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Ultrafiltration as urban wastewater tertiary treatment for water reuse at semi-industrial scale

Jiaqi Yang

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Laboratoire de Modélisation, Mécanique & Procédés Propres (UMR 7340)

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Docteur de l'École Centrale de Marseille
Spécialité : Génie des Procédés

Ultrafiltration as urban wastewater tertiary treatment for water reuse at semi-industrial scale

Présentée et soutenue publiquement par

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Affidavit

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Jiaqi YANG



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Table of Contents

TABLE OF CONTENTS	1
INTRODUCTION	1
CHAPTER 1. STATE OF THE ART	6
1.1. Definition of water reuse from urban wastewater, applications and regulations.....	7
1.1.1. Interest and definition of water reuse	7
1.1.2. Water reuse applications and guidelines	8
1.1.3. Focus on the pollution by microplastic particles	13
1.2. Membrane-based technology for water reuse.....	14
1.2.1. Performances of Membrane-Based Treatment Processes for Municipal Water Reuse	15
1.2.2. MF/UF-based Treatment Processes After Secondary Treatment of WWTP	17
1.2.3. MBR-Based Treatment for Water Reuse.....	26
1.2.4. NF/RO/FO-Based Treatment Processes for Water Reuse	31
1.2.5. Water Recovery and Energy Consumption of Membrane-Bases Processes for Water Reuse.....	37
1.2.6. Summary about membrane-based processes for water reuse	38
1.3. UF membrane fouling control and management by different types of backwashes	41
1.4. Thesis statement	46
CHAPTER 2. MATERIALS AND METHODS	48
2.1. Feed water quality	49
2.2. Lab-scale filtration equipment	49
2.3. Semi-industrial pilot plant.....	51
2.4. Membrane information in UF pilot plant	53
2.5. Membrane cleaning	54
2.6. Filtration conditions	56
2.7. Hydraulic filtration performance of UF membrane.....	58
2.7.1. Permeability	58
2.7.2. Fouling resistances	58
2.7.3. Fouling Reversibility.....	59
2.7.4. Water recovery rate	60
2.7.5. Impact of air backwash on irreversible resistance increase	60

2.7.6. Fouling removal efficiency of backwashes	61
2.8. Water quality assessment	61
2.8.1. Microplastics	61
2.8.2. Analysis of other parameters	65
CHAPTER 3. OPTIMIZATION OF UF OPERATING CONDITIONS FOR WASTEWATER	
REUSE	68
3.1. Introduction	69
3.2. Comparison of membranes' performances in lab-scale	72
3.2.1. Flux decline	72
3.2.2. Permeate quality	73
3.2.3. Fouling resistance	74
3.3. Understanding of UF feed variability and potential impact on UF performance in pilot-scale.....	75
3.4. Impact of filtration time and flux on hydraulic performances in pilot scale	78
3.4.1. Overview of permeability variations with CB and AB	78
3.4.2. Impact of filtration time and flux on fouling reversibility and hydraulic performances	79
3.5. Impact of air backwash frequency	84
3.6. Hydraulic filtration performance in long-term in optimized conditions	87
3.7. Conclusion	89
CHAPTER 4. IMPACT OF UF ON WATER QUALITY	90
4.1. Water quality analysis for reuse purpose	91
4.2. Detection of microplastic particles (MPs) in WWTP effluent and removal by UF.....	94
4.2.1. Selection of rinsing water for MP analysis.....	94
4.2.2. Selection of scanning pixel resolution of microscopy	95
4.2.3. Digestion protocols	97
4.3. Conclusion	101
CHAPTER 5. IMPACTS OF NAOCL-ASSISTED BACKWASH ON UF FOULING	
MANAGEMENT AND TOTAL ECONOMIC COSTS.....	103
5.1. Impact of chlorinated-assisted backwash and air backwash on ultrafiltration fouling management for urban wastewater tertiary treatment	104
5.1.1. Permeability variation	104
5.1.2. Fouling resistance.....	108
5.1.3. Fouling reversibility by physical backwashes	111
5.1.4. Turbidity and TOC removal rates by backwash	115

5.1.5. The influence of NaOCl concentration on air backwash efficiency of condition NNNY	118
5.1.6. Discussion	119
5.1.7. Conclusion.....	120
5.2. Capital expenditure and operational expenditure of semi-industrial scale UF for water reuse	121
5.2.1. Economic cost in operating condition J60t60BW1/3	121
5.2.2. Impact of different NaOCl addition on economic cost.....	127
CONCLUSIONS AND PERSPECTIVES	130
REFERENCES	137
LIST OF TABLES	159
LIST OF FIGURES	161
APPENDIX I :TECHNICAL STRATEGY	163
APPENDIX II: PUBLICATIONS AND CONFERENCES.....	164
ULTRAFILTRATION AS URBAN WASTEWATER TERTIARY TREATMENT FOR WATER REUSE AT SEMI-INDUSTRIAL SCALE -ABSTRACT.....	168
REUTILISATION DE L’EAU PAR ULTRAFILTRATION EN TANT QUE TRAITEMENT TERTIAIRE D’EAUX USEES URBAINES A L’ECHELLE SEMI-INDUSTRIELLE - RESUME	168

Introduction

Water reuse is an enduring topic that benefits the society and future generations of mankind. Qualified reuse water not only reduces the occurrence and spread of diseases, but also contributes to the environment protection. In Europe, approximately 2.4% of the total treated wastewater effluents was reused in 2015 [1], while France only reused around 1% [2]. As the reuse proportion differs a lot among different countries and regions, water reuse market is still young and limited which needs to be highly improved in the future. In this work, water reuse from urban wastewater only, will be considered. Among urban wastewater treatment, primary treatment mostly with physical processes can remove approximately 50–70% of total suspended solids (TSS) and 25–50% of biological oxygen demand after 5 days (BOD₅) [3]. The secondary treatment combines biologic and chemical processes to remove the soluble organic matter and the residual TSS [4]. However, the secondary effluent is limited in reuse applications with risks for human health and public environments [5]. To be reused, the secondary effluent needs tertiary and advanced treatments which are considered as the advanced level of treatment and mostly include membrane processes, advanced oxidation processes, disinfection or any combination of them to remove chemical and biologic constituents. Nowadays, more and more advanced treatment technologies for municipal wastewater reuse have emerged, and membrane filtration is one of the main unit technologies. Ultrafiltration (UF) has been accepted as one of the most cost-effective technology to deal with secondary effluent on non-potable reuses, such as agricultural irrigation, landscape irrigation, urban reuse, and process water for industries [6–9]. UF enables high removal efficiency on TSS, turbidity, organic matters, and microorganisms, etc [6,10]. Based on scientific consensus and best available evidence, the guidelines from the World Health Organization (WHO) and the French water reuse standards can be both taken into account [11,12]. However, regulations are becoming stricter in terms of concentrations of *E. coli*, BOD₅, TSS and turbidity such as in the higher reclaimed water quality class (A) of the recent regulation (EU) 2020/741 of the European Parliament and of the Council of 25 May 2020 on minimum requirements for water reuse [13]. Therefore, there is a need to confirm if UF can meet these new standards and to define for the first time a range of effective operating conditions.

Indeed, additionally, membrane fouling limits the economical operation of UF process on wastewater treatment [14]. To control fouling, filtration conditions (flux, filtration time, cleaning) need to be optimized because they can directly affect either the formation and nature of the fouling layer, or the removal mechanism of the foulants. Chemical washing is effective

on irreversible foulants removal through the reaction between fouling and chemicals. The choice of chemical agents, mostly using acids, alkalis, or oxidants, is based on the membrane materials and fouling types [15]. The conventional chemical cleaning plays an important role on irreversible fouling separation by chemical reagents soaking/reacting with membrane fouling which can damage the foulant-foulant and membrane-foulant interactions [16]. However, using of high concentrated acids, soda, and/or oxidants/disinfectants and longer soaking time will cause irreversible damages gradually on membrane properties and filtration performances in long-term operation [15,17,18]. Moreover, physical cleanings such as classic backwash (CB) and air assisted backwash (AB) are commonly used methods in membrane fouling control [19]. CB can loosen and detach fouling cake partially from membrane surface, and AB is a more effective anti-fouling method compared to CB [20–22]. In this case, a chemically assisted maintenance backwash, the combined physical backwash with lower concentrated oxidants/disinfectants (compared to chemical cleaning) in backwash water, has been developed for permeability maintenance improvement recently, so as to reduce the need for intensive chemical cleanings [23,24]. In result, it is therefore interesting to alternate cleaning by CB and AB during filtration and it is very important to study the optimum backwash frequency of AB and CB with or without chlorine addition as it has never been done on municipal secondary wastewater effluent. Additionally, the consumption of capital expenditure and operational expenditure provides a direct reference to the industrial scale establishment. Therefore, it is necessary to estimate the economic budget of the project before production and application in industrial scale.

The aim of this thesis is to fully assess the performances of a semi-industrial UF pilot plant as an urban wastewater tertiary treatment for water reuse in terms of: (i) optimization of filtration operating conditions, (ii) permeate water quality, (iii) membrane fouling control and management through a combination of potentially chlorinated CB and/or AB, and (iv) brief calculation of total costs. The drawn conclusions are believed to be applicable for practical operation of semi-industrial UF systems. This subject aims to make comprehensive consideration of the impact conditions on UF system in order to achieve the most economical and efficient filtration and qualified water production. A brief overview of each chapter in the thesis is described below. It is important to note that some parts of this work are synthesized in 3 research articles and one review paper whose references are given in Appendix II.

Chapter 1 gives a comprehensive literature review. It first discusses water quality levels for different reuse applications. The state of the art then mainly provides a comprehensive summary of the membrane-based treatment processes, mainly focused on the hydraulic filtration performance, water quality as respect to water reuse guidelines, reuse purpose, fouling resistance potential, resource recovery and energy consumption. Various types of membrane technologies (microfiltration, ultrafiltration (UF), nanofiltration, reverse and forward osmosis) have been used successfully in centralized and decentralized waste reuse applications. The desired end use of the treated water dictates the degree of water quality and influences the membrane process necessary to achieve that goal and in this work, UF is the most suitable. Chapter 1 also assesses the efficiency of a potential combination of different types of backwashes for UF including classic backwash, air backwash and chlorinated backwash. Finally, this chapter raises the issue of the pollution by microplastic particles. Indeed, as microplastic contamination is one of the world's most pressing environmental concerns in oceans, rivers, and plants, and health concerns in animal and human bodies [25–27], it is necessary to study the microplastics fate in wastewater treatment plant effluent for water reuse.

Chapter 2 introduces the methodology, equipment, and materials used in this thesis together with the detailed information of manufacture, testing institute, and testing process. This chapter summarised the materials, analytical methods, and comparative conditions for the experiments from Chapter 3 to Chapter 5.

Chapter 3 is mainly focused on the optimal operating conditions for sustainable filtration performance. The operating parameters to be optimized include flux, filtration time and backwash sequence which consists of a combination of CB and AB during filtration. The target of this study is to find out the optimized filtration conditions that can be long-term applied on secondary wastewater treatment for water reuse with guarantee of adequate permeate quality, sustainable hydraulic filtration performance, and high net permeate production. 15 different filtration conditions are designed to identify the effects of flux, filtration cycle time, and backwash sequences on hydraulic filtration performance. The filtration performances will be described through the analysis of hydraulic filtration performance, fouling composition and reversibility, water recovery rate, and the impact of AB frequency on irreversible fouling control.

Chapter 4 shows the results of water quality before and after the UF system. The permeate quality, analysed several times and during different seasons, will be compared to the water reuse

guidelines or standards of the World Health Organization (WHO), France, and the recent EU regulation. In addition, the coronavirus Sars-CoV-2 which causes the Covid-19 pandemic and the microplastic particles which become new threats for humans are also tested.

Chapter 5 investigates the influence of chlorine (NaOCl) addition in backwash water on backwash cleaning efficiency and filtration performances to better control the UF membrane fouling. Additionally, this chapter also discusses the influence of chlorinated backwashes on membrane structures, and cleaning mechanisms. The comprehensive analysis of the capital expenditure and operation expenditure of the semi-industrial UF system for water reuse is also discussed in this chapter.

In the final conclusions and perspectives section, all the results obtained in this thesis are summarized, and some further studies about this topic are proposed.

CHAPTER 1. State of the Art

1.1. Definition of water reuse from urban wastewater, applications and regulations

1.1.1. Interest and definition of water reuse

Water scarcity is growing under the pressures of population growth, climate changes and increased pollution. From the Global Risks report of 2019 provided by the World Economic Forum, more than 650 million people in 500 cities are projected to face declines in freshwater availability of at least 10% in the 2050s, especially in Middle East and North Africa [28]. Since unsafe wastewater reuse could cause serious catastrophic epidemics of waterborne diseases such as Asiatic cholera and typhoid, the researches on potable water protection and terminal wastewater treatment was then developed, such as the reservoirs or aqueduct systems and filtration [29]. The US Environmental Protection Agency (EPA) defines wastewater reuse as: using wastewater or reclaimed water from one application for another application [30]. The deliberate use of reclaimed water or wastewater must comply with applicable rules for a beneficial purpose. In fact, qualified reuse water not only reduces the occurrence and spread of diseases, but also contributes to the environment protection. Therefore, recovering water, energy, nutrients and other precious materials embedded in wastewater, is a key opportunity to be seized [31,32]. Water reuse could also help to regulate climate by increasing natural flows in surface waters (with cascading positive effects on ecosystem health and biodiversity) and may contribute to rising groundwater tables through water reused for crop or landscaping irrigation [33].

As reported by Bixio et al. in 2005 [34], over 3000 water reuse projects distributed in over 60 countries were assessed in an advanced planning phase. Most water recycling schemes are located in Japan (> 1 800) and the US (> 800), then followed by Australia (> 450), Europe (> 200), the Mediterranean and the Middle East area (> 100) and Latin America (> 50). However, the projects number is likely to be higher considering the rapid development of water reuse in China, India and the Middle East [35]. According to J. Yang [7], the reuse proportion varies greatly from country to country, mainly due to geography, technology, and water supply and demand. As reported by Global Water Market in 2017, the total volume of water reuse was approximately $14.2 \text{ billion m}^3 \cdot \text{y}^{-1}$ worldwide, less than 4% of domestic wastewater (250 to 350

billion $\text{m}^3 \cdot \text{y}^{-1}$) [33,36]. Thus, there is a great potential to establish water reuse projects, improve reused water quality and then contribute to reduce water crisis worldwide.

1.1.2. Water reuse applications and guidelines

As the European Parliament estimated in its recent regulation setting minimum requirements for water reuse in agriculture in May 2020, increasing water reuse in agricultural irrigation from 1.7 billion $\text{m}^3 \cdot \text{y}^{-1}$ to 6.6 billion $\text{m}^3 \cdot \text{y}^{-1}$ by 2025 could reduce water stress by 5% [13]. France, as one of the main member states in the European Union, reused merely 1% of the treated wastewater in 2019 according to the Office International de l'Eau [37]. To improve water reuse rates, the French government aimed to triple the volumes of reused water by 2025 for non-potable purposes by introducing regulatory changes [37]. Generally, non-potable reuse is more likely to build the degree of familiarity and trust required by the public for acceptance, compared to potable reuse [38]. In the global water reuse market, 97.7% of water production is reused for non-potable purposes, of which 52% is for irrigation and 20% is for industrial process water recirculation. Since the potential for human contact increases with water reuse, advanced treatments beyond secondary urban wastewater treatments are more required, among which membrane filtration is one of the key unit technologies for water reuse as discussed in the next section.

Currently, water reuse has become an attractive option for conserving and extending available water supply, and its applications are shown in Table 1-1 [39]. Overall, water reuse plays a significant role in non-potable applications, including non-potable urban reuse, irrigation, recreation impoundment, environmental enhancements, industries and groundwater recharge. Mainly, the share of reclaimed water is mostly for irrigation, 52% in total, including agricultural irrigation and landscape irrigation [39,40]. Additionally, approximately 20% of reclaimed water is reused for the industrial self-water-circulation systems for resource and money savings.

Municipal or urban wastewater or domestic sewage as the main wastewater sources usually have high levels of organic and inorganic material, pathogenic organisms, nutrients and many toxic elements, including heavy metals [41], as stated in Table 1-2.

Table 1-1 Major water reuse applications and constraints worldwide [39,40].

Types	Application Examples	Major Constraints	Percentage Contribution
Potable reuse	Indirect and direct drinking	Public perception issues	2.3%
Non-potable urban reuse	Public parks and schoolyards Highway medians Residential landscapes Fire protection Toilet flushing	Dual distribution system costs The requirement for dual piping systems The greater burden on cross connection control	8.3%
Agricultural Irrigation	Nonfood crops Commercial nurseries Pasture lands	Seasonal demand Usually away from the point of water reclamation Public perception issues High-total dissolved solids (TDS) reclaimed water can adversely affect plant health	32%
Landscape Irrigation	Parks and schoolyards Roadway medians Residential lawns Golf courses Cemeteries Greenbelts	Seasonal demand Usually away from the point of water reclamation High TDS reclaimed water can adversely affect plant health	20%
Recreation	Ponds and lakes Golf courses	Site specific	6.4%
Environmental enhancements	Artificial wetlands Natural wetlands Stream flows	Site specific	8%
Industries	Process water Boiler water makeup Cooling tower water Geothermal energy	Constant demand, but site-specific Limited demand Treatment required depends on end-use	19.3%
Groundwater recharge	Groundwater replenishment Barrier against brackish or seawater intrusion Ground subsidence control	Appropriate hydrogeological conditions needed High level of treatment required Potential for water quality degradation in the subsurface	2.0%
Others			1.7%

Table 1-2 Constituents present in wastewater with their associated risks [42–45].

Wastewater Constituents	Contents	Risks
Microorganisms	Pathogenic bacteria, viruses and worms' eggs	Risk when exposed to humans and animal by inhalation or drinking
Micropollutants	Pesticides, pharmaceuticals, fuel additives, cyanotoxins, personal care products, detergents	Environmental as well as further expected impacts on humans, such as genotoxic, immunotoxic, carcinogenic and fertility-impairing effects
Suspended solids	Particles, solids, colloids	Carrying pollutants and pathogens
Biodegradable organic matters	Organic carbon, sugar, protein, ammonia	Oxygen depletion in rivers and lakes; fish death; odors
Other organic matters	Fat, oil and grease, coloring, solvents, phenols	Toxic effect, esthetic inconveniences, bioaccumulation in the food chain
Other Nutrients	Nitrogen (ammonium, nitrates), phosphorus	Eutrophication, oxygen depletion, toxic effect
Metals	Hg, Pb, Cd, Cr, Cu, Ni	Toxic effect, bioaccumulation
Others inorganic materials	Acids, for example, hydrogen sulfide, bases	Corrosion, toxic effect
Thermal effects	Hot water	Changing living conditions for flora and fauna
Odor (and taste)	Hydrogen sulfide	Esthetic inconveniences, toxic effect
Radioactivity		Toxic effect, accumulation

The reclaimed water should fulfill the criteria of health safety, appearance, environmental acceptance and economic feasibility, for reuse [46]. However, the components and pollutants in wastewater are changeable from source-to-source. Therefore, the treatment processes should vary with the components to be removed. Importantly, different reuse applications require different water quality specifications. Therefore, treatment technologies developed from simple processes into more advanced ones [47]. Since not all the wastewater treatment processes could remove contaminants from wastewater completely, the reused water needs to meet the water reuse standards formulated by the government or other qualified authority institutions that work for the applications or emission of effluents. However, there are no uniformly enforceable

international water reuse guidelines to control the quality of the reclaimed wastewater. Even for the same water reuse application in the U.S., guidelines vary from state to state. Therefore, countries are looking forward to new regulations referring to international guidelines from the United Nations (UN), the EU Commission or the US EPA to estimate the different criteria needed for better water management [35,48,49].

Based on scientific consensus and best available evidence, the World Health Organization (WHO) has set the guidelines for the safe use of wastewater, excreta and greywater, promoting the public health benefits of wastewater, excreta and greywater use in agriculture and aquaculture and is now widely accepted as a benchmark [11]. Additionally, the European Parliament published a legislative resolution on May 2020, regulation (EU) 2020/741 of the European Parliament and of the Council on minimum requirements for water reuse (15301/2/2019—C9-0107/2020—2018/0169(COD)) [13]. This official regulation project defines the water quality limits and obligations of wastewater treatment plant (WWTP) operators, for water reuse in the field of agricultural irrigation only. The above-mentioned reuse guidelines, as the representatives concerning the parameters of water quality, are presented in Table 1-3. To be noted, some parameters mentioned in the EU regulation are not included in Table 1-3. These parameters and associated limits are the *Legionella spp.* should $<1\ 000\ \text{CFU}\cdot\text{L}^{-1}$ where there is risk of aerosolisation in greenhouses, and the Intestinal nematodes (Helminth eggs) should $\leq 1\ \text{egg}\cdot\text{L}^{-1}$ for irrigation of pastures or forage.

In fact, most countries supplement and develop their own guidelines, based on WHO guidelines—and considering their potential specifications. Overall, the following parameters: pH, total suspended solids (TSS), biological oxygen demand after 5 days (BOD_5), turbidity, total coliform and fecal coliforms are important parameters to evaluate the water quality all over the world, and they will be considered to evaluate the ability of some water reuse technologies in the following parts of this study. The choices of wastewater treatment technologies may be affected by the following factors [50]: reclaimed water objectives for application, wastewater characteristics, compatibility with existing conditions, process flexibility, operating and maintenance requirements, energy and chemical requirements, economical requirements, residual disposal options and environmental constraints. In general, in a WWTP, if the wastewater, after primary and/or secondary treatment, cannot be reused for target purposes, it should be treated to tertiary level or with more advanced processes to achieve reuse standards.

Table 1-3 Water reuse categories and typical application (WHO guidelines and EU Parliament regulation).

Organization	Category	Typical Application	pH	TSS (mg·L ⁻¹)	Turbidity (NTU)	BOD ₅ (mg·L ⁻¹)	Residual Cl ⁻ (mg·L ⁻¹)	Fecal Coliform (<i>E. coli</i> as an Indicator) (100 mL ⁻¹)
EU Parliament	Agriculture irrigation	A		≤10	≤5	≤10		≤ 10 / below detection limit
		B			–			≤100
		C		≤35 Directive 91/271/EEC	–	≤25 Directive 91/271/EEC		≤1000
		D			–			≤10,000
WHO guidelines	Agriculture irrigation	Food crop irrigation (uncooked)	6–9	ND	≤2	≤10	1	ND
		Non-food crops and crops consumed after processing	6–9	≤30	–	≤30	1	≤ 200
	Landscape irrigation	Parks; schoolyards; Playgrounds	6–9	ND	≤2	≤10	1	ND
		Golf courses; Cemeteries; Greenbelts; Residential	6–9	≤30	–	≤30	1	≤200
	Industrial recycling and reuse	Cooling water; boiler feed; Process water; Heavy construction	6–9	≤30	–	≤30	–	≤200
	Groundwater	Groundwater replenishment; saltwater intrusion control; Subsidence control			Site-specific; specific guidelines do not exist.			
	Recreational Environmental uses	Lakes and ponds; marsh enhancement; streamflow augmentation; fisheries; snowmaking	6–9	ND	≤2	≤10	1	ND
	Non-potable urban uses	Fire protection; air conditioning; toilet flushing	6–9	ND	≤2	≤10	1	ND
	Potable uses	Blending in water supply reservoirs; Blending in groundwater; Direct pipe-to-pipe water supply			Meet requirements for safe drinking water; specific guidelines do not exist.			

ND, not detected; NTU, nephelometric turbidity units; ABCD represents the different water quality levels, A being the best, D being the worst.

1.1.3. Focus on the pollution by microplastic particles

Even if microplastic particles (MPs) are not mentioned yet in the water reuse standards or guidelines as they are not even mentioned in drinking water standards because standards for detection and quantification are just being elaborated, it is a pollutant of main interest. Indeed, currently, more and more researches, especially on microplastic sources, pollutions, transport, harmfulness, retention, recovery methods, and analytical methods, have been developed [51]. Defined by Frias et al. (2019), MPs are either synthetic solid particles or polymeric matrix, with size ranging from 1 μm to 5 mm, from primary or secondary manufacturing origin, and are insoluble in water. MPs can be easily swallowed by organisms and accumulated due to their physical properties such as small particle sizes and widespread distribution (Rillig, 2012). Normally, plastics have stable chemical properties that can stay for hundreds to thousands of years in nature. MPs do not cause fatal effects on living organisms, but they would cause chronic toxicity, which is considered as a key issue for long-term exposure. There are several mechanisms for microplastics to cause toxic effects. First, some polymer materials can directly release toxicity, such as polyvinyl chlorides (PVC), Polyurethanes (PUR), polyacrylonitriles (PAN), and Polystyrene (PS) etc. PVC can release dioxins, phthalates, vinyl chloride, ethylene dichloride, and other toxic chemicals. PS is found to be translocated in blood circulation and cause reproductive disruption for marine filter feeders [54,55]. All of them can leech harmful chemicals into the water or food and get into human bodies through food chains. Second, microplastics can carry toxic and harmful substances. MPs are prone to be gathered with other particles or suspended solids in waters as plastics tend to be more hydrophobic [56]. Highly toxic substances such as Dichloro-diphenyl-trichloroethane (DDT), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), nonylphenols and other nonpolar substances can adsorb on microplastic surfaces that can be transported and bioaccumulated through food chain [57,58]. Besides, some additives added by manufacturers such as plasticizers, stabilizers and pigments to plastics, mostly are hazardous substances. Third, MPs can be further degraded to the smaller pieces, for example, nanoplastics ($< 1\mu\text{m}$), and may become more dangerous for human health [59]. Ragusa et al. [60] have already discovered the presence of MPs (5-10 μm) in human placenta, reflecting that the MPs can be absorbed into human bloodstream and be transferred among organs which will potentially reduce the bodies defense mechanisms against pathogens.

Nowadays, the demand of plastics in our daily life is largely increased, global plastics production has reached up to 368 million tons in 2019 (PlasticsEurope 2020), thus lots of plastic waste eventually flows into the oceans and wastewater system [62–64]. Liu et al. [65] investigated 38 WWTPs in 11 countries and found that the abundance of MPs in influent ranged from $0.28 \text{ MP}\cdot\text{L}^{-1}$ to $3.14 \times 10^4 \text{ MP}\cdot\text{L}^{-1}$, while that in the effluent ranged from $0.01 \text{ MP}\cdot\text{L}^{-1}$ to $2.97 \times 10^2 \text{ MP}\cdot\text{L}^{-1}$. For example, in raw wastewater, $130 \text{ MP}\cdot\text{L}^{-1}$ was detected by Ben-David et al. [66] and $58 \text{ MP}\cdot\text{L}^{-1}$ was detected by Lares et al. [67]. In this case, wastewater treatment plants (WWTPs) serve as an important route for MPs to the environment. Many researchers reported that WWTPs with tertiary treatment process or membrane bioreactor treatment could achieve higher removal of MPs from raw wastewater. For example, Ben-David et al. [66], Lares et al. [67], and Simon et al. [68] all confirmed that the WWTPs can remove more than 97% of 10-500 μm sized MPs. In fact, the presence of smaller MPs increases the difficulty of detection and tracking, the difficulty of removal, and also increases the potential threat for natural environment and organisms. Therefore, it is necessary to find more accurate analysis method and the effective treatment process to reduce the MPs exposure to human and organisms.

1.2. Membrane-based technology for water reuse

In the context of municipal (urban) wastewater treatment, primary treatment can be considered as physical processes such as sedimentation and flotation [69]. The process can remove approximately 50–70% of total suspended solids (TSS) and 25–50% of BOD_5 [3]. The secondary treatment combines biologic and chemical processes to remove the soluble organic matter and the residual TSS after the primary process, such as activated sludge, oxidation ponds and rotating biologic contactor [4]. However, the secondary effluent is limited in reuse applications with risks for human health and public environments [5]. A tertiary treatment could then be applied to remove associated hazards and health risks [5]. Tertiary and advanced treatments are considered as the advanced level of treatment and mostly include membrane processes, advanced oxidation processes, disinfection or any combination of them to remove chemical and biologic constituents. In order to make reuse cost-effective, the level of treatment must be “fit for purpose”. Treatment technologies are employed either alone or in combination to achieve wastewater reuse levels. However, as the potential for human contact increases,

advanced treatment beyond secondary treatment becomes more accepted, and membrane filtration is one of the key unit technologies for water reuse. Although several review articles have given overview summaries of the previously reported techniques for municipal wastewater reuse [6,70–73], they either mainly focused on greywater reuse or on only one reuse level or on only one type of technology. Therefore, membrane-based municipal wastewater treatment techniques for reuse have not been extensively reviewed.

This section aims to provide a comprehensive view on membrane-based processes used in municipal wastewater treatment for water reuse. Particularly, this review is expected to provide an overall summary of reported research cases of feasible membrane-based technologies on urban wastewater treatment for reuse, mainly focusing on membrane hydraulic performance, contaminants removal ability, reuse purposes and energy consumption. Key benefits and challenges of current implementations will be also described after the summary.

1.2.1. Performances of Membrane-Based Treatment Processes for Municipal Water Reuse

Membrane-based technologies are advanced treatment process for municipal wastewater treatment [74]. Pressure-driven membrane processes are presented in Table 1-4 and can be classified into four main categories based on the different selective pore size: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), [6,75–78]. The selection of material is affected by pore size distribution, wetting susceptibility, porosity, mechanical strength, cost, polymer flexibility, fouling resistance, stability, durability and chemical resistance [79]. As the pores get smaller, the membrane needs more driving force to be operated [79,80]. Membrane fouling always occurs during the filtration process, followed with an increase in transmembrane pressure (TMP) to maintain a constant flux or followed with a decrease in flux for a constant TMP. According to Guo et al. [81], membrane fouling is often defined based on the type of pollutants present in the feed stream and can be classified into four categories, including: (1) particulate/colloidal fouling, when suspended solids and/or colloids clog the pores of a membrane or adhere to its surface; (2) organic fouling, when reactive dissolved organic components or colloids attach to the membrane by adsorption; (3) inorganic fouling, also known as scaling or precipitation fouling, caused by the presence of crystallized

salts, oxides or hydroxides in the feed solution that are prone to precipitate onto membrane surface due to pH change or oxidation; and (4) biofouling, caused when microorganisms, plants, algae or other biologic contaminants grow on or in the membrane surface and pores. Each type of fouling can be either reversible or irreversible by membrane cleaning step and permanently compromise its performance. All the fouling types can take place simultaneously, and the interaction between them usually increases filtration resistance [82].

Table 1-4 Membrane separation characters for water purification [76,87–90].

Characteristics	MF	UF	NF	RO (low-pressure)
Separation mechanism	Sieve	Sieve	Sieve, solution/diffusion, electric repulsion	Solution/diffusion,
Membrane	Porous	Porous	Finely porous asymmetric/composite	Nonporous asymmetric/composite
Molecular weight cut-off	> 300 kDa	1–300 kDa	200–1000 Da	< 200 Da
Retained compounds	Colloids, TSS, turbidity, some protozoan oocysts, cysts, some bacteria and some viruses under specific conditions	Macromolecules, proteins, colloids, bacteria, viruses	LMWC, mono-, di- and oligo-, saccharides; polyvalent anions, some hardness, viruses	LMWC, sodium, chloride, glucose, amino acids, hardness, ions
Transmembrane pressure (TMP)	< 5 bar	< 10 bar	< 20 bar	< 100 bar
Flow modes	Crossflow or Dead-end	Crossflow or Dead-end	Crossflow	Crossflow

HMWC: high molecular weight compounds; LMWC: low molecular weight compounds.

Therefore, in each membrane process, fouling issues need to be considered and optimized to obtain sustainable conditions and excellent filtration performance. Additionally, an important notion to take into account is the threshold flux, the flux at or below which a low and near constant fouling rate occurs, but above which the fouling rate increases markedly [83,84]. The number of studies on critical, sustainable and threshold flux of different kinds of wastewater filtrated by certain membranes are expanding recently because these concepts of flux are highly related to control low fouling rates on membranes [83,85,86]. Especially for commercial and industrial production, the operation at sustainable flux could definitely control fouling and at the same time, give an optimal balance between moderate operating costs and moderate capital costs [83]. The definition of the sustainable conditions appears as a challenge for each new membrane-based treatment processes for municipal water reuse.

1.2.2. MF/UF-based Treatment Processes After Secondary Treatment of WWTP

MF and UF can remove particles larger than their pore size mainly through sieving mechanism [79,91]. In general, the MF process alone can effectively remove high molecular weight organic matters, suspended solids, colloids, bacteria and thus reduce turbidity. Compared to MF, UF membrane has wider separation ranges with smaller pore size and enhanced removal ability for particles, colloids, and more importantly, bacteria with high removal rate and viruses. In order to clearly understand the difference between MF and UF, Tchobanoglous et al. [80] quoted by the review of Warsinger et al. [6], summarized the rejection characteristics when filtrating the same secondary effluent by MF and UF, on bacteria, viruses and other essential water quality parameters such as TSS, chemical oxygen demand (COD) and TDS. It should be noted at first that the data gave general information with certain limitations: the actual performance may vary related to different conditions, such as temperature, flow rate and transmembrane pressure (TMP). From the review [6], MF is almost as efficient as UF in removing TSS and BOD₅, of which the removal rates range from 95% to 99.9% and from 75% to 90%, respectively. Furthermore, the efficiencies of UF on COD and total organic carbon (TOC) concentrations removal are about 5% to 20% higher than MF. More importantly, UF provides almost complete removal of bacteria, protozoa and viruses, which is one of the main

advantages compared to MF. MF can partially remove viruses and protozoa through adhering to impurities in wastewater like protein, colloids or coliforms, but microorganisms can still pass through MF membranes since the pore size is larger than the virus size [34]. In 1995, Madaeni et al. [92] have verified that the presence of bacteria (*E. coli*) and turbidity highly contributed to the *poliovirus* rejection through 0.22 μm MF under TMP between 0.5 and 1 bar. Huang et al. [93] demonstrated that virus (MS2 bacteriophage as an indicator) is more likely to be removed through 0.1 μm -MF in waters with more organic matters or higher concentration of large molecular weight organic foulants. Herath et al. [94] successfully observed a considerably high virus rejection rate at their isoelectric point when using a 0.2 μm MF membrane to filtrate the specific virus (19 to 80 nm) solutions due to the virus–virus coagulation and virus–protein coagulation. In addition, the physicochemical properties of the virus, the membrane material, the feed, as well as the hydraulics of the filtration process could affect virus removal by MF [93,95,96]. For example, Huang et al. [93] recorded that when adding sodium or calcium to feed water, the virus removal capacity decreased immediately. Besides, low pH value increases virus removal while higher pH value makes removal rate decrease [94]. Anyway, the removal of viruses through MF is significant but not complete because the virus are still able to pass through membrane pores [97]. It is reported by Warsinger et al. [6] that the log removal value of MF processes on viruses ranges from 0 to 2 (equals to 0% to 99%). Furthermore, according to Warsinger et al. [6], UF can support up to 6 log removal of bacteria and up to 7 log removal of viruses, and if there are protozoa, UF can remove protozoan cysts and oocysts to more than 6 log reduction. Therefore, UF process provides more safer and more reliable water product than MF process. At present, ultrafiltration (UF) has been widely accepted as one of the most cost-effective technology to deal with secondary effluent on non-potable reuses, such as agricultural irrigation, landscape irrigation, urban reuse, car washing, and sometimes as process water for industries [6,98]. Table 1-5 summarizes the real cases of MF/UF-based processes applied after secondary treatment for water reuse. In results, among the literature about the MF/UF-based process applied on water reuse (Table 1-5), 60% of the lists are UF-based treatment processes that achieved water reuse and met standards. UF enables high removal efficiency on total suspended solids (TSS), turbidity, and partial removal efficiency on organic matters [10]. More importantly, UF can retain microorganisms significantly, including bacteria, protozoa, and viruses [6,99]. In early 1998, G. Tchobanoglous et al. [100] had already applied the UF

membrane (hollow fiber, polysulfone) on treatment of secondary and tertiary municipal wastewater with good results. J. Arévalo et al. [101] confirmed the effectiveness of both hollow fiber and flat-sheet UF membranes on removal of particles, organics, and microorganisms from wastewater. On reuse purpose, the investigation of Muthukumaran et al. (2011) [102], and Pollice et al. (2004) [103] or Falsanisi et al. (2010) [98] all confirmed that UF (with prefilter) can be successfully applied on secondary effluent treatment for reuse in agriculture with respect to WHO guidelines in terms of permeate quality respectively with synthetic and real secondary effluent as feed. These three studies were all conducted in pilot-scale operation ranging from 2 months to 2 years and with pore sizes ranging from 0.002 μm (1kD) to 0.03 μm . In Table 1-5, only one reference demonstrates MF process alone on achieving water reuse. Ahn et al. [104] showed that MF permeate can be used to clean building floors or flush toilets, but they did not state the water reuse standard that they referred to. When comparing their permeate quality with WHO guidelines, the detected parameters are incomplete and substandard, e.g., the turbidity in both cases exceeds the limit of WHO guidelines (turbidity ≤ 2 NTU). The other listed MF processes were all combined with chemical and physical processes, or UF process to finally achieve water reuse purposes.

Moreover, chemical and/or physical processes such as sedimentation, adsorption, flocculation and coagulation [105–107] can be used as pretreatment of membrane processes to decrease membrane fouling potential and improve filtration performance [106,108,109]. For example, $\text{Al}_2(\text{SO}_4)_3$, $\text{Fe}_2(\text{SO}_4)_3$, FeCl_3 and polyaluminum chloride can be used as remarkable coagulants [108]. Activated carbon (AC) is a widely accepted adsorbent for adsorption [110]. It can be used as a powder (PAC) in dispersion or as granules (GAC) in fixed bed. Pretreatment process helps to clean the feed water for MF/UF and contributes to remove the fouling potentials before MF/UF membranes, thus contributing to prolong the whole system operational time, improving permeability recovery, and decreasing membrane cleaning chemicals consumption. As stated by Zheng et al. [111], when studying the time required to increase TMP from 0.22 bar to 0.7 bar on secondary effluent treatment (constant flux of 50 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$), direct UF process took only 12 h while the sand-UF system could extend to 30 days. In addition, Fan et al. [112] designed parallel tests of MF with/without pretreatment to treat secondary effluent under constant TMP (0.70 bar), the results confirmed that the coagulation-prefiltration (1.5 μm)-MF systems could improve flux recovery (J/J_0) between 20% and 30% higher than MF alone due to

the hydraulically irreversible fouling (internal adsorptive fouling) reduction by coagulation. First, it should be noted that in some pretreated processes, for example coagulation with settling, the re-growth of flocs could conversely cause severe cake fouling on membranes [113]. It is necessary to add another prefilter after coagulation, flocculation and settling to remove the flocs, colloids and other particles before the membranes. Second, it has been proven that pretreatment processes could effectively remove natural organic matter (NOM) and colloids [114,115]. As described previously, dissolved organic carbon (DOC) cannot be effectively retained by MF or UF due to membrane pore sizes being much larger than the related organic molecules [116]. Fan et al. [112] reported that MF could only remove 3.9% of DOC, while UF could remove 24.7%. Therefore, when pre-coagulated with $5 \text{ mg}\cdot\text{L}^{-1}$ of Al^{3+} , the MF system could increase 10% to 15% DOC removal; and when pretreated with $10 \text{ mg}\cdot\text{L}^{-1}$ anion exchange resin in wastewater which removes primarily lower molecular weight cut-off (MWCO) and negatively charged organic fractions, the MF and UF systems could improve DOC removal to 58.8% and 68.3%, respectively [112]. Third, pretreatment processes contribute to the removal of dissolved nutrients, mainly nitrogen and phosphorus. Hybrid precipitation–MF system by Lu et al. [117] made use of calcium salt to increase precipitates involved with phosphorus and fluorine and then separated solid–liquid phases by a following crossflow MF for separation. Guo et al. [106] compared the performance of MF with and without pretreatment for wastewater reuse, the results showed that MF alone only removed 20% of TOC and 5% of PO_4^{3-} , while pretreated with flocculation and adsorption, the removal efficiency of TOC reached 99.7% and more than 97% of PO_4^{3-} . In addition, the UF system coupled with PAC can be used to eliminate both DOC and micropollutants. Though MF/UF alone is inadequate for micropollutant removal, in PAC–membrane processes, contaminants (including natural disinfection byproduct precursors) can be reduced through adsorption onto the activated carbon particles, which are then separated from water by either UF or MF [34]. Among MF-based treatments in Table 1-5, the hybrid physical/chemical process with MF can improve the water quality to meet with reuse standards. However, based on the WHO water reuse guidelines, MF permeate is mostly applied in low levels of non-potable reuse, such as toilet flushing, machine cleaning, irrigation for non-food crops or more likely reused as process water for industries, such as washing, cooling and circulating. Among UF-based treatments in Table 1-5, the hybrid flocculation, coagulation or adsorption process with UF processes, could almost completely remove turbidity, TSS, color

from secondary treated municipal WWTP, and the removal of COD, DOC, metal and salt ions was also improved and higher than MF. In addition, pretreatment is mostly used to decrease fouling potential for UF especially when the feed water quality is not very good, in order to provide sustainable filtration performance and reduce operating cost of the whole system. Additionally, pretreatment can also bring troubles. When pretreated with adsorption, the disadvantages include the expensive regeneration of the adsorbents (such as PACs) and loss of adsorbents [118]. Moreover, pretreatment with coagulation, flocculation and adsorption need higher energy cost with increased sludge volume production during treatment [119].

Generally, effluents from WWTPs still carry high TSS and NOMs which are prone to cause the formation of fouling on the MF/UF membrane easier. In fact, dissolved organic matters cannot be removed effectively by MF or UF systems, but conversely it can be the main cause of fouling formation on the membrane, finally resulting in shortening membrane lifespan, reducing flow rate, increasing TMP and energy consumption [120,121]. Considering the hydraulic filtration performances (Table 1-5), almost all researchers were interested in fouling control. Under constant permeate flux of $20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, reported by Falsanisi et al. [98], the TMP on UF would increase fast from 0.3 bar to almost 1.2 bar in only 20 min filtration, and permeability decreased from 150 to about $10 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$. While, according to Pollice et al. [103], with a periodic filtration cycle of the permeate (90–360 s) and a backwash of (30–40 s) in continuous operational process on secondary effluent treatment in autumn, the system could slow down the increase of TMP on UF which increased from less than 0.1 bar to about 0.7 bar in more than 30 days under almost stable flux at $20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$. Therefore, periodic backwashes could prolong the UF operation time, reduce the frequency of chemical cleaning uses and save energy during long term operation. Therefore, to support sustainably high performance of MF/UF on wastewater treatment in long term operations, it is necessary to conduct suitable physical and chemical cleaning on membranes.

In summary, direct MF is less used for water reuse treatment due to lower removal efficiency on microorganism and organic matters. The hybrid MF with physical/chemical processes sometimes can improve the water product quality to reuse levels but are low levels due to more healthy concerns. In comparison, UF is more recommended for water reuse treatment than MF. UF has higher removal capacity than MF on various water parameters, such as TOC, COD, turbidity, etc. Moreover, UF can theoretically completely remove

microorganisms while MF cannot. Additionally, either UF process alone or hybrid with physical/chemical process can produce qualified water for reuse purpose. The pretreatment before UF is mainly to decrease fouling potential especially when the feed water contains more TSS, organic matters, or particles but the pretreatments need higher costs. This is why hybrid MF or UF was not selected in this work. The limitation of UF-based treatment on potable reuse may be the residual dissolved organic matters, micropollutants, residual nutrients such as phosphorus, nitrates and ammonium, and specific toxic ions produced from coagulants or flocculants (such as chloride ions). In this case, the permeate of UF as the main technology for reclamation is commonly reused on non-potable applications. UF-based processes are more acceptable with respect to water reuse guidelines compared to potable reuse.

Table 1-5 MF/UF processes alone or combined with pretreatments after secondary treatment for water reuse applications.

Process (Scale and Operation Duration)	Feed Wastewater	Operating Conditions	Feed Characteristics	Permeate Quality/Removal Rate	Application for Reuse	Standard Basis	Ref.
MF (pilot plant, 120 d)	Domestic wastewater (septic tank effluent)	Capacity: 10 m ³ ·d ⁻¹ MF: 0.1 µm; TMP: 0.20–0.50 bar	COD: 10–622 mg·L ⁻¹ BOD ₅ : 25–110 mg·L ⁻¹ TOC: 2.8–22.6 mg·L ⁻¹ TSS: 5–645 mg·L ⁻¹ ; Turbidity: 2.7–123 NTU Color: 5–109 CU	COD: 1–30 mg·L ⁻¹ , 92.8% BOD ₅ : 1–7 mg·L ⁻¹ , 92.9% TOC: 0.4–8.1 mg·L ⁻¹ , 65.8% TSS: 0–2 mg·L ⁻¹ , 99.8% Turbidity: 0–4.2 NTU, 99.4% Color: 2–32 CU, 76.2%	Toilet flushing	Not mentioned	[104]
UF (pilot plant, 2 years)	Secondary effluent of WWTP	TMP: 0.1–0.7 bar UF: 0.03 µm	TSS: 96–165 mg·L ⁻¹ COD: 167–307 mg·L ⁻¹ PO ₄ -P: 1.0–3.9 mg·L ⁻¹ NH ₄ -N: 3.0–33 mg·L ⁻¹ N _{org} : 9–16 mg·L ⁻¹	TSS: 3–9 mg·L ⁻¹ COD: 42–103 mg·L ⁻¹ PO ₄ -P: 0.8–3.4 mg·L ⁻¹ NH ₄ -N: 4.0–33 mg·L ⁻¹ N _{org} : 2–5 mg·L ⁻¹ LRV (total coliforms): 3.7 LRV (fecal coliforms): 4.2 LRV (<i>E. coli</i>): 3.7	Crops irrigation (tomato and fennel)	Meeting WHO guidelines	[103]
Prefilter +UF (pilot plant, two months)	Secondary effluent of WWTP	UF: 0.01 µm (200 kDa), UF modes: crow flow Inlet flow: 10 m ³ ·h ⁻¹ TMP: 0.30–1.20 bar	pH: 6.3–7.5 T: 19–25 °C EC: 1584–1950 µS·cm ⁻¹ Turbidity: 1–7 NTU TSS: 1–8 mg·L ⁻¹ COD: 26–69 mg·L ⁻¹ <i>E. coli</i> : 3000–36,000 CFU·100 mL ⁻¹ Total coliforms: 9100–96,000 CFU·100 mL ⁻¹	Turbidity: < 0.2 NTU TSS: < 0.2 mg·L ⁻¹ COD: 20–60 mg·L ⁻¹ <i>E. coli</i> : 0 CFU·100 mL ⁻¹ Total coliforms: 0 CFU·100 mL ⁻¹	Agriculture irrigation	Meeting WHO guidelines	[98]

Prefilters +UF (pilot scale)	Synthetic secondary sewage effluent	UF: 1 kDa/0.002 μm (tubular); 25 kDa/0.008 μm (spiral wound) UF modes: cross flow TMP: 1.0–3.3 bar Flow velocity: 0.2 $\text{m}\cdot\text{s}^{-1}$	COD: 18.5–67 $\text{mg}\cdot\text{L}^{-1}$ Turbidity: 9.43–46.4 NTU TSS: 13–30 $\text{mg}\cdot\text{L}^{-1}$ Color: 41–81 EC: 320–366 $\mu\text{S}\cdot\text{cm}^{-1}$ A(254 nm): 0.25–1.051 pH: 7.7–7.81	COD: 64.38–80.4% Turbidity: 96.75–99.61% Color: 0–53.49% Absorbance (254 nm): 76.6–91.94%	Non-potable reuse (not detailed)	Meeting WHO guidelines	[102]
GAC+MF (flat sheet) (lab-scale, 140 d)	Secondary treated water of the sewage treatment plant in Sungkyunkwan University	MF: 0.22 μm Flux: 98 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ TMP: 0–0.4 bar	pH: 7.62–8.02 Turbidity: 2.2–10.3 NTU TSS: 4–20 $\text{mg}\cdot\text{L}^{-1}$ UV ₂₆₀ : 0.28–0.32 cm^{-1} DOC: 6–8 $\text{mg}\cdot\text{L}^{-1}$ COD: 10–30 $\text{mg}\cdot\text{L}^{-1}$ TN: 30–50 $\text{mg}\cdot\text{L}^{-1}$ TP: 15–30 $\text{mg}\cdot\text{L}^{-1}$	Turbidity: 0.1–0.4 NTU, 100% TSS: 100% UV ₂₆₀ : 0.26–0.3 cm^{-1} , 60% DOC: 2–4 $\text{mg}\cdot\text{L}^{-1}$, 40–46% COD: 8–25 $\text{mg}\cdot\text{L}^{-1}$, 53% TN: 20–40 $\text{mg}\cdot\text{L}^{-1}$, 15% TP: 10–20 $\text{mg}\cdot\text{L}^{-1}$, 13%	Not mentioned	WHO guidelines	[122]
Submerged GAC-MF (Semi-batch, 60 d)	Biologically treated sewage effluent	MF: 0.14 μm , flat sheet NF: 700 Da, flat sheet TMP: \leq 4 bar Flux: 2.5 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ GAC: 10% daily replacement	pH: 6.8–7.6 EC: 520–1120 $\mu\text{S}\cdot\text{cm}^{-1}$ DOC: 3.6–7.7 $\text{mg}\cdot\text{L}^{-1}$	DOC: 2.4 \pm 0.2 $\text{mg}\cdot\text{L}^{-1}$, 53 \pm 5% PPCPs: < 5 $\text{ng}\cdot\text{L}^{-1}$ (for each)	Not mentioned	Australian and New Zealand Guidelines for Fresh and Marine Water Quality	[123]
Coagulation +MF (lab-scale, 5 months)	Secondary effluent from WWTP	Coagulant: 10–50 $\text{mg}\cdot\text{L}^{-1}$ alumina MF: 0.1, 0.22 μm ; TMP: 0.34 bar	Turbidity: 19.7 \pm 87.9 NTU TOC: 7.2 \pm 6.5 $\text{mg}\cdot\text{L}^{-1}$ pH: 7.0 \pm 0.2 UV ₂₅₄ : 0.040–0.058 cm^{-1} Alkalinity: 202.8 \pm 12.2 $\text{mg}\cdot\text{L}^{-1}$ as CaCO_3 TSS: 14.4 \pm 25.8 $\text{mg}\cdot\text{L}^{-1}$	Turbidity: 0.11–0.13 NTU, >93%; TOC: 1.30–1.56 $\text{mg}\cdot\text{L}^{-1}$, 23.5–35.5% UV ₂₅₄ : 0.019–0.02 cm^{-1} , 52.5–54.5%	Not mentioned	Not mentioned	[124]

Coagulation + PAC + UF (lab scale)	Secondary effluent from WWTP	UF: 50 kDa TMP: 1 bar Coagulant: FeCl ₃	pH: 7.4 Turbidity: 18 NTU TSS: 35 mg·L ⁻¹ BOD ₅ : 30 mg·L ⁻¹ COD: 77 mg·L ⁻¹ EC: 1350 μS·cm ⁻¹ Zeta potential: 4.118 mV	COD: 13.33–21 mg·L ⁻¹ Turbidity: 0.5–0.8 NTU Zeta potential: -0.332–0.166 mV	Not mentioned	Not mentioned	[125]
PAC+UF (lab-scale, ~1 h)	Secondary effluent from domestic WWTP	UF: 100 kDa TMP: 1 bar	pH: 7.1–7.6 Turbidity: 0.9–1.5 NTU TOC: 3.3–5.2 mg·L ⁻¹ UV ₂₅₄ : 0.09–0.12 cm ⁻¹ COD: 25–32 mg·L ⁻¹ Color: 18–24 CU Coliforms: 300–700 mL ⁻¹	DOC: 22.2–28.8% UV ₂₅₄ : 33.7–38.3%	urban reuse, agricultural, landscape and industrial reuse	Not mentioned	[126]
MF+UF	Secondary effluent from urban WWTP	MF: 0.2 μm, hollow fiber, 0.2–0.8 bar (TMP) UF: 0.05 μm, flat sheet, 0.2–0.6 bar (TMP)	Turbidity: 4–20 NTU TSS: 11–87 mg·L ⁻¹ T-UV _{253.7} : 11–41% Nematode eggs: 0–200 Un·L ⁻¹ <i>E. coli</i> : 10 ⁴ –10 ⁶ CFU·100 mL ⁻¹ Fecal coliforms: 10 ⁴ –10 ⁶ CFU·100 mL ⁻¹ Coliphages: 10 ³ –10 ⁴ PFU·100 mL ⁻¹	Turbidity: 0–0.9 NTU TSS: 1–7 mg·L ⁻¹ T-UV _{253.7} (%): 0 Nematode eggs: 0 <i>E. coli</i> : 0 CFU·100 mL ⁻¹ Fecal coliforms: 0–9 CFU·100 mL ⁻¹ Coliphages: 0–1 PFU·100 mL ⁻¹	Not mentioned	Water reuse guidelines of US EPA	[99,127]

A: absorbance; CFU: colony forming unit; EC: electrical conductivity; LRV: log10 removal value; SAC: spectral absorption coefficient; PPCPs: pharmaceuticals and personal care products

1.2.3. MBR-Based Treatment for Water Reuse

Besides of chemical and physical processes, biologic processes could also be combined with a membrane system based on its advantage in the degradation of microorganisms [128]. The activated sludge process (CAS) is a biologic process that is mainly used for reduction of organic matters in the wastewater, and usually includes an aeration tank used for biologic degradation and a secondary clarifier (sedimentation tank), where the sludge is separated from the treated effluent [129]. The effect of the membrane is to increase the concentration in the biologic reactor, to retain the particulate phase within the bioreactor and allow permeate to the next process or to be discharged/reused [130]. In MBRs, biodegradation and membrane separation are performed simultaneously, thus maximizing wastewater treatment efficiency [131]. A review by Wu et al. [70] detailed the non-potable and potable application of various MBR systems for greywater reclamation with numerous MBRs case analysis. The reclaimed water is proved to satisfy the guidelines of US EPA and WHO reuse applications. It is reported that the MBRs performed better in the removal of organic matters, resulting in relatively less energy and economy consumption compared with conventional CAS processes. In addition, Wu et al. [70] and Atasoy et al. [132] both stated that greywater produced less organic foulant for MBRs together with the lower driving force and lower membrane fouling. Studies summarized in Table 1-6 are focused on MBRs for municipal wastewater treatment and reuse applications. MBRs have been reported to consistently achieve removal rates of 90–95% for COD, 80–99% for $\text{NH}_4\text{-N}$ and 70–99% for total phosphorus (TP), respectively [133]. Although feed characteristics among various municipal WWTPs showed large differences, the results of organic removal ratios in the MBRs presented only slight dissimilarities. The application of reclaimed municipal water can be in non-potable uses with MBRs or additional processes. In addition, MBRs can be operated at wider organic loading rates, higher concentration of mixed liquor suspended solids (MLSS) and larger amount of feed flows. This view can be supported by Tam et al. [134] who described the separately MBR and MF processes on the same sewage treatment, when the water production both achieved the reuse level for toilet flushing, the MF feed was secondary treated effluent whereas the MBR feed was just degrittied sewage. Nevertheless, MBRs are confirmed to produce high qualified water production even under higher concentration of COD, BOD_5 , TSS, TN, TP and turbidity in the feed. Studies have demonstrated that MBR treatment removed different types

of microorganisms, including enteric viruses in some studies, more effectively than in conventional secondary treatment, on both pilot-scale and within full-scale municipal WWTPs [130,135,136]. Chaudhry et al. [137] demonstrated the MBR process could provide more than 4-log reduction of adenovirus and norovirus GII and over 5-log reduction of F⁺ coliphage, and provided evidence for assigning virus disinfection credit to similar MBRs for reclaim wastewater [137]. It is important to note that the removal value is a function of the inlet concentration and outlet after MBR and the inlet concentration is at least two times higher in MBR treatment than in CAS treatment. Nevertheless, the above cases confirmed the stable effectiveness of MBRs on certain bacteria removal whatever the pore sizes, but it is not for all microorganisms. Other studies draw a similar conclusion on the removal of seeded viruses in MBRs with LRVs ranging greatly from 0.4 to 5.8 with membrane pore sizes ranging from 0.1 to 0.4 μm [138,139]. However, size exclusion by the membrane is probably the dominant removal mechanism for bacteria and viruses in MBRs with intact membranes, but not exclusively. Other removal mechanisms, such as adsorption to the biomass, pore blocking and pore constriction may also play an important role [140].

Furthermore, the effective biodegradation integrated with membrane performance leads to more advantages than separated processes, i.e., less organic foulants on the membrane, smaller footprint, faster plant activation, no biologic sedimentation units and less sludge production [141,142]. As a result, (1) particle separation can be achieved without sedimentation before MBR systems, but with membrane filtration [143]; (2) sludge production from MBRs is lower than CAS [82,133]; (3) MBRs can provide footprint savings due to a higher organic loading rate and greater reactor depth compared to CAS. Membrane fouling remains a major obstacle that hinders faster commercialization of MBRs due to its influences on flux decline, membrane lifetime span, backwash frequency, energy cost, and even permeate quality. Generally, membrane fouling in MBRs should be blamed for both membrane pore-clogging and sludge cake deposition on membranes which is usually the predominant fouling component [144]. To control fouling, aeration is effective to remove cake fouling thus enhances the membrane flux, especially for submerged membrane [131,145–148]. But high aeration also brings impacts on biomass characteristics, since colloids and solutes would be the major membrane foulants which cannot be removed effectively by increasing shear stress [149]. In summary, MBRs provide qualified permeate for non-potable water reuse, while improvements need to be found to decrease

membrane fouling and the related energy consumptions. Moreover, the operating conditions for biological degradation and membrane purification are coupled and more difficult to control.

Anaerobic MBR (AnMBRs) are getting increasing attention and interest. Unlike MBR, AnMBR consists of a primary anaerobic bioreactor and a secondary MF/NF membrane bioreactor. First, the microorganisms in the anaerobic bioreactor convert organic carbon and BOD₅ in wastewater into biogas (methane) and CO₂. Second, membranes separate the microorganisms and other suspended solids from the treated effluent. The main advantage of wastewater treatment by AnMBRs is the natural aspects of the bioprocess such as the reduction of the overall energy demand, wide loading rate decreasing the need for aeration, increasing energy recovery from methane production and producing less secondary sludge [72,150,151]. Once Song et al. [72] tested the performance of AnMBR on synthetic domestic wastewater at the lab scale, the results showed that although the effluent after AnMBR achieved the high removal efficiency on COD ($98.4 \pm 0.4\%$), TOC ($98.7 \pm 0.3\%$) and TN ($20.4 \pm 11\%$), the quality cannot meet reuse standards due to the high concentration of COD ($101.5 \pm 22.9 \text{ mg}\cdot\text{L}^{-1}$) and TN ($132.5 \pm 16.9 \text{ mg}\cdot\text{L}^{-1}$) in the effluent. It showed the water production may contain more organic and inorganic matters from the AnMBRs than from MBRs. In addition, it has been reported that AnMBRs caused serious membrane fouling potential with high mixed liquor suspended solid (MLSS) concentration [151] and need longer biomass retention time to support the slow-growing anaerobic microorganisms than aerobic MBRs [152]. Therefore, further treatment process after AnMBR, such as physically adsorption, precipitation, advanced membrane filtration or disinfection, is necessary to control the fouling potential and increase the treatment efficiency of AnMBRs, when applied to water reuse processes [153].

In summary, UF/MF-based treatment and MBR processes can achieve non-potable applications. MBR can work as the secondary treatment in WWTPs but with high removal efficiency of contaminants. However, the fouling potential will be higher in submerged MBR than separate MF or UF due to the sludge degradation and separation and the optimization/control of operating conditions must be done simultaneously (biological and membrane processes). Consequently, for more advanced reuse applications, complementary treatments such as NF, RO or forward osmosis (FO) are often needed, and these will be the subject of the following sections.

Table 1-6 Membrane bioreactors (MBR) for water reuse applications.

Membrane Process (Scale and Operating Duration)	Operating Conditions	Feed Characteristics	Permeate Quality/Removal Rate (in Average)	Application for Reuse	Standard Basis	Ref.
Aerobic MBR+ GAC (water recycle plant, 12 months)	Feed: Primary effluent of municipal wastewater recycling plant UF: 0.04 μm HRT: 3.2 h (MBR) + 0.58 h (GAC)	BOD ₅ : 46.2–262.1 $\text{mg}\cdot\text{L}^{-1}$ COD: 142.0–512.0 $\text{mg}\cdot\text{L}^{-1}$ SS: 47.5–240 $\text{mg}\cdot\text{L}^{-1}$	BOD ₅ : < 1.9 $\text{mg}\cdot\text{L}^{-1}$, >96% COD: < 48.3 $\text{mg}\cdot\text{L}^{-1}$, >65.9% SS: < 7.2 $\text{mg}\cdot\text{L}^{-1}$, >85% Fecal coliforms: 0.3 CFU·100 mL^{-1} Phages: 3.9–5.6 log reduction	Non-potable	California Department of Public Health	[154]
Aerobic submerged MBR (pilot-scale, 50 days)	Feed: black water from household MF: 0.4 μm Flux: 30–40 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ HRT: 36 h	pH: 7.6 BOD ₅ : 406 $\text{mg}\cdot\text{L}^{-1}$ Total COD: 1218 $\text{mg}\cdot\text{L}^{-1}$ Soluble COD: 417 $\text{mg}\cdot\text{L}^{-1}$ TN: 188 $\text{mg}\cdot\text{L}^{-1}$ NH ₄ ⁺ -N: 155 $\text{mg}\cdot\text{L}^{-1}$ TP: 21.3 $\text{mg}\cdot\text{L}^{-1}$ TSS: 560 $\text{mg}\cdot\text{L}^{-1}$ Total coliform: >10 ⁶ 100 mL^{-1}	BOD ₅ : 8 ± 4 $\text{mg}\cdot\text{L}^{-1}$, 98% Total COD: 42 ± 8.81 $\text{mg}\cdot\text{L}^{-1}$, 96% TSS: 2 ± 1.19 $\text{mg}\cdot\text{L}^{-1}$, 99% TN: 19 ± 4.73 $\text{mg}\cdot\text{L}^{-1}$, 89% NH ₄ ⁺ -N: 11 ± 3.76 $\text{mg}\cdot\text{L}^{-1}$, 92% NO _x -N: 8 ± 3.1 $\text{mg}\cdot\text{L}^{-1}$ Total coliforms: 0, 100%	Toilet flushing, Cleaning, Irrigation	Water reuse standards of EPA, EU, WHO, Turkey	[132]
Aerobic submerged MBR (pilot plant, 6 months)	Feed: hotel greywater UF: 0.04 μm , hollow fiber Flux: 20 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$	COD: 41–500 $\text{mg}\cdot\text{L}^{-1}$ BOD ₅ : 36–295 $\text{mg}\cdot\text{L}^{-1}$ TN: 2.6–25 $\text{mg}\cdot\text{L}^{-1}$ Ammonia N: 0.3–14 $\text{mg}\cdot\text{L}^{-1}$ Total count: 1.5 × 10 ⁷ –4.1 × 10 ⁷ CFU·100 mL^{-1} Total coliforms: 1.4 – 4.1 × 10 ⁶ CFU·100 mL^{-1} <i>E. coli</i> : < 1.1 × 10 ⁶ CFU·100 mL^{-1}	COD: <36 $\text{mg}\cdot\text{L}^{-1}$ TN: <10 $\text{mg}\cdot\text{L}^{-1}$ Ammonia N: <8 $\text{mg}\cdot\text{L}^{-1}$ Total count: 5.8 × 10 ³ –1.6 × 10 ⁵ CFU·100 mL^{-1} Total coliforms: 0.27 × 10 ² –2.1×10 ² CFU·100 mL^{-1} <i>E. coli</i> : <1.1 × 10 ² CFU·100 mL^{-1} <i>Legionella spp.</i> : < 1 CFU·100 mL^{-1}	Non-potable reuse	Spanish water reuse standard	[155]

Aerobic MBR (External): Pilot plant for 162 days	Feed: urban wastewater UF: 0.02 $\mu\text{m}/300$ kDa Flux: 75–150 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ HRT: 5 d SRT: 5–30 days	COD: 200–800 $\text{mg}\cdot\text{L}^{-1}$ SS: 100–600 $\text{mg}\cdot\text{L}^{-1}$ NH ₃ -N: 10–30 $\text{mg}\cdot\text{L}^{-1}$ Coliform: 10 ⁵ –10 ⁶ ·L ⁻¹ Turbidity: 50–70 NTU pH: 7.5–8.5	COD: 9.4 $\text{mg}\cdot\text{L}^{-1}$, 97%; SS: nd,100%; NH ₃ -N: 0.2–1.3 $\text{mg}\cdot\text{L}^{-1}$, 96.2%; Turbidity: < 2 NTU Coliform: nd pH: 8.2	Directly for municipal purposes or indirectly for industrial uses after additional treatment	Water reuse standard of China	[156]
Vertical submerged MBR (pilot scale for 600 days)	Feed: municipal wastewater in Korea UF: 0.45 μm Flux: 6.2 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ HRT: 8 h SRT: 60 days	COD: 232 ± 41 $\text{mg}\cdot\text{L}^{-1}$ TSS: 220 ± 52 $\text{mg}\cdot\text{L}^{-1}$ TN: 42 ± 5 $\text{mg}\cdot\text{L}^{-1}$ TP: 3.2 ± 0.4 $\text{mg}\cdot\text{L}^{-1}$ Volatile fatty acids: <1.0 $\text{mg}\cdot\text{L}^{-1}$ pH: 7.3 ± 0.1 Alkalinity as CaCO ₃ : 145 ± 47 $\text{mg}\cdot\text{L}^{-1}$	COD: 9.0 ± 3.6 $\text{mg}\cdot\text{L}^{-1}$, 96% TSS: 220 ± 52 $\text{mg}\cdot\text{L}^{-1}$ TN: 10.6 ± 2.6 $\text{mg}\cdot\text{L}^{-1}$, 74% TP: 0.7 ± 0.2 $\text{mg}\cdot\text{L}^{-1}$, 78% Total colony counts: 24 CFU·mL ⁻¹ Turbidity: 0.18 NTU, pH: 7.3	Urban or rural reuse, such as toilet flushing, sprinkling and car washing	Drinking water standards of Korea and the WHO	[157]

HRT: hydraulic retention time; SRT: solids retention time; nd: not detected.

1.2.4. NF/RO/FO-Based Treatment Processes for Water Reuse

Nanofiltration (NF) or reverse osmosis (RO) have been recognized as an effective means to provide safe and reliable source of water supplies for both potable water and non-potable water purposes [120]. NF/RO technologies are outstanding in wastewater reuse applications, especially in potable reuse levels, because NF/RO can eliminate effectively organic micropollutants such as endocrine-disrupting compounds, pharmaceutically active compounds, pesticides, etc. [158,159]. NF/RO processes also show significant removal efficiency on conductivity, TDS, alkalinity, salinity, hardness and ions from the feed which can be of particular interest for irrigation and potable purposes. In addition, forward osmosis (FO) is a promising membrane technology in the wastewater treatment field using a similar membrane to RO or NF membranes, but an osmotic pressure difference as a driving force [160–162]. Table 1-7 summarizes the NF/RO-based membrane treatment studies applied to municipal wastewater reuse and most of them using non-conventional pretreatment processes before NF/RO.

The separation characteristics of NF include pore-size steric mechanism (convective flow), solution–diffusion mechanisms and electrostatic interactions [87]. Dalar et al. [163] demonstrated that the coupled MBR and NF system could remove almost all divalent and multivalent ions, such as PO_4^{3-} and SO_4^{2-} , but only removed less than 60% monovalent ions, such as NO_3^- , Na^+ and Cl^- . Similar results were also obtained by Bunani et al. [164]. Actually, ion rejection by NF membranes depends on the combination of electrostatic and steric interactions associated with charge shielding, Donnan exclusion and the degree of ion hydration [165]. In addition, rejection mechanisms by NF highly depend on membrane properties, physicochemical properties of solutes and feed characteristics. Whereas RO was originally conceived as a method of producing potable water from the sea or brackish sources from the 1960s, the technology now is also widely applied on a large scale in municipal wastewater treatment and reuse [166,167]. The rejection mechanism of RO includes size exclusion, charge exclusion and physical–chemical interactions between solute and solvent [168,169]. RO retention capacities are better than NF due to its almost nonporous nature, reflected in higher removal of organic compounds, monovalent ions, TDS and conductivity [163,170,171]. As listed

in Table 1-7, the conductivity removal by NF is usually effective, but not completely, ranging between 40% and 90%, while RO shows excellent separation performance on almost complete salinity removal [172,173]. However, it should be noted that some NF and RO membranes materials are selective in removing micropollutants probably related to their rejection mechanisms [174]. For instance, the removal efficiency diminished considerably for micropollutants with a neutral or positive charge, of which the removal through RO was near 100% for naproxen, versus 20% for acetaminophen and 60% for atenolol [175]. Forward osmosis (FO) also called as direct osmosis is a new membrane-separation process in which water moves spontaneously across a semi-permeable membrane from the feed solution (lower osmotic pressure) side to the draw solution (higher osmotic pressure) side [176,177]. Compared to NF/RO, FO is an osmotically driven technology operated at very low or even non-hydraulic pressure during wastewater treatment [178], and FO membranes have a lower membrane fouling potential due to loose formation and less compaction of cake foulants in the absence of hydraulic pressure [179]. Moreover, FO membranes present high contaminant rejection rates, high flux recovery after cleaning and high water recovery using low-grade energy resources [180,181]. However, it should be noted that when applying FO processes to wastewater treatment, further treatment such as NF, RO, MBR or membrane distillation, is required to simultaneously separate product water for reuse and recover draw solution [182–184]. One of the most common combinations is the hybrid FO-RO system, in which the inlet wastewater firstly diffuses into high concentrated draw solution through the FO membrane, and then the diluted draw solution will be filtered by RO to separate water production and continuously reconstitute the draw solution [180].

However, due to their tiny and/or nonporous characteristics, the requirements of feed water quality for NF and RO membranes are much higher than MF and UF, because even small particles such as the NOM and submicron particulates in the feed are prone to cause severe fouling problems on these membranes [185,186]. In particular, biofouling, the irreversible adhesion on a membrane caused by microorganisms and the extracellular polymers, has been considered as the main obstacle during long term NF/RO operation [186–189]. In order to improve filtration performance, pretreatment processes ahead of NF/RO are required to reduce the potential of fouling development [120,190]. Municipal wastewater treatment by hybrid pretreatment with NF/RO processes improves the produced water quality to potable reuse level.

Table 1-7 NF / RO/ FO coupled with pretreatment processes for wastewater reuse applications.

Process (Scale and Operation Duration)	Feed Water	Operating Conditions	Feed Characteristics	Final permeate quality (Removal rate)	Application for Reuse	Standard Basis	References
Submerged MBR+ NF	Municipal wastewater after primary treatment	UF: 200 kDa, hollow fiber NF: 150–300 Da, TMP: 0.1–0.5 bar (UF); 41 bar (NF)	(raw wastewater) EC: $1174 \pm 2 \mu\text{S}\cdot\text{cm}^{-1}$ pH: 7.22 ± 0.11 TSS: $488 \pm 48 \text{ mg}\cdot\text{L}^{-1}$ Turbidity: $248 \pm 11 \text{ NTU}$ DOC: $126.6 \pm 7.3 \text{ mg}\cdot\text{L}^{-1}$ COD: $478 \pm 132 \text{ mg}\cdot\text{L}^{-1}$ F: $0.096 \pm 0.003 \text{ mg}\cdot\text{L}^{-1}$ Cl ⁻ : $156.0 \pm 2.4 \text{ mg}\cdot\text{L}^{-1}$ NO ₂ ⁻ : $64.35 \text{ mg}\cdot\text{L}^{-1}$ NO ₃ ⁻ : $144.53 \pm 42.17 \text{ mg}\cdot\text{L}^{-1}$ PO ₄ ³⁻ : $9.631 \pm 1.428 \text{ mg}\cdot\text{L}^{-1}$ SO ₄ ²⁻ : $36.33 \pm 0.84 \text{ mg}\cdot\text{L}^{-1}$ Na ⁺ : $71.14 \pm 0.48 \text{ mg}\cdot\text{L}^{-1}$ K ⁺ : $11.85 \pm 0.14 \text{ mg}\cdot\text{L}^{-1}$ Mg ²⁺ : $22.05 \pm 0.04 \text{ mg}\cdot\text{L}^{-1}$ Ca ²⁺ : $110.7 \pm 0.2 \text{ mg}\cdot\text{L}^{-1}$ SAR: 1.61 ± 0.01	EC: $397 \mu\text{S}\cdot\text{cm}^{-1}$ pH: 8.06 TSS: $0 \text{ mg}\cdot\text{L}^{-1}$ Turbidity: 0.23 NTU DOC: $0.35 \text{ mg}\cdot\text{L}^{-1}$ COD: $< 5 \text{ mg}\cdot\text{L}^{-1}$ F: n.a. $\text{mg}\cdot\text{L}^{-1}$ Cl ⁻ : $63.77 \text{ mg}\cdot\text{L}^{-1}$ NO ₂ ⁻ : $0.3728 \text{ mg}\cdot\text{L}^{-1}$ NO ₃ ⁻ : $63.1 \text{ mg}\cdot\text{L}^{-1}$ PO ₄ ³⁻ : n.a. $\text{mg}\cdot\text{L}^{-1}$ SO ₄ ²⁻ : $0.464 \text{ mg}\cdot\text{L}^{-1}$ Li ⁺ : nd Na ⁺ : $38.01 \text{ mg}\cdot\text{L}^{-1}$ NH ₄ ⁺ : nd K ⁺ : $5.9 \text{ mg}\cdot\text{L}^{-1}$ Mg ²⁺ : $3.04 \text{ mg}\cdot\text{L}^{-1}$ Ca ²⁺ : $29.9 \text{ mg}\cdot\text{L}^{-1}$ SAR: 1.77	Agricultural Irrigation (50% of MBR effluent and 50% of NF permeate)	WHO and FAO guidelines	[163]
NF	MBR effluent from domestic wastewater	Lp ₀ : $6.2 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ TMP: 8 bar	UV _{254 nm} : 0.148–0.155 UV _{210 nm} : 1.579–3.207 TOC: 6.0–8.0 $\text{mg}\cdot\text{L}^{-1}$ COD: 12–13 $\text{mg}\cdot\text{L}^{-1}$ Mg ²⁺ : 8.9–9.8 $\text{mg}\cdot\text{L}^{-1}$ Ca ²⁺ : 25.0–28.4 $\text{mg}\cdot\text{L}^{-1}$ EC: 631–894 $\mu\text{S}\cdot\text{cm}^{-1}$	UV _{254 nm} > 95% UV _{210 nm} : 75–81% TOC: 82–95% EC: 92–94%	Not mentioned	Not mentioned	[170]

UF+NF	Synthetic municipal wastewater after aerobic activated sludge	UF: 30 kDa, PES NF: 270 Da, PA TMP: 1–6 bar	COD: 243.34 mg·L ⁻¹ TP: 7.53 mg·L ⁻¹ NH ₃ -N: 0.67 mg·L ⁻¹ NO ₂ -N: 4.32 mg·L ⁻¹ NO ₃ -N: 34.43 mg·L ⁻¹	COD: 3.68 mg·L ⁻¹ TP: 0.19 mg·L ⁻¹ NH ₃ -N: 0.14 mg·L ⁻¹ NO ₂ -N: 0.14 mg·L ⁻¹ NO ₃ -N: 1.37 mg·L ⁻¹	Inner industrial reuse, Garden irrigation	China municipal water reuse standards	[191]
Forward osmosis (FO) + NF	Secondary effluent from WWTP	Flux: 2.4 L·m ⁻² ·h ⁻¹ for FO Flux: 3.3 or 6.6 L·m ⁻² ·h ⁻¹ for NF	<i>E. coli</i> : 0 CFU·100 mL ⁻¹ TSS: < 1 mg·L ⁻¹ Turbidity: 0.22 NTU EC: 5.33 dS·m ⁻¹ SAR: 10.6 meq·L ⁻¹ B: 1.17 mg·L ⁻¹ Arsenic: 0.0015 mg·L ⁻¹ Chrome: 0.0041 mg·L ⁻¹ Copper: 0.002 mg·L ⁻¹ Manganese: 0.018 mg·L ⁻¹ Molybdenum: 0.002 mg·L ⁻¹ Nickel: 0.0016 mg·L ⁻¹ Selenium: <0.004 mg·L ⁻¹	Conductivity: 1 mS·cm ⁻¹ B: < 0.4 mg·L ⁻¹ SAR: 1.98 meq·L ⁻¹	Agricultural irrigation	Spanish water reuse legislation (RD1620/2007)	[184]
OMBR+RO	Synthetic wastewater	FO: flat sheet, thin-film composite (TFC), 0.5-M NaCl draw solution. RO: flat sheet, TFC, polyamide Flux: 4–8 L·m ⁻² ·h ⁻¹	Glucose: 100 mg·L ⁻¹ Peptone: 100 mg·L ⁻¹ KH ₂ PO ₄ : 17.5 mg·L ⁻¹ MgSO ₄ : 17.5 mg·L ⁻¹ FeSO ₄ : 10 mg·L ⁻¹ CH ₃ COONa: 225 mg·L ⁻¹ Urea: 35 mg·L ⁻¹ 31 TrOCs: 5 µg·L ⁻¹ for each TrOC	The system achieved the effective removal of bulk organic matter, nutrients and almost complete removal of all 31 trace organic contaminants investigated	Not mentioned	Not mentioned	[192,193]
RO	MBR effluent from domestic wastewater	TMP: 4–12 bars Initial Permeability: 3.6 L·m ⁻² ·d ⁻¹ ·bar ⁻¹	UV _{254 nm} : 0.148–0.155 UV _{210 nm} : 1.579–3.207 TOC: 6.0–8.0 mg·L ⁻¹ Mg ²⁺ : 8.9–9.8 mg·L ⁻¹ Ca ²⁺ : 25.0–28.4 mg·L ⁻¹ EC: 631–894 µS·cm ⁻¹	UV _{254 nm} > 95% UV _{210 nm} : 90–97% TOC: 91–98% EC: 96–98%	Not mentioned	Not mentioned	[170]

MF+RO	Secondary treated effluent from sewage	MF: 26 m ³ ·d ⁻¹ ; 0.4 μm RO: 19 m ³ ·d ⁻¹	TSS: 2 mg·L ⁻¹ BOD ₅ : 3 mg·L ⁻¹ COD: 23 mg·L ⁻¹ TKN: 3.1 mg·L ⁻¹ Nitrate N: 4.7 mgN·L ⁻¹ TDS: 364 mg·L ⁻¹ pH: 7.2 Color: 44 Hazen Alkalinity: 71 mg·L ⁻¹ Si: 11.7 mg·L ⁻¹ Turbidity: 0.6 NTU <i>E. coli</i> : 2.8 × 10 ⁵ CFU·100 mL ⁻¹ Virus: 97 PFU·100 mL ⁻¹ Total Estrogens: 38 μg·L ⁻¹ Odor: 2	TSS: < 2 mg·L ⁻¹ BOD ₅ : < 2 mg·L ⁻¹ COD: < 2 mg·L ⁻¹ TKN: 0.3–0.4 mg·L ⁻¹ Nitrate N: 0.71–1.43 mgN·L ⁻¹ TDS: 17–24 mg·L ⁻¹ pH: 5.3–5.5 Color: < 2.5 Hazen Alkalinity: 2.7–3.3 mg·L ⁻¹ Si: 0.3–0.7 mg·L ⁻¹ EC: 24–33 μS·cm ⁻¹ <i>E. coli</i> : nd Virus: nd Total Estrogens: < 4.4 μg·L ⁻¹ Odor: 1	Both for potable and non-potable reuse	EPA and WHO guidelines	[134]
MBR+RO (pilot scale, 112 days)	Primary municipal wastewater	UF: 0.04 μm, hollow fiber. TMP: 0.42 bar (UF), 15.2 bar (RO)	TSS: 100–1930 mg·L ⁻¹ Turbidity: 7–308 NTU COD: 122–2205 mg·L ⁻¹ DOC: 2.12–10.21 mg·L ⁻¹ (after MBR) UV ₂₅₄ : 0.30–4.00 TN: 12.6–205 mg·L ⁻¹ Pb: 1–16 μg·L ⁻¹ Ni: 1–33.7 μg·L ⁻¹ Cu: 1–1345 μg·L ⁻¹ Cr: 1–746 μg·L ⁻¹	TSS: <1 mg·L ⁻¹ Turbidity: 0.01–0.13 NTU COD: < 32 mg·L ⁻¹ DOC: 1.04–4.1 mg·L ⁻¹ UV ₂₅₄ : 0.001–0.01 TN: 17–21 mg·L ⁻¹ Pb: < 1 μg·L ⁻¹ Ni: < 1 μg·L ⁻¹ Cu: < 1 μg·L ⁻¹ Cr: < 1 μg·L ⁻¹	Not mentioned	Not mentioned	[194]
AnMBR+RO (lab scale)	Synthetic municipal wastewater	Flux: 20 L·m ⁻² ·h ⁻¹	COD: 400 mg·L ⁻¹ NH ₄ ⁺ -N: 45 mg·L ⁻¹ PO ₄ ³⁻ -P: 5 mg·L ⁻¹ NaHCO ₃ : 500 mg·L ⁻¹ CaCl ₂ ·2H ₂ O: 45 mg·L ⁻¹ MgSO ₄ ·7H ₂ O: 20 mg·L ⁻¹ FeSO ₄ ·7H ₂ O: 20 mg·L ⁻¹ FeCl ₃ ·7H ₂ O: 1.5 mg·L ⁻¹	NH ₄ ⁺ -N: 2.1 mg·L ⁻¹ PO ₄ ³⁻ -P: 0.03 mg·L ⁻¹ TOC: 0.13 mg·L ⁻¹ Sodium: 3.2 mg·L ⁻¹ Calcium: 0.05 mg·L ⁻¹ Iron: < 0.005 mg·L ⁻¹ Chloride: 4.7 mg·L ⁻¹ Sulfate: 0.5 mg·L ⁻¹	Discharge to reservoirs for indirectly potable reuse	Guidelines for NEWater in Singapore	[195]

NF-MBR+RO	Municipal wastewater	NF: 200–300 Da	COD: 389.8 ± 169.9 $\text{mg}\cdot\text{L}^{-1}$	NF permeate: COD: $99.6 \pm 0.8\%$ DOC: $0.5\text{--}2.5$ $\text{mg}\cdot\text{L}^{-1}$, $97.5\% \pm 1.8\%$ Biopolymer: nd Humic substances: 0.1 $\text{mg}\cdot\text{L}^{-1}$	Not mentioned	Not mentioned	[196]
		Flux: 10 $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$	DOC: 48.3 ± 13.9 $\text{mg}\cdot\text{L}^{-1}$ Ca^{2+} : 28.9 ± 3.5 Mg^{2+} : 7.9 ± 1.2 $\text{mg}\cdot\text{L}^{-1}$ Na^+ : 105.2 ± 6.9 $\text{mg}\cdot\text{L}^{-1}$ TN: 40.7 ± 6.1 $\text{NH}_4\text{-N}$: 40.9 ± 4.5 $\text{mg}\cdot\text{L}^{-1}$ $\text{NH}_3\text{-N}$: nd PO_4^{3-} : 23.6 ± 3.2 $\text{mg}\cdot\text{L}^{-1}$ EC: 859.5 ± 18.9 $\text{mS}\cdot\text{cm}^{-1}$				
		MLSS: 513 ± 96 $\text{mg}\cdot\text{L}^{-1}$					

SAR: sodium adsorption ratio; PPCPs: pharmaceuticals and personal care products; TKN: total Kjeldahl nitrogen; FAO: Food and Agriculture Organization; TrOCs: trace organic contaminants.

For instance, the hybrid MBR with NF or RO treatment system could both significantly remove all organic compounds investigated in this study with over 95% removal efficiency for all, thus producing an adequate permeate for indirect drinking water reuse [197]. The combination of AnMBR and RO systems could also achieve water reuse with direct energy recovery from municipal wastewater with minimized sludge production and significant energy savings [153,195].

In summary, NF/RO processes are usually used for higher level of water quality demand, such as potable water reuse. NF/RO has excellent removal capacity on removal of macro and micro components in wastewater, but the operation, maintenance and cleaning of membranes are more complicated and expensive which are not favorable for companies or industries. Although FO is developing fast recently, there are not many related reports and cases on the application of municipal wastewater treatment and reuse, and it usually needs a co-treatment process such as NF or RO which in turns increases the difficulty of realization. Therefore, the choice of membrane system should be based on feed water quality and its application purpose to forego unnecessary expenses. When the reuse level is not as high as potable reuse, it is not recommended to use NF/FO/RO processes, UF-based treatment process is more likely to be chosen.

1.2.5. Water Recovery and Energy Consumption of Membrane-Bases Processes for Water Reuse

The desired goal of researchers and companies on wastewater treatment for water reuse is to maximize economic benefits and provide optimum parameters for long term stable operation during membrane filtration. Although many articles are published in the field of municipal wastewater reuse, most of them focus on the feasibility and innovation of the processes, the hydraulic performance, the removal efficiency and optimization of the processes. Less are focused on the real productivity, wastewater recovery rate, energy consumption or total cost during long term operations. Among all the references mentioned in the above tables of this work, only a few of them stated the relevant energy consumption, mostly estimated in pilot scales (Table 1-8). The water recovery rate from wastewater by membrane-based technologies may be

influenced by feed water characteristics, the permeate consumption by physical and chemical cleaning and filtration time, etc. The net energy consumption may depend on the treatment technologies, the chemical reagents consumption and other supplementary energy production (e.g., methane), etc.

From Table 1-8, low-pressure driven membrane technologies (MF, UF, MBR) support higher productivity per unit area and present higher water recovery, over 85%. The water recovery for high-pressure driven membranes (NF, RO) and FO are relatively lower. Generally, the water recovery by RO itself only ranges from 50–75% because of the highly increased osmotic pressure during filtration [198]. In net energy comparison, low-pressure driven membrane technologies always have lower energy consumption. According to Atanasova et al. [155], with an inlet flowrate to an MBR over $15 \text{ m}^3 \cdot \text{d}^{-1}$, the net energy consumption was less than 0.5 kWh m^{-3} , even including the cost of disinfection. The net energy costs of an MBR on municipal wastewater treatment are relatively low ranging from 0.19 to $1.07 \text{ kWh} \cdot \text{m}^{-3}$ [147,199]. It should be noted the above data can only be used as a reference, not as the actual energy consumption for municipal wastewater reuse treatment. In addition, the processes with NF, RO and FO directly increased the energy consumption without consideration of biogas for energy recovery (Table 1-8), because of the increased TMP, membrane maintenance, etc.

In conclusion, as seen in real cases of municipal wastewater reuse treatment, the wastewater recovery rate and net energy consumption are more likely considered as the two main factors, on the basis of meeting required reuse level, because they are important indicators to determine whether the method is cost effective or not. In the future, it could be more interesting to focus on the economic prediction of membraned-based technologies, which will be the reference for the future development of wastewater reuse field.

1.2.6. Summary about membrane-based processes for water reuse

This above state of art gives a comprehensive insight on the global water reuse situation, reuse regulation and membraned-based treatment performance on wastewater reuse. The advantages and limitations of them on reuse purposes, are listed in Table 1-9 Membrane separation processes provide superior quality to the treated wastewater to meet local reuse

guidelines. As the legal national French context is not favorable to potable reuse, it was chosen to focus on non-potable reuse in this work and so advanced treatment such as NF, RO or FO will not be studied. In summary, UF-based membrane treatment and MBRs are both efficient approaches for non-potable reuses. Considering the state of art and the local context of wastewater management of this PhD, it was chosen to focus on UF as tertiary treatment after conventional activated sludge treatment. Indeed, UF can directly filtrate the secondary effluent without change of local WWTPs designs. Therefore, this Ph.D. work will focus on the performances and sustainability of UF membrane as the main urban wastewater tertiary treatment for water reuse applications. To be sustainable, the process needs optimized operating conditions including efficient backwash procedures which will be the topic of the next section.

Table 1-8 Wastewater recovery rate and energy consumption of membrane-based technologies.

Process	Productivity	Membrane Flux	Recovery Rate	Energy Consumption	Ref.
GAC + MF		30 L·m ⁻² ·h ⁻¹	98%		[122]
UF (pilot plant, 2 years)	< 0.7 m ³ ·h ⁻¹	17–22 L·m ⁻² ·h ⁻¹			[103]
UF (pilot plant, two months)	7.7–9.0 m ³ ·h ⁻¹	330–380 L·m ⁻² ·h ⁻¹	82.3–96.5%		[98]
UF	1.0 m ³ ·h ⁻¹	28.5 L·m ⁻² ·h ⁻¹	About 85%		[99]
FO + NF	0.16 (FO) and 0.187–0.35 (NF) m ³ ·h ⁻¹	FO: 2.4 L·m ⁻² ·h ⁻¹ ; NF: 3.3 or 6.6 L·m ⁻² ·h ⁻¹		3.44–4.57 kWh·m ⁻³	[184]
RO	0.79 m ³ ·h ⁻¹		65–75% (RO itself)		[134]
RO	0.04 m ³ ·h ⁻¹		50% (RO itself)		[194]
NF–MBR + RO		MBR permeate flux: 10 L·m ⁻² ·h ⁻¹ ; RO permeate flux: 20 L·m ⁻² ·h ⁻¹	90%	0.739 kWh·m ⁻³	[196]
UF–MBR + RO		MBR permeate flux: 10 L·m ⁻² ·h ⁻¹ ; RO permeate flux: 20 L·m ⁻² ·h ⁻¹	75%	0.732 kWh·m ⁻³	[196]
MBR + RO				0.518 kWh·m ⁻³	[153]
AnMBR + RO		AnMBR permeate flux: 10–20 L·m ⁻² ·h ⁻¹ RO permeate flux: 20 L·m ⁻² ·h ⁻¹	RO: 75% (RO itself)	0.333 kWh·m ⁻³ (consider energy recovery with methane)	[153]

Table 1-9 Advantages and disadvantages of various membrane-based treatment processes.

Treatment processes	Reuse level	Advantages	Disadvantages
MF/UF-based treatment	Non-potable reuse: toilet flushing, urban uses, irrigation, etc.	<ul style="list-style-type: none"> • Low-pressure driven process, high flux and high permeability. • Low energy cost • Effective removal of high molecular weight matters, bacteria and viruses. • UF can almost eliminate all the bacteria, protozoa and viruses, compared to MF. • MF/UF membranes pretreated with physical/chemical process results in low fouling potential, long term operation and higher load rate. 	<ul style="list-style-type: none"> • Health risks potential for humans. • Incomplete removal of low molecular weight matters, dissolved organics, salinity and micropollutants, etc. • Fouling potential
MBRs	Non-potable reuse: Toilet flushing, cleaning, process water, urban uses, irrigation, etc.	<ul style="list-style-type: none"> • Low-pressure driven process, high flux and high permeability. • Low energy cost • Effective removal of organics, TSS, nutrients like N, P, S in various forms, surfactants and micropollutants from various wastewater with biologic process. • Less organic foulants on the membrane, smaller footprint, faster plant activation, no biologic sedimentation units and less sludge production compared to CAS process. • Particularly, AnMBRs hold significant potential to reduce the overall energy demand, together with resource recovery 	<ul style="list-style-type: none"> • Health risks potential for humans. • Incomplete removal of low molecular weight matters, dissolved organics, salinity and micropollutants, etc. • Fouling potential with membrane pore-clogging and sludge cake deposition
NF/RO-based treatment	Non-potable and potable reuse: agricultural irrigation, groundwater recharge, indirect potable water, etc.	<ul style="list-style-type: none"> • High removal efficiency of micropollutants, microorganisms and salinity, EC, other dissolved organic and inorganic matters. • Reduce human health concerns • High level of reuse applications 	<ul style="list-style-type: none"> • High-pressure driven process, with low flux and low permeability. • High energy cost. • Pretreatment demand • Biofouling caused by microorganisms.
FO-based treatment	Non-potable and potable reuse: agricultural irrigation, groundwater recharge, indirect potable water, etc.	<ul style="list-style-type: none"> • FO: non external pressure driven process • Lower membrane fouling potential than RO due to less formation and compaction of cake layers on FO membranes in the absence of hydraulic pressure • High flux recovery after cleaning and high water recovery using low-grade energy resources • Combination of municipal wastewater treatment with seawater desalination 	<ul style="list-style-type: none"> • FO is a process of dilution, which needs further separation treatment. • High requirements for the selection of draw solution and membrane material

1.3. UF membrane fouling control and management by different types of backwashes

While the optimization of UF operating parameters can control membrane fouling, membrane cleaning is more important on the fouling removal. Physical cleanings such as classic backwash (CB) and air assisted backwash (AB) are commonly used methods in membrane fouling control [19]. CB can loosen and detach fouling cake partially from membrane surface, and AB is a more effective anti-fouling method compared to CB [20–22]. Normally, AB can be operated either by air sparing in backwash water, or by air injection into membranes fibres. However, physical backwash is mostly powerless on irreversible fouling removal during long term filtration, and the residual foulants after backwashes would be recompressed after multi-cycle filtration [131]. The conventional chemical cleaning plays an important role on irreversible fouling separation by chemical reagents soaking/reacting with membrane fouling which can damage the foulant-foulant and membrane-foulant interactions [16]. However, using of high concentrated acids, soda, and/or oxidants/disinfectants and longer soaking time will cause irreversible damages gradually on membrane properties and filtration performances in long-term operation [15,17,18]. In this case, two new concept appear as promoter (i) an air injection before classical backwash to strongly reduce the fouling and to destroy organic matter [200–202] and (ii) a chemically assisted maintenance backwash, the combined physical backwash with lower concentrated oxidants/disinfectants (compared to chemical cleaning) in backwash water, has been developed for permeability maintenance improvement recently, so as to reduce the need for intensive chemical cleanings [23,24].

Sodium hypochlorite (NaOCl) improperly often called “chlorine” is one of the commonly used oxidants/disinfectants, which can inactivate the microorganisms and oxidize the organic foulants to be more hydrophilic and easier to detach from the membrane surfaces [203–205]. Studies have also found that the presence of ClO^- is effective in terms of extracellular polymeric substances (EPS) disruption and microbial cell damage [206]. Researchers usually use the concentration total chlorine or free chlorine to illustrate the quantity of chlorine they add in solutions. Free chlorine is the amount of chlorine that is able to sanitize contaminants. Besides, combined chlorine refers to chlorine that has combined directly with the contaminants to intermediate breakdown products.

Total chlorine is basically the sum of free chlorine and combined chlorine [207]. In this study, the commonly mentioned chlorine concentration refers to total concentration unless otherwise specified as free chlorine. NaOCl is an oxidant which is unstable under atmosphere with high temperature, light, strong acid, or other reductants. NaOCl is also a strong base and weak acid salt that can hydrolyze and produce HOCl, $\text{NaOCl} + \text{H}_2\text{O} \rightarrow \text{Na}^+ + \text{HOCl} + \text{OH}^-$. The microbicidal activity of chlorine is largely attributed to undissociated HOCl. The dissociation of HOCl to the less microbicidal form (OCl^-) depends on pH. The disinfecting efficacy of chlorine decreases with increasing pH, which is parallel to the conversion of undissociated HOCl to OCl^- [208,209]. Fukuzaki et al. [210] reported that the bactericidal activity of $2.38 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ (free chlorine) NaOCl was increased with pH decreasing from 9.3 to 5.7 because the HOCl amount was increased from 13% (pH 9.3) to 98% (pH 5.7). In conventional chemical cleanings, NaOCl is usually used at high levels ($300\text{-}3000 \text{ mg NaOCl} \cdot \text{L}^{-1} / 286\text{-}2860 \text{ mg Cl}_2 \cdot \text{L}^{-1}$) and in long time soaking for powerful membrane permeability recovery and irreversible fouling removal [23,211,212]. However, high dosage of NaOCl will accelerate the adversely effects on membrane properties and performance [212], by-products formation, and higher operating costs [9, 20]. Normally, the accumulated amount of NaOCl in contact with membranes can be expressed as a total dose calculated in total chlorine concentration multiplied by contact time (CT) which can provide reference for the degradation progress of membranes. Hanafi et al. reported HClO and $\cdot\text{OH}$ are the responsible species for the chain-scission of polysulfone, polyamide, polyethersulfone (PES) membranes [214–217]. Therefore, the accumulated amount of NaOCl in membrane can cause degradation of membrane materials and membrane aging. PES membranes are considered highly tolerant to oxidants [218]. Wienk et al. [219] detected the aging of PES/PVP membrane under exposure to $6000 \text{ mg} \cdot \text{L}^{-1} \cdot \text{day}$ of NaOCl (approximately equal to $5718 \text{ mg Cl}_2 \cdot \text{L}^{-1} \cdot \text{day}$). Yadav et al. [220] observed the PES membrane surface damage under exposure to $10\,000\text{-}25\,000 \text{ mg} \cdot \text{L}^{-1} \cdot \text{day}$ of NaOCl (approximately equal to $9530\text{-}23853 \text{ mg Cl}_2 \cdot \text{L}^{-1} \cdot \text{day}$), and the damage become worse with increasing amount of NaOCl under the same conditions. While ultrafiltration is mechanically and chemically stressed, the combined oxidant with physical backwash may accelerate physical damage and/or chemical degradation of membrane materials [221]. According to Fukuzaki *et al.* [222], low concentrations of OCl^- could destruct the exoplasmic organic matrix, while high concentrations of OCl^- led to diffusions of OCl^- into cells and disruption of cell metabolisms. Therefore, the new chemically assisted maintenance backwash firstly proposed by Wang *et al.*

[23] with low dose of chlorinated disinfectants was developed and investigated on membrane cleaning efficiency. The effectiveness of the combined backwash with low dosage ($<300 \text{ mg}\cdot\text{L}^{-1}$) of NaOCl on membrane fouling removal has been mentioned in several publications (Table 1-10). Some studies used to add low level of NaOCl into backwash water for disinfection but without special research on the impact of NaOCl addition on cleaning efficiency, such as Wang *et al.* [223], and Liu *et al.* [224]. In 2001, Decarolis *et al.* [225] demonstrated that backwashing with chlorine addition significantly improved UF membrane productivity. Afterwards, Wang *et al.* [23] investigated the impacts of NaOCl-assisted backwash on the hydraulic filtration performances of membrane bioreactor (MBR) under different NaOCl loads. Yue *et al.* [24] did a same study scheme as Wang *et al.* [23] but with a different bioreactor which was anaerobic ceramic membrane bioreactor. In result, they both confirmed that backwash with low dosage of NaOCl enhanced the organic foulant degradation and inhibited microbial regrowth on membranes. Wang *et al.* [23] also mentioned that the NaOCl-assisted backwash at lower NaOCl concentrations and higher backflush frequencies made less adverse effects on the functional groups of the active layer of the membranes. Besides, Zhang *et al.* [226] stated that $95.3 \text{ mg Cl}_2\cdot\text{L}^{-1}$ NaOCl solution exhibited the best performance in removing the irreversible fouling resistance ($88.4\% \pm 1.1\%$) from the algal-fouled UF membrane, compared to the solutions with $500 \text{ mg}\cdot\text{L}^{-1}$ NaOH, $500 \text{ mg}\cdot\text{L}^{-1}$ HCl and $150 \text{ mg}\cdot\text{L}^{-1}$ ethylene diamine tetra acetic acid. This might be attributed to the fact that NaOCl could eliminate major foulants such as carbohydrate-like and protein-like materials on the membrane surface. In result, the cases in Table 1-10 confirmed the cleaning effectiveness of disinfectants (mostly NaOCl) addition in backwash, but their concentrations varied under different conditions.

The membrane fouling strategies fed by municipal secondary effluent includes organic fouling, inorganic fouling, and microbial fouling [15,227,228]. Organic fouling is the main cause of irreversible fouling for membranes. Nonetheless, the efficiency and practicality of NaOCl-assisted backwash has not been studied in real UF pilot scale and even less with periodic classic backwash (CB) and air backwash (AB). Therefore, this subject will investigate the influence of NaOCl-assisted CB and/or AB on the hydraulic filtration performance and fouling management in semi-industrial UF pilot plant. Besides, the impacts on physicochemical properties of membrane material under different addition frequency of NaOCl and the performance of chlorinated CB and AB will also be discussed.

Table 1-10 Studies on the combined physical backwash with low concentrated oxidants/disinfectants in backwash water

Backwash types	Concentration	Backwash time × flux or TMP	Filtration time × flux	Feed water	Membrane process (pore size/area)	Remarks	Ref.
1 NaOCl-assisted backwash	23.8 mg Cl ₂ ·L ⁻¹	24 s / (207-241.5 kPa)	(15-30 min) × (34-102 L·m ⁻² ·h ⁻¹)	Tertiary treated wastewater	UF (150 kDa/1.9 m ²)	<ul style="list-style-type: none"> • Frequent backwash with chlorine addition significantly improved membrane productivity, primarily due to enhanced foulant removal by organic oxidation and bio-growth control. 	[225]
2 NaOCl-assisted backwash	0.191 mg Cl ₂ ·L ⁻¹ (optimized)	15 min × 8.33 L·m ⁻² ·h ⁻¹	12h × 6 L·m ⁻² ·h ⁻¹	Synthetic municipal wastewater	MBR (0.01 μm/0.1 m ²)	<ul style="list-style-type: none"> • NaOCl backflush enhanced the detachment of biopolymers from the fouled membranes, and enhanced the denitrification of MBR. • Low level NaOCl assisted backflush has slight or few adverse effects on sludge and membranes 	[23]
3 NaOCl-assisted backwash	0.953 mg Cl ₂ ·L ⁻¹ (optimized)	30s × 30 L·m ⁻² ·h ⁻¹	9 min × 10 L·m ⁻² ·h ⁻¹	Domestic wastewater	anaerobic ceramic MBR (0.08 μm/0.08 m ²)	<ul style="list-style-type: none"> • The biodegradability of organics in the wastewater and the microbial activities of biomass were improved with low level of NaOCl assisted backwash. • High level of NaOCl assisted backwash deteriorated cell metabolism and led to excessive production of cell lytic products. 	[24]

4	Backwash with chlorinated water	3 mg Cl ₂ ·L ⁻¹	2 min × 60 L·m ⁻² ·h ⁻¹	58 min × 20 L·m ⁻² ·h ⁻¹	Lake water/the Yangtze River water/micro-polluted water/municipal secondary effluent	UF (100 kDa/29.0 cm ²)	<ul style="list-style-type: none"> • The use of chlorinated-water backwashing decreased the number of microorganisms in the biofouling layer, but increased the level of EPS; thus, the membrane fouling resistance decreased by 8.6%. 	[203]
5	NaOCl-assisted backwash	95.3 mg Cl ₂ ·L ⁻¹	1 h soaking	(TMP up to 40 kPa) × 20 L·m ⁻² ·h ⁻¹	Algal-rich water	UF (0.01 μm/0.025 m ²)	<ul style="list-style-type: none"> • Among the tested cleaning reagents (NaOH, HCl, EDTA, and NaOCl), 95.3 mg Cl₂·L⁻¹ NaOCl exhibited the best performance (88.4% ± 1.1%) in removing the irreversible fouling resistance. • The surface morphology of the fouled membrane almost recovered the original state of new membrane after cleaning with NaOCl. 	[229]
6	NaOCl-assisted backwash (CEB)	284 mg Cl ₂ ·L ⁻¹ (optimized)	5.22 L·m ⁻² ·h ⁻¹	-	Synthetic medium contained (NH ₄) ₂ SO ₄ , NaNO ₂ and some trace elements	MBR (0.01 μm/0.05 m ²)	<ul style="list-style-type: none"> • The best cleaning effect was evident at 284 mg Cl₂·L⁻¹ of NaOCl (optimized). 	[230]

MBR: membrane bioreactor.

The units of chlorine used in different references were all uniformed to mg Cl₂·L⁻¹, based on the relationship between NaOCl and Cl₂: 1 mg·L⁻¹ NaOCl = 0.953 mg Cl₂·L⁻¹.

1.4. Thesis statement

This Ph.D. work is aimed to assess the performances of a semi-industrial UF pilot plant as urban wastewater tertiary treatment for water reuse, especially on best filtration conditions, water quality including microplastic particles removal by UF, and membrane fouling control and management. The flow chart of the technical strategy points for this dissertation is given in Appendix I.

According to Chapter 1, UF appears to be an effective technology for tertiary treatment with the objective of reusing water. Although UF has already been studied in the wastewater reuse field in various regions and showed good potentiality, there are still some problems that need to be explored and improved in order to be widely used.

This thesis focused on the following issues:

1) Most publications are experimented in ideal environment or lab-scale and this work will use a semi-industrial pilot plant continuously fed with real WWTP effluent. This was possible thanks to a partnership with la Société des Eaux de Marseille Métropole which gave access to a conventional WWTP in Châteauneuf-les-Martigues (13), France.

2) Few references considered the overall operating conditions (such as flux, filtration time, backwash sequence) for best performances of UF system in this specific application and this thesis will provide these optimal operating conditions including best flux, filtration time and AB frequency.

3) The water reuse standards of different regions are becoming stricter, in particular in Europe with a new regulation released in 2020 and this work will compare the UF permeate quality with this new regulation among others. It will also assess the type, size and concentration of microplastic particles in waters with a dedicated quite new technique developed by PerkinElmer. This analytical technique will be tested on wastewaters and improved in this work.

4) Membrane fouling is an evitable problem during filtration which needs to be furtherly controlled by adapted backwash strategy (combination of classic, air and chlorinated backwashes). In addition to optimized filtration conditions, this work will improve further the performance of UF thanks to adequate backwash strategy including chlorinated backwashes.

5) This subject aims to make comprehensive consideration of the impact conditions on UF system in order to achieve the most economical and efficient operation effect and qualified water production and thus brief calculations of total costs at semi-industrial scale will be provided.

CHAPTER 2. Materials and Methods

2.1. Feed water quality

The feed water used both for lab-scale and semi-industrial-scale experiments is the secondary effluent of a municipal wastewater treatment plant (WWTP) located in Châteauneuf-les-Martigues (13), France. The WWTP was started from 2002, it uses a conventional activated sludge process followed by sedimentation tanks to treat raw wastewater. The treatment capacity of the station is 16 000 inhabitant equivalents, of which the treatment capacity is $3\,200\text{ m}^3\cdot\text{d}^{-1}$, and the average flow rate is $133\text{ m}^3\cdot\text{h}^{-1}$. The WWTP is in long-term stable operation, the secondary treated effluent can be discharged without risk into the natural environment. Table 2-1 shows the quality of the inlet and outlet effluent of the WWTP. This outlet effluent was the UF feed of this work. The atmospheric temperature in Châteauneuf-les-Martigues per year probably ranged from 3°C to 32°C . The temperature of UF feed which is secondary effluent from WWTP was between 15°C and 26°C , thereby reducing the influence of water temperature on filtration performances.

2.2. Lab-scale filtration equipment

In order to choose a suitable UF membrane for this project in the pilot-scale, which will be developed in Chapter 3, the filtration performances of two flat sheet membranes and a hollow fibre UF membrane with molecular weight cut-off of 30 kDa, 100 kDa and 200 kDa respectively, are first compared in lab. The detailed information of membranes is listed in Table 2-2. The material of the three membranes is polyethersulfone (PES). The flat sheet membranes are conducted with the 500 mL Amicon stirred cell with concentrate volume of 420 mL as shown in Figure 2-1(a). Hollow fibre membrane is operated with the vertical module with inside membrane volume of 1.9 mL as shown in Figure 2-1 (b). These tests are operated under same TMP of 0.3bar and the same feed water. The initial membrane permeability was measured for the three membranes and as expected, the highest MWCO exhibited the highest initial permeability. The experiments were all operated in dead-end filtration mode whatever the geometry of membrane.

Table 2-1 Raw wastewater and UF feed quality

Parameters	Campaign 1 (October 2019 – March 2020)		Campaign 2 (July 2020 – April 2021)	
	WWTP influent	Outlet effluent (UF feed)	WWTP influent	Outlet effluent (UF feed)
<i>E. coli</i> (CFU 100mL ⁻¹)	1.6×10^8	$(3.4 \pm 2.6) \times 10^4$	$> 1.4 \times 10^6$	$(9.4 \pm 8.5) \times 10^3$
Enterococci (CFU 100mL ⁻¹)	2.2×10^7	$(1.3 \pm 1.0) \times 10^4$	$(3.4 \pm 3.1) \times 10^7$	$(9.5 \pm 8.6) \times 10^2$
Anaerobic sulphito-reducers (spores) (CFU 100mL ⁻¹)	5.6×10^3	268 ± 253	$(1.7 \pm 1.3) \times 10^4$	$(9.6 \pm 5.1) \times 10^2$
Specific F-RNA bacteriophages (PFP 100mL ⁻¹)	4.5×10^3	< 30	$(1.8 \pm 1.7) \times 10^3$	< 30
COD (mgO ₂ ·L ⁻¹)	1124	20 ± 9	552 ± 61	45 ± 21
BOD ₅ (mgO ₂ ·L ⁻¹)	n.m.	n.m.	205 ± 82	< 3
TSS (mg·L ⁻¹)	77	4 ± 2	367 ± 166	12.1 ± 8
TOC (mgC·L ⁻¹)	n. m.	18 ± 9	n.m.	n.m.
Turbidity (NTU)	n. m.	2.3 ± 0.9	208 ± 38	2.1 ± 1.6
NH ₄ ⁺ (mgN·L ⁻¹)	3.8 ± 2.4	-	3.8 ± 2.4	-
pH	n. m.	7.2 ± 0.4	7.4 ± 0.3	7.5 ± 0.1
Conductivity (μS·cm ⁻¹)	n. m.	$1\ 168 \pm 128$	n.m.	n.m.

CFU: colony-forming unit; PFP: Polyhedral, filamentous, and pleomorphic; n. m.: not measured. COD: Chemical oxygen demand; BOD₅: Biochemical oxygen demand TSS: Total suspended solids; TOC: Total organic carbon.

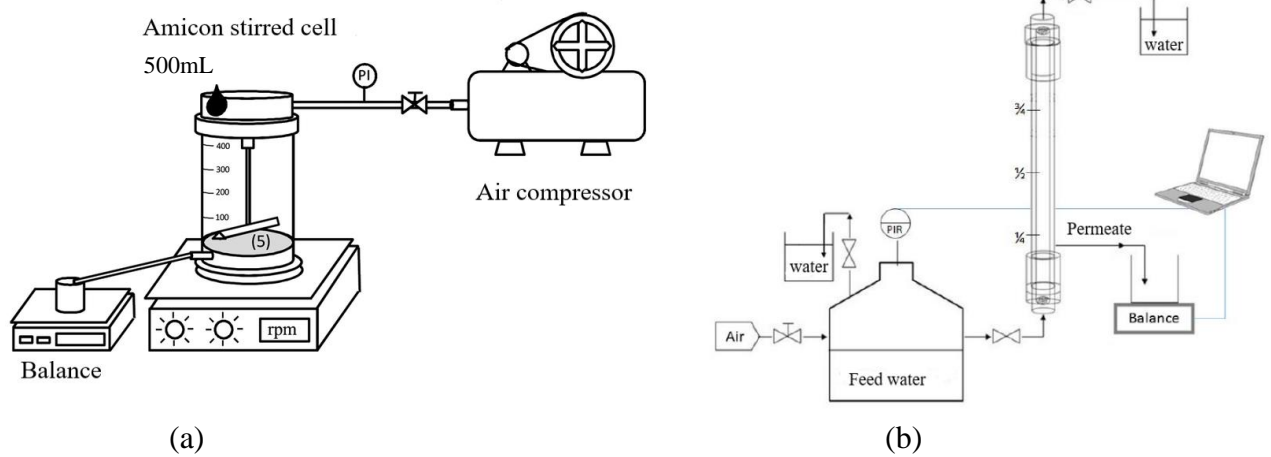


Figure 2-1 Filtration instruments in lab (a) flat sheet membrane, (b) hollow fibers

Table 2-2 Membrane types and operating conditions in Lab scale

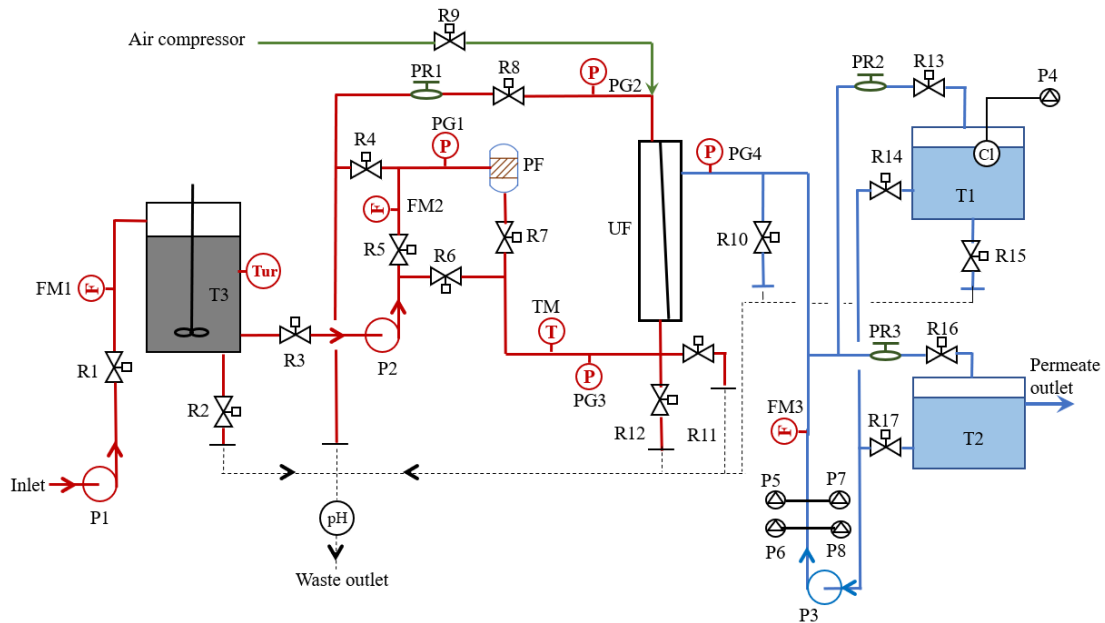
No.	Manufacturer	Material	MWCO (kDa)	Filtration Area (m ²)	Permeability @20°C (L·h ⁻¹ ·m ⁻² ·bar ⁻¹)	Geometry	Concentrate volume (mL)	Operated Pressure (bar)	Agitation rate
1	Millipore		30	0.00418	365	Flat-sheet	420		1-2 rpm
2	Millipore		100	0.00418	773				
3	ALTEON Aquasource®	PES	200	0.00843	958	Multi-channel hollow fiber	1.9	0.3	None

2.3. Semi-industrial pilot plant

Mainly used in this thesis, a semi-industrial UF pilot plant manufactured from Aquasource has been established with automatically operation and recording skills. The nominal capacity of the pilot plant is 20 m³ of filtrated water per day. The flow diagram is shown in Figure 2-2. To be better controlled, the pilot plant can be conducted in manual mode, half automatic mode and automatic mode. The switch of valves in the system are pneumatic controlled by solenoid valves. To control the filtration conditions and detect the filtration performance, pressure sensors, temperature sensors, chlorine sensor, turbidity sensor of feed, pH sensor of effluent, and flowmeters are all connected to the data logger. The chlorine probe is a Dulcotest Type CGE 2-mA-10 ppm as Cl₂

from ProMinent measuring free chlorine and organically bound chlorine (to cyanuric acid in the form of trichloroisocyanuric acid or sodium dichloroisocyanurate-bound chlorine). To be noted, the unit “ppm” in our study represents $\text{mg}\cdot\text{L}^{-1}$.

As shown in Figure 2-2, the secondary effluent of WWTP as feed will be drawn into a feed tank T3 automatically when the feed water drops to the lowest settled level in the pilot plant system. Tank T1 and T2 are the tanks for permeate. Differently, T1 is a single tank with a constant chlorine concentration of $5 \text{ mg Cl}_2\cdot\text{L}^{-1}$ which is used for air backwashes (ABs) and chemical enhanced backwashed (CEBs). T2, without chlorine, is used for classic backwashes (CBs) and is equipped with an overflow going to sewage or storage tank for reuse. A $200 \mu\text{m}$ disk prefilter is designed before UF membrane in order to intercept the biggest particles to avoid UF fiber clogging. Figure 2-3 shows the real view of the pilot plant.



- | | | | |
|------------------------------------|----------------------|-------------------|----------------------|
| R1~R17: Valves | Tur: Turbidity meter | T3: Feed tank | P6: Coagulation pump |
| PG1~PG4: Pressure Gauge | PH: pH meter | P1/P2: Feed pump | P7: Soda pump |
| PR1~PR3: Pressure regulating valve | TM: Thermometer | P3: Backwash pump | P8: Chlorine pump |
| FM1~FM3: Flowmeter | T1: Permeate tank 1 | P4: Chlorine pump | |
| Cl: Chlorometer | T2: Permeate tank 2 | P5: Acid pump | |

Figure 2-2 Diagram of semi-industrial pilot plant



Figure 2-3 Picture of pilot plant

2.4. Membrane information in UF pilot plant

Table 2-3 UF membrane and module characteristics

Characteristics	Data
Material	PES (polyethersulfone)
Pore size - MWCO	0,02 μm – 200 kDa
Length	1.2 m
Internal diameter ID	0.9 mm
Number of channels	7
Filtration Surface	9 m^2
Volume of fibers	2.0 L
Maximum TMP	2.5 bar
pH tolerant value	1-13

The membrane module used for the filtration purpose is an ALTEONTM I (Aquasource) multichannel hollow fiber UF module. Table 2-3 shows the detailed information of the UF module and membrane. The module is operated in dead-end filtration mode with an inside-out configuration. The best range of operational transmembrane pressure (TMP) on UF should be less than 1 bar. The hydraulic resistance of new membrane modules was measured with pure water to be $4 \times 10^{11} \text{ m}^{-1}$ at 20°C (pure water permeability = $900 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$). The maximum permeability of UF membrane when filtrated with the feed water could reach $700 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$ at

20°C and the average value is $600 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ at 20°C. Therefore, each filtration condition was started from initial L_p at around $600 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, and the corresponding membrane resistance of $6 \times 10^{11} \text{ m}^{-1}$ at 20°C was considered for future calculations.

2.5. Membrane cleaning

The review by Chang et al. (2017) [19] critically addressed the critical function and parameters of backwashing on low pressure-driven membranes (mainly MF/UF) in drinking water treatment, and pointed out the optimal range of parameters that affecting the backwash performance. The main difference in our test is using low pressure-driven membrane-UF to treat secondary WWTP effluent for reuse, instead of for drinking water treatment. The parameters of classic backwash in this study were shown in Table 2-4.

Table 2-4 Optimal parameters for classic backwash

Operating conditions	Optimal range values in general [19]	CB parameters in this study
Backwash interval	10-60 min	20-60 min
Backwash duration	10-90 s	52 s
Backwash strength*	1-5	2.5-5
Backwash water	Permeate, deionized water	Permeate

Backwash strength*: The ratio of backwash pressure to permeate pressure or the ratio of backwash flux to permeate flux.

To eliminate fouling in this PhD, three types of membrane cleanings were automatically carried out by the pilot. The detailed procedures of CB and AB are shown in Table 2-5.

(1) Classical backwashes (CB) use permeate as backwash water with a flow rate of $2.5 \text{ m}^3\cdot\text{h}^{-1}$. The backwash water velocity is $0.64 \text{ m}\cdot\text{s}^{-1}$. When using CB without chlorine, the backwash water for membrane heads cleaning is pumped from T2 without chlorine, when using CB with chlorine, the backwash water is pumped from T1 with chlorine. The duration and the volume of water used during a CB is respectively 52 s and 36 L.

(2) Air backwashes (AB) consists of 3 steps: 1. emptying the fibers by opening all the valves of the membrane module; 2. air injection into membrane to dry the fouling layer until reaching a pressure of 0.3 bar. The air is supplied with an air compressor and purified. The air velocity during injection is around $0.6 \text{ m}\cdot\text{s}^{-1}$. 3. backwash at the top, bottom, and both heads with permeate with or without chlorine. This type of backwash was described by Cordier et al. [200,231]. In chapter 3, AB added $5 \text{ mg Cl}_2\cdot\text{L}^{-1}$ of NaOCl in permeate for backwash, the duration and the volume of water used is respectively 67 s and 52 L. In chapter 5, AB adds different concentration of NaOCl ($10 \text{ mg Cl}_2\cdot\text{L}^{-1}$) in backwash water, the duration and the volume of water used is respectively 67 s and 36L. Similarly, when using AB without chlorine, the backwash water for membrane heads cleaning is pumped from T2 without chlorine, when using CB with chlorine, the backwash water is pumped from T1 with chlorine.

(3) Finally, before starting with new operating conditions or if the permeability dropped below $200 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ at real temperature, a chemical enhanced backwash (CEB) was conducted. A CEB starts with a CB with chlorine, followed by chemical injection which includes the top injection and bottom injection. Next, pilot plant closes all the valves and membrane soaking happens with chemicals to degrade foulants. Chemicals are either sulfuric acid ($[\text{H}^+] = 1000 \text{ mg}\cdot\text{L}^{-1}$), or sodium hydroxide ($[\text{OH}^-] = 800 \text{ mg}\cdot\text{L}^{-1}$) and chlorine ($[\text{Cl}_2] = 50 \text{ mg Cl}_2\cdot\text{L}^{-1}$). The duration of soaking is 1200 s. After soaking, the system discharges the soaking solutions by gravity and then starts with a long-term CB without chlorine until pH of discharge water comes back to a neutral value. The duration and the volume of water used during a CEB is respectively 1560 s and 250 L.

In addition of the above-mentioned backwashes to remove membrane fouling, a flush was operated at the middle of each filtration step for about 10 s allowing feed water to circulate in the membrane in bottom-up-outside mode with flow rate between $2.0 \text{ m}^3\cdot\text{h}^{-1}$ and $3.0 \text{ m}^3\cdot\text{h}^{-1}$ (corresponding to) The flush, sometimes called “rinsing”, has been proved to be efficient in preventing particle deposition on membrane through circulating concentrate out with potential fouling materials in dispersed phase [232]. In result, the cooperation of flush and backwash could finally slow down fouling velocity, increase permeability, and reduce energy consumption [233].

Table 2-5 Physical backwash parameters

	AB with CI (RL A1)	AB without CI (RL A2)	CB with CI (RL C1)	CB without CI (RL C2)
Step 1	Air injection into the fibers (2 min)	Air injection into the fibers (2 min)		
Step 2	Decompression before standard backwash	Decompression before standard backwash	Decompression before standard backwash	Decompression before standard backwash
Step 3	Backwash top head (31 s)	Backwash top head (31 s)	Backwash top head (31 s)	Backwash top head (31 s)
Step 4	Backwash 2 heads (3 s)	Backwash 2 heads (3 s)	Backwash 2 heads (3 s)	Backwash 2 heads (3 s)
Step 5	Backwash bottom head (18 s)	Backwash bottom head (18 s)	Backwash bottom head (18 s)	Backwash bottom head (18 s)
Step 6	Backwash prefilter (12 s)	Backwash prefilter (12 s)	Backwash prefilter (12s)	Backwash prefilter (12 s)
Permeate tank (volume)	T1 (29.25 L) + T2 (6.75 L)	T2 (36 L)	T1 (29.25 L) + T2 (6.75 L)	T2 (36 L)
Duration	184 s	184 s	64 s	64 s

2.6. Filtration conditions

In chapter 3, the experiments were all operated in automatic mode, in dead-end filtration mode at constant flux ranging from 20 to 100 L·h⁻¹·m⁻². Dead-end ultrafiltration (UF) has been considered as a more energy efficient operation mode compared to cross-flow filtration in large-scale water treatment systems [234]. For several months of operation, filtration parameters including feed turbidity, feed temperature, flux, transmembrane pressure, calculated permeability at 20°C were monitored continuously to investigate the impact of filtration cycle time, backwash sequence and constant flux on membrane hydraulic performances. The backwash sequence, represented by “BW 1/n” in the following contents, such as 1/3, 1/4, 1/6, and 1/9, represents n times of classic backwashes (CB) followed with 1 air backwash (AB). Different designed conditions mainly focused on filtration cycle time, filtration flux, and backwash sequence were studied in this test, as shown in Table 2-6. The total 15 different filtration conditions were separated into 3 main groups to study the impact of filtration cycle time, flux, and backwash sequence on membrane filtration performance, such as permeate quality, permeability, fouling reversibility by backwash, fouling resistance, economic costs..... Each filtration condition was conducted continuously for more than 40h. After each operating condition, several CEBs were operated manually to remove the membrane fouling until the initial permeability (Lp) reached about 600 L·m⁻²·h⁻¹·bar⁻¹, so as to maintain the similar initial membrane state for the next filtration conditions. In chapter 5, the optimized

condition based on the results of chapter 3 was selected: a flux of $60 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, a filtration time of 60 min and an AB frequency of 1/3. The variable in chapter 5 is the chlorine concentration in backwash water. Four backwash conditions were investigated in this test, shown in Table 2-7. Each condition was conducted continuously for 5-7 days to obtain stable filtration performance. To be noted, the parameters including flux and L_p , that could be affected by the temperature have been normalized to a standard temperature (20°C) to account viscosity fluctuations with these parameters.

Table 2-6 Filtration conditions in chapter 3

	Flux (J) ($\text{L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$)	Filtration time t (min)	Backwash sequence (Air Backwash/ Classic Backwash)
Impact of Filtration cycle time on filtration performance	60	20	1/3
		30	
		40	
	80	60	
		30	
		40	
Impact of Flux on filtration performance	30	30	1/3
	60		
	80		
	100		
Impact of backwash sequence on filtration performance	60	60	1/3
			1/4
			1/6
			1/9
			No air backwash (No AB)

Table 2-7 Backwash conditions in chapter 5 on condition J60t60BW1/3

	CB	CB	CB	AB	Name
1	No Cl	No Cl	No Cl	No Cl	NNNN
2	10 mg Cl ₂ ·L ⁻¹ Cl	10 mg Cl ₂ ·L ⁻¹ Cl	10 mg Cl ₂ ·L ⁻¹ Cl	No Cl	YYYN
3	No Cl	No Cl	No Cl	10 mg Cl ₂ ·L ⁻¹ Cl	NNNY
4	10 mg Cl ₂ ·L ⁻¹ Cl	10 mg Cl ₂ ·L ⁻¹ Cl	10 mg Cl ₂ ·L ⁻¹ Cl	10 mg Cl ₂ ·L ⁻¹ Cl	YYYY

2.7. Hydraulic filtration performance of UF membrane

2.7.1. Permeability

In this manuscript, the units are not those of the international system, but the practical units measured on the pilot and used by industrials.

As the flux J is constant in the semi-industrial system, the relationship between J (L·m⁻²·h⁻¹) and Lp (L·m⁻²·h⁻¹·bar⁻¹) can be expressed as Eq. (1).

$$Lp = \frac{J}{TMP} \quad (1)$$

The pressure difference over a membrane is called the Transmembrane Pressure (TMP), unit *bar*, which is calculated by Eq. (2)

$$TMP = \frac{P_{top\ head} + P_{bottom\ head}}{2} - P_{permeate} \quad (2)$$

Permeability recovery:

The cleaning efficiency (%) of CEB can be reflected by recovery of permeability and defined as Eq. (3):

$$Permeability\ recovery = \frac{Lp_{After}}{Lp_{Before}} \times 100\% \quad (3)$$

where Lp_{Before} and Lp_{After} are the membrane permeabilities recovered before and after the CEB cleaning.

2.7.2. Fouling resistances

According to Darcy's law [235], hydraulic resistance of the fouled membrane was measured with Eq. (4):

$$R_t = R_{irr} + R_{re} + R_m = \frac{TMP}{\mu \cdot J} = \frac{1}{\mu \cdot Lp} \quad (4)$$

where, TMP is the transmembrane pressure, μ is the viscosity of permeate and J is the applied flux. Actually, the total membrane resistance (R_t) which is finally calculated by viscosity (μ) and permeability (Lp) includes three parts of resistances: irreversible fouling resistance (R_{irr}) which cannot be removed by CB, reversible fouling resistance (R_{re}) which can be removed by CB and membrane resistance (R_m). The unit of resistances is m^{-1} .

The flux at 20°C is calculated through Eq. (5) [236]:

$$J(20^\circ\text{C}) = \frac{\mu(T)}{\mu(20)} \cdot J(T) = J(T) \times e^{[0.0239 \times (20-T)]} \quad (5)$$

In two adjacent filtration cycles, it can be considered that the composition of fouling resistance in end of cycle (n-1) is

$$R_{\text{end}(n-1)} = R_m + R_{\text{irr}(n-1)} + R_{\text{re}(n-1)} \quad (6)$$

While after a CB, the total resistance in the beginning of cycle (n) is

$$R_{\text{ini}} = R_m + R_{\text{irr}(n-1)} \quad (7)$$

Through difference in Eq. (6) (7) and knowing R_m , values of R_{irr} and R_{re} in the n-1 filtration cycle can be found.

2.7.3. Fouling Reversibility

Filtration performance was evaluated by parameters such as backwash effectiveness (fouling reversibility), fouling rate, and fouling resistance [237]. Backwash effectiveness can be indicated by fouling reversibility which was calculated after each filtration cycle n according to Chang et al. (2016) [19,238]. Reversibility after each filtration cycle could then be calculated using the initial TMP and final TMP values (TMP_{end}^n and TMP_{ini}^n) of the cycle n as well as the initial TMP of the next filtration cycle ($TMP_{\text{ini}}^{(n+1)}$). As described above, the flux and viscosity at 20°C are both constant during each filtration condition. Therefore, the reversibility can be calculated as follow Eq. (8).

$$\text{Reversibility}(n) = \frac{TMP_{\text{end}}^n - TMP_{\text{ini}}^{(n+1)}}{TMP_{\text{end}}^n - TMP_{\text{ini}}^n} = \frac{R_{\text{end}}^n - R_{\text{ini}}^{(n+1)}}{R_{\text{end}}^n - R_{\text{ini}}^n} \quad (8)$$

During one operating condition, there were multiple CBs and ABs. For each condition, a mean value of CB reversibility and AB reversibility, with the related standard deviation were calculated. In chapter 5, statistical analysis was performed to determine if there was a statistically significant linear regression between backwash

reversibility and R_{irr} before backwash. The data were analysed by XLSTAT (Addinsoft), and an ANOVA test was performed.

2.7.4. Water recovery rate

The recovery rate represents the proportion of net produced water quantity when compared to the total inlet feed water quantity. This was calculated with Eq. (9).

$$Recovery\ rate = \frac{V_{net}}{V_{inlet}} \times 100 = \frac{V_{inlet} - V_{CB} - V_{AB} - V_{CEB} - V_{Con}}{V_{inlet}} \times 100\% \quad (9)$$

where V_{inlet} refers to the volume of inlet feed water, V_{net} refers to the net volume produced. The calculation of V_{inlet} is related to the filtration flux and filtration time, as in Eq (13). The volume unit is the liter.

$$V_{inlet} = J \times t_f \quad (10)$$

V_{Con} refers to the concentrate volume, which is known to be 2 L (volume of fibers presented in Table 2-3) and V_{CB} , V_{AB} , and V_{CEB} represent the volume of permeate consumed for CB, AB and CEB which are 36 L, 52 L and 250 L, respectively.

2.7.5. Impact of air backwash on irreversible resistance increase

To find out the influence of air backwash frequency on UF membrane filtration performance, the variation of irreversible fouling during operation can be an interesting parameter to follow. The relationship of R_{irr}/R_m variation with feed water turbidity integration (Eq. (11)) will be considered to investigate the influence of backwash sequence on fouling formation as the function of the quality of feed water changes:

$$\frac{(R_{irr}/R_m)_n}{(R_{irr}/R_m)_{initial}} = f(\sum T_{ur}.dt) \quad (11)$$

where the turbidity integration with time is shown as follows with n=number of filtration cycle, and t_f the duration of a filtration cycle.

$$\sum_{i=1}^n \left[\int_0^{t_f} Turbidity. dt \right]_i \quad (12)$$

Obviously, the membrane fouling is not limited to the quantity of suspended matter given by the measurement of turbidity but an order of magnitude was obtained. In fact, it was shown during this study that the variation in turbidity had a direct impact on the

variation of the permeate flux. In this case, parameters such as the presence of organic matters are not considered.

2.7.6. Fouling removal efficiency of backwashes

In chapter 5, the removal efficiency of physical backwashes on turbidity and TOC (removal rates) are illustrated to furtherly verify the effectiveness of physical backwashes. For organic matters, the accumulation of dissolved organic matters was already integrated with filtration time by Lin et al. [239]. Therefore, the turbidity and TOC are considered as accumulated amounts in membrane during the filtration cycles, the removal rates of these parameters can be calculated with the following equations based on the amount of substance balance.

(1) Turbidity:

$$R_{\text{Tur}} = \frac{\int (\text{Tur}_{\text{Feed}} - \text{Tur}_{\text{permeate}}) dV_0}{\int (\text{Tur}_{\text{BW}} - \text{Tur}_{\text{permeate}}) dV_{\text{BW}}} \times 100\% = \frac{(\text{Tur}_{\text{Feed}} - \text{Tur}_{\text{permeate}}) \times J \times S \times t}{(\text{Tur}_{\text{BW}} - \text{Tur}_{\text{permeate}}) \times \Delta V_{\text{BW}}} \times 100\% \quad (13)$$

(2) TOC:

$$R_{\text{TOC}} = \frac{\int (\text{TOC}_{\text{Feed}} - \text{TOC}_{\text{permeate}}) dV_0}{\int (\text{TOC}_{\text{BW}} - \text{TOC}_{\text{permeate}}) dV_{\text{BW}}} \times 100\% = \frac{(\text{TOC}_{\text{Feed}} - \text{TOC}_{\text{permeate}}) \times J \times S \times t}{(\text{TOC}_{\text{BW}} - \text{TOC}_{\text{permeate}}) \times \Delta V_{\text{BW}}} \times 100\% \quad (14)$$

here, V_0 is the filtration volume (L) through the UF membrane during one filtration cycle.

V_{BW} is the physical backwash volume during one physical backwash, $V_{\text{BW}} = 36 \text{ L}$.

J is the filtration flux, $J = 60 \text{ L} \cdot \text{h}^{-1} \cdot \text{m}^{-2}$.

S is the surface area of UF membrane, $S = 9 \text{ m}^2$.

t is the filtration time during one filtration cycle, $t = 1 \text{ h}$.

2.8. Water quality assessment

2.8.1. Microplastics

2.8.1.1 Principle of the analysis and main detection parameters

The detection of microplastics in this study was achieved by the PerkinElmer FT-IR spectrometer with spectrum Spotlight 400 FT-IR microscope (USA). During this thesis and in partnership with PerkinElmer, the development of this analytical technique was

carried out. Therefore, this study was realized from tests on ultrapure water to the identification and quantification of microplastics in seawater treatment plants, pharmaceutical wastewater treatment plants, together with the samples from the WWTP in Chateauneuf-les-Martigues. The analytical process includes sample preparation, digestion of organic matters, filtration, identification and quantification of microplastics, and finally to calculate the removal rate of microplastic particles under different treatment process. The full information will be presented in another scientific paper and not in this thesis because of the relevance and continuity of the subject content. Only the detection of microplastics in the feed and permeate of the UF plant will be presented. The detection system is mainly composed of four parts: the FT-IR spectrometer, the spectrum Spotlight 400 FT-IR microscope, the stage control for sample view movement in automatic and manual, and the computer for data treatment, shown in Figure 2-4. The core component of FT-IR spectrometer is the Michelson interferometer that can generate an interferogram by making measurements of the signal at many discrete positions of the moving mirror. The light beam uses the middle IR in this FT-IR spectroscopy with wave number ranges from 400 to 4000 cm^{-1} that mainly focused on molecular vibrational energy levels (~ 80 to 8000 cm^{-1}). The related software here is Spectrum IMAGE. The computer conducts Fourier transform calculations on the interferogram and displays the spectrum as absorbance (A) or transmittance (T) versus wavenumber. In microscope, the detector is a liquid nitrogen cooled MCTs (LN-MCTs) with high sensitivity on smaller particles ($<10 \mu\text{m}$). Every day before starting to analyze samples, the MCT detector was refilled with liquid nitrogen to cooldown during prolonged use.

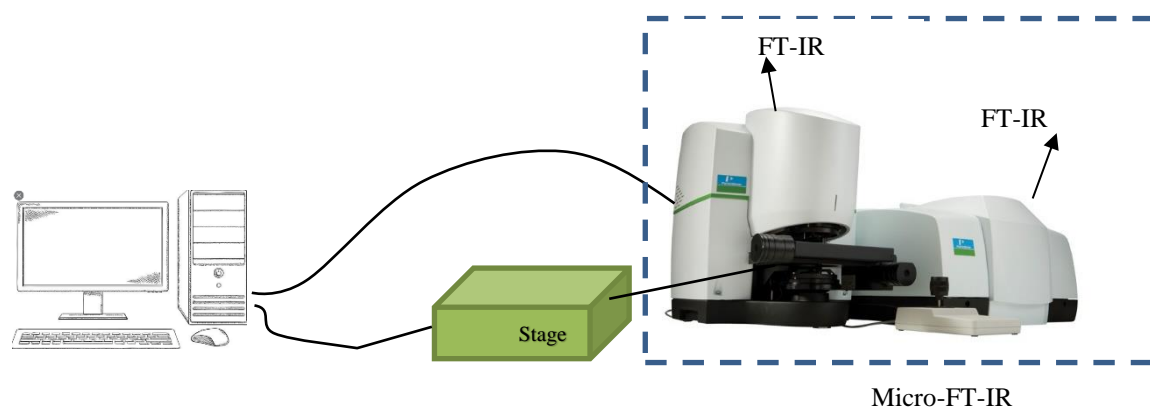


Figure 2-4 The microscopic-equipped FT-IR spectroscopy system

In this work, FT-IR spectroscopic imaging in reflectance mode using a focal plane array (e.g., 64×64 pixels) has been applied to the identification properties. This detection mode enables the rapid analysis of thick and opaque samples and are therefore highly suitable for detecting microplastics in environmental samples [240,241]. Spectral resolution is a measure of how well a spectrometer can distinguish closely spaced spectral features. According the investigation on infrared spectra differences of polypropylene under different resolutions (2 cm^{-1} , 4 cm^{-1} , 6 cm^{-1} , 8 cm^{-1} , 12 cm^{-1} , 32 cm^{-1} and 64 cm^{-1}) by Zheng et al. (2021), the 16 cm^{-1} spectral resolution was optimized and thus is selected in this study as the analytical devices used in both studies were the same. A background spectrum was collected for each sample from a blank area of the gold filter at a spectral resolution of 16 cm^{-1} with pixel size both at $6.25 \mu\text{m}$ or $25 \mu\text{m}$. The other identification parameters of micro-FT-IR in this study are as follows: 2 scans per pixel, an interferometer speed of $2.2 \text{ cm}\cdot\text{s}^{-1}$, IR spectral range of $4000 \text{ cm}^{-1} - 690 \text{ cm}^{-1}$ and the resolution of pixel size selected at $6.25 \mu\text{m}$ or $25 \mu\text{m}$ depending on the filters used ($25 \mu\text{m}$ stainless steel filter with $25 \mu\text{m}$, $3 \mu\text{m}$ and $5 \mu\text{m}$ gold coated filters with $6.25 \mu\text{m}$). The scanning area ranged from $6000 \mu\text{m} \times 6000 \mu\text{m}$ to $10000 \mu\text{m} \times 10000 \mu\text{m}$ which were selected from the edges of the sample deposited on each filter. For each sample, the FT-IR system will generate an absorbance image with an infrared spectrum information on each pixel.

To identify microplastics' structure easier and faster, a freeware, siMPle, developed by Aalborg University, Denmark and Alfred Wegener Institute, Germany, is adopted in this study. The reference database contains both plastic polymers and natural materials such as protein and cellulose, totally in 23 types of materials given in Table 2-8. The siMPle software is created based on the combination of MPHunter [243] and AWI Automatic Pipeline [244] for MP analysis that compares the IR-spectrum of each map pixel with the spectra of a reference database and assigns a score to the quality of each match [244,245]. The parameters than can be detected from siMPle include the polymer types, range of abscissa and ordinate, numbers of pixel, minor/major dimension, surface area, and estimated volume and mass of each particle. The major dimension is analysed for the longest distance between pixels of the particle. The minor dimension is found by assuming the particle shape is an ellipse and knowing the area of the particle in the scan.

Table 2-8 The types of materials recorded in siMPle and their abbreviations

No.	Polymer and natural material	Abbreviation in siMPle
1	Polyethylene	pe
2	Polypropylene	pp
3	Polystyrene	ps
4	Polyamide (Nylon)	pa
5	Polyvinyl Chloride	pvc
6	Acrylonitrile Butadiene Styrene	abs
7	Polyethylene Terephthalate	pet
8	Polymethylmethacrylate (Acrylic)	pmma
9	Protein	protein
10	Polyetheretherketone	peek
11	Poly Lactic Acid	pla
12	Ethylene Vinyl Acetate	eva
13	Polyoxymethylene (Acetal)	pom
14	Stearate	stearate
15	Stearate + Glyceride	stearate + glyceride
16	Polyisoprene	polyisoprene
17	Polytetrafluoroethylene	ptfe
18	Cellulose	cellulose
19	Polyether Block Amide or Polyester Block Amide	peba
20	Silicone	silicone
21	CaCO ₃	caco3
22	Glyceride	glyceride
23	Wax	wax

2.8.1.2 Sample preparation

Sample pretreatment is necessary to be considered before analysis. Pretreatment consists of digestion of organic matters which could hide MPs on the filters. As the composition of the samples can directly affect the digestion methods and duration, it is necessary to find the suitable digestion protocol of each type of water. The digestion methods used in this study was H₂O₂. First, samples can be digested with 30% (w/v) H₂O₂ at a volume ratio of 1:1 for 1-10 days at room temperature. During stirring, the glass bottle was covered with aluminum foil to prevent airborne contamination. The rinsing water used in this study is ultrapure water. This water was selected after a comparison of different clean waters (HPLC water, distilled water, Evian water, tap water, etc.).

After the preparation, the water samples are ready to be filtered. There are 2 types of filters used, the 3 µm gold-coated polyester membranes (i3 TrackPor P, Germany),

and 25 μm stainless steel filter. In reflectance mode, the filter type needs to reflect IR radiation without any significant absorptions. Normally, the gold-coated filters show high energy in reflectance, and the stainless steel slide can reflect all infrared light without contributing to the infrared spectrum [246]. The samples are filtrated through a vacuum apparatus, shown in Figure 2-5. The apparatus is composed of filtering cup, filtration mask, spring clamp, filter, glass stopper, and conical flask. The filtration masks are round metal molds with hollow square in the center to regulate the area of the filters which will be analysed by FT-IR. The dimensions of the square on filtration masks are 6000 μm \times 6000 μm and 10000 μm \times 10000 μm , respectively. In this experiment, the mask with 6000 μm \times 6000 μm square is mainly used on 3 μm gold coated filters, the mask with 10000 μm \times 10000 μm square is used on 25 μm stainless steel filter. For the samples with visible particles and solids, it is better to firstly filtrate them through the 25 μm stainless steel filter, and secondly filtrate with a 3 μm gold filter and then perform the analysis of both filters to make results more reliable. For clean water samples as UF permeates, it can be directly filtrated through the 3 μm gold filter. Before and after testing on FT-IR spectroscopy, the filter with collected sample will be put in an evaporating dish to avoid external pollution.

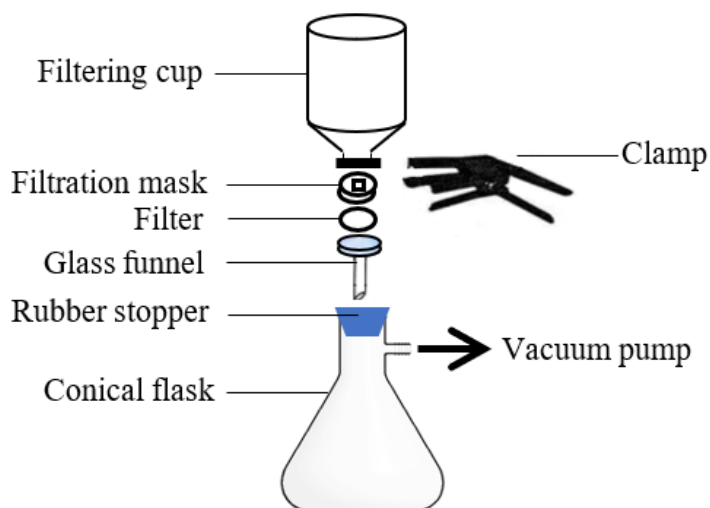


Figure 2-5 Filtration apparatus - vacuum büchner funnel

2.8.2. Analysis of other parameters

Total suspended solids (TSS) were measured with a 47 mm glass-fiber-filter (Whatman). The filter was firstly rinsed with distilled water and dried during minimum

2h at 105°C after filtration of the sample. The TSS concentrations were then obtained by calculating the difference of weight before and after filtration divided by the volume of sample.

The turbidity of feed was measured and recorded every minute of the pilot plant using a probe TurbiMax W CUS31(Endress Hauser). Besides, the turbidity of permeate was also tested punctually in laboratory with a turbidity meter (Turb 550 IR, WTW, Germany).

The electrical conductivity value was determined with a conductometer (Sension+EC7, Hach, USA). The pH was measured with a pH-meter (Sension+pH31, Hach, USA).

During Campaign 1, COD analysis was conducted through reagent vials (COD cell test C3/25), with test range from 10 to 150 mg·L⁻¹. A volume of 3 mL of a homogeneous water sample was injected into a COD digestion reagent vial. Then, the vial contents were stirred and subsequently placed in the COD reactor at 148°C for 2h. Finally, the samples after cooling were tested by a spectrophotometer— (190 – 1100 nm, Photolab 6600 UV-Vis, WTW).

A TOC-L machine (*Shimadzu, Japan*) based on the 680° C combustion catalytic oxidation method was used to measure the concentration of total organic carbon (TOC). The non-purgeable organic carbon (NPOC) method with a detection limit of 4 µg L⁻¹ was used.

During Campaign 2, COD and BOD₅ were tested by an external laboratory (*Carso – Laboratoire Santé Environnement Hygiène de Lyon*) certified by the French Ministry of Health for water analysis. The methods for COD and BOD₅ are respectively ISO 15705 and NF EN ISO-5815-1.

The microbiological analyses were also tested by *Carso* laboratory and consisted in the measurements of *E. Coli* (*NF EN ISO 9308-1, NF EN ISO 9308-3*), *Enterococci* (*NF EN ISO 7899-1, NF EN ISO 7899-2*), anaerobic sulphito-reducers (spores) (NF EN 26461-2), specific F-RNA bacteriophages (NF EN ISO 10705-1), and SARS-CoV-2 by real-time quantitative polymerase chain reaction (qPCR). For SARS-CoV-2 analysis, the envelope protein gene E is first tested and the nucleoprotein gene N is tested only if the envelope protein gene E is detected. The detection methods NF EN ISO 9308-1

for *E. coli*, and NF EN ISO 7899-2 for *Enterococci* are used in samples during Campaign 1 with both detection limits at $< 1 \text{ CFU} \cdot 100\text{mL}^{-1}$. The detection methods NF EN ISO 9308-3 for *E. coli*, and NF EN ISO 7899-1 for *Enterococci* are used in samples of Campaign 2 with both detection limits at $< 56 \text{ CFU} \cdot 100\text{mL}^{-1}$. Inlet and outlet (UF feed) of the WWTP and permeate were analysed at the same time.

CHAPTER 3. Optimization of UF operating conditions for wastewater reuse

3.1. Introduction

As reviewed in Chapter 1, the membrane-based technologies used for municipal wastewater treatment for water reuse usually include microfiltration/ultrafiltration (MF/UF)-based technologies, membrane biological reactor (MBR) and nanofiltration/forward osmosis/ reverse osmosis (NF/FO/RO)-based technologies [7]. Currently, it is logically predicted that non potable reuse is more successful to build the degree of familiarity and trust required by the public for acceptance [38]. In non-potable water treatment, ultrafiltration (UF) has been widely accepted as one of the most cost-effective technology to deal with secondary effluent on non-potable reuses, such as agricultural irrigation, landscape irrigation, urban reuse, car washing, and sometimes as process water for industries [6,98]. UF enables high removal efficiency on total suspended solids (TSS), turbidity and to a smaller extend on total organic carbon (TOC). More importantly, UF can retain microorganisms significantly, including bacteria, protozoa, and viruses [6]. The investigation of Muthukumaran et al. [102], Pollice et al. [103] and Falsanisi et al. [98] all confirmed that UF (with prefilter) can be successfully applied on secondary effluent treatment for reuse in agriculture with respect to WHO guidelines in terms of permeate quality. To be reliable, the three publications were all conducted in pilot-scale operation ranging from 2 months to 2 years, and with UF pore sizes ranging from 0.002 μm (1kD) to 0.03 μm . Additionally, both Pollice et al. [103] and Falsanisi et al. [98] used real secondary effluent as feed water and Muthukumaran et al. [102] used the synthetic secondary effluent.

Nowadays, membranes are committed to combine material properties and self-forming mechanism for better filtration processes. The selection of material is affected by pore size distribution, wetting susceptibility, porosity, mechanical strength, cost, polymer flexibility, fouling resistance, stability, durability, and chemical resistance [79]. Currently, polymer membranes are widely used among membranes with advantage of superior mechanical strength and low cost. Additionally, PES is one of the most commonly used polymer material for UF and also as supporting substrates for NF and RO processes approved with excellent permeability, mechanical stability, and chemical resistance [247]. To find out a suitable UF membrane, the beginning of this chapter aims to investigate the secondary effluent filtration performances (including flux, fouling formation, and permeate quality) of UF at different molecular weight cut-off (MWCO) in the lab-scale.

In secondary wastewater treatment in full scale, a main drawback limiting the economical operation of UF process on wastewater reuse treatment is the fouling resistance, especially the irreversible fouling which is mainly deposited by the organic matters in the secondary municipal wastewater[14]. In general, two types of fouling phenomena are distinguished for UF membranes in secondary effluent treatment. The first type is the macro-solute or particle deposition on UF membranes mainly caused by suspended solids in feed is mostly external fouling or cake formation on the top surface of the membrane, which is usually reversible and non-adhesive [21]. The second type is the organic matters caused pore blocking and internal pore adsorption on UF membranes, which is usually irreversible and adhesive [248].

In the field of fouling management, the feed water quality, and filtration conditions (filtration flux, filtration cycle time (backwash interval), and cleaning mode play important roles due to their direct effects either on the formation and nature of the fouling layer, or on the removal of the foulants. Normally, less concentration of dissolved organic matters and suspended solids is in feed water, the more beneficial it is to control fouling formation. Therefore, we use secondary treated water as the feed water and a 130 μm prefilter is set in front of UF. During filtration, the filtration flux and filtration time will be incorporated to affect the thickness and density of the foulants deposited on the membranes [21]. To maintain a sustainable and effective treatment performance, frequent cleaning of the membranes is required including physical and chemical washing. Chemical washing is generally applied to remove hydraulically irreversible foulants. The choice of chemical agents on membranes, mostly using acids, alkalis, or oxidants, should be based on the membrane materials and fouling types [15]. Physical cleaning like classic backwash (CB) and air assisted backwash (AB) are commonly applied in most UF process designs as standard operating strategies to limit fouling [19]. Besides, forward flushing with a high flow rate of water can be incorporated with physical backwashing to decrease aggregation or dense attachment of particulates on the membrane surface [249]. CB is commonly used to loosen and detach fouling cake partially from membrane surface. In addition, air assisted backwash is accepted as a more effective anti-fouling physical cleaning compared to CB. While the CB is expected to detach the cake layer from the hollow fiber, air injection contributes to break up and detach foulants away from the membrane surface [233,250]. Normally, AB can be conducted either by air scouring in backwash water, or by air injection into membranes fibres. In air scouring process, air is injected into the membrane with the feed stream to generate a gas-liquid two-phase flow, leading to higher

shear stress near the membrane surface[251]. In air injection process, air can be injected into the concentrate side (inside the lumen of the fiber) without feed stream [252]. Assisted by air scouring, a previous study by Ye et al. [20] demonstrated the air assisted backwash system in moderate air flow rate (e.g. $154 \text{ mL} \cdot \text{min}^{-1}$) could remove most of the foulants more effectively while the loosen and residual foulants after classic backwash would be recompressed when filtration restarts. Air scouring is more likely to be used in MBR [251,253]. Salerno et al. [253] compared air scouring with flow rate of 150, 250, and $500 \text{ mL} \cdot \text{min}^{-1}$ MBR process and concluded that the $150 \text{ mL} \cdot \text{min}^{-1}$ considered as an optimal compromise between high effluent quality and limited cleaning frequency. Assisted by air injection, P.J. Remize et al. [254] also stated that backwash procedure was hydraulically enhanced by air which was injected into the concentrate side, the remaining particulate fouling was significantly lowered and leads to an increase in process runtime. Additionally, C. Cordier *et al.* [231] confirmed the higher removal ability of AB with air injection, and it could destroy the cells' integrity by air injection, such as oysters' oocytes and spermatozoa in seawater. However, frequent air assisted backwash can bring some disadvantages. 1. Frequent and/or high flow rate of air scouring backwash would not remove macromolecules effectively, and in contrast might cause serious pore blocking inside membranes [20]. 2. Because of the harsh requirement of air backwash for membranes (e.g., low bubble point and easily rewetting) [250], air assisted backwash may lead to partial drying for certain membranes, which can cause embrittlement and membrane integrity problems [19]. 3. Air assisted backwash requires more energy and equipment cost due to the requirements of air blowers, and more complex operation and maintenance of air backwash. Considering both the pros and cons of classic backwash and air assisted backwash, a strategy that maximizes benefits may incorporate periodic classic backwash, and air assisted backwash together in a filtration mode (named as periodical backwash sequence in this test) to provide better fouling removal effects. Through retrieval, there are very few publications about the performance of alternative backwash and air assisted backwash during one filtration condition, nor the optimization of filtration conditions. Although C. Cordier et al. [202] has reported the several backwash sequences (1/3, 1/5, 1/7, and 1/9) with this article focused on UF membrane filtration, the test was fed with aquaculture seawater, which is different from the composition of municipal secondary effluent wastewater shown in this work.

Therefore, it is very meaningful to set-up on-site a semi-industrial UF pilot plant with periodic classic backwashes (CB) and air backwashes (AB) on real secondary effluent for water reuse

application. The target of this study is to find out the optimized filtration conditions that can be long-term applied on secondary wastewater treatment for water reuse with guarantee of adequate permeate quality, sustainable hydraulic filtration performance, and high net permeate production. 15 different filtration conditions are designed to identify the effects of flux, filtration cycle time, and backwash sequences on hydraulic filtration performance. The filtration performances will be described through the analysis of fouling reversibility and composition, water recovery rate, and the impact of AB frequency on irreversible fouling control. The analysis of permeate quality will be discussed in chapter 4.

3.2. Comparison of membranes' performances in lab-scale

There are three tested ultrafiltration membranes (shown in Table 2-2) to be compared in lab, with MWCO of 30 kDa, 100k Da and 200 kDa, respectively. The tested WWTP (Châteauneuf-les-Martigues, France) effluent contained $8.9 \text{ mgC}\cdot\text{L}^{-1}$ of total organic carbon (TOC) and with mean turbidity of 3.5 NTU. The initial permeabilities at 20°C of 200, 100 and 30 kDa membranes are $960, 770$ and $365 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}\cdot\text{bar}^{-1}$, respectively. During filtration experiments, the transmembrane pressure (TMP) was set to 0.3 bar, and permeate samples were collected for analysis.

3.2.1. Flux decline

Due to concentrate volume difference on different membrane modules (i.e. geometry), Figure 3-1 uses the “permeate volume/filtration area” to replace the commonly used volumetric concentration factor (VCF) as the X axis. Y axis reflects flux variation with the ratio of the instantaneous flux to the initial flux, where J_p is the instantaneous permeate flux during filtration, J_0 is the initial permeate flux. Overall, the three curves show similar decreasing trend on flux changes during filtration by different membranes when filtrating the same feed water. During filtration, permeability of UF membranes decreased from rapidly to gradually versus permeate volume because of the inevitable membrane fouling [255].

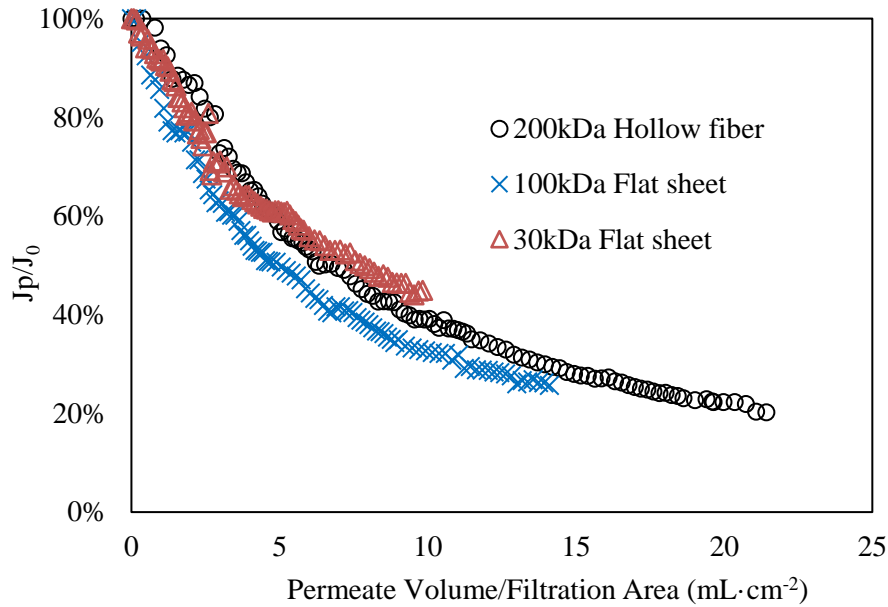


Figure 3-1 Permeability variation of UF membranes at different MWCOs

3.2.2. Permeate quality

To compare the permeate quality from different membranes, parameters including turbidity and TOC rejection rates were tested. As a result, all membranes can decrease the turbidity from feed water to less than 0.8 NTU with more than 90% removal rates. With the extension of filtration, all the membrane showed increased turbidity retention rate which indicated that the formed cake foulant on membranes helped to hold up more contaminants in feed water. The 100 kDa hollow fiber membrane showed the highest and most stable turbidity retention rate compared to 30 kDa and 100 kDa flat sheet membranes, which resulted in > 99.9% removal of turbidity. For TOC rejection, 200 kDa hollow fiber membranes showed the highest removal efficiency with more than 95% removal rates, while the 30 kDa and 100 kDa flat sheet membranes could just remove about 40% and 50% of TOC from the feed. Here to be noted, the removal rates were calculated by the TOC concentration in permeate with the TOC concentration in the volume near the membrane. As the hollow fiber module has smaller concentrate volume (1.9 mL) than the Amicon stirred cell (420 mL), the TOC concentration in concentrate of hollow fiber module will be much higher than that in concentrate of flat sheet membrane module, this is why there is a big difference on TOC

removal between the hollow fiber and flat sheet membranes. Actually, the concentration of TOC in all permeates had less difference and were all around 4~7 mg·L⁻¹.

3.2.3. Fouling resistance

To further compare, the total resistance on membrane increase on UF membranes during filtration was shown in Figure 3-2. According to Darcy's law, the intercept on Y axis represents the initial membrane resistance of each membrane, the slope of each fitted curve represents the increasing rate of total fouling resistance. Through comparison, 200kDa membrane showed the lowest fouling increasing rate during filtration, which reflects the better anti-fouling character than the others. In order to evaluate the fouling types, a backwash experiment was carried out with the hollow fiber module with 1/3 volume of total permeate under 2 bar. The results showed that the total fouling resistance was composed of 97% of reversible fouling and 3% of irreversible fouling in the lab-scale which is quite promising for full-scale experiments.

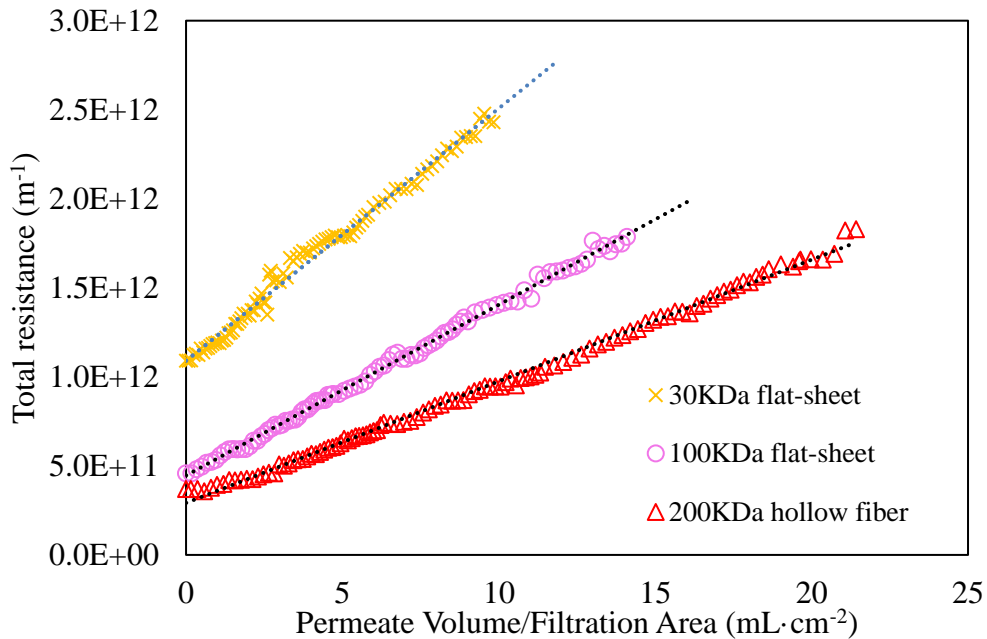


Figure 3-2 Fouling resistance formation on different membranes.

In summary, the MWCO does not seem to impact the flux decline shapes with this secondary effluent and does not seem to impact the turbidity rejection rates. The 200 kDa multi-channel hollow fiber membrane showed the highest TOC rejection rates, the lowest fouling increasing rate

and of course a large membrane specific area due to the geometry, thus it is considered to be the most adequate membrane for the semi-industrial pilot plant experiments.

3.3. Understanding of UF feed variability and potential impact on UF performance in pilot-scale

Before studying the performance of the UF pilot plant, it is very important to explain the WWTP running operation (because the UF feed water is function of it). The flowrate of raw wastewater in the WWTP strongly changes continuously between day and night and the temperature too. Classically, the peak periods of residential water use all over a day generally occur at 7:00 to 10:00, 12:00 to 14:00 and 18:00 to 20:00, while the trough period occurs between 0:00 to 4:00 where the flowrate is divided by a factor 20. In general, periods with higher flowrate mean higher load rate for the biological wastewater treatment processes, such as activated sludge process in the tested WWTP. According to Aygun et al. (2007) [256], when increasing the organic loading rate in raw wastewater (increasing COD), the performance of treatment process became powerless thus the COD removal efficiency decreased. In addition, higher flowrates lead to reduced sedimentation times in the clarifier which can decrease the particles' removal thus increasing turbidity. This can be observed in Figure 3-3 presenting the variations of inlet secondary wastewater flowrate and outlet turbidity. Moreover, the weather, such as a rainfall may significantly increase the flowrate and change the compositions of wastewater, and thus also further affect wastewater treatment performance and effluent quality [257] (i.e. feed of UF).

In order to show the influence of this feed water quality on UF performance, the variations of permeability, feed water turbidity and temperature obtained for regular days in condition J60t20BW1/3 (permeate flux of $60 \text{ L h}^{-1} \text{ m}^{-2}$, duration of filtration step 20 min and 1 AB followed by 3 CB) are shown in Figure 3-4 and Figure 3-5. This condition is taken as a representative example of all conditions. As demonstrated by M. Raffin et al. [258] and Huang et al. [259], the feed water quality was one of the main factors on membrane fouling formation, and turbidity, as an indicator of water quality, directly reflects the potential of fouling. In Figure 3-4, the membrane permeability has converse variation trends with turbidity variation. The partial increase of permeability maybe caused by the periodical variation of feed water flowrate and quality. From

above, trough periods with lower turbidity of feed produced less fouling load on membrane thus the permeability decreased slower than during peak periods. Periodical backwash can remove the previous cake foulant thus improving the permeability recovery after backwash. This phenomenon may explain some significant increase of permeability during the filtration process that occurred usually at the trough periods of a day (red circles in Figure 3-4).

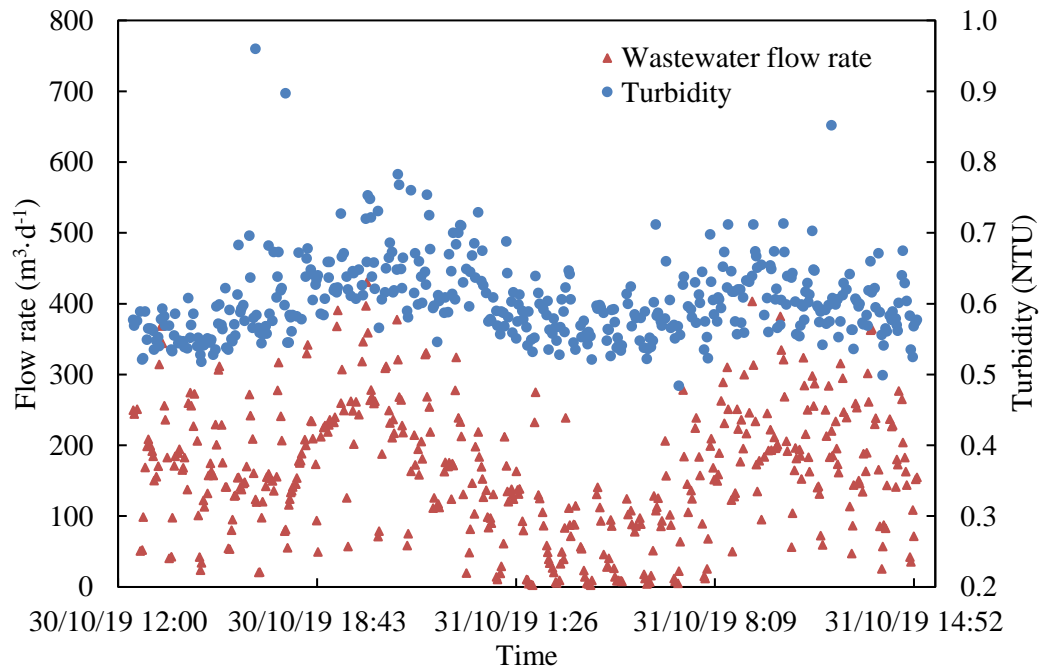


Figure 3-3 Example of raw wastewater flowrate and feed water turbidity variation throughout the day and night

Additionally, the process temperature can also affect the permeability. In this study, the filtration was conducted in constant flux mode whatever the temperature changes as shown in Figure 3-5. The variation of permeability was shown in highly-positive correlated trend with the feed water temperature variation from Figure 3-5. The liquid viscosity is decreased with temperature increase [260], therefore, the permeability at real temperature will be in positive relationship with temperature variation based on Darcy's law. Moreover, as early as in 1880s, Tarnawski and Jelen [261] found a non-linear permeability increase for a polymeric UF membrane in the temperature region from 20°C to 45°C even after compensating for the viscosity decrease, and they attributed this increase partly to a thermal expansion of the membrane material. Generally, the variation of UF permeability is the combined effects by many factors, such as flow velocity,

concentration of substances to be removed, and the feed water temperature. Except specific weather influence, each experiment suffered the almost same change of feed water quality, flow rate, and temperature variation throughout experiments. Therefore, it was considered that the different experiments at different operating conditions presented in the next sections could be compared.

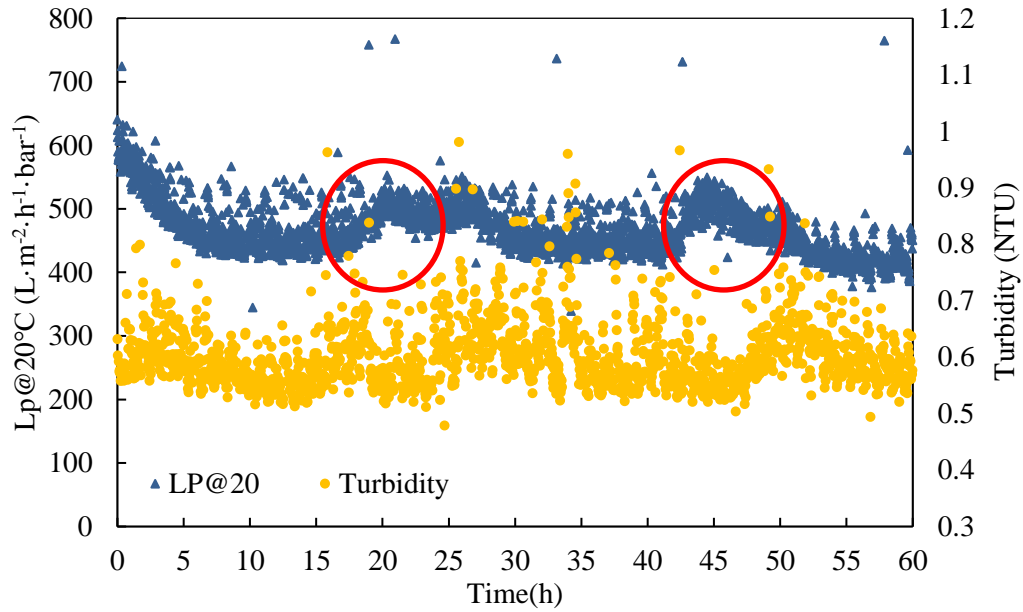


Figure 3-4 Permeability and feed water turbidity variations in condition J60t20BW1/3

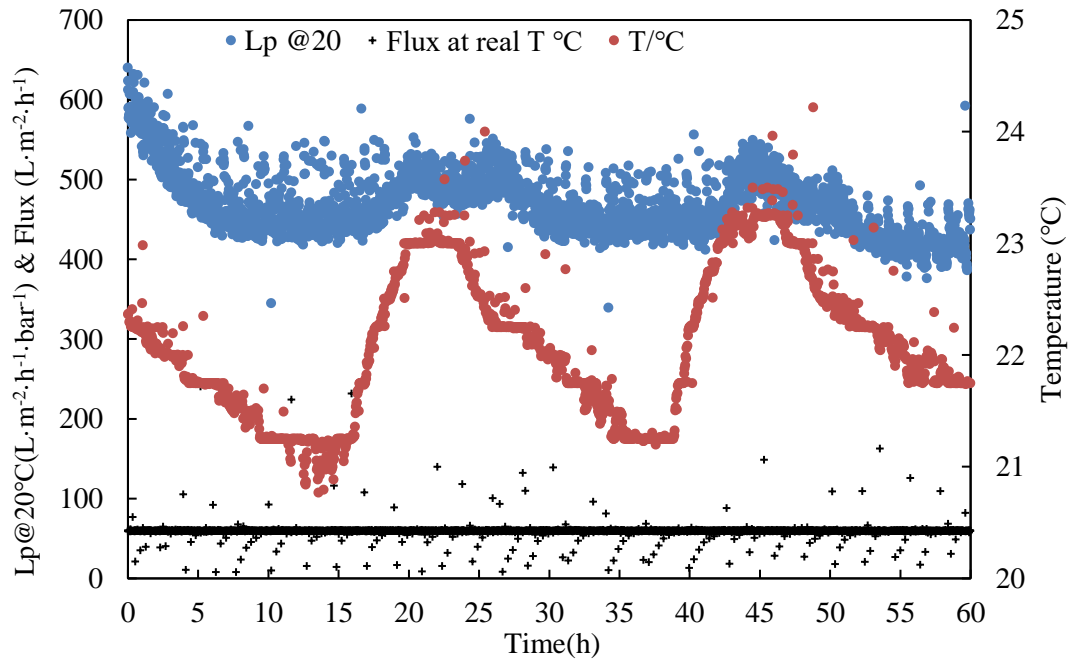


Figure 3-5 Permeability and feed water temperature variations in condition J60t20BW1/3

3.4. Impact of filtration time and flux on hydraulic performances in pilot scale

3.4.1. Overview of permeability variations with CB and AB

Each operational condition in our test (Table 2-6) was carried out with sustainable filtration process for more than 40h. Before explaining in details the different results obtained, the condition J60t60BW1/3 is taken as an example (Figure 3-6), to show the permeability variation curves. It is observed that the filtration system was operated in periodically backwash, with three times of CB between adjacent AB which was related to the fixed backwash sequence in each condition. The other conditions were all operated in the similar status but with different filtration conditions. If a CEB happened, the backwash sequence setting returned to the initial state. According to Figure 3-6, the permeability was partially recovered regularly by cleaning methods. In Figure 3-6 (b), a better fouling removal efficiency by AB than by CB is obtained because the initial L_p after each AB is higher than initial L_p after pervious CB.

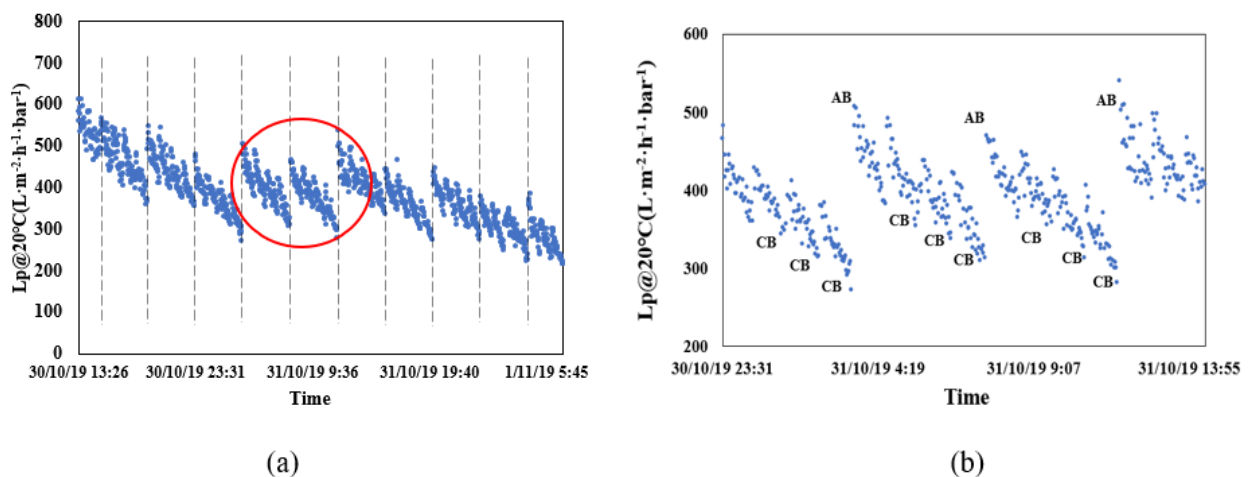


Figure 3-6 Permeability variation versus time in J60t60BW1/3:(a) the whole filtration process, (b) a selected zoomed period

Figure 3-7 (a)(b)(c)(d) shows the permeability variation in 4 representative conditions J30t30BW1/3, J60t40BW1/3, J80t40BW1/3, and J80t60BW1/3, respectively. It is more obvious to observe the significance of AB during filtration in conditions J60t40BW1/3, J60t60BW1/3, and J80t40BW1/3, compared to J30t30BW1/3 and J80t60BW1/3. Through comparison of the four

conditions in Figure 3-7, the overall permeability decreased and stabilized at a smaller value with increased flux and filtration time. Moreover, from Figure 3-7, all these conditions were shown in regular and stable variation of permeability.

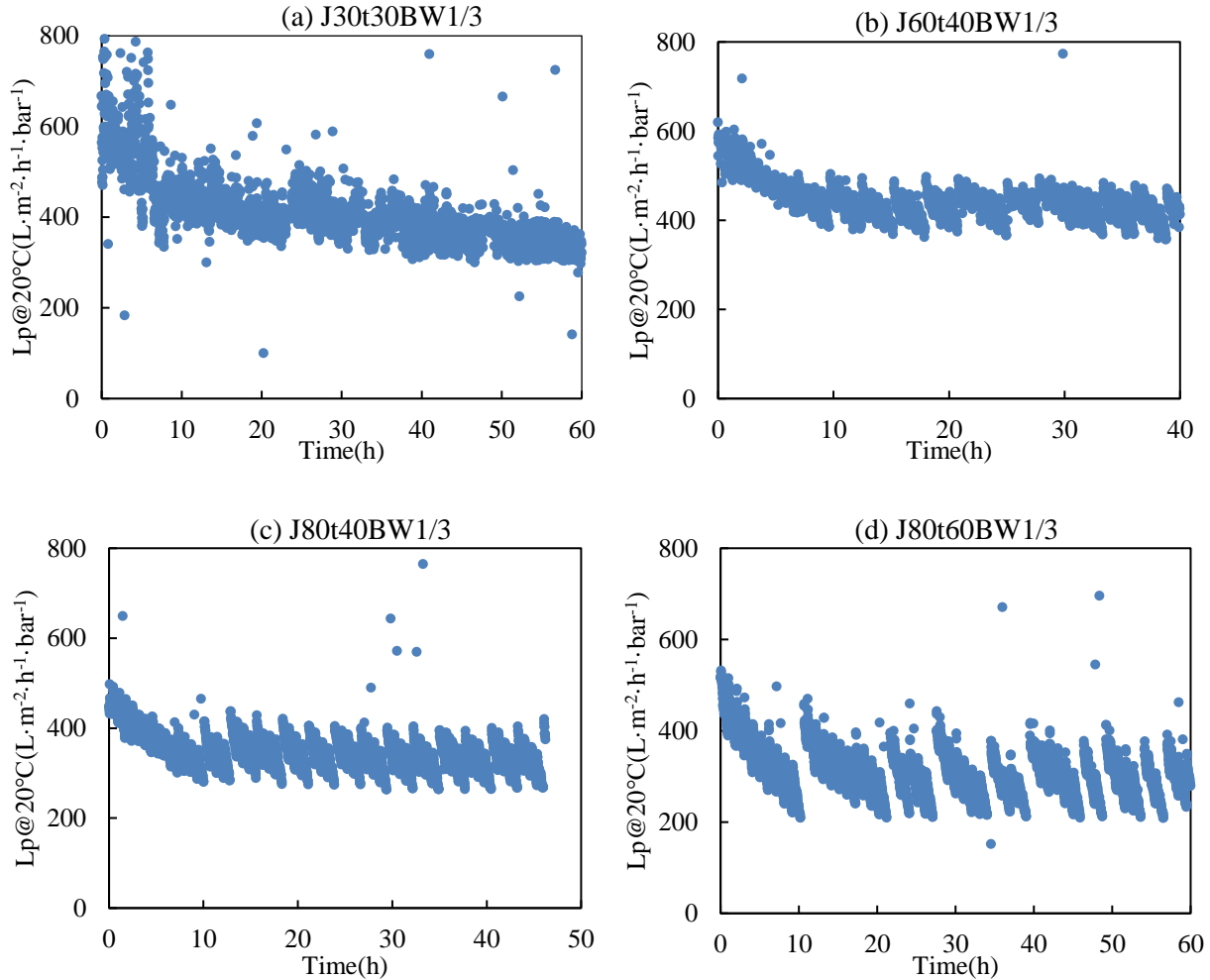


Figure 3-7 Permeability variation in conditions: (a) J30t30BW1/3, (b) J60t40BW1/3, (c) J80t40BW1/3, (d) J80t60BW1/3

3.4.2. Impact of filtration time and flux on fouling reversibility and hydraulic performances

Filtration performance was evaluated by parameters such as backwash effectiveness (fouling reversibility, Eq.8), fouling rate, and fouling resistance. Generally, the total fouling resistance in UF filtration is mainly composed of hydraulic resistance of cake resistance R_c [262], therefore both

increase of flux and/or longer backwash interval can convert more solids, particles and organic matters, into cake fouling resistance over a filtration cycle. To evaluate the influence of flux and filtration cycle time, conditions with gradual increase of filtration cycle time (t) at flux of $60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ (or $80 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$), and with gradual increase of filtration flux (J) at constant t of 30 min (or 60 min) were designed and tested as shown in Table 2-6. The average fouling reversibility by CB and AB obtained under different filtration cycle time and different flux conditions with all over 40 hours of operation is shown in Figure 3-8, which is indicative of the overall backwash effectiveness.

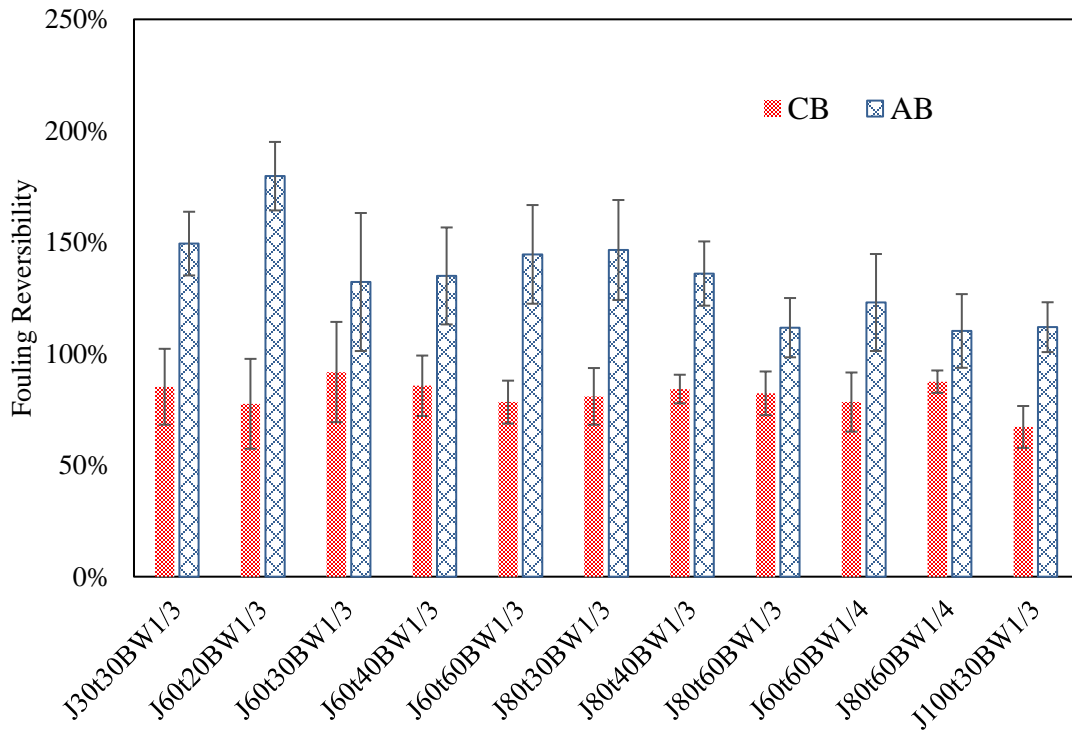


Figure 3-8 Fouling reversibility by CB and AB in different conditions

From the Figure 3-8, fouling reversibility by AB fluctuates from 110% to 180% in all conditions which was conspicuously higher than fouling reversibility by CB (between 70% and 90%). It is therefore very interesting to use AB in long-term fouling management.

A classification of the different conditions is proposed in Table 3-1 based on the fouling reversibility, the total filtrated volume during a cycle (V_{f-cyc}), the initial TMP of a cycle and the number of CEB in 40 hours of operation. From Figure 3-8, J30t30BW1/3 and J60t20BW1/3 exhibited higher AB removal capacity than the others with reversibility values of 150% and 180%,

respectively. These conditions with low flux or low filtration time had the smallest filtrated volume and initial TMP as well as no CEB needed during 40 h operation. Consequently, these conditions are referred as soft conditions. Some other conditions, such as J80t60BW1/3, J80t60BW1/4, and J100t30BW1/3, were resulted in relatively weaker removal ability of CB and AB. These conditions with high flux or high filtration time were also the only conditions where at least 5 CEBs were needed during 40 hours of operation. Thus, these conditions are referred as harsh conditions.

Table 3-1 Classification of the different operating conditions considering the total filtrated volume during a cycle, the initial TMP and the number of CEB in 40h-operation.

Conditions	V_{f-cyc} (L ⁻¹)	Initial TMP* (bar)	CEB times in 40h operation	Types
J30t30BW1/3	135	0.05	0	Soft conditions
J60t20BW1/3	180	0.1	0	
J60t30BW1/3	270	0.1	0	Standard conditions
J60t40BW1/3	360	0.1	0	
J60t60BW1/3	540	0.1	0	
J60t60BW1/4	540	0.1	0	
J80t30BW1/3	360	0.133	0	
J80t40BW1/3	480	0.133	0	
J80t60BW1/3	720	0.133	6	Harsh conditions
J80t60BW1/4	720	0.133	5	
J100t30BW1/3	450	0.167	10	

Initial TMP*: calculated when L_p is $600 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$ at 20°C .

Despite the soft and harsh conditions, the rest is considered as standard conditions with CB reversibility ranging between 80% and 90%, and AB reversibility ranging between 120% and 145%. The harsh conditions of J80t60BW1/ 3, J80t60BW1/4 could filtrate the highest volume (720 L) during one cycle which also means the large quantity of fouling load would be retained by membranes. The highly increased fouling resistance thereby reduced the fouling reversibility of CB and AB. This phenomenon can be explained by the higher filtration volume over a filtration cycle (V_{s-cyc}) as a combined result of flux and time, which means more solids, particles and organic matters retained by membrane thus increasing both irreversible and reversible fouling.

For J100t30BW1/3 condition, even if the total filtrated volume in a cycle V_{f-cyc} (450 L) was lower than in standard conditions (such as J60t60BW1/3 and J80t40BW1/3), the reversibility of both CB and AB were still lower, and with more frequent CEBs. According to Darcy's law, the increase of flux ($100 \text{ L} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$) under constant filtration cycle time can directly increase TMP

(0.167 bar) from the beginning of the filtration process. The increase of TMP may compress the fouling layer (i.e. cake density) which will promote the smallest particles penetration into membrane pores thus enhancing irreversible fouling [258,263]. In this case, although backwashes could flow away the cake foulant on membrane surface significantly, they could not remove the smallest compounds clogging into membrane effectively, thus decreasing CB and AB reversibility [20]. As the irreversible fouling is mainly caused by increased TMP, the irreversible fouling was more sensitive to flux increase than filtration cycle time extension in these tests: this is in agreement with Akhondi et al. [21].

In order to confirm the above results, the variations and composition of fouling resistances before backwash versus time are proposed for each condition (Figure 3-9). J60t20BW1/3, J60t60BW1/3, and J100t30BW1/3 are taken as representatives of the 3 types of conditions.

- In soft condition J60t20BW1/3, membrane resistance R_m was in dominant position during the whole filtration, and R_{irr} did not reach to one-third of R_m .

- In standard condition J60t60BW1/3 with 40 min filtration time longer than J60t20BW1/3 in a cycle, the irreversible fouling R_{irr} gradually reached the value of R_m after 35 hours of operation and stayed in the same order of magnitude than R_m .

- In harsh condition J100t30BW1/3 with the highest flux among all tested conditions, the irreversible fouling R_{irr} increased fastest from beginning and exceeded the value of R_m . Higher values of reversible resistances than in soft or standard conditions are also to be noted.

Through comparison, increase of flux and filtration cycle time both contribute to the increasing rate of R_{irr} during filtration process. The increase of filtration flux (comparison between J60t60 and J100t30) seems to have a higher impact on R_{irr} increasing rate than the increase of filtration time (comparison between J60t20 and J60t60).

From above, the harsh conditions all resulted in occurrence of CEB during 40 h's operation, especially in condition J100t30BW1/3 with the most frequent CEB needed. In practical uses, the CEB interval usually ranges from hours to weeks among different UF processes [264,265]. Because one CEB consumes more chemical agents, permeate water, and energy compared with an AB or a CB, it is necessary to prolong the CEB interval during filtration. It has been previously estimated that sustainable operating conditions lead to less than 2 CEB occurrences per day

[200,231]. Therefore, the harsh conditions cannot be considered as the optimized conditions for long term operation.

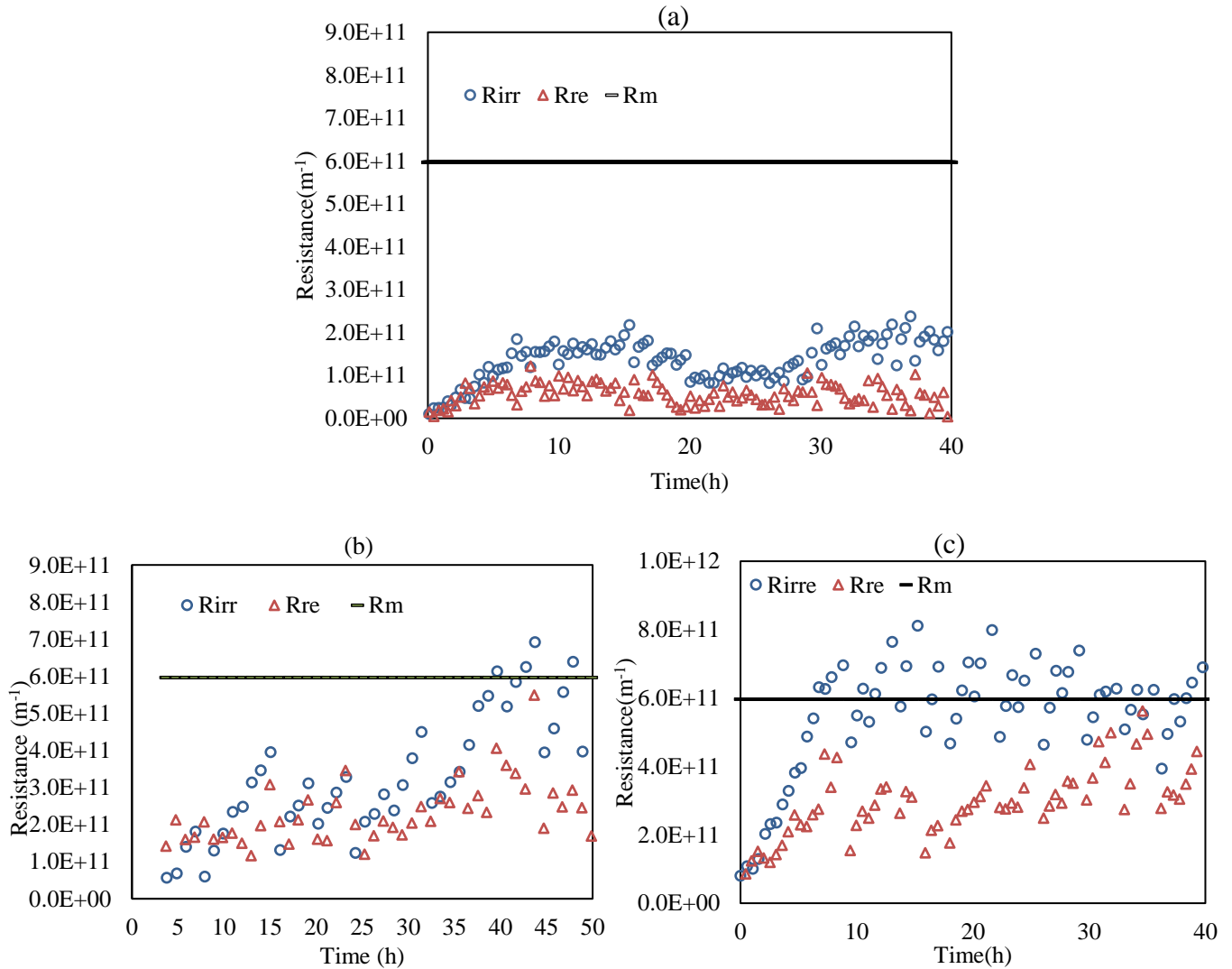


Figure 3-9 Resistances' variations versus time at conditions of (a) J60t20BW1/3, (b) J60t60BW1/3 and (c) J100t30BW1/3

Furthermore, the permeate water was produced for reuse, but it was also used for cleanings. The permeate consumption in CB, AB, and CEB conversely decreased the productivity of the system. Therefore, it is necessary to investigate the water recovery rate (Eq.9) in different conditions to make a comprehensive comparison, shown in in Figure 3-10 all conditions of AB frequency of 1/3. Through calculation, the highest recovery rates occurred in the standard

conditions, especially in conditions of J60t60BW1/3 and J80t40BW1/3 which resulted in 92.7% and 94.5% water recovery, respectively. In addition, the soft conditions showed the lowest recovery rates between 70.7% and 72.2% because of frequent AB and CB, and low flux.

The water recovery rates of harsh conditions J100t30BW1/3 and J80t60BW1/3, were at an intermediate level, ranging from 84.7% to 89.9%. Except the conditions in backwash frequency of 1/3, the conditions J60t60BW1/4 and J80t60BW1/4, resulted in water recovery rates of 92.8% and 90.8% respectively (not shown in the Figure 3-10). The condition J60t60BW1/4 showed almost the same water recovery rate with J60t60BW1/3, with only 0.1% difference because of different permeate consumption between AB and CB. Therefore, the higher water recovery rates among these conditions were obtained from the conditions of J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3 with potential to be the optimized operating conditions in this study. Apart from filtration time and flux, ABs were operated in this study and the AB frequency can have an impact of UF hydraulic performance as discussed in the next section.

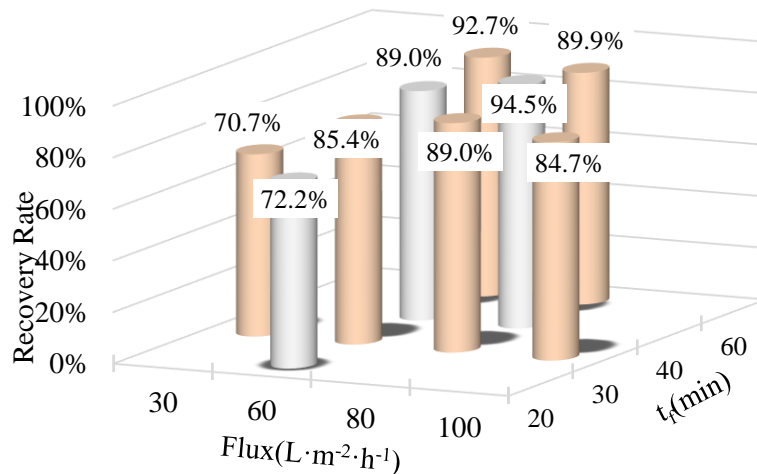


Figure 3-10 Water reuse recovery rate: conditions at backwash frequency of 1/3. The uncertainty of each column is between 1 and 2%.

3.5. Impact of air backwash frequency

Based on the above analysis, AB was definitely resulted in higher reversibility compared to CB, thus reflecting the higher removal efficiency on cake fouling. Indeed, many publications confirmed the high removal efficiency by air-assisted backwash including air drying and air

scouring applied on low-pressure driven membranes [20–22,233,250,266,267]. To evaluate the impact of AB frequency on filtration performance and confirm that the frequencies 1/3 and 1/4 previously studied were the best, 5 different backwash frequencies (1/3, 1/4, 1/6, 1/9, and No AB) (Table 2-6) were tested, all under the constant flux and filtration cycle time ($J = 60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and $t = 60 \text{ min}$). The variation of $(R_{\text{irr}}/R_{\text{m}})_n/(R_{\text{irr}}/R_{\text{m}})_{\text{ini}}$ versus turbidity integration based on Eq.(11) and Eq. (12) reflecting the R_{irr} increasing rate was calculated and is shown in Figure 3-11. The turbidity integration enables to consider feed water quality variation over time considering turbidity as a main indicator of feed quality. It should be noted that the filtration process between 2 CEBs was considered as a new and independent filtration phase to be calculated. Therefore, turbidity integration will be re-calculated from 0, and corresponding to the re-calculated $(R_{\text{irr}}/R_{\text{m}})_{\text{ini}}$. This explains why some conditions present several phases. In Figure 3-11 and Figure 3-12, two main variations can be described. On one hand, Figure 3-11 with AB frequency of 1/9 and no AB presents very high $(R_{\text{irr}}/R_{\text{m}})_n/(R_{\text{irr}}/R_{\text{m}})_{\text{initial}}$ increase with turbidity integration and 2 CEBs occurring during 2 days of filtration. On the other hand, Figure 3-12 with AB frequencies of 1/3, 1/4 and 1/6 where $(R_{\text{irr}}/R_{\text{m}})_n/(R_{\text{irr}}/R_{\text{m}})_{\text{initial}}$ slowly increased with turbidity integration. AB frequency in conditions of BW1/3, BW1/4 are the best with excellent R_{irr} removal abilities, which can maintain membrane permeability over $200 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ in 2 days of filtration. No CEB occurred in these conditions. Therefore, conditions BW1/3 and BW1/4 are more suitable for long term operation at industrial scale. This result is similar to the result on seawater [202], who concluded that AB was more effective to control fouling resistance in conditions of BW1/3 and BW1/5, than that in conditions with lower AB frequency BW1/7 and BW1/9, all under the same $J = 60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and $t = 60 \text{ min}$.

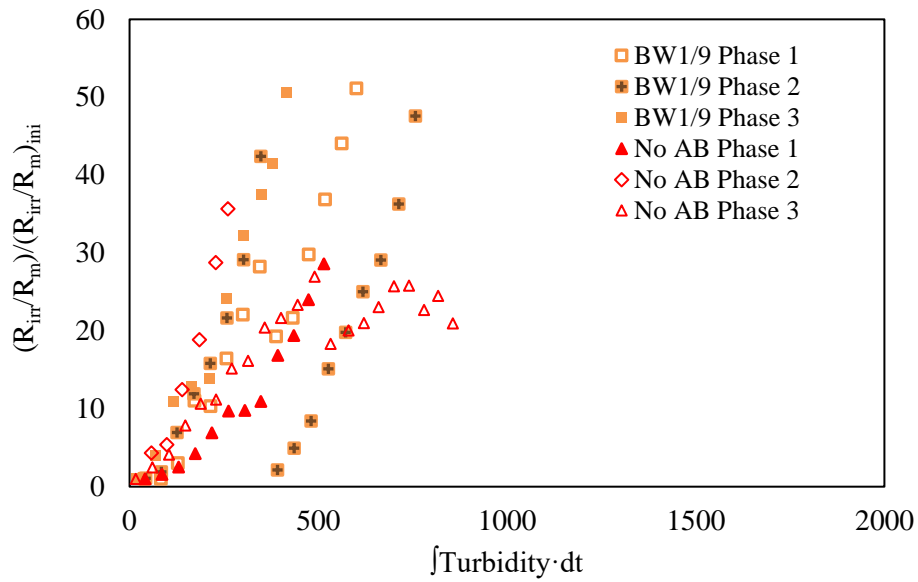


Figure 3-11 Variation of irreversible resistance as a function of the integral of turbidity vs. time for different BWs (1/9 and No AB) at flux=60 L·m⁻²·h⁻¹ and t = 60 min

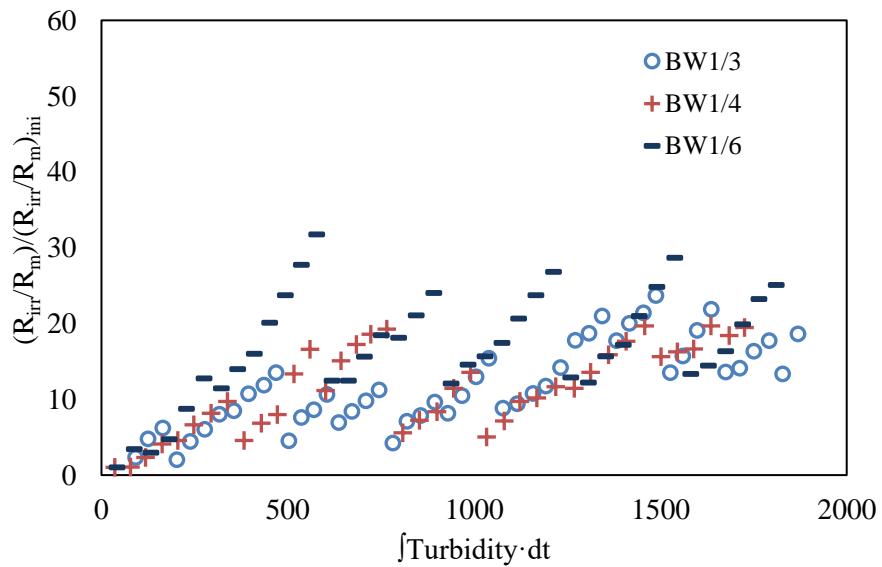


Figure 3-12 Variation of irreversible resistance as a function of the integral of turbidity vs. time for different BWs (1/3, 1/4, and 1/6) at flux=60 L·m⁻²·h⁻¹ and t = 60 min

3.6. Hydraulic filtration performance in long-term in optimized conditions

From the above sections, the three best operating conditions on short-term experiments are J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3. To verify the feasibility and sustainability of the optimized conditions, long terms tests were conducted on condition J60t60BW1/3 both in winter (from 26/2/2020 to 18/3/2020) and in summer (from 1/7/2020 to 27/7/2020). Figure 3-13 and Figure 3-14 show the permeability variation for more than 20 days, respectively, in winter and in summer. From Figure 3-13 and Figure 3-14, the UF pilot plant showed stable and continuous filtration performance, and great permeability recoveries (by CB, AB, and CEBs) during long-term operation, both in summer and winter. The occurrence of CEB were all less than once per day which meets sustainable objectives. However, the frequency of CEB is unstable, which is found to be related to the feed water quality and temperature changes. In summer, the permeability drops seem faster than in winter, but it also seems that permeability recoveries by CEB are higher than in winter. Faster permeability drops in summer can be related to more algae observed in the effluent of WWTP resulting in more suspended solids and organic matters. Therefore, the fouling potential on membrane in summer was relatively higher than in winter but at the same time the mean water temperature in summer was 27.7 ± 1.3 °C, while in winter it was 20.2 ± 3 °C which could lead to better reversibility of cleanings including AB, CB, and CEB [268–270]. Indeed, the mean reversibility of AB in summer and in winter are respectively $144 \pm 46\%$ and $135 \pm 21\%$. Besides, the mean permeability recoveries by CEB in summer and in winter are respectively $308 \pm 15\%$ and $246 \pm 30\%$. However, the water temperature increase had little effects on CB performance: the mean reversibility of CB in summer and winter are all around 80%. Although there was higher fouling potential on membrane in summer, the corresponding temperature increase improved cleaning efficiency of AB and CEB, finally resulting in sustainable permeability recovery and reasonable CEB frequency.

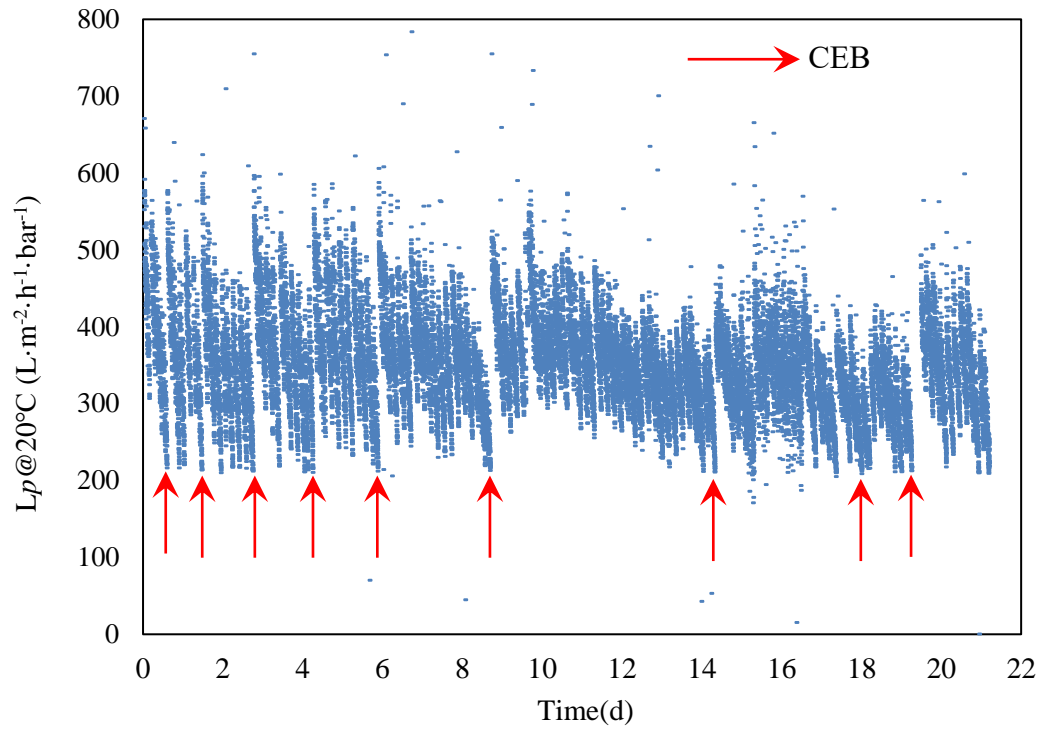


Figure 3-13 Permeability variation in winter from 26/2/2020 to 18/3/2020

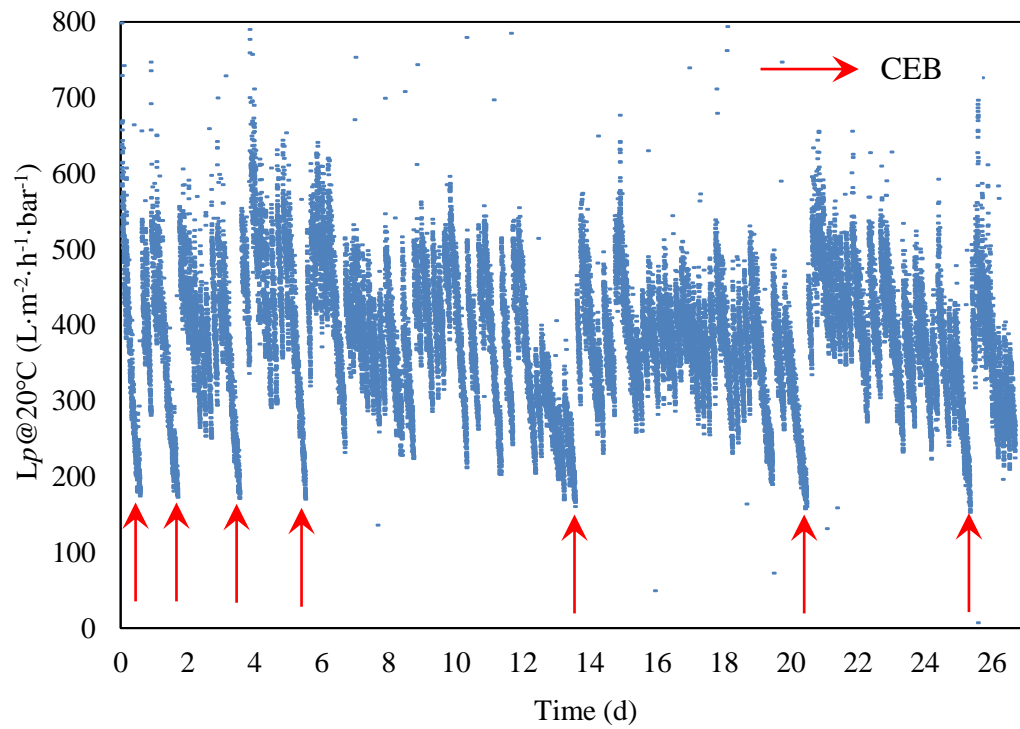


Figure 3-14 Permeability variation in Summer from 1/7/2020 to 27/7/2020

3.7. Conclusion

In this chapter, ultrafiltration is considered as tertiary treatment for municipal wastewater reuse in semi-industrial operation and for the first time some conclusions can be given:

(i) Concerning the most suitable operating conditions for UF: soft conditions with lower flux or frequent physical cleaning (short filtration time) resulted in too little productivity and were discarded. Harsh conditions with higher flux or longer filtration time were also discarded because of high occurrence of CEB which led to difficult permeability stabilization and high consumption of permeate and chemicals. Finally, standard conditions J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3 stood out from the others with higher overall performances. Of course, these conditions are function of the WWTP and feed water quality, but still, this study defined some ranges of adequate operating conditions for future water managers and engineers for municipal wastewater tertiary treatment before reuse.

(ii) By analyzing the influence of AB frequency on irreversible fouling management, the irreversible fouling resistance increased faster with the decrease of AB frequency during operation. Optimum AB frequencies of BW1/3 and BW1/4 under constant flux of $60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and t of 60 min found in (i) were confirmed.

(iii) Concerning the selected sustainable conditions, more than 20 days of operation in condition J60t60BW1/3 in winter and in summer confirmed that the UF pilot plant could provide sustainable and adaptable filtration performance regardless of the temperature and feed water quality.

CHAPTER 4. Impact of UF on water quality

4.1. Water quality analysis for reuse purpose

Permeate quality analysis is critical to the realization of water reuse. To ensure the safety and reliability of reused water in the frame of this study, the permeate quality was compared to the guidelines for water reuse. Over the course of the thesis, changes in standards, the pandemic and the emergence of pollutants (microplastics) and the possibility of developing a new analysis technique in partnership with PerkinElmer: the analysis evolved over time. Table 4-1 shows the UF permeate quality during both Campaign 1 (October 2019 – March 2020) and Campaign 2 (July 2020 – April 2021) compared to WHO guidelines for water reuse, French reuse standard, and European parliament regulations for water reuse [11–13]. The removal rates between UF feed and UF permeate were calculated to evaluate the benefit of UF. Moreover, as required by the standards, the removal rates between WWTP feed and UF permeate were also calculated when possible. As a result, the UF system can greatly retain the microorganisms, including *E.coli*, *Enterococci*, spores of anaerobic sulphito-reducers, and bacteriophages, all under the detection limitation of the analysis methods. Particularly, the treatment processes in WWTP together with UF system achieved more than 5 log removal of bacteria *E.coli* and *Enterococci*. Besides, the COD, BOD₅, TSS concentrations and turbidity of permeate all fulfil the three standards. However, because of the detection limit, the log removals of the spores of anaerobic sulphito-reducers and virus (Specific F-RNA bacteriophages) cannot fully reach more than 4. In fact, the concentrations of the spores of anaerobic sulphito-reducers and virus in permeate were very low and under the detection limits ($< 1 \text{ CFU} \cdot 100\text{mL}^{-1}$ for bacteria or $< 1 \text{ PFP} \cdot 100\text{mL}^{-1}$ for virus). In addition of the parameters in Table 4-1, the parameters of *Salmonella*, *Legionella spp.*, and viable pathogenic helminth eggs, were tested only during Campaign 2 as mentioned in the EU regulation, and they were not found in any of the permeates. Therefore, regardless of the filtration conditions and the variation of feed water quality in this study during the first experimental campaign, UF as tertiary treatment can provide high-quality water for reuse as it reaches the WHO guidelines for reuse and the French reuse standards with quality “A”. According to the results of the second experimental campaign, UF permeate also reaches the very recent EU regulation for reuse with quality “A”.

Additionally, because of the international Covid-19 pandemic, coronavirus SARS-CoV-2 was measured in the raw wastewater of WWTP, UF feed, and UF permeate in November 2020. Coronaviruses are continuously detected in different types of wastewater contaminated through

several sources, such as handwashing, sputum, vomit, and feces from disease carriers [271–273]. Among coronaviruses, SARS-CoV-2 responsible for the Covid-19 disease can cause possible transmission via contaminated waste surfaces and aerosols from wastewater systems [274–276]. Generally, coronavirus in wastewater can be active for several days. As reported by Bivins et al. [277], 90% of SARS-CoV-2 in wastewater can be reduced in 1.6-2.1 days under room temperature, and 99% of SARS-CoV-2 in wastewater can be reduced in 2.9 – 6.5 days under room temperature. To ensure the safety of the water production for reuse, the detection of SARS-CoV-2 from the influent to end effluent is necessary. The results showed that the raw wastewater was tested positive with the envelope protein gene E above the detection limit (17 000 genome units per liter) but below the quantification limit (170 000 genome units per liter) and the nucleoprotein gene N was not detected. The UF feed and UF permeate were tested negative to SARS-CoV-2 by these PCR assays. Because the limit of detection of this PCR method is quite high, it is uncertain to say that the virus is completely absent in the waters or not. However, according to recent publications in the field, the findings indicate that secondary wastewater treatment can contribute to decrease SARS-CoV-2 concentration in wastewater, and tertiary treatment or disinfection enhance the inactivation of SARS-CoV-2 in WWTPs [278–281]. Bogler et al. (2020) stated briefly in their review that SARS-CoV-2 should be removed reliably by UF. Based on the above analysis and detection, the UF permeate after UF treatment in this test has no infectivity of SARS-CoV-2 and can be reused safely. Besides, in order to fully assess UF as a barrier to SARS-CoV-2, lower detection limits in PCR should be sought or new analytical tools should be developed to quantify infectious SARS-CoV-2 for example.

Table 4-1 Mean UF feed and permeate qualities compared to WHO guidelines, and French reuse standards for irrigation

Parameters	Campaign 1 (October 2019 – March 2020)			Campaign 2 (July 2020-April 2021)			WHO guidelines	French standard (A)*	European parliament regulation (A) *
	UF permeate			UF permeate					
	Mean ± SD	RRs* by WWTP+UF (%)	RRs* by UF (%)	Mean ± SD	RRs* by WWTP+UF (%)	RRs* by UF (%)			
<i>E. coli</i> (CFU·100mL ⁻¹)	< 1	> 6.7 (log*)	> 4.8(log*)	< 56	> 6.5 ± 1.4(log*)	> 2.0 ± 0.5(log*)	≤ 200	≤ 250	≤ 10 / ≥ 5 (log*)
<i>Enterococci</i> (CFU·100mL ⁻¹)	< 1	> 6.2 (log*)	> 4.4(log*)	< 56	> 5.4± 1.4(log*)	> 1.1 ± 0.3(log*)	-	≥ 4 (log*)	-
Anaerobic sulphito-reducers (spores) (CFU·100mL ⁻¹)	< 1	> 4.1 (log*)	-	< 1	> 3.5 ± 0.3(log*)	> 2.8 ± 0.1(log*)	-	≥ 4 (log*)	≥ 5 (log*) /Absence
Specific F-RNA bacteriophages (PFP·100mL ⁻¹)	< 1	> 3.6 (log*)	-	< 30	> 2.6 ± 0.8(log*)	-	-	≥ 4 (log*)	≥ 6 (log*) /Absence
COD (mgO ₂ ·L ⁻¹)	< 10	> 98 %	>44%	< 30	> 95.4± 2.0%	-	-	< 60	-
BOD ₅ (mgO ₂ ·L ⁻¹)	-	-	-	< 3	> 98.1± 0.3%	-	-	-	≤ 10
TSS (mg·L ⁻¹)	< 2	> 97%	> 56%	< 2	> 98.5± 0.8%	> 84 ± 6%	≤ 30	< 15	≤ 10
TOC (mgC·L ⁻¹)	4.61 ± 0.61	94 ± 3%	70± 11%	-	-	-	-	-	-
Turbidity (NTU)	0.42 ± 0.35	-	85± 7%	< 0.31	> 99.9± 0.0%	> 86 ± 11%	≤ 2	-	≤ 5
pH	7.46 ± 0.07	-	-	7.5 ± 0.1	-	-	-	-	-
Conductivity(μS·cm ⁻¹)	1056 ± 70	-	-	-	-	-	-	-	-

CFU: colony-forming unit; PFP: Polyhedral, filamentous, and pleomorphic; log*: log removal calculated from the raw wastewater quality; -: not measured.

(A)*: There are ABCD four different levels of water quality in French water reuse standards and EU regulation. Level A being the best.

RRs*: Removal rates

4.2. Detection of microplastic particles (MPs) in WWTP effluent and removal by UF

4.2.1. Selection of rinsing water for MP analysis

To reduce external microplastic contamination, it is necessary to select a qualified water for rinsing during pretreatment and filtration processes described in section 2.8.1.2. In this part, the microplastic particles in Evian water, ultrapure water, distilled water (DW), and tap water were all detected more than twice for comparison. Before each filtration, the glass devices will be first rinsed with ultrapure water and acetone, then rinsed with the tested water before filtration. The distribution of microplastics in different types of water samples under $6.25 \mu\text{m}/\text{pixel}$ is shown in Table 4-2. Each test consisted in filtrating 1 L of sample through the $3 \mu\text{m}$ gold filter. Except microplastics, all particles shown in Table 4-2 contains mainly protein and cellulose, and a bit of CaCO_3 based on siMPle database. During the filtration of the samples through $3 \mu\text{m}$ gold filters, the filtration processes were all rapid, and in apparent constant flux. The time for filtration of 1 L sample were all under 30 seconds. Therefore, the filtration apparatus supports effective filtration efficiency on clean water samples.

Looking at MPs in Table 4-2, Evian, and tap water contains less numbers and types of microplastics compared to the others while distilled water contains the most. Because the distilled water and ultrapure water are made from tap water through some advanced purification process, the increase of microplastics in distilled water and ultrapure water may come from process contamination, such as the used polymer pipes, tapes, and membranes. Looking at all particles (MPs + cellulose, salts, organics...), ultrapure water contains the lowest while tap water contains the most. In Evian water the microplastic dimensions are larger than ultrapure water and tap water. Since larger particles and/or larger amounts of natural particles such as proteins and cellulose can increase the chance of microplastics to be covered on filters, the tap water and Evian water are not recommended as rinsing waters. Through analysis, the ultrapure water can therefore be considered as the best rinsing water. To avoid the external contamination of microplastics during rinsing, the use of ultrapure water is controlled within 20 mL, with less than 1 microplastic involved averagely.

Table 4-2 Detection results in different types of water

Water types	Particles	Particle concentration in each test (Particles·L ⁻¹)							Average ± SD	Types of MP	Major dimension of MP (µm)	Minor dimension of MP (µm)
		1	2	3	4	5	6					
Ultrapure water	Only MP	11	22	16	38	40	4	22±3	pe, pp, ps, pa, pet, pvc,	20-150	10-60	
	All particles	-	-	-	158	272	205	212±47	silicone			
DW	Only MP	31	23	12	38			26±10	pa, pp, ps, pet, pvc, abs, pe,	20-250	10-120	
	All particles	714	498	197	825			558±239	eva, silicone			
Evian	Only MP	19	2	4				8±7	pa, pe, pp, ps, pet	20-200	16-100	
	All particles	348	530	171				350±146				
Tap water	Only MP	3	9					6±3	ps, pa, pp, pvc	45-80	10-40	
	All particles	1566	1254					1410±156				

4.2.2. Selection of scanning pixel resolution of microscopy

The pixel size of focal array plane detector (pixel resolution) is a combination function of native focal plane array (FPA) detector element size, objective magnification and Intermediate optics magnification. On imaging of micro-FT-IR, the selection of pixel size can be 6.25 µm, 25 µm and, 50 µm. The pixel size of 50 µm is not recommended for imaging because the maximum pore size of filter in this study is 25 µm from the stainless- steel filter. To compare the differences on spectra map under different pixel size, the ultrapure water was both imaged under pixel size of 6.25 µm and 25 µm. A volume of 1 L for all the samples was filtered through a 3 µm gold coated filter with a filtration square at 6000 µm × 6000 µm.

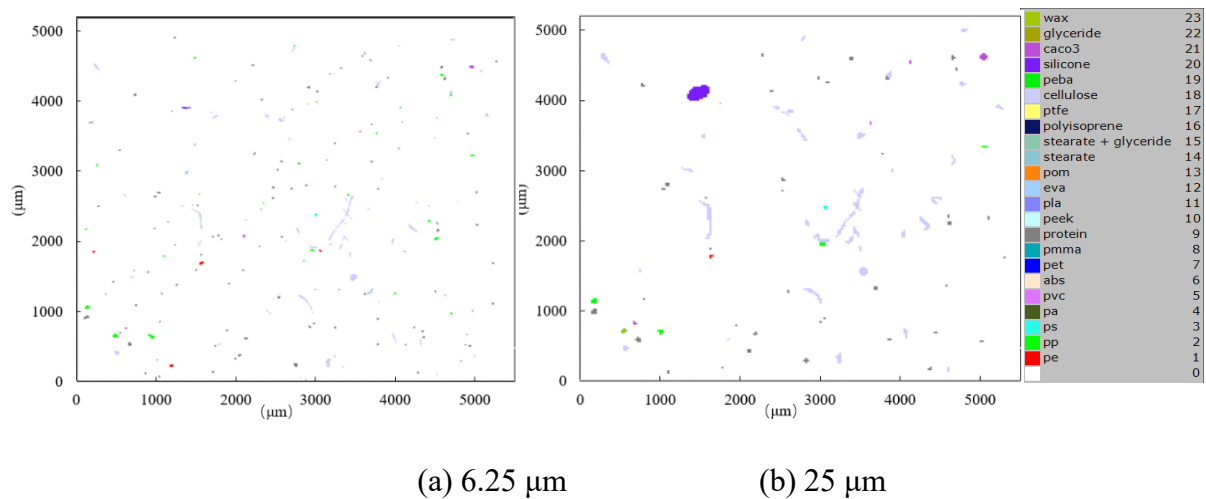


Figure 4-1 Spectra map of ultrapure water with all particles by siMPLe (a) pixel size at 6.25 μm (b) pixel size at 25 μm

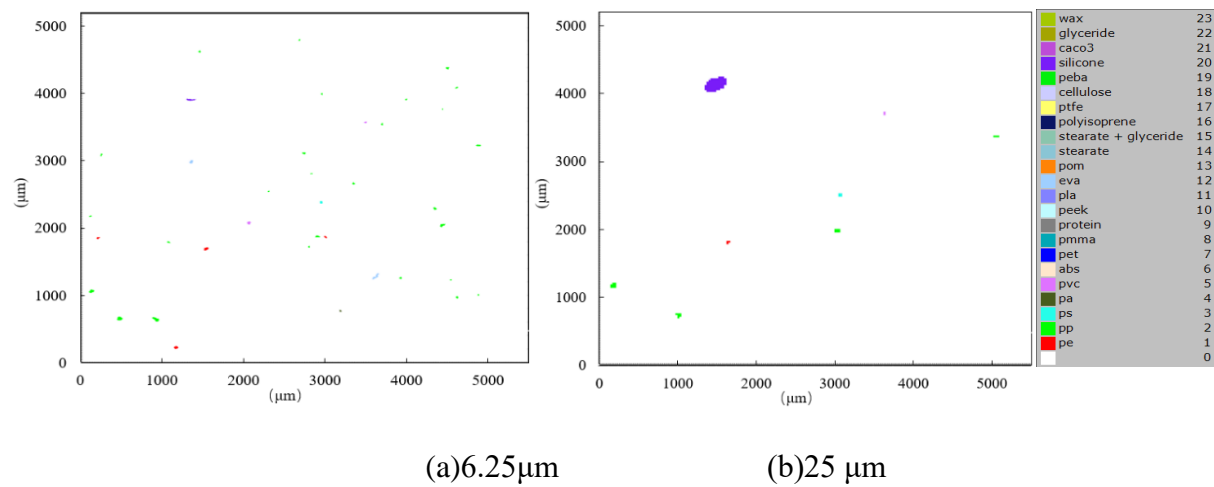


Figure 4-2 Spectra map of ultrapure water with only microplastics (a) pixel size at 6.25 μm (b) pixel size at 25 μm

Figure 4-1 shows the spectra maps with all particles of ultrapure water by siMPLe at 6.25 μm pixel and 25 μm pixel. In comparison, the position, shape, and material types of all particles at 25 μm pixel are the same as that at 6.25 μm pixel. But the dimension and counts of particles are quite different. The dimensions of all particles at pixel size of 25 μm is larger than pixel size of 6.25 μm . There were more smaller particles discovered at pixel size of 6.25 μm , while the smaller particles were ignored at pixel size of 25 μm . Besides, detection under larger pixel size sometimes may consider several similar small particles as one large particle because low

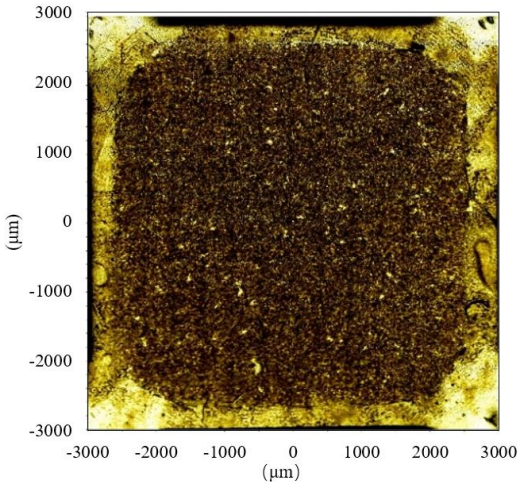
resolution cannot clearly distinguish the edges of small particles. In spectra map Figure 4-2, only 8 microplastics were discovered under 25 μm pixel while 38 microplastics discovered under 6.25 μm pixel. In spectra map Figure 4-1, 85 particles were discovered under 25 μm pixel while 206 particles discovered under 6.25 μm pixel. The image with 6.25 μm pixel size can detect particles with dimension size of less than 10 μm , while the particles detected under 25 μm pixel are all larger than 25 μm . In result, the choice of pixel size has significant impacts on the counts and dimension sizes of microplastics, especially on smaller particles. As scientific research requires precise and rigorous attitude, it is better to use 6.25 μm per pixel for imaging. Although it takes longer time and larger space for storage, the smaller pixel will provide more accurate numbers, dimensions, and identification of particles. The imaging under 25 μm per pixel is good at shorter detection time and lower storage space, it is recommended to be used when the known size of microplastics is larger than 25 μm or the pore size of the filter is larger than 25 μm . In this study, the image by focal array plane detector on 25 μm filters uses the 25 μm pixel resolution, and the image on 3 μm and 5 μm filters uses the 6.25 μm pixel resolution in the next sections.

4.2.3. Digestion protocols

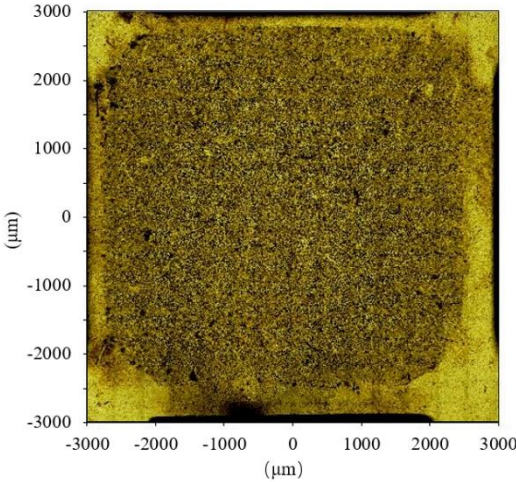
In this study, the UF permeates are clean enough to be filtrated directly for detection without digestion process. Indeed, the quality of UF permeate is quite good, with turbidity lower than 1 NTU, total organic carbon (TOC) lower than 10 $\text{mgC}\cdot\text{L}^{-1}$, and non-visible suspended particles. The filtration volume of these samples on the detection filter can reach up to 500 mL to 1 L with clear visible survey by microscope. The UF feed from WWTP was filtrated in two steps to ensure all microplastics can be detected correctly. Because there are more organic matters in the UF feed than UF permeate, the UF feed sample needs to be digested. An optimisation of this digestion was realised to define the duration and the volume of reagent. Therefore, as digestion with H_2O_2 can reduce the influence of organic matters on MP detection[240,282], the UF feed sample was mixed with 30% H_2O_2 with volume ratio at 1:1, then the samples were analysed under different digestion duration. The mixture was kept in room temperature without stirring. Figure 4-3 shows the visible images of the filters by FT-IR microscopy. From case (a) to case (c), the samples were all filtrated through the 3 μm filter with 50 mL of feed water (100 mL in total with 50 mL UF feed and 50 mL H_2O_2) from the same sampling bottle. It is obvious to see that the organic matters were increasingly digested with H_2O_2 as the reaction time extends. In

view of Figure 4-3, the deposit in case (a) and case (b) is quite dense, the microplastics may be covered and failed to be detected. The samples in case (a) and case (b) were for 1 and 7 days. After 10 days reaction, the visible images in case (c) and case (d) were much clearer than that in case (a) and case (b). However, a point worth to be noted here is the organic deposit especially in case (a) and case (b) is not mainly composed of proteins or cellulose based on the spectra maps generated by siMPle shown in Figure 4-4.

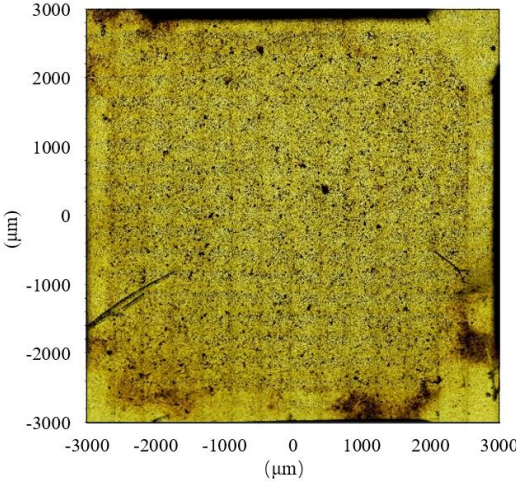
Figure 4-3 UF feed deposits on 3 μm filter under different digestion duration by H_2O_2 (a) 50mL net UF feed after 1d digestion (b) 50mL net UF feed after 7d digestion (c) 50mL net UF feed after 10d digestion (d) 100mL net UF feed after 10d digestion



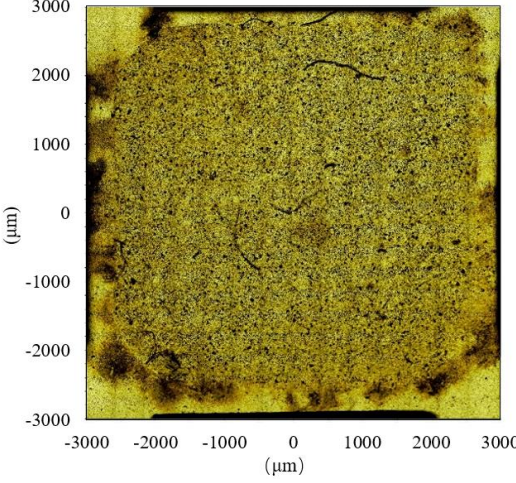
(a) 50 mL – 1 day digestion



(b) 50 mL – 7 days digestion



(c) 50 mL – 10 days digestion



(d) 100 mL – 10 days digestion

Through detection by siMPle, only 3 and 5 microplastic particles was identified respectively in case (a) and case (b) because some particles may be covered by the unknown organic matters, while microplastics in case (c) and case (d) were detected at 11 MPs and 26 MPs, respectively. Actually, case (d) was filtrated with double volume of sample than case (c), the counts of MP were almost in proportional relationship. This result not only confirmed the effectiveness of digestion but also verified the accuracy and reliability of the results. Therefore, digestion with H₂O₂ can reduce the influence of organic matters on MP detection, 10 days digestion with H₂O₂ is recommended for the secondary treated effluent from WWTP.

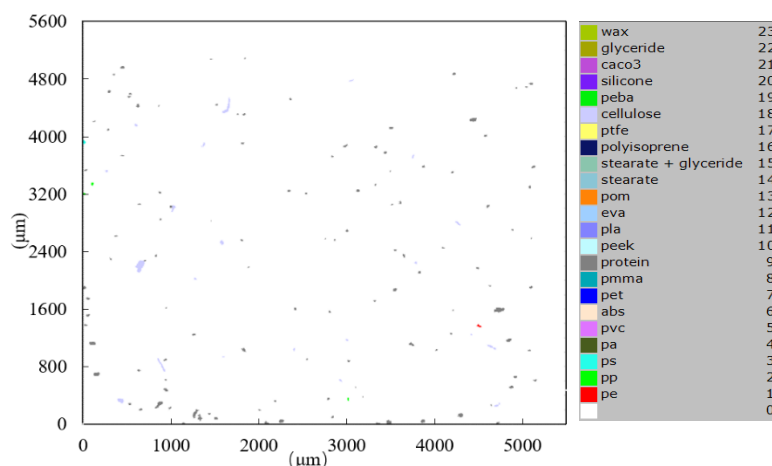


Figure 4-4 Spectra map of detected particles distribution in case (a) by siMPle software

Municipal wastewater treatment plants are considered as a significant source of various microplastics to enter in aquatic environment [283,284]. To reduce microplastic pollution, the removal efficiency of microplastics by the 200 kDa UF membrane in operation in the pilot-plant was investigated in this section. Table 4-3 shows the microplastics concentration both in quantity and in total surface area for UF feed and UF permeate. In result, the UF membrane was confirmed to be effective on microplastics removal, resulting in 86.0 ± 0.6 % removal of microplastic in quantity and 97 ± 2 % removal in surface area. From Figure 4-5, averagely 81.5% of microplastic particles in secondary effluent were small particles with minor dimension $< 40 \mu\text{m}$ and major dimension $< 100 \mu\text{m}$. Through UF membrane, the microplastic particles with minor dimension $> 80 \mu\text{m}$ and major dimension $> 200 \mu\text{m}$ were completely removed. From Figure 4-6, the main components of microplastics in secondary effluent include PA, PE, PP, PS, and PVC. Through comparison of microplastics removal, the removal rate in total surface area is more than that in

quantity which is similar to the UF process in seawater treatment plant (not presented in this study). The result furtherly illustrates that the UF membrane filtration process may cause microplastic fragmentation similar to the above-mentioned UF filtration in seawater treatment plant. The size of the microplastics after UF is very high in comparison with the pore size (factor $\times 1000$). Actually, microplastics are everywhere and it is difficult to remove them all, as the container of the membrane and the pipes for water transportation are all made of plastics.

Table 4-3 The microplastics concentration in quantity and in total surface area of samples from municipal WWTP

Samples	Concentration (MP·L ⁻¹) (mean ± SD)	Concentration in covered surface area (μm ² ·L ⁻¹) (mean ± SD)
UF feed	249 ± 25	(6 ± 2) × 10 ⁵
UF Permeate	35 ± 5	(2.8 ± 0.3) × 10 ⁴
Removal rates by UF	(86.0 ± 0.6) %	(97 ± 2) %

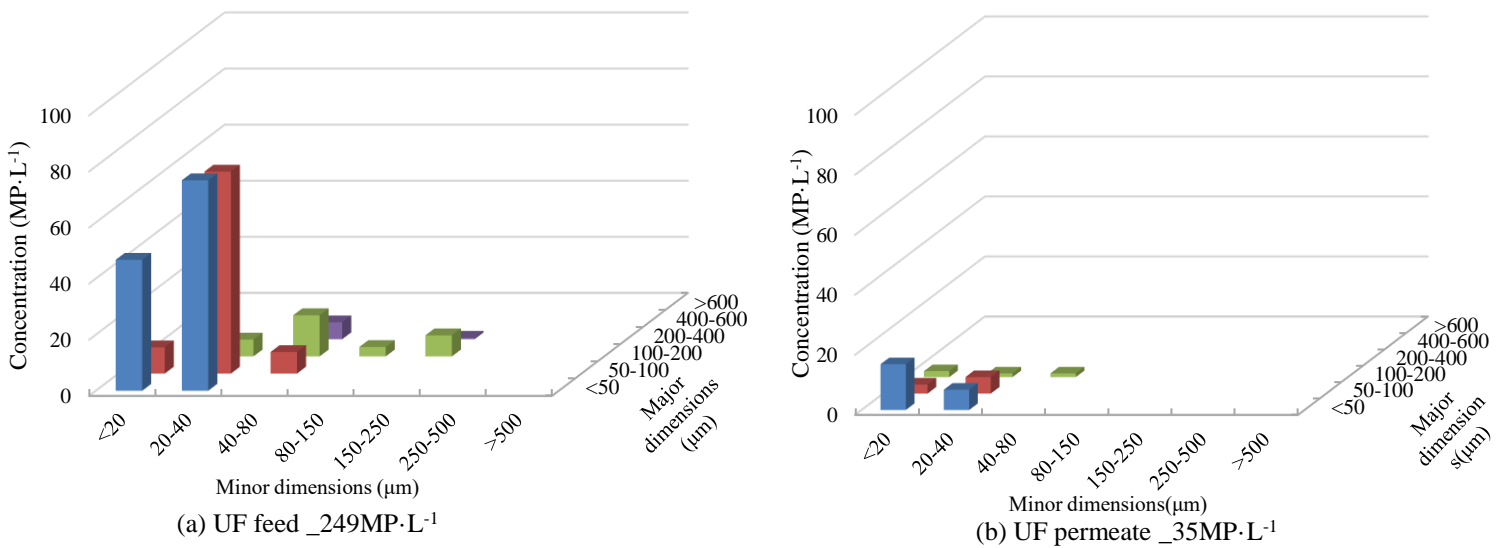


Figure 4-5 Dimensions distribution of microplastics in municipal WWTP samples

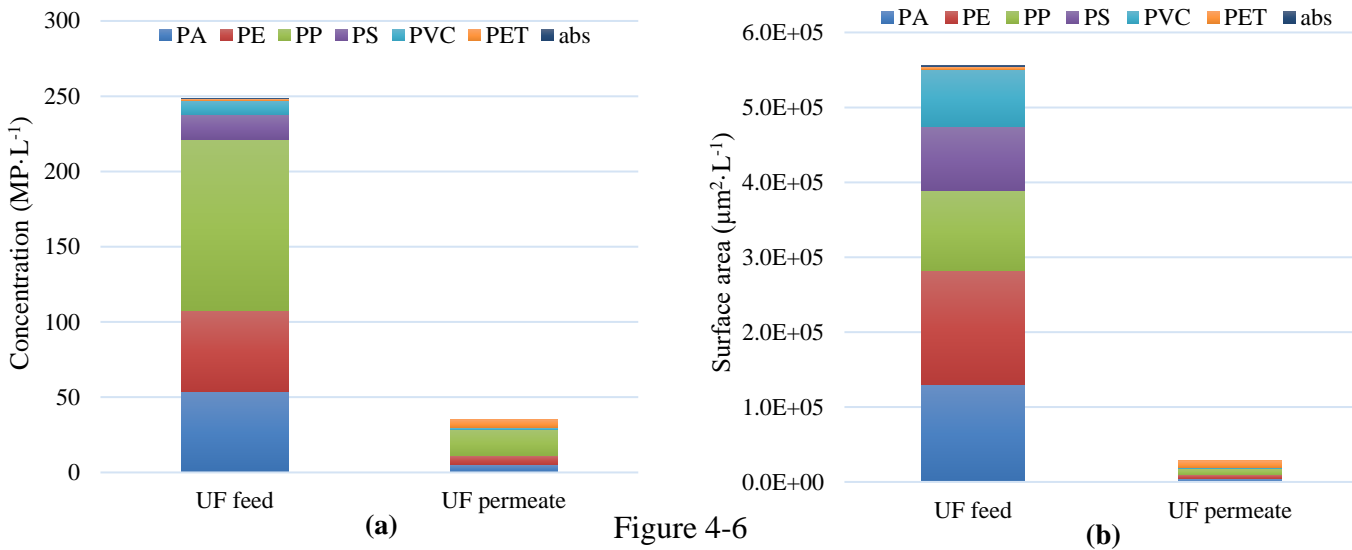


Figure 4-6
Abundance of Microplastic material in both quantity and surface area in samples from municipal WWTP. (a) In microplastic numbers (b) in microplastic surface area

4.3. Conclusion

In summary, the UF permeate quality was detected to be good enough to be reused in non-potable applications as it met reuse guidelines of the WHO, reuse standards of France and the most recent EU regulation for agricultural irrigation. The whole treatment process provided high removal capacity on microorganism removal. The UF permeate was resulted in more than 5 log removal rates of bacteria *E.coli* and *Enterococci* compared to the raw wastewater. Besides, the concentrations of the spores of anaerobic sulphito-reducers and virus in permeate were as low as to be under the detection limits of analytical methods ($< 1 \text{ CFU} \cdot 100\text{mL}^{-1}$ for bacteria or $< 1 \text{ PFP} \cdot 100\text{mL}^{-1}$ for virus). This works confirms the great interest of UF as tertiary treatment of conventional WWTP for water reuse. Considering the international Covid-19 pandemic right now, the UF permeate after UF treatment in this test would have no infectivity of SARS-CoV-2 and could be reused safely. Recently, microplastic contamination have attracted worldwide attention as a new emerging pollutant. The study made the optimisation on rinsing water and scanning pixel resolution selection to obtain more accurate results. The Ultrapure water as selected as rinsing water due to its lower quantity of all particles and the smaller dimensions of the tested samples compared to Evian, tap water, and distilled water. The scanning pixel size at $6.25 \mu\text{m}$ pixel resolution shows more accurate identification and quantification of particles

compared to 25 μm pixel resolution. The imaging under 25 μm per pixel is good at shorter detection time and larger storage space, it is recommended to be used when the known size of microplastics is larger than 25 μm or the pore size of the filter membrane is larger 25 μm . The image on 3 μm and 5 μm filters uses the 6.25 μm pixel resolution. In results, The UF membrane could provide 86.0 ± 0.6 % removal of microplastic in quantity and 97 ± 2 % removal in surface area of UF feed. However, the UF system showed effective removal capacities and considerable concentration of microplastics, but the microplastics fragmentation during each treatment process seems hard to avoid. The smaller the microplastics, the greater the difficulty of removal and the greater the potential harmfulness which needs to be highly investigated in the future. Consequently, this works confirms the great interest of UF as tertiary treatment of conventional WWTP for water reuse.

CHAPTER 5. Impacts of NaOCl-assisted backwash on UF fouling management and total economic costs

5.1. Impact of chlorinated-assisted backwash and air backwash on ultrafiltration fouling management for urban wastewater tertiary treatment

To improve membrane fouling management, the NaOCl-assisted backwash has been developed to improve permeability maintenance and reduce the need for intensive chemical cleanings. This section is aimed to focus on the efficiency of NaOCl-assisted backwash in real UF pilot scale and with periodic classic backwash (CB) and air backwash (AB). The impacts on hydraulic filtration performance, physicochemical properties of membrane material under different addition frequency of NaOCl, and the performance of chlorinated CB and AB will be discussed. The membrane foulants fed by municipal secondary effluent includes organic fouling, inorganic fouling and microbial fouling [15,227,228]. Organic fouling is the main cause of irreversible fouling for membranes. Nonetheless, the efficiency and practicality of NaOCl-assisted backwash has not been studied in real UF pilot scale and even less with periodic classic backwash (CB) and air backwash (AB). Therefore, this study aims to investigate the influence of NaOCl-assisted CB and/or AB on the hydraulic filtration performance and fouling management in the semi-industrial UF pilot plant with maximum flow rate of $20 \text{ m}^3 \cdot \text{d}^{-1}$ described in Chapter 2. To this aim, 4 different conditions were studied as mentioned in Table 2-7, and recalled here: Conditions NNNN, NNNY, YYYN, and YYYY for CB CB CB AB with (Y-Yes) or without (N-No) chloride. Besides, the impacts on physicochemical properties of membrane material under different addition frequency of NaOCl and the performance of chlorinated CB and AB will also be discussed.

5.1.1. Permeability variation

During filtration, the permeability variation gives the hydraulic filtration performance of the UF membrane, as shown in Figure 5-1. According to Figure 5-1, the turbidity of UF feed was stabilized around 1 NTU in all conditions. Yet the turbidity shows slight periodical variation with time in a day because the feed water quality seems positive correlated to the variations of

raw wastewater flow rate as demonstrated in Chapter 3. Besides, the temperature of 4 conditions is 21 ± 2 °C and there is also a cyclical variation of temperature during the day and night. Nevertheless, the variation of feed water quality was undergone of all conditions, thus the feed water quality can be considered to have the similar effect on the 4 conditions. From CEB occurrences, condition NNNN was resulted in the highest frequency of CEB, with 1 CEB per day. Additionally, condition YYYY had 1 CEB in 5 days, condition NNNY had in 1 CEB in 5 days, and condition YYYN has 1 CEB in 6 days operation but these 3 last conditions are similar. From permeability stability, condition NNNN showed the fastest decrease of permeability to $200 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ which triggered CEB cleaning. Condition NNNY showed stable variation of permeability, the addition of NaOCl only in AB largely improved the UF filtration performance compared to NNNN, with only 1 CEB occurrence in 5 days. In this condition, the CEB occurred at the 1st day of operation and there is no more CEB in the last 5 days. The reason for faster permeability decreases at 1st day may be due to the variation of feed water quality, or the newly cleaned membrane was sensitive to cake deposition which was in higher chances of pore blocking and lower reversibility of backwash. Similarly, Ye *et al.* [285] found less reversible fouling in the first few cycles filtration of seawater than in the following cycles. In whole filtration, the permeability in condition NNNY showed a gradual downward trend. The permeability after stabilization in condition YYYN showed the most sustainable variation during one-week filtration, stabilized at $260\text{-}580 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, while the permeability in condition NNNN, NNNY, and YYYY were stabilized on average at around $200\text{-}500 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, $230\text{-}580 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, and $300\text{-}650 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, respectively (some individual highs and lows and initial permeability are not taken into account). It seemed that the efficiency of physical backwashes in YYYN was just balanced to the accumulation of fouling rates especially after 2-day operation and the permeability after backwash could achieve to 100% recovery. So far, the addition of NaOCl in either AB or CB could improve the membrane filtration performances greatly, including more sustainable permeability variation and less CEB occurrences.

The recovered permeability in condition YYYY after backwashes was higher than the others which can achieved to $700 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ in the first two days. The CEB in condition YYYY was triggered on the 2nd day of filtration, the fast decrease of permeability was mainly occurred in the 5-6 filtration cycles before CEB. Except the rapid permeability decrease on the 2nd day, the

peak of permeability and average permeability were both higher than the other conditions. Considering that the variation of turbidity in that period was in regular ranges, the exceptional rapid decrease of permeability may be caused by other parameters that are not taken in account, such as the organic matters in the feed water.

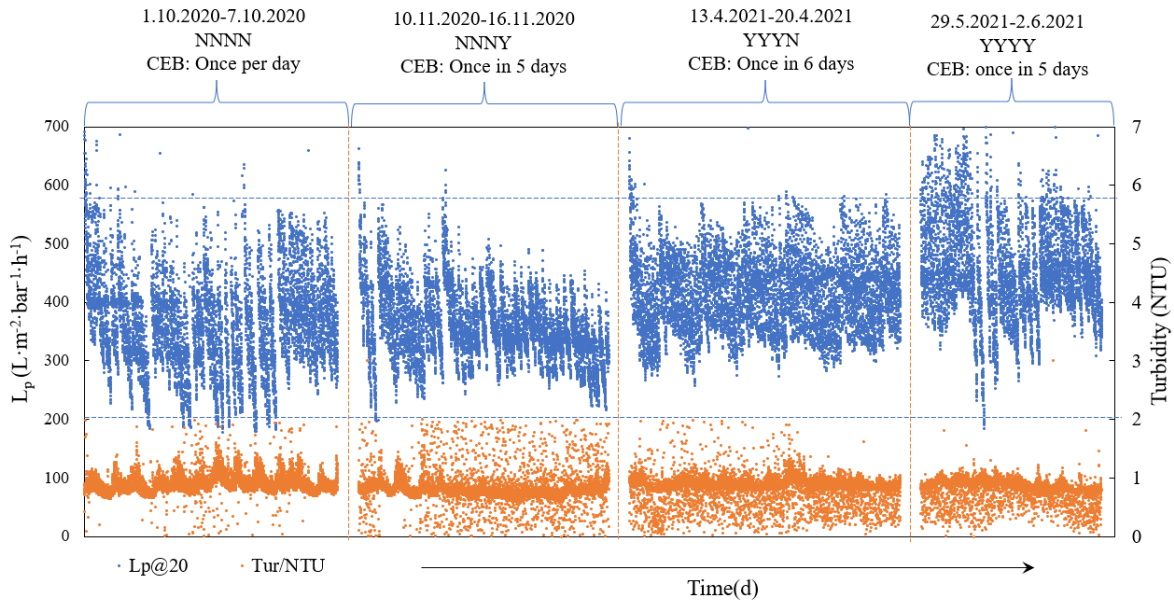


Figure 5-1 Permeability, feed water turbidity and temperature variation versus time in different backwash conditions

A difference of YYYY condition with the other 3 conditions was the peak permeability values. The UF membrane was strongly cleaned by CEB before each condition started which indicated that both the reversible and irreversible fouling can be removed greatly. The fouling formation in the first few cycles of each condition may be mainly because of pore blocking which is irreversible, thus the permeability decreased faster at the beginning of filtration than in the following cycles [20, 21], the physical backwash normally cannot recover the permeability back to the initial levels because of their poor separation strength on irreversible foulant. From Figure 5-1, it can be seen obviously the fast decrease of permeability at the first few cycles in conditions NNNN, NNNY, and YYYN, and even after automatic CEB, the permeability hardly reached $650 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$. In condition YYYY, the permeability could be easily recovered to the $700 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ with all NaOCl-assisted physical backwashes. Before condition YYYY, the pilot plant was just finished with the condition YYYN that already maintained the membrane in very good

permeability variation. With NaOCl addition both in CB and AB, it seems that the condition YYY Y can reach to more than 100% recovery of the decreased permeability in a filtration cycle. The increase of permeability was also observed by Alhweij *et al.* and Wienk *et al.* [219,286]. The UF membrane showed quite good removal efficiency on parameters such as turbidity, microplastics, bacteria, virus and organic matters of UF feed in all conditions, the permeate quality was detected to be good enough to be reused in non-potable applications as it met reuse guidelines of the WHO, reuse standards of France and the most recent EU regulation for agricultural irrigation as shown in Chapter 4.

To summarize the disinfection efficiency of NaOCl, values for concentration multiplied by contact time (CT) considering backwash and CEB occurrences based on the above performance of each condition, were estimated for a simulated 10 years of operation (Table 5-1). The frequency of CEB occurrence of NNNN, NNNY, YYYN, and YYY Y here are estimated after feedback of this study as once per day, once in 5 days, once in 6 days, once in 5 days.

Table 5-1 The consumption of NaOCl of each condition in 10 years

Name	Estimated total CT in 10 years (mg Cl ₂ ·L ⁻¹ ·h)	CT of backwash in 10 years (mg Cl ₂ ·L ⁻¹ ·h)	Equivalent consumption of NaOCl [gCl ₂ ·m ⁻³ (permeate)]	CEB frequency	Maximum NaOCl CT value of UF module mg Cl ₂ ·L ⁻¹ ·day
NNNN	60 833	0	272	Once per day	
NNNY	15 225	3058	175	Once in 5 days	23853
YYYN	19 630	9490	414	Once in 6 days	
YYYY	24 820	10544	546	Once in 5 days	

Referring on impacts of chlorinated disinfectants on membranes, many studies have reported that NaOCl can attack the chemical bonds of polymeric membranes and degrade the membrane material due to long term exposure or excessive dosage of chemicals [206, 214, 286–288]. Similarly, excessive chlorine can impact the PES membranes, but the degree of impact varies a lot in different studies. Some studies pointed out the occurrence of a PES-chain scission and PES material degradation though excessive chlorination, such as after 28500 mg·L⁻¹ total free chlorine of NaOCl (27160 mg Cl₂·L⁻¹) contact for 8 months [289], or after 200-2400 mg·L⁻¹ total free chlorine of NaOCl (190-2287 mg Cl₂·L⁻¹)contact for 10-60d [214]. Fu and Zhang carried out the proposed PES degradation mechanisms that the PES may undergo a chain scission of the

backbone structure into sulfonic acid groups and phenyl chloride groups [221]. Hanafi *et al.* [214] showed that exposure of PES/PVP membranes to NaOCl even led to the appearance of macrovoids in the membrane sub-layer. In contrary, some researchers found slight modification on PES membranes. Alhweij *et al.* [286] found that only slight changes in PES surface roughness was observed after exposure to 238 mg Cl₂·L⁻¹ NaOCl for 3 days. The permeability of PES membrane increased by 74% after chlorine exposure, but SEM images confirmed that there were no observed structural damages on the membrane. Wypyssek *et al.* [290] also stated that a damaged separation skin does not have a significant negative influence on filtration performance of PES membrane. Yadav *et al.* [220] also mentioned that the changes of PES membrane surface properties was observed after exposure to 10000-25000 mg·L⁻¹·day of NaOCl (9530-23825 mg Cl₂·L⁻¹·day) at pH 9-12, but no great changes were observed on membrane mechanical properties and tensile strength. However, according to Wang *et al.* [291], long time exposure to chlorine could impact physical and chemical properties of membranes regardless of the chlorine concentration. This phenomenon can be explained by the study of Abdullah and Bérubé [292] who stated that the more accurate relationship of chemical exposure to membranes should be CⁿT, not the CT. “n” is the power coefficient and was determined <1 from their experiment, which indicated that the contact time of cleaning had a more severe impact on the changes in the physical/chemical characteristics of the membranes than the NaOCl concentration. From Table 5-1, condition NNNY represented the lowest CT of NaOCl and equivalent consumption per m³ of permeate compared to the others while condition NNNN represented the highest. Although the CT with NaOCl in condition YYYYY was less than NNNN, they provided higher contact frequency of NaOCl contact to membranes. Therefore, it is not recommended to apply YYYYY and NNNN condition for long-term filtration purposes in case of the irreversible changes on the membrane.

5.1.2. Fouling resistance

This part investigates the influence of chlorinated backwash on fouling composition. Figure 5-2 shows the variation of fouling resistance at the end of each filtration cycle in the four conditions, membrane resistance (R_m) was considered related to the initial permeability in each

condition ($650 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$) thus was constant at $5.51\times 10^{11} \text{ m}^{-1}$ in all conditions. From Figure 5-2, the R_{irr} and R_{re} in condition NNNN increased rapidly between neighbouring CEBs which can easily increase over R_{m} . This situation can refer to Akhondi *et al.* [21] who confirmed the increase of fouling rate with periodic physical backwash under constant flux and dead-end filtration mode, and the fouling rate could be highly increased when the fouling achieves to a certain amount, together with poor reversibility and cleaning efficiency. In contrary, the variation of R_{irr} and R_{re} in conditions with NaOCl in backwashes (NNNY, YYYN, YYYYY) were mostly under the R_{m} . Actually, it is important to limit the fouling deposition on membrane, especially the R_{irr} on membrane due to its negative impacts on hydraulic filtration performance and cleaning efficiency. In condition NNNY, the addition of NaOCl in AB largely decreased R_{irr} and R_{re} compared to NNNN. Ignoring the filtration from the beginning to the 1st CEB, the overall R_{irr} variation can be separated into two parts, the slower increasing rate of overall R_{irr} from the 2nd to 5th day and the faster increasing rate of overall R_{irr} from the 5th to 7th day shown by the red dashed lines in Figure 5-2. In filtration from 2nd to 5th day, the overall R_{irr} increasing rate was in slow and steady growth during constant flux dead-end filtration. From each backwash sequence (4 filtration cycles with CB, CB, CB, AB), the R_{irr} accumulated on membrane increased in the first 3 filtration cycles with CBs and significantly decreased after an AB with NaOCl which controlled the overall R_{irr} at lower increasing rate. CB without NaOCl could carry away partial cake foulant from membrane surface, but could not remove the smallest compounds adsorbed onto the membrane material effectively, when filtration restarts the residual foulant will adhere to the membrane surface again and accumulated continuously [20]. AB itself has powerful strength with air injection to loosen the foulant layer and to break and/or carry away the smaller particles blocked and absorbed into membrane [231]. Then after air injection, backwash with NaOCl can inactivate the microorganisms and oxidize organic matters deposited on membrane. The cooperation of air flow and NaOCl greatly improved the backwash efficiency on foulant cake removal. When the fouling resistance achieved to a certain value, the AB and CB cleaning efficiency was decreased and the accumulation of integral foulant on membrane became faster than before. Thus, in filtration from 5th to 7th day, a higher overall R_{irr} rate was observed and the fouling cake seemed hardly to be removed by CBs and unable to be controlled by ABs. This suggests that as the continuous accumulation of fouling deposition, the foulant cake maybe changed to a more compact structure. The increased fouling rate may be caused by increased

local flux due to loss of effective filtration area by irreversible fouling. The strength of physical backwash was powerless in front of the increased fouling layer, and maybe the exposure of NaOCl to fouling layer became useless due to the low concentration and short contact time.

The distribution of R_{irr} and R_{re} in condition YYYN were in the most stable variation, and R_{irr} was lower than R_{re} and lower than R_m ($R_{irr} < R_{re} < R_m$) .i.e. so it appears that adding chlorine to 3 CB is similar to performing an AB. The addition of NaOCl in CB showed an advantage on R_{irr} control from the beginning of the filtration compared to the previous conditions NNNN and NNNY. The variation of R_{irr} fluctuated slight around the overall irreversible fouling trendline with periodic ups and downs in YYYN which may be due to the periodic peak and trough periods of influent flow, secondary effluent quality, and temperature during a day as stated in Chapter 3. The fouling rate was measured to be very low and the fouling layer deposited in a filtration cycle seemed to be fully removed by backwashes. There is no more accumulation of R_{irr} after CBs in a backwash sequence in contrast to condition NNNY, thus the fouling removal efficiency of CB was largely improved by NaOCl addition. Normally, the PES/PVP blended membranes shows superior hydrophilicity, anti-fouling, oxidative and thermal stability as well as good mechanical property [293,294]. But when looking at the condition YYY, although the value of R_{irr} and R_{re} were both under R_m , the variation of R_{irr} was in high volatility compared to YYYN. The R_{irr} sometimes increased very fast, sometimes it was even lower than that in YYYN. Otherwise, the total fouling resistances ($R_{irr} + R_{re}$) in conditions NNNY, YYYN, and YYY were in similar ranges, around $4-7 \times 10^{11} \text{ m}^{-1}$ except the points around CEB in condition NNNY and YYYN. As the CEB is triggered at $TMP \geq 0.3 \text{ bar}$ for 60s, the corresponding total fouling resistances is at $1.41 \times 10^{12} \text{ m}^{-1}$ at 20 °C. The total fouling resistances in conditions NNNN was higher, which resulted in frequent occurrence of CEB.

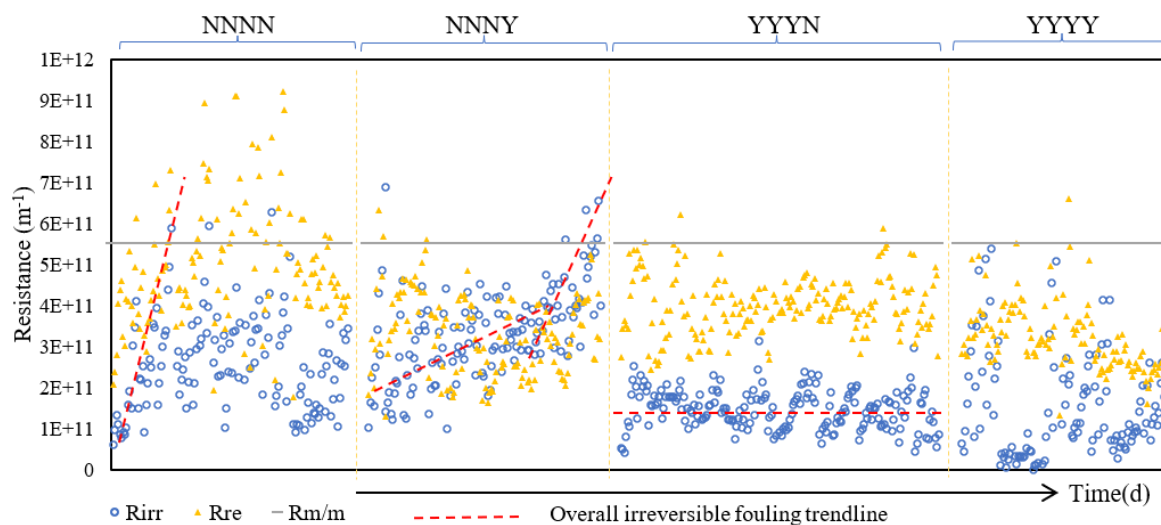


Figure 5-2 Fouling resistance variation versus time in different backwash conditions

5.1.3. Fouling reversibility by physical backwashes

From the previous analysis, the addition of NaOCl in CB and AB improves the backwash cleaning efficiency, but it is not clear to know how far it can be improved. Therefore, it is necessary to compare the fouling reversibility (Eq.(8)) in different backwash conditions. According to the membrane filtration model by Kalboussi *et al.* [295], the mass of the cake layer tends towards a constant value as the number of filtration cycles increase. According to the TMP variation in dead-end filtration with periodic backwashes [21,252], the fouling resistance at the end of each filtration cycle would be in increasing trend with filtration cycles continues, therefore the fouling condition before AB is much closer to that before the 3rd CB. Therefore, the fouling distribution before AB is much closer to that before the 3rd CB. Figure 5-3 gives the reversibility of AB and 3rd CB ranges in different conditions and Table 5-2 shows the comparison between them. It can be confirmed that AB has dominant position in reversibility compared to CB with or without NaOCl, even the addition of 10 mg Cl₂·L⁻¹ NaOCl in CB could not reach the reversibility of AB without NaOCl. Moreover, condition NNNY offered the highest average reversibility of AB (133 ± 16%) and the stable reversibility range (98%-174%), and the second highest was condition YYYY. YYYN is resulted in the lowest AB reversibility. The addition of 10 mg Cl₂·L⁻¹ NaOCl in AB improved the reversibility of AB by 15%-18% in average through comparison in parallel conditions NNNN and NNNY, and in conditions YYYN and YYYY.

However, the addition of 10 mg Cl₂·L⁻¹ NaOCl in CB improved the reversibility of 3rd CB by 5% in average through comparison in parallel conditions NNNN and YYYN, and the 3rd CB reversibility in condition NNNY and YYYY was very close to each other. The lowest reversibility of AB in condition YYYN may be because of the highest reversibility of 3rd CB compared to the other conditions and/or the lack of chlorine. The cleaning performance of CB and AB mutually restricts and influences each other as the reversibility in percentage is related to the previous filtration cycle. From the fouling resistance distribution in section 5.1.2, the R_{irr} was controlled in the lowest ranges in YYYN compared to the others with chlorinated CB and indicated that the membrane was rather clean, therefore, the total foulant cake before AB in YYYN would be less than that in conditions NNNN and NNNY which dealt with both the cake layer formed in one filtration cycle and accumulated foulant after the previous three CBs. Therefore, the cleaning performance by AB in YYYN did not exert its best removal ability compared to that in NNNY condition. On the other hand, although backwash condition YYYN largely improved the fouling management, there was still residual R_{irr} left on membrane after chlorinated CB, but non-chlorinated AB failed to remove these residual foulant which reflected its limitation in particular on R_{irr} removal, while chlorinated AB in condition YYYY could achieve much lower value of R_{irr} if the sensitivity of the membrane to fouling is ignored.

Table 5-2 Reversibility ranges of backwash in different conditions

Conditions	3 rd CB			AB		
	Min	Max	Mean ± SD	Min	Max	Mean ± SD
NNNN	76%	102%	91 ± 7%	92%	140%	118 ± 12%
NNNY	75%	104%	86 ± 8%	98%	174%	133 ± 16%
YYYN	85%	125%	96 ± 7%	81%	135%	107 ± 12%
YYYY	64%	114%	87 ± 14%	85%	165%	125 ± 21%

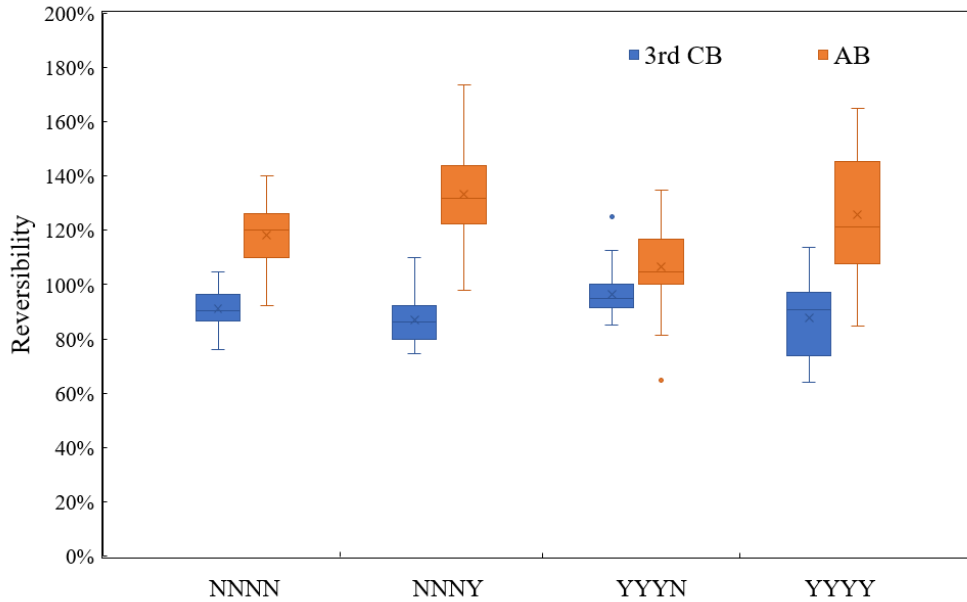


Figure 5-3 Reversibility ranges of AB and 3rd CB in different conditions

To furtherly investigate the impacts of irreversible fouling on cleaning efficiency by physical backwash, the relationship of fouling reversibility versus the R_{irr} on membrane just before the corresponding AB or CB and the dynamic trendlines were shown in Figure 5-4 and Figure 5-5. The cases of CB in Figure 5-5 includes the 1st CB, 2nd CB, and 3rd CB of all backwash sequences. The statistical test showed positive linear regression between AB reversibility and R_{irr} before AB in all conditions (p -value<0.05), and showed significant negative linear regression on CB reversibility and R_{irr} before CB in condition NNNN, YYYN, and YYYY; only the statistical test for condition NNNY ($0,05 < p=0.066$) is not significant. However, the data here were mainly shown to know the variation tendency (increase or decrease not necessarily linearly) of the AB or CB reversibility versus total irreversible fouling resistance before the backwash. Therefore, the significances of both AB and CB under 90% confidential interval can be totally supported.

The reversibility of AB whether with or without NaOCl was shown in positive correlation to R_{irr} from Figure 5-4, while the reversibility of CB with or without NaOCl was shown in negative correlation to R_{irr} from Figure 5-5. It means that the cleaning capacity of AB will be increased with the increase of R_{irr} leading to better filtration performance, but the removal capacity of CB will be decreased with the increase of R_{irr} causing harsh filtration performance. From Figure 5-4, the increasing rates of AB reversibility with R_{irr} in all conditions were similar, the reversibility

of AB under the same R_{irr} was order in YYYY higher than NNNY, higher than NNNN and YYYN. The addition of NaOCl in AB indeed improved its cleaning efficiency as NNNY and YYYY were both in higher reversibility compared to NNNN and NNNY whatever the value of R_{irr} .

From Figure 5-5, the reversibility of CB was decreased with the deposited R_{irr} increase and validated the higher performance of AB in comparison to CB. The slight decline of CB reversibility in conditions NNNN and NNNY was very close to each other and both conditions used non-chlorinated CB for cleaning. The CB of YYYN and YYYY showed higher reversibility with lower R_{irr} deposition, but the CB reversibility decreased faster with R_{irr} increase compared to condition NNNN and NNNY. On the one hand, the chlorinated CB offered higher reversibility that could better control R_{irr} in lower levels. On the other hand, although chlorinated CB was good at R_{irr} controlling, it has poor adaptability and lower reversibility when R_{irr} increased. Looking back to NNNN and NNNY, the cleaning efficiency of non-chlorinated CB was more stable and sustainable with R_{irr} variation. Condition YYYN although had the best fouling resistance management, non-chlorinated AB was resulted in lower reversibility which was much closer to the performance of chlorinated CB. Both conditions YYYN and YYYY showed faster decrease of CB reversibility with R_{irr} increase. From this point, condition NNNY is more recommended as optimized backwash condition which showed relatively higher effective and stable reversibility variation both of AB and CB compared to other conditions.

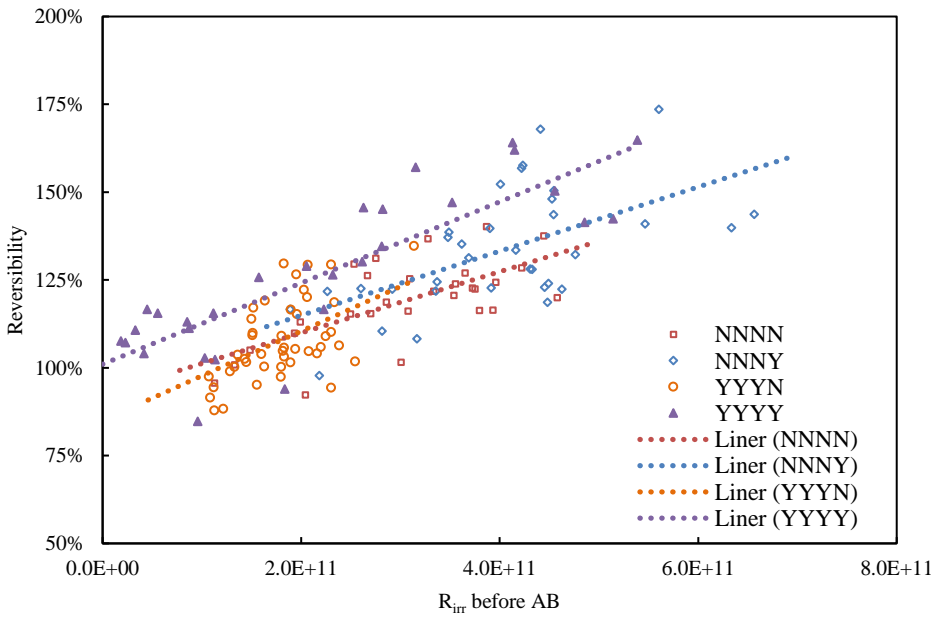


Figure 5-4 Reversibility of AB versus R_{irr} on the membrane just before AB

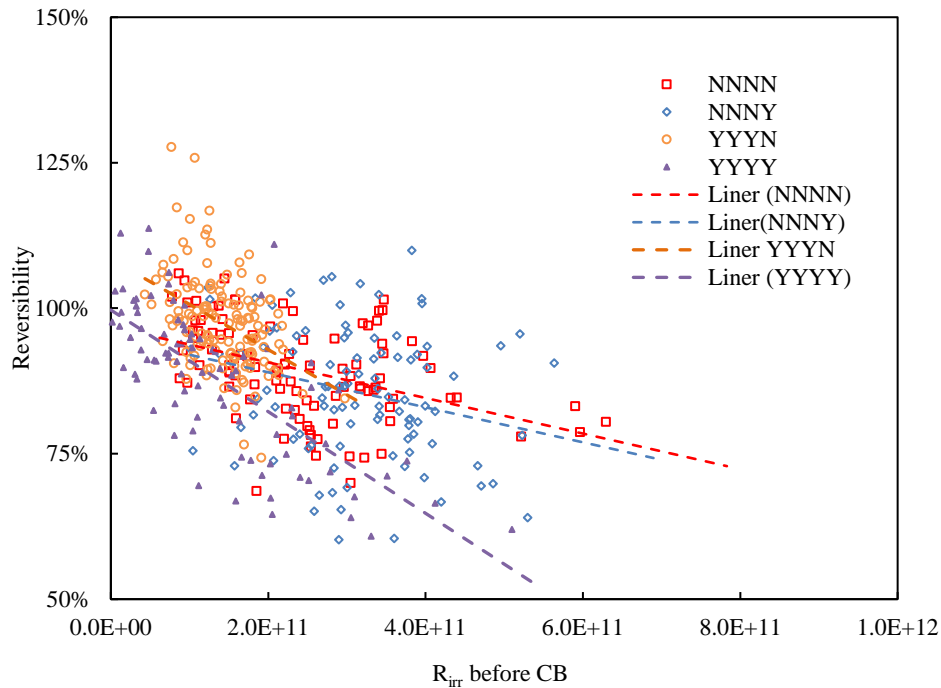


Figure 5-5 Reversibility of CB versus R_{irr} on the membrane just before CB

5.1.4. Turbidity and TOC removal rates by backwash

In general, as the cake layer mass grows in proportion to the concentration of foulants in the feed, and to the total volume of feed passing through the membrane, the cake layer resistance is proportional to the mass accumulated on the membrane surface [295]. Table 5-3 shows the water quality of AB, 3rd CB, UF feed, and UF permeate in the middle of filtration in different conditions and Figure 5-6 (a) and (b) are the turbidity and TOC removal rates by AB and 3rd CB through mass balance in a filtration cycle.

Figure 5-6 (a) shows that the percentage of turbidity recovered by 3rd CB is always below 80% which indicated there would be an increased foulant deposition of this residual turbidity after CB together with the new turbidity accumulation for the next filtration cycle. The turbidity removal capacity by 3rd CB was improved by 15%-30% with 10 mg $Cl_2 \cdot L^{-1}$ NaOCl addition

compared to non-chlorinated 3rd CB. Additionally, the turbidity removal performance of AB was much better than CB, as the lowest removal rate is around 88% averagely. The chlorinated AB in condition NNNY showed the highest, 136% removal of turbidity from concentrate which means the chlorinated AB removed the total turbidity accumulated in its own filtration cycle completely and removed part of the residual turbidity from the previous filtration cycles. The turbidity recovery performances of AB in conditions NNNN, YYYN, and YYYY were very similar, the chlorinated AB in condition YYYY showed no improvement on turbidity removal. The removal efficiency of AB in condition NNNY was the highest compared to the other 3 conditions almost in similar efficiency.

From Figure 5-6 (b), it is obvious to see the high improvement of TOC recovery in conditions with NaOCl compared to NNNN. In condition NNNN, the 3rd CB and AB both without NaOCl showed similar and low removal efficiency on TOC. TOC recovery rate by chlorinated AB was highly improved to more than 200% while by 3rd CB, it was only 50% in condition NNNY. Otherwise, the addition of NaOCl in CB also greatly improved the TOC recovery rates as in conditions YYYN and YYYY. Through comparison, the addition of NaOCl in CB showed higher removal capacity on TOC compared to turbidity removal.

Table 5-3 The Turbidity and TOC in different types of backwash water and UF feed and permeate

Condition	Water types	Turbidity (NTU)	TOC (mgC.L ⁻¹)
All conditions	UF feed	1.9 ± 0.9	6.6 ± 0.4
	UF permeate	0.4 ± 0.3	6.3 ± 0.3
NNNN	AB	20.2 ± 2	9.3 ± 1
	3 rd CB	10.1 ± 2	9.4 ± 0.6
NNNY	AB	30.9 ± 6	16.4 ± 2
	3 rd CB	14.1 ± 4	8.6 ± 0.6
YYYN	AB	20.5 ± 3	18.9 ± 3
	3 rd CB	17.2 ± 2	17.6 ± 2
YYYY	AB	20.2 ± 9	14.7 ± 6
	3 rd CB	17.0 ± 8	16.1 ± 0.4

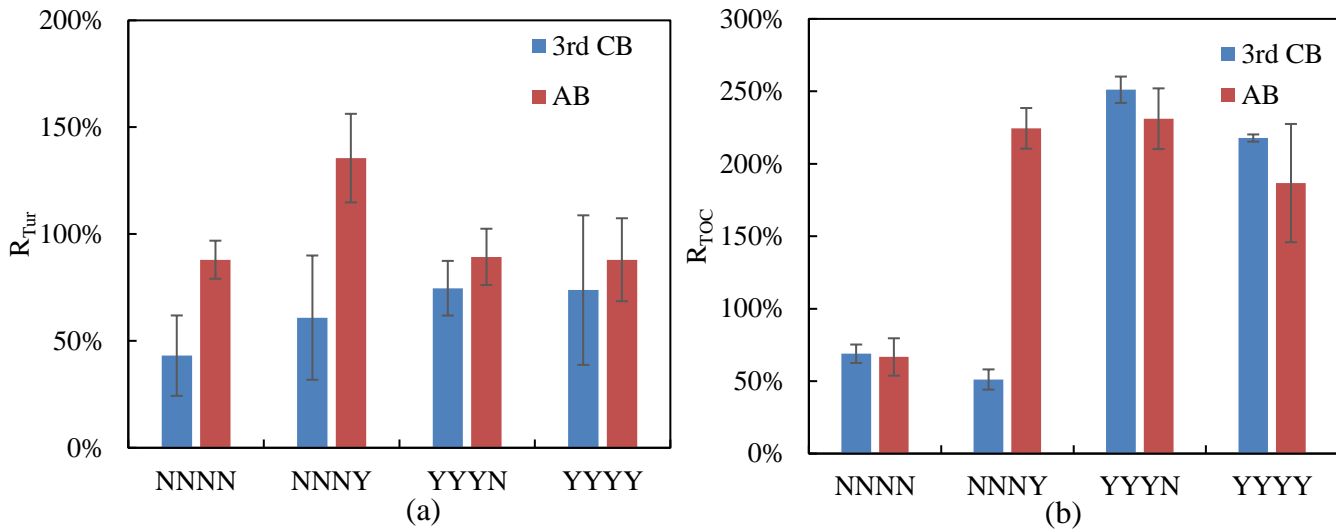


Figure 5-6 (a) Turbidity removal rates of AB and 3rd CB (b) TOC removal rates of AB and 3rd CB

Therefore, for TOC and turbidity removal rates: (i) when no NaOCl is added in CBs (NNNN and NNNY), the AB, either with or without NaOCl, will greatly improve the removal rates thus compensating the low removal efficiency of CB without NaOCl; (ii) when adding NaOCl in CBs (YYYN and YYYY), CB with NaOCl will improve the removal rates previous to AB, thus AB either with or without NaOCl is proportionally less effective compared to CB with NaOCl.

Through comparison, the condition NNNN was not suggested for operation because of the frequent CEB occurrence, fastest R_{irr} increase and poor cleaning efficiency by CB and AB. Condition YYYY was not recommended because of its potential damage on membrane material with most frequent contact with NaOCl, unstable permeability and R_{irr} variation, and unstable cleaning efficiency of CB. YYYN was resulted in the best fouling resistance control and permeability recovery, but the chlorinated CB showed lower reversibility when faced with higher R_{irr} on the membrane. Additionally, the addition of NaOCl in condition YYYN will be higher than NNNY under the same operation period, which would increase the operating costs and may cause faster damage on membrane structures in the long term. Although the control of R_{irr} in condition NNNY was not as good as in condition YYYN, it was in slight and sustainable increase resulted in only 1 CEB occurrence in 6 days. The comparison of total costs depending on the 4 conditions will be discussed at the end of this chapter but condition NNNY seem to be the most

cost-effective backwash condition in this study mainly due to less consumption of NaOCl, sustainable and adaptable filtration performance, and less potential damage on the physicochemical characteristics of membrane. Therefore, the condition NNNY is recommended for long-term operation. The influence of NaOCl concentration on backwash cleaning will be discussed in the next part.

5.1.5. The influence of NaOCl concentration on air backwash efficiency of condition NNNY

As backwash sequence has been optimized in condition NNNY, it is necessary to furtherly investigate the performance of NNNY condition with different concentration of NaOCl addition. The concentration of NaOCl at 5 mg Cl₂·L⁻¹ and 10 mg Cl₂·L⁻¹ in condition NNNY are compared on the filtration performance. The long-term filtration results of 5 mg Cl₂·L⁻¹ NaOCl condition has already been shown in Chapter 3, of which the frequency of CEB occurrence was once in 3 days. In this study, the CEB occurred once in 6 days in NNNY with 10 mg Cl₂·L⁻¹ NaOCl. To be compared, the variation of R_{irr} at the end of each filtration cycle in 5 mg Cl₂·L⁻¹ NaOCl and 10 mg Cl₂·L⁻¹ NaOCl conditions was shown in Figure 5-7. The Y axis R_{irr-end} (n) / R_{irr-end} (1) represents the increasing ration of R_{irr-end} during filtration, and X axis represents the foulant cake mass accumulation during filtration. Within 1000 NTU·min integration, the variation of R_{irr-end} (n) / R_{irr-end} (1) in condition with 5 mg Cl₂·L⁻¹ and 10 mg Cl₂·L⁻¹ of NaOCl was closer to each other, and the ration of R_{irr-end} (n) / R_{irr-end} (1) was all under 5. After this, the R_{irr} increasing rate in 5 mg Cl₂·L⁻¹ NaOCl condition was increased very fast than before 1000 NTU·min integration, while the variation in 10 mg Cl₂·L⁻¹ NaOCl condition was continuously in sustainable and slight increase. The R_{irr} variation in condition NNNY with 5 mg Cl₂·L⁻¹ and 10 mg Cl₂·L⁻¹ NaOCl was indeed from 1.02×10¹¹ m⁻¹ to 9.17×10¹¹ m⁻¹, and from 1.35×10¹¹ m⁻¹ to 6.57×10¹¹ m⁻¹, respectively. The faster increase of R_{irr-end} after 1000 NTU·min integration in 5 mg Cl₂·L⁻¹ NaOCl condition may be due to more complex structure of foulant cake layer and increased local flux due to loss of effective filtration area, as stated in section 5.1.2 of condition NNNY. Higher concentration of NaOCl at 10 mg Cl₂·L⁻¹ increased the chance to inactivate organisms, detach cake foulant, extend the time to form more complex fouling, and improve the load capacity on

feed water quality. Therefore, the increase of NaOCl concentration in condition NNNY indeed improved the AB cleaning efficiency and the overall R_{irr} control during filtration. $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl is more effective on fouling control in condition NNNY.

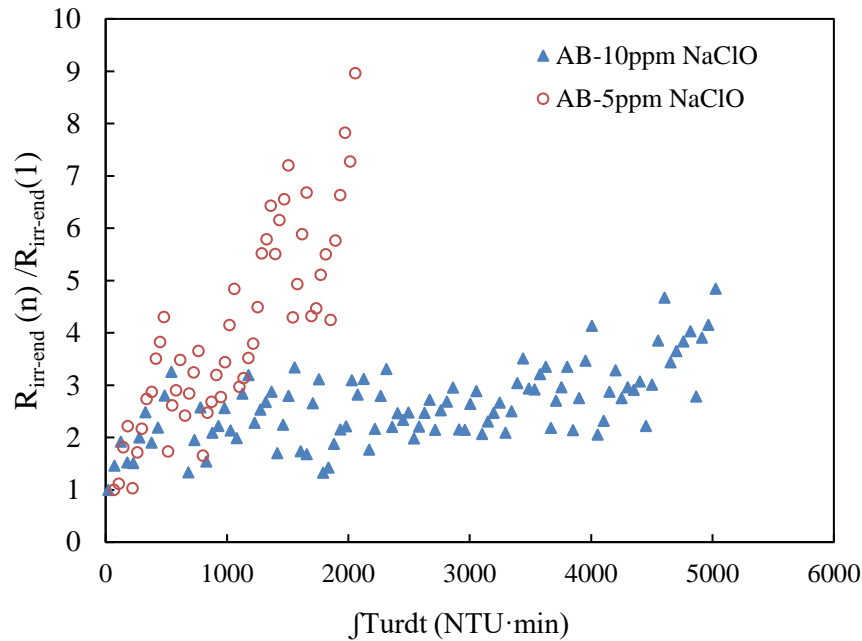


Figure 5-7 Variation of $R_{irr-end}$ of filtration cycle (n) to the initial cycle (1) a function of the integral of turbidity vs. time

5.1.6. Discussion

In this study, $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl addition in backwash water can positively improve the filtration performance of UF pilot plant. The addition of NaOCl had significant improvement on AB cleaning efficiency when comparing condition NNNN and NNNY, or comparing condition YYYN and YYYY, resulting in 15-18% increasing of reversibility. However, NaOCl addition itself almost had fewer improvement on 3rd CB cleaning, with averagely 1-5% increasing of reversibility when comparing condition NNNN and YYYN, or condition NNNY and YYYY. In this case, it seems that air injection contributes to enhance the NaOCl-assisted backwash cleaning. In the process of this study, air injection with strong forces to break the foulant layer on membrane to be less dense thus increase the foulant contact area with backwash water and NaOCl. The cooperation of air injection and NaOCl oxidation on fouling highly improved the cleaning efficiency of backwash. Additionally, comparing the performance of AB and 3rd CB in

all conditions, the reversibility of AB (with or without NaOCl) was completely higher than that of CB (with or without NaOCl). Therefore, it seems that the effects of air injection on CB were more significant than NaOCl assistance in CB water, this can be considered as AB without NaOCl in this study has higher cleaning efficiency than CB with NaOCl, see as in condition YYYN. Based on the comparison on efficiency of chemical cleaning and physical cleaning from Park et al. [296], extending contact time of chemical cleaning was one of the important factors on improvement of membrane permeability recovery compared to physical cleaning. In our study, the contact time of chlorinated backwash water through membrane is 52s in total. The disinfection and oxidation reactions between NaOCl and cake foulants cannot be completely reacted in shorter contact time, thus the cleaning efficiency of chlorinated CB was limited in this study. In contrast, the physical assistance by air injection could greatly improve the cleaning efficiency. As from Guigui et al. [266], air injection even at very low air velocity ($0.08 \text{ m}\cdot\text{s}^{-1}$) could result in a high backwash efficiency. And normally, the duration of air injection does not need too long, less than 2 min is enough. Based on above analysis, air injection assistance on CB (AB) indeed provides higher contributions on fouling removal than $10 \text{ mg Cl}_2\cdot\text{L}^{-1}$ NaOCl. However, the comparison conditions of AB and NaOCl assisted CB in this study could be completed with consideration of all impact factors. The impact factors include air injection duration, air velocity, NaOCl concentration in backwash water, temperature, and NaOCl contact time with membrane. Additionally, the co-effects among these factors are complex and could be investigated in the future.

5.1.7. Conclusion

It is the first time that a comparison between air injection and/or NaOCl addition was realised in semi-industrial conditions. In conclusion, $10 \text{ mg Cl}_2\cdot\text{L}^{-1}$ NaOCl addition in backwash water is confirmed to greatly improve the overall filtration performance and backwash cleaning efficiency based on the analysis on filtration permeability, fouling resistance distribution, backwash reversibility, and cake layer removal ability. During experiment, the chronological order of conditions is NNNN, NNNY, YYYN, and finally YYYY. The type of backwash among all conditions is 3 CBs followed with 1 AB. N represents a backwash either CB or AB without NaOCl and Y represents a backwash either CB or AB with NaOCl. First, condition NNNY stands out from the others due to better control of R_{irr} , less NaOCl consumption in 10 years prediction,

sustainable and adaptable filtration performance, and less potential damage on the physicochemical properties of membrane. The secondary best condition is YYYN which resulting in the best fouling control and permeability recovery, but it has higher potential to damage membrane structures under frequent NaOCl addition. Second, in all conditions, AB showed positive correlation of reversibility and R_{irr} increase, but CB showed negative correlation of reversibility and R_{irr} increase. Besides, CB even added with $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl hardly exceeded the reversibility of AB without chlorine. These results reflect that AB itself whether with or without NaOCl has high cleaning efficiency on fouling removal, especially better on R_{irr} control. Through comparison of $5 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ and $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl in condition NNNY, higher NaOCl concentration increased the chance to inactivate organisms, detach cake foulant, extend the time to more complex fouling formation, and improve the load capacity on feed water quality. Therefore, condition NNNY with $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl was mostly recommended for long term operation in this study on secondary effluent treatment. To further assess the applicability of the process, the total costs of semi-industrial scale UF for water reuse under different operating conditions will be assessed in the next section.

5.2. Capital expenditure and operational expenditure of semi-industrial scale UF for water reuse

5.2.1. Economic cost in operating condition J60t60BW1/3

Based on results of chapter 3, the condition J60t60BW1/3 with $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl addition in AB water is recommended for the future treatment of secondary effluent of WWTP. Before production and application in reality, it is necessary to estimate the economic budget of the project. The calculations with $10 \text{ mg Cl}_2 \cdot \text{L}^{-1}$ NaOCl addition as recommended in the previous section will be given in section 5.2.3. The daily volume of wastewater to the WWTP is $3200 \text{ m}^3 \cdot \text{d}^{-1}$ and the average flow rate is $133 \text{ m}^3 \cdot \text{h}^{-1}$. The flow rate of the pilot plant is $540 \text{ L} \cdot \text{h}^{-1}$. The calculation of expenditure in this project is based on the calculation by Guilbaud et al. [297] who

estimated the costs of UF for another application and for the size of the pilot plant used. Operating Expenses (OPEX) is the day-to-day cost for running a product, business, or system. Costs related to replacing consumables (e.g., ultrafiltration membranes), consumption of chemicals and/or energy, the maintenance, and the labor were taken into account for the OPEX in this study. UF pilot plant and UF module lifespan are estimated at 10 years, but modules are considered to be changed every 5 years to ensure the good condition of the membranes. Capital expenditure (CAPEX) is determined as the depreciation of the fixed assets, including the cost of the investment and the UF modules. The following budget calculation was firstly based on 1 pilot plant and 1 UF membrane module under the optimized condition J60t60BW1/3 with 5 mg Cl₂·L⁻¹ NaOCl addition in AB water. The flow rate for one UF membrane module is 540L·h⁻¹. The equation of CAPEX can be translated as follows:

$$\text{CAPEX} = \$ (\text{pilot plant}) + \$ (\text{UF module})$$

Table 5-4 shows the related factors and costs of CAPEX. In industrial production, a set of spare membrane module needs to be bought in case of special conditions occurrences. The final CAPEX of the project is 2940 €·yr⁻¹.

Table 5-4 Calculation of CAPEX

UF Membrane	Values
Lifespan of UF	10 years
Change duration	5 years
Unit price of UF module	2200 €
Number of module (include spare module)	2
UF investment total cost before change	4400 €
\$ (UF module)	440 €·yr ⁻¹
Pilot plant	
Lifespan	10 years
Pilot plant price	25000 €
\$ (pilot plant)	2500 €·yr ⁻¹
CAPEX	2940 €·yr⁻¹

Based on the definition, the equation of OPEX can be translated as follows:

$$\text{OPEX} = \$ (\text{UF module replacement}) + \$ (\text{chemical consumption}) + \$ (\text{energy consumption}) + \$ (\text{maintenance} + \text{labor})$$

To calculate maintenance and labor costs, a value equivalent to 10% of the fixed capital expenditure was considered [297,298]. The cost for UF module replacement [$\$$ (UF module replacement)] is calculated the same as $\$$ (UF module) in CAPEX as shown in Table 5-4. The process to generate OPEX is mainly focused on CEB cleaning, AB, CB, filtration process, and pumps. Table 5-5-Table 5-9 listed the practical information of the above processes for energy and chemical cost calculation. The calculation process and method were presented in the remarks of Tables, but the unit conversion was not included. The chemical consumption includes the use of 14% NaOCl solution, 30% NaOH solution and 50% H₂SO₄ solution. The consumption of 30% NaOH solution and 50% H₂SO₄ solution was related to the quantity of chemicals used in CEB. The consumption of NaOCl was calculated at the ideal state with total usage in CEB (50 mg Cl₂·L⁻¹) and air backwash (5 mg Cl₂·L⁻¹). The potential consumption of NaOCl was not taken into account which includes the complex reaction with ammonia or ammonium compounds in permeate, self-decomposition and chlorine gas volatilization, and usage in manual cleaning of equipment. The consumption of energy was mainly from pumps and air compressor used in cleaning processes, filtration process, and chemical injection process. The consumption of energy all was converted into cost of electricity. In conclusion, the pilot plant in condition J60t60BW1/3 with 5 mg Cl₂·L⁻¹ NaOCl addition in air backwash was resulted in CAPEX of 2940 €·yr⁻¹ and OPEX of 3324 €·yr⁻¹. The net price of unit production in this study is 1.39 €·m⁻³ to implement UF as tertiary treatment in semi-industrial scale for water reuse. It is clear that this cost would be much lower at industrial scale thanks to economy of scale.

Table 5-5 Calculation of energy and chemical costs of CEB

Codename	CEB Parameters	Values	Remarks
A0	Frequency	Once every 3 days	
A1	Number of CEB in 1 year	122 yr ⁻¹	B=365 d·yr ⁻¹ /3d
A2	Permeate consumption of each CEB	250 L	Set value
A3	Soda+chlorine inject duration in CEB	160 s	Set value
A4	Acid inject duration in CEB	160 s	Set value
A5	Backwash duration without air injection	128 s	Set value

A6	soaking time in CEB	2400 s	Set value
A7	Relax duration in CEB	480 s	Set value
A8	Duration of each cleaning	3328 s	$A8=A3+A4+A5+A6+A7$
A9	Power of pump (P3) for backwash, chemical injection in CEB at $2.5\text{m}^3\cdot\text{h}^{-1}$	0.83 kW	Product parameter
A10	Working duration of P3 for CEB per year	$15.14\text{ h}\cdot\text{yr}^{-1}$	$A12=(A3+A4+A5)\times A1$
A11	Unit cost of energy	$0.182\text{ €}\cdot(\text{kWh})^{-1}$	
A12	C_{NaOH} for soaking in CEB	$800\text{ mg}\cdot\text{L}^{-1}$	Set value
A13	C_{NaOCl} for soaking in CEB	$50\text{ mg Cl}_2\cdot\text{L}^{-1}$	Set value
A14	$C_{\text{H}_2\text{SO}_4}$ for soaking in CEB	$1000\text{ mg}\cdot\text{L}^{-1}$	Set value
A15	Permeate volume for dosing chemicals	60 L	
A16	Unit price of 30% (w/w) NaOH solution	$50.2\text{ €}\cdot\text{L}^{-1}$	
A17	Unit price of 14% (w/v) NaOCl solution	$11.1\text{ €}\cdot\text{L}^{-1}$	
A18	Unit price of 50% (v/v) H_2SO_4 solution	$20\text{ €}\cdot\text{L}^{-1}$	
A19	Concentration of 30% (w/w) NaOH solution	$300\,000\text{ mg}\cdot\text{L}^{-1}$	
A20	Concentration of 14% (w/v) NaOCl solution	$140\,000\text{ mg Cl}_2\cdot\text{L}^{-1}$	
A21	Concentration of 50% (v/v) H_2SO_4 solution	$640\,000\text{ mg}\cdot\text{L}^{-1}$	
A22	Cost of 30% NaOH solution	$980\text{ €}\cdot\text{yr}^{-1}$	$A22=A12\times A15/A19\times A1\times A16$
A23	Cost of 14% NaOCl solution	$29\text{ €}\cdot\text{yr}^{-1}$	$A23=A13\times A15/A20\times A1\times A17$
A24	Cost of 50% H_2SO_4 solution	$229\text{ €}\cdot\text{yr}^{-1}$	$A24=A14\times A15/A21\times A1\times A18$
A25	Pump for relaxation (P2) power at $540\text{ L}\cdot\text{h}^{-1}$	0.55kW	
A26	Working duration of P2 for CEB per year	16.3h	$A26=A7\times A1$
$\$_{\text{E-CEB}}$	$\$$ (Energy consumption in CEB)	$2.4\text{ €}\cdot\text{yr}^{-1}$	$\$_{\text{E-CEB}} = A11\times A10\times A9$ $A25\times A26\times A11$
$\$_{\text{C-CEB}}$	$\$$ (Chemical consumption in CEB)	$1238\text{ €}\cdot\text{yr}^{-1}$	$\$_{\text{C-CEB}} = A22+A23+A24$

Table 5-6 Calculation of energy and chemical costs of CB

Codename	CB Parameters	Values	Remarks
B0	Frequency	3 times every 4h	
B1	Number of cleanings in 1 year	6510 yr ⁻¹	
B2	Duration of cleaning	64 s	Set value
B3	Permeate consumption of each cleaning	36 L	Set value
B4	Pump power for CB (P3) at 2.5m ³ ·h ⁻¹	0.83 kW	
B5	Working duration of P3 for CB per year	116 h·yr ⁻¹	B5=B1×B2
\$ _{E-CB}	\$(Energy consumption in CB)	17 €·yr ⁻¹	\$ _{E-CB} = B5×B4×A11
\$ _{C-CB}	\$(Energy consumption in CB)	0	

Table 5-7 Calculation of energy and chemical costs of AB

Codename	AB Parameters	Values	Remarks
C0	Frequency	Once every 4h	
C1	Number of cleanings in 1 year	2130 yr ⁻¹	
C2	Duration of cleaning	184 s	
C3	Air injection duration in AB	120 s	
C4	Backwash duration without air injection	64 s	
C5	Water consumption of each cleaning	36 L	
C6	Air compressor power	1.5 kW	
C7	Air compressor working time for AB	71 h·yr ⁻¹	C7=C3×C1
C8	Pump power for AB (P3) at 2.5m ³ ·h ⁻¹	0.83 kW	
C9	Working duration of P3 for AB per year	38 h·yr ⁻¹	C9=C4×C1
C10	C _{NaOCl} for AB	5 mg Cl ₂ ·L ⁻¹	
C11	Permeate volume with NaOCl addition	100 L	
C12	Volume of 14% (w/v) NaOCl solution	7.6 L·yr ⁻¹	C12=C10×C11×C1/A21
\$ _{E-AB}	\$(Energy consumption in AB)	26 €·yr ⁻¹	\$ _{E-AB} = (C6×C7+C8×C9) ×A11
\$ _{C-AB}	\$(Chemical consumption in AB)	85 €·yr ⁻¹	\$ _{C-AB} = C12×A18

Table 5-8 Calculation of energy and chemical costs of filtration

Codename	Parameters	Values	Remarks
D0	Number of P1	1	
D1	Number of P2	1	
D2	Filtration flow rate	540 L·h ⁻¹	Constant
D3	Filtration time in one cycle	1 h	Set value
D4	Total filtration duration in a year	8423 h·yr ⁻¹	D3=8760h·yr ⁻¹ -A1×A8-B1×B2-C1×C2
D5	Filtration cycles in a year	8423 yr ⁻¹	D5=D4/D3
D6	Flush numbers in one filtration cycle	1	
D7	Flush duration in one filtration cycle	15 s	Set value
D8	Flush flow rate	2.5 m ³ ·h ⁻¹	
D9	Feed water volume for flush in a year	88 m ³	D9=D8×D7×D5
D10	Working flow of feed pump (P1)	2.5 m ³ ·h ⁻¹	
D11	Workload of P1 per year	4618 m ³	D11=D2×(D3-D7) ×D5+D9
D12	Working time of P1 per year	1847 h·yr ⁻¹	D12=D11/D10
D13	P1 (feed pump) power	1.00 kW	
D14	Working time of P2 for flush per year	35 h·yr ⁻¹	D14=D7×D5
D15	Working time of P2 for filtration per year	8388 h·yr ⁻¹	D15=D4-D14
D16	P2 power at 540 L·h ⁻¹	0.55 kW	Manual
D17	P2 power at 2.5 m ³ ·h ⁻¹	0.83 kW	Manual
D18	Energy consumption of P1 per year	336 €·yr ⁻¹	D18=D12×D13×A11
D19	Energy consumption of P2 per year	845 €·yr ⁻¹	D19=(D16×D15+D17×D14) ×A11
D20	Air compressor working frequency for valves	Once every 10 min	Experience value
D21	Air compressor working time each time for valves	10s	Experience value
D22	Energy consumption of air compressor for valves	40 €·yr ⁻¹	D22=D21×6×8760h·yr ⁻¹ ×C6×A11
D23	Net Permeate production per year	4188 m ³ ·yr ⁻¹	D23=D2×D15-A1×A2-B1×B3-C1×C5
\$ _{E-filtration}	\$(Energy consumption of filtration)	1221 €·yr ⁻¹	\$ _{E-filtration} = D18+D19+D22

Table 5-9 Energy consumption of Chemical pumps

Codename	Parameters	Values	Remarks
E0	Soda pump (P7) working time	5.41 h·yr ⁻¹	E0=A3×A1
E1	Chlorine pump (P8) working time	5.41 h·yr ⁻¹	E1=A3×A1
E2	Acid pump working time	5.41 h·yr ⁻¹	E2=A4×A1
E3	Chlorine pump (P4) working time	60 h·yr ⁻¹	Estimated value
E4	All chemical pumps power	0.03 kW	
$\$_{E-cp}$	\$(Energy consumption of chemical pumps)	0.5 €·yr ⁻¹	E5=E4×(E0+E1+E2+E3) ×A11

Table 5-10 Economic cost

Type	Cost	Remarks
\$ (UF module replacement)	440 €·yr ⁻¹	Refer to Table 5-4
\$ (chemical consumption)	1323 €·yr ⁻¹	$\$ = \$_{C-CEB} + \$_{C-AB}$
\$ (energy consumption)	1267 €·yr ⁻¹	$\$ = \$_{E-CEB} + \$_{E-CB} + \$_{E-AB} + \$_{E-Filtration} + \$_{E-cp}$
\$ (maintenance + labor)	294 €·yr ⁻¹	$\$ = 10\% \times \text{Depreciation(CAPEX)}$
OPEX	3324 €·yr ⁻¹	
CAPEX	2940 €·yr ⁻¹	
Total price	5824 €·yr⁻¹	Total price=OPEX+CAPEX-\$ (UF module replacement)
Unit price of permeate production	1.39 €·m⁻³	Unit price of production=Total price/D23

5.2.2. Impact of different NaOCl addition on economic cost

In J60t60BW1/3, based on the performance of NaOCl addition conditions NNNN, NNNY, YYYN, and YYYY, the main difference on economic cost is the chemical cost due to the frequency of CEB occurrence and NaOCl addition frequency in physical backwashes changes. Besides, the concentration of NaOCl was 10 mg Cl₂·L⁻¹ added in the backwash water in these

conditions. Table 5-11 recalls the cost of each condition through applying the changes into the formula from Table 5-4 to Table 5-10 in section 5.2.1. Figure 5-8 shows the components of CAPEX and OPEX. The CAPEX of all the conditions is the same as they use the same pilot plant and UF membrane module. The OPEX of condition NNNN will spend the most because the frequent CEB will consume more chemical cost although there is no addition of NaOCl in backwashes. Condition NNNY resulted in the most economical OPEX and the lowest cost of unit product compared to the others. The OPEX of condition NNNY is 2917.3 €·yr⁻¹ and the unit price of permeate is 1.28 €·m⁻³. This unit price of permeate is very important in comparison with the production cost of drinking water by UF which is around 0.5 €·m⁻³. The difference is due to the smaller size of the plant in this study, the cost of the plant (include the module) represents more than 58% of the unit price. Moreover, due to the quality of the feed water (from WWTP), the cost of chemical is also important to represent around 23%. In the same condition J60t60BW1/3 and NNNY, the OPEX and unit cost of product of condition with 10 mg Cl₂·L⁻¹ NaOCl addition in backwash water is finally lower than that with 5 mg Cl₂·L⁻¹ NaOCl addition. In view of economic cost, condition J60t60BW1/3 with 10 mg Cl₂·L⁻¹ NaOCl addition in AB (NNNY) is recommended for the most cost-effective and optimized condition in long-term operation.

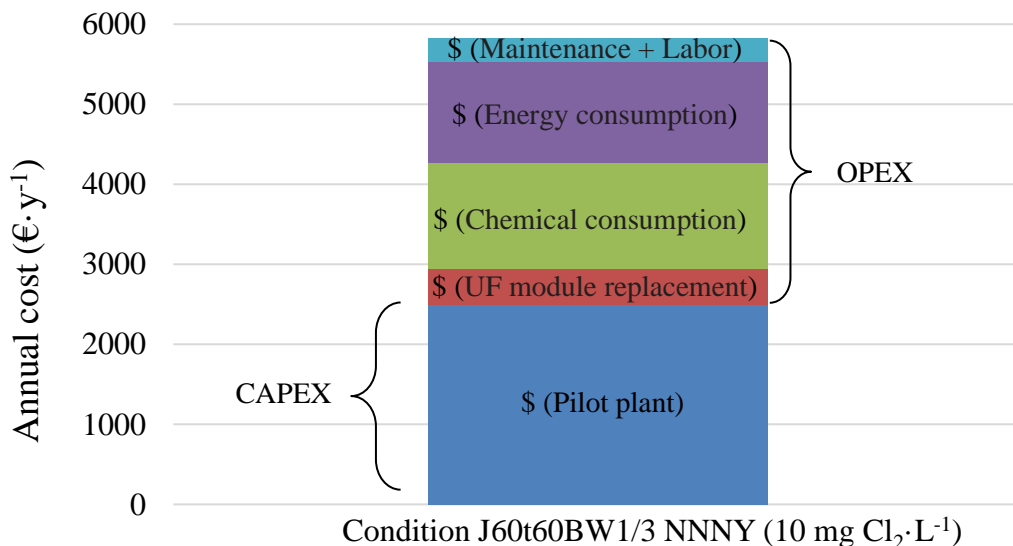


Figure 5-8 Cumulative costs for CAPEX and OPEX

Table 5-11 Expenditure cost on different backwash conditions

	NNNN	NNNY	YYYN	YYYY	
CEB frequency	Once per day	Once in 5 days	Once in 3 days	Once in 6 days	Once in 5 days
Number of CEB in a year (yr ⁻¹)	365	73	122	61	73
Number of AB in a year (yr ⁻¹)	2008	2154	6510	2160	2154
Number of CB in a year (yr ⁻¹)	6388	6534	2130	6540	6534
NaOCl concentration in backwash water (mg Cl₂·L⁻¹)	0	10	5	10	10
\$ _{E-CEB} (€·yr ⁻¹)	5.8	1.3	2.4	1.1	1.3
\$ _{C-CEB} (€·yr ⁻¹)	3703	741	1238	619	741
\$ _{E-CB} (€·yr ⁻¹)	17	18	17	18	18
\$ _{C-CB} (€·yr ⁻¹)	0	0	0	678	811
\$ _{E-AB} (€·yr ⁻¹)	24	26	26	26	26
\$ _{C-AB} (€·yr ⁻¹)	0	171	85	0	171
\$ _{E-filtration} (€·yr ⁻¹)	1200	1226	1221	1227	1227
\$ _{E-cp} (€·yr ⁻¹)	1.5	2.1	0.5	2	2.1
\$ (chemical consumption) (€·yr ⁻¹)	3703	912	1323	1297	1723
\$ (energy consumption) (€·yr ⁻¹)	1246.8	1271.3	1267	1272.1	1272.3
CAPEX (€·yr ⁻¹)	2940	2940	2940	2940	2940
OPEX (€·yr ⁻¹)	5683.8	2917.3	3324	3303.1	3729.3
Net production (m ³)	4023	4221	4188	4230	4222
Unit price of product (€·m⁻³)	2.03	1.28	1.39	1.37	1.48

Conclusions and Perspectives

The objective of this thesis was to investigate the feasibility of the UF technology as tertiary treatment for municipal wastewater reuse on non-potable purposes, such as pipe washing, street cleaning, underground water recharge, etc. Meanwhile, the thesis aimed to investigate the influence of filtration parameters such as flux, filtration time, backwash sequence including classical and air backwashes with or without NaOCl on UF membrane filtration performances and fouling management, in order to find out the optimized filtration conditions of the semi-industrial UF pilot system and provide guidance for future industrial application. The assessment of optimum conditions is mainly based on comparison of the permeate quality, water recovery rate, backwash efficiency, fouling control, and capital/operational expenditure. The conclusions are listed as follows.

Selection of UF system on wastewater treatment for reuse purpose. After a large state of the art, ultrafiltration has been successfully applied on industrial scale and wastewater reuse cases around the world. The UF membranes enable high removal efficiency on total suspended solids, turbidity, microorganisms, and partial removal efficiency on organic matters. Generally, UF membranes provide higher removal capacity on contaminants and qualified permeate production compared to MF membranes. UF membranes are less sensitive to fouling formation especially irreversible fouling compared to NF and RO, together with lower requirement on feed water quality, transmembrane pressure, installation and commissioning. Currently, UF process is considered as one of the most cost-effective technology to deal with secondary effluent on non-potable reuses. Therefore, UF system is suggested in this thesis and a semi-industrial UF pilot plant was installed after the secondary effluent of a French WWTP.

Selection of UF membrane. The filtration performances of three UF membranes were compared in lab-scale. The 200 kDa hollow fiber membrane stands out from the others with a high initial permeability at $960 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}\cdot\text{bar}^{-1}$, the lowest fouling increasing rate during filtration, and larger membrane specific areas due to the geometry. Thus, the 200 kDa hollow fiber is considered to be the most adequate membrane for semi-industrial pilot scale and more especially after the settling tank (clarifier) directly without pretreatment.

Qualified water quality for reuse. The WWTP with UF treatment process provided significant removal capacity on turbidity, organic matters, microorganisms, and microplastics. The UF system provided higher than 5 log removal rates of *E.coli* and *Enterococci* compared to

the raw wastewater, and the spores of anaerobic sulphito-reducers and virus in permeate were removed to be under the detection limits of analytical methods ($< 1 \text{ CFU} \cdot 100\text{mL}^{-1}$ for bacteria or $< 1 \text{ PFP} \cdot 100\text{mL}^{-1}$ for virus). Considering the international Covid-19 pandemic, the UF permeate was free from infectious SARS-CoV-2. Concerning microplastic particles, the UF membrane provided $86.0 \pm 0.6 \%$ removal of microplastics in quantity and $97 \pm 2 \%$ removal in filter surface area from UF feed. However, although the UF system showed effective removal capacities of microplastics, the microplastics' fragmentation during each treatment process seems hard to avoid and needs to be furtherly investigated. Finally, the UF permeate quality was confirmed to be good enough to be reused in non-potable applications as it met reuse guidelines of the WHO, reuse standards of France and the most recent EU regulation for agricultural irrigation, all with the highest quality level "A". This works confirms the great interest of UF as tertiary treatment of conventional WWTP for water reuse for different non potable application.

Effects of flux and filtration time on UF system hydraulic filtration performance. 15 filtration conditions were conducted for investigation of flux and filtration time, and all conditions were shown in constant and stable variation of permeability for 40 hours. While filtrating with real secondary effluent from WWTP, the overall permeability decreased and stabilized to a smaller value with an increased flux and/or filtration time. In constant flux mode, the increase of flux (related to a higher TMP on membrane) and the increase of filtration time both lead to more potential foulants brought onto the membrane. Based on the filtration performances, these conditions were classified into 3 types: soft conditions with lower flux or frequent backwash (short filtration time) resulting in too little productivity and which were discarded. Harsh conditions with higher flux or longer filtration time were also discarded because of high occurrence of CEB which led to difficult permeability stabilization and high consumption of permeate and chemicals. Finally, standard conditions J80t40BW1/3, J60t60BW1/4, and J60t60BW1/3 stood out from the others with higher overall performances based on the comparison of AB and CB reversibility, irreversible fouling resistance increase, occurrence of CEBs and water recovery rates. The optimized conditions resulted in higher than 90% water recovery rates, the fouling reversibility by AB and CB were in ranges of 135%-145% and 78%-84%, respectively. The fouling resistances were better controlled in slower increase compared to the other conditions resulting in less CEB. Of course, these conditions are function of the

WWTP and feed water quality, but for the first time, this study defined some ranges of operating conditions for future water managers and engineers for municipal wastewater tertiary treatment before reuse.

Effects of Air backwash frequency on filtration performance. The air injection process in AB greatly increased the cake layer removal efficiency compared to CB. Besides, 5ppm NaOCl addition in backwash water of AB of all condition furtherly improved the cleaning efficiency of AB due to the function of disinfection and oxidation. Therefore, AB showed higher reversibility efficiency than CB, which was around 1.25~ 2 times the reversibility of CB in average. By analysing the influence of AB frequency (1/3, 1/4, 1/6, 1/9, and no AB) on irreversible fouling management, the irreversible fouling resistance increased faster with the decrease of AB frequency during operation. AB frequencies of BW1/3 and BW1/4 under constant flux of $60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and t_c of 60 min showed better permeability recovery and less CEB occurrence during filtration, thus were recommended for long-term filtration test.

Sustainable and long-term filtration investigation. After optimization of the filtration conditions, the condition J60t60BW1/3 was operated for more than 20 days both in winter and in summer. The occurrences of CEB were all less than once per day which meets sustainable objectives. In summer, feed water quality became worse compared to that in winter with more bacteria, algae, and insects contributing to higher fouling potential, meanwhile, the higher temperature in summer improved the cleaning effects of CEB, CB, and AB which in turn maintained the filtration performance in sustainable permeability recovery and reasonable CEB frequency. The results confirmed that the UF pilot plant could provide sustainable and adaptable filtration performance regardless of the temperature whatever the feed water quality.

Impact of NaOCl -assisted backwashes on membrane fouling management. In this subject, 4 conditions in chronological order are NNNN, NNNY, YYYN, and finally YYYYY. N (No) represents a backwash either CB or AB without NaOCl and Y (Yes) represents a backwash either CB or AB with NaOCl . The other parameters for all conditions are 3 CBs followed with 1 AB, under constant flux of $60 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and t_c of 60 min. The addition of NaOCl in backwash water either for AB or CB is confirmed to greatly enhance the backwash cleaning efficiency thus improving the hydraulic filtration performance according to analysis on filtration permeability, fouling resistance distribution, backwash reversibility, and cake layer removal ability. In all

conditions, the estimated consumption of NaOCl in 10 years were lower than the amount that causes the damage on membrane materials and structure. Condition NNNY stands out from the others due to better control of R_{irr} , less NaOCl consumption, sustainable and adaptable filtration performance, and less potential damage on the physicochemical properties of membrane. The secondary best condition is YYYN which resulting in the best fouling control and permeability recovery, but it has higher potential to damage membrane structures under frequent NaOCl addition. Second, in all conditions, there was a positive correlation between fouling reversibility by AB and R_{irr} increase, and a negative correlation between fouling reversibility by CB and R_{irr} increase. Besides, CB with NaOCl hardly exceeded the reversibility of AB without NaOCl . These results reflect that AB itself whether with or without NaOCl has excellent cleaning efficiency on fouling removal, especially to control the increase of R_{irr} . Through comparison of 5 ppm and 10 ppm NaOCl in condition NNNY, higher NaOCl concentration increased the chance to inactivate organisms, detach cake foulant, extend the time to more complex fouling formation, and improve the load capacity on feed water quality. Therefore, condition NNNY with 10 ppm NaOCl was mostly recommended for future operation.

Estimation of CAPEX and OPEX on UF pilot plant. To furtherly assess the applicability of the process, the total costs of semi-industrial scale UF for water reuse were estimated. OPEX is the day-to-day cost, including the cost of replacing consumables (e.g., ultrafiltration membranes), chemicals and energy, maintenance, and labour. CAPEX is determined as the depreciation of the fixed assets, including the cost of the investment and the UF modules. In result, the pilot plant in condition J60t60BW1/3 with 5 ppm NaOCl addition in air backwash was resulted in CAPEX of 2940 $\text{€}\cdot\text{yr}^{-1}$ and OPEX of 3324 $\text{€}\cdot\text{yr}^{-1}$. The net price of unit production is 1.39 $\text{€}\cdot\text{m}^{-3}$ to implement UF as tertiary treatment in semi-industrial scale for water reuse. In backwash conditions NNNN, NNNY, YYYN, and YYYY, the OPEX of condition NNNY is the lowest which is 2917.3 $\text{€}\cdot\text{yr}^{-1}$ and the unit price of permeate is 1.28 $\text{€}\cdot\text{m}^{-3}$. In condition J60t60BW1/3 under NNNY, the OPEX and unit cost of product of condition with 10 ppm NaOCl addition in backwash water is finally lower than that with 5 ppm NaOCl addition. In view of economic cost, condition J60t60BW1/3 with 10 ppm NaOCl addition in AB (NNNY) is recommended for future operations. However, among all conditions, the unit price of permeate is very important in comparison with the production cost of drinking water by UF which is lower than 0.5 $\text{€}\cdot\text{m}^{-3}$ but

in agreement with the state of the art. The difference is due to the smaller size of the plant in this study, the cost of the plant (including the module) represents more than 58% of the total price. Moreover, due to the quality of the feed water (from WWTP), the cost of chemicals is also high and represents around 23%. It is clear that this cost would be much lower at industrial scale thanks to economy of scale.

Perspectives. From the end of the 20th century, the benefits of promoting water reuse from wastewater to supplement water resources have been recognized by most countries and regions. Although many treatment processes have been reported to achieve wastewater reuse standard or guidelines, the water reuse projects presently implemented are still very few compared to the large quantity of wastewater discharged all over the world. The total volume of water reuse is approximately 14.2 billion m³·y⁻¹ worldwide reported in Global Water Market 2017, which is less than 4% of the total volume of domestic wastewater. However, the ultimate decision to promote wastewater reuse depends on economic support, government regulations and policies, public acceptance, security and requirements for reliable water supply in local conditions. The wastewater reuse guidelines are not unified in the world even for the same water reuse purpose. Therefore, continuous improvement in the existing reuse standards and guidelines by revising/expanding is necessary. Control and monitoring during reuse applications along with the public feedback is also necessary for safety assessment. It is also worth noting that cultural and religious differences guide public opinion and acceptance of wastewater reuse. In addition, some developing countries still implement the direct use of treated wastewater without accordance with standards and associated risks. In this case, it is suggested to enact wastewater quality standards step-by-step, over suitable periods and according to treatment capabilities, for different countries and regions.

Membrane fouling and its consequences in terms of operating costs and plant maintenance remains the critical limiting factors affecting the widespread application of UF for wastewater treatment. Backwash is the commonly used and effective methods for membrane fouling control. Compared to CEB, the conduction of backwash is fast, short, periodic, and leads to low damage to membranes. Although intensive efforts have been dedicated to the study on membrane fouling mechanisms and control, it is still necessary to develop more effective and easier cleaning methods to control and minimize membrane fouling especially in full-scale applications. The

assisted process for backwash such as air injection or chemical assistance are both confirmed effective for improvement of backwash efficiency. However, the comparison on effectiveness of air backwash and chemical-assisted backwash and the influencing factors between them has not been studied yet. Normally, air injection even at very low air velocity (0.08 m/s) could result in a high backwash efficiency within shorter contact time[266]. Although chemical-assisted backwash, such as NaOCl -assisted backwash provides higher removal efficiency compared to backwash, extending contact time is an important factor for cleaning improvement compared to physical cleaning according to from Park et al[299]. It would be interesting to investigate the degree of cleaning efficiency improvement by impact factors such as air injection duration, air velocity, NaOCl concentration in backwash water, temperature, and NaOCl contact time with membrane. The co-effects among these factors are complex and the effects of physical assistance or chemical assistance on backwash efficiency and their influencing factors could be seriously investigated in the future.

This dissertation emphasized the performances of the UF system on contaminants removal capacity, microorganisms and microplastics removal capacity, membrane fouling potential, cleaning efficiency, water recovery rate and economic cost. The UF pilot plant was confirmed to successfully meet the water reuse standards both from France and from the European parliament. Furthermore, future research focusing on micropollutants removal, fouling mechanism strategies, and life cycle assessment of membrane-based municipal wastewater treatments, for potable and non-potable reuse applications, continues to need further systematic investigation.

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List of Tables

Table 1-1 Major water reuse applications and constraints worldwide [39,40].	9
Table 1-2 Constituents present in wastewater with their associated risks [42–45].	10
Table 1-3 Water reuse categories and typical application (WHO guidelines and EU Parliament regulation).	12
Table 1-4 Membrane separation characters for water purification [76,87–90].	16
Table 1-5 MF/UF processes alone or combined with pretreatments after secondary treatment for water reuse applications.	23
Table 1-6 Membrane bioreactors (MBR) for water reuse applications.	29
Table 1-7 NF / RO/ FO coupled with pretreatment processes for wastewater reuse applications.	33
Table 1-8 Wastewater recovery rate and energy consumption of membrane-based technologies.	39
Table 1-9 Advantages and disadvantages of various membrane-based treatment processes.	40
Table 1-10 Studies on the combined physical backwash with low concentrated oxidants/disinfectants in backwash water.	44
Table 2-1 Raw wastewater and UF feed quality	50
Table 2-2 Membrane types and operating conditions in Lab scale	51
Table 2-3 UF membrane and module characteristics	53
Table 2-4 Optimal parameters for classic backwash	54
Table 2-5 Physical backwash parameters	55
Table 2-6 Filtration conditions in chapter 3	57
Table 2-7 Backwash conditions in chapter 5 on condition J60t60BW1/3	58
Table 2-8 The types of materials recorded in siMPle and their abbreviations	64
Table 3-1 Classification of the different operating conditions considering the total filtrated volume during a cycle, the initial TMP and the number of CEB in 40h-operation.	81
Table 4-1 Mean UF feed and permeate qualities compared to WHO guidelines, and French reuse standards for irrigation	93

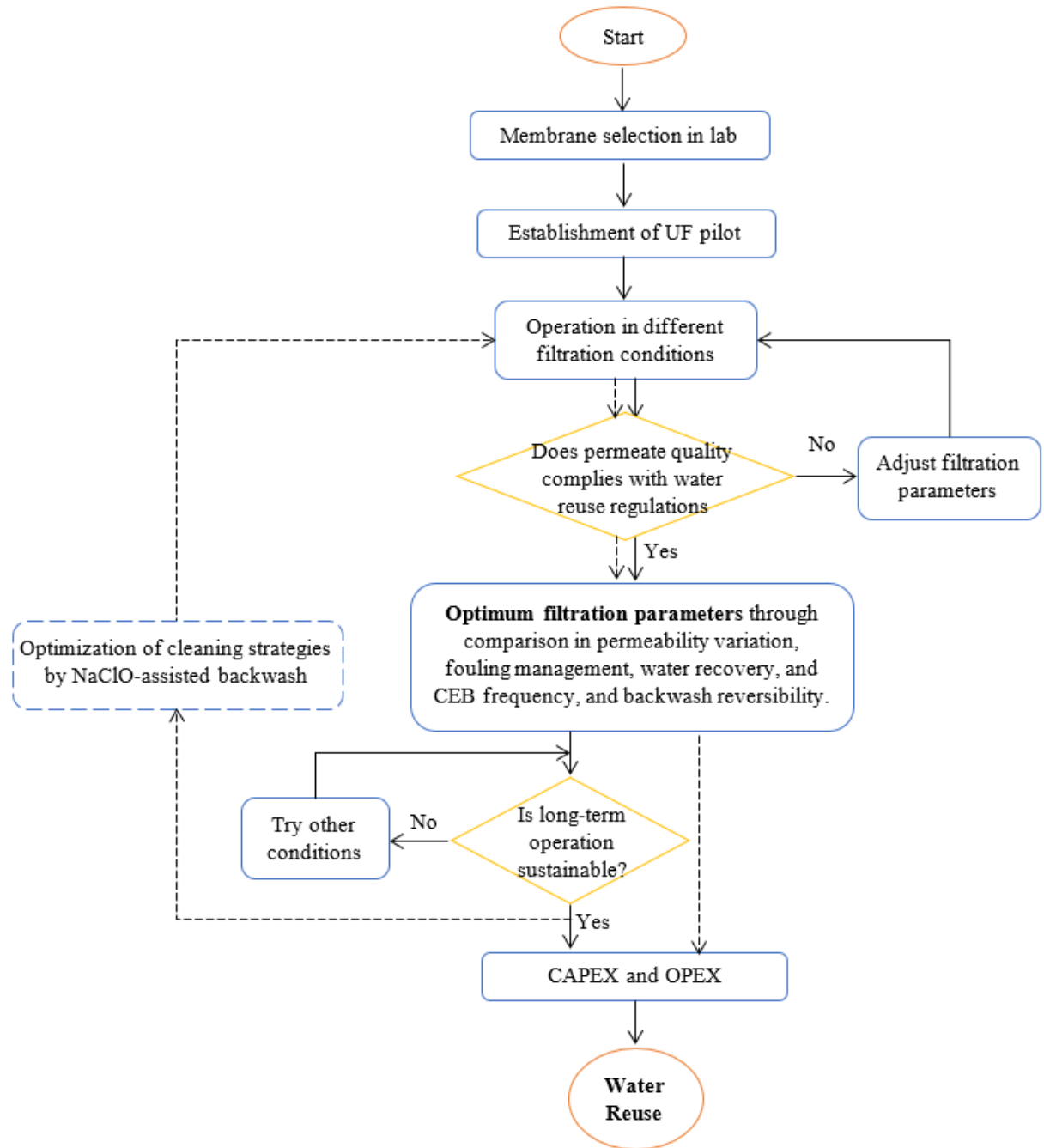
Table 4-2 Detection results in different types of water.....	95
Table 4-3 The microplastics concentration in quantity and in total surface area of samples from municipal WWTP.....	100
Table 5-1 The consumption of NaOCl of each condition in 10 years	107
Table 5-2 Reversibility ranges of backwash in different conditions	112
Table 5-3 The Turbidity and TOC in different types of backwash water and UF feed and permeate	116
Table 5-4 Calculation of CAPEX	122
Table 5-5 Calculation of energy and chemical costs of CEB	123
Table 5-6 Calculation of energy and chemical costs of CB.....	125
Table 5-7 Calculation of energy and chemical costs of AB	125
Table 5-8 Calculation of energy and chemical costs of filtration.....	126
Table 5-9 Energy consumption of Chemical pumps.....	127
Table 5-10 Economic cost	127
Table 5-11 Expenditure cost on different backwash conditions	129

List of Figures

Figure 2-1 Filtration instruments in lab (a) flat sheet membrane, (b) hollow fibers	51
Figure 2-2 Diagram of semi-industrial pilot plant.....	52
Figure 2-3 Picture of pilot plant	53
Figure 2-4 The microscopic-equipped FT-IR spectroscopy system	62
Figure 2-5 Filtration apparatus - vacuum büchner funnel	65
Figure 3-1 Permeability variation of UF membranes at different MWCOs.....	73
Figure 3-2 Fouling resistance formation on different membranes.	74
Figure 3-3 Example of raw wastewater flowrate and feed water turbidity variation throughout the day and night	76
Figure 3-4 Permeability and feed water turbidity variations in condition J60t20BW1/3	77
Figure 3-5 Permeability and feed water temperature variations in condition J60t20BW1/3	77
Figure 3-6 Permeability variation versus time in J60t60BW1/3:(a) the whole filtration process, (b) a selected zoomed period.....	78
Figure 3-7 Permeability variation in conditions: (a) J30t30BW1/3, (b) J60t40BW1/3, (c) J80t40BW1/3, (b) J80t60BW1/3.....	79
Figure 3-8 Fouling reversibility by CB and AB in different conditions	80
Figure 3-9 Resistances' variations versus time at conditions of (a) J60t20BW1/3, (b) J60t60BW1/3 and (c) J100t30BW1/3	83
Figure 3-10 Water reuse recovery rate: conditions at backwash frequency of 1/3. The uncertainty of each column is between 1 and 2%.	84
Figure 3-11 Variation of irreversible resistance as a function of the integral of turbidity vs. time for different BWs (1/9 and No AB) at flux=60 L·m ⁻² ·h ⁻¹ and t = 60 min.....	86
Figure 3-12 Variation of irreversible resistance as a function of the integral of turbidity vs. time for different BWs (1/3, 1/4, and 1/6) at flux=60 L·m ⁻² ·h ⁻¹ and t = 60 min	86
Figure 3-13 Permeability variation in winter from 26/2/2020 to 18/3/2020.....	88
Figure 3-14 Permeability variation in Summer from 1/7/2020 to 27/7/2020.....	88

Figure 4-1 Spectra map of ultrapure water with all particles by siMPle (a) pixel size at 6.25 μm (b) pixel size at 25 μm	96
Figure 4-2 Spectra map of ultrapure water with only microplastics (a) pixel size at 6.25 μm (b) pixel size at 25 μm	96
Figure 4-3 UF feed deposits on 3 μm filter under different digestion duration by H_2O_2 (a) 50mL net UF feed after 1d digestion (b) 50mL net UF feed after 7d digestion (c) 50mL net UF feed after 10d digestion (d) 100mL net UF feed after 10d digestion.....	98
Figure 4-4 Spectra map of detected particles distribution in case (a) by siMPle software	99
Figure 4-5 Dimensions distribution of microplastics in municipal WWTP samples.....	100
Figure 4-6 Abundance of Microplastic material in both quantity and surface area in samples from municipal WWTP. (a) In microplastic numbers (b) in microplastic surface area....	101
Figure 5-1 Permeability, feed water turbidity and temperature variation versus time in different backwash conditions	106
Figure 5-2 Fouling resistance variation versus time in different backwash conditions	111
Figure 5-3 Reversibility ranges of AB and 3rd CB in different conditions	113
Figure 5-4 Reversibility of AB versus R_{irr} on the membrane just before AB.....	115
Figure 5-5 Reversibility of CB versus R_{irr} on the membrane just before CB	115
Figure 5-6 (a) Turbidity recovery rates of AB and 3 rd CB (b) TOC recovery rates of AB and 3 rd CB.....	117
Figure 5-7 Variation of $R_{\text{irr-end}}$ of filtration cycle (n) to the initial cycle (1) a function of the integral of turbidity vs. time	119
Figure 5-8 Components for CAPEX and OPEX.....	128

Appendix I : Technical strategy



Appendix II: Publications and Conferences

List of publications

[1] J. Yang, M. Monnot, L. Ercolei, P. Moulin, Membrane-Based Processes Used in Municipal Wastewater Treatment for Water Reuse: State-of-the-Art and Performance Analysis, *Membranes*. 10 (2020) 131. <https://doi.org/10.3390/membranes10060131>.



membranes



Review

Membrane-Based Processes Used in Municipal Wastewater Treatment for Water Reuse: State-of-the-Art and Performance Analysis

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Abstract: Wastewater reuse as a sustainable, reliable and energy recovery concept is a promising approach to alleviate worldwide water scarcity. However, the water reuse market needs to be developed with long-term efforts because only less than 4% of the total wastewater worldwide has been treated for water reuse at present. In addition, the reclaimed water should fulfill the criteria of health safety, appearance, environmental acceptance and economic feasibility based on their local water reuse guidelines. Moreover, municipal wastewater as an alternative water resource for non-potable or potable reuse, has been widely treated by various membrane-based treatment processes for reuse applications. By collecting lab-scale and pilot-scale reuse cases as much as possible, this review aims to provide a comprehensive summary of the membrane-based treatment processes, mainly focused on the hydraulic filtration performance, contaminants removal capacity, reuse purpose, fouling resistance potential, resource recovery and energy consumption. The advances and limitations of different membrane-based processes alone or coupled with other possible processes such as disinfection processes and advanced oxidation processes, are also highlighted. Challenges still facing membrane-based technologies for water reuse applications, including institutional barriers, financial allocation and public perception, are stated as areas in need of further research and development.

Keywords: membrane processes; municipal wastewater reuse; disinfection efficiency; fouling; water recovery

[2] J. Yang, M. Monnot, T. Eljaddi, L. Ercolei, L. Simonian, P. Moulin, Ultrafiltration as tertiary treatment for municipal wastewater reuse, *Sep. Purif. Technol.* 272 (2021) 118921. <https://doi.org/10.1016/j.seppur.2021.118921>.

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Ultrafiltration as tertiary treatment for municipal wastewater reuse

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ABSTRACT

Water reuse is an enduring topic that benefits the society and future generations of mankind. Ultrafiltration (UF) is one of the most cost-effective treatment technologies for improving water quality. In this study, a semi-industrial UF pilot plant with periodical classic backwash (CB) and air backwash (AB) was operated automatically to evaluate its feasibility and sustainability for municipal wastewater reuse and find out the optimized filtration condition. This study carried out 15 filtration conditions to investigate the impacts of flux (J in $L \cdot h^{-1} \cdot m^{-2}$), filtration cycle time (t in min), and air backwash frequency (BW) on membrane hydraulic filtration performance and membrane fouling management. Through comparative analysis of all conditions in water quality, permeability variation, irreversible fouling management, and water recovery rates, the sustainable conditions J80t40BW1/3 (flux of $80 L \cdot h^{-1} \cdot m^{-2}$, filtration cycle time of 40 min, 1 AB followed with 3 CBs), J60t60BW1/4 (flux of $60 L \cdot h^{-1} \cdot m^{-2}$, filtration cycle time of 60 min, 1 AB followed with 4 CBs), and J60t60BW1/3 (flux of $60 L \cdot h^{-1} \cdot m^{-2}$, filtration cycle time of 60 min, 1 AB followed with 3 CBs), stood out from the others with higher overall performances. Additionally, air backwash showed excellent reversibility on membrane fouling control, which was around 1.25–2 times that of CB in average. After all, long term operation on condition J60t60BW1/3 in winter and in summer confirmed that the UF system could provide sustainable and adaptable filtration performance regardless of the temperature and feed water quality. The UF permeate quality is good enough to be reused in non-potable applications as it met reuse guidelines of the World Health Organization, reuse standards of France and the most recent EU regulation for agricultural irrigation. This work confirms the great interest of UF as tertiary treatment for water reuse and gives operational indications for future industrial-scale production of reclaimed water.

[3] J. Yang, M. Monnot, L. Ercolei, P. Moulin, Impact of Chlorinated-Assisted Backwash and Air Backwash on Ultrafiltration Fouling Management for Urban Wastewater Tertiary Treatment, *Membranes*. 11 (2021) 733. <https://doi.org/10.3390/membranes11100733>.

Article

Impact of Chlorinated-Assisted Backwash and Air Backwash on Ultrafiltration Fouling Management for Urban Wastewater Tertiary Treatment

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Abstract: To improve membrane fouling management, the NaClO-assisted backwash has been developed to improve permeability maintenance and reduce the need for intensive chemical cleanings. This study is aimed to focus on the efficiency of NaClO-assisted backwash in real UF pilot scale and with periodic classic backwash (CB) and air backwash (AB). The impacts on hydraulic filtration performance, physicochemical properties of membrane material under different addition frequencies of NaClO, and the performance of chlorinated CB and AB will be discussed. In result, 10 mg Cl₂ L⁻¹ NaClO addition in backwash water is confirmed to greatly improve the overall filtration performance and backwash cleaning efficiency. One condition stands out from the other due to better control of irreversible fouling, less NaClO consumption in 10 years prediction, sustainable and adaptable filtration performance, and less potential damage on the physicochemical properties of the membrane. Additionally, it can be inferred from this experiment that frequent contact with NaClO induced some degradation on the PES-made UF membrane surface properties. To retain the best state of UF membrane on anti-fouling and qualified production, the optimized condition with more frequent NaClO contact was not suggested for long-term filtration.

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Keywords: NaClO-assisted backwash; UF membrane; irreversible fouling; permeability; membrane properties

[4] Y. Jiaqi, M. Monnot, T. Eljaddi, L. Simonian, L. Ercolei, P. MOULIN, Réutilisation des eaux par ultrafiltration après une filière conventionnelle de traitement des eaux usées, *Eau Ind. Nuis.* 440 (2021) 95.

[5] J. Yang, M. Monnot, T. Eljaddi, L. Ercolei, P. Moulin, Water Reuse from Municipal Secondary Effluent by Ultrafiltration Becomes a Reality More than Ever, *Open Access J. Environ. Soil Sci.* 5 (2020) 655–659.

List of conferences

1. 26ème congrès de l'Ecole doctorale Sciences de l'Environnement (EDSE, 2019)
Poster: Municipal wastewater treatment by Ultrafiltration for water reuse Applications
2. 17ème congrès de la Société Française de Génie des Procédés (SFGP, 2019)
Abstract: Ultrafiltration as tertiary municipal wastewater treatment for water reuse (Oral presentation)
3. 13ème congrès international du GRUTEE
Abstract: Performance of an ultrafiltration pilot plant for municipal wastewater reuse (Oral presentation)
4. 1st GREENERING international conference (2021)
Poster: Performance and permeate quality of ultrafiltration as tertiary urban wastewater treatment for water reuse
5. 24th International Congress of Chemical and Process Engineering CHISA Virtually (2021)
Abstract: Semi-industrial ultrafiltration as tertiary treatment for water reuse: water quality and optimization of operating conditions (Oral presentation)

Ultrafiltration as urban wastewater tertiary treatment for water reuse at semi-industrial scale -Abstract

Water reuse is a sustainable development strategy that benefits society and future generations. In this study, a semi-industrial ultrafiltration (UF) pilot plant established at the outlet of a wastewater treatment plant was studied to assess its feasibility and sustainability for non-potable water reuse. The optimization of operating conditions made it possible to support reliable and sustainable filtration performance, the operating conditions were optimized through comparative analysis in terms of water quality, permeability variation, irreversible fouling management, and water recovery rate. The best conditions were J80t40BW1/3 (flux of $80 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, filtration cycle time of 40 min, 1 air backwash followed by 3 classical backwashes), J60t60BW1/4 and J60t60BW1/3. The long-term study on condition J60t60BW1/3 provides sustainable and adaptable filtration performance regardless of the temperature and feed water quality variation. In addition, the air backwashes enabled excellent reversibility of membrane fouling, which was approximately 1.25 to 2 times higher than of classic backwashes in average. The quality of the UF permeate was good enough to be reused in non-potable purposes as it met reuse guidelines of the World Health Organization, reuse standards of France, and the most recent EU regulation for agricultural irrigation. A specific study of membrane cleaning has shown that the addition of NaClO in backwash water can greatly increase cleaning efficiency of air backwashes. Finally, the calculation of the capital expenditure (CAPEX) and operational expenditure (OPEX) of the UF system under optimized conditions gives a profitable net unit price for water production. Through this thesis, UF is confirmed to be a reliable tertiary treatment for water reuse and the results give operational indications for the industrial scale and provides proposals for the management of membrane fouling by air backwash with chemical assistance.

Keywords: Ultrafiltration, water reuse, fouling control, air backwash, reuse standard

Réutilisation de l'eau par ultrafiltration en tant que traitement tertiaire d'eaux usées urbaines à l'échelle semi-industrielle - Résumé

La réutilisation de l'eau, dans le cadre d'un développement durable est nécessaire pour la société et pour les générations futures. Dans cette étude, une unité d'ultrafiltration (UF) semi-industrielle placée en sortie de station d'épuration a été étudiée pour évaluer sa faisabilité et sa durabilité. L'optimisation des conditions opératoires a permis de donner pour la première fois des contraintes d'exploitations fiables et durables qui se dégagent par une analyse comparative de l'impact de 15 conditions opératoires différentes sur la qualité de l'eau, la variation de perméabilité, la gestion du colmatage irréversible et le taux de conversion de l'eau. Les meilleures conditions ont été J80t40BW1/3 (flux de $80 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$, durée du cycle de filtration de 40 min, 1 rétrolavage essoré suivi de 3 rétrolavages classiques), J60t60BW1/4 et J60t60BW1/3. Une étude sur du long terme avec la condition J60t60BW1/3 a fourni des performances de filtration durables et adaptables quelles que soient la température et la qualité de l'eau d'alimentation. De plus, les rétrolavages essorés à l'air ont permis une excellente réversibilité du colmatage des membranes, qui était d'environ 1,25 à 2 fois supérieure celle des rétrolavages classiques en moyenne. La qualité du perméat d'UF a été suffisamment bonne pour être réutilisée dans des applications non potables car elle répond à la meilleure qualité des directives de réutilisation de l'Organisation mondiale de la santé, aux normes de réutilisation de la France et à la réglementation européenne la plus récente pour l'irrigation agricole. Une étude spécifique du nettoyage des membranes a permis de montrer que l'utilisation de chlore pouvait avoir une efficacité accrue lorsqu'il est couplé à l'effet mécanique des rétrolavages essorés. Finalement, le calcul des dépenses d'investissement (CAPEX) et opérationnelles (OPEX) du système d'UF donne, dans les conditions optimisées, un prix unitaire net de production d'eau rentable. Au travers de cette thèse, l'UF apparaît comme un traitement tertiaire fiable pour la réutilisation de l'eau et les résultats donnent des indications opérationnelles pour l'échelle industrielle et des propositions pour la gestion du colmatage des membranes par rétrolavage à l'air assisté chimiquement.

Mots clés : Ultrafiltration, réutilisation de l'eau, contrôle du colmatage, retrolavage à l'air, normes de réutilisation