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Establishment of High-Rate MBRs for Effective Recovery of Organic Matter from Municipal Wastewater: Fouling Mitigation for Long-term Operation

都市下水からの効率的有機物回収を行う高速 MBR の確立：
長期運転のためのファウリング軽減

Michael J. ROCCO

Division of Environmental Engineering
Hokkaido University

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Advisor: Professor KIMURA Katsuki

By

Michael J. ROCCO

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Division of Environmental Engineering
Graduate School of Engineering
Hokkaido University
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ABSTRACT

Wastewater should be regarded as a resource for the establishment of a sustainable society as opposed to a waste product that needs to be dealt with. A combination of high-rate activated sludge and membrane separation (high-rate membrane bioreactors (HR-MBRs)) is considered a viable option for efficient recovery of organic matter from wastewater. Simultaneously, high effluent water quality can be achieved. However, operation of HR-MBRs suffers from extremely severe membrane fouling caused by short solid and hydraulic retention times. The establishment of the HR-MBR as a working process for organic matter recovery was aimed for in this study, and it was believed that fouling control should be the starting point. Although fouling observed during operation has been reported in studies focusing on HR-MBRs, a clear lack of research on fouling mitigation techniques applied to HR-MBRs was evident.

Control of membrane fouling in HR-MBRs, while extremely difficult, was nonetheless attempted. The effectiveness of a strategy combining granular scouring and frequent chemically enhanced backwashing was assessed. This approach was made possible through the use of robust flat-sheet ceramic membranes. Experiments were conducted at an existing wastewater treatment plant, where the MBR was operated as a high-rate system by setting solid residence time (SRT) and hydraulic residence time (HRT) at 0.5 days and less than 2 hours, respectively. With these settings in place and after the initial stages of operation had had their fouling controlled, an attempt was made to optimize the cleaning protocol for the HR-MBR mentioned previously. Improvements to the reactor design allowed for increased membrane area to be used, and a lower flux was needed to achieve the required HRT (i.e., 0.8 hours). To facilitate a long-term operation (e.g., 1 month) of the HR-MBR under the condition of a practical net flux, a novel in-situ CEB, termed air-exposed CEB (AECEB), was developed in this study. Optimization of the AECEB parameters including choice and concentration of chemicals, duration, and flux were investigated. Following these developments, due to the operational conditions employed, the concentrated stream was found to still be high in volume and low in volatile solids for subsequent resource recovery (e.g., anaerobic digestion). Addition of a secondary process was thought to be a suitable solution. Due to its reduced energy consumption and relative ease of implementation, gravity

separation process with overflow into the reactor was employed to try and solve the concentration issue. The effect of fouling induced by the secondary process was focused on to ensure that the process could still function effectively.

Even though a relatively high flux of 20 L m^{-2} h^{-1} was applied, the proposed HR-MBR exhibited a very low fouling rate of 1.3 kPa/day in the first four days of operation. In this timeframe, the system could recover more than 70 percent of the organic matter from raw wastewater, whereas the concentration of chemical oxygen demand (COD) in the effluent was lowered to less than 20 mg/L. After significant fouling was observed on the membrane, optimized AECEB achieved almost complete cancellation of the fouling developed in long-term experiments. However, transmembrane pressure surged when the operation of the HR-MBR was resumed after implementing the AECEB. It was found that scouring agents made from polyethylene glycol released foulants during the AECEB. Based on this finding, alternative scouring agents made from polyurethane were used. Following this result, when gravity separation was introduced as a secondary concentration process, an increase in concentration by 8.3 times and a reduction in volume by 11.5 times showed that the combination was successful in concentrating the sludge. However, gravity separation increased fouling in the HR-MBR which resulted in a much faster transmembrane pressure increase and the deterioration of filterability in the reactor. This addition also lowered the total amount of recovered organic matter because of the increased SRT and mineralization.

The low SRT and HRT set in this study enabled efficient recovery of organic matter, whereas the severe fouling in the HR-MBR could be controlled by the intensive membrane cleaning. The performance of the proposed HR-MBR observed in this study was clearly superior in the initial stages to those reported in previous studies with related operational conditions. The combination of AECEB and granular scouring using the polyurethane agents worked exceptionally well. The fouling in the HR-MBR could be almost completely controlled and long-term operation was deemed possible. Additionally, AECEB did not negatively impact the HR-MBR operation.

Although concentration of the organic matter was achieved, the drawbacks show a need for a more optimal system to concentrate the organic matter without having the negative impact

observed in this study. Other options, such as chemically enhanced clarification or direct membrane filtration, should also be considered for a secondary concentration process, although they come with significant drawbacks such as the cost of coagulants or additional energy requirements.

The study highlighted significant improvements in the HR-MBR process, particularly in COD removal, loss reduction, and recovery rates, demonstrating superior performance compared to HRAS systems due to the complete separation of particulate and colloidal matter by the membranes. The HR-MBR achieved COD removal rates of approximately 91% without the primary sedimentation basin and 95% with its inclusion, surpassing other HR-MBRs under similar conditions. The inclusion of the primary sedimentation basin increased the capture of organic matter by 10%, from 68% to 78% of the total incoming COD.

The study also demonstrated the potential of HR-MBR technology to transform wastewater treatment plants into resource recovery facilities, enhancing recovery rates and reducing energy consumption. The study successfully restored the permeability of severely fouled membranes through an on-line cleaning process, minimizing downtime and enhancing the feasibility of organic matter recovery. The final length of operation achieved was 29 days of continuous operation, with the next step being to study the effects of influent, fouling, filterability, and sludge characteristics in more detail to optimize the process and maximize recovery.

Keywords: HR-MBR, Carbon Recovery, Wastewater Treatment, Granular Scouring, Chemically Enhanced Backwash, Air-exposed CEB, Fouling Mitigation

TABLE OF CONTENTS

ACKNOWLEDGEMENTS I

ABSTRACT II

TABLE OF CONTENTS V

LIST OF FIGURES XI

LIST OF TABLES XIII

ABBREVIATIONS XIV

CHAPTER 1. INTRODUCTION 1

1.1 RESEARCH BACKGROUND 1

1.1.1 Resource consumption and recovery 1

1.1.2 Adsorption/ bio-oxidation processes 3

1.1.3 High-rate activated sludge processes for organic matter recovery 3

1.1.4 Definition of the HR-MBR 4

1.1.5 Fouling during the membrane process 5

1.1.6 Foulant removal and cleaning 6

1.2 RESEARCH PROBLEM 6

1.3 AIMS, OBJECTIVES AND RESEARCH QUESTIONS 7

1.4 NOVELTY AND SIGNIFICANCE 8

1.5 LIMITATIONS 8

1.6 STRUCTURAL OUTLINE 10

CHAPTER 2. LITERATURE REVIEW 11

2.1 THE HIGH-RATE MEMBRANE BIOREACTOR 11

2.1.1 History of HR-MBRs for up-concentration of organics 11

2.1.2 Operational conditions of the HR-MBR 12

2.1.3 Effluent water quality and organic matter recovery in HR-MBRs 13

2.1.4 Seeding of suspended biomass from conventional MBRs 13

2.2 FOULING IN THE HR-MBR 13

2.2.1 Foulants 13

2.2.2 Chemical Flocculation and Bioflocculation 14

2.3 FOULING MITIGATION TECHNIQUES 15

2.3.1 Relaxation and hydraulic backwashing 15

2.3.2 Aeration, crossflow and scouring 16

2.3.3 Chemically enhanced backwash (CEB) 16

2.3.4 Granular scouring materials 17

2.4 APPLICABILITY OF THE FOULING MITIGATION TECHNIQUES AND CERAMIC MEMBRANES 17

2.4.1 Membrane type and the effect of flux on the severity of fouling 18

2.4.2 Membrane material 18

2.5 CONCENTRATION LEVELS IN THE HR-MBR 19

CHAPTER 3. GENERAL METHODOLOGY 20

3.1 EXPERIMENTAL SETUP 20

3.1.1 The bench scale HR-MBR 20

3.1.2 Operational conditions 21

3.1.3 Frequent chemically enhanced backwash (CEB) 22

3.1.4 Granular materials and aeration 22

3.1.5 HR-MBR modifications for increased performance 22

3.2 ASSESSMENT OF MEMBRANE FOULING 23
3.2.1 Transmembrane pressure (TMP) evolution 23
3.2.2 Clean water permeability measurement 23
3.2.3 Filtration cell 25
3.2.4 Assessment of distribution of fouling 25
3.2.5 Additional fouling layer characterization through resistance measurements 26
3.2.6 Clean membranes and new membranes 27
3.3 EXPERIMENTAL DETAILS 27
3.3.1 Chapter 4: Initial Attempts at Fouling Control in HR-MBRs 27
3.3.2 Chapter 5: Effective Reset of Fouling through Air-exposed CEB 28
3.3.3 Chapter 6: Gravity Settling for Additional Concentration of the Recovered Organic Matter 33
3.4 WASTEWATER QUALITY 34
3.5 FRACTIONATION OF SAMPLES AND BIOFLOCCULATION ANALYSIS 36
3.6 MASS BALANCE ANALYSIS 36
3.6.1 Mass Balance Calculation 36
3.6.2 Accumulated Mass Balance 37
3.7 ANALYTICAL METHODS 38
3.7.1 Carbon and organic matter measurement techniques 38
3.7.2 Extracellular polymeric substance (EPS) extraction 38
3.7.3 Fourier transform infrared (FTIR) spectroscopy analysis 38
3.7.4 Dissolved organic matter analysis 39
3.7.5 Scanning Electron Microscopy 39

CHAPTER 4. RESULTS AND DISCUSSION OF THE IMPLEMENTATION OF THE HR-MBR BASED ON ESTABLISHED FOULING MITIGATION TECHNIQUES 40

4.1 INDIVIDUAL EFFECTS OF FOULING MITIGATION TECHNIQUES (RUN1) 40

4.2 EFFECTIVENESS OF THE COMBINATION OF GRANULAR SCOURING AND CEB WITH VARIOUS CONCENTRATION OF SODIUM HYPOCHLORITE (RUN 2) 41

4.2.1 Fouling evolution in Run 2 41

4.2.2 Analysis of fouling Resistance in Run 2 43

4.2.3 Bioflocculation in Run 2 43

4.3 LONGER OPERATION OF THE HR-MBR (RUN 3) 45

4.3.1 Fouling evolution in Run 3 45

4.3.2 Carbon recovery and effluent quality achieved by the HR-MBR 47

CHAPTER 5. RESULTS AND DISCUSSION OF THE EFFECTIVE RESET OF FOULING IN A HR-MBR BY IN SITU, AIR EXPOSED AND CHEMICALLY ENHANCED BACKWASH 48

5.1 RUN 4 (IMPLEMENTATION OF AECEB-1 WITH A SHORT INTERVAL) 48

5.2 FIRST ATTEMPT TO OPTIMIZE THE AECEB 49

5.3 APPLICATION OF THE SEMI-OPTIMIZED AECEB-2 IN A LONG-TERM OPERATION (RUN 5) 52

5.4 APPLICATION OF THE OPTIMIZED AECEB-3 IN A LONG-TERM OPERATION (RUN 6) 53

5.5 LONG-TERM OPERATION WITH POLYURETHANE GRANULAR AGENTS (RUN 7) 55

CHAPTER 6. CONCENTRATION OF THE RECOVERED ORGANIC MATTER WITH GRAVITY SETTLING 59

6.1 PERMEATE WATER QUALITY DETAILS 59

6.2 FINAL ORGANIC MATTER CONCENTRATION 59

6.3 MASS BALANCE ANALYSIS RESULTS 61

6.4 DETERIORATION OF FILTERABILITY 62

6.5 FOULING LAYER INVESTIGATION 63

CHAPTER 7. COMPARATIVE ANALYSIS OF HR-MBR: ORGANIC MATTER RECOVERY AND ENERGY EVALUATION 68

7.1 COMPARISON OF HR-MBR EFFECTIVENESS 68

7.2 EVALUATION OF THE ENERGY BALANCE AND THE FEASIBILITY OF THE HR-MBR AS AN EFFECTIVE TECHNOLOGY 71

7.2.1 Primary treatment 71

7.2.2 Secondary treatment and organic matter recovery 72

7.2.3 Additional treatment and other resource recovery 74

CHAPTER 8. CONCLUSIONS, IMPLICATIONS AND FUTURE PROSPECTS 76

8.1 CONCLUSIONS 76

8.2 IMPLICATIONS AND FUTURE PROSPECTS 78

REFERENCES 80

APPENDIX A. SUPPLEMENTARY MATERIALS 98

S1. Literature review of the HR-MBR 98

S2. Mass balance during Run 3.1 and 3.2 104

S3. Mass balance during Run 4 105

S4. Clean water permeability results of the long-term experiments (Runs 5-7) 105

S5. Optimization experiments for the reduction of chemical concentrations in the air-exposed CEB. 106

S6. Accumulated mass balance during Run 7. 107

APPENDIX B. PUBLICATIONS AND CONFERENCE PROCEEDINGS 108

Publications 108

Conference Proceedings 108

LIST OF FIGURES

Figure 1. Schematic flow diagrams of the process trains during the CAS process (Top) and the adsorption/ bio-oxidation process (Bottom). 2
Figure 2. Categorization of membrane technologies for up-concentration of organic matter. 5
Figure 3. Schematic flow diagram of the experimental system. The shaded area indicates the closed loop water circulator for temperature control. 21
Figure 4. Schematic flow diagram of the experimental setup for the clean water permeability analysis. The water in the hatched area was separate from the water in the clean water tank. 24
Figure 5. Representation of a conventional CEB (left) and the AECEB (right). 29
Figure 6. Schematic flow diagram of the experimental setup of the HR-MBR installed at Soseigawa WWTP. 29
Figure 7. Schematic flow diagram of parallel HR-MBRs equipped with ceramic flat-sheet membranes utilized for organic matter recovery. 33
Figure 8. The transmembrane pressure (TMP) evolution during the operation of Runs (a) 1.1 with only CEB and (b) 1.2 with only granular scouring. 40
Figure 9. TMP evolution in Runs (a) 2.1, (b) 2.2, (c) 2.3 and (d) 2.4, in which the concentrations of NaClO in the backwash solution were set at 1000 ppm, 500 ppm, 100 ppm and 50 ppm, respectively. In these runs, granular scouring was also carried out. 41
Figure 10. Distribution of fouling resistances assessed at the termination of Runs 2.1, 2.2, 2.3 and 2.4. 42
Figure 11. Size distributions (particulate, colloidal and soluble) of COD in the biomass suspension just before termination of Runs 2.1 to 2.4. 44
Figure 12. Increases in TMP in Runs 3.1 and 3.2. The dotted lines represent the linear regression of TMP evolutions. During Run 3.1, at around 2.5 days of operation, a power outage that lasted 4 hours stopped all activity in the experimental system. 45
Figure 13. TMP evolution of the extended operation of Run 3.2. 46
Figure 14. Increase in TMP in Run 4. The arrow indicates the implementation of AECEB-1. 48
Figure 15. TMP evolution in Run 5. The arrow indicates the implementation of AECEB-2. 52

Figure 16. TMP evolution in Run 6. The arrow indicates the implementation of AECEB-3. 53

Figure 17. FTIR spectra of samples from Run 6 of the fouling layer (A) and the suspended solids before (B) and after (C) the AECEB compared with the spectrum of the PEG standard (D). 55

Figure 18. TMP evolution from the long-term operation of an HR-MBR (Run 4). The black arrow shows the start of the AECEB when TMP reached 30 kPa. 56

Figure 19. FTIR spectra of different fouling layers (A) found on the membrane surface and suspended solids in the reactor (B) before and 24 hours after AECEB-3 in Run 7..... 57

Figure 20. Concentration of organic matter and solids in the concentrated discharge (A) and the mixed liquor (B) of the HR-MBR with gravity separation (MBR 1) and without gravity separation (MBR 2). The left margin shows the respective energy content of the recovered organic matter (A) and the VS/TS ratio of the mixed liquor organic matter (B). 60

Figure 21. Daily mass balance analysis of both MBR1 (A) and MBR 2 (B). 61

Figure 22. Comparison of LC-OCD chromatograms showing the soluble fraction of the mixed liquor (ML) suspension in MBRs 1 and 2. 62

Figure 23. TMP evolution of the two reactors during operation (A) and resistance measured at the termination of the seven-day operation (B) of two membranes from each reactor. The models generated from the additional resistances measured at varying fluxes from the membranes in MBR1 (C) and MBR2 (D) are shown. 64

Figure 24. SEM observations of the surface of the fouled membranes at the end of operation. Different magnifications of the average surface of the membranes from MBR 1 (A images) and from MBR 2 (B images). 66

Figure 25. Influent COD recovery compared with the SRT in different high-rate systems treating different wastewaters. High-rate contact stabilization combined with a membrane bioreactor (HRCS-MBR). 69

Figure 26. Influent COD recovery compared with the SRT in different high-rate systems treating different wastewaters. 70

Figure 27. Influent COD loss percentage compared with the SRT in different high-rate systems treating different wastewaters. 71

LIST OF TABLES

Table 1. Details of the AECEB examined in each run. 31
Table 2. Average characteristics of the wastewater (primary sedimentation effluent) fed into the HR-MBR in the experiments conducted in Chapter 4. 34
Table 3. Average wastewater characteristics of the influent and effluent from the primary sedimentation basin during experiments of Chapter 5. 35
Table 4. Average wastewater characteristics of the influent and effluent from the primary sedimentation basin during experiments of Chapter 6. 35
Table 5. Preliminary air-exposed CEB optimization tests for different solutions. 50
Table 6. Recoveries of the permeability achieved by different conditions of AECEB using NaClO. 50
Table 7. Energy consumption and recovery of wastewater treatment using the HR-MBR and the primary sedimentation basin 73

ABBREVIATIONS

AB	adsorption/ bio-oxidation
AECEB	air-exposed CEB
CAS	conventional activated sludge
CEB	chemically enhanced backwash
COD	chemical oxygen demand
CST	capillary suction time
DMF	direct membrane filtration
DN	dissolved nitrogen
DO	dissolved oxygen
DOC	dissolved organic carbon
DOM	dissolved organic matter
EPS	extracellular polymeric substances
FTIR	Fourier transform infrared spectroscopy
HRAS	high-rate activated sludge
HRCS	high-rate contact stabilization
HR-MBR	high-rate membrane bioreactor
HRT	hydraulic residence time
LC-OCD	liquid chromatography with organic carbon detection
LMH	liters per meter squared per hour
MBR	membrane bioreactor
ML	mixed liquor
PEG	polyethylene glycol
PLC	programmable logic controller
PTFE	polytetrafluoroethylene
PU	polyurethane
SEM	scanning electron microscopy
SMP	soluble microbial products
SRT	solid residence time
SS	suspended solids
TMP	transmembrane pressure
TN	total nitrogen
TOC	total organic carbon
TS	total solids
VCF	volumetric concentration factor
VS	volatile solids
VSS	volatile suspended solids
WWTP	wastewater treatment plant

CHAPTER 1.
INTRODUCTION

1.1 RESEARCH BACKGROUND

Resource recovery from wastewater has gained increased attention due to the shift away from only performing wastewater treatment and organic matter removal. Many researchers have sought to recover the vast amount of resources found in wastewater, be it energy or nutrients, instead of conventional disposal focused methods. However, these conventional methods are not effective for a recovery focused sustainable approach. Therefore, new methodologies are essential to further improve the recovery of these resources. This research aims to evaluate and improve one of these new technologies, the high-rate membrane bioreactor (HR-MBR). This chapter will introduce the background, aims and scope of the research, while also commenting on its limitations.

1.1.1 Resource consumption and recovery

The main source of energy in the world is fossil fuel combustion, which releases stored carbon and contributes to the climate change crisis [1]. To mitigate the release of carbon into the atmosphere, it is important to reduce the consumption of electricity that is mainly produced with fossil fuels and create renewable sources [2]. Wastewater treatment plants (WWTPs) are significant contributors to energy consumption in cities. It is estimated that WWTPs consume 1-3% and 3-4% of the total energy in Europe and in the United States of America, respectively [3]. These are some of the reasons why reduction of electricity consumption in WWTPs is of the upmost importance.

New options for converting wastewater treatment into a net-positive energy process have been investigated in recent studies. Instead of eliminating the energy source by using conventional activated sludge (CAS) treatment, capturing organic matter in wastewater into a concentrated stream has drawn significant attention [4]. From the concentrated stream,

energy can be efficiently recovered from wastewater using currently available technology (e.g., anaerobic digestion (AD)) [5,6]. Sarpong et al. [7] reported that by capturing organic matter at the beginning of the treatment trains, conversion of WWTPs into net-positive energy production facilities could be achievable. It is therefore important to establish an efficient technology for capturing organic matter in wastewater.

Some technologies that have been used to capture organic matter in wastewater are chemically enhanced primary treatment [8], membrane processes [9], contact stabilization activated sludge [10], and high-rate activated sludge (HRAS) [11]. Some combinations of these technologies have gained significant attention due to the added benefits derived from combining them, especially the combination of membrane systems with the other organic capture technologies.

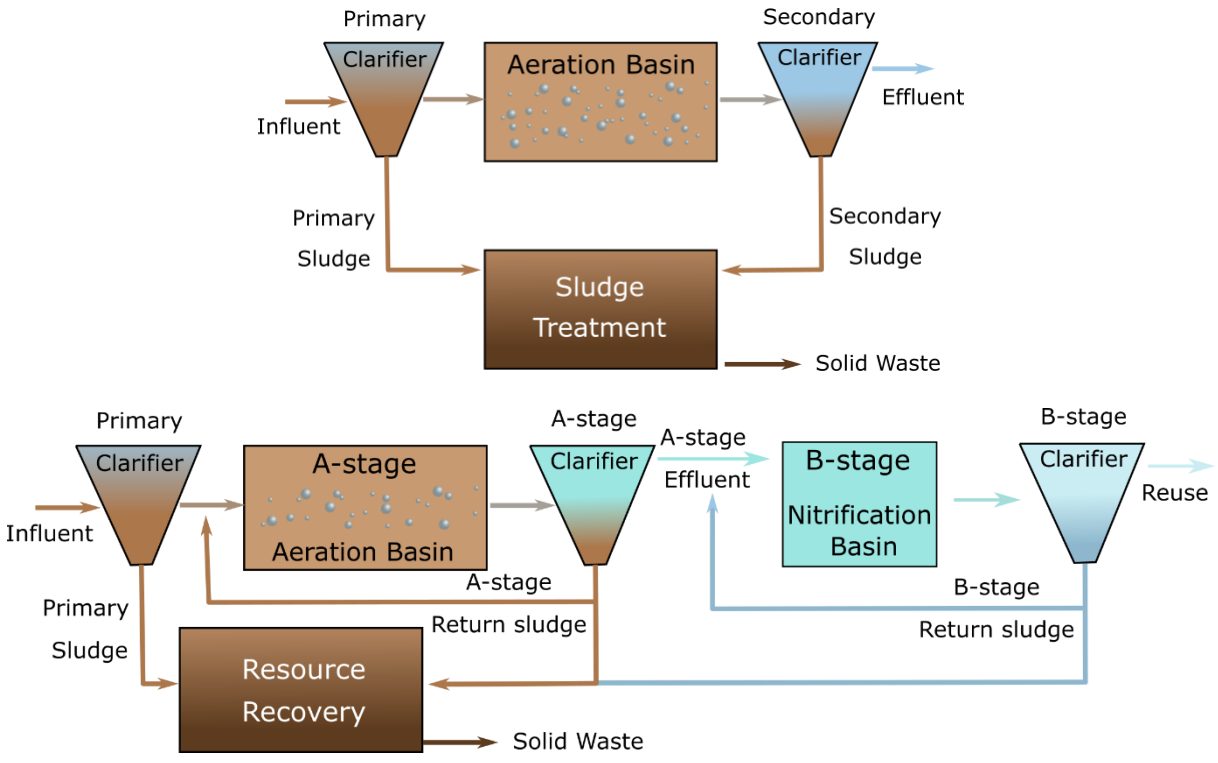


Figure 1. Schematic flow diagrams of the process trains during the CAS process (Top) and the adsorption/ bio-oxidation process (Bottom).

1.1.2 Adsorption/ bio-oxidation processes

Figure 1 shows both schematic flow diagrams from the process trains of the CAS system and adsorption/ bio-oxidation system (i.e., AB system). The main difference between these two systems is the mindset discussed previously. While the CAS system only aims to remove all unwanted constituents from the wastewater, the AB system's goal is to collect the available resources from wastewater and use them. To achieve this goal, the process was divided into two parts, the adsorption stage and the bio-oxidation stage. The adsorption stage (A-stage) is similar to CAS where the incoming influent is aerated and bioflocculation (i.e., flocculation promoted by the biomass in activated sludge) is promoted to achieve separation of solids from the liquid phase. However, the difference lies in that this is the only task which must be completed by the A-stage. Because the relative goal of this stage is limited, the task can be completed quickly, and organic matter is not lost to mineralization during long residence times. Therefore, most of the organic matter is redirected to resource recovery before it is mineralized. Subsequently, the bio-oxidation stage (B-stage) is where the remaining constituents, mainly the nitrogen compounds, are removed and recovered.

Although the AB system should theoretically be much better than CAS. In practice, several problems arose with the constituent removal and nitrogen removal during the A-stage portion and B-stage portion, respectively. The rise of single reactor aerobic/anoxic systems as the preferred technology and a lack of technologies to address the problems encountered created the downfall of the AB system and only a few operating plants using this system remain today [12]. Recently, there has been a resurgence of the A-stage from the AB system as new technologies have been invented to solve the issues first encountered. Because of the lack of a B-stage, the A-stage has had a change of naming to high-rate activated sludge (HRAS).

1.1.3 High-rate activated sludge processes for organic matter recovery

High-rate processes enable the capture of colloidal and dissolved organic matter into microbial flocs without degradation [10,13]. These processes achieve this by controlling the amount of time the liquid and solid fractions of the wastewater remain on average inside the bioreactor, known as the solid residence time (SRT) and hydraulic residence time (HRT), respectively.

HRAS processes are, specifically, biological treatment processes characterized by an extremely short SRT (0.5 to 2 days) and an extremely short HRT (less than 5 hours) [11,12]. Through the capture of colloidal and dissolved organic matter, high-rate systems can therefore increase the amount of organic matter transferred to AD [14]. However, this process has some drawbacks that need to be addressed.

Generally, in high-rate processes, shorter SRT leads to higher carbon capture rates and longer SRT leads to better effluent quality (i.e., a higher degree of mineralization of organic carbon) [15]. Due to this tradeoff, simultaneously achieving a high recovery rate of carbon capture and a sufficiently high quality of effluent would be difficult with only an HRAS process. This issue is aggravated by the settling problems (i.e., bulking) in the solid separation stage, especially under low temperature conditions [16–18]. Thus, the definition of the HRAS system's short SRT is directly contrary to that of a CAS system, where longer SRTs promote better separation during clarification. Additionally, at shorter HRTs needed for concentration, higher aeration rates and reduced separation of mixed liquor suspended solids are also observed [19]. Integration of membrane filtration into the high-rate process may solve this issue, and successfully separate and concentrate the organic matter in wastewater.

1.1.4 Definition of the HR-MBR

There are some technologies involving membranes for particle separation in the field of organic matter recovery. Figure 2 shows some of the different ways to categorize these technologies, and some of the similarities and differences considered. The effectiveness of these combined technologies is still under investigation in recent years. To further improve these technologies, it is necessary to clearly define them. For this point, the definition of the key parameters that outline what is and is not considered as HR-MBRs is essential.

A combination of membrane filtration and biological treatment is defined as a membrane bioreactor (MBR). In contrast, when biological treatment is not the focus of the treatment, it is considered direct membrane filtration (DMF) [20]. During operation of the MBR, some other details affect the definition of the type of MBR used. For example, when two tanks are used for the aerator instead of one, it is termed contact stabilization [10]. Thus, different parts of the process inform the term used.

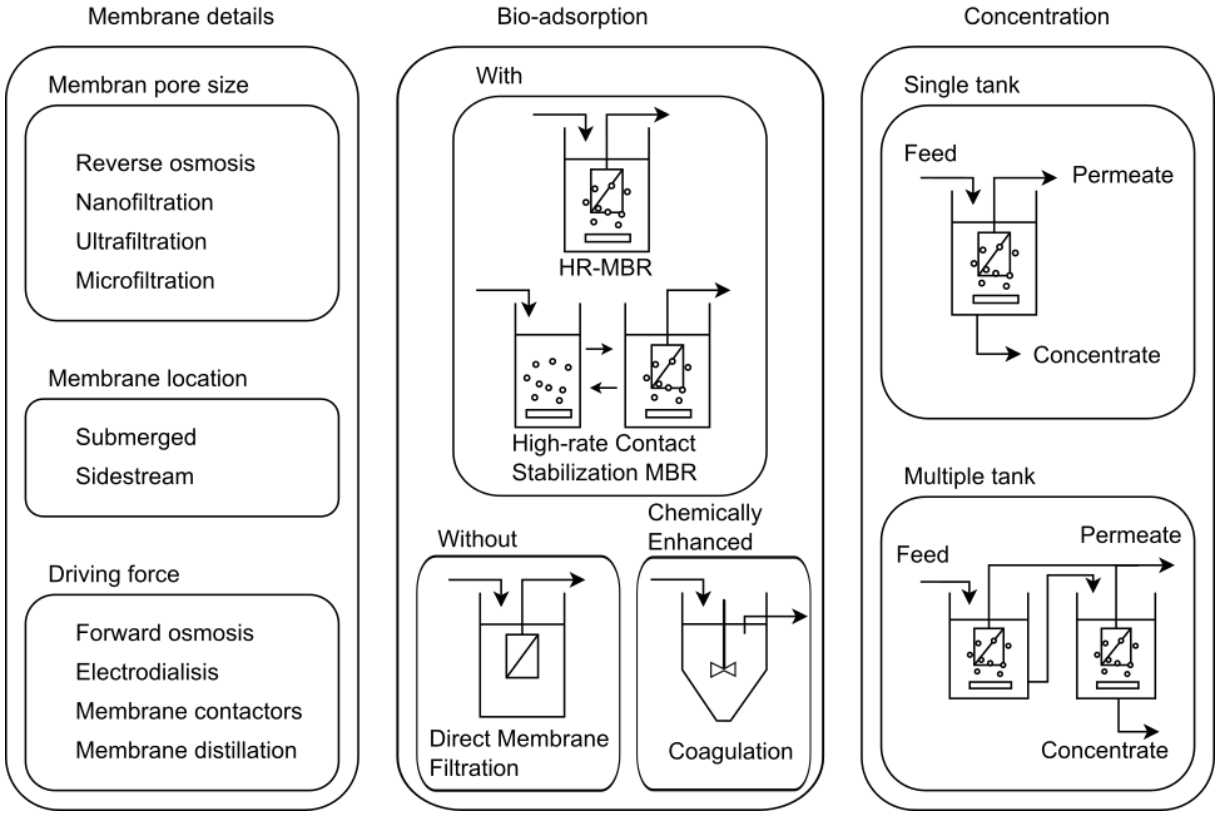


Figure 2. Categorization of membrane technologies for up-concentration of organic matter.

Conventional MBRs, a direct evolution from CAS, utilize long SRTs to ensure complete denitrification and bio-oxidation, as well as to reduce fouling. In contrast, operating an MBR with the high-rate mode (i.e., under a short SRT and HRT), termed HR-MBR, can achieve both a high rate of recovery of organic matter in wastewater and sufficiently reduce the concentration of organic matter in the treated water (permeate) [21–23]. Other names referring to HR-MBRs were high loaded MBRs [22] or Super-Fast MBR [24].

1.1.5 Fouling during the membrane process

Fouling is still one of the leading problems with membrane processes [25–28]. Concerning HR-MBRs, fouling is especially problematic because of the reduction in filtration performance. In HR-MBRs, short HRT is essential to be able to achieve the concentration of the organic

matter. Higher membrane fluxes can help achieve these shorter HRTs, but with higher fluxes, severe membrane fouling is expected.

1.1.6 Foulant removal and cleaning

Off-line cleaning refers to the process of removing the membrane plates from the reactor and conducting a cleaning protocol before reintroduction. Off-line cleaning is conducted when fouling has reached its maximum allowed transmembrane pressure (TMP). In contrast, on-line membrane cleaning refers to the process of cleaning the membrane units without moving the membrane module.

1.2 RESEARCH PROBLEM

Feasibility is already well established in the literature for HR-MBRs, including synthetic wastewater [21,24], municipal wastewater [22,23,29] and greywater [30,31]. Recently, a few studies have conducted research on the operating parameters which directly affect the operation of the HR-MBR. These parameters include the SRT, HRT, DO, microbial community, bioflocculation, temperature, etc [23,31–34]. However, one of the main problems of HR-MBRs is the increased fouling propensity with extremely short SRT [30,34], which is commonly only discussed as a consequence of the operational condition. A more detailed analysis of the fouling in HR-MBRs and especially the fouling mitigation processes is necessary. Although previous studies reported on the mechanisms of fouling in HR-MBRs [32,35,36], mitigation of fouling in HR-MBRs has rarely been investigated. It was decided that a detailed investigation into what fouling mitigation techniques could be used in a HR-MBR would be a way to fill this gap in the current research.

HR-MBRs have been found to be feasible, however, in order to progress to pilot and full-scale applications, extensive work on fouling mitigation is necessary. Operation length directly affects the applicability of a treatment process and unless operation can be maintained for a long period (i.e., about one month), the process will most likely not advance from laboratory scale. Additionally, initial testing of the operational conditions used in this study showed that final organic matter concentrations were below 3 g-SS/L. One of the main

ways to recover energy from the concentrated organic matter is anaerobic digestion, however, this concentration makes it difficult to conduct anaerobic digestions directly [37] without a secondary concentration step. Therefore, an additional concentration step and the effect this additional step would have on fouling in the HR-MBR was thought to be important for this study.

1.3 AIMS, OBJECTIVES AND RESEARCH QUESTIONS

The establishment of the HR-MBR as a functional treatment process was the aim of this study. To accomplish this aim, the process needs to ensure both a high recovery of concentrated organic matter and high permeate water quality from actual municipal wastewater. Practical objectives were set to accomplish this primary goal. Despite time constraints, two key areas were focused on to try to achieve the study's aim: the duration of operation and the final concentration of organic matter. As the operation length directly influences various performance aspects of the HR-MBR, long-term operation (i.e., about one month) without off-line cleaning was deemed an important first goal.

For the first key area of investigation, increasing operational length, assessment of the application of appropriate fouling mitigation techniques and the development of new fouling mitigation methods was employed. The suitability of ceramic membranes for concentration of organic matter and implementation of the fouling mitigation techniques, was also evaluated. Additionally, it was imperative that the overall efficiency of the HR-MBR was preserved throughout the entire process to maintain the HR-MBR's overall high performance. This ensured high-quality permeate and a high recovery rate. The important research questions that arose from these first objectives were as follows:

- What are the effective fouling mitigation techniques that could be used to increase the operational length and reduce fouling rates in a flat-sheet HR-MBR?
- What is the longest operation time that could be achieved by the HR-MBR without off-line cleaning?
- Can the HR-MBR maintain high effluent quality and high recovery percentage simultaneously while implementing fouling mitigation techniques?

To ensure an additional concentration step was possible, the impact of concentration techniques on fouling within the HR-MBR was examined and the concentration increase provided by a gravity separation basin was evaluated. Gravity separation as a concentration step was chosen as the initial starting point for increased concentration because of the minimal energy required to be operated. The next set of objectives provided more research questions:

- Can ceramic membranes conduct organic matter concentration, and can this concentration be further increased by a secondary process or gravity settling?

1.4 NOVELTY AND SIGNIFICANCE

The use of ceramic membranes for wastewater treatment, particularly for organic matter recovery, is currently under investigation. Although ceramic membranes have been used in many different applications, this study represents the first instance of submerged flat-sheet ceramic membranes being used for HR-MBRs during organic matter recovery.

The novel application of fouling mitigation techniques contributes to increased cleaning, reduced chemical usage, and the advancement of technologies such as HR-MBRs towards more sustainable futures. Despite these technologies being available for use today, costs and practicality remain as limiting factors. Incremental improvements enhance the feasibility of a technology, preparing it for widespread adoption, step by step. Thus, the development and implementation of novel fouling mitigation techniques with real wastewater are both important for advancing the establishment of these new processes. Additionally, a novel approach to fouling mitigation, which involves the complete in-situ cancellation of severe fouling, was developed and implemented in this study.

1.5 LIMITATIONS

In this study, bench scale experiments were conducted. Therefore, it is important to note that these results may not be directly applicable to pilot or full-scale operations. This

limitation is particularly relevant when energy calculations are considered, as larger and longer periods of study are required for more accurate results.

In our experiments, a single type of ceramic flat-sheet microfiltration membrane was utilized. The influence of the choice of membrane on the results is significant. Different results might be obtained if other membrane materials (e.g., titanium oxide or silicon carbide) and types (e.g., ultrafiltration, forward osmosis, etc.) were used. The potential use of hollow fiber membranes or other membrane systems has been demonstrated in other studies.

When considering the type of wastewater, it was found that a more realistic context for implementing a treatment process is offered by real wastewater than synthetic wastewater. However, due to the global variability in wastewater composition, the results of this study cannot be generalized without the treatment process being first applied to different wastewaters.

Constraints were also experienced in terms of the operational conditions used to operate the HR-MBR in our study. Certain operational conditions (such as SRT or filtration time) were fixed to improve fouling mitigation techniques and could not be altered. This means that all conclusions drawn from these results primarily pertain to the operational conditions described in this study and may differ if other operational conditions were used by other researchers.

A significant factor encountered was the variability in the influent wastewater between subsequent runs. Despite the best efforts made to ensure reproducibility, it was found that investigations will always have limitations when different feed solutions are used. At present, the exact effects of real wastewater are not replicated by synthetic wastewater. Consequently, it was concluded that the fouling from real wastewater cannot be substituted by the fouling produced by an MBR with synthetic wastewater. Thus, real municipal wastewater was chosen even though variability was known to have a strong effect on results and replication would be difficult.

Finally, time was a constraining factor in our study. This led to a reduction in the scope of the project's concentration phase, and only one type of concentration process was explored. Additionally, although replication was found to be difficult, a reduced number of runs were done due to time constraints. The limited number of runs in this study could restrict its

applicability. To enhance this study, it is suggested that additional runs exploring more parameter variations and feed types should be considered.

1.6 STRUCTURAL OUTLINE

After the introduction chapter, Chapter 1, Chapter 2 will review the relevant literature. Following this, Chapter 3 will go through all the materials and general methodologies which were used in this study. Next, Chapters 4, 5 and 6 contain the results and discussions pertaining to the experimental sections. In Chapter 4, initial implementation of conventional fouling mitigation techniques was explored. Chapter 5 directly follows, in which new fouling mitigation techniques were developed to extend operation length even more. Chapter 6 focuses on increasing the concentration of organic matter after the initial concentration by the HR-MBR. Following that, Chapter 7 compares the results obtained with other HR-MBR studies. Finally, Chapter 8 contains the conclusions of the study and the implications.

CHAPTER 2.
LITERATURE REVIEW

2.1 THE HIGH-RATE MEMBRANE BIOREACTOR

Chapter 2 consists of a literature review. The history of HR-MBRs was explored to give a broad overview of why it is an important technology. Next, the main operational conditions are investigated. Also, to establish the opportunities and drawbacks of fouling mitigation, a deep dive into fouling and mitigation techniques was conducted. The application of these techniques was subsequently discussed. Finally, the concentration of the recovered organic matter was studied.

2.1.1 History of HR-MBRs for up-concentration of organics

A detailed history of the HRAS process is described by Orhon et al. [12]. Where they describe how it was developed as a strategy to reduce the energy requirements and decrease operation length in conventional activated sludge (CAS). This process, known as the AB system at the time, was quickly replaced by a wide application of the anoxic/aerobic single stage reactor. This was due to the complications found during nitrification in the B-stage and a lack of technologies to deal with the issue at the time.

This AB process has recently reemerged as HRAS from the conceptual basis of the A-stage [10,11,38,39]. The same goals are stated in the new implementation; however, the same drawbacks still apply to the process. As described in these reports [12,18], significant improvements to particle separation, rather than gravity sedimentation, are necessary for this technology to be applicable to the current standards of constituent removal. Therefore, membrane filtration is a clear step to achieve these improvements [40].

2.1.2 Operational conditions of the HR-MBR

Initial reports have demonstrated the effectiveness of utilizing membrane systems as particle separation during the implementation of HRAS in HR-MBRs. Table S1 presents a collection of the studies that focused on the HR-MBR and the highlights of those research studies. These studies focused on operational parameters to evaluate the performance of the HR-MBR under different conditions. The aim was to identify the optimal parameters for conducting organic matter recovery and energy recovery while evaluating constituent removal.

The Volumetric Concentration Factor (VCF), defined as the ratio of influent flowrate over the wasted flowrate of a membrane system [46]. In the absence of backwash, the VCF becomes the ratio of SRT over HRT, which was the focus of a significant portion of the studies depicted in Table S1. The VCF can be used as a benchmark for the maximum organic matter that could be recovered by the HR-MBR [22,23]. However, when backwash is conducted, a different calculation of the mass balance is necessary

SRT, a crucial aspect of the HR-MBR, controls various system aspects such as microbial community, recovery efficiency, final concentration, and bioflocculation [23,30,35,47]. Higher SRTs (i.e., more than 1 day) appear to be less effective at maintaining a high recovery efficiency of the organic matter [23,31,36]. Conversely, shorter SRTs (i.e., less than 1 day) tend to result in lower permeate quality and lower final concentrations, which may necessitate additional concentration steps after the HR-MBR [24,36]. A significant tradeoff exists when choosing the right SRT for a specific system, and careful consideration should be given when making this decision.

Membrane systems offer the advantage of completely separating the HRT from the SRT, allowing the HRT to be modified to control the performance of the HR-MBR. The HRT is primarily controlled in two ways: by modifying the total filtration flowrate or by changing the effective biomass suspension volume inside the reactor. The total filtration flowrate can be increased either by introducing additional membrane area or by increasing the net flux. An increase in net flux, intended to increase the flowrate, is expected to result in higher fouling rates. In contrast, the utilization of a larger membrane area is expected to result in higher membrane costs. Therefore, the implementation of better membrane fouling mitigation techniques may allow for the use of higher fluxes, enabling the use of smaller membrane areas and reducing the overall cost of the membranes to achieve the same HRT.

2.1.3 Effluent water quality and organic matter recovery in HR-MBRs

Since the initial stages of HR-MBR development, organic matter removal has been high. In the first study, even when exploring 0.25 days SRT, Ng et al. [21] had much lower total COD in the effluent of the HR-MBR compared to the completely mixed HRAS reactor [21]. Researchers have found that without membrane separation, it is very difficult to achieve high effluent quality at lower SRTs [10,19].

2.1.4 Seeding of suspended biomass from conventional MBRs

Biomass in CAS is seeded into a new bioreactor by inserting sludge from an operating system (i.e. inoculation). The main objective of doing so is to maintain a similar community of microorganisms which are known to achieve the required biodegradation. In addition, slow growing strains are also important when seeding as the time necessary for these strains (e.g., nitrifiers) to accumulate to the required level can be drastically reduced [48]. Compared to CAS, conventional MBRs appear to have reduced need for inoculation, however, starting the MBR without inoculation can have negative impacts on fouling during startup [49].

In HR-MBRs, seeding of biomass has been conducted in most of the studies, even though the microorganism communities have been clearly shown to quickly change from the original communities found in the inoculum [42,43]. Therefore, seeding in HR-MBRs may not be necessary. Seeding of biomass presents an additional problem for fouling mitigation studies, as discussed in the previous paragraph, the initial stages are very important in terms of fouling rates. When seeding is used, the filterability of the seeded sludge is commonly higher than raw wastewater, altering the initial stages and preventing the observation of fouling during these stages.

2.2 FOULING IN THE HR-MBR

2.2.1 Foulants

Similar biological processes are observed in the operation of HR-MBRs as those in conventional MBRs, leading researchers to the belief that the fouling in HR-MBRs is probably similar to the fouling that occurs in conventional MBRs. Therefore, the foulants that are

observed in conventional MBRs are considered a good starting point when exploring which foulants have a greater impact on HR-MBR operation. Some fouling and fouling mitigation details observed in the studies focused on HR-MBRs are also shown in Table S1. The mechanisms of fouling observed during HR-MBR operation were a substantial part of most of these studies. An important role was proposed to be played by submicron particles by Faust et al. [23], and a correlation with fouling was found [23]. Additionally, it was proposed by a large portion of the studies that SMPs were a major part of fouling.

Recently, Faridizad et al. [35] explored the direct impact of SRT on fouling in HR-MBRs. They observed an increase in concentration of SMP when increasing SRT from 0.5 days to 2 days during operation with synthetic municipal wastewater [35]. In contrast, Yilmaz et al. [34] found that with industrial dye wastewater, slowly biodegradable SMPs tended to accumulate when SRT was reduced from 5 days to 0.5 days. This accumulation was thought to be the cause of the increased fouling in lower SRTs [34]. Although both studies have contrasting opinions on the SRT which causes the highest fouling rate in HR-MBRs, both reach the same conclusion that there is a correlation between increased SMPs in the HR-MBR biomass suspension and higher fouling rates. This point coincides with high fouling observed in conventional MBR operation when EPS, especially SMP is found in the biomass suspension [50,51]. Organic material such as biopolymers, proteins and polysaccharides are the main components of EPS and SMP [52]. Fouling mitigation which focuses on organic foulant removal could be an important starting point for HR-MBRs.

2.2.2 Chemical Flocculation and Bioflocculation

Coagulation and flocculation are commonly used to increase the efficiency constituent removal during later stages in biological processes. Coagulation refers to the introduction of chemicals to a suspension which neutralizes the surface charge of the constituents, allowing them to form flocs (i.e., flocculate) [46]. Chemical coagulation can form a significant part of the cost of a separation process, Taboada-Santos et al. [8] compared a chemically enhanced primary treatment to the effectiveness of a HRAS process and found that the effectiveness of recovery was counteracted by the cost [8]. Bioflocculation on the other hand, is mainly attributed to aeration which is already employed in MBR setups and is not normally the primary driver for aeration demand.

Bioflocculation is the process by which microorganisms in the biomass suspension entrap organic matter through production of EPS and SMP [52]. This EPS-mediated bioflocculation plays an important role in the effectiveness of the HR-MBR, as was determined previously [22,30–32]. However, as described in the previous section, SMP (i.e., the dissolved part of EPS) plays a significant role in fouling during HR-MBR operation. Thus, flocculation, be it biological or chemical, is interconnected with fouling in HR-MBRs.

Chemical flocculation and/ or bioflocculation in HR-MBRs affects the outcome in two ways, they increase the removal of the soluble and colloidal organics by adsorption and reduce fouling of the membranes. Increasing the adsorption of soluble and colloidal organic matter onto the sludge flocs makes it easier for the HR-MBR to separate the organic matter into concentrated stream [22]. Additionally, larger sludge flocs produce a more porous cake layer that allows for less restricted filtration, as opposed to a denser gel layer formed by smaller organic constituents [31]. Therefore, flocculation must be encouraged when the HR-MBR is in operation.

2.3 FOULING MITIGATION TECHNIQUES

Various reviews have explored the state of fouling mitigation techniques available for submerged membrane systems [53–55]. Some of the common fouling mitigation techniques used in membrane systems are relaxation, hydraulic or chemically enhanced backwashing, and scouring by aeration. Additionally, emergent mitigation techniques include granular scouring, quorum quenching, oscillation and ultrasonic vibration. A combination of different fouling mitigation techniques is a commonly accepted way to deal with fouling in submerged membrane systems. In Table S1, some of the applied fouling mitigation techniques are shown. These mainly include the common techniques, however the work by He et al. [29] show the effectiveness of coagulation and powdered activated carbon on fouling reduction [29]. More studies that focus on new ways of combining the fouling mitigation techniques are needed.

2.3.1 Relaxation and hydraulic backwashing

Relaxation of a membrane refers to the time when filtration is stopped to allow for back-transfer of constituents from the boundary layer near the membrane. It helps the filtration

process to avoid additional fouling by reducing concentration polarization. However, an increase in the instantaneous flux is needed to achieve the same volume of filtrate production as that when relaxation is used [56]. The same could be said for hydraulic backwashing and any other fouling mitigation technique where the filtration has to be stopped for the technique to be applied. Therefore, the shorter the time needed to conduct the cleaning, the lower the instantaneous flux needed to achieve the same HRT. Furthermore, backwashing has been shown to be a better alternative than relaxation when compared one to one [57], indicating that in the case where a reduction in filtration time is necessary, backwashing may be chosen over relaxation.

2.3.2 Aeration, crossflow and scouring

In CAS, aeration serves two primary functions: mixing and oxygenation. The rate of organism growth is increased and the removal of constituents is accelerated through the oxygenation of the sludge, either by adsorption via bioflocculation or through oxidation. Mixing is facilitated by the upward movement of bubbles exiting the aerators, which transport nutrients to other parts of the biomass suspension. In conventional MBRs, in addition to mixing and oxygenation, aeration also creates cross flow for scouring the membrane surface, thereby removing fouling and enhancing permeability [58].

These principles are also applied during HR-MBR operation to prevent fouling and achieve bioflocculation. However, due to the reduced oxygen demand [41], HR-MBRs require less oxygenation. Akanyeti et al. [22] proposed the use of coarse bubble aeration, as opposed to fine bubble aeration, in response to this reduced need for oxygenation and increased need for fouling control [22]. Fine bubbles have a larger surface area for mass transfer of oxygen from the bubbles to the liquid phase, than coarse bubbles [59]. For this reason, they are preferable in conventional MBRs [60,61] when the sludge concentration increases, as they transfer oxygen more effectively to the biomass suspension. However, during HR-MBR operation, the reduction of mineralization is encouraged, and fouling control becomes the main focus of the aeration system, making coarse bubbles preferable [62].

2.3.3 Chemically enhanced backwash (CEB)

CEB is defined as the addition of chemicals to the backwash solution, with the aim of promoting foulant removal. In full-scale conventional MBRs, the use of CEB is a standard

practice for mitigating fouling [63,64]. Chemicals commonly used include sodium hydroxide (NaOH), citric acid, hydrochloric acid (HCl), sodium hypochlorite (NaClO), and ozone (O_3) [37,65–69]. When these chemicals are applied to the backwash solution, they come into direct contact with the foulants inside the membrane and the fouling layer on the membrane surface, making CEB a direct method for applying chemicals to the foulants. However, the use of CEB in MBRs presents challenges. While it can remove biofouling and inorganic fouling on membranes, the introduction of the CEB solution can also cause damage to the biomass, which drives the biological process for constituent removal [70,71]. Especially at high concentrations, adverse effects on the biomass suspension have been shown to be produced by CEB [72,73].

In the case of HR-MBRs, not only is constituent removal an important function of the biomass suspension, but bioflocculation and adsorption are also crucial for both recovery and fouling control. Direct contact with the CEB solution can have adverse effects on bioflocculation [72], reducing the effectiveness of the biomass suspension and, in some cases, even inducing additional fouling [74,75].

2.3.4 Granular scouring materials

Previously, many studies have used granular scouring materials to reduce fouling and improve outcomes of membrane processes [71,76–81]. Scouring materials include plastic beads, granular or powdered activated carbon, biocarriers and aerobic granular sludge (AGS) [55]. Although the positive effect on fouling control has been recognized, some studies have shown damage caused by granular materials on polymeric membranes [76,79].

2.4 APPLICABILITY OF THE FOULING MITIGATION TECHNIQUES AND CERAMIC MEMBRANES

The fouling in HR-MBRs could not be effectively controlled in previous studies [23,30,36]. The aforementioned problem of membrane fouling in HR-MBRs, however, may be addressed by intensive membrane cleaning. Very intensive membrane cleaning for conventional MBRs by using a combination of granular scouring and chemically enhanced backwashing (CEB) has been proposed [71,73]. The use of robust flat-sheet ceramic membranes enabled the implementation

of such intensive cleaning. It was thought that this approach might also be effective for the HR-MBRs used for capturing carbon from municipal wastewater, in which extremely severe membrane fouling occurs.

2.4.1 Membrane type and the effect of flux on the severity of fouling

As discussed previously, HRT is mainly controlled by total filtration flowrate in HR-MBRs. Because HR-MBRs need HRT to be as low as possible, depending on total membrane area, high filtration fluxes are necessary to produce the required flowrates. Initially, the use of flat sheet membranes was common with HR-MBRs [22,23,30], however, after the operational conditions were slowly optimized, hollow fiber membranes had the advantage due to the higher membrane area and packing density [29,32,34]. Thus, lower HRTs could be achieved without having to increase the flux.

Flat sheet membranes, in contrast, suffer from reduced membrane packing, which necessitates higher fluxes to achieve the same HRTs as hollow fiber membranes. However, they have the benefit of easier cleaning, removal and replacement, complimenting the above drawbacks.

2.4.2 Membrane material

Polymeric membranes are the most common type of membrane used in the world today [82,83]. The cost of polymeric membranes has been declining steadily in recent years. However, some fouling mitigation techniques are not suitable for the use of polymeric membranes. Harsh chemicals, such as the ones described for CEB, especially at high concentrations, have been known to damage polymeric membranes [63]. New developments in polymeric membranes have allowed them to have higher resistances to chemicals and have been used more frequently with CEB [37,68,84].

Ceramic membranes are membranes produced with inorganic materials. Some of the main ones are alumina (aluminum oxide), titanium oxide, silicon carbide, etc [85,86]. These membranes are known for their high resistance to chemicals, temperature and abrasion [87], making them perfect for intensive cleaning techniques that can remove stubborn fouling from membranes [73,88].

2.5 CONCENTRATION LEVELS IN THE HR-MBR

As discussed previously, the concentration level highly depends on the combination of HRT and the SRT. In addition, fouling mitigation techniques which require filtration to stop, also directly affect the concentration amount of the biomass suspension. Most of the studies depicted in Table S1 used low HRTs (below 2 hours) which increases the volumetric factor and allows for the recovery of organic matter [23,29]. However, SRT is the main driver of the VCF because of the longer times involved. Therefore, high concentration was mainly achieved in previous studies when SRTs were longer. The tradeoff of longer SRTs to reduced recovery efficiency is still largely misunderstood and more research is needed.

CHAPTER 3.
GENERAL METHODOLOGY

3.1 EXPERIMENTAL SETUP

Chapter 3 will go into detail about the methodology used throughout the study. The methodology is structured where the physical details are first, such as the experimental setup and fouling layer characterization techniques. Next, influent water quality and experimental details are outlined. Followed by bioflocculation and mass balance analysis. Finally, additional methodologies, including organic matter measurement analyses, are at the end.

3.1.1 The bench scale HR-MBR

Bench-scale experiments were carried out at an existing wastewater treatment plant (Sapporo, Japan) connected to a combined sewer system. Flat-sheet ceramic membranes (Meidensha, Tokyo, Japan) with a nominal pore size of 0.1 μm and total membrane area of 0.2048 m^2 were immersed in the reactor. Figure 3 shows a schematic flow diagram of the experimental setup. Clearances between the membranes were 10 mm, allowing free movement of the granules for efficient scouring of the membrane surface.

Temperature has been found to greatly affect the results of a HR-MBR [45]. It was decided that temperature would be maintained as close to 20°C as possible to avoid uncertainties derived from temperature variations. The temperature inside the reactor was maintained at 19.4 ± 0.8 °C by a thermostatic system (MT-1000, Eyela, Tokyo, Japan). Concentrate (i.e., recovered organic matter) was withdrawn directly and intermittently from the HR-MBR by using a peristaltic pump. A screen (3 mm holes) was installed in the concentrate recovery line to prevent the loss of granules. The concentrate pump extracted the suspension for 10 minutes and stopped for 10 minutes. This allowed two important processes, the flowrate of the concentrate pump could be increased to ensure there weren't any blockages in the tube of the concentrate line. Second, granular materials that would get attached to the screen could be

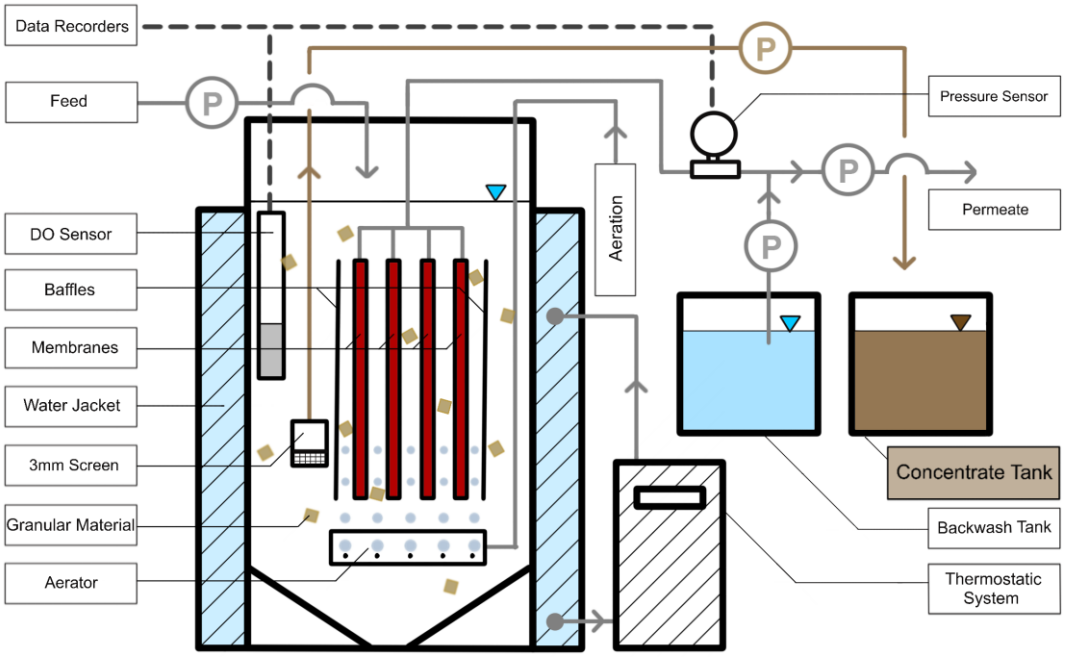


Figure 3. Schematic flow diagram of the experimental system. The shaded area indicates the closed loop water circulator for temperature control.

released and would not get stuck in the holes. This was necessary because the granular materials used were gel-like in nature and deformable. With enough pressure applied, they could deform and wedge into the holes of the screen. To prevent this point further, the end of the concentrate line had a bell shape to increase the surface area of the inlet and reduce the flowrate near the screen.

3.1.2 Operational conditions

Each filtration cycle was composed of 15 minutes of filtration followed by 4 minutes of CEB. Preliminary tests indicated this condition was viable and that relaxation was not necessary for fouling mitigation when CEB was employed. Therefore, relaxation was omitted to improve the total net filtration time. The HR-MBR was operated as constant flux mode and was manually measured every 24 hours and adjusted to maintain the correct flux. A peristaltic pump (Masterflex L/S Pump, Avantor, United States) was used to create the vacuum for filtration. The net flux was set at 20 L m^{-2} h^{-1} to achieve an HRT of 1.6 hours,

resulting in a VCF of 6.7. SRT in this study was set at 0.5 days. As described in Chapter 2, shorter SRT are believed to have better resource recovery than longer SRTs. Although fouling is expected to be worse than if longer SRTs were used, it was believed that the fouling mitigation techniques could mitigate the fouling enough to continue the operation.

3.1.3 Frequent chemically enhanced backwash (CEB)

Chemically enhanced backwash was used throughout the study. During the initial experiments in Chapter 4, a CEB protocol of 50 ppm NaClO solution was pumped backwards through the membrane at a flux of 8 LMH for the 4 minutes where filtration was stopped. This process was conducted for every filtration cycle. To produce the CEB solution, tap water was used and was mixed with NaClO concentrated solution at 5% concentration. In must be noted that the solution for frequent CEB was prepared every 24 hours and there is a small loss of strength throughout the day until the next solution is prepared.

3.1.4 Granular materials and aeration

Granular materials were used in this study for two main reasons, the first and most important was to scour the membrane surface to remove fouling. The other reason was the reduction of organic suspension in the reactor.

The main granules used for scouring the membrane surface in this study were cylindrical granules (4 mm in diameter and length) made from polyethylene glycol (BCN, Nisshinbo Chemical Inc., Tokyo, Japan). Based on the results obtained in preliminary experiments, granular materials were placed in the reactor so that the volume of the granules was 30% of the reactor's total volume. The granules were fluidized by a coarse bubble aeration flow (4 L/min) provided through two half-inch tubes placed perpendicular to the membrane plates at the bottom of the reactor. Each tube had 5 holes of 2 mm in diameter which provided coarse bubble aeration for oxygenation, scouring and granule fluidization.

3.1.5 HR-MBR modifications for increased performance

After the results obtained in Chapter 4, it was thought that a lower HRT would benefit the overall performance of the HR-MBR. Increasing the number of membranes units inside the reactor could be used to reduce HRT. Spacing between the membrane sheets was reduced

from 10 mm to 8 mm. In flat-sheet configurations of membrane modules, the closer the membranes are, the more membranes can be inserted into each membrane module (i.e., the packing density). The spacing between membranes is what determines the packing density and because of the limits of maximum flux in submerged systems treating wastewater, it is what controls the minimum HRT. In addition, the factor that controls the minimum spacing of membranes in this case is the size of the granular materials and the ability to fluidize them. Fluidization of granular materials was confirmed to be possible with the reduced spacing which allowed for the modification to be realized. Reduced spacing allowed for introduction of additional membrane plates (from 4 to 8) to increase membrane area inside the reactor and reduce HRT. Due to the increase in membrane plates, tank volume was also reduced from 8.4 L to 7.3 L, reducing the effective biomass suspension somewhat.

3.2 ASSESSMENT OF MEMBRANE FOULING

3.2.1 Transmembrane pressure (TMP) evolution

During membrane filtration, TMP evolution was used to assess the degree of fouling during the HR-MBR operation. Transmembrane pressure was measured with a pressure sensor (GC-61, Nagano Keiki, Japan) between the membrane module and the peristaltic pump, as seen in Figure 3. The data logging was set to record the voltage every minute, allowing for visualization of the low and high points of the TMP graph which showed reversible and irreversible fouling (details described later).

The system panel was changed from a completely electromechanically controlled system to a programable logic controller (PLC) system. This meant that the timings controlled by the electromechanical timer relays were changed to digital timers, ensuring accuracy of timings during filtration. This allowed TMP evolution graphs, which had some artifacts from the slightly irregular timings, to become smoother.

3.2.2 Clean water permeability measurement

The current methods used to characterize the fouling layer can be divided into invasive and non-invasive analyses. In the former, the procedure damages the fouling layer to obtain

the data, and, in the latter, the fouling layer would be preserved for further analysis. Therefore, a non-invasive method to characterize the fouling layer must be used in order to compare both before and after the cleaning technique. If a destructive method would be used, this comparison would be impossible. Additionally, membrane fouling in membrane processes is heterogeneous, meaning, each membrane will have slightly different fouling layer characteristics than the others from the same module [89]. Therefore, the only way to make sure the comparison is correct would be to compare the fouling layer on the same membrane before and after a cleaning technique.

Clean water permeability measurements were conducted in this study to assess the filtration resistance caused by fouling on/in the membrane. Based on Darcy's Law, filtration resistance of the membranes was calculated using Equation 1.

$$J = \frac{\Delta P}{\mu R} \tag{1}$$

where R is the total filtration resistance (m^{-1}), ΔP is transmembrane pressure (TMP) (Pa), J is the flux ($m^3\ m^{-2}\ s^{-1}$) and μ ($g\ m^{-1}\ s^{-1}$) is the permeate viscosity.

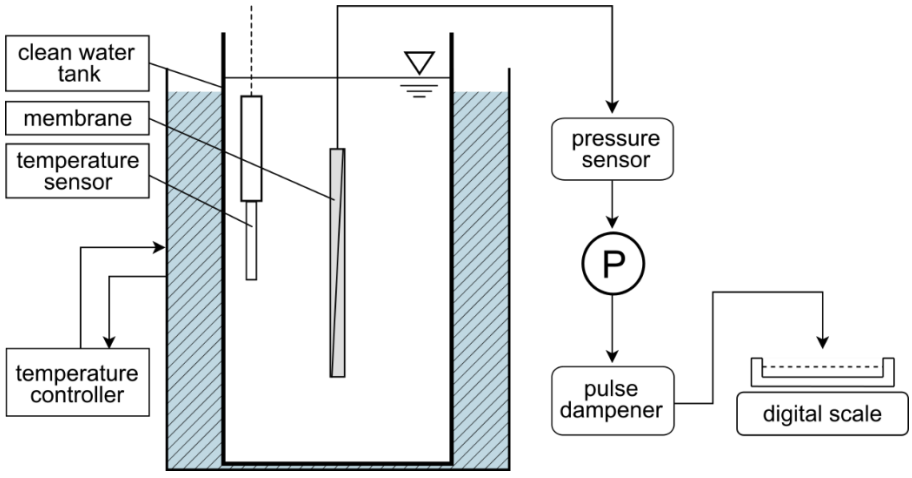


Figure 4. Schematic flow diagram of the experimental setup for the clean water permeability analysis. The water in the hatched area was separate from the water in the clean water tank.

3.2.3 Filtration cell

Figure 4 shows the experimental setup used for the clean water permeability analysis. The membrane unit was completely submerged in clean water and connected to the system by a short tube segment. The digital pressure sensor (GC-31, Nagano Keiki, Japan) and the digital scale were both connected to a computer for data logging. The pump (Masterflex L/S Standard Digital Pump, Avantor, United States) was controlled manually. Because a peristaltic pump was used, a pulse dampener was added in line after the pump to convert the flow from pulse flow to constant flow. The temperature controller maintained a constant temperature of the tap water between 18 and 22 degrees. The temperature of tap water inside the clean water tank was monitored and recorded for the calculation.

During filtration measurement, tap water was used as filtrate solution. Tap water is more economical and readily available than reagent grade water. Additionally, ultrapure water has been shown to have better cleaning capabilities than tap water during backwash experiments [90]. This may be caused by the lack of cations and natural organic matter in ultrapure water, which are present in tap water. Thus, tap water was preferred to prevent any alteration to the fouling layer during the test. An important assumption was that the increase in fouling layer during the clean water permeability measurement utilizing tap water, was negligible during the short measurement.

3.2.4 Assessment of distribution of fouling

Utilizing the resistance in series model [46], the total resistance calculated from Equation 1 was separated into three sources of resistance: intrinsic resistance of the membrane (R_m), reversible resistance (R_r) mainly caused by the cake and gel layers, and irreversible resistance (R_{irr}) from the intrapore foulants [91]. To separate the three resistances, three measurements were conducted. First, permeability was measured when the membranes were clean before the operation (R_m) (i.e., intrinsic membrane resistance). Next, a measurement was conducted after the membranes were fouled (R_f). Lastly, physical cleaning was conducted with a soft sponge to remove the cake layer, and the permeability was measured once again (R_{pc}). Physically reversible fouling resistance (R_{re}) and irreversible fouling resistance (R_{ir}) were calculated as follows:

$$R_{ir} = R_{pc} - R_m \quad (2)$$

$$R_{re} = R_f - R_{pc} \quad (3)$$

3.2.5 Additional fouling layer characterization through resistance measurements

Although the measurement of clean water permeability is equivalent to a dead-end filtration cell test, it differs in two key aspects. The first difference is that it is assumed the fouling layer does not increase as filtration volume increases. The other difference is that not all solids removed from the permeate, remain part of the fouling layer. When we conduct filtration in a submerged MBR, fouling mitigation techniques are used to prolong the operation length (e.g., cross flow shear stress, aeration and bubble scouring, CEB, etc.), which remove foulants from the membrane. Therefore, a different way to model the fouling layer of a fouled membrane during the measurement of the clean water permeability was necessary.

Multiple measurements of the clean water permeability analysis were conducted at varying fluxes to show how the compressibility of the fouling layer reacted to the measurements [92]. During preliminary tests, the shape of the data showed similarities to the power model employed for dead-end filtration, however, obtaining the model with methods outlined in those studies [93,94] proved to be difficult. The difficulty arose from the fact that both the bulk concentration is variable, and the formation of the fouling layer is variable depending on the fouling mitigation technique's effectiveness. Therefore, an empirical generalized linear model approach was used to fit the data.

A generalized linear model [95] is a class of model which utilizes the maximum likelihood methodology in statistics to generate linear predictors where the resulting equations are not necessarily linear. In this case, the linear predictors of the resistance dependent on flux were estimated through the Poisson family and log link function. The model resulted in the following equation:

$$\ln R(J) = \beta_0 + \beta_1 J \quad (4)$$

where R is the filtration resistance, J is the flux, and β_0 and β_1 are the linear predictors estimated for the model. The data was fitted to the model through the statsmodel python

package [96,97]. After generating the model, a clear non-linear relationship is observed which can tell us more about the fouling layer than only one measurement could.

3.2.6 Clean membranes and new membranes

Prior to the initiation of each experiment, all membranes were thoroughly cleaned by a 24 h soak in a 1000 ppm sodium hypochlorite (NaClO) solution followed by a 24 h soak in a 2000 mg/L citric acid (or oxalic acid solution). After the two soakings, these membranes were designated as "clean membranes". This process was done to ensure that the filtration resistance of the cleaned membranes was the same as that of a new membrane. Filtration resistance was assessed for all clean membranes prior to use. The initial resistance of the membranes used was $4.5 \pm 0.1 \times 10^{11}$ m^{-1}, which is close to the resistance of a new membrane ($3.9 \pm 0.1 \times 10^{11}$ m^{-1}).

3.3 EXPERIMENTAL DETAILS

3.3.1 Chapter 4: Initial Attempts at Fouling Control in HR-MBRs

In all experiments of Chapter 4, net filtration flux, aeration rate, and SRT were fixed at 20 L m^{-2} h^{-1} (LMH), 4 L/min, and 0.5 days, respectively. Three series of experiments, Runs 1, 2 and 3, were conducted in Chapter 4. Run 1.1 only with CEB and Run 1.2 only with granular scouring, were conducted to establish the effectiveness of each of the fouling mitigation techniques in isolation. Subsequently, Run 2 was carried out to investigate concentrations of NaClO for CEB at which stable operation of the HR-MBR could be sustained. A NaClO concentration of 500–3000 mg/L is typically used to remove irreversible fouling in submerged MBRs [98]. In this study, 1000 mg/L and 500 mg/L were chosen as high concentrations of NaClO for CEB. A low concentration of 50 mg/L and an intermediate concentration of 100 mg/L were also investigated. Run 2 was conducted to investigate the extent of carbon recovery achievable by the proposed HR-MBR using ceramic membranes. In Run 3, the operation was carried out for 4 days, corresponding to 8 times the SRT. It should be noted that SRT in the HR-MBR was very short in this study (12 h). Therefore, the HR-MBR could be started up within a few days and no seed sludge was added. Run 3.1 was stopped at four days of operation

as it was concluded that a successful run had been achieved. Next, Run 3.2 was conducted to haver reproducibility, however, after four days of operation, it was deemed opportune to extended the operation for verification of the maximum length of operation achievable by the HR-MBR with the current membrane fouling techniques utilized.

3.3.2 Chapter 5: Effective Reset of Fouling through Air-exposed CEB

3.3.2.1 Development of the Air-exposed CEB

In the previous chapter, with intensive cleaning, the HR-MBRs could be operated under a relatively high flux of 20 LMH for several days. However, frequent off-line chemical membrane cleaning would be necessary to continue the operation for a long period (e.g., 1 month). Further improvement in cleaning is necessary to present HR-MBRs as a practical option. In this study, we developed a novel on-line cleaning methodology for HR-MBRs, which was termed air-exposed CEB (AECEB). The protocol of the AECEB consisted of three steps. First, the organic suspension is drained from the reactor and the membranes are exposed to air. Then chemical reagents are introduced from the permeate side: namely, CEB is conducted. Finally, the chemical solution is drained from the reactor and the operation is resumed with new feed.

The cleaning efficiency of AECEB should be higher than that of conventional CEB for the following reasons (Figure 5). In the case of conventional CEB, the chemicals immediately disperse into the organic suspension when they are delivered to the membrane surface. The contact between the chemicals and the fouling layer is therefore limited. Additionally, in conventional CEB, the permeating chemicals will have negative impacts on the microorganisms in the organic suspension [75], which may cause unnecessary membrane fouling [72,74]. In contrast, in the case of AECEB, chemicals introduced from the permeate side eventually drip down the surface of the fouling layer without dilution, which significantly improves the contact between the chemicals and the fouling layer. In the operation of HR-MBRs, frequent draining of the organic suspension is not disadvantageous because the drained suspension can be immediately used for energy recovery (e.g., anaerobic digestion) and because it does not take a long time to start up the reactor.

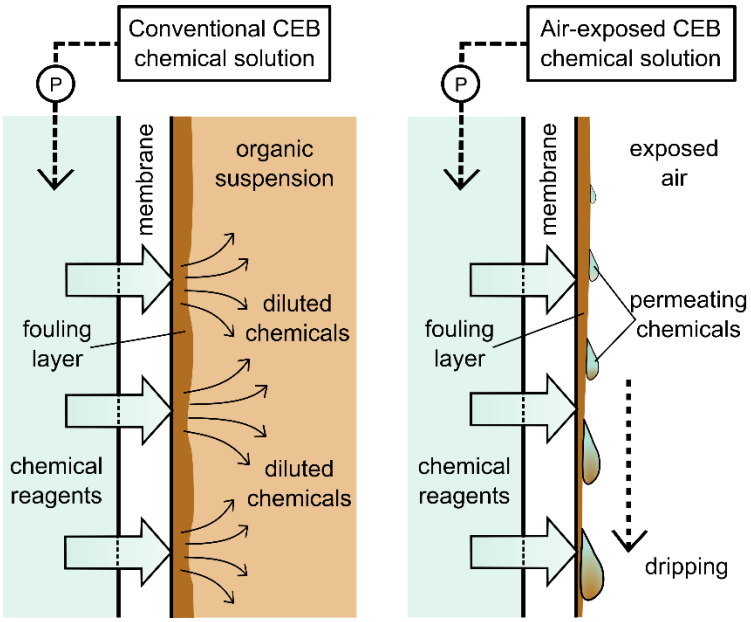


Figure 5. Representation of a conventional CEB (left) and the AECEB (right).

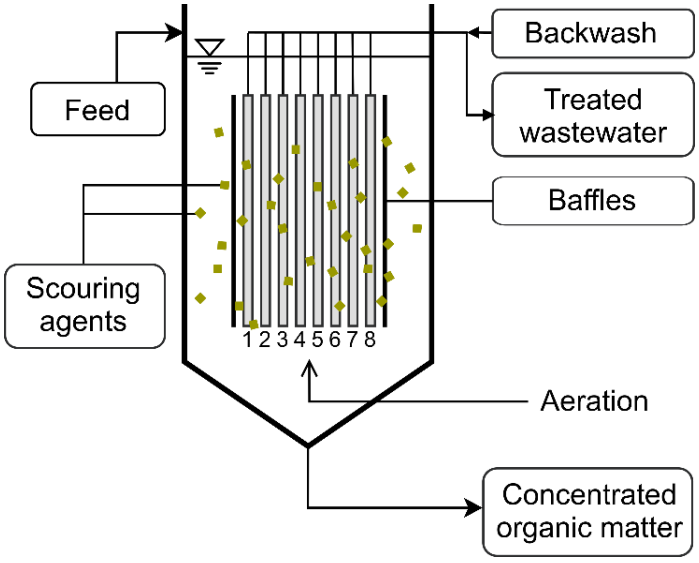


Figure 6. Schematic flow diagram of the experimental setup of the HR-MBR installed at Soseigawa WWTP.

3.3.2.2 Modification to the HR-MBR during implementation of the AECEB

In Chapter 5, four experimental runs (Runs 4-7) were conducted under identical operational conditions except for the manner of AECEB. Figure 6 shows the modified experimental setup used after the results of Chapter 4 (water jacket not shown). The effective tank volume of the HR-MBR was 7.3 L. Eight ceramic flat-sheet membranes (Meidensha, Japan) were inserted in the reactor. From the optimized fouling mitigation in Chapter 4, granular scouring was carried out in the subsequent chapters. Two different types of granular scouring agents were tested in this study. In Runs 4-6, polyethylene glycol (PEG) granules (BCN, Nisshinbo Chemical Inc., Tokyo, Japan) were used, whereas polyurethane (PU) granules (Organo, Tokyo, Japan) were used in Run 7. In Runs 4-6, PEG granules corresponding to 30% (v/v) of the tank volume were placed into the tank and were fluidized by aeration. In Run 7, probably due to the porous nature of the PU granules, 30% (v/v) of the PU granules could not be fluidized. Therefore, the volume of PU granules introduced was reduced to 20% (v/v). The system was operated under the conditions of SRT and HRT of 0.5 days and 0.8 hours, respectively. Instantaneous membrane flux was fixed at 20 LMH, resulting in a net flux of 16 LMH. Due to the addition of membrane plates, additional holes were added to the aerator tubes. To enable fluidization of granules, the aeration provided had to be increased to a fixed flow rate of 7.6 L/min from 4 L/min, resulting in a dissolved oxygen concentration in the reactor of 6.0±0.9 mg-O_2/L.

3.3.2.3 Permeability recovery and the optimization protocol

Permeability recovery achieved by a membrane fouling cleaning protocol is defined as follows:

$$Permeability\ Recovery\ (\%) = \left(\frac{R_f - R_b}{R_f - R_m}\right) \times 100, \quad (5)$$

where R_f is the total filtration resistance, R_b is the resistance remaining after the cleaning protocol and R_m is the intrinsic membrane resistance determined with the clean membranes before operation. Clean water permeability was measured for two membranes (taken from positions 3 and 6 in Figure 6) before and after AECEB was carried out in long-term operations

(Runs 5-7). By using a separate tank, it was possible to assess the cleaning efficiency without the impact of the surrounding organic suspension. The protocol of the measurement of clean water permeability mentioned above was also used in an attempt to determine the optimum conditions of AECEB, as described in the next section.

Table 1. Details of the AECEB examined in each run.

	Starting	**Details of the AECEB**		
	Condition	**Solutions**	**Flux**	**Duration**
Run 4 (AECEB-1)	When TMP reached 10 kPa	0.025 M HCl followed by distilled water	75 LMH	5 minutes with each solution
Run 5 (AECEB-2)	When TMP reached 30 kPa	5000 ppm NaClO followed by 0.1 M oxalic acid	25 LMH	30 minutes with each solution.
Runs 6 and 7 (AECEB-3)	When TMP reached 30 kPa	1000 mg/L oxalic acid followed by 1000 ppm NaClO	25-100 LMH [a]	5 minutes with each solution

a: In Runs 6 and 7, four cleaning cycles were repeated with different backwash fluxes. The CEB flux was increased gradually from 25 LMH to 100 LMH by 25 LMH intervals (i.e., 25, 50, 75 and 100 LMH). After the fourth CEB, hydraulic backwash was carried out (flux at 100 LMH and duration for 5 minutes) to remove any remaining chemical solution left in the membrane and tank.

3.3.2.4 Implementation of the AECEB for Fouling Control in HR-MBRs

As mentioned before, AECEB was performed after the organic suspension had been drained from the tank to increase the contact time between the CEB solution and the foulants. Table 1 summarizes the protocols of AECEB applied in Runs 4-7. A 3-mm screen installed at the bottom of the reactor prevented the loss of granular materials from the system during the AECEB events. The new feed was introduced to the reactor after it was confirmed by visual inspection that all chemical solutions, which had permeated through the membrane and subsequently dripped down, had exited from the reactor.

After Runs 4 and 5, optimization of the AECEB was attempted with fouled membranes prepared in separate operations in which the HR-MBR unit was operated under the same conditions as those outlined in Section 3.3.2.3. The membranes used for the optimization experiments were obtained when TMP reached 30 kPa. It was therefore assumed that the degrees of fouling of the membranes used for the optimization of AECEB were similar. Subsequently, the fouled membranes were cleaned by AECEB under a variety of conditions. Clean water permeability was measured before and after each examined backwash, following the methodology described in Section 3.2.2, and the permeability recovery efficiencies of each backwash were assessed.

The initial attempt to enhance the cleaning efficiency of the AECEB used in Run 4 (designated as AECEB-1), in which 0.025 M HCl was used, was to use multiple chemical reagents. Sequential CEB with different chemicals is believed to yield synergistic effects [68,99]. This AECEB, in which NaClO was followed by oxalic acid, was designated as AECEB-2 and was used in Run 5. After Run 5, further improvements were attempted to reduce the chemical agents used and address the problems in AECEB-2 (details described in Chapter 5). In the newly established protocol of AECEB, designated as AECEB-3 hereafter, CEB was repeated four times with variable fluxes and a fixed short duration of 5 minutes. The first cycle of AECEB-3 was initiated by acid backwashing (1000 mg/L of oxalic acid) under a flux of 25 LMH, which was followed by oxidant backwashing (1000 ppm of NaClO) under the same flux. After the first cycle of CEB using the two chemicals, three more CEB cycles were repeated with a gradual increase in backwashing flux in AECEB-3. In each step, the backwash flux was increased by 25 LMH. Namely, backwashing flux in the fourth cycle of AECEB-3 was 100 LMH. This style of AECEB was developed to reduce the amount of chemicals used and to address the issues of uneven distribution of chemicals inside the flat-sheet membrane elements, which was found in preliminary tests. AECEB-3 was completed by conducting a 5-minute hydraulic backwash at 100 LMH to rinse the leftover chemicals on/in the membranes after the fourth cycle using oxalic acid and NaClO.

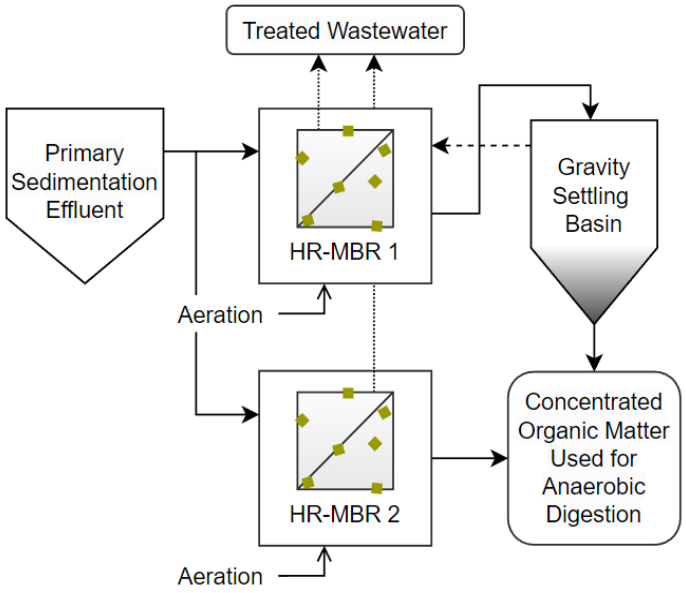


Figure 7. Schematic flow diagram of parallel HR-MBRs equipped with ceramic flat-sheet membranes utilized for organic matter recovery.

3.3.3 Chapter 6: Gravity Settling for Additional Concentration of the Recovered Organic Matter

Two HR-MBR reactors as described in Section 3.1.1 were operated in parallel (Figure 7). One of the reactors, HR-MBR 1, was operated with a separate 4 L gravity settling basin, which redirected the overflow back to the HR-MBR. Furthermore, half of the concentrated sludge was manually removed from HR-MBR 1 every 24 hours. Due to the inclusion of the settling basin, there were variations in solids retention time (SRT) and hydraulic retention time (HRT). Specifically, SRTs were 12 hours for both HR-MBR 1 and 2 and 48 hours for the settling basin. Meanwhile, HRTs were 1.2 hours for HR-MBR 1 combined with the settling basin, and 0.9 hours for HR-MBR 2. Before the start of the operation, the two systems were run for 7 days with the same conditions but different sets of membranes to prepare the mixed liquor.

3.4 WASTEWATER QUALITY

The effluent from the primary sedimentation basin of the wastewater treatment plant (Soseigawa Wastewater Treatment Plant, Sapporo City, Japan) was used as the feed in this study. Use of primary settling for energy conservation was found to be better than without in activated sludge processes [40]. The average wastewater properties of the influent for each of the experimental sections are shown in Tables 2, 3 and 4 for Chapters 4, 5 and 6, respectively. Analysis was carried out for daily 24-hour composite samples. Grab sampling of primary sedimentation influent after Chapter 4 was conducted to ascertain the total recovery from raw wastewater.

Table 2. Average characteristics of the wastewater (primary sedimentation effluent) fed into the HR-MBR in the experiments conducted in Chapter 4.

Effluent of primary sedimentation (feed)					
pH		7.1	±	0.2	
Temperature	T	19.6	±	3.2	(°C)
Total chemical oxygen demand	CODt	226	±	50	(mg/L)
Total organic carbon	TOC	42.4	±	11.4	(mg/L)
Total phosphorus	T-P	5.6	±	1.9	(mg/L)
Total nitrogen	T-N	20.8	±	4.4	(mg/L)

Table 3. Average wastewater characteristics of the influent and effluent from the primary sedimentation basin during experiments of Chapter 5.

Influent of primary sedimentation					
Total chemical oxygen demand	CODt	464	±	215	(mg/L)
Effluent of primary sedimentation (feed)					
pH		7.09	±	0.11	
Temperature	T	12.6	±	1.2	(°C)
Total chemical oxygen demand	CODt	239	±	65	(mg/L)
Soluble chemical oxygen demand	CODs	60	±	27	(mg/L)
Total organic carbon	TOC	90.5	±	13.4	(mg/L)
Dissolved organic carbon	DOC	51.3	±	8.7	(mg/L)
Total nitrogen	TN	19.7	±	3.9	(mg/L)
Dissolved nitrogen	DN	14.9	±	3.3	(mg/L)

Table 4. Average wastewater characteristics of the influent and effluent from the primary sedimentation basin during experiments of Chapter 6.

Influent of primary sedimentation					
Total chemical oxygen demand	CODt	306	±	86	(mg/L)
Soluble chemical oxygen demand	CODs	97	±	17	(mg/L)
Effluent of primary sedimentation (feed)					
pH		7.0	±	0.1	
Temperature	T	14.9	±	2.1	(°C)
Total chemical oxygen demand	CODt	181	±	37	(mg/L)
Soluble chemical oxygen demand	CODs	67	±	16	(mg/L)

3.5 FRACTIONATION OF SAMPLES AND BIOFLOCCULATION ANALYSIS

The quantity of organic matter adsorbed onto sludge flocs (i.e., bioflocculation) was also assessed in this study. The method used for this assessment was similar to other organic matter redirection studies [22,36], where the difference in particulate COD between the influent and the concentrate was regarded as adsorption of colloidal and dissolved COD onto the flocs. COD in the tank was fractionated into particulate, colloidal and soluble fractions in the following manner. Mixed liquor samples were centrifuged at 4000 rpm for 5 min (CT-6E, Hitachi, Tokyo, Japan) and supernatants were obtained. The soluble fraction was obtained by filtering the supernatants through 0.45 μm PTFE membranes (Advantec, Tokyo, Japan). The particulate COD fraction was determined by the difference between the total COD and the supernatant COD, whereas the colloidal COD fraction was determined by the difference between the supernatant COD and the soluble COD.

3.6 MASS BALANCE ANALYSIS

3.6.1 Mass Balance Calculation

Mass balance of organic matter around the HR-MBR was assessed based on the COD measurements of the influent wastewater (considered the total incoming mass), the effluent, the accumulation in the reactor and the concentrated organic matter. Sampling was minimized as much as possible and any difference between the total influent mass and the rest of the parameters was considered as losses. The daily mass calculation was based on the following equation:

$$M_I - M_S = M_C + M_P + M_A + M_L \quad (6)$$

where M_I, M_S, M_C, M_P, M_A, and M_L are the daily mass rates of the influent wastewater, samples, concentrate, permeate, accumulation and loss, respectively. Each of which is calculated from the following equations:

$$M_I = (COD_I \cdot Q_I), \tag{7}$$

$$M_C = (COD_C \cdot Q_C), \tag{8}$$

$$M_P = (COD_P \cdot Q_P), \tag{9}$$

$$M_A = \frac{\left(COD_{ML}^{f} - COD_{ML}^{i}\right)V_R}{t} \tag{10}$$

$$M_S = \frac{COD_{ML}^{f} \cdot V_S}{t}, \tag{11}$$

where COD_I, COD_C, COD_P, COD_{ML} refer to COD concentrations in the influent, concentrate, permeate and mixed liquor, respectively. Superscripts "*i*" and "*f*" refer to the initial and final concentrations of the mixed liquor during the period t between samplings. Q_I, Q_C and Q_P refer to the flow rates of the influent wastewater, concentrate and permeate, respectively. V_s and V_R refer to the volumes of the samples and the reactor, respectively.

3.6.2 Accumulated Mass Balance

After the initial calculation of the masses, each total mass was then multiplied by a 24-hour period to obtain daily masses. The daily percentage of total mass refers to the ratio of mass accumulated (i.e., continuously added) up until that point, described by the following equation:

$$Accumulated\ Mass\ (\%) = \frac{\sum_{n=0}^{N} M_{x_n}}{\sum_{n=0}^{N} M_{I_n}} \times 100, \tag{12}$$

where M_x refers to the different masses calculated in Equations 6-11 and N refers to the day where the accumulated mass is being calculated.

3.7 ANALYTICAL METHODS

3.7.1 Carbon and organic matter measurement techniques

Measurements of total organic carbon (TOC) were carried out using a TOC analyzer (TOC-L, Shimadzu, Kyoto, Japan). A commercially available analytical kit (HACH, Colorado, United States, Method 8000) was used for chemical oxygen demand (COD) measurements. Amounts of suspended solids (SS) and volatile suspended solids (VSS) were determined using standard methods [100].

3.7.2 Extracellular polymeric substance (EPS) extraction

EPS extraction was conducted through a modified protocol of Frølund et al. (1996) utilizing cation exchange resin. In short, 18.5 g of cation exchange resin per g-VSS is mixed with phosphate buffer for 1 hour at 300 rpm and 4°C. Next, the supernatant is removed from the sample through centrifugation at 4,000 rpm and 4°C for 15 minutes and is replaced with phosphate buffer. The buffer is removed from the washed exchange resin and the sample solution is mixed in for 1 hour at 300 rpm and 4°C. Finally, the solution is centrifuged at 10,000 rpm at 4°C for 1 minute to remove the resin and for 15 minutes to remove all solid particles (twice in different containers).

3.7.3 Fourier transform infrared (FTIR) spectroscopy analysis

A Fourier transform infrared (FTIR) spectrophotometer (IR-7500S, Shimadzu, Kyoto, Japan) equipped with an attenuated reflection (ATR) accessory (Quest ATR, Specac, UK) was used to obtain IR spectra of samples. FTIR spectra were acquired from wavenumbers from 800 to 4000 cm^{-1} with 2 cm^{-1} spectral resolution. Spectral data were processed using LabSolutions IR software (Shimadzu, Kyoto, Japan).

Suspended solids in the HR-MBR and fouling layers scraped from the fouled membranes were subject to FTIR analysis. To collect samples from the fouling layer, first, a delicate flow of distilled water was used to remove loose debris and a new polyurethane brush was used to scrape the fouling layer from the membrane surface. For the suspended solids in the HR-MBR, samples were collected as grab samples directly from the reactor. The samples were frozen and freeze-dried (FDU-2100, Eyela, Japan) before FTIR analysis. FTIR analysis was also

carried out for the PEG standard (laboratory grade 8000 MW PEG powder, MP Biomedicals, USA).

3.7.4 Dissolved organic matter analysis

Liquid chromatography with organic carbon detection (LC-OCD) with a modified dual column setup (Tominaga et al., 2022) was used to determine the size distribution of the molecules (Model 8, DOC-LABOR, Karlsruhe, Germany). The dissolved organic matter samples were obtained by filtering the supernatants through 0.45 µm PTFE membranes (Advantec, Tokyo, Japan).

3.7.5 Scanning Electron Microscopy

To observe the fouling layer on the surface and cross-section of the membranes, we utilized scanning electron microscopy (SEM) (S-4000 instrument, Hitachi, Tokyo, Japan). The preparation method used was shown in a previous study [102]. Prior to SEM observation of the membrane surfaces, the samples were dried (EM-CPD300, Leica microsystems, Wetzlar, Germany) and coated with platinum (JFC-1200, JEOL, Tokyo, Japan).

CHAPTER 4.
RESULTS AND DISCUSSION OF THE IMPLEMENTATION OF THE HR-MBR BASED ON ESTABLISHED FOULING MITIGATION TECHNIQUES

4.1 INDIVIDUAL EFFECTS OF FOULING MITIGATION TECHNIQUES (RUN1)

Run 1 was the initial implementations of the granular materials and of the selected CEB protocol. Although TMP increased very rapidly and became uncontrollable within several hours in the control experiments in which either granular scouring or CEB was omitted (Figure 8), each of the fouling mitigation techniques had some initial effect on the fouling. The HR-MBR which utilized granular materials had better fouling control compared with the one with only CEB. However, compared with the following runs which utilized both fouling mitigation techniques, both operations from Run 1 were severely lacking.

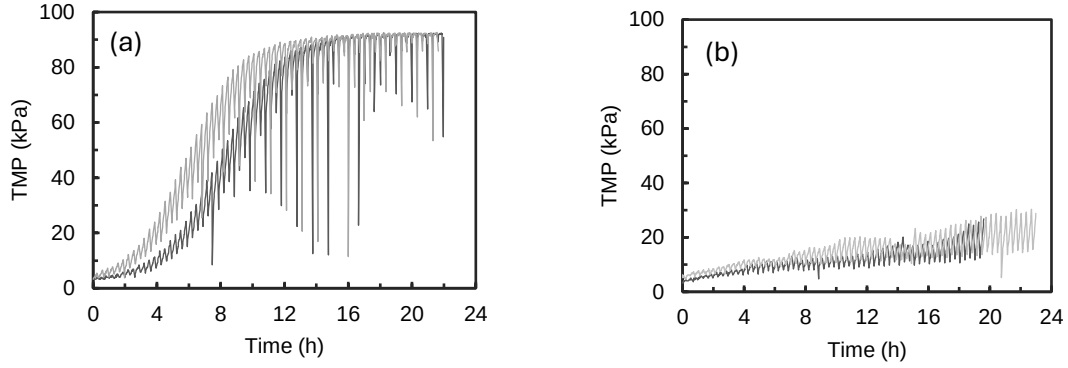


Figure 8. The transmembrane pressure (TMP) evolution during the operation of Runs (a) 1.1 with only CEB and (b) 1.2 with only granular scouring.

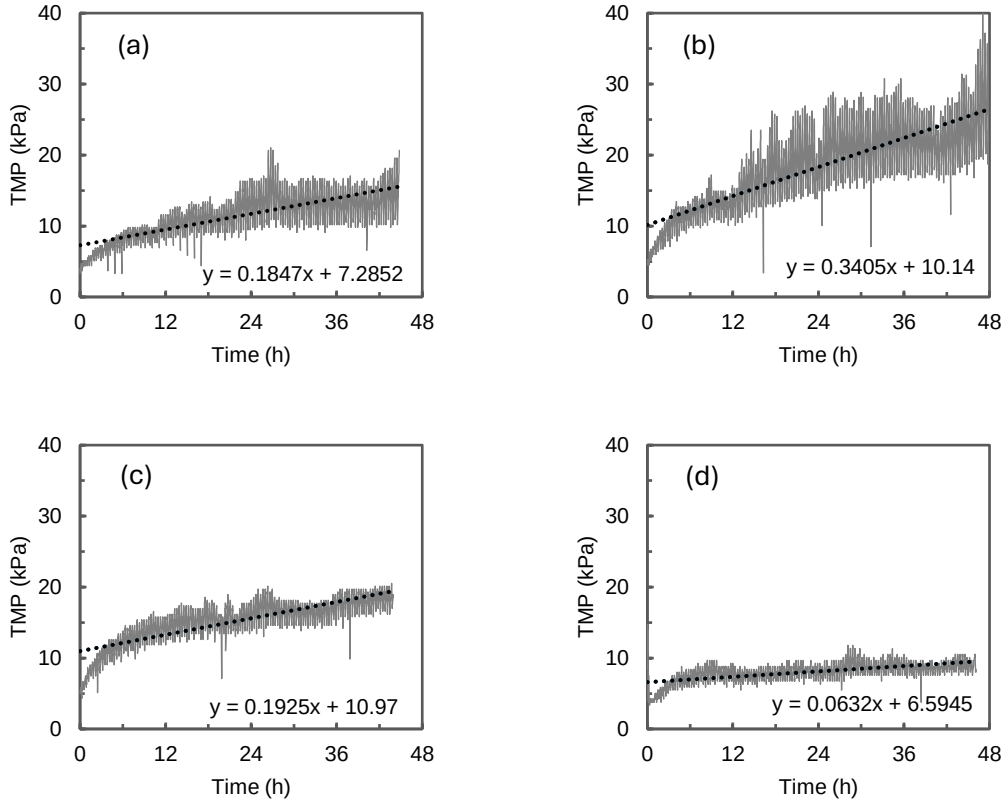


Figure 9. TMP evolution in Runs (a) 2.1, (b) 2.2, (c) 2.3 and (d) 2.4, in which the concentrations of NaClO in the backwash solution were set at 1000 ppm, 500 ppm, 100 ppm and 50 ppm, respectively. In these runs, granular scouring was also carried out.

4.2 EFFECTIVENESS OF THE COMBINATION OF GRANULAR SCOURING AND CEB WITH VARIOUS CONCENTRATION OF SODIUM HYPOCHLORITE (RUN 2)

4.2.1 Fouling evolution in Run 2

In Run 2, the effectiveness of the combination of granular scouring and CEB for fouling control in the HR-MBR was examined with various CEB intensities (various NaClO concentrations in the backwash solution). Figure 9 shows the TMP increases in Run 2 where both control strategies were used at the same time. The two control strategies could work in

different ways: granular scouring and CEB were effective for removing foulants on the membrane surface and inside the micropores, respectively. Therefore, that combination could work complimentarily. The slope of the linear regression line (dotted lines in Figure 9) of the TMP increase shows a rough estimation of the fouling rate in kPa per day. Fouling rates in Runs 2.1, 2.2, 2.3 and 2.4, calculated from the equation of the linear regression, were 4.43 kPa/d, 8.17 kPa/d, 4.62 kPa/d and 1.52 kPa/d, respectively. Interestingly, the lowest concentration of NaClO in the CEB tested, 50 ppm, exhibited the lowest fouling rate. Conventional MBRs treating municipal wastewater are normally operated at a fouling rate of less than 2 kPa/day [51,103]. It should be noted that the HR-MBR in this study was operated at a practical net flux of 20 LMH [83,104]. Therefore, the fouling rate of 1.52 kPa/d observed in Run 2.4 suggests that long-term operation of an HR-MBR is feasible, assuming that there is still plenty of room for further optimization of operational parameters or fouling mitigation techniques.

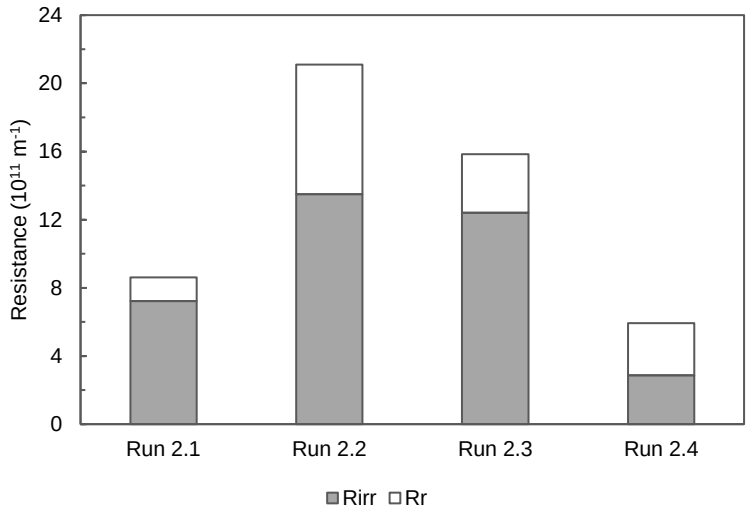


Figure 10. Distribution of fouling resistances assessed at the termination of Runs 2.1, 2.2, 2.3 and 2.4.

4.2.2 Analysis of fouling Resistance in Run 2

Figure 10 shows the distribution of fouling resistances assessed at the termination of Runs 2.1, 2.2, 2.3 and 2.4. The resistance occurring inside the membrane pores (i.e., irreversible fouling) was dominant in these runs except for Run 2.4. As mentioned before, the evolution of fouling was effectively controlled in Run 2.4, in which the lowest CEB intensity (50 ppm NaClO) was applied. This was largely attributed to the slowest evolution of irreversible fouling as shown in Figure 10. Kimura et al. [73] showed that CEB using NaClO could induce the release of dissolved organic matter (DOM) from biomass in an aerobic MBR and that the released DOM caused fouling in ceramic membranes submerged in the MBR [73]. Similarly, Sun et al. [105] reported that intensive in-line membrane cleaning in an MBR using ozonation induced the re-lease of DOM from the mixed liquor suspension, which accelerated fouling of a crossflow microfiltration module [105]. It is thought that CEB with a high intensity (more than 100 ppm NaClO in the backwashing solution) induces release of DOM from biomass in the HR-MBR, leading to the evolution of irreversible fouling. However, when the highest CEB (1000 ppm NaClO in Run 2.1) was examined, the degree of irreversible fouling was considerably lower. A possible explanation for this is that the high cleaning efficiency with 1000 ppm NaClO could offset the release of DOM from biomass to some extent. The degree of irreversible fouling with 1000 ppm NaClO was still higher than that with 50 ppm NaClO. Based on these observations, the lowest CEB concentration of 50 ppm was used in the subsequent test (Run 3). It should be noted that the degree of reversible fouling was the lowest with 1000 ppm NaClO. A sufficiently high CEB intensity may reduce the degree of reversible fouling, although economic feasibility and the evolution of irreversible fouling are potential problems. This point should be investigated in the future.

4.2.3 Bioflocculation in Run 2

When bioflocculation occurs in HR-MBRs, adsorption of the soluble and colloidal organic matter onto the particulate fraction is promoted and the soluble and colloidal COD fractions in a mixed liquor suspension are subsequently reduced. Faust et al. [23] suggested that bioflocculation plays an important role in the maintenance of the high filterability of the mixed liquor in HR-MBRs [23]. Bioflocculation in HR-MBRs should, therefore, be maximized

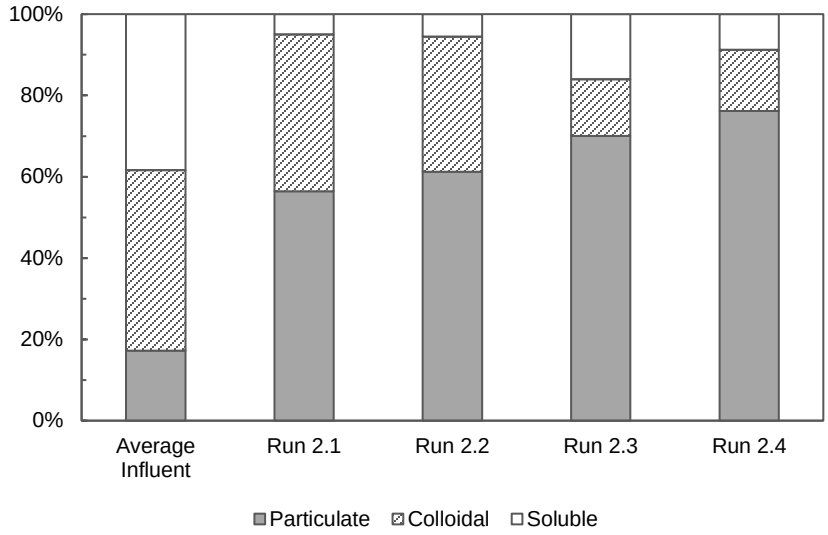


Figure 11. Size distributions (particulate, colloidal and soluble) of COD in the biomass suspension just before termination of Runs 2.1 to 2.4.

to carry out stable operation. Figure 11 shows the size distribution of organic matter (expressed as COD) in the HR-MBR in Run 2. The assessment, the results of which are shown in Figure 11, was carried out by the method used in previous studies [22]. Figure 11 shows that the presence of colloidal COD was substantial in Runs 2.1 and 2.2, indicating that bioflocculation was poor in those runs. High concentrations of NaClO in the CEB solution used in Runs 2.1 and 2.2 probably inhibited efficient bioflocculation by suppressing microbial activity. Sun et al. (2021) observed a reduction in the average particle size (i.e., deflocculation) after the implementation of CEB using NaClO in a conventional MBR (SRT of 30 days), which led to deterioration in the filterability of the mixed liquor [72]. The high concentration of colloidal COD could also explain the rapid evolution of fouling in Runs 2.1 and 2.2. It has been reported that the presence of colloidal organic matter in MBRs is closely related to the evolution of membrane fouling [106,107] and the reduced filterability of the mixed liquor [108]. In contrast, when the CEB intensity in an HR-MBR was reduced, bioflocculation could be promoted and high filterability of the mixed liquor was maintained. Enhanced bioflocculation is also beneficial as it increases the amount of carbon that can be utilized for biogas generation.

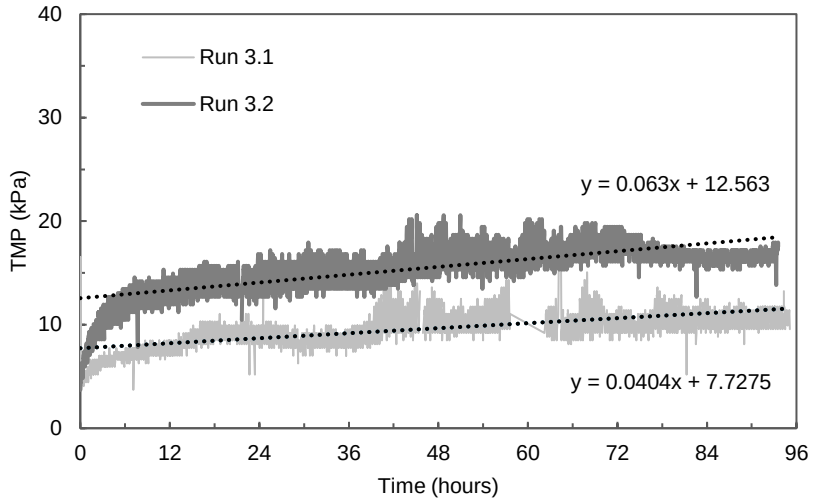


Figure 12. Increases in TMP in Runs 3.1 and 3.2. The dotted lines represent the linear regression of TMP evolutions. During Run 3.1, at around 2.5 days of operation, a power outage that lasted 4 hours stopped all activity in the experimental system.

Thus, the importance of a balance between the maintenance of bioflocculation and the cleaning efficiency of CEB for fouling control in HR-MBRs was demonstrated.

4.3 LONGER OPERATION OF THE HR-MBR (RUN 3)

4.3.1 Fouling evolution in Run 3

As stated in the previous section, it was demonstrated that the combination of granular scouring and CEB could be effective for the control of membrane fouling in the HR-MBR. It was also shown that the concentration of NaClO in the CEB solution had a significant impact on the evolution of fouling in the HR-MBR. Based on the results obtained in Run 2, long-term operations of the HR-MBR were attempted (Run 3). The concentration of NaClO in the CEB solution was set at 50 ppm based on the results of Run 2. Figure 12 shows the increases in TMP observed in Run 3. Operations lasting for 4 days were carried out twice (Run 3.1 and 3.2) under the same operational conditions as those tested in Run 2.4. In Runs 3.1 and 3.2, increases in TMP were relatively rapid during the first 5 h of the operation. It was possible

that the rapid increase at the beginning of operation was due to the absence of seeding. However, even with seeding in conventional MBRs, high fouling at startup has been observed [109]. Still more investigation is needed to ascertain the reason why this initial fouling occurred.

After that, the rate of TMP increase became slower, and the operation could be carried out stably over a period of four days. Fouling rates during the initial four days of operation in Runs 3.1 and 3.2 were 1.0 kPa/d and 1.5 kPa/d, respectively. Fouling rates in Runs 3.1 and 3.2 were slightly different. This was attributed to the difference in the quality of the influent wastewater. It should be noted that real municipal wastewater was used in this study: variation in the influent wastewater was uncontrollable.

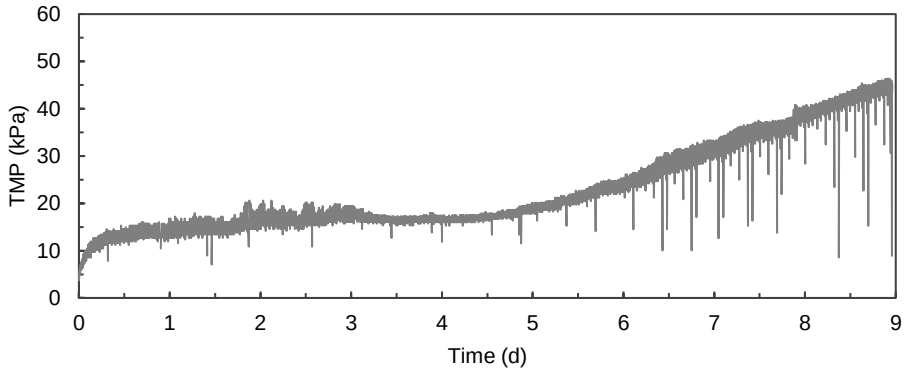


Figure 13. TMP evolution of the extended operation of Run 3.2.

Figure 13 shows the TMP evolution during Run 3.2, which was terminated when the TMP reached 40 kPa and the constant flux could not be maintained. With the fouling mitigation techniques and the operational conditions used, operation could be conducted for 9 consecutive days without off-line cleaning. Although the initial operation period could be conducted with low fouling rate, at around 5 days, the TMP evolution started to rise at a steady state. This rise occurred when the fouling mitigation techniques could no longer control fouling and irreversible fouling started to accumulate. It was hypothesized that if it was

possible to completely remove fouling with an on-line cleaning on day 4 or 5, operation could be restarted with minimal loss of efficiency.

4.3.2 Carbon recovery and effluent quality achieved by the HR-MBR

By conducting longer-term operations (Run 3), it became possible to assess the carbon recovery achieved by the HR-MBR. On average, 58.9% of the COD in the primary sedimentation effluent could be recovered by the HR-MBR (see Figure S1 in Supporting Material). According to the annual report from the WWTP where this study was carried out, the primary treatment removed one-third of organic matter in the influent to the sedimentation basin. Thus, in total, 73.3% of organic matter in the raw wastewater could be recovered by combining primary sedimentation and the HR-MBR, and it could be used for methane generation. As discussed in the subsequent section, the carbon recovery achieved by the HR-MBR in this study was improved compared to that of previous related studies by implementing the primary sedimentation tank.

In Run 2, the average COD concentration in the permeate was 18.8 mg/L. About 95% of the COD in the influent to the WWTP was removed in the permeate of the HR-MBR. This removal efficiency is comparable to that reported for conventional MBRs (SRT of 15–60 days) treating municipal wastewater [63,83], indicating that the frequent CEB carried out in this study had a limited impact on microbial performance. The quality of the effluent obtained in this study with the HR-MBR was much higher than that found with other competitive technology for carbon recovery from municipal wastewater. It has been reported that HRAS systems (A-stage systems) can achieve a COD removal rate of 52–64% [38,110]. As another emerging technology for carbon capture, direct membrane filtration (DMF) (in which no biological treatment is pro-moted) could not produce treated water with a low concentration of COD (<30 mg/L) without using coagulation [111].

CHAPTER 5.
RESULTS AND DISCUSSION OF THE EFFECTIVE RESET OF FOULING IN A HR-MBR BY IN SITU, AIR EXPOSED AND CHEMICALLY ENHANCED BACKWASH

5.1 RUN 4 (IMPLEMENTATION OF AECEB-1 WITH A SHORT INTERVAL)

Figure 14 illustrates the TMP evolution in Run 4. Implementations of the AECEB are indicated by arrows in Figure 14. As clearly shown in Figure 14, the operation of the HR-MBR was very stable in Run 4, in which AECEB-1 was performed when TMP reached 10 kPa. Analysis of the COD mass balance around the HR-MBR (Figure S2) showed that about 60% of the organic matter in the feed (effluent from the primary sedimentation basin) was recovered as the concentrate in Run 1. Combined with the organic matter settled in the

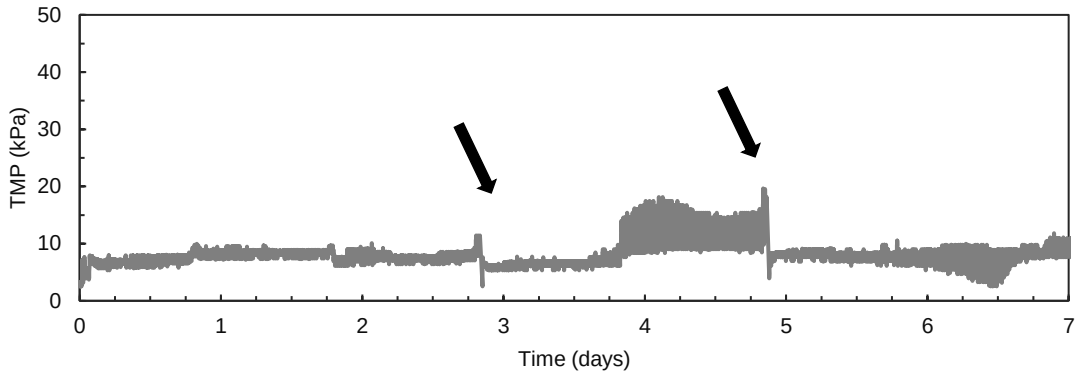


Figure 14. Increase in TMP in Run 4. The arrow indicates the implementation of AECEB-1.

primary sedimentation basin, 70-75% of the organic matter in the raw wastewater could be recovered in Run 4, and that percentage is comparable to the percentages in previous related studies [29,32]. Thus, it was confirmed that implementation of AECEB did not negatively affect the recovery of organic matter by the HR-MBR. The concentration of COD in the permeate observed in Run 4 was 45±3 mg/L, corresponding to 90% removal of COD in the raw wastewater (See Table 3). Thus, both sufficient recovery of organic matter and sufficient reduction of COD level in the treated water were achieved in Run 1.

It should be noted that a significant portion (approximately 25%) of organic matter in the feed to the HR-MBR was lost due to mineralization. The aeration rate of 7.6 L/min set in the modified HR-MBR reactor might be too high, resulting in a dissolved oxygen (DO) concentration of around 6 mg-O_2/L in the tank. However, when the aeration rate was reduced, fluidization of the granules became unstable with the experimental apparatus used in this study. There are significant contradictions in previous studies concerning the impacts of DO concentration on recovery of organic matter in high-rate processes. Faust et al. (Faust et al., 2014b) reported that higher DO resulted in better bioflocculation efficiency and Wan et al. [32] also found higher organic recovery in HR-MBRs with higher DO. In contrast, He et al. [29] demonstrated that a low DO concentration in an HR-MBR combined with chemical coagulation significantly enhanced the recovery of organic matter. More investigation on the impact of DO on the recovery of organic matter by HR-MBRs is needed.

5.2 FIRST ATTEMPT TO OPTIMIZE THE AECEB

As described in the previous section, the AECEB was found to be effective for long-term operation of the HR-MBR. However, frequent implementation of AECEB (e.g., every 2-3 days) may reduce the feasibility of the HR-MBR. Intervals between AECEB events should be expanded as long as possible. This means that AECEB should be effective for more severely fouled membranes than those experienced in Run 4. Preliminary tests showed that AECEB-1 could not restore the permeability sufficiently when TMP reached 30 kPa. Therefore, we attempted to further optimize the AECEB by the protocol described in Section 3.3.2.3.

Table 5. Preliminary air-exposed CEB optimization tests for different solutions.

Solution	Concentration	Flux (LMH)	Duration (min)	Permeability Recovery (%)
NaClO	5000 ppm	100	30	93
Oxalic acid	0.1 M	100	30	60
HCl	0.025 M	100	30	63
NaOH	0.1 M	100	30	62
H_2O_2	5000 ppm	100	30	47

Table 6. Recoveries of the permeability achieved by different conditions of AECEB using NaClO.

NaClO concentration (ppm)	Backwash flux (LMH)	Duration (minutes)	Permeability recovery (%)	Chemicals delivered to the membrane (g-NaClO/m^2)
5000	100	30	93	250
5000	50	30	93	125
5000	25	30	87	62.5
5000	100	15	87	125
2500	100	30	86	125
1250	50	30	81	31.3
5000	50	15	79	62.5
1250	25	30	72	15.6
5000	100	7.5	70	62.5
2500	50	15	68	31.3
1250	100	30	60	62.5
1250	25	15	56	7.8
1250	100	7.5	41	15.6
2500	50	7.5	35	15.6
1250	25	7.5	21	3.9

First, performances of various chemicals were assessed with similarly fouled membranes, which were prepared in the manner described in Section 3.3.2.4. Table 5 summarizes the preliminary comparison among chemicals commonly used for backwashing. Oxalic acid, hydrochloric acid (HCl) and sodium hydroxide (NaOH) exhibited similar recoveries of the permeability (around 60%). Sodium hypochlorite (NaClO) exhibited much better performance (above 90% permeability recovery) in these tests. Therefore, NaClO was used as the primary CEB solution in the subsequent tests.

By using NaClO, the optimum concentration, optimum duration, and optimum flux for the AECEB using NaClO were investigated in the manner described in Section 3.3.2.3. Table 6 shows a summary of the results of these tests. As expected, higher concentrations and longer durations resulted in high recoveries of the permeability. The best result (93% permeability recovery) was obtained with the combinations of 5000 ppm/ 100 LMH/ 30 minutes and 5000 ppm/ 50 LMH/ 30 minutes. In terms of the amount of NaClO used for a specific membrane area, the former and the latter combinations were equivalent to 250 g-NaClO/m^2 and 125 g-NaClO/ m^2, respectively. The combination of 5000 ppm/ 25 LMH/ 30 minutes using 62.5 g-NaClO/m^2 also resulted in a relatively high permeability recovery of 87%. Based on these results, the combination of 5000 ppm/ 25 LMH/ 30 minutes was used as the primary condition in the subsequent optimization experiments.

It is widely known that different chemical reagents remove different types of constituents from fouled membranes. NaClO is good at removing organic matter from fouled membranes [113] but is not good at removing inorganic matter. The results of our previous study suggested that inorganic matter, primarily iron [114], also played an important role in the evolution of fouling in the HR-MBR. Acid is known to be effective for removing inorganic matter from fouled membranes [65,68]. Thus, it was expected that further improvements would occur if acid was also used with NaClO in AECEB. Oxalic acid was used since previous studies in which similar membranes were used showed that it exhibited good cleaning performance [65,69,113]. After implementing AECEB with NaClO (5000 ppm/ 25 LMH/ 30 minutes), an additional AECEB using oxalic acid (0.1 M/ 25 LMH/ 30 minutes) was also carried out for the fouled membrane prepared in the manner described in Section 3.3.2.3. This combination exhibited an excellent cleaning efficiency: 97.1% recovery of the permeability was observed. Based on these results, we decided to carry out AECEB with these conditions,

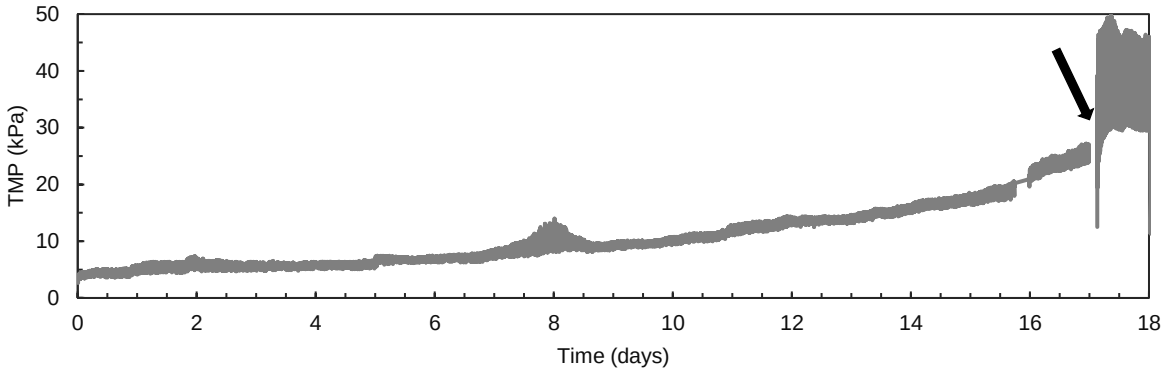


Figure 15. TMP evolution in Run 5. The arrow indicates the implementation of AECEB-2.

designated as AECEB-2 (Table 1), in the subsequent long-term operation of the HR-MBR (Run 5).

5.3 APPLICATION OF THE SEMI-OPTIMIZED AECEB-2 IN A LONG-TERM OPERATION (RUN 5)

Figure 15 illustrates the TMP evolution in Run 5, in which AECEB-2 was conducted when TMP reached 30 kPa. Operation of the HR-MBR could be continued stably for 17 days. On day 17, AECEB was carried out. Recovery of clean water permeability achieved by AECEB-2 in Run 5 was separately assessed and was found to be 91% (Figure S3). Thus, it was confirmed that AECEB-2 could sufficiently cancel the fouling in Run 5. However, when the operation of the HR-MBR was restarted, such high efficiency in the cleaning was not reflected in the TMP profile: TMP increased very rapidly, and the operation quickly became unstable. The value of TMP exceeded 40 kPa within a single filtration cycle (i.e., less than 10 minutes).

A notable drop in pH was observed when the feed was introduced to the tank after the implementation of AECEB-2. The value of pH in the tank was 7.1±0.5 before implementation of AECEB-2, whereas it dropped to 1.9 when the new feed was introduced. Although the value of pH in the tank became normal (7.5) after 24 hours of operation, the TMP increase was still uncontrollable. It has been reported that low pH in biological treatment processes can induce

cell lysis [115], which might cause the sudden increase in TMP after AECEB-2 in Run 5. Also, it was reported that contact of highly concentrated NaClO with biomass suspension in MBRs induced the release of constituents with high fouling potential [74]. It was hypothesized that the chemicals with high concentrations used for AECEB-2 remained in the tank, which eventually led to the rapid increase in TMP in Run 5.

5.4 APPLICATION OF THE OPTIMIZED AECEB-3 IN A LONG-TERM OPERATION (RUN 6)

The experience in Run 5 suggested that the chemical solutions with high concentrations used in AECEB-2 were the cause of the rapid increase in TMP. It was therefore desirable to reduce the concentrations of the chemicals in the AECEB. However, merely reducing the concentrations of chemicals would result in less efficient cleaning, as was explored previously (Table 5). We carried out additional optimization tests and could modify the AECEB-2 (Details can be found in the supplementary materials Section S3.). Although the improved AECEB-3 (Table 1) was carried out with lower chemical consumption, recovery of clean water permeability achieved by the improved AECEB-3 was comparable to that by AECEB-2. The next long-term operation (Run 6) was carried out with AECEB-3, which was performed when TMP reached 30 kPa.

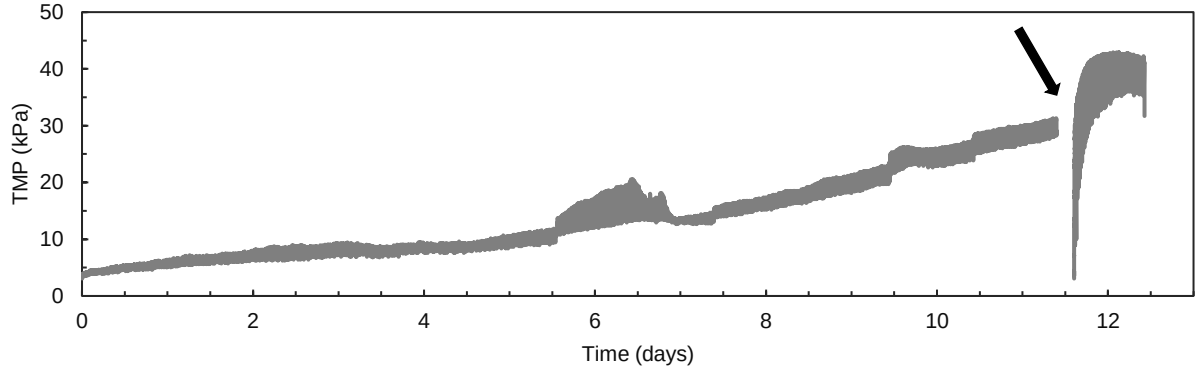


Figure 16. TMP evolution in Run 6. The arrow indicates the implementation of AECEB-3.

Figure 16 shows the increase in TMP in Run 6. It took 11 days before TMP reached 30 kPa in Run 6, and AECEB-3 was then performed. In a separate assessment of clean water permeability, it was found that AECEB-3 recovered 98% (Figure S3) of permeability. It should be noted that the amount of chemicals used in AECEB-3 was considerably reduced compared with that in AECEB-2: the amounts of NaClO used in AECEB-2 and AECEB-3 were 62.5 g/m^2 and 20.8 g/m^2, respectively, while those of oxalic acid were 112.5 g/m^2 and 20.8 g/m^2, respectively. Nevertheless, permeability recovery achieved by AECEB-3 was excellent.

As mentioned in the previous section, a notable pH drop after AECEB-2 might have induced the rapid increase in TMP in Run 5. It was confirmed that the value of pH was 7.0 when the operation of the HR-MBR was restarted after AECEB-3: the countermeasures taken to address the pH drop were found to be effective. Nevertheless, as shown in Figure 16, TMP increased very rapidly after AECEB-3. The result observed in Run 5 was reproduced in Run 6.

To identify the constituents causing the severe fouling after the implementation of AECEB-3 in Run 6, the fouling layer was collected from the membrane surface one day after resuming the operation and was subject to FTIR analysis. Figure 17 shows FTIR spectra obtained for (A) the fouling layer, suspended solids in the reactor collected (B) before and (C) after AECEB-3 and (D) the PEG standard. The FTIR spectrum of PEG, the base material of the scouring agents used in Runs 4-6, was similar to that reported previously [116,117]. Intense sharp peaks were observed at the wavenumbers of 2872 cm^{-1} and 1089 cm^{-1}, which are characteristic for this compound and highly distinguished from other biopolymers [116].

The FTIR spectrum of the fouling layer was considerably different from that of the suspended solids in the reactor collected before the implementation of AECEB-3. Rather, a high similarity was recognized between the FTIR spectrum of the fouling layer and that of the PEG. The FTIR spectrum of the suspended solids collected just after the implementation of AECEB-3 also had features similar to those of the spectrum of the PEG standard, which somewhat diminished after 24 hours. These results suggest that the PEG granules used for granular scouring were degraded by the chemicals used for AECEB-3, and PEG fragments were released from the granules. A part of the PEG fragments remained in the tank after AECEB-3 and subsequently caused the severe fouling when the operation was resumed. It is known that PEG has strong gelling properties [118,119]. These PEG fragments generated by

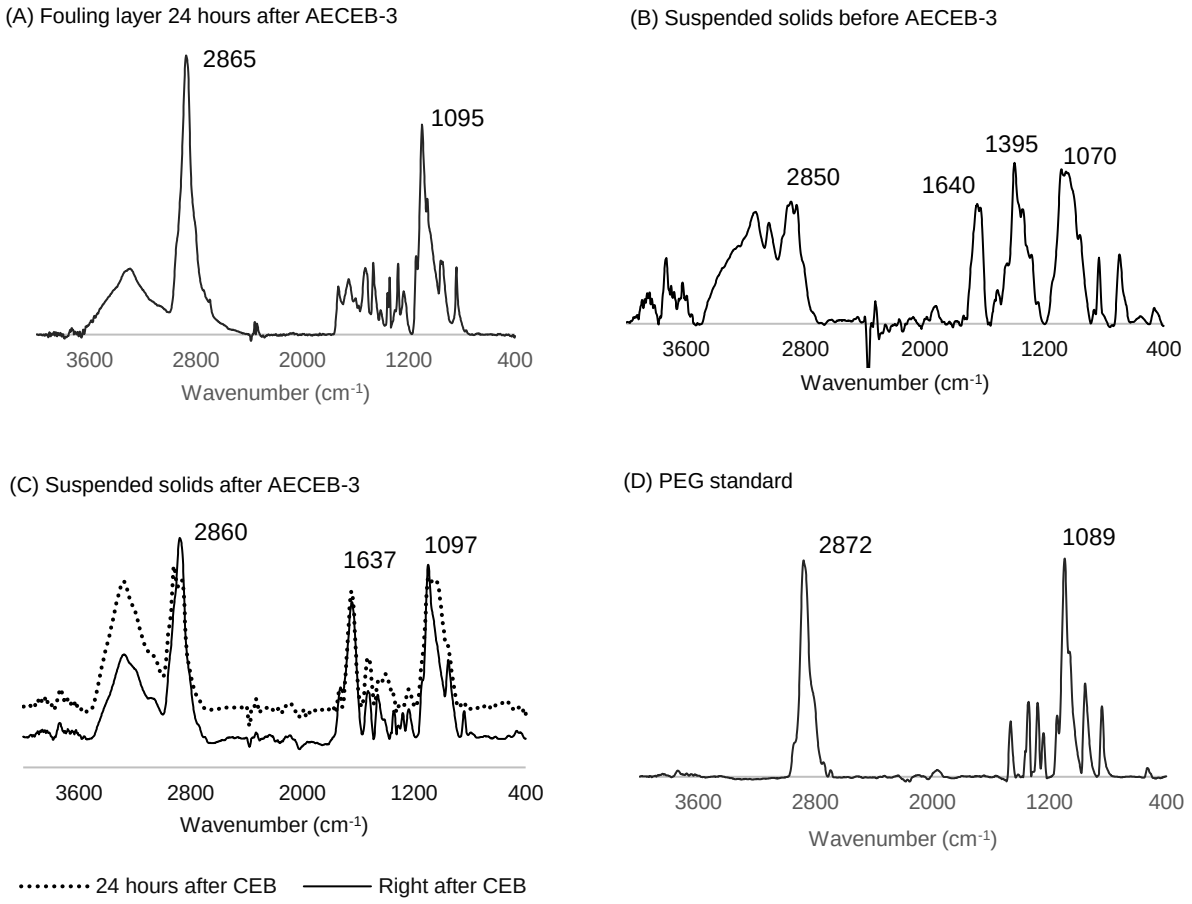


Figure 17. FTIR spectra of samples from Run 6 of the fouling layer (A) and the suspended solids before (B) and after (C) the AECEB compared with the spectrum of the PEG standard (D).

AECEB-3 might be involved in the formation of the gel on the membrane surface, which caused the surge of TMP in Runs 5 and 6.

5.5 LONG-TERM OPERATION WITH POLYURETHANE GRANULAR AGENTS (RUN 7)

As mentioned before, the effectiveness of AECEB-2 and AECEB-3 was confirmed in separate assessments. However, that effectiveness was hindered when the operation was

resumed in Runs 2 and 3. Based on the results of FTIR analysis carried out in Run 3, it was postulated that scouring agents made from PEG released foulants when AECEB-2 and AECEB-3 were carried out and caused the rapid TMP increase in Runs 2 and 3. It was hypothesized that the HR-MBR could be operated very stably with the AECEB when granular agents that were tolerant to the CEB were used. The next long-term operation (Run 4) was conducted to prove this hypothesis.

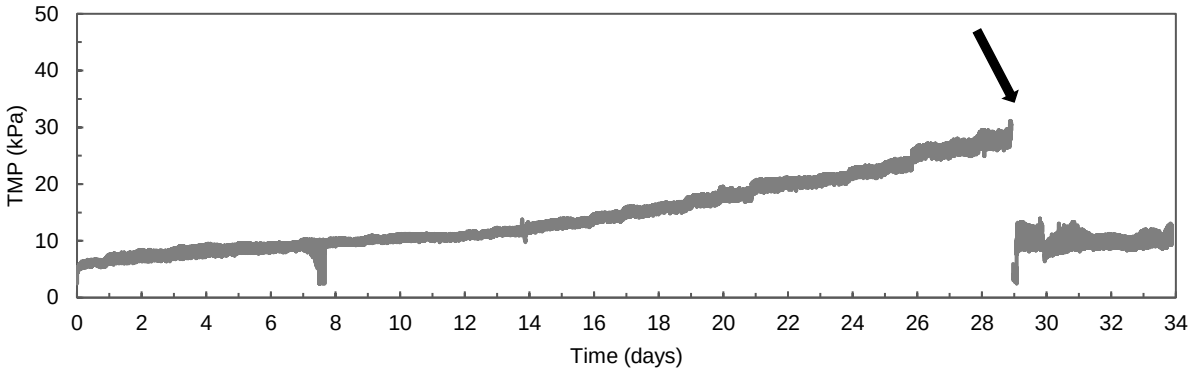


Figure 18. TMP evolution from the long-term operation of an HR-MBR (Run 4). The black arrow shows the start of the AECEB when TMP reached 30 kPa.

In Run 7, PU granules were used instead of the PEG granules. The operational conditions in Run 7 were the same as those set in Run 6: AECEB-3 was carried out in Run 7. Figure 18 shows the TMP increase observed in Run 7. After 29 days of operation when TMP reached 30 kPa, AECEB-3 was carried out. Permeability recovery achieved by AECEB-3 in Run 7, assessed in a separate tank, was 96% (Figure S3). This was very close to the recovery achieved in Run 6. As shown in Figure 18, the TMP profile after implementation of AECEB-3 in Run 7 was totally different from that observed in Run 6. In Run7, AECEB-3 was not accompanied by a surge of TMP. This result suggests that the aforementioned hypothesis, where PEG was responsible, is correct.

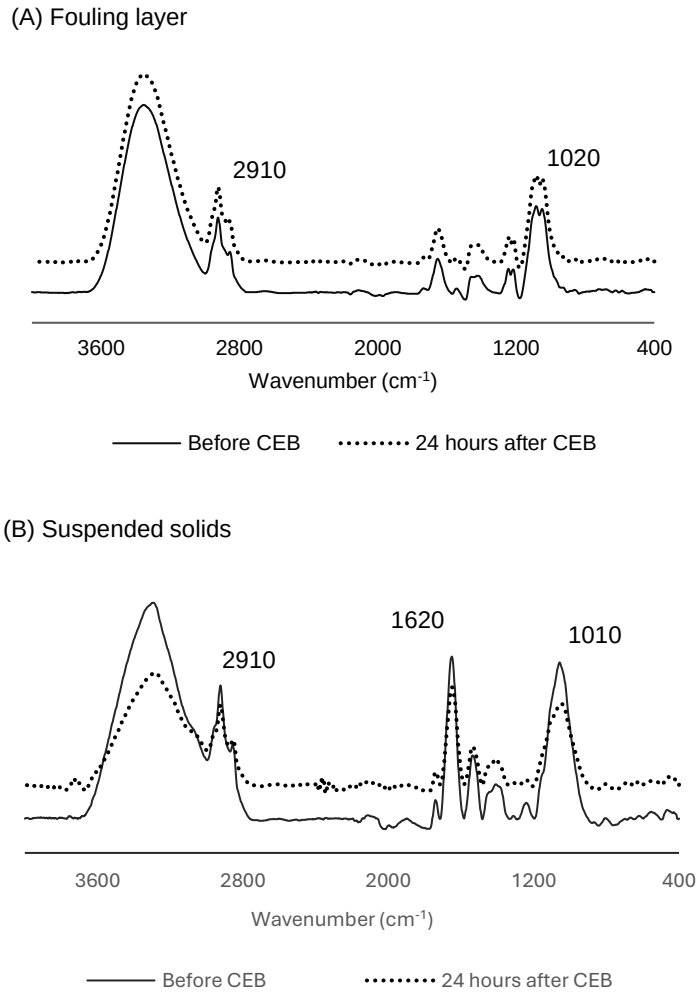


Figure 19. FTIR spectra of different fouling layers (A) found on the membrane surface and suspended solids in the reactor (B) before and 24 hours after AECEB-3 in Run 7.

Figure 19 shows FTIR spectra of the fouling layers collected before and 24 hours after the implementation of AECEB-3 in Run 7. Apparently, there was no similarity between the PEG standard and the fouling layer in Run 7 in terms of FTIR spectra. The characteristic sharp peaks around the 2870 cm^{-1} and 1089 cm^{-1} for the PEG standard wavelength were not seen in these FTIR spectra. It is notable that the FTIR spectra of the fouling layer collected before and after AECEB-3 were very similar in Run 7. Figure 19 also shows FTIR spectra of the suspended solids in the reactor before and 24 hours after AECEB-3 in Run 7. The same

conclusion as that for the fouling layer can be reached for the suspended solids, for which the characteristic peaks of the PEG standard were not observed. This implies that AECEB-3 did not alter the characteristics of the fouling layer or the suspended solids in Run 7, which were totally different from the observations in Run 6.

Mass balance analysis on COD in Run 7 showed consistent recovery of organic matter: about 78% of COD in the feed was recovered on average as concentrate in Run 7 (Figure S4). The use of the PU granules in Run 7 did not negatively affect the mass balance. The permeate quality was also not negatively affected by the use of PU granules. The concentration of COD in the permeate was 16±7 mg/L in Run 7, corresponding to 97% removal of the total COD from the raw wastewater. Permeate quality was analyzed right after the AECEB, it maintained a COD of below 25 mg/L, which meant that permeate quality was only slightly affected by AECEB.

CHAPTER 6.
CONCENTRATION OF THE RECOVERED ORGANIC MATTER WITH GRAVITY SETTLING

6.1 PERMEATE WATER QUALITY DETAILS

Conventional MBRs treating municipal wastewater exhibit COD removal of >95% [63,83]. Both MBRs in our study achieved a COD removal rate exceeding 93%, resulting in permeate COD concentrations of 22±8 mg/L for MBR 1 and 20±6 mg/L for MBR 2. It's important to note that MBRs already effectively remove colloidal and suspended organic matter, underscoring the significance of soluble COD in HR-MBR performance [29]. In our study, the removal of soluble COD reached 77.8% for MBR 1 and 79.3% for MBR 2. These results align with a recent pilot-scale HR-MBR study that utilized chemical coagulation [29], highlighting the effectiveness of our current system with its high aeration.

6.2 FINAL ORGANIC MATTER CONCENTRATION

Figure 20-A shows the solids from the discharged organic matter of both systems. As intended, concentration of COD in organic matter discharged from MBR 1 could be increased substantially. MBR 1 with the gravity separation system was able to increase the concentration from around 1 g/L to more than 10 g/L, which is seen in conventional MBR sludge with a 30-day SRT. The concentration of volatile solids (VS) in the gravity separated sludge (Figure 20) increased by 8.3 times, while volume of sludge decreased by 11.5 times, in comparison to the HR-MBR without gravity separation.

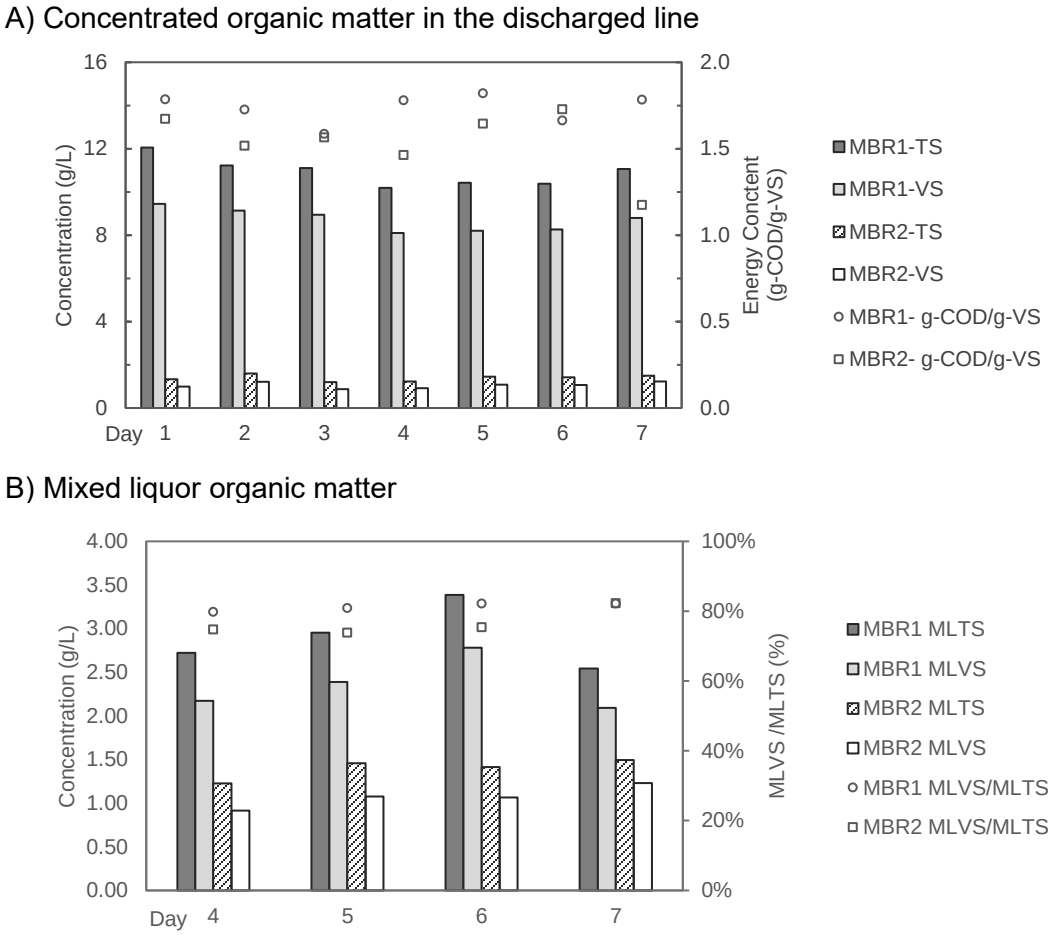


Figure 20. Concentration of organic matter and solids in the concentrated discharge (A) and the mixed liquor (B) of the HR-MBR with gravity separation (MBR 1) and without gravity separation (MBR 2). The left margin shows the respective energy content of the recovered organic matter (A) and the VS/TS ratio of the mixed liquor organic matter (B).

Figure 20-B shows the differences of organic matter content in the mixed liquor. Only the last 4 days were measured to allow the system to completely stabilize after adding the clean membranes. In most cases presented in Figure 20, MBR 1 exhibited higher values of energy content (g-COD/g-VS) and MLVS/MLTS.

A higher g-COD/g-VS value signifies a greater amount of extractable energy from the organic matter within the sample. The higher MLVS/MLTS ratio can imply one of two

possibilities: either more solids in the sample were utilized to support the growth of the microorganism community, or there will be an increased availability of organic matter during anaerobic digestion. The latter hypothesis is supported by the prior observation of g-COD/g-VS, as an increase in the biomass fraction of VS might lead to a reduction in energy content. A direct investigation into the digestibility of the concentrated matter could help validate this hypothesis.

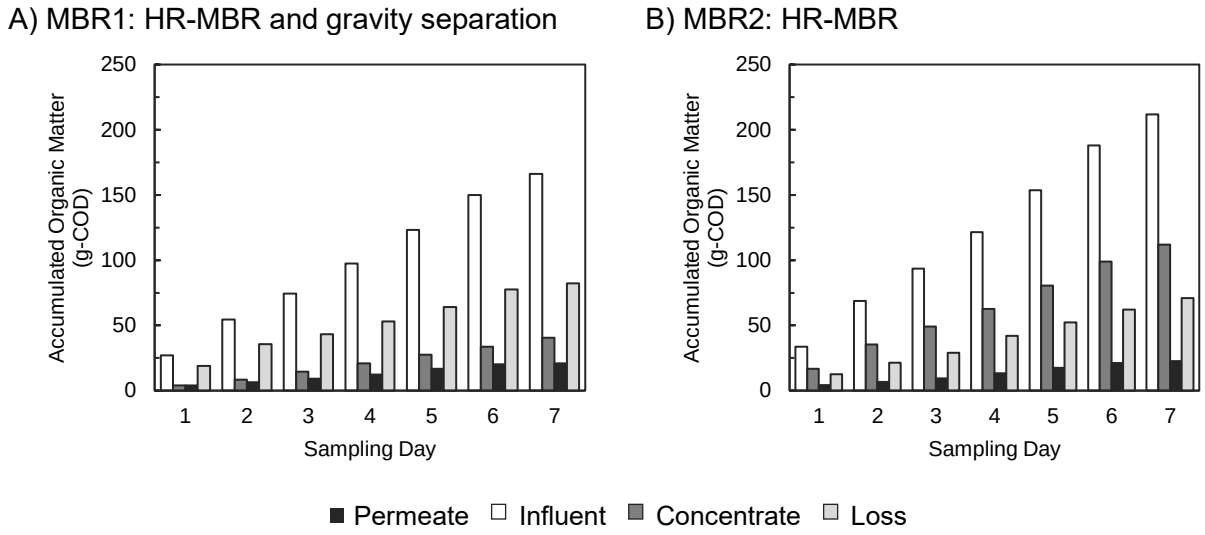


Figure 21. Daily mass balance analysis of both MBR1 (A) and MBR 2 (B).

6.3 MASS BALANCE ANALYSIS RESULTS

Including the organic matter settled in the primary sedimentation basin, the recovery rates were 56.9±4.7% for MBR 1 and 73.3±2.2% for MBR 2. Consequently, the implementation of gravity separation led to a substantial reduction in the recoverable organic carbon. The difference in organic matter accumulation between MBR 1 and 2 indicates that organic matter is retained within the system. This can likely be attributed to an increase in the total solids content of the mixed liquor, as illustrated in Figure 20-B. Secondly, the accumulation of fouling on the membranes was counted as losses, not as accumulation because it was never

released into the suspension. The decrease in recovery is likely attributed to both the losses mentioned previously and mineralization, influenced by the extended HRT and SRT within the HR-MBR combined with the gravity separation tank. A shorter time in the gravity settler may be a solution to this issue however, there is a balance between the time for settling and the losses to mineralization which could be better optimized.

6.4 DETERIORATION OF FILTERABILITY

The CST measurement was employed to assess the filterability of the mixed liquor [107,120]. CST values obtained from MBR 1 (304 ± 115 s) were much higher than those from MBR 2 (15 ± 4 s), indicating considerably reduced filterability in MBR 1. Extraction of EPS from the mixed liquor suspension was conducted for both MBRs during the final two days of operation. The extracted EPS exhibited COD concentrations of 90 ± 34 mg-COD/L for MBR 1 and 20 ± 6 mg-COD/L for MBR 2.

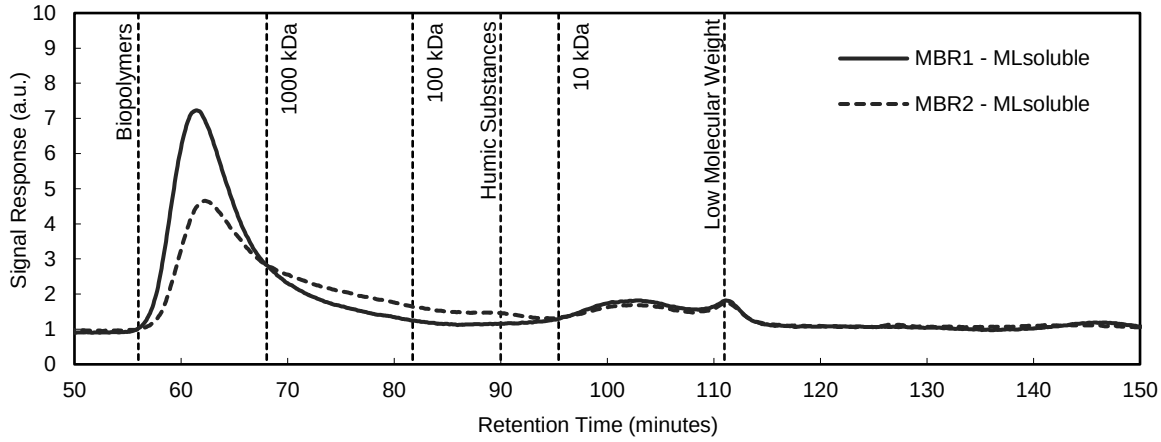


Figure 22. Comparison of LC-OCD chromatograms showing the soluble fraction of the mixed liquor (ML) suspension in MBRs 1 and 2.

The degradation in filterability observed in MBR 1 may be attributed to the higher presence of EPS in the mixed liquor [50,51] and an elevated concentration of biopolymers (more than 10 kDa sized molecules) in the dissolved fraction (Figure 22). This is notable due to the heightened fouling potential of these biopolymers [121]. It is hypothesized that the gravity separation basin, which reduced dissolved oxygen concentration and extended the HRT, could have induced the release of protective EPS from microorganisms.

A contrary observation was made in a study by Dai et al. [36], where a high-rate contact stabilization MBR with extended HRT and reduced dissolved oxygen in the stabilization tank exhibited less fouling compared to a high-rate MBR [36]. However, in our case, the removal of the concentrated particulate fraction allowed the soluble and colloidal fraction to overflow back into the MBR. Typically, this fraction is removed through adsorption, which may have amplified the adverse effects on fouling.

6.5 FOULING LAYER INVESTIGATION

The introduction of gravity separation for organic matter enrichment had a detrimental effect on fouling evolution in the HR-MBR. In Figure 23-A, the TMP evolution revealed a rapid increase in MBR 1, reaching 30 kPa within 5 days, whereas MBR 2 only reached 12 kPa. At this junction, MBR 1 would face difficulty processing the same volume of wastewater and would require cleaning to restore permeability. In contrast, MBR 2 could continue operating for a longer period due to its lower TMP values.

In many research studies, TMP graphs typically display maximum, minimum, or averaged TMP values per cycle. However, for HR-MBRs, fouling characteristics can vary greatly during each filtration cycle, necessitating consideration of how each cycle impacts the fouling layer. In this chapter, the addition of gravity separation led to distinct fouling evolution in both MBRs, which would not have been apparent if average TMP values were used to graph the same data. The thickness of the TMP plots illustrates how pressures at the cycle's start and end were affected by fouling and other conditions. The initial TMP of each cycle (at the graph's bottom) represents fouling that mitigation strategies couldn't remove. The final TMP of each cycle (at the graph's top) demonstrates both the rapid recompression

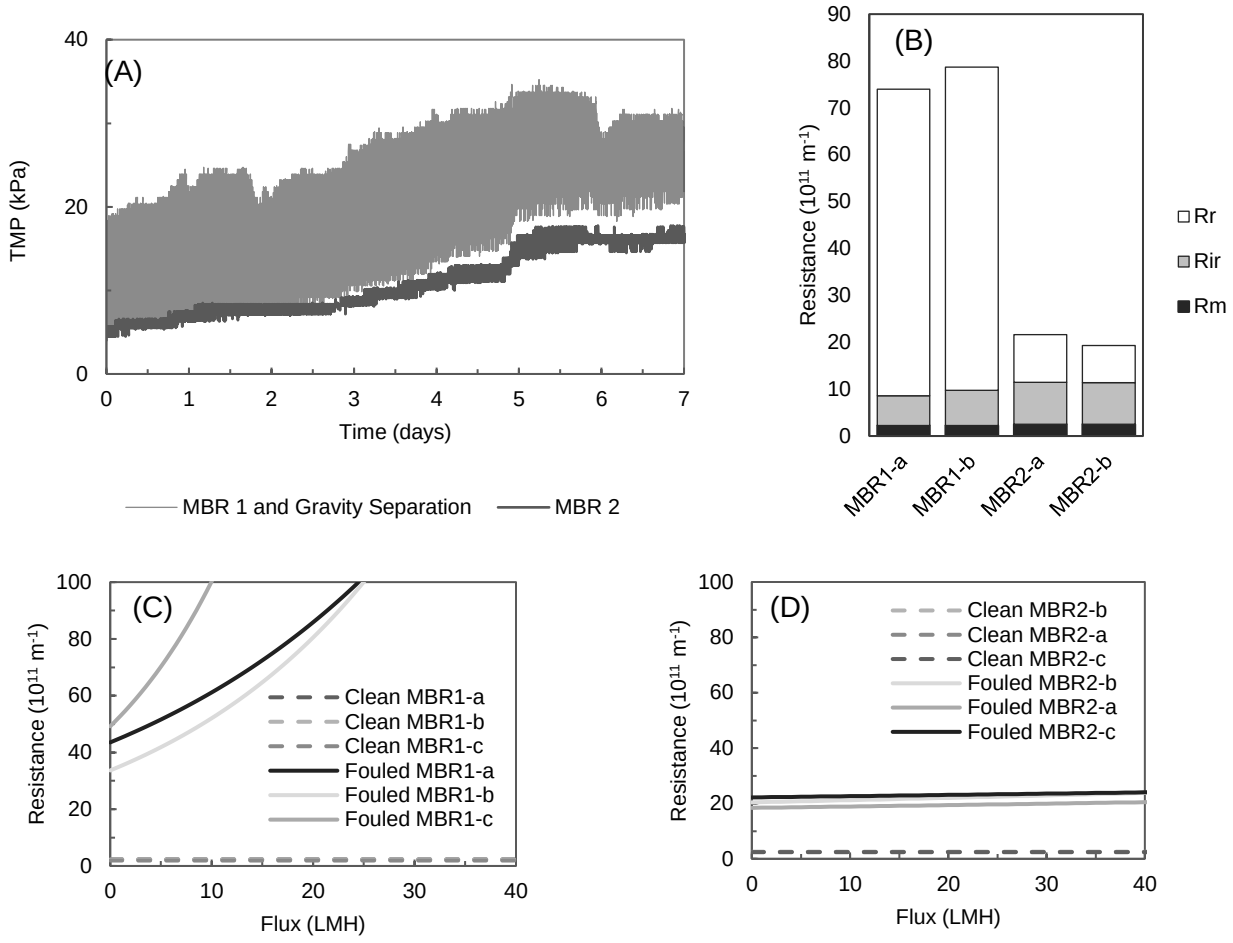


Figure 23. TMP evolution of the two reactors during operation (A) and resistance measured at the termination of the seven-day operation (B) of two membranes from each reactor. The models generated from the additional resistances measured at varying fluxes from the membranes in MBR1 (C) and MBR2 (D) are shown.

of the fouling layer combined with fouling accumulated during the filtration cycle, and how these impacted the membrane. By analyzing both parts of the graph, a more detailed view of the fouling can be extracted from the graph.

In Figure 23-A, MBR1 exhibited a much thicker graph than the one of MBR2. An explanation for the greater thickness of the TMP graph of MBR1 is a reduced mixed liquor filterability. As the filtration cycle commences, contaminants rapidly adhere to the membrane

surface and the TMP quickly climbs. Because most of the fouling is superficial, it can then be removed or released by CEB. An important observation is that the TMP graph's thickness remained consistent from the start of the operation, suggesting that the system's filterability was consistently problematic, even from the beginning.

During the CEB phase, the solution is pumped out of the membrane, serving two key purposes. First, it allows the fouling layer to decompress, akin to a relaxation phase. Second, it facilitates fouling layer cleaning by enabling chemical interaction with the fouling layer and mechanical detachment. When the TMP graph exhibits significant thickness, as seen here, it indicates that the fouling layer remains mostly intact, and CEB does not effectively clear it. This aligns with the subsequent SEM observations (Figure 24).

At the termination of operations, two membranes were taken from each MBR, and clean-water permeability measurements were conducted, as displayed in Figure 23-B. MBR1 exhibited significantly higher total resistance, corroborating the high TMP values noted earlier. The reversibility of the resistance (i.e., R_r/R_{ir}) indicates the proportion of fouling that constitutes a cake or gel layer, removable by sponge cleaning, versus intrapore fouling. For MBR 1 and 2, the reversibilities were 9.7 and 1.0, respectively. Such high reversibility highlights the thickness of the cake layer, contributing to the TMP graph's variability and the inefficacy of the CEB protocol.

The assessment of clean water permeability is recognized as a valuable indicator of membrane fouling and the performance of fouling mitigation. From the clean water permeability measurements, a resistance model was generated, illustrating the resistance at various fluxes. This model is depicted in Figures 23-C and 23-D for both MBRs at the termination of the runs. A large compressibility of the fouling layer in the MBR with gravity settling is demonstrated by the models when the flux is increased. This outcome implies that to maintain a constant flowrate, the flux and pressure must be significantly higher in MBR2, where no gravity settling was conducted. The models also facilitate a comparison between the membranes used for SEM observations and those where physical cleaning was conducted. The results indicate that the fouling on the membranes was comparable, allowing for a direct comparison between the observations made in the SEM and the resistance measurements and reversibility.

The high total resistance in MBR1, along with the observed TMP evolution and high reversibility, collectively point to substantial fouling issues within this system. In contrast, MBR 2 exhibited lower resistance and more favorable characteristics. This study underscores the complex interplay between various factors affecting membrane fouling in HR-MBR systems. Further investigation into the specific mechanisms behind the observed trends in fouling evolution and the impact of different operating conditions is warranted to optimize HR-MBR performance for wastewater treatment.

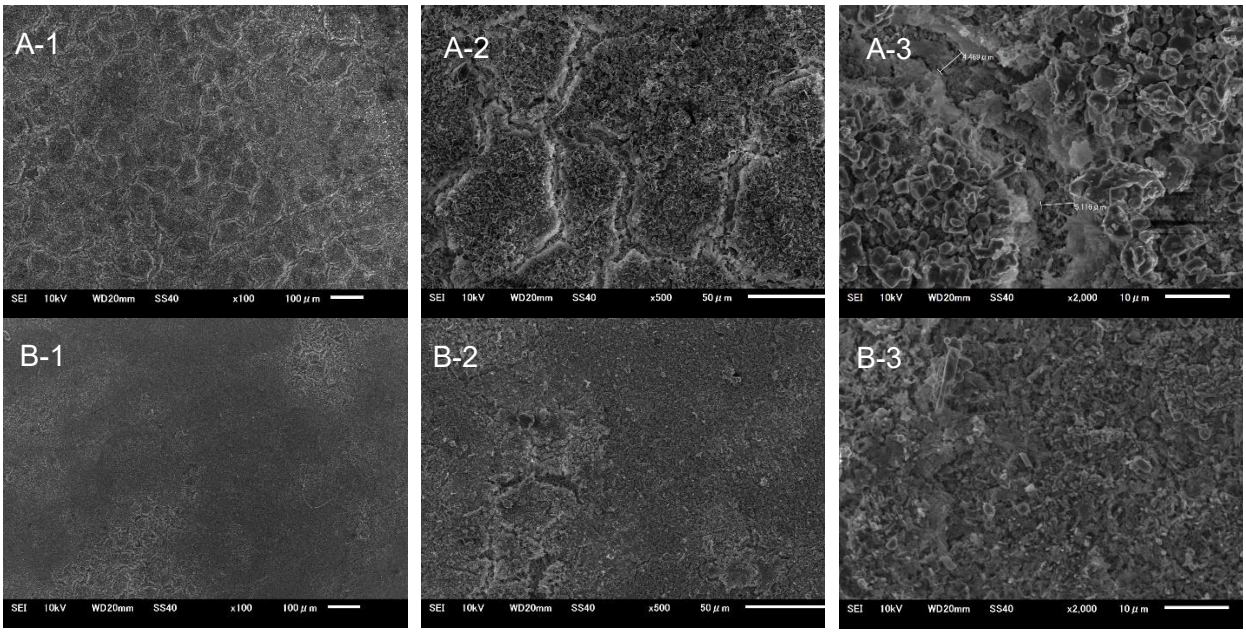


Figure 24. SEM observations of the surface of the fouled membranes at the end of operation. Different magnifications of the average surface of the membranes from MBR 1 (A images) and from MBR 2 (B images).

As the fouling layer thickness increases, two potential mechanisms come into play for backwashing: the formation of channels or the detachment of large sections of the fouling layer. In the case of MBR2, where the fouling layer is considerably thinner than that of MBR1, some areas exhibit channel formation, though much of the fouling layer is evenly distributed

across the membrane surface. Consequently, the TMP evolution (Figure 23-A) displays a smaller variation between the highest and lowest values per cycle, resulting in a thinner plot.

Meng, et al. [122] reported the presence of channels in the fouling layer of backwashed membranes fouled with bovine serum albumin. However, this phenomenon was not observed in backwashed membranes with fouling layers formed from sodium alginate or humic acids. These findings suggest that the fouling encountered in MBR 1 and 2 likely consists primarily of proteins. Further investigation is needed to develop a cleaning procedure tailored to this type of fouling.

CHAPTER 7.
COMPARATIVE ANALYSIS OF HR-MBR: ORGANIC MATTER RECOVERY AND ENERGY EVALUATION

7.1 COMPARISON OF HR-MBR EFFECTIVENESS

In comparing recovery rates of the HRAS systems, it was found that the average recovery reported in the literature ranges between 20 and 55% of the influent COD [10]. The highest recovery percentages in these studies are achieved at SRTs between 0.25 and 2 days. Consequently, most HR-MBR studies conducted in the past decade have focused on these SRTs. Some studies have reported higher recoveries closer to an SRT of 2 days, while others have found the highest recoveries closer to an SRT of 0.5 days.

During the operation of HRAS, the quality of the effluent is commonly considered the limiting factor for recovery. The loss of organic matter in the effluent constitutes a significant portion of the losses that keep HRAS recovery below 60%. This is attributed to the fact that the HRT can be dramatically reduced, which increases the total concentration of organic matter but reduces the effectiveness of treatment. Conversely, if the SRT is allowed to increase, the treatment becomes more effective, but losses due to mineralization begin to impact the overall effectiveness of the system. This balance between SRT and HRT is imperative to find the optimal solution for carbon capture, however, in the case of HR-MBRs, the separation of SRT and HRT allows more freedom and flexibility in this balance which then is translated into higher recovery and treatment capabilities at the cost of the increased energy demands.

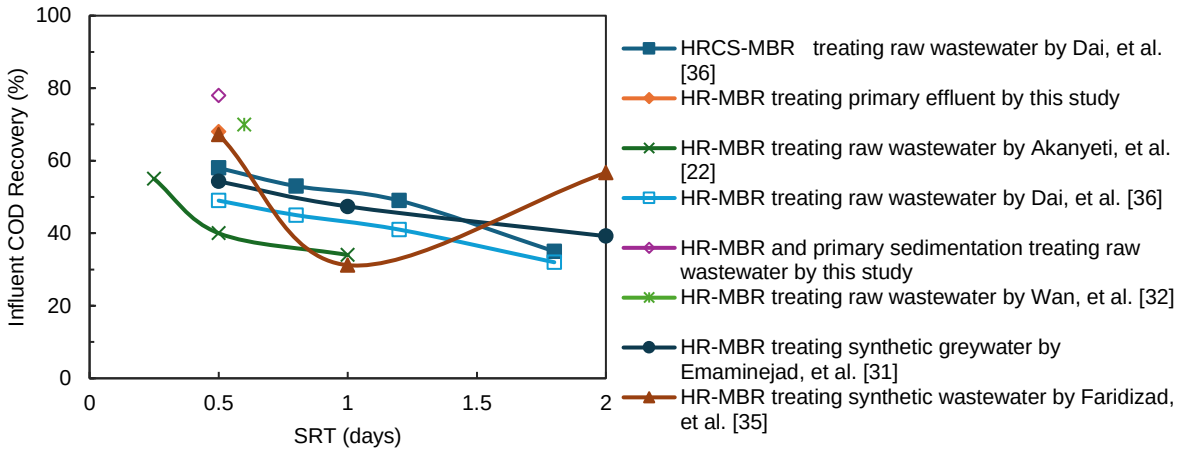


Figure 25. Influent COD recovery compared with the SRT in different high-rate systems treating different wastewaters. High-rate contact stabilization combined with a membrane bioreactor (HRCS-MBR).

Most HR-MBRs reported in the literature demonstrate recovery percentages that are similar to or higher than those of HRAS systems. This is likely due to the complete separation of the particulate and colloidal matter by the membranes, which ensures that a larger portion of the influent COD, that would otherwise be lost in HRAS, is collected in the concentrated sludge.

Figure 25 illustrates that the recovery in HR-MBRs operating with SRTs between 0.25 and 0.5 days ranges from 30% to 70%. The rate of carbon recovery achieved by the HR-MBR in this study was also found to be comparable to the rates reported for other HR-MBRs. Orhon et al. [12] demonstrated through simulation that the use of primary sedimentation could improve the recovery of the HRAS system. This was confirmed to also be true with HR-MBR with the results of the combined recovery of the primary sedimentation and the HR-MBR. The results obtained in Run 7 indicate that it is possible to increase the capture of organic matter by 10%, from 68% to 78% of the total incoming COD, by utilizing primary sedimentation. Allowing to elevate the recovery above other reported HR-MBR systems.

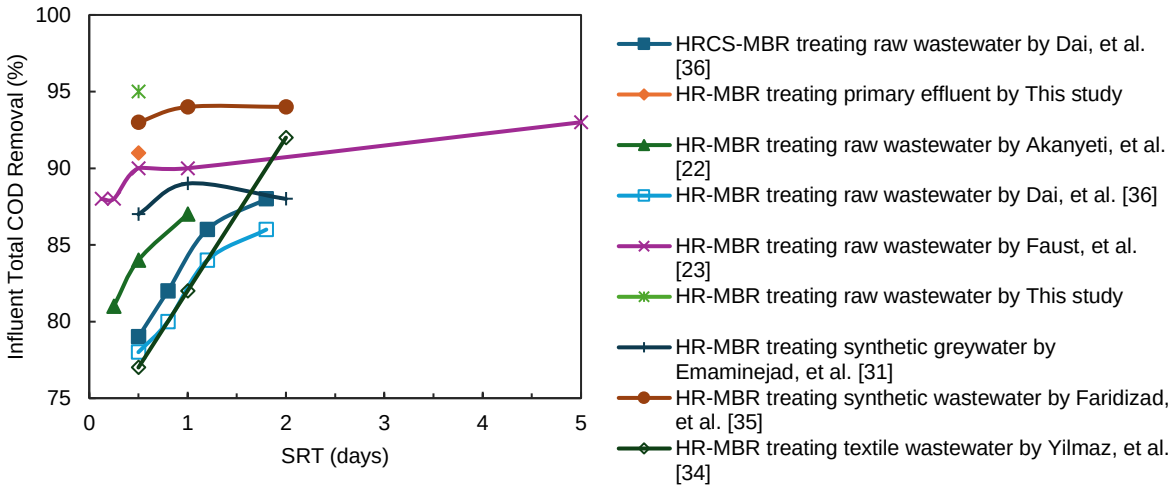


Figure 26. Influent COD recovery compared with the SRT in different high-rate systems treating different wastewaters.

The primary function of the HR-MBR is the removal of organic matter, while the removal and recovery of nutrients are delegated to other processes. Therefore, the efficiency of the HR-MBR is primarily compared through COD removal. This process has been the focus of most HR-MBR studies in recent years. Figure 26 provides a summary of the total COD removal achieved by other HR-MBRs examined in recent studies, as well as the performance of the HR-MBR used in this study. The HR-MBR in this study demonstrated a COD removal rate of approximately 91% without the effect of the primary sedimentation basin and 95% with its inclusion. This resulted in a COD removal rate that surpasses those reported for other HR-MBRs operating under similar conditions, including some with higher SRTs.

Figure 27 shows the losses of recent HR-MBR studies. Most of the losses depicted are based on fouling and mineralization observed during operation. A clear trend is seen where the losses increase as SRT becomes longer. Additionally, synthetic wastewaters have significantly lower losses compared to real wastewater, a deeper investigation into this phenomenon might be necessary.

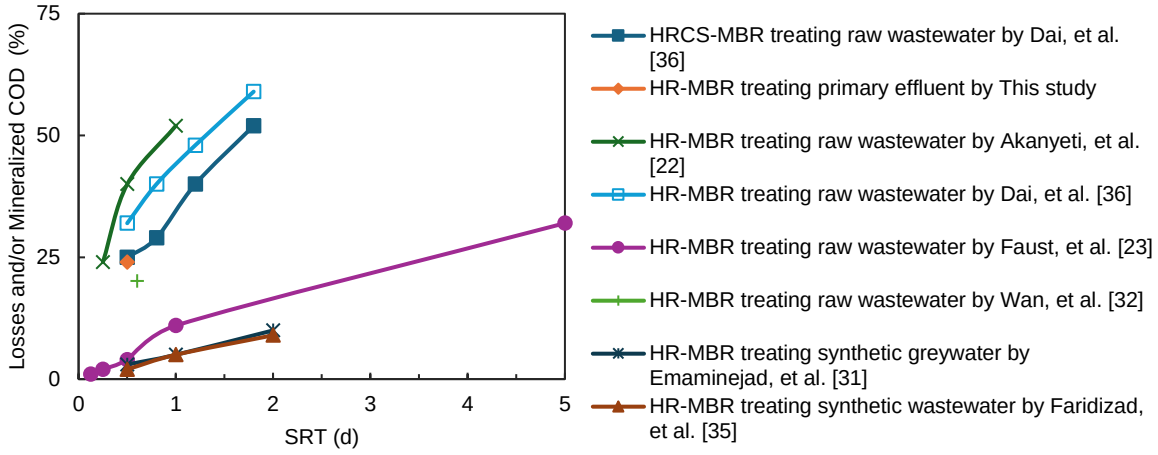


Figure 27. Influent COD loss percentage compared with the SRT in different high-rate systems treating different wastewaters.

From the comparison with other HR-MBRs, we can see that the improvements proposed by this study have had important effects on the effectiveness of the HR-MBR process. Losses based on cake layer formation have been dramatically reduced and the removal efficiency allows for less COD escaping into the effluent. These two improvements combined are what allowed the recovery to reach such a high level. Additionally, the inclusion of the primary sedimentation basin into the process is one of the important aspects of this research. Although recovery during the HR-MBR process becomes significantly more difficult, the added benefits of the low energy demand primary treatment will be shown in the next section, which is focused on the energy balance.

7.2 EVALUATION OF THE ENERGY BALANCE AND THE FEASIBILITY OF THE HR-MBR AS AN EFFECTIVE TECHNOLOGY

7.2.1 Primary treatment

Resource recovery and energy recovery are the primary goals of the HR-MBR and other technologies focused on the shift from wastewater treatment plant to wastewater resource

recovery plant. Primary systems and treatments, such as pumping and grit separation, are not included in this chapter's discussion, as they are necessary for all water treatment or resource recovery plants. The critical question in primary treatment is whether to conduct a specific treatment like primary sedimentation or proceed directly to secondary treatment, which might be more effective at treatment or recovery.

As previously discussed (Section 7.1), adding a primary sedimentation basin prior to the HR-MBR could increase recovery. According to Mizuta and Shimada [123], primary treatment accounts for an average of 10-20% of a wastewater treatment plant's energy consumption. Besides the added recovery, the energy required for this treatment is significantly lower than for subsequent treatments due to the fact that energy consumption during the primary treatment is primarily due to pumping. This means that each kilowatt-hour per cubic meter captured in primary treatment is more efficient compared to the energy captured in later processes.

7.2.2 Secondary treatment and organic matter recovery

When the HR-MBR technology is chosen as the secondary treatment, the organic matter is captured, and other nutrients such as nitrogen and phosphorus are deferred to a different recovery process, which will be discussed later. The benefits of using the HR-MBR include reductions in energy consumption compared to conventional MBRs. However, it should be noted that as a membrane process, higher energy consumption is expected compared to a CAS system. The crucial point would be whether the recovery could outweigh the additional energy consumption. A significant deciding factor in this case would be the size of the treatment plant.

Energy net neutrality, and even some net positive energy plants, have already been demonstrated in certain scenarios [124,125]. Medium to large scale wastewater treatment typically relies on anaerobic digestion and biogas production as the primary methods to recover energy from organic matter [126].

In Japan, it has been observed that larger plants exhibit better specific energy consumption than smaller ones [123]. These larger plants can leverage substantial capital investments, such as the construction of digesters and Combined Heat and Power (CHP) engines, which may be prohibitively expensive for smaller recovery plants. Therefore, HR-

MBR systems could be an effective strategy to incorporate into these larger plants. This is because the capital investment required for membrane systems is often higher than for other types of technologies.

Table 7. Energy consumption and recovery of wastewater treatment using the HR-MBR and the primary sedimentation basin

Total energy consumption (kWh/m^3)	**-0.118**
Pumping (kWh/m^3)	-0.008[a]
Aeration (kWh/m^3)	-0.110[b]
Total energy generation (kWh/m^3)	**0.204[c]**
Influent COD (kg-COD/m^3)	0.306[d]
Recoverable COD (kg-COD/m^3)	0.239[e]
Theoretical energy captured from COD in wastewater (kWh/m^3)	0.834[f]

[a] Average pumping energy consumption in a conventional MBR [104].

[b] Average aeration energy consumption in a conventional MBR (0.22 kWh/m3) [104] reduced by 50% through granular scouring [76,77] as well as the HR-MBR utilizing less aeration overall.

[c] Theoretical energy produced from COD in wastewater multiplied by both the anaerobic digestibility of up-concentrated sludge (70%) [127,128] and the efficiency of the combined heat and power engine (35%) [129].

[d] Influent COD in the primary sedimentation tank (Chapter 6).

[e] Influent COD multiplied by the recovery rate of 78% (Chapter 6).

[f] Stoichiometric chemical content in municipal wastewater (3.49 kWh/ kg-COD) [130] multiplied by the recoverable COD.

Table 7 shows a preliminary estimation of the energy balance for wastewater treatment using the HR-MBR. Although it is a primitive analysis with several assumptions, a large potential of HR-MBRs as technology that enables transformation of wastewater treatment from a big consumer of energy into a net energy generator has been demonstrated. It should be noted that there is plenty of room for optimization in the operation of the HR-MBR. To estimate energy consumption in the HR-MBR, the data for optimized full-scale conventional

MBRs [104] are used. For the HR-MBR tested in this study, 50% reduction of the aeration energy, compared with that of conventional MBRs, was assumed with the use of granular scouring [76,77] as well as the reduced aeration demand. Further reduction in aeration rates may be possible, as optimization of granular scouring for MBRs has never been studied. Regarding energy production, many references focusing on biogas production from organic matter are available as cited in the footnotes of Table 7, and they were adequately used.

Wastewater treatment plants, excluding MBRs, can have energy consumption as low as 0.25 kWh/m^3 for constructed wetlands [131], and as high as 3.74 kWh/m^3 for advanced wastewater treatment plants [123]. This represents a wide range, where balancing at the higher end would be extremely challenging.

In this context, it is important to compare the effectiveness of conventional MBRs, which can produce very high-quality effluent, with the energy consumption of the HR-MBR. Optimized MBRs have significantly lower energy consumption due to efficiency improvements in recent years [132]. For instance, some wastewater treatment plants in China that utilize MBRs have reduced specific energy consumption to as low as 0.52 kWh/m^3 [133] while larger systems can go even lower to 0.40 kWh/m^3 [126]. The result presented in Table 7 would lower the final specific energy consumption to close to 0.29 - 0.41 kWh/m^3, a total reduction of about 20-25%.

On the other hand, while CAS systems can have low energy recovery, some conventional wastewater treatment plants using CAS and anaerobic digestion have achieved an energy production of 0.17 kWh/m^3 [123]. Compared to the results presented, an increase of 20% energy recovery is also expected. Therefore, the improvements proposed with the HR-MBR technology can reduce energy requirements and increase biogas production, thereby reducing the total gap between consumption and generation.

7.2.3 Additional treatment and other resource recovery

This study did not explore the removal and recovery of other resources, such as phosphorus, nitrogen, and potassium. However, these resources are a significant part of the costs associated with the energy and chemicals of a wastewater resource recovery facility. Additional energy and/or chemical costs are associated with the recovery of the remaining constituents. For instance, the removal of phosphorus and nitrogen can be achieved through

chemical precipitation [134,135], but the added costs may make this type of treatment prohibitive. The rising costs of fertilizers containing these resources could create a more favorable landscape for investing in technologies that aim to recover these nutrients, thereby helping to reduce the total cost of implementation.

Due to the short SRT, the collection and removal of resources from the HR-MBR are primarily accomplished through the adsorption and solid separation pathway. Most of the nitrogen in the influent, especially the dissolved fraction, passes through to the effluent. A novel approach to nitrogen removal that has emerged in recent years is the membrane contactor. Some pilot-scale and full-scale facilities have already been installed, achieving a removal of 70-96% of the ammonium nitrogen [136–138]. Membrane contactors are energy-efficient because they only require the energy for crossflow pumping, and the acid with the captured nitrogen can yield high-purity results for resource recovery. These new technologies can be employed to further reduce energy consumption while creating viable income streams to offset the cost of the remaining energy balance.

CHAPTER 8.
CONCLUSIONS, IMPLICATIONS AND FUTURE PROSPECTS

8.1 CONCLUSIONS

The application of granular scouring and frequent CEB, which are too intensive for polymeric membranes, was enabled by the use of ceramic membranes. The efficient movement of granules was promoted by the flat-sheet configuration, making an HR-MBR using flat-sheet ceramic membranes a promising technology.

The performance of the HR-MBR observed in the initial stages of this study was characterized by stable membrane filtration at a relatively high flux of 20 LMH, high recovery of organic matter (more than 70%), and a low concentration of COD in the permeate (less than 20 mg/L). Efficient recovery of organic matter was enabled by the low SRT and HRT set in this study (0.5 days and less than 2 hours, respectively), while severe fouling in the HR-MBR could be controlled by intensive membrane cleaning. Subsequently, an attempt was made to operate the HR-MBR under a high flux of 16 LMH for longer than was achieved in the initial experiments (i.e., around one month).

A newly developed CEB protocol, Air-Exposed CEB (AECEB), exhibited excellent cleaning efficiencies, and the severe fouling that developed in the HR-MBR was almost completely cancelled. However, it was found that the strong chemicals used for AECEB degraded the PEG granules. Although these granules greatly contributed to the mitigation of fouling in the HR-MBR in long-term operation via their scouring effects, severe membrane fouling was caused when the operation was restarted by PEG fragments generated during the AECEB. This problem was adequately addressed by replacing the PEG granules with PU granules that were tolerant to the chemicals.

With the optimized cleaning protocol, the fouling of the HR-MBR was excellently controlled, and long-term operation was possible without implementing off-line cleaning. This

work has clearly demonstrated the feasibility of HR-MBRs to recover organic matter in wastewater treatment and has provided important information for converting WWTPs into resource recovery centers.

The installation of gravity separation was examined to increase the concentration of organic matter recovered by an HR-MBR. This method was successful at increasing this concentration by a large margin. The energy content and Volatile Solids/Total Solids (VS/TS) ratio were also enhanced. If these were the only important aspects, the process would be optimal because of its low energy use. However, it was shown that the combination examined led to deterioration of the mixed liquor, and eventually caused very severe membrane fouling in the HR-MBR. Also, the combination reduced the amount of recoverable organic carbon due to increased losses in the mass balance. Other solutions for enrichment of organic matter discharged from HR-MBRs must be examined.

Finally, significant improvements in the HR-MBR process were highlighted by the study, particularly in COD removal, loss reduction and recovery rates. Superior performance was demonstrated by the HR-MBR compared to HRAS systems, primarily due to the complete separation of particulate and colloidal matter by the membranes. It was also found that COD removal rates of approximately 91% without the primary sedimentation basin and 95% with its inclusion were achieved by the HR-MBR, surpassed the rates reported for other HR-MBRs under similar conditions. Additionally, the capture of organic matter was increased by 10%, from 68% to 78% of the total incoming COD, by the inclusion of the primary sedimentation basin. The high recovery rates achieved were attributed to the improvements implemented in this study, such as reducing losses due to cake layer formation and enhancing removal efficiency.

Significant potential in transforming wastewater treatment plants into resource recovery facilities is demonstrated by the HR-MBR technology proposed in this study. Recovery rates are enhanced, and energy consumption is reduced by the primary treatment, particularly the inclusion of a primary sedimentation basin. Therefore, primary sedimentation should be included in future studies to explore the real effect during larger scales. The HR-MBR technology, with its potential for optimization of secondary treatment, has shown promise in reducing energy requirements and increasing biogas production, thereby narrowing the gap between energy consumption and generation. Additionally, the importance of recovering

other resources, such as phosphorus and nitrogen, to further enhance the cost-effectiveness and sustainability of wastewater resource recovery facilities is underscored by the study.

8.2 IMPLICATIONS AND FUTURE PROSPECTS

In this study, we successfully restored the permeability of the membranes that were severely fouled in the long-term operation of the HR-MBR by an on-line cleaning process: membranes were not taken out from the system during cleaning. It is therefore possible to minimize the downtime in long-term operation of the HR-MBRs during the membrane cleaning, enhancing the feasibility of recovery of organic matter from wastewater by the HR-MBR process. It should be noted that the membrane flux set in this study (16 LMH) was considerably higher than the filtration fluxes set in related studies [22,30,32]. The efficiency of the cleaning developed in this study is apparently high. The cleaning protocol for HR-MBRs, AECEB, is still not fully optimized. There is plenty of room for optimization of AECEB for HR-MBRs, implying that the feasibility of the recovery of organic matter from wastewater by HR-MBRs is likely to be further enhanced.

The final length of operation achieved in this study was 29 days of continuous operation during Run 7. Implementation of the AECEB showed the effective reset of the TMP to a level where operation would basically restart without needing to move the membrane plates. The restart would theoretically present an indefinite operational period where better observations of the effects of influent, fouling, filterability, sludge characteristics, etc. can be studied in more detail. Thus, the next step of the project would be to study these effects and optimize the process to maximize recovery and evaluate the potential of the organic matter and nutrients that can be extracted through this technology.

Most previous HR-MBR studies used hollow fiber membranes to have a large membrane area, which was necessary to operate the system under a lower flux. In this study, in contrast, stable operation of the HR-MBR could be carried out for a long period (i.e., about one month) under a high flux of 16 LMH. This was largely attributed to the effectiveness of granular scouring, which is not easily used with hollow fiber membranes due to trapping of granules inside the membrane bundles. Also, the hard contact between granules and the membrane

can cause damage of polymeric membranes [76,77]. The air-exposed CEB developed in this study may also be used with hollow fiber membranes. However, much attention needs to be paid to the concentrations of chemicals in the application of AECEB for HR-MBRs using hollow fiber membranes. Generally, polymeric hollow fiber membranes are much weaker than ceramic membranes. The use of flat-sheet ceramic membranes could avoid these problems and take full advantage of the protocols developed.

It is certainly desirable to reduce the amount of chemicals used in AECEB. In this study, NaClO and oxalic acid were chosen partly because they have been commonly used in full-scale applications of membrane processes [64,139]. However, they are toxic when they are discharged directly to natural environments. Therefore, alternative agents for AECEB should be investigated. Various types of "green" chemicals have been investigated, and some of them were reported to be effective for membrane cleaning [140]. They should be examined with HR-MBRs to enhance the feasibility of the process. Nanobubbles have been used for cleaning various industrial components, including membranes [141]. It was reported that nanobubbles could be used for backwashing of membranes in MBRs [102]. Application of those new chemicals that have less impact on the environment should be examined for AECEB for HR-MBRs in future studies. If frequent implementation of AECEB is accepted (Run 1 in this study), the fouling in an HR-MBR could be controlled much more easily. The feasibility of the operation of HR-MBRs with frequent implementation of AECEB should also be investigated in future studies.

It should be noted that there is still significant potential for optimization of the operating conditions of the HR-MBR. Investigation into how to incorporate processes of nutrient recovery in combination with the HR-MBR is necessary. Additional concentration of the recovered organic matter is also necessary to improve the transportation and availability of the organic matter.

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APPENDIX A. SUPPLEMENTARY MATERIALS

S1. Literature review of the HR-MBR

Table S1. Collection of studies that have reported on HR-MBRs specifically.

Authors	Year	Title	Highlights	Fouling and mitigation
Ng et al. [21]	2005	Membrane bioreactor operation at short solids retention times: performance and biomass characteristics	▪ Total COD removal is above 94% in MBR compared to CMAS. ▪ Nitrification stopped below a 2.5 d SRT.	▪ Fouling was not discussed. ▪ Mitigation by relaxation and coarse bubble aeration.
Akanyeti et al. [22]	2010	Feasibility of bioflocculation in a high-loaded membrane bioreactor for improved energy recovery from sewage	▪ Feasibility was confirmed for at least 35% total COD recovery. ▪ Mentions lower aeration energy consumption due to lower aeration demand. ▪ Call for further optimizations, especially in fouling mitigation and process conditions.	▪ High fouling observed especially below 1 d SRT. ▪ Mitigation by relaxation and coarse bubble aeration.
Hernandez Leal et al. [30]	2010	Bioflocculation of grey water for improved energy recovery	▪ Excellent bioflocculation of greywater at short	▪ Complete fouling before 10 hours of operation in all conditions evaluated.

		within decentralized sanitation concepts	SRTs was demonstrated. ▪ Low mineralization (11%) in greywater was observed.	▪ Mitigation by physical cleaning twice per day.
Faust et al. [23]	2014a	High loaded MBRs for organic matter recovery from sewage: Effect of solids retention time on bioflocculation and on the role of extracellular polymers	▪ Found 0.5 -1 d SRT to be the best for organic matter recovery. ▪ Showed EPS proteins were the main drivers of bioflocculation.	▪ Separate filtration experiments showed higher resistances with lower SRTs. ▪ Higher bioflocculation at longer SRTs showed better fouling performance. ▪ Mitigation by relaxation and coarse bubble aeration.
Faust et al. [41]	2014b	Effect of dissolved oxygen concentration on the bioflocculation process in high loaded MBRs	▪ Higher DO (4 mg/L) performed better for bioflocculation compared with lower DO (1 mg/L). ▪ All parameters of the biomass suspension were better with higher DO.	▪ Higher DO supernatant and concentrate had better filterability than lower DO. ▪ Mitigation by relaxation and coarse bubble aeration.
Başaran et al. [42]	2014	Kinetic characterization of acetate utilization and response of microbial population in super	▪ SRT influenced microbial communities, but they did not have a negative effect on HR-MBR performance.	▪ A side stream membrane of pore size of 20 nm was used help maintain the TMP below 200 kPa.

		fast membrane bioreactor	▪ Suggested low SRT was more beneficial for lower SMP production than effective COD capture.	▪ Mitigation by frequent backwash every cycle and chemical cleaning every day.
Faust et al. [43]	2015	Characterization of the bacterial community involved in the bioflocculation process of wastewater organic matter in high-loaded MBRs	▪ The bacterial community during an SRT of 0.125 d accumulated in the supernatant. ▪ Higher SRTs shifted the communities to the sludge flocs which in turn increased sludge bound EPS and better bioflocculation.	▪ Fouling was not discussed. ▪ Mitigation by relaxation and coarse bubble aeration.
Sözen et al. [33]	2015	Performance and microbial behavior of submerged membrane bioreactor at extremely low sludge ages	▪ The microbial community was directly affected by the SRTs selected. ▪ Lower SRTs selected the microbial community for faster growth.	
Sözen et al. [44]	2016	Toward a novel membrane process for organic carbon removal—fate of slowly biodegradable substrate in super	▪ Model simulation of the HR-MBR confirmed complete consumption of hydrolysable products in all configurations tested.	▪ A side stream membrane of pore size of 20 nm was used help maintain the TMP below 200 kPa.

		fast membrane bioreactor		▪ Mitigation by frequent backwash every cycle and chemical cleaning every day.
Sözen et al. [24]	2018	A novel process maximizing energy conservation potential of biological treatment: Super fast membrane bioreactor	▪ Model simulations confirmed removal of soluble biodegradable COD components and partial removal of slowly biodegradable COD. ▪ Model estimation highlights increased energy recovery and increased energy conservation achieved by the HR-MBR, compared to a wide range of processes for wastewater.	▪ Fouling was not discussed.
Dai et al. [36]	2018	High-Rate Contact Stabilization Process-Coupled Membrane Bioreactor for Maximal Recovery of Organics from Municipal Wastewater	▪ Comparison of HRCS-MBR with HR-MBR suggested better performance of the contact stabilization system due to higher bound EPS production. ▪ Suggested an optimal SRT of 1.2 days for high-rate processes.	▪ Higher fouling rates observed with lower SRTs, due to reduced bioflocculation. ▪ High correlation between colloidal and SMP with TMP increase. ▪ Mitigation by coarse bubble aeration.

Emaminejad et al. [31]	2019	Gaining deeper insights into the bioflocculation process occurring in a high loaded membrane bioreactor used for the treatment of synthetic greywater	▪ Suggested a maximum bioflocculation efficiency reached when SRT was 2 days. ▪ Correlation analysis suggested protein and tightly bound fractions of EPS had strong effect on bioflocculation, while carbohydrates and lightly bound fraction of EPS had a negative impact on energy recovery.	▪ Fouling was not quantified during the study, but it was suggested the lowest fouling would be during an SRT of 0.5 days. ▪ Mitigation by relaxation, coarse bubble aeration and physical cleaning 4 times per day.
Wan et al. [32]	2020a	High-Loaded Bioflocculation Membrane Reactor of Novel Structure for Organic Matter Recovery from Sewage: Effect of Dissolved Oxygen on Bioflocculation and Membrane Fouling	▪ Higher EPS content was found in the high DO (6-8 mg/L) system compared to the low DO (1-2 mg/L) system. ▪ Similar but higher bioflocculation efficiency was obtained from the high DO system.	▪ Fouling in the higher DO system was more serious than in the lower DO system. ▪ The reactor with low DO operated for 130 hours before reaching 30 kPa. ▪ Mitigation by relaxation, coarse bubble aeration and physical cleaning at 30 kPa.
Wan et al. [45]	2020b	High Loaded Bioflocculation Membrane Reactor of	▪ Poor bioflocculation efficiency (65%) was obtained in the low	▪ Fouling in the lower temperature system was more serious than

		Novel Structure for Organic Matter Recovery from sewage: Effect of Temperature on Bioflocculation and Membrane Fouling	temperature reactor (8°C) compared to 85% in the high temperature one (15°C). ▪ This low bioflocculation was attributed to the low EPS production in the low temperature system.	in the high temperature system. ▪ They suggest good bioflocculation could greatly mitigate membrane fouling. ▪ Mitigation by relaxation, coarse bubble aeration and physical cleaning at 30 kPa.
Faridizad et al. [35]	2022	The use of membrane bioreactors in high rate activated sludge processes: How and why sludge retention time affects membrane fouling	▪ SMP was the highest correlated parameter with an increase in membrane fouling. ▪ 0.5 d SRT was found to be the best SRT in terms of fouling, mineralization, yield and COD recovery out of the ones examined.	▪ Fouling was found to increase when SRT increased. ▪ Mitigation by relaxation, coarse bubble aeration and physical cleaning at 30 kPa.
He et al. [29]	2023	Chemically enhanced high-loaded membrane bioreactor (CE-HLMBR) for A-stage municipal wastewater treatment: Pilot-scale experiments and practical feasibility evaluation	▪ The practical feasibility of the chemically enhanced HR-MBR was confirmed on a pilot scale system.	▪ Stable operation (8-10 LMH) was achieved for more than a month ▪ Mitigation by relaxation, aeration, PAC coagulation and activated carbon addition.

Yilmaz et al. [34]	2024	Performance of a high-rate membrane bioreactor for energy-efficient treatment of textile wastewater	▪ The HR-MBR was found to be promising at removing COD from industrial dye wastewater. ▪ Membrane fouling was found to be one of the mayor limiting factors, especially at low SRTs and HRTs.	▪ Sludge filterability decreased during periods with 0.5-1 d SRT due to accumulation of slowly biodegradable substrates and SMP. ▪ Mitigation by relaxation and aeration. Physical and chemical off-line cleaning when necessary.

S2. Mass balance during Run 3.1 and 3.2

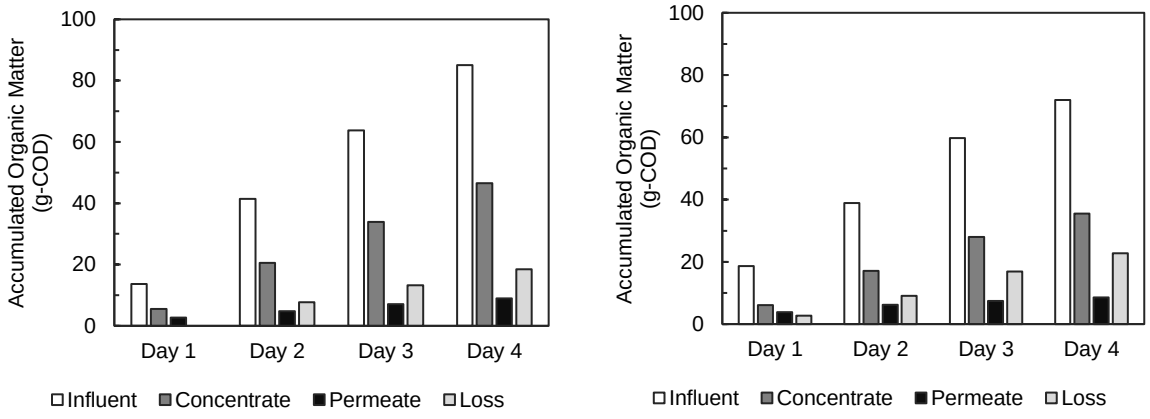


Figure S1. COD mass balance in Run 3.

S3. Mass balance during Run 4

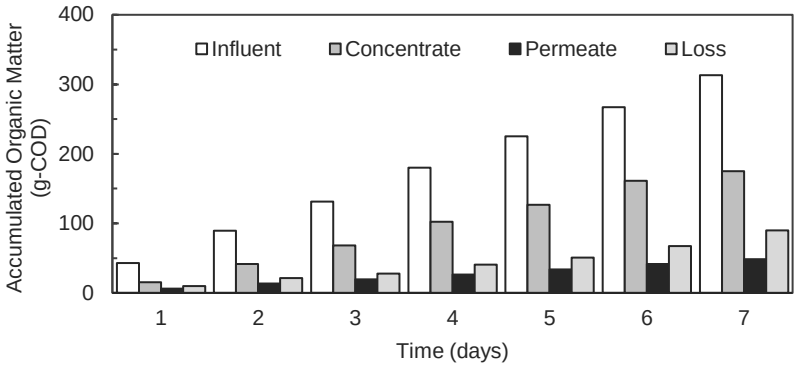


Figure S2. COD mass balance in Run 4.

S4. Clean water permeability results of the long-term experiments (Runs 5-7)

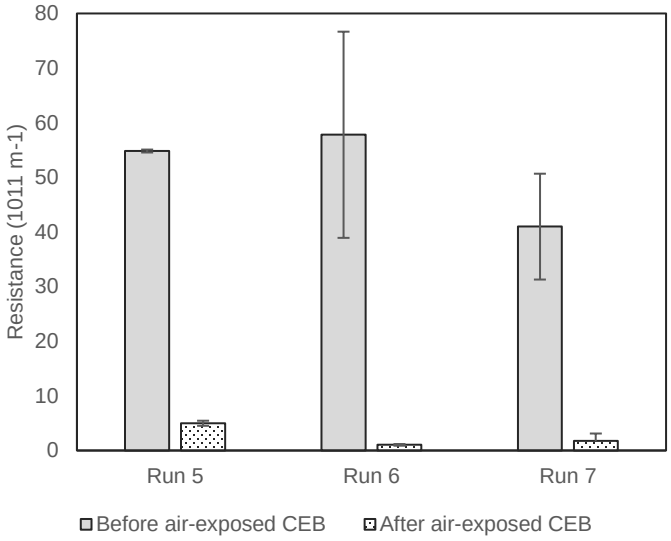


Figure S3. Fouling layer resistances measured before and after the air-exposed CEB measurements for Runs 5-7. Intrinsic membrane resistance has been subtracted from all results. Two membranes were subject to this analysis to ascertain the spatial differences in the reactor.

S5. Optimization experiments for the reduction of chemical concentrations in the air-exposed CEB.

Table S2. Details of the air-exposed CEB procedures used during optimizations S.1 and S.2 in this study.

	Starting Condition	CEB Details		
		Solution	Flux	Duration
Op. S1	When TMP reached 30 kPa	1000 mg/L oxalic acid followed by 1000 ppm NaClO	25 LMH	5-min backwashing for each solution
Op. S2	When TMP reached 30 kPa	1000 mg/L oxalic acid followed by 1000 ppm NaClO	25-100 LMH	5-min backwashing for each solution.

Two additional optimization tests (Section 3.3.2.3) were conducted to optimize for low concentrations. The details are outlined in Table S.1. The first optimization test was carried out to investigate what would happen if instead of doing the air-exposed CEB in two separate blocks, shorter air-exposed CEBs were done while changing from one solution to the other and back again (i.e., first acid, then NaClO, then acid again and repeating). After four repetitions of both oxalic acid and NaClO, a permeability recovery of 49.4% was achieved. Therefore, another improvement would be necessary to achieve better permeability recovery.

During the initial optimization test with a low concentration, a spot of clean membrane was observed, which, after a short while, would not extend, even when solutions were exchanged. It appeared as if all of the CEB solution was exiting the membrane in the same clean spot and not permeating throughout the membrane surface. Preliminary tests showed that an increase in flux could help increase the area where cleaning would take place. Thus, the second optimization test was carried out in the same conditions but with increasing fluxes that would solve the previously detailed problem.

The second optimization test for the low concentration condition used increasing flux, in steps, for each cycle. Starting at 25 LMH, both 1000 mg/L oxalic acid and 1000 ppm NaClO were backwashed for 5 minutes each. Five minutes were allotted between each backwash to change the solution in the system manually. The second time the oxalic acid solution was introduced, the flux was increased to 50 LMH and both solutions were used for backwashing, once again. This procedure was repeated for 75 and 100 LMH. After the four cycles, permeability recovery of 74% was achieved, and the appearance of cleaning was more evenly spread across the membrane surface.

S6. Accumulated mass balance during Run 7.

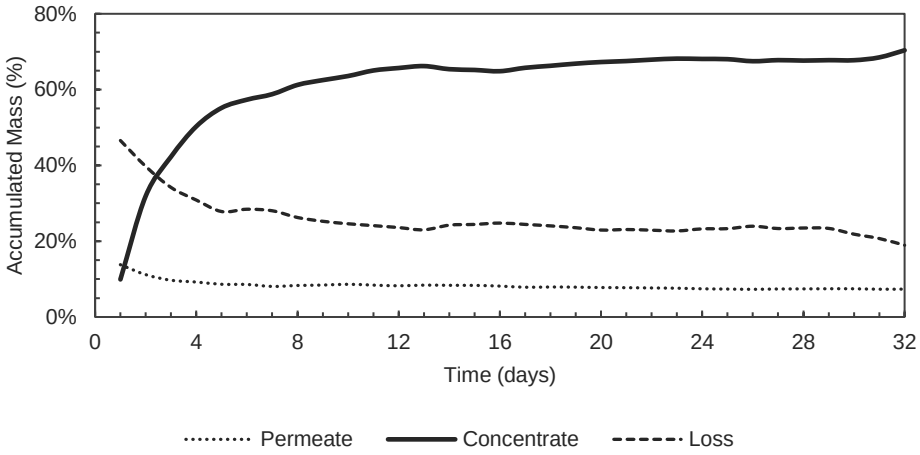


Figure S4. Accumulated mass percentages compared to accumulated influent COD mass of Run 7 in which the air-exposed CEB was conducted at day 29.

APPENDIX B. PUBLICATIONS AND CONFERENCE PROCEEDINGS

Publications

Published:

Rocco, M.J., Hafuka, A., Tsuchiya, T., Kimura, K., 2023. Efficient Recovery of Organic Matter from Municipal Wastewater by a High-Rate Membrane Bioreactor Equipped with Flat-Sheet Ceramic Membranes. Membranes 2023, Vol. 13, Page 300 13, 300. https://doi.org/10.3390/MEMBRANES13030300

Under Review

Rocco, M.J., Makizuka, H., Hafuka, A., Tsuchiya, T., Kimura, K., 2024. Effective reset of fouling in a high-rate MBR by in-situ, air-exposed and chemically enhanced backwash: a cautionary tale for granular scouring use. Journal of Water Process Engineering

Conference Proceedings

“Enrichment of Organic Matter Discharged from a High-Rate MBR Treating Municipal Wastewater”, IWA Particle Separation Conference, Boksburg, South Africa, 2023 (Oral presentation)

“Carbon recovery from municipal wastewater by a high-rate MBR operated with primary sedimentation”, The 60th Japan Annual Technical Conference on Sewerage (JATCS), Sapporo, Japan, 2023 (Oral presentation)