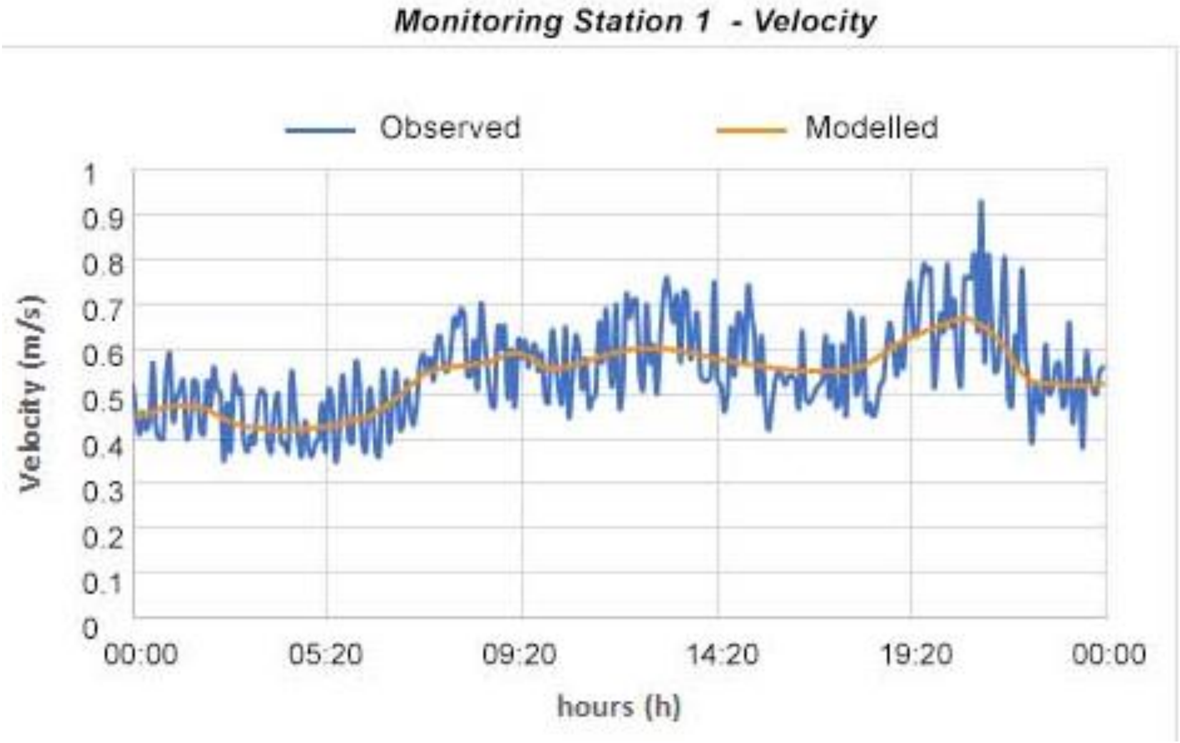

Figure S6. 7: Calibration results in terms of velocity for monitoring station n.1

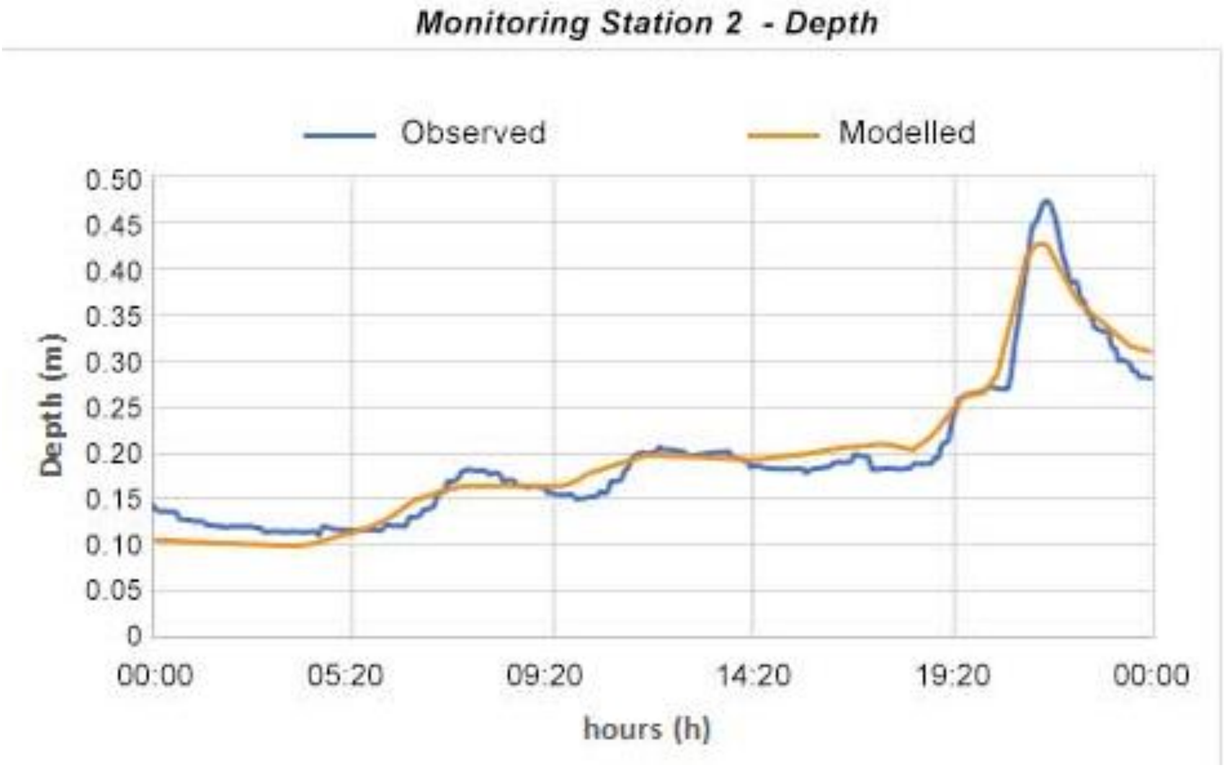

Figure S6. 8: Calibration results in terms of depth for monitoring station n.2

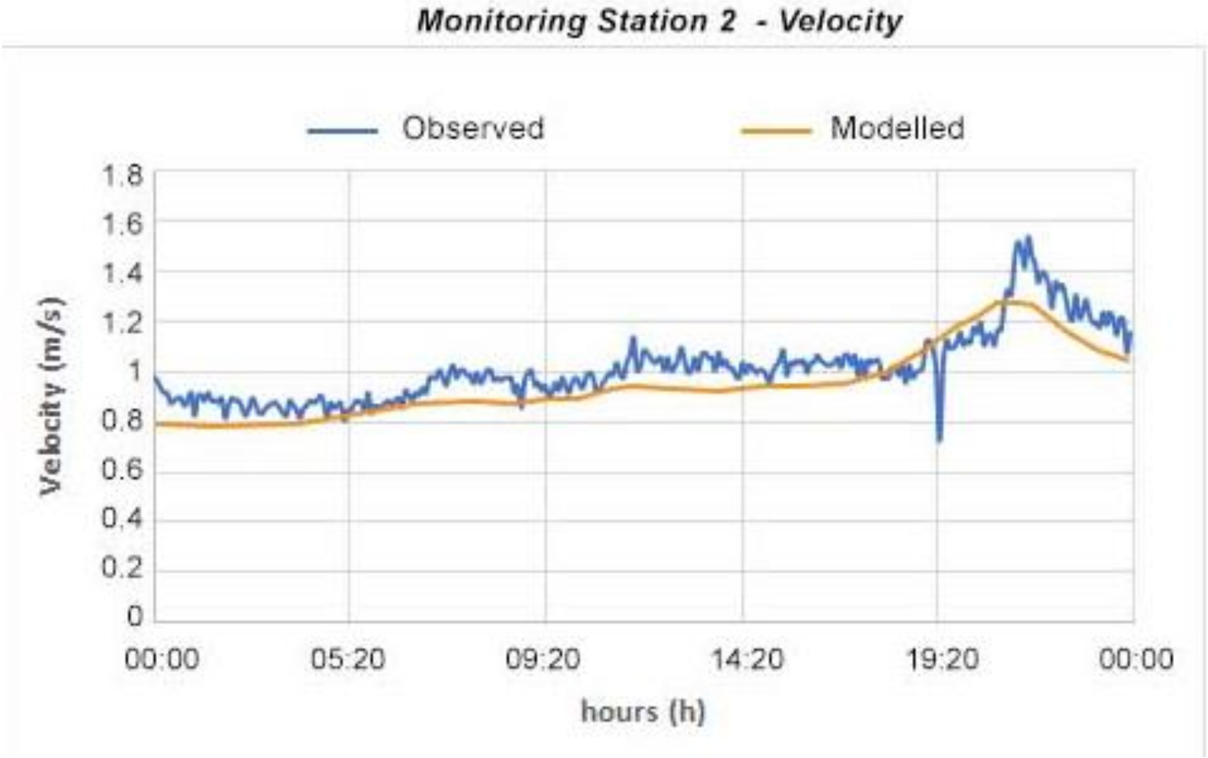

Figure S6. 9: Calibration results in terms of velocity for monitoring station n.2

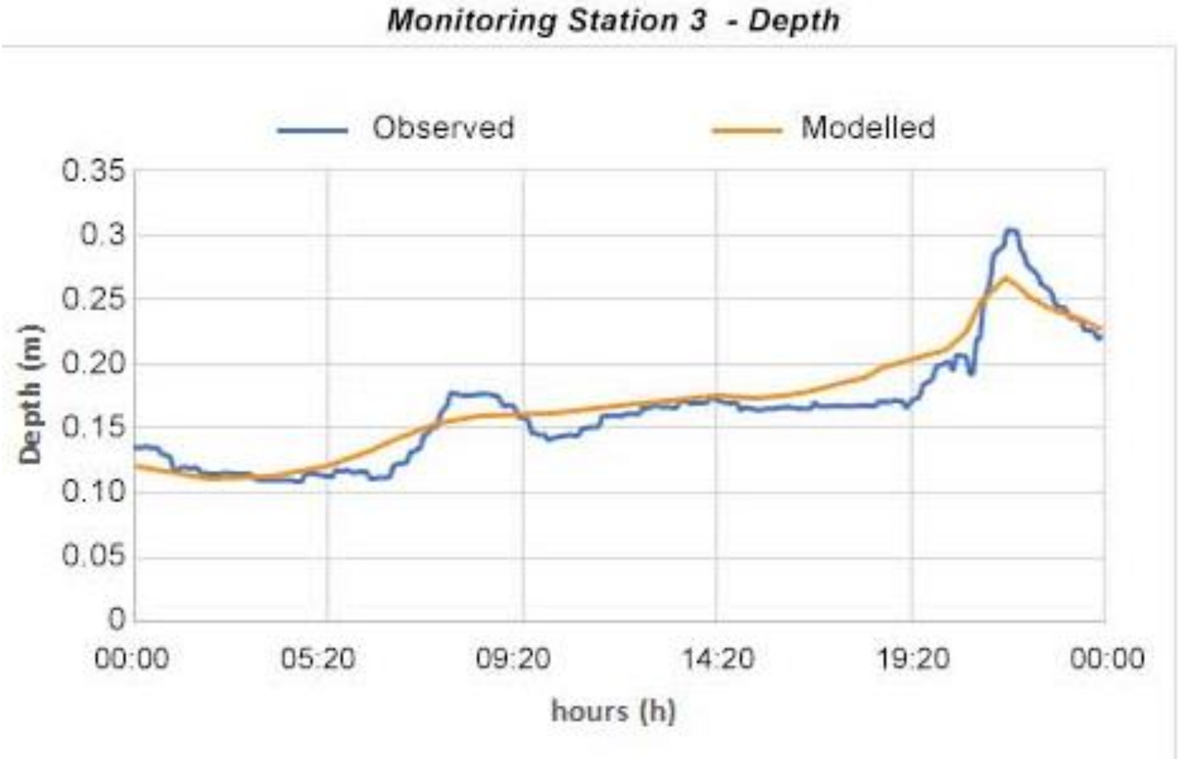

Figure S6. 10: Calibration results in terms of depth for monitoring station n.3

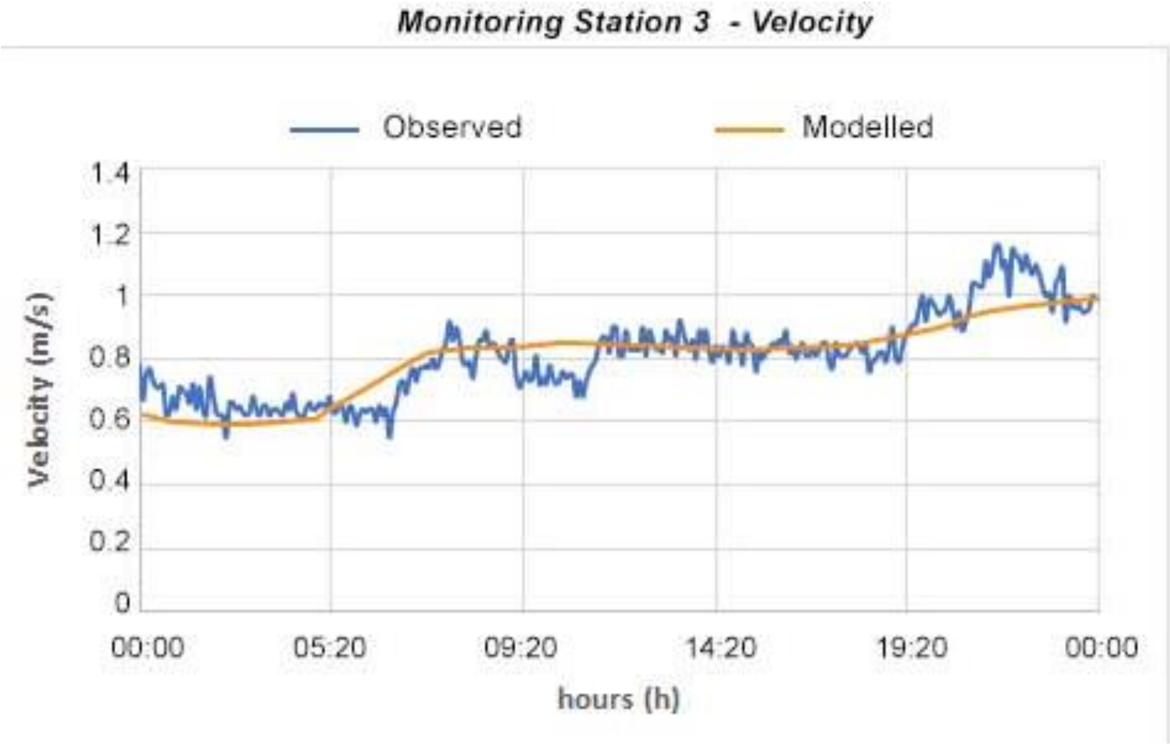

Figure S6. 11: Calibration results in terms of velocity for monitoring station n.3

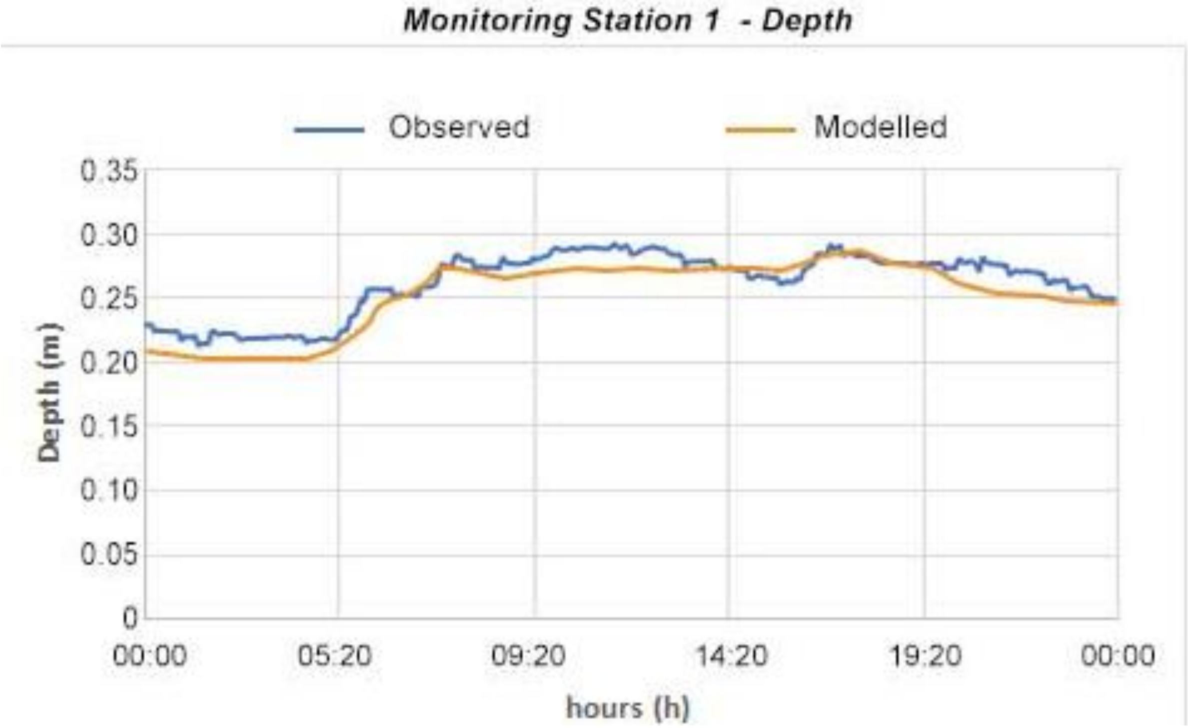

Figure S6. 12: Validation results in terms of depth for monitoring station n.1

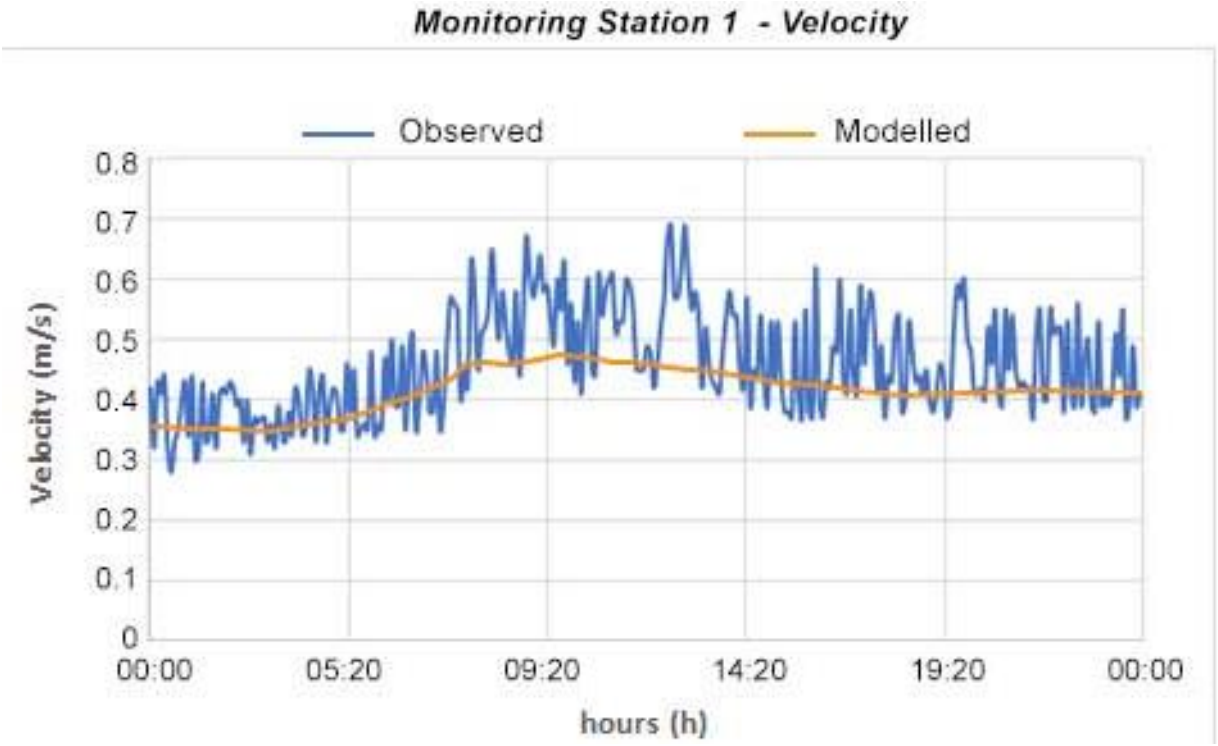

Figure S6. 13: Validation results in terms of velocity for monitoring station n.1

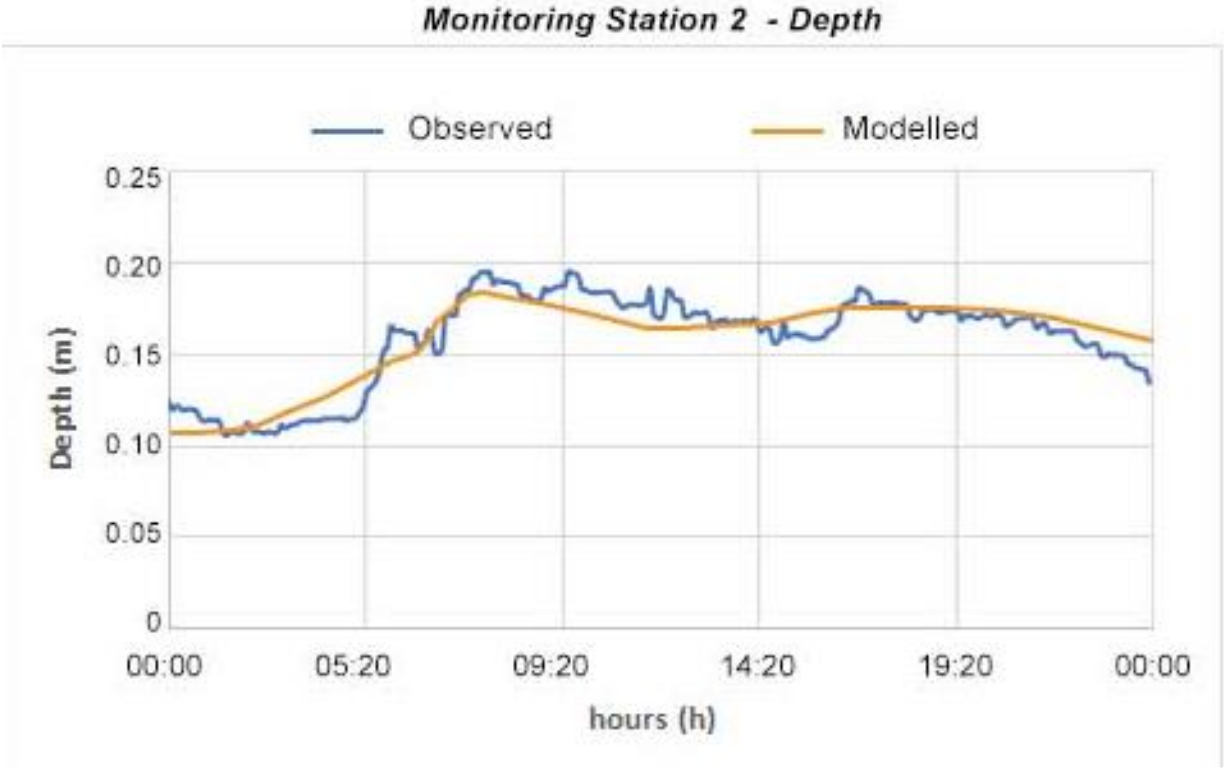

Figure S6. 14: Validation results in terms of depth for monitoring station n.2

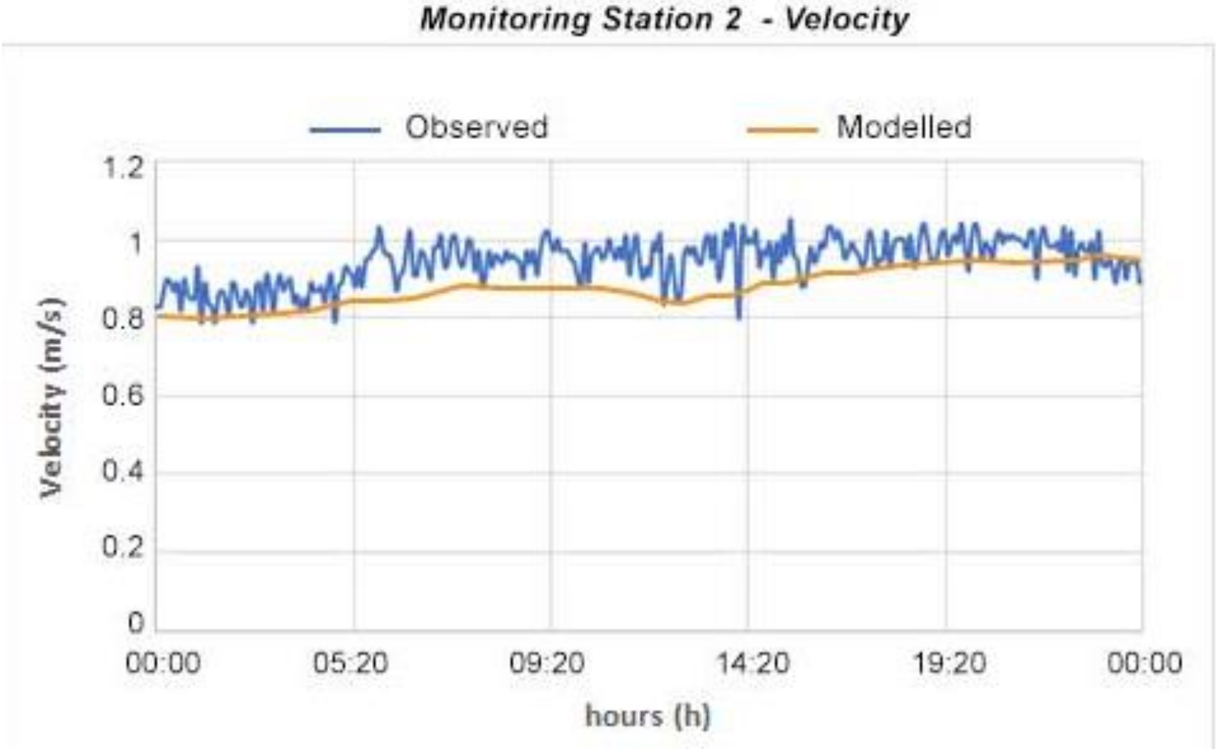

Figure S6. 15: Validation results in terms of velocity for monitoring station n.2

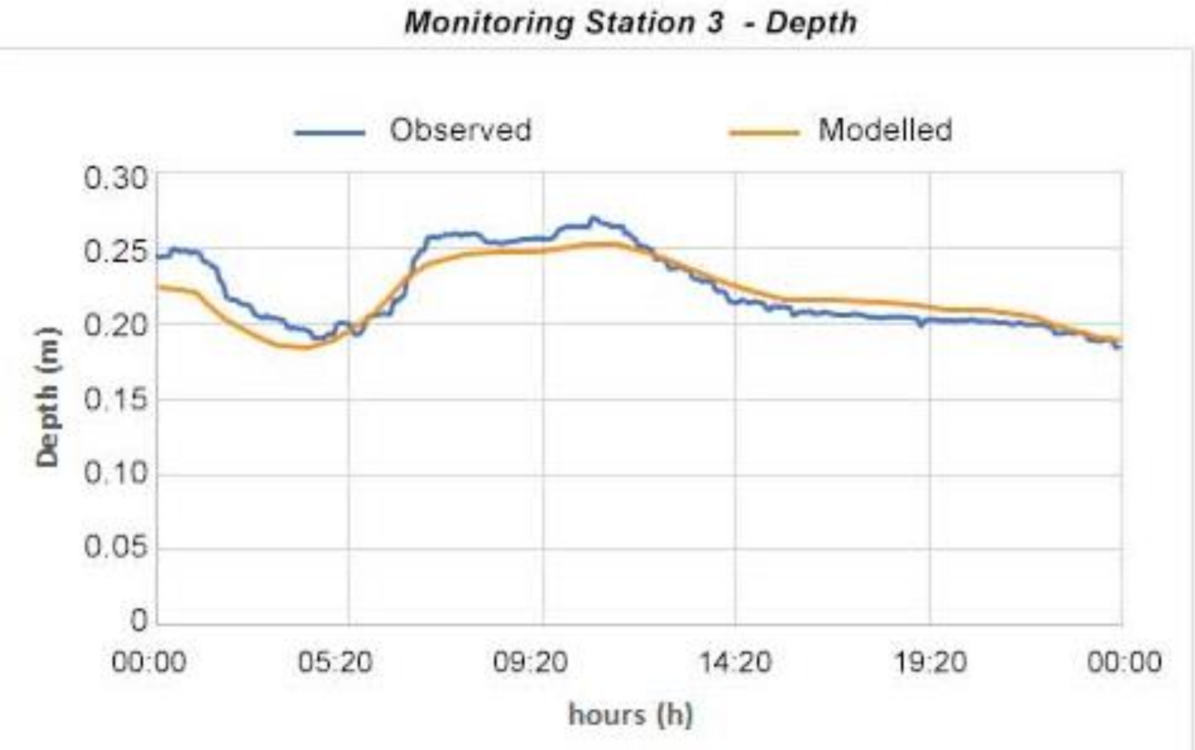

Figure S6. 16: Validation results in terms of depth for monitoring station n.3

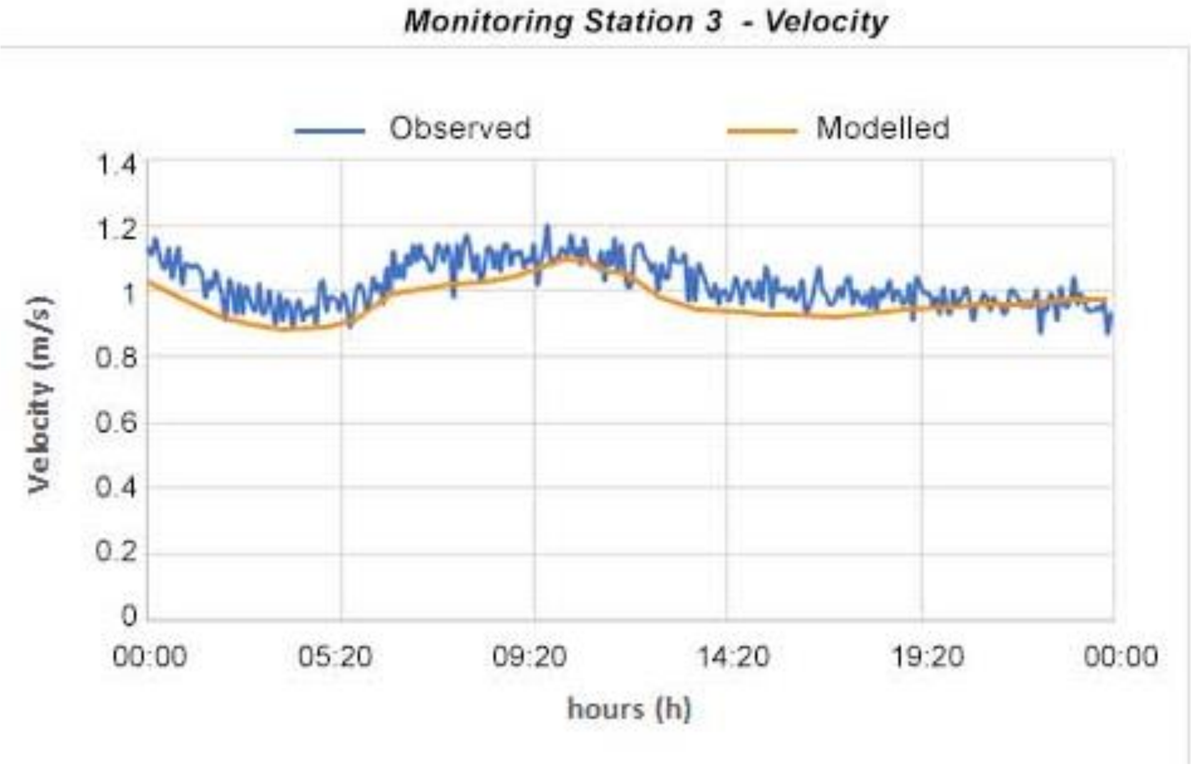

Figure S6. 17: Validation results in terms of velocity for monitoring station n.3

Appendix D: Preliminary treatment assessment

In the last decade, different strategies were developed to control CSO discharge including 1) source controls reducing the flow directed to the combined sewers, 2) conveyance controls storing or delaying the flow of excessive amounts of stormwater, and 3) end-of-pipe controls developing water treatments or adopting physical separation methods at the end of a flow conveyance system or outfall. When choosing the treatment configuration for CSO wastewater several factors should be considered. The most important criteria are related to the quality of the wastewater and the amount of overflow wastewater. It is also essential to verify the cost-effectiveness of the alternatives proposed to overcome CSO challenges. To date, limited information is available for the cost of CSO control strategies. In the Long-Term Control Plan (EPA, 2007a), it was reported that the cost of sewer separation is one of the most expensive approaches. In Randolph (VT), 2,660,000 USD had been spent to separate 95% of its combined sewers, while in Seaford (DE), 2,200,000 USD were used to separate approximately 40 percent of its combined sewer system. Under the Metropolitan Council's Environmental Servicies Division (MCES), the cities of St. Paul, South St. Paul, and Minneapolis spent 331,000,000 USD to complete a 10-year sewer separation program (EPA, 2007b). In 2009, the City of Quebec (Quebec, Canada) adopted a CSO Long-Term Control Plan (LTCP) planning to install 14 storage tanks and one tunnel, for a total storage volume of 45 million gallons, and a cost of 89,812,500 USD (with an exchange rate of 0.72 CAD/USD) (Olivier Fradetz et al., 2011).

In appendix L of the Long-Term Control Plan, EPA estimated a default value for different CSO treatment strategies. For chemical flocculation, a cost of 40,000 USD for every million gallons treated/day using aluminum additive, and 1,030,000 USD for for every million gallons treated/day using ferrous sulfate was estimated. Reynolds et al. (Reynolds et al., 1981) compared three different CSO control alternatives: 1) CSO storage control strategy, 2) transport and treatment of overflows, and 3) screening and disinfection treatment. It was pointed out that the most cost-effective method for CSO control was decreasing the amount of overflow by storage tank while transporting the remaining flow to the treatment plant for secondary treatment.

Based on direct contact with the vendors, the price per ton of cationic polymer, PAC and Zeolite were 1,000 CAD, 400 CAD and 300 CAD, respectively. A preliminary cost assessment for the treatment proposed in this study led to a cost of about 14.5 USD of powdered activated carbon, 32 USD of polymer (with an exchange rate of 0.74 CAD/USD), and 400 USD of zeolite for every million gallons of wastewater treated/day. A cost of 45,455 –190,000 USD per million gallons of wastewater treated/day (with an exchange rate of 1.09 EUR/USD) can be estimated for filtration and disinfection treatment based on previous studies (Iglesias et al., 2010). Moreover, it is worth noting that the proposed treatment could be exploited to deal with multiple CSO contaminants and the associated impacts on the receiving bodies (Venditto et al., 2020). This new approach, still not explored in other studies, has a great potential to address CSO and nutrient pollution with a single capital upgrade, which may be very important for municipalities facing CSO challenges. Therefore, it can be concluded that the proposed treatment train is an alternative to current solutions, which is competitive in terms of cost-effectiveness compared with sewer separation or storage tank applications. It is worth noting that most of the data available and reported above were derived from research conducted on a laboratory scale, and the operational/maintenance costs were not considered. Consequently, further experiments on a pilot scale are needed to quantify the overall treatment cost associated with the proposed treatment.

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Venditto, T., A.K.Ray, M.Ponzelli, D.Santoro, Treatment strategies for combined sewer overflows in multifunctional reactors. 9th International Young Water Professionals Conference, June 23-27, 2019, London, ON, Canada. (presenting author: Tiziana Venditto)

Venditto, T., A.K.Ray, M.Ponzelli, D.Santoro, Bluewater in wet weather: SWMM, RBF and UV disinfection to address the CSO impact. IWA Specialized International Conference, ecoSTP18, June 25-28, 2018, London, ON, Canada. (presenting author: Tiziana Venditto)

Venditto, T., A.K.Ray, F. Daynouri-Pancino, D.Santoro, Bluewater in wet weather: SWMM, RBF and UV disinfection to address the CSO impact. AquaHaking, United for Lake Erie, October 10, 2017, Waterloo, ON, Canada. (presenting author: Tiziana Venditto)

Venditto, T., Ponzelli, M., Sarathy, S., Ray, A.K., Santoro, D., 2020. *A microsieve-based filtration process for combined sewer overflow treatment with nutrient control*: Modeling and experimental studies. Water Res. 170, 115328. https://doi.org/10.1016/j.watres.2019.115328