

CRANFIELD UNIVERSITY

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Anaerobic Membrane Bioreactors for water reuse using municipal
wastewater: the role of post-treatment

Cranfield Water Science Institute
School of Water, Energy and Environment

PhD
Academic Year: 2018 - 2021

Supervisor: Dr. Marc Pidou
Associate Supervisor: Prof. Paul Jeffrey
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This thesis is submitted in partial fulfilment of the requirements for
the degree of PhD

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ABSTRACT

Anaerobic Membrane Bioreactors (AnMBRs) are seen as a promising alternative to Aerobic Membrane Bioreactor (AeMBR) based water reuse schemes as they better support a circular economy paradigm with the potential for recovery of energy and nutrients. However, evidence of their application for water reuse is very limited which significantly restricts their potential deployment. This research aimed to identify the current challenges of using AnMBRs for water reuse with respect to their ability to achieve the quality requirements in state of the art national and regional standards. The work investigates the performance and feasibility of technologies commonly applied as a post-treatment stage for AeMBRs and ultimately to provide references for possible treatment trains for future water reuse implementations. A critical review and controlled pilot scale AnMBR and AeMBR operations followed by lab-scale post-treatment trials were conducted to understand the performance of the investigated post-MBR processes and their potential role in AnMBR based water reuse applications. The distinctive matrices of AnMBR and AeMBR effluents, in particular the different nitrogen species as ammonia in the AnMBR effluent and nitrate in the AeMBR effluent, were found to influence different performance across the investigated post-MBR technologies. The presence of ammonia caused a higher membrane fouling and a potential failure to meet the standard for potable reuse during the RO filtration of the AnMBR effluent. When chlorinated, the AnMBR effluent provided a controllable residual ammonia and chlorine concentration while exhibiting lower disinfection by-products formation potential compared to the AeMBR effluent. UV/TiO₂ delivered a selective removal of organic and nutrient compounds as a function of varying the UV intensity and TiO₂ dose from the AnMBR effluent. These findings highlight the potential to combine these processes to achieve more sustainable treatment trains producing high quality effluents for various water reuse applications. In particular, the combination of AnMBR-Chlorination shows promise as a circular economy approach to municipal wastewater treatment for agriculture irrigation.

Keywords:

Anaerobic membrane bioreactor (AnMBR), Sustainable wastewater treatment; Standards; Agriculture irrigation; Indirect potable water reuse (IPR); Reverse osmosis (RO); Disinfection by-products (DBPs); UV/TiO₂; Treatment train.

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LIST OF ABBREVIATIONS

3D EEM FRI	Fluorescence excitation–emission matrix regional integration
AC	Activated carbon
Ae	Aerobic
AeMBR	Aerobic membrane bioreactor
AH	Animal husbandry
An	Anaerobic
AN	AnMBR with no sparging
AnMBR	Anaerobic membrane bioreactor
AnMDBR	Anaerobic membrane distillation bioreactors
AnOMBR	Anaerobic osmotic membrane bioreactor
AOC	Aromatic organic compounds
AOPs	Advanced oxidation processes
AP	Aromatic protein
APr	Aromatic protein tyrosine-like
APU	At point of use
APy	Aromatic protein tryptophan-like
ARB	Antibiotic resistant bacteria
ARGs	Antibiotic resistance genes
ASR	Augmentation of Surface Water Supply Reservoirs
BAP	Biomass associated products
BCAA	Bromochloroacetic acid
BDCAA	Bromodichloroacetic acid
BDCM	Bromodichloromethane
BDL	Below detection limit
BOD ₅	Five-day biochemical oxygen demand
C	Ceramic
C-DBPs	Carbonaceous DBPs
CG	Conventional Granular
CH ₄	Methane
CIP	Ciprofloxacin
COD	Chemical oxygen demand

CRFC	Combined residual free chlorine
CSTR	Completely Stirred Tank Reactor
CT	Ceramic Tubular
DBAA	Dibromoacetic acid
DBCAA	Dibromochloroacetic acid
DBCM	Dibromochloromethane
DBP	Disinfection by-products
DBPFP	Disinfection by-product formation potential
DCAA	Dichloroacetic acid
DFF	Downflow Floating media Filter
DI	Deionised
DM	Dynamic Membrane
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
DPR	Direct potable reuse
Dv50	Size point below which 50% of the material is contained
EC/E. Coli	Escherichia coliforms
ECs	Emerging contaminants
EDs	Endocrine disruptors
EDS	Energy dispersive spectroscopy
EDS	Energy dispersive spectroscopy
EEM	Excitation–emission matrices
EPSs	Extracellular polymeric substances
EU	European Union
F	Flat sheet
F	Flocculant Sludge
FA	Fulvic acid-like
FBR	Fluidized-Bed Reactors
FC	Faecal coliforms
FCN	Non food crops
FCP	Processed food crops
FCR	Food crops consumed raw
FO	Forward osmosis

FP	Formation potential
FRC	Free residual chlorine
FRI	Fluorescence regional integration
FTIR	Fourier transform infrared spectroscopy
G	Granular Sludge
GAC	Granular Activated Carbon
GACF	Granular Activated Carbon Fluidized
GC-MS	Gas chromatography mass spectrometry
GC- μ ECD	Gas chromatography with a micro electron capture detector
GRI	Groundwater recharge by injection into potable aquifers
GRS	Groundwater recharge by spreading into potable aquifers
GS	Gas Sparging
H	Hollow Fibre
HA	Humic acid-like
HAAs	Haloacetic acids
HRT	Hydraulic retention time
IC	Inorganic carbon
ICP-MS	Inductively coupled plasma-mass-spectrometry
IEX	Ion exchange
IND	If not disinfected
IPR	Indirect potable water reuse
LMH	Litres per square metre hour
LRV	Log reduction value
Ma	Maximum
MA	Methanogenic archaea
MBAA	Monobromoacetic acid
MBR	Membrane bioreactor
MCAA	Monochloroacetic acid
MD	Membrane distillation
Me	Median
MF	Microfiltration

MRW	Mixed raw sewage
MSA	Membrane surface area
MW	Molecular weight
MY	Methane yield (L CH ₄ /g COD _{removed})
N	Nitrogen
N/A	Not available
NCl ₃	Trichloramine
N-DBPs	Nitrogenous DBPs
NDMA	N-Nitrosodimethylamine
NF	Nanofiltration
NGS	Non Gas Sparging
NH ₂ Cl	Monochloramine
NH ₃ -N	Ammonia nitrogen
NH ₄ ⁺	Ammonium nitrogen
NHCl ₂	Dichloramine
NO ₂ -N	Nitrite nitrogen
NO ₃ -N	Nitrate nitrogen
NOM	Natural organic matter
OC	Once-through Cooling
OFMSW	Organic fraction of municipal solid waste
OLR	Organic loading rate
OMPs	Organic micro-pollutants
Or-N	Organic nitrogen
Or-P	Organic phosphorus
P	Phosphorus
PAC	Powdered Activated Carbon
PCPs	Personal care products
PFPS	Plug Flow Partially Stirred
PhCs	Pharmaceutically compounds
PO ₄ ³⁻	Phosphate
PS	Particle Sparging
PVDF	Polyvinylidene difluoride
R ²	Coefficient of determination value

RA	Restricted area
RCT	Recirculation Cooling Towers
RM	Rotating Membrane
RO	Reverse osmosis
ROC	RO concentrate
ROP	RO permeate
ROS	RO membrane surface
RS	Raw sewage
RSW	Raw surface water
SAFMBR	Staged Anaerobic Fluidized Membrane Bioreactor
SBR	Sequencing batch reactor
SEM	Scanning electron microscope
SEM	Scanning electron microscopy
SG	Sponge Assisted-Granular
SiCF	Silicon Carbide Ceramic Flat sheet
SMF	Street maintenance and firefighting
SMPs	Soluble microbial products
SPE	Synthetic primary effluent
SRB	Sulfate reducing bacteria
SRT	Sludge retention time
SS	Settled sewage
SUVA	Specific ultraviolet absorbance
SVI	Sludge volume index
SW	Synthetic wastewater
T	Tubular
TBAA	Tribromoacetic acid
TBM	Bromoform
TC	Total carbon
TC	Total coliforms
TCAA	Trichloroacetic acid
TCM	Chloroform
TDS	Total dissolved solids
THAAFP	Total HAAFP

THMs	Trihalomethanes
TKN	Total Kjeldahl nitrogen
TN	Total nitrogen
TOC	Total organic carbon
TP	Total phosphorus
TrOCs	Trace organics compounds
TSS	Toilet sewage
TSS	Total suspended solids
TSW	Treated surface water
TTHMFP	Total THMFP
UA	Unrestricted area
UAP	Utilization associated products
UASB	Upflow Anaerobic Sludge Bed
UF	Ultrafiltration
UP	Ultrapure
USA	United states of America
VOC	Volatile organic compounds
WQIs	Water quality indicators

1 Introduction

1.1 Background

Water reuse is widely recognised as a useful response to increasing global water scarcity (Rebelo et al., 2018; Salgot & Folch, 2018), offering opportunities to maximise the value of wastewater as well as recover nutrients, energy, and other constituents (Smol et al., 2020; Song et al., 2018). Reuse can help reduce the cost and footprint of water and wastewater treatment (Aquim et al., 2019; Xu et al., 2020), and ultimately transform conventional linear water cycles into a sustainable circular economy approach (The European Commission, 2015; Vivaldi et al., 2022; Voulvoulis, 2012). To achieve this ambition, advancement of progressive standards and regulations has been made at national or regional levels to boost the implementation of water reuse schemes (ISO, 2018; Sgroi et al., 2018). Such regulatory requirements commonly set a quantitative threshold for a wide range of water quality indicators (WQIs) to ensure the quality of the recycled water is fit for purpose, covering organics, solids, nutrients and pathogenic indicators. Recovered water can be used for a variety of applications including potable uses (e.g. direct potable reuse (DPR) into drinking water supply and indirect potable reuse (IPR) for the recharge of surface water or groundwater aquifers), sub-potable uses such as toilet flushing, and most commonly for irrigation (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; PUB, 2020; USEPA, 2012, 2017).

Identified challenges relating to progressing a more circular economy model for water resources management range from public perception issues to financial viability of schemes, lack of regulatory maturity, and development of appropriate technologies and systems (Kirchherr et al., 2018; Voulvoulis, 2018). Perhaps the principal area of concern around technologies is the challenge of developing practicable treatment trains which are capable of generating water which meets regulated levels of contaminants (specific to its use) but avoids the intensive energy consumption often associated with membrane based reuse systems (Morris et al., 2021; Tang et al., 2018a). More research is needed to identify and develop effective treatment options for reuse which are also capable of

performing against financial and environmental performance metrics (Capocelli & Piemonte, 2021). In this context, Membrane Bioreactor (MBR) technologies have been studied to act as a means of producing a solids and pathogen free effluent, while maintaining a robust effluent quality sufficient for various non-potable reuse applications (Delanka-Pedige et al., 2020; Winward et al., 2008). In addition to studies of the more typical Aerobic MBR (AeMBR), the performance attributes of Anaerobic Membrane Bioreactors (AnMBR) for the treatment of municipal wastewater have also been assessed (Shin & Bae, 2018; Visvanathan & Abeynayaka, 2012).

Current water reuse schemes for the treatment of municipal wastewater commonly consist of primary, secondary, tertiary and advanced treatment process as the treatment train to ensure the effluent meets the requirement (Shareefdeen et al., 2015). AeMBR based applications both provide a treatment function at secondary and tertiary stages for non-potable water reuse applications while also acting in other roles. Examples of such ancillary benefits include serving as a reliable protector of reverse osmosis (RO) membranes in potable reuse schemes by removing the fine solids and colloids to prevent RO channel blockage (CWB, 2021; Visvanathan et al., 2000), or as a stable effluent producer before the disinfection step in agricultural irrigation, eliminating a significant amount of organic matter to increase the efficiency of disinfection process (Xu et al., 2020; Zaouri et al., 2021), or as a durable upstream promoter for advanced oxidation processes (AOPs) in potable / sub-potable reuse implementations, providing a higher quality permeate to reduce the catalyst and energy consumption in photocatalytic AOPs (Baumgarten et al., 2007; Sarp et al., 2011). The AnMBR configuration is seen as a competitive challenger to AeMBRs, which are widespread in modern water reuse applications, and recognised as the dominant technology employed in reuse schemes treating municipal wastewater (Capocelli & Piemonte, 2021; Jeffrey et al., 2022; Shareefdeen et al., 2015). However, knowledge about the performance of AnMBRs for water reuse is fragmented and incomplete compared with that for AeMBRs. This knowledge gap constrains the potential exploitation of AnMBRs in full scale reuse schemes.

The oxygen free operational environment of an AnMBR has no aeration associated energy demand and the methane rich biogas generated can be converted to heat or electricity to further reduce the energy demand of the overall treatment train (Robles et al., 2018; Song et al., 2018). Indeed, AnMBRs can achieve a similar organic pollutant removal as AeMBRs, while maintaining a low solids production and hence a reduced cost in sludge treatment, as well as offering great potential for irrigation reuse applications as the nutrients (Nitrogen and Phosphorus) remain in the effluent (Foglia et al., 2019; Robles et al., 2018; Shin & Bae, 2018; Visvanathan & Abeynayaka, 2012; Wang et al., 2018). However, there remains uncertainty as to whether AnMBR effluent could satisfy the quality requirements set in the reuse standards for irrigation purposes. As such, while offering the advantage of avoiding the intensive energy consumption of current water reuse applications, the AnMBR effluent may offer a different set of challenges to the performance of post-treatment technologies compared to those generated by an AeMBR. As noted above, the specific end use of recycled water will determine the water quality criteria imposed by regulators. Hence, this study adopts two sets of well-established and internationally influential standards as reference points for desirable effluent water quality: Singapore's PUB NEWater standard (PUB, 2020) for IPR and the USA's EPA standard (USEPA, 2012) for non-potable reuse.

Two important areas of understanding remain unexplored in this context. Firstly, AnMBR effluent exhibits a different matrix in comparison to AeMBR effluent due to the dissimilar biological process before the membrane barrier. In particular, the unremoved nutrients found in AnMBR effluents has been shown to be one of the key differences between AnMBR and AeMBR performance (Skouteris et al., 2012; Wang et al., 2018; Xu et al., 2020). Secondly, most reuse standards have set a compulsory 1 mg/L residual chlorine concentration to give an additional protection against the microbial risk for the delivered water (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; PUB, 2020; USEPA, 2012, 2017) but there is yet to be studies evaluating the potential for disinfection and disinfection by-product formation in anaerobically treated effluents. Overall, this highlights the need to better understand the impact of AnMBR effluent produced during

municipal wastewater treatment on advanced treatment technologies for water reuse. It should be noted that the nutrients present in the AnMBR effluent will have to be removed in post-treatment processes to achieve an adequate quality for the reuse applications that have a strict nutrient consents; for example the <1 mg/L ammonia nitrogen concentration in Singapore's PUB NEWater standard for IPR (PUB, 2020). Hence understanding the impact of unremoved nutrients on the performance of candidate post-AnMBR treatment options becomes of importance. Processes such as RO and photocatalysis processes applied for potable uses may be affected differently when treating an AnMBR effluent rather than an AeMBR effluent.

The few previously reported studies on the use of AnMBRs for reuse have only investigated these aspects of AnMBR performance in passing and / or under restricted operating conditions. For example, investigating the post-treatment of AnMBR effluent, Mai et al. (2018) reported excellent removal of Ciprofloxacin (CIP) and organic compounds by activated carbon adsorption. Also, Augsburger et al. (2021) demonstrated the potential application of UV/H₂O₂ to facilitate the removal of emerging contaminants in the AnMBR effluent, the levels of three pharmaceutical compounds, antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARGs) were reported as effectively reduced. Shin et al. (2021) evaluated the ammonia nitrogen removal from AnMBR effluent by RO membranes and demonstrated that the pH significantly affects the ammonia rejection rate by changing the ammonium/ammonia speciation and membrane surface charge. However, the majority of relevant studies available in the literature were either conducted with a synthetic wastewater (Mai et al., 2018b), or operated under mesophilic (35°C) temperatures (Augsburger et al., 2021; Mai et al., 2018b). In particular, none of these studies focused on the post-treatment of AnMBR effluent for water reuse implementations. It should also be noted that there is no literature on other post-treatment options for MBR effluents such as chlorination or UV/TiO₂ as applied to AnMBRs. Moreover, there is very limited information available on the direct comparison of the post-treatment of AnMBR and AeMBR effluents which would help established clearer distinctions between

the two in terms of their relative benefits and challenges. Overall, this raises the following research questions:

- What are the potential and limitations of AnMBR treating municipal wastewater for water reuse?
- How does the AnMBR effluent matrix impact the performance of post-treatment technologies and how does this compare to AeMBR?
- How does the inclusion of AnMBR influence treatment train selection for future sustainable water reuse scheme implementations?

Answering these questions will provide the intellectual contribution to better understand AnMBRs in the context of water reuse and then support treatment train selection for future sustainable water reuse scheme implementations.

1.2 Aim and objectives

The aim of this thesis is to understand the impact of the AnMBR effluent matrix from the treatment of municipal wastewater on selected post-treatment processes including reverse osmosis, chlorination, and UV/TiO₂ within the context of water reuse.

In this regard, four objectives will be pursued (also see Figure 1-1):

1. Understand the potential and limitation of AnMBR treating municipal wastewater for water reuse applications, compare and contrast existing literature with current water reuse standards;
2. Determine the performance of selected post-treatment technologies for the treatment of AnMBR effluent, with qualitative and quantitative analysis of the fate of key contaminants monitored in water reuse regulations;
3. Deliver a comparative study of the impact of AnMBR and AeMBR effluents on the post-treatment options selected for various water reuse applications;
4. Define sustainable treatment trains comprising the examined post-treatment processes for AnMBR based water reuse schemes and highlight the advantages and disadvantages when compared to current practice.

1.3 Thesis structure

This thesis is structured with chapters formatted as papers for publication in academic journals, with in addition Chapter 1 as the introduction providing the background and context of the work, Chapter 6 as the overall discussion of the work, and Chapter 7 as the conclusion of the conducted research as well as highlighting suggested future work. The title and publication progress of each chapter with the corresponding objectives are highlighted in Figure 1-1 also listed in Table 1-1.

All chapters were written by the first author, Yu Huang and edited by the supervisors Dr. Marc Pidou and Professor Paul Jeffrey. All laboratory work was planned and conducted by Yu Huang, with the help from Miss Tamazon Cowley for the DBP analysis in Chapter 4, and Mrs Nuannat Simmons for the trace elements analysis in Chapters 3 and 4.

Chapter 2: Municipal wastewater treatment with Anaerobic Membrane Bioreactors for sub-potable reuse: a review. This chapter is a critical review of the state of the art on AnMBRs applied to municipal wastewater treatment to understand the potential and challenges of their implementation for sub-potable water reuse applications.

Chapter 3: A comparative evaluation of reverse osmosis membrane performance when combined with Anaerobic or Aerobic membrane bioreactors for indirect potable reuse applications. In this chapter, the filtration performance and fouling behaviour of reverse osmosis membrane treating the AnMBR and AeMBR effluents for indirect potable implementations is evaluated.

Chapter 4: Understanding chlorine based disinfection processes and disinfection by-product formation in Anaerobic and Aerobic MBR effluents for reuse applications. In this chapter, a comparative analysis of chlorination of AnMBR and AeMBR effluents is carried out providing a detailed evaluation of DBP formation behaviour.

Chapter 5: UV/TiO₂ photocatalysis as a post-treatment option for anaerobic membrane bioreactor effluent in the context of reuse. This chapter aims to

understand the potential and limitation of TiO_2 photocatalytic process for the treatment of AnMBR and AeMBR effluents and evaluate the impact of the different matrices in terms of organics and nutrients on the process.

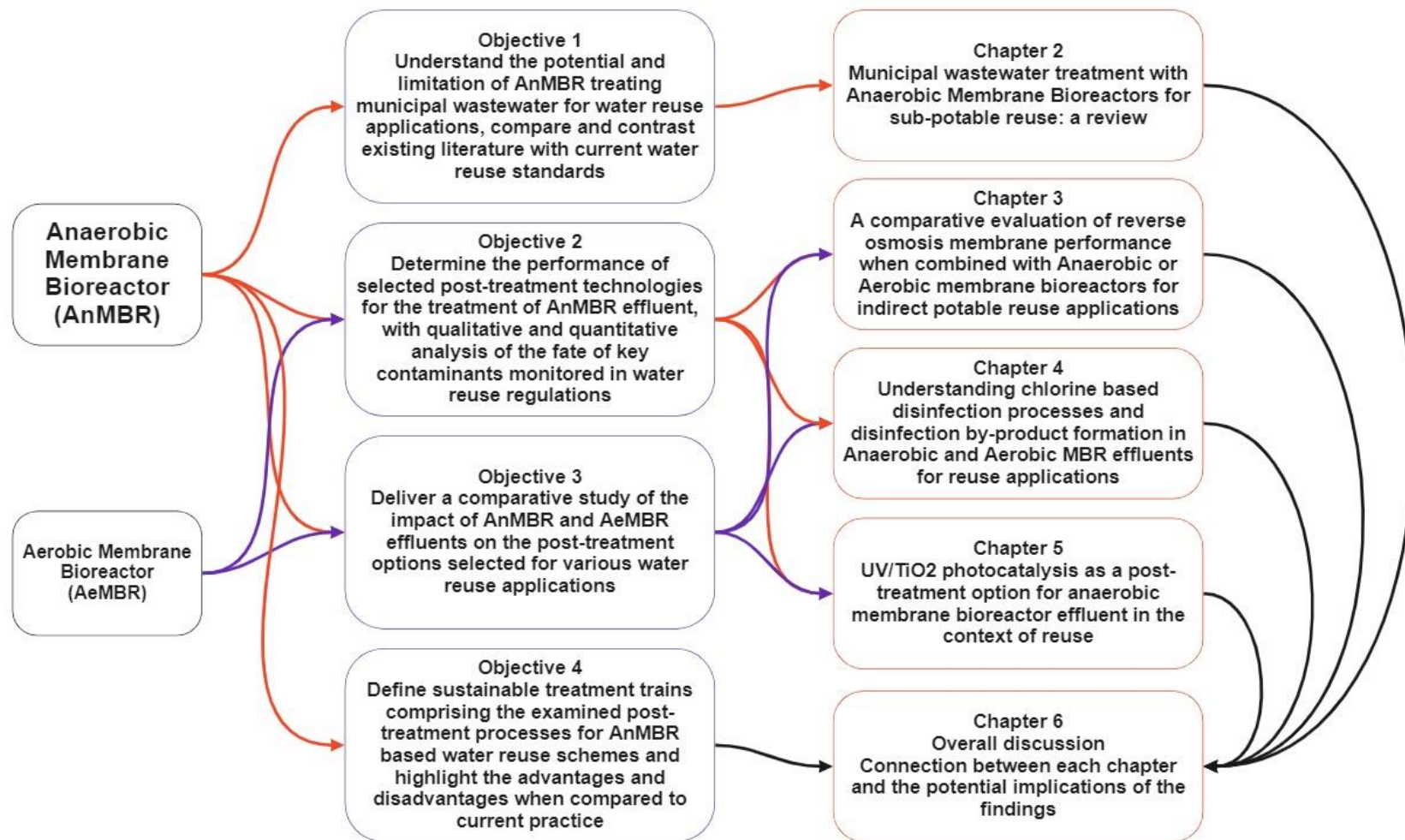


Figure 1-1 Thesis objectives and related chapters

Table 1-1 Thesis contents and the publication progress

Chapter	Title	Link to Objectives	Target journal
1	Introduction	-	-
2	Municipal wastewater treatment with Anaerobic Membrane Bioreactors for sub-potable reuse: a review	1	Critical Review of Environmental Science and Technology
3	A comparative evaluation of reverse osmosis membrane performance when combined with Anaerobic or Aerobic membrane bioreactors for indirect potable reuse applications	2,3,4	Journal of membrane Science
4	Understanding chlorine based disinfection processes and disinfection by-product formation in Anaerobic and Aerobic MBR effluents for reuse applications	2,3,4	Water Research
5	UV/TiO ₂ photocatalysis as a post-treatment option for anaerobic membrane bioreactor effluent in the context of reuse	2,3,4	Chemosphere
6	Overall discussion	1,2,3,4	-
7	Conclusion	-	-

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2 Municipal wastewater treatment with Anaerobic Membrane Bioreactors for sub-potable reuse: a review

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Abstract

Anaerobic Membrane Bioreactors (AnMBRs) are seen as a promising technology for application in water reuse schemes. However, the evidence base for their potential and efficacy in this regard is fragmented. We draw together this disparate knowledge base to offer a state of the art review of municipal wastewater treatment with AnMBRs and evaluate the technology's potential application for water reuse. Water quality regulations and standards from different regions of the world are used as performance metrics to compare and contrast the treatment performance of pilot and laboratory scale AnMBR systems reported in the literature (n=50). Findings indicate that under stable operation, AnMBRs have the potential to produce water for agricultural reuse. However, without post-treatment, AnMBRs are incapable of delivering water that meets other non-potable reuse standards across a range of important parameters such as COD, BOD₅, NH₃-N and TP. Analysis of key operational parameters cover hydraulic retention time, temperature, membrane configuration and material. An assessment of candidate post treatment technologies suggests a trade-off between the cost and effluent quality based on the reuse application requirement. We conclude by discussing a number of challenges and limitations to the use of AnMBRs for reuse applications in order to outline a pathway to maturity for effective treatment trains.

Keywords:

water reuse, anaerobic membrane bioreactor, standards, post-treatment

2.1 Introduction

Water stress is an increasingly urgent and widespread global challenge with water shortages highlighting the need for sustainable water management strategies (Song, Luo, Hai, et al., 2018). Water reuse is a useful foil to such water scarcity, offering opportunities to recover nutrients and energy, helping to offset the cost of wastewater treatment (Winpenny et al., 2010), and leading to a transformation of conventional linear water cycles into circular economy solutions with enhanced economic value (Smol et al., 2020; Voulvoulis, 2012). Progressive regulations and policies are needed to ensure the success of water reuse implementation (Sgroi et al., 2018) and recent years have seen major advances in the provision of supportive regulatory frameworks at national and supra-national levels. For example, the European Union has made water reuse a prominent theme in its circular economy policy to provide significant environmental, economic and social benefits (The European Commission, 2015) and China has underpinned growth in water reuse schemes by publishing standards on reused water quality (CNEPA, 2021). Other jurisdictions have updated existing guidelines in response to growth in the reuse sector (USEPA, 2012). These regulatory interventions are typically focused on forms of reuse which show particular promise or opportunity within a national or regional context and set quantitative limits for a range of water quality parameters, such as organics, solids, nutrients and pathogenic indicators (CNEPA, 2021; EUPC, 2020; NRMMC, 2008; USEPA, 2012).

Conventional water and wastewater treatment trains usually include up to three stages: primary (preliminary), secondary and tertiary treatment (Shareefdeen et al., 2015), aiming to remove settleable solids, suspended solids, biodegradable organic matter, nutrients, and pathogens. However, these processes in isolation often cannot produce an effluent that meets non-potable standard consents and therefore advanced treatment is needed to deliver a final effluent with sufficient quality for reuse. Several advanced treatment technologies including physical (absorption and membrane), biological (algae, wetland and anaerobic reactor) and chemical (ion exchange, advanced oxidation processes) processes have been reported to produce effluents of sufficient qualities for different reuse

purposes (Aquim et al., 2019). Among these options, membrane processes have spurred most interest in recent years as they offer easy operation and a high quality effluent with a high degree of selectivity (Sgroi et al., 2018). Membrane Bioreactors (MBRs) combine membrane filtration with conventional activated sludge treatment, the membrane rejects the solid materials which are developed by the biological process, effectively removing solids, organics, nutrients, and turbidity as well as achieving a noteworthy level of disinfection (Judd, 2011).

MBRs can be operated in both aerobic and anaerobic conditions depending on the presence or absence of oxygen in the biological reactor (Huang & Lee, 2015). Anaerobic treatment offers a number of advantages over aerobic comparators as, for example, the process leads to lower levels of biosolids to dispose of due to the low growth yield of the anaerobic biomass (Giménez et al., 2011). Degradation of organic matter and its conversion to methane-rich biogas provides a renewable source of energy but depends on the symbiotic relationship among different groups of microorganisms such as fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenotrophic methanogens and aceticlastic methanogens (Chen et al., 2016). Of these microorganism groups, methanogens play arguably the most important role for biogas production by converting intermediate products to methane gas which can be used to offset the energy demand for wastewater treatment (McCarty et al., 2011). Since anaerobic treatment converts nutrients to chemically available forms as ammonia and phosphate, AnMBRs can facilitate nutrient recovery via subsequent precipitation (Shin & Bae, 2018). By integrating membrane separation processes such as microfiltration (MF) or ultrafiltration (UF) with anaerobic reactors, the hydraulic retention time (HRT) can be decoupled from the sludge retention time (SRT) (Pileggi & Parker, 2017), allowing the slow growth of anaerobic bacteria and also maintaining higher biomass concentration to provide higher treatment capacity (Berkessa et al., 2018). Thereby, more precise process control such as high organic loading rate and short HRT can be achieved, resulting in an efficient conversion of organic compounds to biogas due to increased active biomass densities in the bioreactor (Tassew et al., 2019). Additionally, as a membrane based system, AnMBRs eliminate TSS and turbidity concerns and have been

shown to be effective in the removal of both bacteria (MF) and viruses (UF) (Mai et al., 2018a).

Historically, AnMBRs have been typically deployed to deal with high strength wastewaters (Futselaar et al., 2013), with multiple studies proving their benefits when applied to the treatment of municipal wastewater (Shin & Bae, 2018; Song et al., 2018; Wang et al., 2018), and demonstrating that their use can reduce asset footprint, energy consumption and HRT (Shareefdeen et al., 2015). Useful reviews reporting AnMBR fundamentals, system configuration and performance for wastewater treatment can be found in Skouteris et al. (2012), Shin & Bae (2018), and Song et al. (2018). However, and of significance to this contribution, the exploitation of AnMBRs in the context of water reuse has attracted little attention from researchers and technologists. Of those explicitly referencing reuse as part of the performance objective, Gu et al. (2019) report that the product water of an AnMBR followed by reverse osmosis (RO) and ion exchange (IEX) is better than Singapore's NEWater quality standard for industry and indirect potable reuse. Nevertheless, there remain several significant challenges in the development of AnMBRs for resource recovery from wastewater, particularly municipal wastewater. These include low organic and nutrient loads in municipal wastewater, process operation stability and effluent water quality, membrane fouling, along with further post-treatment required to meet the higher quality reuse standards (Evans et al., 2019; Mai et al., 2018a; Shin & Bae, 2018; Song et al., 2018).

Given the promise of AnMBRs as a facilitating technology for resource recovery and the need to premise future research on a thorough understanding of what has been a fragmented and poorly focused literature base, the following sections critically review the performance of AnMBRs in the context of water reuse. We respond to two queries: What evidence is there that AnMBRs have potential as a treatment option for water reuse applications? and Where should future research be focused to further develop the opportunities for AnMBR deployment in reuse applications? Regulations and standards published in different countries and jurisdictions are used to understand water quality targets and the potential of

AnMBR technology to produce effluent meeting existing standards for reuse applications. We also discuss potential post-treatment processes that might be used to meet stricter consents. Finally, remaining gaps in understanding which could further unlock the potential application of AnMBRs for water reuse are identified.

2.2 Sub-potable water quality standards

In order to provide quantitative pollutant removal benchmarks for assessing the performance of AnMBRs we have collected and collated data from water reuse regulations issued by a number of national and regional bodies (Supporting information 1-1). Our intention here is not to generate a comprehensive global listing of sub-potable reuse regulations but rather to provide a sample of frequently referenced regulations from parts of the world where reuse is more widespread. These quality standards provide maximum pollutant levels for water delivered for a variety of uses. Limits are prescribed for a range of pollutants including routine physical and chemical parameters such as solids, organics, nutrients, residual disinfectants etc., aesthetic parameters such as colour, and microbial parameters. Standards generally reflect the principle that those uses with higher likelihood of human contact are subject to more stringent control. Indicator bacteria levels are prescribed in all standards not involving aquifer recharge, with faecal coliforms being the most common parameter. Additional protection against microbial risk is often secured through the presence of a chlorine residual in the delivered water with a required concentration above 1 mg/L in most cases (although noteworthily not in the Chinese and Canadian standards which require only 0.5 mg/L for most uses). In all standards, required pH values are in a similar range irrespective of end use; between 5.5 to 9.0. However, there is significant variation in other regulated parameter values across the various standards (e.g. BOD₅, TSS and nutrients). Emerging organic contaminants such as pharmaceutical residues, endocrine disruptors and personal care product residues are poorly covered in reuse water quality standards with only the Australian national guidelines for the augmentation of drinking water supplies with recycled water referencing them (NRMMC, 2008).

Water reuse applications are generally divided into non-potable and potable groups (ISO, 2018) with standards reflecting the various risks emergent from using the recovered water for a specific purpose. For example, limits for nutrients such as total nitrogen and total phosphorus are commonly listed when the reuse application involves discharge to environmental and recreational water bodies so as to prevent eutrophication. To illustrate, Arizona and New Jersey in the USA have a limit of 10 mg/L in TN and the Chinese standard has limits of 15 mg/L in TN, 5 mg/L in $\text{NH}_3\text{-N}$ and 1 mg/L for TP. For municipal and commercial uses where the risk of human contact is highest (e.g. toilet flushing, street cleaning, fire-fighting), indicator bacteria consents are often listed as non-detectable. Reuse for agricultural irrigation is one of the most common end uses of reused water (Asano et al., 2007) with specific water quality classifications characterising regulatory approaches. The USA, Australia and the EU use a three class system; food crops consumed raw (FCR), processed food crops (FCP) and non food crops (FCN) whereas China divides the end uses more broadly into farming, forestry and animal husbandry. Faecal coliform limits vary widely from 200 CFU/100ml to non-detectable and 10000 CFU/100ml to 2000 CFU/100ml in the USA and Chinese standards, respectively. Organic consents referencing both biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD) are universally listed in reuse standards. The consents for BOD_5 in environmental and recreational uses provide a useful example of how problematic comparison of standards is across jurisdictions when only using headline water use descriptors. The lowest limit for BOD_5 for water used for 'recreational purposes' can be found in China at 6 mg/L whereas the respective value in the USA is 30 mg/L. The difference can be explained by the fact that the two countries have different definitions for this type of reuse. The Chinese regulations allow for the water to be used as a centralized drinking water source whereas the USA definition allows water-contact activities but not use as a drinking water source (CNMEE, 2002; USEPA, 2012). For municipal uses, the BOD_5 limit in our selected standards ranges from 10 mg/L to 30 mg/L, driven by minor differences in the end use application (e.g. toilet flushing has a lower limit compared with urban grounds irrigation). A summary of BOD_5 limit concentrations taken from

different regulations (Table 2-1) illustrates the broad relationship between human exposure and stricter water quality control as well as providing a quantifiable scale with which to evaluate and compare the performance of different AnMBR systems.

Table 2-1 BOD₅ concentration in various non-potable water reuse standards (mg/L)

≤35	China: Agricultural (Farming & Forestry).	
≤30	USA: Environmental, Recreational (RA), Landscape irrigation (RA), Agricultural (FCP & FCN), Industry (OC & RCT).	
≤25	EU: Agricultural (FCP & FCN), Industry.	
≤20	China: Landscape irrigation. Australia: Landscape irrigation, Agricultural (FCP & FCN).	
≤15	China: SMF, Industry (cooling & washing & Boiler).	
≤10	USA: Recreational (UA), Toilet flushing, SMF, Agricultural (FCR). China: Environmental, Toilet flushing, Agricultural (AH). Israel: Recreational, Landscape irrigation (UA & RA), Toilet flushing (UA). Canada: Toilet flushing. EU: Agricultural (FCR).	
≤6	China: Environmental (lakes), Recreational.	
Notes	RA: Restricted area; UA: Unrestricted area; FCR: Food crops with raw consumer; FCP: Food crops with processed consumer; FCN: None Food crops; AH: Animal husbandry; OC: Once-through Cooling; RCT: Recirculation Cooling Towers; SMF: Street maintenance and firefighting.	
Sources	USA	(USEPA, 2012)
	EU	(EUPC, 2020)
	China	(CNEPA, 2007)
	Australia	(NRMCC, 2008)
	Canada	(Health Canada, 2010)
	Israel	(ISO, 2018)

2.3 Methodology

Based on the success of early bench scale studies (Skouteris et al., 2012; Smith et al., 2012), research on AnMBRs moved on to make use of pilot-scale facilities (Shin & Bae, 2018). Most reported AnMBR studies were designed with commercial membrane modules and were fed with real domestic wastewaters to reflect diurnal and seasonal changes in the feed. However, to the authors' knowledge there are only three publications to date specifically focusing on the implementation of AnMBRs for water reuse; two of which have explored the performance of (respectively) a gas sparging AnMBR (Martinez-Sosa et al., 2011)

and an AnMBR followed by RO and IEX treating municipal wastewater (Gu et al., 2019) and a third lab scale project which assessed the use of an AnMBR to treat cheese whey with the treated effluent being recovered and used for site cleaning purposes (Ribera-Pi et al., 2020). However, AnMBR performance in treating municipal wastewater has been extensively explored and can be used in combination with the few studies noted above to provide a more detailed picture of the technology's potential application to water reuse.

Our evidence acquisition strategy involved (i) identifying suitable research paper repositories, (ii) application of selected search terms (in several stages) and rationalisation of the database records to remove duplicates, quality control entries, and ensure comparability of sources, and (iii) structuring of the resulting studies to facilitate data analysis. Scientific publication databases Scopus, Mendeley and Google Scholar and citation tracing were used to source literature initially using the keywords 'anaerobic' + 'membrane bioreactor' + 'wastewater'. A second search used the keywords 'AnMBR' + 'municipal wastewater' and a third search the keywords 'AnMBR' + 'water reuse'. Retrieved papers were filtered to identify studies which reported performance data at either pilot or full scale. The operational data from these studies were extracted to populate a database and a subset of tested technologies was identified (n=50) which allowed a comparative analysis of performance to be conducted. Table 2-2 provides a list of the AnMBR systems evaluated in this review. Additional technology and operational parameters are listed in Supporting information 1-2 including reactor volume, membrane surface area, hydraulic retention time, sludge retention time, organic loading rate, temperature, and flux along with a comparison of influent and effluent concentrations of typical water quality parameters. As stated above, water reuse regulations typically target BOD₅ as the organic content indicator. However, most studies focusing on wastewater treatment for discharge use COD and only 16 out of the 50 studies have recorded the effluent BOD₅ concentration whereas 37 out of 50 studies have COD data. For the purpose of the evaluation, considering the municipal wastewater effluent as a stable compensated wastewater, BOD₅ values were estimated from the respective COD values reported and the calculated average effluent BOD/COD ratio of 0.3 (± 0.1)

obtained from the 16 studies where both COD and BOD₅ measures were available.

Table 2-2 AnMBR systems included in this study

Reference	Source	Reference	Source
GS-F1	(Martinez-Sosa et al., 2011)	ANH1	(Pretel et al., 2016)
GS-H1	(Giménez et al., 2011)	GL-T1	(Dolejš et al., 2017)
GS-H3	(Robles et al., 2013)	ANH2	(Ferrari et al., 2019)
GS-H5	(Mei et al., 2017)	ANF1	(Vyrides et al., 2010)
GS-H6	(Dong et al., 2015)	GS-F3	(Zhou et al., 2016)
GS-H7	(Gouveia et al., 2015)	GS-F4	(Thanh et al., 2017)
GS-H8	(Gouveia et al., 2015)	GS-F5	(Kunacheva, Soh, & Stuckey, 2017)
PS-H1	(Shin et al., 2014)	GS-F6	(Trzcinski & Stuckey, 2016)
RM-H1	(Ruigómez et al., 2016)	GS-FPAC1	(Akram & Stuckey, 2008)
GS-C1	(Song et al., 2018)	GS-F7	(Fox & Stuckey, 2015)
GS-H15	(Wang et al., 2018)	GS-H16	(Martin Garcia et al., 2013)
GSG-H1	(Wang et al., 2018)	GSG-H2	(Martin Garcia et al., 2013)
GSF-H1	(Wang et al., 2019)	GS-F8	(Zhang et al., 2017)
GS-H9	(Wang et al., 2019)	GS-F9	(Xiao et al., 2017)
GS-H10	(Evans et al., 2019)	GS-F10	(Kunacheva et al., 2017)
GACF-H1	(Evans et al., 2019)	GS-H11	(Khan et al., 2019)
PFPS-H1	(Bair et al., 2015)	GS-H12	(Peña et al., 2019)
CSTR-H1	(Wei et al., 2014)	GS-F11	(Chen et al., 2019)
DFF-T1	(Seib et al., 2016)	GS-H13	(Seco et al., 2018)
FBR-CT1	(Seib et al., 2016)	GS-SiCF1	(Cho et al., 2019)
UASB-H1	(Petropoulos et al., 2019)	GSDM-F1	(Hu et al., 2018)
NGS-T1	(Cerón-Vivas & Noyola, 2017)	SAFMBR-1	(Chen et al., 2019)
GS-T1	(Cerón-Vivas & Noyola, 2017)	ANH3	(Ding et al., 2019)
CGH1	(Chen et al., 2017)	ANH4	(Ding et al., 2019)
SGH1	(Chen et al., 2017)	GS-H14	(Gu et al., 2019)
Notes	GS: Gas Sparging; PS: Particle Sparging; RM: Rotating Membrane; DM: Dynamic Membrane; NGS: Non Gas Sparging; F: Flat sheet; H: Hollow Fibre; C: Ceramic; T: Tubular; CT: Ceramic Tubular; SiCF: Silicon Carbide Ceramic Flat sheet; PAC: Powdered Activated Carbon; GACF: Granular Activated Carbon Fluidized; AN: AnMBR with no sparging; PFPS: Plug Flow Partially Stirred; CSTR: Completely Stirred Tank Reactor; FBR: Fluidized-Bed		

	Reactors; DFF: Downflow Floating media Filter; UASB: Upflow Anaerobic Sludge Bed; CG: Conventional Granular; SG: Sponge Assisted-Granular; G:Granular Sludge; F: Flocculant Sludge; SAFMBR: Staged Anaerobic Fluidized Membrane Bioreactor.
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2.4 Anaerobic Membrane Bioreactor performance

The 50 systems captured in our review demonstrate significant variability in the ranges of pollutant removal efficacy across a number of parameters (Figure 2-1). A consistent range of measured parameters is not available across all the 50 reviewed systems as some papers only record nutrient removal instead of organics; a fact reflected in the presented data sets below.

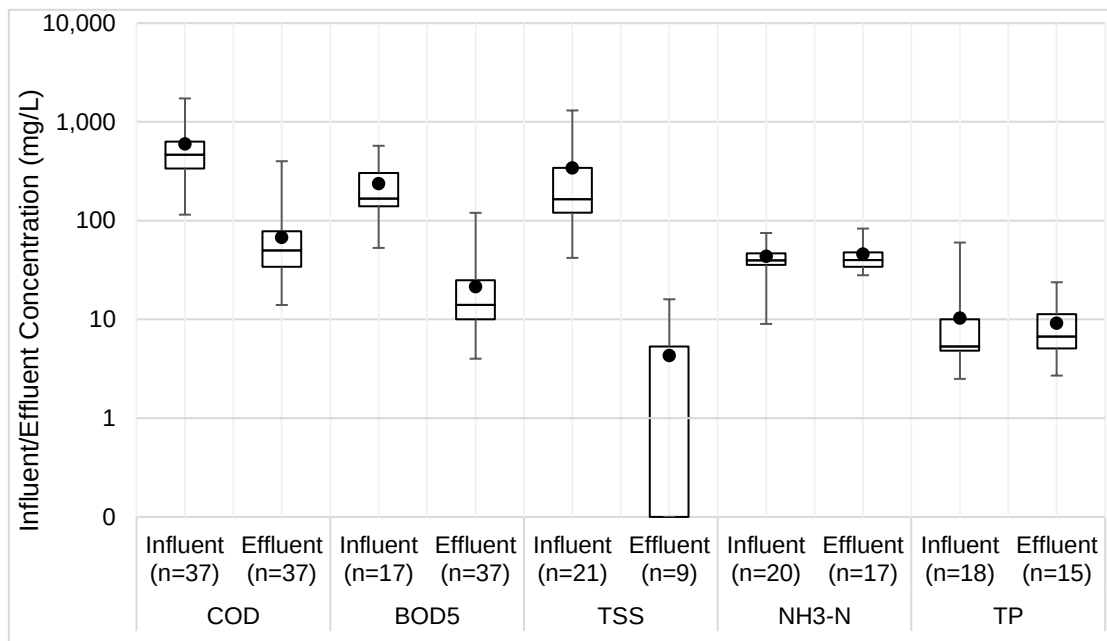


Figure 2-1 Influent/Effluent Concentration of AnMBR treating municipal wastewater

Notes: n represents the number of data sets available for each parameter. The black spot stands for the mean value for each parameter. The box represents the distribution of the concentration for the first quartile (bottom), median (middle) and third quartile (top) of the data and the error bar represent the maximum and minimum concentrations.

As AnMBRs possess an integrated membrane unit, there should be no TSS concern in the effluent water, which may explain the lack of data on this

parameter in the reported studies. Two systems reported relatively high effluent TSS concentrations of 16 mg/L (NGS-T1) and 15 mg/L (GS-T1) (Cerón-Vivas & Noyola, 2017). These unusual values, which would prevent the effluent being used for a number of reuse purposes, were reported to be due to biofilm growth on the permeate pipe walls in the study. Other reported systems are able to meet the TSS concentration limit for every reuse purpose in the various standards with five of them reporting not detected, GS-H9 and GS-H13 reported 5.33 mg/L and 2 mg/L, respectively (Chen et al., 2019; Martin Garcia et al., 2013; Rongwong et al., 2019; Shin et al., 2014; Wang et al., 2019).

It is also important to note that, as demonstrated by many previous studies (e.g. (Song et al., 2018) AnMBRs do not remove nutrients ($\text{NH}_3\text{-N}$ and TP). The effluent $\text{NH}_3\text{-N}$ concentrations ranges from 28 mg/L to 83.2 mg/L as the effluent TP concentrations varies from 2.7 mg/L to 23.8 mg/L. Among all non-potable reuse standards, none of AnMBR systems could directly produce an effluent satisfying the nutrients consent of $\text{NH}_3\text{-N}$ and TP. To illustrate, the $\text{NH}_3\text{-N}$ has a limit ranging from 5 mg/L to 20 mg/L for various uses in the Chinese reuse standard but none of the AnMBRs could meet this consent and no system could meet the highest TP limit of 1 mg/L.

Another significant concern for reused water is the pathogen concentration as it constitutes one of the most important criteria for reused water quality (Asano et al., 2007; ISO, 2018; Lazarova, 2015). Pathogen requirements for non-potable water is evaluated using the concentration of three indicator coliform measures: faecal, *Escherichia* (E.) and total coliforms. Every standard has a numerical limit for different indicator bacteria with faecal coliforms being the most common adopted parameter. However, indicator bacteria were rarely recorded in any of the reviewed studies. This can be explained by the presence of the membrane in an AnMBR which removes a significant fraction of bacteria. Interestingly, GS-F7 reported a higher MS-2 phage removal rate (3 LRV) compared with aerobic MBRs (1.2 LRV) using similar membranes, where MS-2 phage is often used to model the behaviour of pathogenic human viruses. This may be due to the action of certain anaerobic solutes which could be further studied (Fox & Stuckey, 2015).

Moreover, GS-F1 and GS-H12, monitored indicator bacteria with the effluent faecal coliforms concentration for GS-F1 and GS-H12 were 49 and 42 ± 48 CFU/100 mL, respectively (Martinez-Sosa et al., 2011; Peña et al., 2019). Both effluents could meet the 200 CFU/100 mL limit for many agricultural uses (except the non-detectable limit for the irrigation of raw consumed food crops) and industrial reuse but will need a disinfection process to meet the stricter consents (typically non-detectable) required by applications such as toilet flushing or street maintenance and fire-fighting. The requirement in many standards for a chlorine residual concentration of between 0.5 and 1.0 mg/L means that a further disinfection process is compulsory for many water reuse applications.

The performance of AnMBRs on organic removal is shown in Figure 2-2 together with the BOD₅ limits taken from our benchmark standards. Compared with the BOD₅ limit of non-potable water reuse standards in Table 2-1, there are 32, 31 and 30 of the 37 systems which could respectively meet the BOD₅ limit of 35 mg/L, 30 mg/L and 25 mg/L with no further treatment required. Consents of 20 mg/L and 15 mg/L, where most standards for agricultural irrigation lie, can be met by 24 and 22 systems respectively and 12 systems meet the consent of 10 mg/L, which includes direct human contact reuse applications. Finally, six systems meet the strictest BOD₅ limit of 6 mg/L for non-potable reuse purposes in all standards.

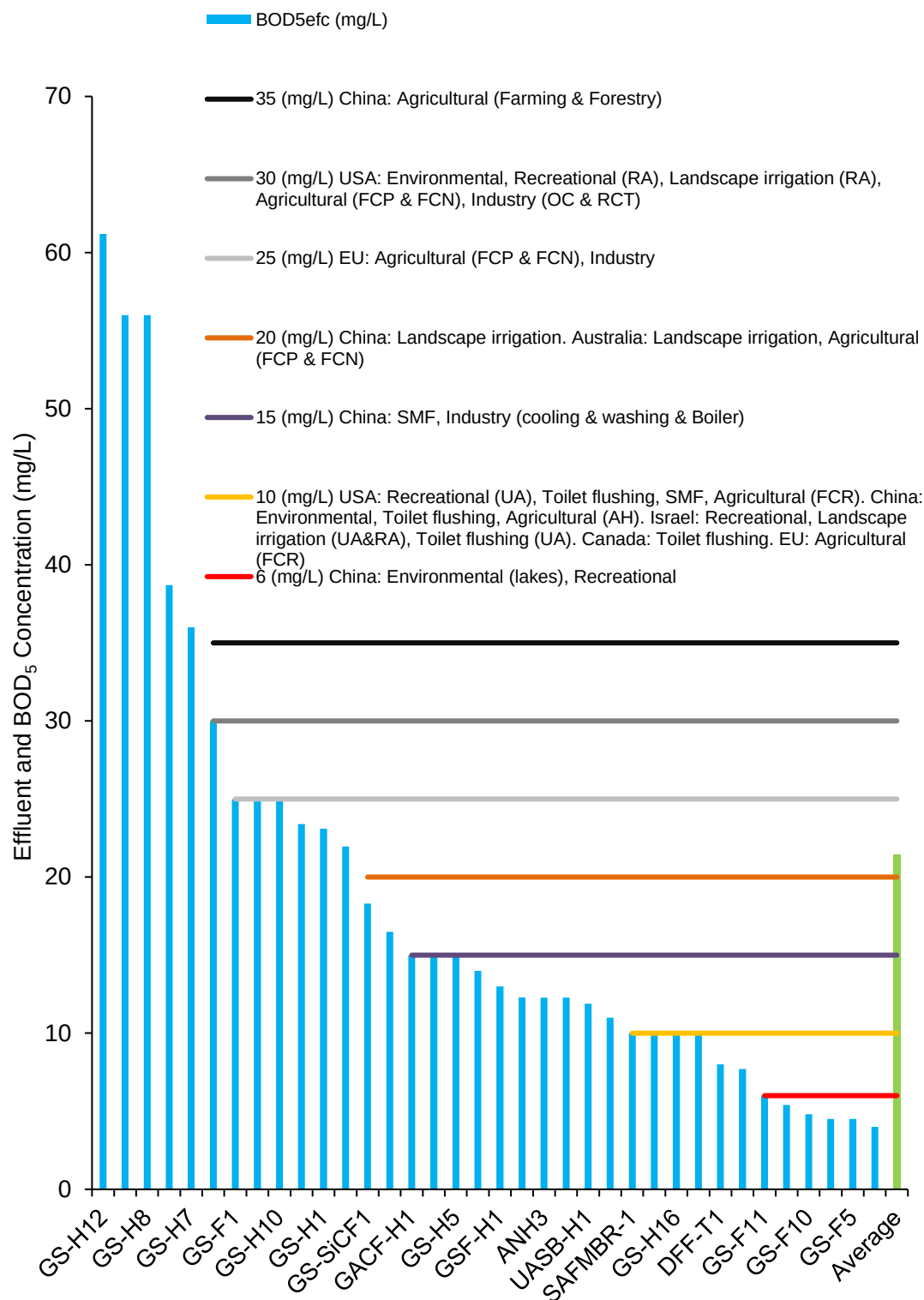


Figure 2-2 Effluent BOD₅ concentration of AnMBR systems against sub-potable water reuse standards

Only the EU and Chinese standards have a COD consent. Agricultural reuse has the most wide-ranging limit for different purposes and 33 systems satisfy the EU's COD limit of 125 mg/L for agricultural irrigation (except where raw consumed food crops are involved), There are 30 and 13 AnMBRs that can respectively satisfy the COD limit set by the Chinese agricultural reuse standards for farming and forestry of 90 mg/L and animal husbandry of 40 mg/L (Supporting information 1-1). Overall these results show that, although there is a wide variability in effluent quality reported in the different studies evaluated, AnMBRs can be operated in such a way that they can ultimately meet the organics limit for any non-potable reuse standard.

2.5 Impact of system configuration and operational variables on performance

The variations in AnMBR performance reported above invite us to explore how other factors influence the operational sweet spot of AnMBRs as well as investigate how their configuration and design might be finessed specifically for water reuse. Such considerations can be extended to assess the role that AnMBRs might play in wider circular economy solutions, supporting recovery of nutrients and energy for example. The following paragraphs explore the influence of pH and salinity, temperature, hydraulic retention time, and reactor configuration on AnMBR performance in the context of meeting water quality criteria for sub-potable applications.

When comparing the effluent pH value of all AnMBR systems with water reuse standards (5.5 to 9), it is obvious that all systems can meet the pH limit. Therefore, unless extreme conditions happen to the influent wastewater (e.g. chemical release), this leaves no concern on balancing the effluents. However, the influent wastewater pH may have a negative impact on AnMBR operation but insignificant influence on effluent quality. To illustrate, higher membrane fouling rate was observed in GS-H11 (Khan et al., 2019), in both acidic and alkaline pH conditions, with the lowest fouling rate was observed at pH 7.0. The effect of pH shock has been studied in GS-F5 (Kunacheva, Soh, & Stuckey, 2017) by changing the feed water pH from 7 to 5 and 11 for 4 hour and 24 hour, respectively. The 4 hour pH

shock did not have a negative effect on the AnMBR performance but there is a significant effect when pH shock is 24 hour. For the pH 5 shock, the system took 24 hours to recover due to high concentrations of carbohydrates found during this period. For the pH 11 shock, the system only took 8 hours to recover as large molecular weight (MW) (1500 kDa-0.2 μ m) protein-like compounds were found. As larger colloids related to protein-like compounds caused more fouling on the membrane, this biofouling layer is an important factor of removing neutral and negatively charged low MW compounds, which is effectively increasing the COD removal in submerged AnMBR. Additionally, the pH shock reduced the ability of removing these compounds therefore having a negative impact on the AnMBR's performance. However, under the pH shock GS-F5 could still stably produce an effluent with 15 mg/L COD which satisfies the organic consent in all non-potable reuse standards. The salinity during operation is also a factor which need to be noticed, high salinity results in lower biological performance and poor COD removal, additionally significant increase of soluble microbial products (SMPs) was observed with the increase of salinity in GS-F11 but the effluent COD concentration stayed under 20 mg/L which also satisfies all organic consent for non-potable reuse (Chen et al., 2019). Consequently, municipal wastewater normally has no extreme pH or salinity conditions, due to the generally large range of pH accepted in reuse standards, there is no concern for AnMBRs to meet the reuse standard in the effluent but significant variations of the pH within that range may affect operation (fouling) and possibly treatment performance (organics), hence a close monitoring of the pH is essential.

AnMBR are more commonly operated under either thermophilic (50–60°C) or mesophilic (30–40°C) conditions (Galib et al., 2016). However, psychrophilic condition (< 20°C) has also been shown to achieve good treatment of municipal wastewater (Robles et al., 2013). The benefit of operating at lower temperature is that there is no need to heat the reactor, especially in temperate climate, and consequently reduce the overall energy consumption of the system and can potentially allow energy neutral operation. However, a number of studies reported that operating AnMBR at low temperature can result in several limitations, including slow contaminant biodegradation, aggravated membrane fouling, low

CH₄ production but high CH₄ solubility in the effluent (Dolejš et al., 2017; Martin Garcia et al., 2013; Smith et al., 2012; Yoo et al., 2014). With the first priority of satisfying water reuse standard, slow contaminant biodegradation directly influencing the effluent quality, leads to longer HRT or additional treatment required. Furthermore, higher membrane fouling decreases the permeate production efficiency and lead to frequent membrane backwash/cleaning which ultimately increases the operational expenditure. Another concern of operating AnMBR under low temperature is a lower energy recovery potential compared to those operating under higher temperature. Hydrolysis of particulate matter into dissolved molecules is limited at low temperature, leading to the accumulation of suspended solids in the reactor and a decrease in methanogenic activity (Chen et al., 2016). The decreased CH₄ production could also be attributed to its increased solubility in the effluent when the temperature decreased to 20°C or below. In addition, the mixed liquor viscosity also increased as the temperature decreased, thus requiring more energy for mixing and pumping which resulted in a slightly lower organic and nutrients removal performance and had a faster membrane fouling rate (Ding et al., 2019). Low temperature tolerance of AnMBR has also been reported with lab-scale (Smith et al., 2015; Yoo et al., 2014) and pilot-scale AnMBRs. The seasonal changes in temperature will result in lower hydrolysis-rates and higher methane dissolution rates in the effluent but the COD removal remained similar during both summer and winter (Giménez et al., 2014). For GS-F1, an increase in the total suspended solids content and soluble COD in the bioreactor was observed when the temperature of the AnMBR was reduced from 35°C to 20°C (Martinez-Sosa et al., 2011). However, the organic removal stayed constant with an effluent COD and BOD₅ no higher than 80 mg/L and 25 mg/L, respectively. Therefore, the permeate water quality could potentially satisfy the 25 mg/L BOD₅ reuse consent under a stable operating condition. Furthermore, the effect of psychrophilic temperature shocks was studied on a gas-lift AnMBR (GL-T1), as the temperature was decreased from 35°C to 15°C. The effluent BOD₅ concentration remains at low level (16.5 mg/L) as the influent COD accumulated in the reactor. Under psychrophilic conditions, more than 80% of the influent COD accumulated in the reactor which is 41% higher than in

mesophilic conditions which led to a lower biogas. Additionally, the application of membrane has a positive effect on the rejection of the COD inside the reactor, only a slightly higher filtration resistance increase was observed due the decreased viscosity of the permeate (Dolejš et al., 2017). Considering the effluent water quality as the priority, it could already produce a permeate satisfying the 20 mg/L BOD₅ reuse consent.

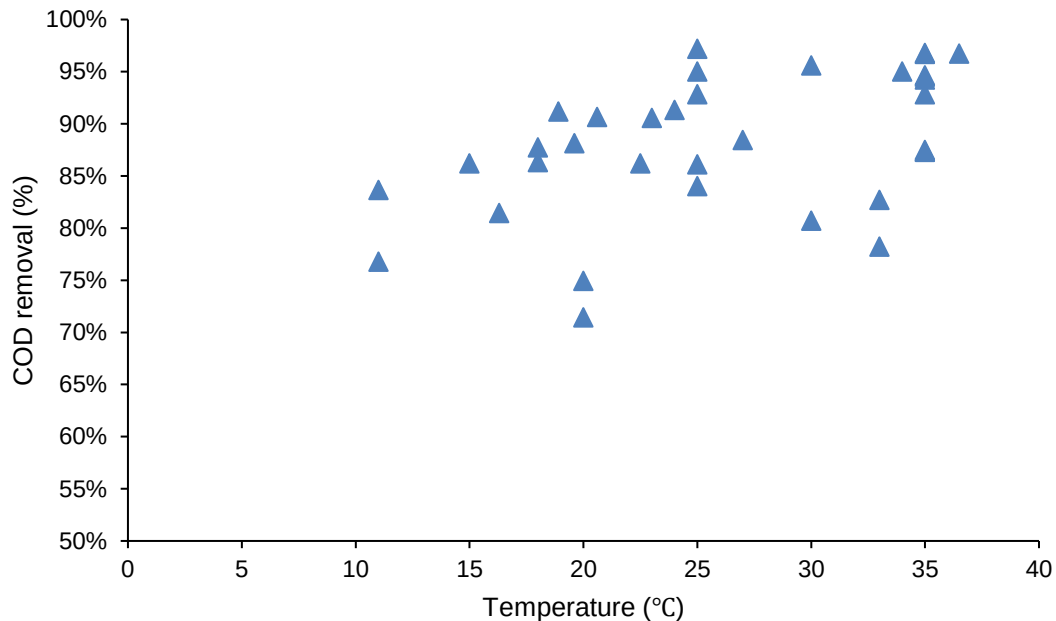


Figure 2-3 COD removal under different Temperature

The AnMBRs operating around 35°C generally had a better organic removal performance than those running at lower temperature especially systems under 25°C (Figure 2-3). Effluent COD concentration of 15 mg/L and 14 mg/L, satisfying all sub-potable reuse standard, was achieved for temperature of 25°C and 35°C for GS-F5 and FBR-CT1, respectively. Additionally, GS-F5 and FBR-CT1 reached COD removal rates of 96.8% and 97.2%, respectively. It should however be noted that GS-F5 was treating a synthetic wastewater and FBR-CT1 was treating synthetic primary effluent which are often shown to be more easily degraded than real wastewaters due to the fact that they have no daily/seasonal variations. Furthermore, the 6 AnMBRs with the lowest BOD₅ effluent concentration were treating synthetic wastewater. Reviewed literature shows a general trend of increasing organic removal performance with concentrated

synthetic wastewater, raw wastewater, mixed raw wastewater, settled wastewater, synthetic wastewater and synthetic primary effluent. This indicates that the difference in influent wastewater type may have an impact on AnMBRs' performance, which could be explained by the more complex matrix of compounds in real wastes and possibly the more biodegradable organics available in synthetic wastewater as demonstrated by the higher BOD₅/COD ratio of synthetic wastewater (0.5-0.7) compared to real wastewater (0.3-0.5). Systems operated under 25°C show a big variation in the organic removal performance. Representatively, the systems operated at 11°C (GSF-H1 and GSG-H1) achieved a lower COD removal rate of 83.7% and 76.8% when compared to others operated under higher temperature. However, GSF-H1 and GSG-H1 could produce an effluent with COD concentrations of 34 mg/L and 39 mg/L, respectively, which equivalently (assuming a BOD₅/COD ratio of 0.3) satisfies the 15 mg/L BOD₅ standard consents including agriculture and landscape irrigation.

These results indicate that good organic removal could be achieved under a psychrophilic temperature range and operating under a higher temperature should be considered to meet a very strict reuse standard consent such as for recreational purposes (BOD₅ 10 mg/L). In terms of operation, the authors of the GS-H15 study argue that membrane permeability was higher under psychrophilic than mesophilic conditions and that lower extracellular polymeric substances (EPSs) and soluble microbial products (SMPs) levels were observed at psychrophilic conditions due to lower biomass activity, which affected not only the three-dimensional floc matrix, but also the fouling propensity (Wang et al., 2018). Similarly, GS-H3 had reported that higher membrane fouling propensities were observed under mesophilic than under psychrophilic conditions due to higher SMP production (Robles et al., 2013). With the aim of reducing energy consumption and as heating the reactor is reported as not viable if targeting energy neutral operation (Mathioudakis et al., 2012; McAdam et al., 2011), operation at lower temperature is preferred. This suggests that as highlighted before for long-term operation, there will be a trade-off between energy use (heating of reactor, membrane operation) and effluent quality but also by managing key parameters of solid concentration and biogas sparging, AnMBR

could be stably operated in psychrophilic temperature for treating municipal wastewater for sub-potable reuse.

Another key parameter for AnMBR operation is the hydraulic retention time. A huge variation of organic removal rate is observed for AnMBRs reviewed in this contribution (Figure 2-4). Most studies reviewed operated in an HRT range of 4 h to 12 h, while 8 h was more commonly applied. Effluent COD (COD removal rate) of 16 mg/L (96.7%), 15 mg/L (96.7%), 15 mg/L (96.8%) and 14 mg/L (97.2%) were achieved respectively with HRTs of 4 h (GS-F10), 12 h (GS-F3), 6 h (GS-F5) and 8 h (FBR-CT1), while other studies also reported COD removals as low as 70-75% for similar HRTs. However, higher HRT delivers more predictable performance, all systems operating with the HRT longer than 15 h had achieved a removal rate higher than 84%. These results show that it is possible to achieve high effluent quality with short HRTs but also highlights that other factors are influencing the treatment performance.

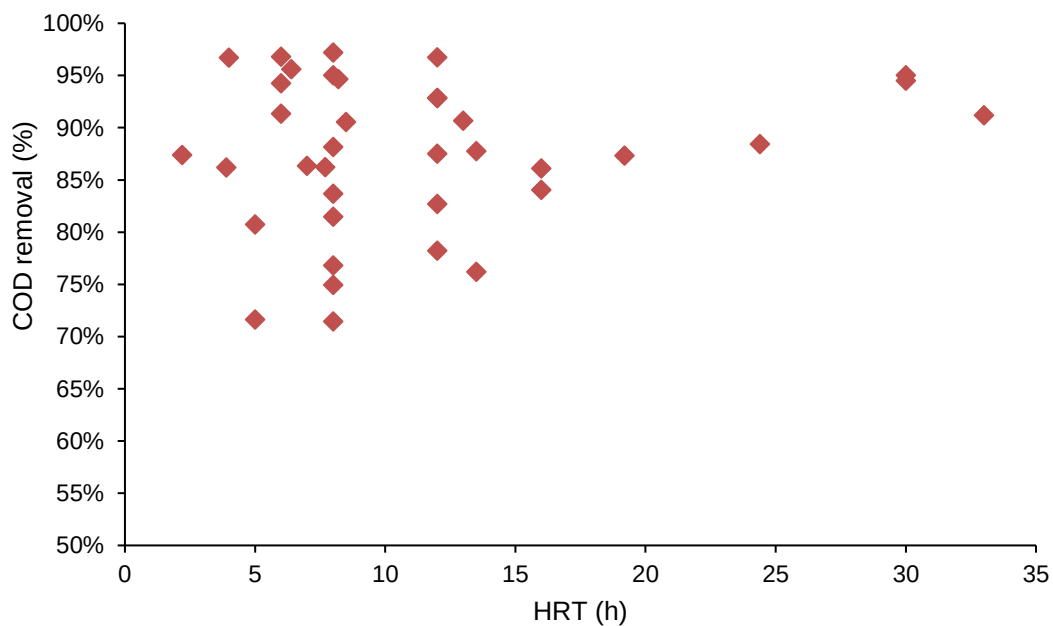


Figure 2-4 COD removal under different HRT

GS-H2 operated with short hydraulic retention time (2.2 h) and a flux of 6 LMH for 340 days without any membrane cleaning but no organic removal performance is recorded in this study (Mei et al., 2017). The lowest COD removal

rate (71.4%) was reported in NGS-T1 operated under an HRT of 8 h, this is mainly due the lack of gas sparging resulted in higher membrane fouling rate also decreased the mixing in the upper part of the UASB reactor therefore reduced the organic removal performance (Cerón-Vivas & Noyola, 2017). In another study, GS-F6 was treating a concentrated synthetic wastewater which had an influent COD concentration of 1410 mg/L, under a 5 h HRT and it could not achieve a complete organic biodegradation (Trzcinski & Stuckey, 2016). In long-term treatment of municipal wastewater, AnMBRs were capable of operating at low HRTs with a tolerance of hydraulic shock loads but a reduced organic removal performance. GS-F4 reported an effluent COD concentration of 16 mg/L was achieved when operating the AnMBR under a 4 h HRT. However, VFAs began to accumulate when the HRT was reduced to 2 h or further down to 1 h, which accounted for 69% and 89% of the effluent COD, respectively (Kunacheva, Soh, & Stuckey, 2017). The study with ANH3 and ANH4 also argued that under a stable HRT, an AnMBR operating in psychrophilic temperature had a slightly lower organic removal performance and had a faster membrane fouling rate (Ding et al., 2019) which had a negative impact on effluent quality. This led to the effluent quality failing to satisfy most sub-potable reuse standards.

Overall these results show that it is possible to operate a system with a very short HRT (5-10 h), hence reducing the footprint of the system, and still achieve very good removal (>90%). Ultimately, it is difficult to conclude on the impact of one specific parameter because the overall performance of these AnMBRs would have been affected by many parameters and all these systems are different. For instance, GS-H12 has both the highest effluent COD and BOD_{5efc} concentrations of 150 and 61.2 mg/L, respectively, but this can be attributed to the fact that the influent wastewater had the highest COD concentration of 1729 mg/L. However, ANH2 with a lower effluent COD concentration (86 mg/L) had similar influent COD concentration of 1720 mg/L. The later was operated under 34°C and 30 h HRT while GS-H12 operated under 24°C and 6 h for temperature and HRT. This confirms that the operating temperature and HRT are expected to have a positive influence on treatment performance and can be adapted to tailor the organic

removal to the effluent quality requirement associated with specific standards for reuse.

Different configuration of AnMBR may enhance the produced water quality with advanced membrane technology. GSDM-F1 configured with a 75 μm pore size nylon mesh dynamic membrane module was reported to be successfully operated under high flux (22.5 L/(m²h)) and 8 h HRT at psychrophilic temperature. When treating municipal wastewater, this system could produce an effluent satisfying the 25 mg/L BOD₅ reuse standard consents (Hu et al., 2018). Compared to the configuration of UASB + hollow fibre/flat sheet membrane reported in most literatures, the best performance of AnMBR reviewed in this paper had a different configuration both in reactor, membrane type and material. The fluidized-bed reactors (FBR) and downflow floating media filter (DFF) AnMBR systems could produce high quality effluents with COD concentration of 14 mg/L and 25 mg/L and BOD₅ concentration of 5 mg/L and 8 mg/L for FBR-ceramic tube membrane and DFF-PVDF tube membrane, respectively. The fixed-film reactors had enhanced the biodegradation of organic compounds also the 0.018 μm pore-sized ceramic tube membrane increased the rejection performance. However, with this configuration it consumed 65%-75% more energy than the conventional AnMBRs with an energy neutral operation (Seib et al., 2016). These results also highlight the trade-off between energy use and operation and treatment performances, especially when strict standards are to be met.

Dense high-pressure membrane processes like NF and RO are excellent technologies for advanced treatment of MBR permeates, as they both deliver high quality water (Mai et al., 2018a). Recent progress to advance AnMBR systems has resulted in the emergence of high retention AnMBR systems. The combination of membranes such as FO, RO, NF with anaerobic reactors is aimed at producing water that can be reused. These include anaerobic membrane distillation bioreactors (AnMDBR) and anaerobic osmotic membrane bioreactor (AnOMBR) (Shin & Bae, 2018). Both AnMDBR and AnOMBR can enhance the removal of contaminants to produce a sufficient effluent quality for water reuse

applications. AnMDBR is an integration of membrane distillation (MD) and anaerobic treatment. MD is a thermally driven separation process, in which the thermal gradient between a feed solution and distillate drives the transportation of water vapour through a hydrophobic, microporous membrane. The competitive advantages of anaerobic processes can be readily utilized when they are combined with the MD process, because the thermophilic operation for anaerobic treatment can reduce extra heat requirement for MD operation (Kim et al., 2015). GS-C1 integrated a MD process with AnMBR, high removal of organic matter and nutrients was achieved with an effluent COD concentration of 101.5 mg/L, which satisfies the COD consent in EU's agriculture reuse standard. The high retention capacity of the MD membrane resulted in significant phosphate accumulation in the feed solution, thereby producing an opportunity for phosphorus recovery from the AnMBR effluent (Song et al., 2018). Similar result was reported by Jacob et al. (2015) with 90% removal of COD and ammonium nitrogen from AnMBR effluent by a direct contact MD process. Further research is necessary to address the issue of MD membrane fouling when coupled with the anaerobic reactor for phosphorus recovery and water reuse applications. It is noteworthy that a reduction of NH_4^+ removal was observed in their study due to its transportation through the MD membrane via ammonia evaporation. This issue can be potentially addressed using a FO and MD hybrid system, where the FO membrane can effectively reject NH_4^+ while MD can be used to regenerate the draw solution and produce clean water for reuse.

FO is the transport of water across a semi-permeable membrane similar to natural osmosis, coupling FO membranes with an anaerobic bioreactor provided a higher quality effluent compared to a conventional AnMBR, especially in terms of nitrogen and phosphorus removal (Chen et al., 2016; Ding et al., 2014). FO was also reported to reject particles, pathogens and emerging contaminants in wastewater (Lutchmiah et al., 2014). Additionally, FO could be both considered as a pre-treatment for AnMBR as FO-AnMBR or replacement for the membrane unit as AnOMBR. During AnOMBR operation, a desalination process, such as nanofiltration (NF) or reverse osmosis (RO), can be used to regenerate the draw solution and produce clean water. Compared to conventional MF and UF

membranes, FO has higher selectivity, lower membrane fouling propensity and better membrane fouling reversibility (Xie et al., 2015). However, FO has a much lower flux and the permeate requires further separation to produce reusable water, arguing FO is not effective at producing potable grade water compared to RO (Ansari et al., 2017). Indeed, the FO process requires a draw solution to drive the osmosis, this also increases the process complexity and cost (Ding et al., 2014). CSTR-H1 was configured with a forward osmosis and heating pump to evaluate the feasibility of energy neutral system treating synthetic municipal wastewater (Wei et al., 2014). The FO unit was set up as a pre-treatment unit together with the heating pump to achieve a mesophilic temperature of AnMBR. The effluent water quality is sufficient for 15 mg/L BOD₅ consents in reuse standard. This suggests the combination of FO+AnMBR could be a promising direction for achieving water reuse via AnMBR. ANH2 studied the feasibility of this configuration and reported with similar result. The FO unit concentrated the wastewater for AnMBR, at 15°C a lower OLR should be applied to maintain stable operation compare to higher temperature. Under the optimum conditions, the effluent water from FO could meet the 25 mg/L BOD₅ reuse consents (Ferrari et al., 2019). However, the high salinity wastewater from the FO unit could have a negative impact on methane production which reduce the energy recovery of AnMBR (Tang & Ng, 2014).

Amongst complex contaminants in wastewater, TrOCs present arguably the most vexing challenge to water reuse (Schwarzenbach et al., 2006). TrOCs are trace organics compounds, also called organic micro-pollutants, including pharmaceuticals, endocrine disruptors, pesticides and personal care products. Recent studies have also demonstrated that the removal of TrOCs by AnMBR varied significantly from negligible to more than 90%. TrOC removal by AnMBR is governed mostly by intrinsic physiochemical properties of the compound (Monsalvo et al., 2014). The removal of 38 TrOCs by AnMBR was studied and reported over 90% removal of nine compounds, while the others were removed by less than 50% (Monsalvo et al., 2014). A predictive framework was successfully developed to assess the removal of TrOCs by AnMBR, which relates to their hydrophobicity and molecular structures (Wijekoon et al., 2015). GS-C1

had reported a complete removal of the TrOCs by integrating AnMBR with membrane distillation. The synergy between the biological treatment and the MD membrane rejection contributed to 76% to complete removal of all 26 selected TrOCs (Song et al., 2018). GS-F9 studied the removal of pharmaceuticals in AnMBR with the addition of powdered activated carbon. The sludge adsorption contributed significantly within 10 days when pharmaceuticals first entering the systems. After that the removal efficiency is depended on the biodegradability (Xiao et al., 2017). With one-time PAC addition, within the first 5 days all five pharmaceuticals removal were also improved by enhancing the biotransformation, but the removal of refractory compounds was still low which requires further treatment to satisfy the stricter reuse consent. SAFMBR-1 examined the removal of pharmaceuticals in municipal wastewater with a staged Anaerobic fluidized membrane bioreactor. By being positively charged at neutral pH and/or hydrophobic, the pharmaceuticals were initially adsorbed onto the sludge, followed by being desorbed when sludge decayed after long operating hour (Chen et al., 2019). Processes responsible for the removals of pharmaceuticals were more complicated as the HRT adjustment had a limited effect rather than apply a post-treatment process. Consequently, TrOCs removal by membrane process is largely governed by their molecular properties, regardless of the presence or absence of oxygen in the bioreactor (Tadkaew et al., 2011; Wijekoon et al., 2015). Therefore, for environmental and recreational reuse or potentially potable water reuse, post-treatment is compulsory to produce an effluent of sufficient quality.

2.6 Energy recovery

It is well established that biogas produced by AnMBR consists of more than 70% CH₄ (Skouteris et al., 2012). During AnMBR treatment, the CH₄ yield increases linearly with the organic loading rate (Yeo et al., 2015). Under optimized condition, AnMBRs can convert up to 98% of the influent COD into biogas, which is equivalent to seven times the energy required for system operation (Song et al., 2018). In practice, actual biogas yields are considerably lower than the theoretical value, due to the high solubility of CH₄ in the effluent and process

inhibition caused by inhibitory substances. CH₄ loss due to its solubility (22.7 mg/L, 20 °C) in the effluent is significant during AnMBR treatment, particularly for low strength municipal wastewater (Smith et al., 2012). Dissolved CH₄ in permeate was approximately 45% of total produced CH₄ at 30°C when an AnMBR was used for treating municipal wastewater with COD of 200 mg/L (Liu et al., 2013). Similar studies have reported 24 – 58% of total produced CH₄ dissolved in the permeate during AnMBR treatment and have demonstrated that AnMBRs can remove 86–88% COD from municipal wastewater (influent COD of approximately 330 mg/L), although 67% of the produced CH₄ is dissolved in the mixed liquor and then released via the permeate (Yeo et al., 2015). Dissolved CH₄ in the permeate does not only reduce the energy efficiency of AnMBR treatment, but also contributes to global warming as the greenhouse potency of CH₄ is 25 times higher than carbon dioxide (Jacob et al., 2015).

Major factors reported to affect biogas production are organic loading rate (OLR) and influent sulfate concentration. No correlation was found here between OLR and organic removal but a link to biogas production performance has been reported. Dissolved CH₄ concentrations were seen to decrease from 54 to 25 mg/L when the organic loading rate of wastewater increased from 0.4 to 3.2 kg COD/m³d, due to the enhanced biogas yield at the higher organic loading rate (Galib et al., 2016). Similarly, improved biogas production was reported for PFPS-H1 with increasing organic loading rate (Bair et al., 2015). The economic and environmental sustainability of increasing OLR of AnMBR treating municipal wastewater with organic fraction of municipal solid waste (OFMSW) was evaluated by ANH1 treating low-sulphate influent (Pretel et al., 2016), the result agreed AnMBR could achieve a net energy producer with GS-F6 reported by Trzcinski & Stuckey (2016). The overall environmental performance was enhanced by a higher methane yield but significant membrane fouling was observed at the same time. In addition to OLR, the methane conversion is also dependent on the presence of sulphide. Low conversion of COD to methane for UASB-H1 was attributed to the presence of SO₄, due to scavenging of up to 50% of COD in the influent and accumulation of un-hydrolysed lipids in the mixed liquor (Petroopoulos et al., 2019). For GS-H1, a higher COD/SO₄²⁻-S ratio was reported

to result in a rapid increase of methane yield. The COD/SO₄-S ratio strongly affects the methane yield by methanogenic archaea (MA) and sulfate reducing bacteria (SRB) competition, as COD is removed by SRB (Giménez et al., 2011). Furthermore, GS-H13 had confirmed when treating sulfate-rich municipal wastewater the SRB could reduce all the influent sulfate to sulphide and the organic matter available for methanogens which also reduce the methane yield. The increase of sulphide level will lead to a decrease of Fe²⁺ bioavailability, the soluble COD increase in the reactor along with the increasing of VFAs level but a decrease of methane yields (Seco et al., 2018). By dosing EDDS in a 1:1M ratio with Fe²⁺, GS-F4 gave an insight of reducing soluble COD and VFAs resulting in the methane yields increased by 9.46% under the same sulphide level (Thanh et al., 2017). Therefore, it is valuable to both maximise the conversion of CH₄ from the influent and subsequently recover from the effluent, whereas increasing operational organic loading rate and maintaining a low influent sulfate concentration would also be beneficial for a better energy potential which subsequently reduce the overall operational cost for reuse application.

2.7 Post-treatment

Although AnMBRs have been shown to produce high quality effluents, those standards requiring tighter consents for organics, nutrients and pathogens cannot always be met and consequently post-treatment will be needed. The produced biogas during anaerobic process, along with the dissolved CH₄, will also need additional treatment to produce safe effluent for reuse and potentially energy recovery. Despite the biogas, removal and recovery of nutrients from AnMBR effluent constitutes another step towards enhanced resource recovery via water reuse schemes. As stated above, AnMBRs do not remove nutrients, anaerobic processes turns nitrogen and phosphorus to the form of ammonium (NH₄⁺) and phosphate (PO₄³⁻), respectively (Song et al., 2018). Recent reviews by Mai et al. (2018) and Rongwong et al. (2019) have described post-treatment technologies for use prior to discharging or reusing anaerobic effluent. Table 2-3 presents potential post-treatment options that are well suited for use post-AnMBR in order to meet sub-potable reuse standards.

Table 2-3 Post-treatment options for AnMBR based sub-potable reuse applications

Technology	Targeted components with removal rate (if available)	Advantages	Disadvantages	Sources
GAC	80% COD; Phenols; VOC; PhCs; Metals; EDs; PCPs	Wide targeting range, easy to apply, low cost, effectively eliminate PhCs, EDs and PCPs	Adsorption of low MW compounds is limited	(Mai et al., 2018b; Nguyen et al., 2012; Snyder et al., 2007; Trzcinski et al., 2011; Vyrides, Conteras, et al., 2010)
PAC	84% COD; Phenols; 80% DOC; VOC; PhCs; Metals; EDs; PCPs	Wide targeting range, effectively removing traces of organic carbon, effectively eliminate PhCs, EDs and PCPs		
Carbon Nanotubes	COD; Phenols; VOC; PhCs; Metals; AOCs; NOM; EDs; PCPs	Wide targeting range, no diffusion required compared to AC, effectively eliminate PhCs, EDs and PCPs	High manufacturing cost	(Amin et al., 2014)
MD	98% COD; 10% NH_4^+ ; >99% PO_4^{3-} ; 65% dissolved methane	Complement very well with AnMBR for energy and water recovery	Flux strongly depends on the temperature and pressure, require further treatment for TN	(Jacob et al., 2015; Kim et al., 2015; Song et al., 2018; Xie et al., 2016)
NF	COD; NH_4^+ ; Up to 95% PO_4^{3-} ; Multivalent ions; 80-92% OMPs;	Produce high quality effluent for reuse	Lower energy demand than RO, effluent contains monovalent ions	(Wei et al., 2015)
RO	88% COD; >99%TOC; 91% TKN; 99% PO_4^{3-} ; Multivalent ions; Monovalent ions; >90% OMPs	Produce the best quality effluent with nearly all removal of organics and nutrient	Very high energy demand, limited removal on hydrophilic organics with low MWs	(Grundestam & Hellström, 2007; Gu et al., 2019; Liu et al., 2020)
FO	61-88% COD; 98-99% NH_4^+ ; 89-94% PO_4^{3-} ; Particles, Pathogens;	Low energy consumption	Draw solution require	(Chen et al., 2014; Ding et al., 2014; Lutchmiah et al., 2014)

	Emerging contaminants;			
Microalgae	67% NH_4^+ ; 98% PO_4^{3-} ; COD	Suitable for high quality effluent from AnMBR, Generated biomass could be used to generate renewable biofuel	Need optimum operation condition	(Ruiz-Martinez et al., 2012)
IEX (Zeolites)	83-94% NH_4^+	Selectively remove and recover of nutrients, Short contact time and small footprint	Regeneration dominated the operational cost and energy consumption	(Deng et al., 2014)
IEX (HAIX)	97% PO_4^{3-} (PhosXnp)			(Sendrowski & Boyer, 2013)
Coagulation-Flocculation (FeCl_3)	96% PO_4^{3-}	Widely applied to Aerobic system for enchanted chemical phosphorus removal	Produce excess wastes	(Penetra et al., 1999)
Membrane Contactor	98.9% Dissolved Methane; 90% NH_4^+	High methane and ammonium recovery	Membrane fouling and wetting, Economic viability and process safety have not been fully evaluated, require further treatment for TP	(Cookney et al., 2016; Rongwong et al., 2019)
Membrane biofilm reactor	96% NH_4^+ ; 98% dissolved methane	Could both remove dissolved methane and TN	Require both energy and membrane cost, also further treatment for TP	(Chen et al., 2015)
Notes: GAC: Granular activated carbon; PAC: Powdered activated carbon; VOC: Volatile organic compounds; PhCs: Pharmaceutically compounds; DOC: Dissolved organic carbon; AOCs: Aromatic organic compounds; NOMs: Natural organic matter; AC: Activated carbon; MW: Molecular weight; EDs: Endocrine disruptors; PCPs: Personal care products; MD: Membrane distillation; NF: Nano-filtration membrane; FO: Forward osmosis membrane; RO: Reverse osmosis membrane; TN: Total nitrogen; TP: Total phosphorus; TKN: Total Kjeldahl nitrogen; OMPs: Organic micropollutants; AOP: Advanced oxidation Process; IEX: Ion exchange; HRT: Hydraulic retention time.				

Post-treatment technologies listed in Table 2-3 target a wide range of contaminants and provide options to improve AnMBR effluent quality in order to satisfy different reuse applications. Carbon based post-treatments could help AnMBRs meet organic consents for sub-potable reuse standards up to the

strictest levels (6 mg/L BOD₅). This provides a promising level of treatment for agricultural or landscape irrigation purposes where a lower organic limit (20 mg/L BOD₅) but no nutrients removal is required. Additionally, carbon based filtration has a wide target contaminant range, also effectively removing traces of organic carbon (eliminating PhCs, EDs and PCPs). Good removal of ciprofloxacin (CIP) and VFAs was achieved by treating AnMBR effluent with Granular Activated Carbon (GAC) and Powdered Activated Carbon (PAC) (Mai et al., 2018b). However, poor removal rates for low molecular weight compounds has been reported for GAC and PAC (Trzcinski et al., 2011; Vyrides, Conteras, et al., 2010), and high manufacturing cost for carbon nanotubes (Amin et al., 2014) are a significant barrier to carbon based post-treatment technologies. Additional membrane processes can deliver high quality effluent with a certain amount of organic, nutrient, and pathogen removal with the produced effluent being suitable for a broad range of reuse purposes such as environmental enhancement and recreational waters. NF can be used to produce a low organic, ammonia and phosphorus concentration effluent (Wei et al., 2015), which satisfies all sub-potable reuse standard consents. Reverse Osmosis can reach even higher standards (4 mg/L BOD₅, 0.2 mg/L NH₃-N) and also removes trace organics (Mamo et al., 2018). However, the energy consumption of RO processes (Gu et al., 2019), means that it may only be suitable for potable reuse applications. Another advantage of membrane based post-treatment (NF, RO, FO and MD) is concentrating N and P compounds in the effluents while producing clean water as a permeate (Rongwong et al., 2019).

In addition to membrane filtration processes, effective removal of nutrients and dissolved methane was also reported with membrane biofilm reactors (90% NH₄⁺; 98.9% Dissolved Methane) (Chen et al., 2015) and membrane contactors (96% NH₄⁺; 98% dissolved methane) (Rongwong et al., 2019), where dissolved methane is not removed by pore rejection (Mai et al., 2018a). Other options available to remove dissolved methane and nutrients include membrane distillation (10% NH₄⁺; >99% PO₄³⁻; 65% dissolved methane) (Song et al., 2018), and electrodialysis (ED) (Xie et al., 2016). The recovered methane could significantly lower the operating energy consumption (Crone et al., 2016)

especially when the higher quality is needed for reuse application. Interestingly, where complete nutrient removal is required for environmental and recreational reuse, membrane biofilm reactors and membrane contactors require further treatment of phosphorus, where membrane distillation requires a further treatment step to remove nitrogen. This therefore suggests the combination between different membrane processes may provide a suitable option for on removal and potentially recover the targeted compounds based on the reuse purpose. As Xie et al. (2016) reported, the integration of FO, MD and ED can potentially achieve complete nutrient recovery. However, none of these technologies have been tested as a post-treatment option for AnMBR effluent and require further study to understand how their energy use can be lowered. Combining and optimizing suitable process steps is again a trade-off between the targeted recovery rate and cost intensity combined with the different effluent quality requirements of reuse applications.

Other post-treatment options that may be beneficial include microalgae, ion exchange, and coagulation-flocculation with FeCl_3 . Microalgae is a promising technology to treat AnMBR effluent to remove COD, NH_4^+ (up to 67%) and PO_4^{3-} (up to 98%) but also produce a biomass for bio-fuel generation (Ruiz-Martinez et al., 2012). IEX processes are reported to have the advantage of lower costs, greenhouse gas reduction and nutrient recovery (Huang et al., 2020; Liu et al., 2016). Coagulation-flocculation with FeCl_3 could be applied as a single stage post-treatment process to remove up to 96% PO_4^{3-} (Penetra et al., 1999) also directly dosing into an Anaerobic reactor to enhance the MBR performance (Dong et al., 2015) and the efficiency of the recovery cleaning protocol (Shin & Bae, 2018).

The ultimate target of combining post-treatment processes should not only be to satisfy reuse standards, but also to create a multi-functional resource recovery treatment train. However, it is not only the post-treatment for effluent quality requirements that needs to be considered, but also the impact which the AnMBR effluent may have on a post-treatment process when specifically targeting a reuse application. For example, there is only one published study that has documented

the use of advanced oxidation processes (AOPs) as a post-treatment step for AnMBR effluent, in which UV/H₂O₂ acted as an efficient and safe strategy to remove both biological and chemical contaminants from an ammonia rich AnMBR effluent (Augsburger et al., 2021). We would note that nutrients and other dissolved low molecular weight components may have an impact on post-treatment processes. For example, the presences of N and P has been associated with biofilm formation in reverse osmosis processes (Liu et al., 2020).

2.8 Summary

Literature documented so far has not reported the application of full scale AnMBRs for municipal wastewater treatment, let alone for water reuse. We aimed to discuss the evidences across different dimension (e.g. speciality, feasibility and capability) that how suitable AnMBR would fit into water reuse applications and how it may improve from conventional process, with a focus on the treatment performance, operational parameters, system configuration, energy recovery, and post-treatment processes, which are summarized below.

Reports from the literature suggest that AnMBR could directly produce an effluent satisfying the strictest consents for all sub-potable reuse application except for nutrients and residual chlorine concentration. However, the wide variability observed between literatures indicated the necessity of finding the optimum operational conditions based on the quality requirements for different reuse purpose. Analysis of documented operating data suggests temperature and hydraulic retention time are the two key operational parameters affecting the organic removal performance, where membrane material, configuration and fouling mitigation are mainly contributing to lower the cost of AnMBR rather than contaminant removal performance. Therefore, finding the right balance between the performance and operational expenditure is the critical point of combining AnMBR with water reuse.

Considering the trade-off between energy use (heating of reactor, membrane operation) and effluent quality, AnMBR may be more suitable for areas with warm (hot) climate such as south east Asia with a higher ambient temperature. The higher environmental temperature can offset the limitations operating under

psychrophilic temperature (Dolejš et al., 2017; Martin Garcia et al., 2013; Smith et al., 2012; Yoo et al., 2014), resulting in better organic removal also higher methane yield and lower dissolved methane in AnMBR effluent. Another design insight could be fully covered if deploying AnMBR in cold temperature area. An underground municipal wastewater treatment plant can improve the public perception and overcome footprint cost, potentially heat up the anaerobic reactor and mitigate the odour of biogas. However, the fully covered system requires a higher construction expenditure, while it also require additional air ventilation system and more complex safety protocol compare to overground plants (Sun et al., 2019).

Membrane configurations recorded in the reviewed literature are hollow fibre, spiral-wound, flat sheet and tubular membrane, where hollow fibre is the most common membrane configuration. A significant variation in effluent COD concentration was observed against the different membrane pore sizes and configurations, this confirmed that although the membrane has a critical role in separation of the mixed liquor from the treated effluent in MBRs, the organic removal performance is less dependent on the membrane configuration and material. Combination of different membrane processes with anaerobic reactor has synchronised the nutrient removal before (FO-AnMBR) or after (AnMDBR) the AnMBR process, produces a lower nutrients concentration effluent which satisfies environmental and recreational reuse quality standard.

As stated above, a higher OLR could enhance the methane yield consequently a better energy recover potential, the settled wastewater is most likely the best option for AnMBR reuse application. Reasons include no TSS and no inorganic COD flows to AnMBR leads to low pump energy consumption, lower membrane fouling rate and higher biodegradability, which ultimately gives a richness influent and reduce the total operational cost. However, AnMBRs with an organic removal rate higher than 95% were testing synthetic wastewater, where no such removal rate is reported for those treating the actual wastewater. Therefore, select authentic wastewater for future study will give a more accurate understanding on process fundamental specifically for reuse application.

As negligible nutrient is removed from the influent, apart from the impact of system configuration and operational variables, this argues the purpose of agriculture and landscape irrigation can be more preferable and attractive as the food crops or plants can directly extract the nutrients from the effluent of AnMBR. However, for other reuse purposes such as recreational and landscape irrigation would require post-treatment process to additionally remove organics and nutrients down to sufficient level.

Among all post-treatment technologies available, limited literatures are focusing on treating AnMBR effluent specifically for reuse. Most literatures are either not focusing on treating AnMBR effluent or aiming only for discharge. Leaving the gap of evaluating the feasibility of different technology treating AnMBR effluent for reuse purposes. However, several studies have reported to combine AnMBR with MD, NF, RO, FO and RO+IEX are able to provide a permeate satisfying all non-potable water reuse standard consents (Lim et al., 2020). Further from post-treatment process aiming to meet reuse consents, combination of resource recovery with post-treatment process brings promising insight of applying AnMBR to water reuse applications. As literature has argued the sulfate-rich municipal wastewater reduced methane yield which resulting a lower energy recovery potential (Giménez et al., 2011; Petropoulos et al., 2019; Pretel et al., 2016), AnMBR would be more preferably to treat low sulfate wastewater. Therefore, combination of pre- or post- treatment process to remove upfront nutrients of AnMBR then recover would not only results in improved effluent quality but also recovering the nutrients for resource reuse. Thus, the AnMBR permeate could both meet the organic and nutrient consents in water reuse standards, also the additional value of applying AnMBR to municipal wastewater reuse is enhanced. Ultimately, continues effort should be made to search for technologies with low cost but high effluent quality also maintaining a remove and recovery of dissolved methane and nutrients towards a circular water reuse application.

2.9 Future research

The foregoing text has summarised and drawn out important features of the performance of pilot and laboratory scale AnMBR systems as they have been

applied to municipal wastewater treatment. We have also evaluated the potential application of AnMBR systems to sub-potable water reuse schemes by comparing their performance with water quality regulations and standards from different regions of the world. Our analysis suggests that hydraulic retention time and temperature are two key operational parameters which affect organic removal and that membrane material, configuration and fouling mitigation need continuing study to lower the cost of AnMBR operation. The trade-off between cost and effluent quality is central to the successful application of AnMBRs to water reuse. Ultimately, with proper post-treatment, AnMBRs can deliver water that meets sub-potable reuse standards but further study is required to outline a pathway to maturity for effective treatment trains. In particular, the following specific knowledge gaps require urgent attention:

- More comprehensive water quality analysis (e.g. turbidity, faecal coliforms) is needed to examine the feasibility of AnMBR effluent for water reuse, this will boost understanding of not only the impact of reusing the AnMBR effluent but also options for post-treatment technologies to enable the achievement of stricter reuse standard consents.
- Optimising the operational sweet spot is suggested when treating municipal wastewater for reuse purposes. Stable operation of AnMBRs requires deeper understanding of how multiple parameters interact. Finding the balance between effluent quality and energy consumption requires further study and improved energy recovery will require pre-treatment of influent sulfate as well as safe and efficient dissolved methane recovery. The search for ways in which a satisfactory effluent quality can be produced with low operational expenditure will involve investigation of new membrane materials, configurations and fouling mitigation methodologies.
- Continuing efforts should be made to understand the feasibility of deploying underground AnMBR systems where the risks of operating AnMBRs and safely recycling the generated biogas require careful evaluation.

- Selecting authentic wastewater as the influent for pilot scale experimental work will provide more accurate and credible findings on which to base system design. Results obtained using synthetic wastewater are unconvincing to regulators and scheme approvers.
- Exploring different AnMBR configurations and couplings with secondary technologies may result in combinations of pre-treatment and post-treatment steps with better overall performance both on contaminant removal and energy recovery. The impact of nutrients and low molecular weight substrates on post-treatment technology performance will require further investigation.
- Advanced understandings of the effectiveness of disinfection processes when coupled with AnMBRs (particularly any differences between aerobically and anaerobically treated wastewaters) will support treatment train design for applications requiring near-potable quality water.
- The fate of OMPs still need further study to understand the pathway in the whole process specially the characterization from a real wastewater sample. This understanding will help to evaluate the risks of reusing the AnMBR effluent.
- The fate of different virus species through AnMBR based treatment trains (perhaps involving post-treatment or disinfection) remains largely unknown. Additional research is required to provide confidence in the inactivation or removal of different virus species, particularly where tighter reuse cycles are being planned.

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2.11 Supporting information 1

2.11.1 Supporting information 1-1

Table 2-4 Water reuse standards in different countries/regions

Parameters	Non-potable reuse													
	Environmental uses		Recreational uses					Landscape irrigation uses						
	USA	China	USA(RA)	USA(UA)	China	Israel	Japan(UA)	USA(RA)	China	Israel(UA)	Israel(RA)	Japan	Portugal(RA)	Australia
pH	-	6.0-9.0	-	6.0-9.0	6.0-9.0	6.5-8.5	5.8-8.6	6.0-9.0	6.0-9.0	6.5-8.5	6.5-8.5	5.8-8.6	-	-
BOD ₅ (mg O ₂ /L)	≤30	≤10(6)	≤30	≤10	≤6	<10	-	≤30	≤20	≤10	≤10	-	-	≤20
COD (mg/L)	-	≤40(30)	-	-	≤30	-	-	-	≤1000	-	-	-	-	-
TSS (mg/L)	≤30	≤20(10)	≤30	-	-	-	-	≤30	-	-	-	-	-	≤30
Colour (colour or Hazen units)	-	≤30	-	-	≤30		≤10	-	≤30	-	-	≤40	-	-
Turbidity (NTU or mg-kaolin/L)	-	-	-	≤2 NTU	≤5 NTU	≤2 NTU(Me), ≤5 NTU(Ma)	<2 mg-kaolin/L	-	≤10 NTU	≤2 NTU(Me), ≤5 NTU(Ma)	≤5 NTU(Me), ≤10 NTU(Ma)	≤2 mg-kaolin/L	-	-
TN (mg N/L)	-	≤15	-	-	≤15	-	-	-	-	-	-	-	-	-
NH ₃ -N (mg N/L)	-	≤5	-	-	≤5	-	-	-	≤20	-		-	-	-

TP (mg P/L)	-	≤1(0.5)	-	-	≤1(0.5)	-	-	-	-	-	-	-	-	-			
Faecal coliforms (CFU/100 mL or MPN/100 mL)	≤200(7d Me) ≤800(Ma)	≤1000(200)	≤200(7d Me) ≤800(Ma)	ND(7d Me) ≤14(Ma)	≤50(ND)	≤0(Me), ≤14 NTU(Ma),	-	-	≤200	≤0(Me), ≤14 NTU(Ma),	≤0(Me), ≤14 NTU(Ma),	-	-	-			
<i>E. coli</i> (CFU/100 mL or MPN/100 mL)	-	-	-	-	-	-	ND	≤200(7d Me) ≤800(Ma)	-	-	-	-	≤200	≤ 1000 (IND)			
Total coliforms (MPN/100 mL)	-	-	-	-	-	-	-	-	-	-	-	≤1000(Temporary)	-	-			
TDS (mg/L)	-	-	-	-	-	-	-	-	≤1000	-	-	-	-	-			
TOC (mg/L)	-	-	-	-	-	-	-	-	-	-	-	-	-	-			
Chlorine residual (mg/L)	≥1.0(90min)	≥0.5	≥1.0(90 min)	≥1.0(90 min)	≥0.05	≥1.0(30min)	FRC >0.1 or CRFC >0.4	≥1.0(90 min)	-	≥1.0(30min)	≥1.0(30 min)	-	-	-			
Parameters		Non-potable reuse															
		Toilet flushing								Street maintenance and firefighting							
		USA		China		Israel(UA)		Japan		Spain		Canada		USA	China	Japan	Spain
pH		6.0-9.0		6.0-9.0		6.5-8.5		5.8-8.6		-		-		6.0-9.0	6.0-9.0	5.8-8.6	-
BOD ₅ (mg O ₂ /L)		≤10		≤10		≤10		-		-		≤10(Me), ≤20(Ma)		≤10	≤15	-	-
COD (mg/L)		-		-		-		-		-		-		-	-	-	-
TSS (mg/L)		-		≤1500		-		-		≤10		≤10(Me), ≤20(Ma)		-	≤1500	-	≤20

Colour (colour or Hazen units)			≤30		-	-	-	-	-	-	≤30	-	-					
Turbidity (NTU or mg-kaolin/L)		≤2 NTU	≤5 NTU		≤2 NTU(Me), ≤5 NTU(Ma)	≤2 mg-kaolin/L	≤2 NTU	≤2 NTU(Me), ≤5 NTU(Ma)		≤2 NTU	≤10 NTU	≤2 mg-kaolin/L	≤10 NTU					
TN (mg N/L)		-	-		-	-	-	-		-	-	-	-					
NH ₃ -N (mg N/L)		-	≤10		-	-	-	-		-	≤10	-	-					
TP (mg P/L)		-	-		-	-	-	-		-	-	-	-					
Faecal coliforms (CFU/100 mL or MPN/100 mL)		ND(7d Me) ≤14(Ma)	-		≤0(Me), 14 NTU(Ma),	-	-	ND(7d Me) ≤200(Ma)		ND(7d Me) ≤14(Ma)	-	-	-					
E. coli (CFU/100 mL or MPN/100 mL)		-	-		-	ND	ND	ND(7d Me) ≤200(Ma)		-	-	ND	≤200					
Total coliforms (MPN/100 mL)		-	≤3		-	-	-	-		-	≤3	-	-					
TDS (mg/L)		-	≤1500		-	-	-	-		-	-	-	-					
TOC (mg/L)		-	-		-	-	-	-		-	-	-	-					
Chlorine residual (mg/L)		≥1.0(90min)	≥1.0(30min) ≥0.2(APU)		≥1(30min)	FRC >0.1 or CRFC >0.4	-	≥0.5		-	-	-	-					
Parameter s	Non-potable reuse																	
	Agricultural											Industry						
	USA(FCR)	USA(FCP)	USA(FCN)	China(Farming)	China(Forestry)	China(AH)	Australia(FCR)	Australia(FCP)	Australia(FCN)	EU(FCR)	EU(FCP)	EU(FCN)	USA(OC)	USA(RCT)	China(Cooling)	China(Washing)	China(Boiler)	EU
pH	6.0-9.0	6.0-9.0		5.5-8.5			-	-		-		6.0-9.0		6.5-8.5	6.5-9.0	6.5-8.5	-	
BOD ₅ (mg O ₂ /L)	≤10	≤30		≤35		≤10	Case by case	<20		≤10	≤25		≤30		≤15	≤15	≤15	≤25
COD (mg/L)	-			≤90		≤40		-		-	≤125		-		≤60			-

TSS (mg/L)	-	≤30		≤30				<30		≤10	≤35		≤30		-	-	-	≤35
Colour (colour or Hazen units)	-	-		≤30				-		-			-		≤10	≤30	≤10	-
Turbidity (NTU or mg-kaolin/L)	-	-		≤10 NTU				-		≤5	-		-		≤5 NTU			-
TN (mg N/L)	-	-	-	-	-	-		-	-	-	-	-	-	-	-	-	-	-
NH ₃ -N (mg N/L)	-	-		≤10				-		-			-		≤10			-
TP (mg P/L)	-	-	-	-	-	-		-	-	-	-	-	-	-	-	-	-	-
Faecal coliforms (CFU/100 mL or MPN/100 mL)	ND(7 d Me) ≤14(Ma)	≤200		≤10000				≤2000	-		-			≤200		≤2000		
<i>E. coli</i> (CFU/100 mL or MPN/100 mL)	-	-		-			E.coli < 1 CFU/100 mL	E.coli < 100 CFU/100 mL		≤10 or BD L	≤100	≤1000	-		-	-	-	≤10000
Total coliforms (MPN/100 mL)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
TDS (mg/L)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

TOC (mg/L)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Chlorine residual (mg/L)	≥1.0(90min)	≥1.0(90min)	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Notes: RA: Restricted area; UA: Unrestricted area; FCR: Food crops with raw consumer; FCP: Food crops with processed consumer; FCN: None food crops; AH: Animal husbandry; OC: Once-through cooling; RCT: Recirculation cooling towers; GRS: Groundwater recharge by spreading into potable aquifers; GRI: Groundwater recharge by injection into potable aquifers; ASR: Augmentation of surface water supply reservoirs. Me: Median; Ma: Maximum; ND: Non detectable; IND: If not disinfected; BDL: Below detection limit; FRC: Free residual chlorine; CRFC: Combined residual free chlorine; APU: At point of use.																		

2.11.2 Supporting information 1-2

Table 2-5 AnMBR systems operation parameters

Reactor	Reactor Volume (L)	MSA (m ²)	HRT(h)	SRT(d)	OLR (kg COD/m ³ /d)	Temperature (°C)	Flux (L/m ² /h)	Influent Type	COD _{in} (mg/L)	COD _{ef} (mg/L)	BO _{D5in} (mg/L)	BO _{D5ef} (mg/L)	TSS _{in} (mg/L)	TSS _{ef} (mg/L)	NH ₃ -N _{in} (mg/L)	NH ₃ -N _{ef} (mg/L)	TP _{in} (mg/L)	TP _{ef} (mg/L)	Faecal Coli. _{in} (CFU/100 mL)	Faecal Coli. _{ef} (CFU/100 mL)	pH	MY
GS-F1	350	3.5	19	680	0.85	35	7	RS	630	80	396	25	N/A	N/A	61.5	67.4	8.2	7.1	4.3*10 ⁶	49	6.8	0.27
GS-H1	800	30	12	70	1	33	10	RS	445	77	N/A	N/A	186	N/A	27	33.4	2.7	3.1	N/A	N/A	6.72	0.07
GS-H3	1300	30	12	70	1.2	33	10	RS	459	100	N/A	N/A	242	N/A	28.6	N/A	3.1	N/A	N/A	N/A	6.75	N/A
GS-H5	25	5.4	2	N/A	3	35	6	RS	396	50	N/A	N/A	N/A	N/A	36.7	34.2	10.4	6.5	N/A	N/A	6.9	0.12
GS-H6	550	5.4	9	70	1	23	17	RS	383	36.2	141	7.7	N/A	N/A	40	28	5.1	2.7	N/A	N/A	6.75	0.1
GS-H7	25	0.93	7	infinite	2	18	17	SS	892	122	573	36	123	N/A	71	74	10	23.8	N/A	N/A	N/A	0.25
GS-H8	284	0.93	13.5	N/A	2.25	18	13	SS	978	120	474	56	83	N/A	75	83.2	10	10	N/A	N/A	7.2	0.23
PS-H1	1760	39.5	5	36	1.75	30	6	SS	280	14	160	10	165	ND	N/A	34	N/A	3.7	N/A	N/A	6.7	0.215
RM-H1	50	0.9	33	270	1.75	19	10	RS	1462	129	N/A	N/A	960	N/A	38.9	36.2	22.5	20	N/A	N/A	7.5	0.212
GS-C1	20	0.09	96	215	N/A	35	2	SW	6252.3	101.5	N/A	N/A	N/A	N/A	34.7	346.3	195.4	213.7	N/A	N/A	7	N/A

GS-H15	42.5	0.93	8	N/A	N/A	16.3	10	SS	221	41	106	11	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	8.2	N/A
GSG-H1	15	0.93	8	N/A	N/A	11	12	SS	168	39	88	10	105	ND	N/A	N/A	N/A	N/A	N/A	N/A	8.1	N/A
GSF-H1	15	0.93	8	N/A	N/A	11	12	SS	208	34	138	13	127	ND	N/A	N/A	N/A	N/A	N/A	N/A	7.7	N/A
GS-H9	N/A	N/A	N/A	N/A	N/A	23.5	N/A	RS	N/A	N/A	N/A	N/A	N/A	5.33	N/A	34.9	N/A	9.42	N/A	N/A	N/A	N/A
GS-H10	1300	12.9	13	60	1.3	20.6	7.6	SS	620	58	250	25	330	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
GAC F-H1	990	60	3.9	11	1.4	22.5	7.9	SS	210	29	140	15	120	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
PFPS-H1	1714	13.2	50.4	140	1	60	2	TS	2700	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.35
CST R-H1	2	0.003	12	1000	0.8	35	6	SW	400	50	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	0.3
DFF-T1	2.3	0.059	8	N/A	N/A	25	6	SP E	500	14	235	4	120	N/A	43	N/A	2.5	N/A	N/A	N/A	N/A	N/A
FBR-CT1	2.3	0.05	8	N/A	N/A	25	6	SP E	500	25	235	8	120	N/A	43	N/A	2.5	N/A	N/A	N/A	N/A	N/A
UAS B-H1	N/A	N/A	0.32	N/A	1.6	15	7	SS	100	39.6	N/A	N/A	244	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	0.09
NGS-T1	700	0.2375	8	N/A	N/A	20	2.5	RS	525	150	N/A	N/A	515	16	N/A	N/A	N/A	N/A	N/A	N/A	7.9	N/A
GS-T1	700	0.2375	8	N/A	N/A	19.6	2.5	RS	657	78	N/A	N/A	1307	47	N/A	N/A	N/A	N/A	N/A	N/A	7.8	N/A
CGH 1	3	0.06	12	N/A	N/A	20	5.3	SW	330	N/A	N/A	N/A	N/A	N/A	6.05	N/A	3.35	N/A	N/A	N/A	7	N/A
SGH 1	3	0.06	12	N/A	N/A	20	5.3	SW	330	N/A	N/A	N/A	N/A	N/A	6.05	N/A	3.35	N/A	N/A	N/A	7	N/A

ANH1	900	31	22	40	N/A	25	15.8	RS	643	N/A	N/A	N/A	260	N/A	49.6	N/A	5	N/A	N/A	N/A	N/A	0.35
GL-T1	13	0.066	30	N/A	0.619	35	4.5	SW	1000	55	400	N/A	N/A	N/A	N/A	33	N/A	16	N/A	N/A	7.75	0.19
ANH2	5.4	0.125	30	103	1.3	34	2.1	SW	1720	86	N/A	N/A	N/A	N/A	39	N/A	60	N/A	N/A	N/A	N/A	N/A
ANF1	5	0.1	12	N/A	2	35	9	SW	465	N/A	N/A	N/A	N/A	N/A	10	N/A	8	N/A	N/A	N/A	N/A	N/A
GS-F3	3.2	0.1	12	200	N/A	36.5	30	SW	460	15	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	6.8	0.3
GS-F4	3.2	0.1	6	100	N/A	36.5	30	SW	500	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	N/A
GS-F5	3	0.116	6	200	N/A	35	15	SW	466	15	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	0.252
GS-F6	3	0.1	5	N/A	N/A	N/A	N/A	OF MS W	1410	400	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
GS-FPAC 1	3	0.1	6	250	16	35	5	SW	4000	70	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	N/A
GS-F7	3	0.1	12	N/A	N/A	N/A	6	SW	460	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	6.95	N/A
GS-H16	1000	12.5	16	100	N/A	25	6	SS	338	54	167	10	84	ND	35	40	N/A	N/A	N/A	N/A	8.1	N/A
GSG-H2	85	0.93	16	100	N/A	25	6	SS	338	47	167	11	84	ND	35	39	N/A	N/A	N/A	N/A	8.1	N/A
GS-F8	N/A	0.11	6	150	N/A	35	26	SW	500	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	N/A
GS-F9	3.2	0.11	6	213	N/A	35	5	SW	500	N/A	N/A	N/A	N/A	N/A	100	N/A	N/A	N/A	N/A	N/A	N/A	N/A

GS-F10	3	0.116	4	200	N/A	35	15	SW	484	12	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	N/A
GS-H11	3.5	0.08	8	60	0.55	22	N/A	SW	550	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	7	N/A
GS-H12	318	1.86	6	N/A	N/A	24	12	RS	1729	150	N/A	N/A	964	N/A	55.4	69.16	9.55	12.6	4*10 ⁶ -2*10 ⁷	42+-48	7.22	N/A
GS-F11	3.5	0.08	6	120	N/A	35	15	SW	347	20	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
GS-H13	1300	31	24.4	140	0.49	27	N/A	SS	510	59	305	14	342	2	42.8	47.9	5.5	6.7	N/A	N/A	N/A	N/A
GS-SiCF1	500	6.02	8.2	250	3	35	10.1	MRW	1138	61	N/A	N/A	942	N/A	9	N/A	14	N/A	N/A	N/A	7	0.09
GSD M-F1	3.5	0.02	8	infinite	0.88	20	22.5	SW	292	73.2	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
SAF MBR-1	9	0.158	4.5	N/A	1.83	N/A	5.6	SW	115	10.6	53	61.2	42	N/A	74	N/A	5	N/A	N/A	N/A	7	N/A
ANH3	8	0.2	12	300	N/A	35	1.68	SW	570.41	40.94	N/A	N/A	N/A	N/A	36.03	44.93	4.84	6.08	N/A	N/A	N/A	N/A
ANH4	8	0.2	12	300	N/A	25	1.68	SW	570.41	40.94	N/A	N/A	N/A	N/A	36.03	42.84	4.84	5.79	N/A	N/A	N/A	N/A
GS-H14	5	0.78	6.4	N/A	N/A	30	10	SW	409	18	N/A	N/A	N/A	N/A	43.9	41.9	4.8	4.4	N/A	N/A	N/A	0.28
Average	322.60	6.20	12.45	194.57	2.08	27.68	9.65	N/A	807.34	68.34	236.94	19.81	339.32	17.58	41.33	64.03	17.99	23.15	N/A	49.00	7.22	0.212

Notes: MSA: Membrane surface area; MY: Methane yield (L CH₄/g COD_{removed}); N/A: Not available; RS: Raw sewage; SS: Settled sewage; TS: Toilet sewage; SW: Synthetic wastewater; SPE: Synthetic primary effluent; OFMSW: Organic fraction of municipal solid waste; MRW: Mixed raw sewage.

3 A comparative evaluation of reverse osmosis membrane performance when combined with Anaerobic or Aerobic membrane bioreactors for indirect potable reuse applications

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Abstract

A combination of membrane bioreactor (MBR) and reverse osmosis (RO) technologies are a promising option for delivering potable water reuse, utilising either aerobic (Ae) or anaerobic (An) MBR systems. Interestingly, the AnMBR offers several advantages over its aerobic counterpart including lower energy demand and the production of a methane rich biogas than can be converted to energy. However, the comparative impact of different effluent matrices from these two types of MBR on RO filtration and fouling behaviour has never been directly addressed. In this study, the filtration performance and fouling behaviour was investigated with a dead-end RO filtration cell fed with the effluent from an AnMBR and an AeMBR treating municipal wastewater. Analysis suggests that both AnMBR and AeMBR followed by RO can produce a water quality sufficient for indirect potable water reuse. However, the unremoved ammonia in the AnMBR effluent may partially go through into the RO permeate and has the potential to exceed the threshold in Singapore's PUB NEWater standard. A fluorescence 3D excitation-emission matrix analysis suggested RO can effectively remove disinfection by-products precursors, ensuring the safety for chlorine based reuse water distribution. During 33 filtration tests with each MBR effluent, the membrane resistance, elemental analysis, and membrane surface imaging results demonstrated that the AnMBR-RO system had higher membrane fouling levels compared to an AeMBR-RO, with the foulant deposition sequence being principally organic and colloidal at first, followed by inorganic substances.

Multivariate analysis of elemental analysis results indicated that the ammonia in the AnMBR and nitrate in the AeMBR effluent had led to a different co-deposition of substances on the membrane surface. The mass balance of major foulants found that TOC and Ca were the most deposited foulants in both effluents, while the deposition rate of ammonia (13.8 %) was significantly higher than nitrate (0.02%). From an application point of view, the findings suggest that the combination of AnMBR with RO offer a more sustainable approach than with the AeMBR but nutrients removal, with the potential of recovery, is recommended before the RO membranes to limit fouling propensity and achieve a permeate of sufficient quality.

Keywords:

Aerobic; anaerobic; indirect potable water reuse; membrane bioreactor; reverse osmosis; membrane fouling

3.1 Introduction

Water reuse has the potential to make a significant contribution to overcoming the challenges of water scarcity and growing demands on fresh water supply (Capocelli & Piemonte, 2021; National Research Council, 2012). There are growing numbers of water reuse schemes providing non-potable supplies for agricultural irrigation and industrial cooling in many countries (Asano et al., 2007; Salgot & Folch, 2018). Furthermore, potable water reuse applications are becoming more widely implemented across the world, including surface and ground water recharge schemes which blend treated effluent with raw water before final abstraction for drinking water production (indirect potable reuse) and directly using treated effluent as a source of drinking water (direct potable reuse) (Lazarova, 2015; Lee & Tan, 2016). To ensure the quality and safety of the purified wastewater, water quality parameters have to be monitored and controlled including organics, nutrients, pathogens, trace elements including for example disinfection by-products (DBPs) (CNEPA, 2021; ISO, 2018; USEPA, 2017).

In this context, membrane bioreactor (MBR) technologies offer several advantages over conventional treatment systems, delivering a robust, solids free effluent which is substantially disinfected (Delanka-Pedige et al., 2020). The combination of a conventional activated sludge process with membrane filtration also significantly reduces the plant footprint (Santos et al., 2011). Typical MBR applications are based on aerobic reactors (AeMBR). Anaerobic membrane bioreactors (AnMBR) operating in an oxygen free environment are an alternative form of MBR which has attracted extensive interest in recent years. In comparison to AeMBR, the biological process of an AnMBR digests the organic compounds in the absence of oxygen which requires no energy demand for aeration, and produces a methane rich biogas which offers the potential to produce energy from the methane, leading to lower energy consumption as well as a further reduced footprint (Shin & Bae, 2018; Wang et al., 2018).

Reverse osmosis (RO) filtration is commonly used for potable reuse applications (Jeffrey et al., 2022; Plevri et al., 2017; Wintgens et al., 2005). With a typical pore

size between 0.1–1.0 nm these membranes allow the rejection of the smallest contaminants and monovalent ions (Shen et al., 2016). The combination of RO with AeMBR has been shown to deliver a reliable and high quality effluent for potable water reuse (Shareefdeen et al., 2015; Wintgens et al., 2005) specifically when strict organic micropollutant threshold values are set (Plevri et al., 2017, 2018). Indeed, the combined systems have been shown to remove almost 99.9% of particles, pathogens, viruses, and organic micro-pollutants (OMPs), as well as the vast majority of nutrients, thus posing no organic and pathogen concerns as the resulting effluent has high bio-stability (Zhai, Liu and van der Meer, 2021). These advantages raise the possibility of combining RO with AnMBRs to produce potable reuse quality effluent but with lower overall energy consumption. In this context, Gu *et al.*, (2019) have reported a combined AnMBR-RO-IE (ion exchange) system able to produce NEWater-like water (PUB, 2019) while recovering energy. However, the effluents from AeMBRs and AnMBRs have different matrices that may impact the RO filtration performance and in particular the fouling mechanism differently. Shin *et al.*, (2021) reported that at pH 6, the total ammonia nitrogen rejection efficiency of RO was optimal at 99.8% and Liu *et al.*, (2020) suggested that the presence of N and P at higher levels may lead to severe membrane fouling due to biofilm formation. Available literature comparing the performance of AnMBRs with AeMBRs is focusing on several aspects, as for example, Wang *et al.*, (2018) demonstrated that AnMBRs have a better energy efficiency treating municipal wastewater compared to AeMBRs, Zaouri *et al.*, (2021) showed that both AeMBR and AnMBR effluents have different organic and nutrient content as well as concentrations of emerging contaminants and heavy metals, and Liu *et al.*, (2020) reported a higher removal of most trace organics (TrOCs) by AeMBR whilst AnMBR effluent has a higher level of TrOCs with nitrogen in their molecular structures. However, no literature has been found which focuses on the comparison of AnMBR and AeMBR effluents for potable reuse applications, specifically on the impact of the different nitrogen species present in the effluents.

Several studies which have looked at potable water reuse through an AeMBR-RO system have focused on the fate of TrOCs (Alturki et al., 2010; Kim et al.,

2007; Mamo et al., 2018). Other studies have reported that the organic matter in MBR effluent plays an important role in the evolution of RO fouling (Kimura et al., 2016) and that the incomplete removal of nitrate by the RO membrane poses a potential concern of catalysing DBP formation (Comerton et al., 2005, 2006). RO processes can simultaneously remove both organic and inorganic DBP precursors thereby providing an important option for DBP minimization (Zazouli & Kalankesh, 2017). Studies on AnMBR-RO systems are very few compared to those on AeMBR-RO. Despite this interest in coupling AnMBRs with RO for reuse, several knowledge gaps remain if we are to better understand the feasibility of AnMBR-RO systems for potable water reuse, specifically the impact which the different effluent matrix, when compared to an AeMBR, may have on treatment performance and fouling mechanisms. The fate of DBP precursors through each technology is also unclear. Therefore, a comparative evaluation of reverse osmosis membranes combined with both anaerobic and aerobic MBR treating municipal wastewater for potable reuse applications would provide significant advances in the field.

This paper then offers the first report of a direct comparison between an AnMBR and an AeMBR treating real municipal wastewater for potable water reuse when coupled with reverse osmosis membranes. The study compares the performance of both options against a comprehensive range of water quality parameters and trace elements listed in PUB's NEWater standard. It also specifically examines the impact of various key species on RO fouling from each effluent. The fate of DBP precursors was also studied to understand the safety of indirect potable water reuse application. The major RO foulants were qualitatively and quantitatively characterized for AnMBR and AnMBR effluent, respectively.

3.2 Materials and methods

3.2.1 Pilot scale anaerobic membrane bioreactor and aerobic membrane bioreactor

AnMBR and AeMBR pilot plants (Figure 3-1) were installed in Cranfield University's National Research Facility for Water and Wastewater Treatment and operated stably for 55 weeks. The systems were fed with the raw wastewater

from the Cranfield University sewage works after settling in a primary settling tank. The AnMBR was configured as a cylindrical upflow sludge blanket bioreactor (UASB-45L) and a side stream membrane tank (30L), seeded with granular sludge obtained from a system treating industrial wastewater from the paper industry (Saica Paper, Manchester, UK). The sludge was acclimatised to the settled wastewater for 21 days until performance stabilised before focused trials started. The settled municipal wastewater was fed by a peristaltic pump (Watson Marlow 620s, UK). Internal recirculation between the membrane tank and the UASB reactor was maintained by a peristaltic pump (Watson Marlow 520s, UK). The AnMBR was operated under ambient temperature with a 0.9 m/h upflow velocity, 8 hours hydraulic retention time (HRT), and infinite sludge retention time (SRT). The AeMBR was configured with a level control tank (10L), aerobic tank (40L) and membrane tank (18L), seeded with activated sludge obtained from the Cotton Valley sewage treatment works (Anglian Water, Milton Keynes, UK). The AeMBR was operated under ambient temperature with 12 hours HRT and 7 days SRT. DO in the aerobic tank was controlled to above 1 mg/L and the mixed liquor suspended solids in the membrane tank was controlled between 8 000-12 000 mg/L. Recirculation was maintained by a peristaltic pump (Watson Marlow 520s, UK) at the rate of 200%. For both systems, a PVDF hollow fibre ultra-filtration (UF) membrane with 0.93 m² surface area and 0.04 µm nominal pore size (GE ZW-10, USA) was placed in the membrane tank and operating with a flux of 10 LMH. Permeate was produced by suction through a timer controlled peristaltic pump (Watson Marlow 520s, UK) with 60 mins permeation and 30 mins relaxation periods. For the AnMBR, nitrogen gas was continuously sparging into the membrane module with a constant specific gas demand of 0.25-1.0 m³m⁻²h⁻¹. For the AeMBR, compressed air was continuously sparging into the aerobic tank and membrane module through a fine air bubble diffusor at a rate of 0.93 m³h⁻¹ and a course air diffusor for a specific aeration demand of 0.25-1.0 m³m⁻²h⁻¹, respectively. Due to the conservative approach in the operation of the membranes, during the 55 weeks of operation of the two MBRs, no membrane cleaning was required.

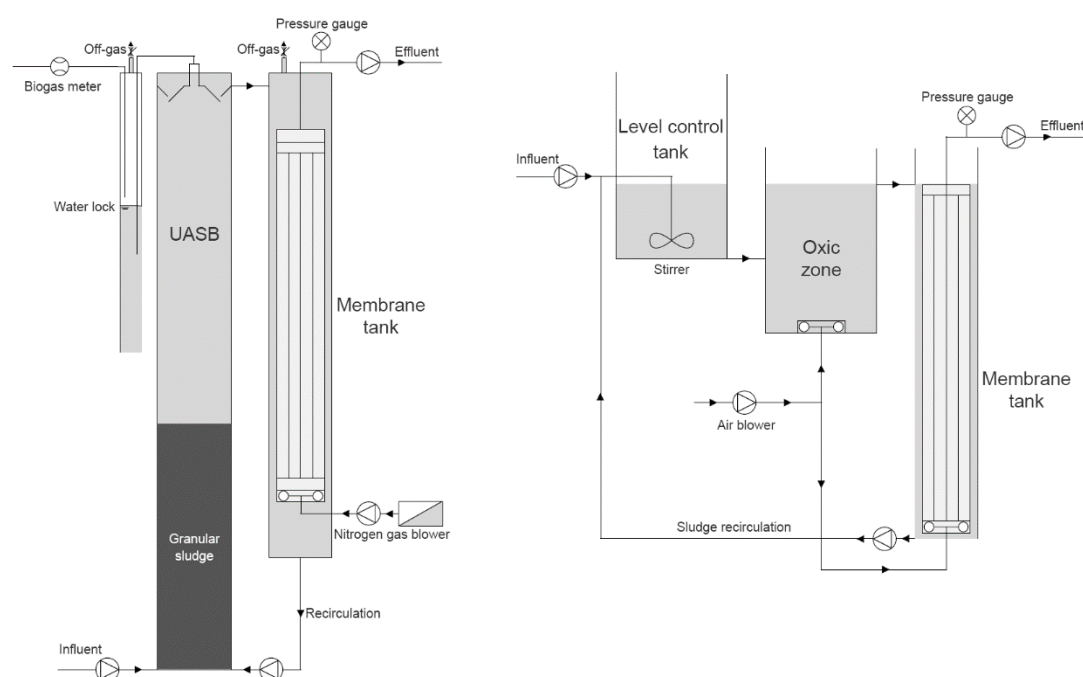


Figure 3-1 Process flow diagram for AnMBR and AeMBR

3.2.2 Bench scale reverse osmosis membrane cell

A dead-end bench scale (300 ml) reverse osmosis (RO) membrane cell system (Sterlitech HP4750, USA) was used for the filtration tests with the effluents from both AnMBR and AeMBR. The pressure driven filtration was achieved with a pre-cut 47 mm diameter polyamide-urea-thin film composite hydrophilic flat sheet reverse osmosis membrane (TriSep X201, USA) with an active membrane area of 14.6 cm². For the tests, a constant 10 Bar pressure was maintained by a regulator (BOC, UK) with bottled oxygen free nitrogen gas (BOC 1066, UK). A constant 400 RPM stirring rate was maintained with a magnetic stirrer (Benchmark 3770, UK) to mix the MBR effluents and limit concentration polarisation at the surface of the membrane. For each test, a 60 ml sample of the AnMBR/AeMBR effluent was sealed into the membrane cell and filtered with a new RO membrane coupon until a recovery of 75% was achieved. After the filtration, the 45 ml RO permeate and 15 ml RO concentrate samples were analysed. The new RO membrane coupon was first soaked in deionised (DI) water (ELGA, UK) for 12 hours prior to the filtration test. Clean water flux tests were carried out with DI water before and after each filtration test to determine

the clean water permeability with and without fouling and allow the calculation of the resistance. For the clean water test, the filtration time for 5 ml of DI water was recorded.

Membrane fouling resistance was calculated with the equation according to Darcy's law:

$$R_f = R_t - R_0 = \frac{P}{\mu \cdot J_t} - \frac{P}{\mu \cdot J_0} \quad (3-1)$$

Where, R_f (m^{-1}) = resistance of fouling layer, R_t (m^{-1}) = total resistance at time t , R_0 (m^{-1}) = clean membrane resistance, t (s) = filtration time, J_0 ($m^3 \cdot m^{-2} \cdot s^{-1}$) = clean membrane flux, J_t ($m^3 \cdot m^{-2} \cdot s^{-1}$) = flux at time t , p (Pa) = pressure, μ ($Pa \cdot s^{-1}$) = DI water viscosity = 0.01.

Deposited foulants mass was calculated with the equation according to mass balance law:

$$m_{ROD} = C_0 \times 60 - C_{ROP} \times 45 - C_{ROC} \times 15 \quad (3-2)$$

Where, m_{ROS} (μg) = mass of the substance deposited on the RO membrane surface (ROS), C_0 ($\mu g/L$) = substance concentration in the RO feed, C_{ROP} ($\mu g/L$) = substance concentration in the RO permeate (ROP), and C_{ROC} ($\mu g/L$) = substance concentration in the RO concentrate (ROC).

The foulant deposition rate was calculated with the equation adapted from (Tang et al., 2014, 2016):

$$R_{\%} = \frac{m_{ROD}}{m_0} \quad (3-3)$$

Where, R_D (%) = the foulant deposition percentage, and m_0 (μg) = mass of the substance in the RO feed.

3.2.3 Analytical measurements

Temperature, pH and total dissolved solids (TDS) were measured by an online probe (Endress+Hauser, Switzerland). Total suspended solids (TSS), 5 day biochemical oxygen demand (BOD_5), sludge volume index (SVI) were measured

based on the Standard Methods (APHA, 1999). Chemical oxygen demand (COD), ammonia nitrogen (NH₃-N), nitrite (NO₂-N), nitrate (NO₃-N), phosphate (PO₄-P), and total phosphorus (TP) were analysed with cell tests and a photo spectrometer (Merck Spectroquant, Germany). Total coliform (TC), Faecal coliform (FC) and *Escherichia coli* (*E. Coli*) were measured by Quanti-Tray (Coliler-18, USA) analysis. Total organic carbon (TOC) and total nitrogen (TN) were measured by a Shimadzu TOC analyser (Shimadzu 3201, Japan). Trace level elements were analysed with an inductively coupled plasma mass spectrometry (ICP-MS) (PerkinElmer NexION 350d, USA). Characterization of the MBRs influents were measured after filtering with 0.45 µm syringe membrane disc filter (Merck Millipore, Germany). Organic composition was evaluated by a fluorescence spectrometer (HORIBA FLuoroMax+, Japan) with a 3D excitation–emission matrices. Scan settings were 200–400 nm excitation and 280–500 nm emission wavelength, 1 nm entrance slit and an integration time of 0.1 second. The 1st and 2nd Rayleigh scatter were masked with 5 nm slit width by FluorEssence software (HORIBA, Japan). The fluorescence intensity maps were masked together by light intensity order to increase the accuracy while minimizing the signal noise and system error. Each map displayed a colour metric bar to quantify the detected peak intensities, where the intensity shifted from low to high within blue-green-yellow-red. Results from Fluorescence spectrometer were plotted as a 2D intensity map and the fluorescence regional integration (FRI) analysis was extracted with a 5 region DOM categories method (aromatic protein (AP): tyrosine-like (APr) excitation/emission (E_x/E_m) = (200–250) nm/(280–330) nm; tryptophan-like (APy) E_x/E_m = (200–250) nm/(330–380) nm; fulvic acid-like (FA) E_x/E_m = (200–260) nm/(380–500) nm; soluble microbial products (SMP) E_x/E_m = (250–280) nm/(310–380) nm; humic acid-like (HA) E_x/E_m = (280–380) nm/(380–500) nm) (Chen et al., 2003; Pan et al., 2021) using an R scrip modified from Lapworth and Kinniburgh (2009). The foulant layer accumulated on the RO membrane coupons during the filtration process was characterized by a scanning electron microscope with energy dispersive spectroscopy (SEM-EDS) for imaging (Tescan S8000G, Czech Republic) and EDS (Oxford Instruments Ultim Max detector, UK) analysis with AZtec software (Oxford Instruments, UK). A

virgin RO membrane and fouled membranes obtained from each filtration batch (5 AnMBR-RO and 5 AeMBR-RO) were dried under constant room temperature before a 1 cm² piece was cut from the centre of the membrane. The cut membrane was then adhered to a carbon tab and coated with 10 nm gold, finally the coated membrane sample was transferred to the SEM-EDS. SEM imaging was done in secondary electron mode for topography, EDS analysis was done for 5 selected areas on each site in SEM images with backscatter mode under low vacuum for atomic number contrast. Raw numerical data was input to JMP 16 pro to obtain a multivariate correlation analysis and statistical analysis.

3.3 Results and discussion

3.3.1 MBRs influent and effluents quality

Typical municipal wastewater parameter values were measured for the influent feeding the two MBRs, with a slightly lower strength ($BOD_5/COD=0.38$) compared to others reported in the literature on MBR systems ($B/C > 0.5$) (Song et al., 2018; Tang et al., 2018; Visvanathan & Abeynayaka, 2012). This can be explained by the fact that the majority of the trials were conducted during the COVID pandemic and that the population on the university campus was lower than usual which would have affected some of the characteristics of the raw sewage. There was no organic phosphate detected during the whole period of the experiment and the concentration of total phosphorus (TP) was equal to that for PO_4-P (Table 3-1).

Table 3-1 Water quality parameters and trace level elements analysis for the influent wastewater, AnMBR and AeMBR effluent during the 55 weeks of operation

Unit	Influent		AnMBR Effluent		AeMBR Effluent	
mg/L	Mean	Std	Mean	Std	Mean	Std
pH	7.9	±0.5	7.6	±0.5	7.5	±0.4
°C	14.3	±5.3	13.3	±6.2	13.4	±6.2
TDS	410.4	±64.8	379.2	±72.6	304.5	±77.5
TSS	112.1	±31.1	ND*		ND*	
COD	218.9	±59.4	37.6	±15.5	10.1	±8.5
BOD ₅	83.3	±39.7	9.3	±6.5	2.7	±1.7

TOC	24.6	±5.5	17.9	±5.8	6.7	±1.3
NH₃-N	23.7	±5.3	25.0	±7.0	ND*	
NO₃-N	0.9	±1.8	ND*		17.6	±4.4
NO₂-N	0.4	±0.6	ND*		ND*	
Or-N	4.5	±1.6	2.3	±0.5	0.5	±0.2
TN	31.3	±4.5	28.8	±6.5	24.1	±3.5
PO₄-P	3.9	±1.4	4.0	±1.2	4.3	±1.5
Or-P	ND*		ND*		ND*	
µg/L	Mean	Std	Mean	Std	Mean	Std
B	54.17	±20.64	73.00	±9.85	64.39	±11.06
Na	44766.67	±9016.80	50780.95	±11015.06	50647.62	±9502.61
Mg	5283.33	±383.54	5373.33	±329.14	5472.38	±485.74
Al	ND*		ND*		ND*	
Si	7473.33	±552.47	7028.57	±974.55	6840.00	±309.71
K	13636.67	±1391.89	13051.43	±2610.50	12712.38	±1522.31
Ca	42733.33	±5845.57	48657.14	±8576.75	49190.48	±9831.27
Mn	11.43	±6.26	2.08	±5.90	ND*	
Fe	47.15	±20.91	41.65	±14.93	34.12	±17.34
Cu	62.17	±12.89	59.10	±11.41	61.80	±19.62
Zn	6.43	±13.03	11.45	±13.81	16.04	±11.36
Sr	171.57	±38.55	198.65	±44.63	200.12	±51.74
Cd	0.67	±1.59	ND*		ND*	
Ba	45.83	±3.09	37.90	±2.58	36.26	±4.40
Pb	0.04	±0.10	ND*		ND*	

Note: ND*, Non detectable.

Similar pH values were measured for the AnMBR (7.6) and AeMBR (7.5) effluents and the temperature was almost the same since both reactors were operated under ambient temperature. Under this pH, ammonia was mostly in the form of ammonium (NH₄⁺) under which conditions a lower ammonium removal (97%-99%) may be expected during RO filtration (Shin et al., 2021). The AnMBR effluent had a slightly higher total dissolved solids (TDS) concentration than for the AeMBR, indicating there was more dissolved content to be sent on to the RO membrane. Due to the UF membrane, total suspended solids (TSS) and coliforms (TC, FC and E. coli) were not detected in both effluents. This is in agreement with previous literature which has highlighted that there is no concern with TSS and

coliforms under undamaged membrane conditions (Foglia et al., 2019), as well as the evidence for satisfying the NEWater standard's threshold for microbial barrier even before the RO filtration. Additionally, this suggested there should be no particulate fouling and limited biofouling formation on the RO. In terms of organic removal performance, the AeMBR achieved a better removal of COD (95%), BOD₅ (97%) and TOC (73%) compared to the AnMBR with 83%, 89% and 27%, respectively. The unremoved organics may cause organic fouling of the RO membrane and decrease the filtration performance since the adsorption of organic substances onto the membrane surface has been shown to form an organic film contributing to fouling (Martínez et al., 2015). Phosphate levels of the two effluents were very similar, although the AeMBR effluent had a slightly higher concentration as a result of high SRT and MLSS. Literature has suggested the presence of phosphate can cause inorganic scaling in particular with calcium phosphate, and simultaneously absorbing silica onto the surface hydroxyl functional groups, which also causes silica deposition on the RO membrane surface since phosphate polymers are structurally very similar to those of silica (Malki & Abbas, 2013). There was no nitrite (NO₂-N) detected in both effluents. However, other nitrogen species, specifically ammonia nitrogen (NH₃-N) and nitrate (NO₃-N), were found to constitute the largest difference between the AnMBR and AeMBR effluents. There was no nitrate detected in the AnMBR effluent and nearly no removal was found for ammonium. Oppositely, for the AeMBR effluent, there was no ammonium and 17.6±4.4 mg/L of nitrate. This can be explained as a function of the different biological processes happening before the membrane modules. Anaerobic biomass converts organics to methane-rich biogas and also reduces the nitrate and nitrite to ammonia in the absence of oxygen while the aerobic process oxidises the ammonia to nitrite then to nitrate with biological nitrification in the presence of oxygen (Gerardi, 2002; Shin & Bae, 2018). A minor difference in the TN removal was found between the two MBR effluents, which may be explained by the different organic nitrogen (Or-N) removal during the biological stage, where the AeMBR removed more Or-N in the presence of oxygen compared to the AnMBR. Interestingly, there is no literature on the impact of ammonia and nitrate on RO fouling and this may be a critical

insight to understand the fundamental role for AnMBR combined with RO as a post-treatment process for high grade water reuse applications.

Trace element analysis displayed similar results for the AnMBR and AeMBR effluents. There was no meaningful removal of Fe or Cu whereas the low levels of Cd and Pb in the influent streams were below the limit of detection. Levels of B, Na, Mg, Si, K, Ca, Cu, Zn, Sr, and Ba in the effluents remained similar to the influent. However, trace levels of Mn were found in the AnMBR effluent but not in the AeMBR effluent. This was due to the different SRT applied to the two MBRs. The removal mechanism for heavy metals is mainly by sludge sorption (Arévalo et al., 2013; Plevri et al., 2018). The AnMBR in this study was operated with an infinite SRT but for the AeMBR the sludge was discharged regularly each week, therefore the AnMBR sludge had possibly reached the breakthrough point for its sorption capacity. Among those elements detected in the MBR effluents, Fe, Ca, Mn, Si and Mg are reported to be major inorganic elements of RO Membrane surface deposits (Tang et al., 2014), causing inorganic scaling of the RO membrane (Tang et al., 2016). Additionally, the co-presence of silica and organic matter can cause severe membrane fouling for RO filtering MBR effluent, whereas silica alone did not cause severe fouling (Kimura et al., 2016). This highlights the need to understand the interactions between silica and specific organic matter such as proteins and soluble microbial products (SMPs). Ultimately, the difference in organic matter and nitrogen species may indicate that higher RO membrane fouling should be expected when coupled with an AnMBR compared to an AeMBR.

3.3.2 RO performance with AnMBR and AeMBR effluents

3.3.2.1 Indirect Potable water reuse parameters and trace elements

Identical analytical measurement was conducted for the RO permeates (ROP) obtained from the AnMBR and AeMBR effluents (Table 3-2). The average pH of the RO permeate from the AnMBR (6.0) and AeMBR (5.8) effluents were slightly lower than the Singapore's PUB NEWater standard requirement (7.0-8.5) (PUB, 2020), which could be stabilised by adding lime (Lee & Tan, 2016) or blending with another source of water (Comerton et al., 2006; Zhai et al., 2021). Significant

removal of TDS, COD, NH₃-N (AnMBR), NO₃-N (AeMBR), Or-N, TN, PO₄-P, and TOC were found for both permeates and a complete non detectable BOD₅ was achieved, whilst the TDS for AnMBR and AeMBR permeates was respectively lower than the threshold in PUB NEWater standard (PUB, 2020). Indeed, only 88% ammonia and 87% nitrate removal were achieved respectively in the AnMBR-ROP and AeMBR-ROP. The incomplete removal of ammonia was reported by Qin, Oo and Coniglio (2005) as the rejection efficiency of ammonia varies with pH and flux due to the presence of deprotonated non-ionic ammonia. Residual ammonia may pose a concern during chlorine based disinfection processes as the formed chloramine can react with aromatic protein-like (AP) substances which results in N-Nitrosodimethylamine (NDMA) formation (Ma et al., 2015). The ammonia concentration in the AnMBR-ROP (4.0 ± 1.9) exceeded the 1 mg/L limit specified in the NEWater standard (PUB, 2020). This suggests that the pre-removal of ammonia, or a post-polishing process is required when combining RO with AnMBR for indirect potable reuse purposes. Indeed, literature has widely reported that buffering the feed water to an optimal pH range from 6 to 6.5 can significantly enhance the ammonia rejection up to 99.8% (Cancino-Madariaga et al., 2011; Masse et al., 2015; Shin et al., 2021). Moreover, the presence of ammonia in the AnMBR effluent can be useful for chloramine production before the RO membrane when a chlorine based fouling control regime is applied (Gare, 2002) since also Huang et al. (2017) suggested that chloramination achieved with the sequence of adding ammonia then chlorine generates less DBP than adding chlorine followed by ammonia. In contrast, nitrate removal by RO under neutral pH may be reduced in the presence of sodium and sulphates, which has been shown to reduce the removal percentage down to 50% (Kapoor & Viraraghavan, 1997). However, the nitrate concentration in the AeMBR-ROP met the 5 mg/L regulation limit (PUB, 2020) but Comerton et al. (2005, 2006) have suggested the residual nitrate may cause a concern in the context of DBP formation.

Table 3-2 RO performance on treating AnMBR and AeMBR effluent

Unit	AnMBR-RO Effluent			AeMBR-RO Effluent			PUB NEWater Standard
mg/L	Mean	Std	Removal%	Mean	Std	Removal%	(PUB, 2020)
pH	6.0	±1.9		5.8	±1.8		7.0-8.5
°C	13.3	±6.2		13.4	±6.2		
TDS	37.5	±11.7	93%	33.3	±10.4	92%	<150
COD	1.9	±2.5	96%	1.3	±2.1	90%	
BOD ₅	ND			ND			
TOC	1.8	±1.0	92%	1.1	±0.4	88%	
NH ₃ -N	4.0	±1.9	88%	ND			<1
NO ₃ -N	ND			3.0	±1.4	87%	<5
Or-N	ND			ND			
TN	5.3	±1.8	86%	4.1	±1.2	87%	
PO ₄ -P	0.3	±0.3	95%	0.3	±0.3	95%	
µg/L	Mean	Std	Removal%	Mean	Std	Removal%	
B	36.73	±11.28	62%	45.04	±12.89	48%	<50
Na	5624.44	±1806.15	92%	7248.89	±2002.88	89%	<20 000
Mg	222.98	±149.47	97%	295.56	±149.96	96%	
Al	ND			ND			<100
Si	475.33	±158.92	95%	541.47	±283.47	94%	<3 000
K	1566.89	±469.39	91%	1814.67	±475.72	89%	
Ca	2024.00	±1226.37	97%	2830.22	±2016.54	96%	<20 000
Mn	ND			ND			<50
Fe	5.38	±1.25	90%	ND			<40
Cu	4.65	±1.16	94%	7.09	±62.38	91%	<50
Zn	1.27	±1.89	92%	1.38	±1.37	94%	<100
Sr	5.90	±5.12	98%	8.77	±5.25	97%	<100
Cd	ND			ND			
Ba	2.34	±1.27	95%	3.16	±1.01	93%	<100
Pb	ND			ND			

Note: ND*, Non detectable.

Excellent removal was observed for Mg, Si, K, Ca, Mn, Fe, Cu, Zn, Sr, and Ba in both permeates and the previously reported traces of Mn and Fe in the AnMBR

and AeMBR effluents were removed, whilst the concentrations of B, Na, Al, Si, Ca, Mn, Fe, Cu, Zn Sr, and Ba in AnMBR-ROP and AeMBR-ROP satisfied the limit in the NEWater standard (PUB, 2020). However, the rejection rate of Na was 92% and 89% for the AnMBR and AeMBR effluent, respectively, which is lower than the manufacturer's specification (98.5%). This can be explained by the more complex feed water matrix and operating conditions used during the study as compared with the manufacturer's testing regime which uses ultrapure water with dissolved high purity sodium chloride. Low Boron (B) removal was found in both permeates with 62% and 48% removal for the AnMBR-ROP and AeMBR-ROP, respectively. This finding agrees with Tu, Nghiem and Chivas (2010) that the Boron rejection rate increases with increasing pH and operating pressure but decreases when temperature and fouling increase. In this study the less preferable feed water pH and operating pressure led to the low boron removal.

Overall, the combination of RO and AeMBR is able to produce a sufficiently high quality permeate for a potential indirect potable water reuse, with inadequate ammonia removal being a notable weakness of the AnMBR-RO combination. The RO step successfully act as the barrier to offset the insufficient organic and phosphate removal in both MBR effluents, as well as the potential heavy metal concern initially observed in the AnMBR effluent.

3.3.2.2 Impact of dissolved organic matter on RO treatment performance

With fluorescence spectroscopy, a wide intense peak present in Zone V indicates the presence of a significant amount of humic acid (HA) (Figure 3-2.a), while the peaks in Zones III and IV confirmed the presence of some fulvic acid-like substances (FA) and soluble microbial products (SMP) respectively. The AnMBR (Figure 3-2.b) and AeMBR (Figure 3-2.c) effluents displayed a similar distribution as both intensity maps have a deep red coloured peak shifted to light green in Zone V, indicating the major substance present is HA. However, the middle left light blue peak in the AnMBR effluent map suggests a higher content of SMP compared to the AeMBR effluent. Both intensity maps of the RO permeates (Figure 3-2.d and 3-2.e) exhibit a large area of blue with noticeable unmasked 2nd order Rayleigh scatter and several light blue peaks in Zone V for HA. However,

there was very little indication of aromatic protein (AP) (Zones I and II) present in the AeMBR-ROP effluent and none at all was found in the AnMBR-ROP effluent. This suggests the better removal of DOM will lead to a higher level of membrane fouling during the filtration of the AnMBR effluent (Chaukura et al., 2020).

The similar peak distribution for the AnMBR and AeMBR effluent indicates that a similar DOM fouling behaviour should be expected during RO filtration. However, the intensity map only displayed the relative concentration detected in each sample, offering no quantitative comparison. A further Fluorescence Regional Integration analysis (FRI) and mass balance was needed to quantify the concentration of each species. The Ca concentration detected in both MBR effluents in this study exceeded 1 mM, which is reported as the critical value of Ca^{2+} under a neutral pH to bind humic acid molecules together through the bridging effect, and partially neutralize the negative charge on humic acid thereby greatly enhancing the collision efficiency of approaching humic material onto membrane surfaces (Tang et al., 2007a, 2007b). This finding ultimately highlighted the need for further analysis using SEM-EDS analysis for membrane surface foulant characterization.

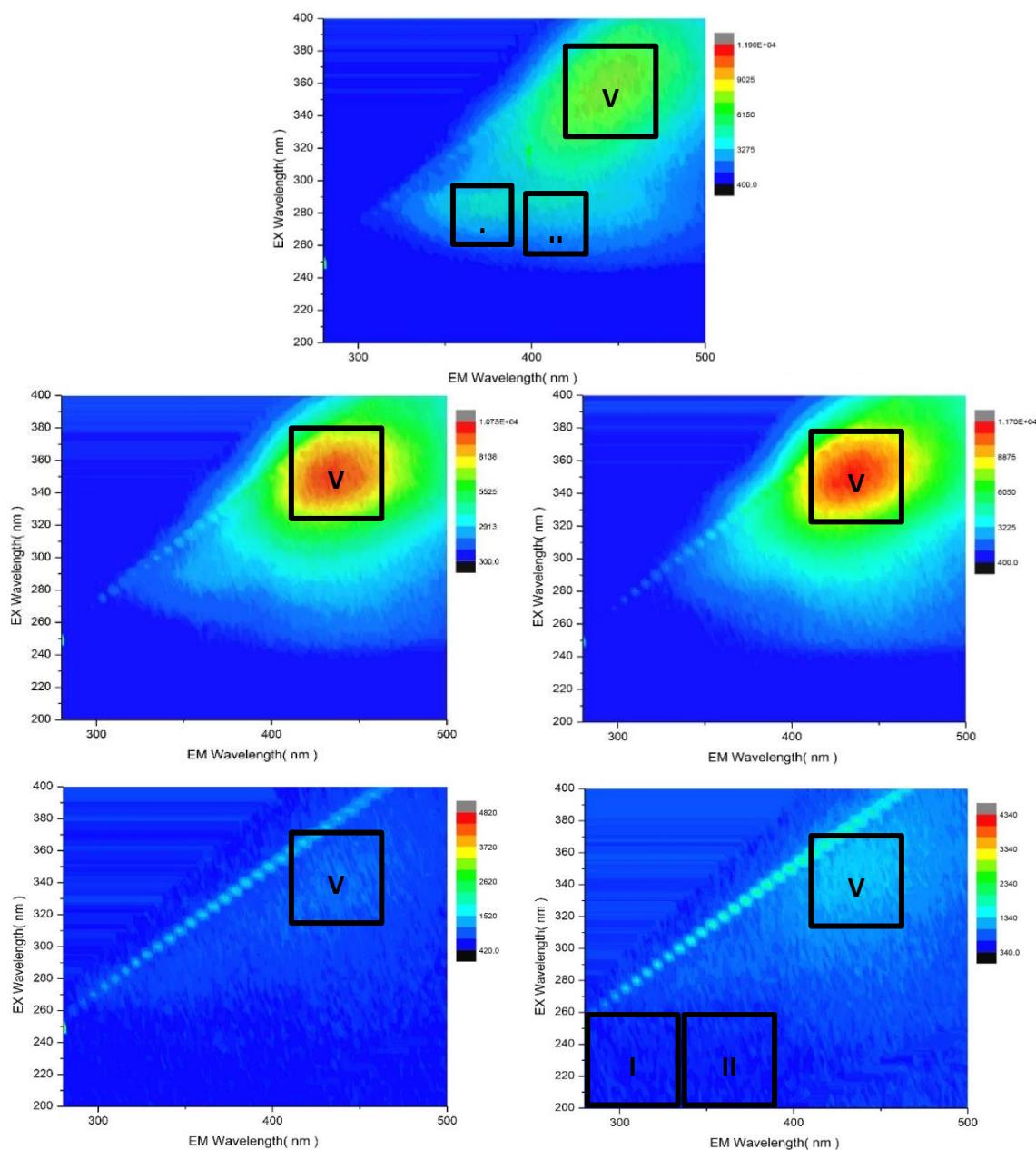


Figure 3-2 DOM characterization of (a) influent wastewater, (b) AnMBR and (c) AeMBR effluent, (d) AnMBR-ROP and (e) AeMBR-ROP

Zone I: tyrosine-like (APr); Zone II: tryptophan-like (APy); Zone III: fulvic acid-like (FA); Zone IV: soluble microbial products (SMP); Zone V: humic acid-like (HA).

As shown in Figure 3-3, the use of UF membranes within the MBRs achieved 50% removal of SMP, the rest of the organic substances presented a low removal

particularly for HA as the main removal mechanism is size exclusion. This finding agrees with Liu et al. (2020) that although UF has a certain retention for all categories of DOM, the removal efficiency is not high. RO filtration achieved a low removal of AP substances due to the low initial concentration. The RO step removed a further 50% of SMP and FA after the RO but a sharp reduction of HA was noted in both MBR permeates. The difference in removal performance is due to the dissimilar size and hydrophobicity of each type of DOM. AP is reported to have a low molecular weight (MW) and hydrophobicity (Rose, 2004); FA (Hiradate et al., 2006) and SMP (Banti et al., 2018) a higher MW compared to AP and neutral hydrophobic and hydrophilic moieties; and HA a high MW and hydrophilic moieties (Ma et al., 2015). Therefore, since a significant amount of HA was rejected by the RO membrane and a very low level of SMP was observed in the permeate produced by both MBRs, this has arguably led to a reduction of primary halogenated DBP precursors (Bond et al., 2011; Ma et al., 2016), which ultimately would lead to low DBP formation during chlorine or chloramine disinfection for reused water distribution. Since similar fluorescence intensity maps were generated, the AnMBR and AeMBR effluent may have a similar fouling behaviour in terms of DOM, while HA can be hypothesised as the major organic foulant during the RO filtration.

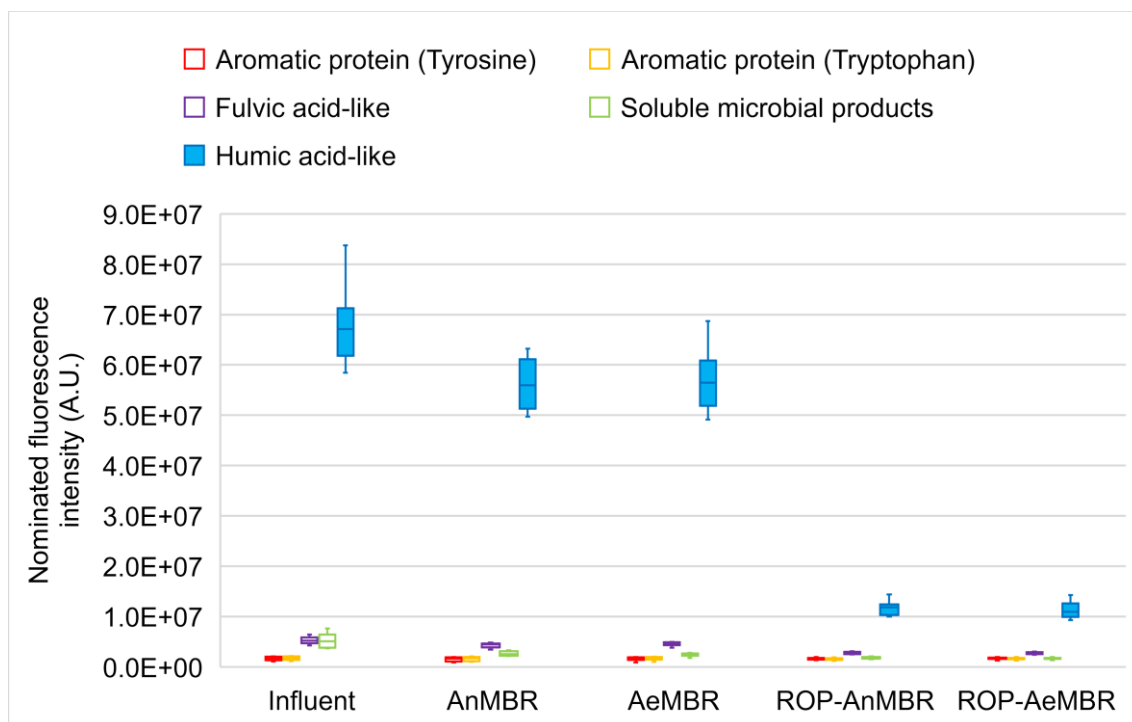


Figure 3-3 Distribution of characterized DOM for influent wastewater, AnMBR and AeMBR effluent, AnMBR-ROP and AeMBR-ROP

It is worth mentioning here the relative effectiveness of pre-RO and post-RO disinfection, particularly as pre-RO disinfection has been reported to be an inefficient tool to mitigate RO biofouling (Anis et al., 2019; Gare, 2002). Moreover, Al-Abri *et al.*, (2019) has reported that pre-RO chlorination may lead to higher overall operational expenditure. The ineffective elimination of generated active chlorine species can cause damage to the membrane polymer, which reduces the lifecycle of RO membranes. However, by controlling the chlorine to ammonia weight dosing ratio (Wahman, 2018) during the pre-RO chlorination, the ammonia concentration in AnMBR effluent can be accurately reduced, coupled with a weaker reactivity chloramine being generated compared to chlorine (Hua et al., 2015) in AeMBR effluent. This highlights a potential mitigation to the unsatisfactory ammonia removal in AnMBR-RO system while producing less DBPs (Farré et al., 2012) as well as keeping the risk of damage to the membrane surface material to a minimum (Cran et al., 2011). On the other hand, the ROP from both MBR effluents in this study exhibited low levels of DBP precursors along with unremoved nutrients and trace elements. Since incomplete removal of

trihalomethanes (THMs) and haloacetic acids (HAAs) but no THM or HAA formation for pre-RO and post-RO disinfected MBR effluent has been reported in various studies (Comerton et al., 2005; Kimura et al., 2003; Zazouli & Kalankesh, 2017), this may highlight the need to understand the impact of unremoved substances, specifically the unremoved nitrogen species, on pre-RO or post-RO disinfection processes such as NDMA formation when RO is combined with AnMBR or AeMBR for potable water reuse applications.

3.3.3 Impact of organic carbon and nitrogen species on RO fouling

Different organics removal performance and different nitrogen species present in the effluent were identified as a distinctive disparity between the performance of the AnMBR and AeMBR. As shown in Figure 3-4, the wide distribution of AnMBR effluent TOC concentrations indicate a relatively unstable performance of the biological stage in the AnMBR, while the AeMBR achieved more effective and more robust organic removal, as evidenced by a narrower distribution. Average membrane resistance values of 12.35 m^{-1} and 8.97 m^{-1} for the AnMBR and AeMBR respectively were recorded, demonstrating a greater fouling propensity of the AnMBR effluent. Noticeably, the average TOC concentration of the AnMBR effluent was 2-3 times higher but the recorded resistance was only about 1.5 time higher compared to AeMBR effluent, suggesting the fouling may not proportionally increase as a function of the TOC concentration. Furthermore, no strong correlation was observed between the membrane resistance and the feed TOC concentration for the AnMBR suggesting that the fouling observed is due to more complex interaction of different compounds. However, the distribution profile partially shows that a higher TOC concentration may lead to higher membrane fouling since the higher resistance was only observed in tandem with higher TOC. In contrast, there was no such correlation observed for the AeMBR operation.

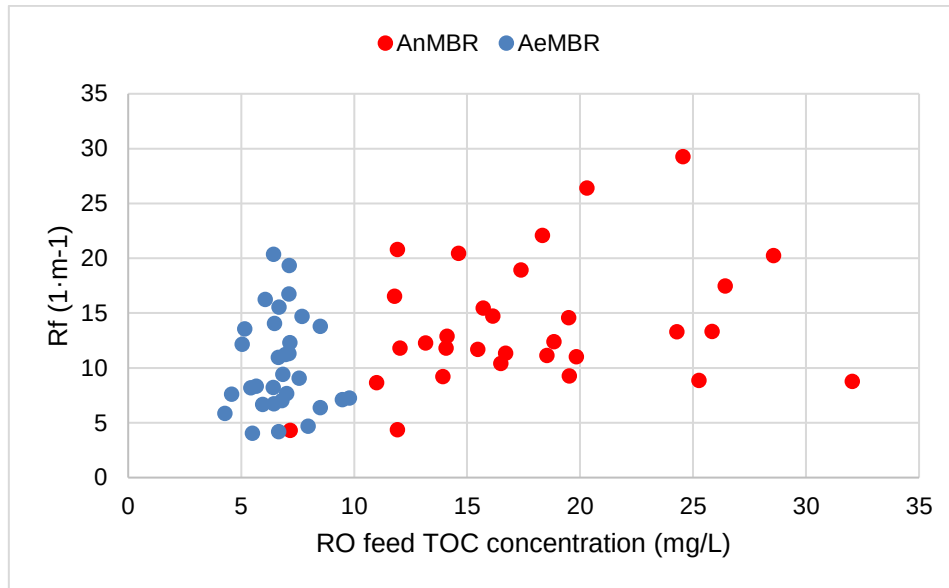


Figure 3-4 RO membrane resistance against feed TOC concentration

A slightly different relationship between the membrane resistance and RO feed characteristics was observed for the AnMBR (Figure 3-5.a) and AeMBR (Figure 3-5.b) effluents. For the AnMBR effluent, low membrane resistance was observed at low ammonia concentration, and higher resistance was observed with higher ammonia concentration. This could be explained by the different surface charge of ammonia and nitrate in the aquatic environment, where aquatic ammonium presents a single positive charge while nitrate is negative. The membrane used in this study had a negative surface charge, which resulted in electrostatic repulsion of anions (Alghoul et al., 2009) and attraction of cations. Therefore ammonium was attracted to maintain a charge balance (Hoang et al., 2010) while simultaneously depositing onto the membrane surface. Furthermore, as more permeate was produced, the pH in the RO cell kept increasing, which led to a constant increase in the ammonia fraction and lowering of the ammonium fraction simultaneously (Shin et al., 2021). This ultimately caused a decrease in overall ammonia removal performance as uncharged ammonia was more likely to go through the membrane which explains the poorer removal performance. Contrastingly, low membrane resistance was observed at high nitrate concentrations, which can be explained by the nitrate being repulsed by a negative membrane surface charge. Since aquatic nitrate is reported as

unreactive (Capocelli & Piemonte, 2021), this suggested nitrate may not have a significant impact on RO fouling.

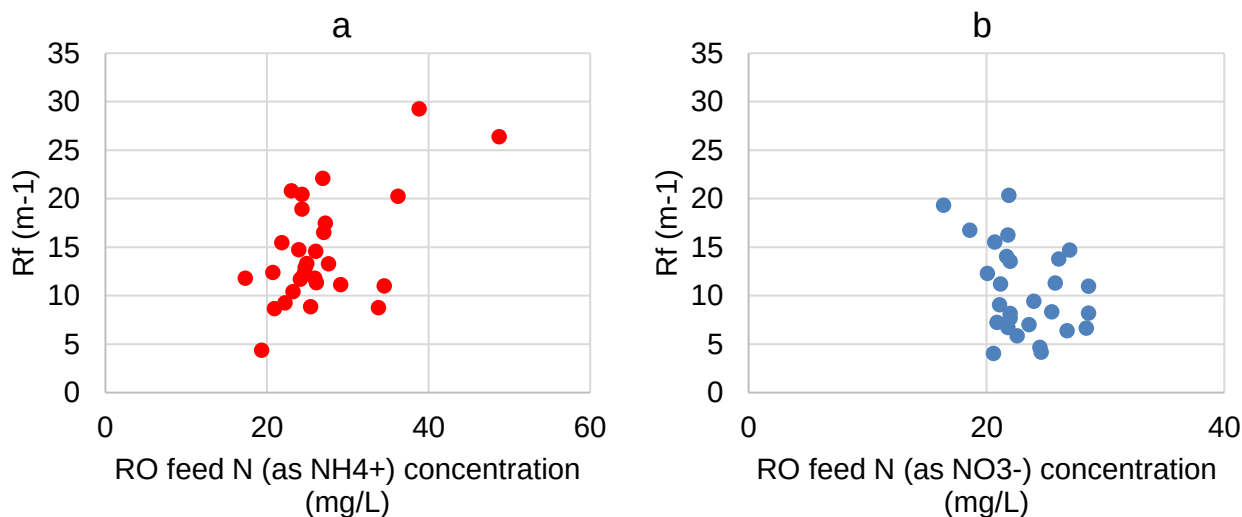


Figure 3-5 RO membrane resistance against feed nitrogen concentration

Overall, a relatively higher membrane resistance was measured during the filtration of the AnMBR effluent due to a higher total feed organic content when compared to the AeMBR. The presence of ammonium nitrogen in the AnMBR effluent may also contribute to the RO fouling, while the nitrate present in the AeMBR effluent may decrease the fouling. However, no strong correlation was found between membrane resistance and any specific compound.

3.3.4 RO foulant analysis

3.3.4.1 Foulant characterization with SEM-EDS

The back scatter electron EDS images for the virgin membrane displayed no bright scatter, which indicated, as expected, that there is no foulant deposited on the membrane surface. After producing 45 ml permeate, the tested membranes of the AnMBR-RO exhibited a large quantity of accumulated bright scatter compared to the AeMBR-RO membranes, indicating more foulant was deposited (Supporting information 2-1). Moreover, the random deposit spots observed on the back scatter images for both effluents suggested there was no connection between foulant deposit and membrane surface morphology.

The secondary electron mode in the SEM images generates a grey scale contrast which displays a transitional level of membrane surface topography. A clear cloth type structure could be seen in the virgin membrane image (Figure 3-6.a). In contrast, Figure 3-6.b and 3-6.c show comprehensive substance deposition on the used AnMBR-RO and AeMBR-RO membrane surfaces, with the membrane irregularities filled with colloidal material and covered by a multilayer poly-structures. This confirmed the foulant types were organic fouling, inorganic scaling, and colloidal fouling (Hu et al., 2014; Jiang et al., 2017; Shafi et al., 2017; Xie et al., 2016).

Both images display a colloidal like layer in between and on top of the original membrane structures. The colloidal layer on the AnMBR-RO membranes exhibited complete coverage over the membrane irregularities, while the original membrane surface features were still visible for AeMBR-RO membranes. The multilayer poly-structures observed on the AnMBR-RO membranes had a thicker fouling layer compared to the AeMBR-RO membranes, indicating again a significant level of inorganic substances deposited onto the membrane surface which consequently led to more inorganic fouling. This confirms that the AnMBR effluent contributed to more RO fouling than the AeMBR effluent. This may suggest the priority of foulant deposition during the filtration of MBR effluent is organic and colloidal fouling followed by multilayer scaling of inorganic substances, which confirmed inorganic fouling occurs when the dissolved substances reach their solubility limit in more concentrated solution (Jiang et al., 2017) at higher recovery rates. There was no organisms observed in all membrane samples for both MBR effluents, suggesting a low potential of biofouling during the filtration period, which meets the expectation from the results of the coliform tests.

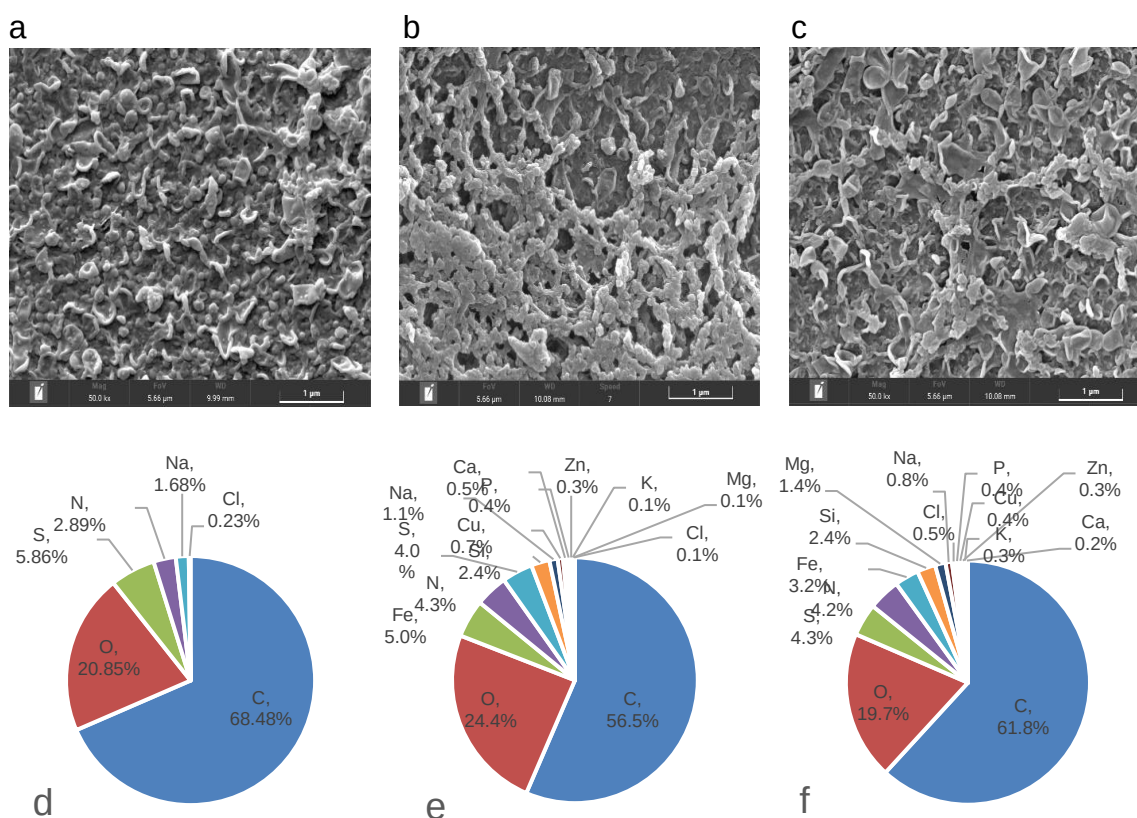


Figure 3-6 SEM image and EDS analysis of (a & d) virgin and tested membranes with the (b & e) AnMBR and (c & f) AeMBR

EDS analysis of the virgin membrane (Figure 3-6.d) agreed with the reported component characterization of a polyamide-urea-thin film RO membrane, where Carbon and Oxygen account for the most weight percentage (Liu et al., 2006). As expected, a variety of deposited substances were detected on both tested membranes, leading to a slight decrease in weight percentage for C, O, S, Na and Cl since more substances were deposited on the membrane surface (Figure 3-6.e and 3-6.f). However, the weight percentage of N for each membrane increased, indicating deposition of ammonia and nitrate from both the AnMBR and AeMBR effluents. Fe (5%), N (4.3%), Si (2.4%), Cu (0.7%), Ca (0.5%), P (0.4%), Zn (0.3%), K (0.1%) and Mg (0.1%) were identified foulants in the AnMBR effluent. In contrast, foulants deposited from the AeMBR effluent were N (4.2%), Fe (3.2%), Si (2.4%), Mg (1.4%), P (0.4%), Cu (0.4%), Zn (0.3%), K (0.2%) and Ca (0.2%). This agreed with the weight percentage of the foulants on an AeMBR-

RO membrane deposit reported by Tang et al. (2014, 2016), with Fe as the highest deposit content by weight. The SEM-EDS analysis also confirms that the AnMBR and AeMBR effluents deposited similar foulants on the RO membranes with the exception of nitrogen species. However, the co-presence of various foulants may have a further impact on RO fouling (Jiang et al., 2017). These observations highlight the need to better understand the impact of ammonia and nitrate on RO fouling mechanisms.

3.3.4.2 Foulant co-presence impact

A multivariate analysis was carried out with the EDS data (n=25) to understand the possible interactions between each deposited substance (Table 3-3), which may cause more severe fouling compared to the presence of any one foulant on its own (Kimura et al., 2016; Malki & Abbas, 2013; Shafi et al., 2017). Although literature sources have commonly reported Ca foulants as calcium sulphate, calcium carbonate, and calcium phosphate (Jiang et al., 2017; Ochando-Pulido et al., 2015), on the AnMBR-RO membranes, no correlation between Ca and either phosphate or sulphur was observed. Similarly, no association was found between the presence of N and other compounds. However, Ca was found to preferably deposit with N and P for the AeMBR-RO. This may suggest the presence of ammonia and nitrate had a different impact on the scaling of Ca. Likewise, Si has been reported to be deposited in association with phosphate salts (Malki & Abbas, 2013) which subsequently causes severe fouling with organic matter (Kimura et al., 2016). Both MBR effluents in this study showed a negative correlation between Si with P and C. However, Si was found to have a significant correlation with O in both effluents, which may suggest silicon dioxide (SiO₂) was formed. Furthermore, Cu was found to not deposit with ammonia on the AnMBR-RO membranes but displayed a high probability to deposit with nitrate on the AeMBR-RO membranes, while phosphate was respectively incongruous to deposit with Fe, Cu and Zn for AnMBR-RO but showed a strong co-deposition correlation on the AeMBR-RO membranes. The positive co-deposition correlation between Mg and O supports previous work which has suggested that the most common Mg foulant is magnesium hydroxide (Patel &

Milligan, 2008). This meets the expectation that the different behaviour observed were a direct implication of the different effluent matrices from the two systems.

Table 3-3 R values of multivariate correlation matrix of identified foulants for AnMBR-RO (top) and AeMBR-RO (bottom)

AnMBR-RO													
C	-0.13	-0.97	-0.59	-0.26	-0.69	0.03	0.84	0.72	-0.15	-0.19	-0.41	0.17	0.13
-0.32	N	-0.05	0.03	-0.21	-0.11	0.48	-0.22	-0.44	-0.10	0.00	-0.10	0.08	0.28
-0.93	0.03	O	0.56	0.32	0.73	-0.12	-0.87	-0.66	0.21	0.23	0.39	-0.34	-0.20
0.08	-0.12	-0.24	Na	-0.11	0.77	-0.16	-0.44	-0.39	-0.14	-0.11	-0.19	-0.12	-0.11
-0.45	-0.11	0.51	-0.02	Mg	0.19	-0.10	-0.34	-0.25	0.67	0.50	0.01	-0.08	-0.10
-0.56	-0.09	0.70	-0.10	0.47	Si	-0.24	-0.60	-0.52	0.15	0.02	-0.19	-0.18	-0.15
-0.47	0.13	0.34	-0.07	-0.11	-0.12	P	-0.03	-0.34	-0.06	-0.16	0.09	0.03	0.26
0.86	-0.33	-0.75	-0.21	-0.30	-0.51	-0.36	S	0.67	-0.39	-0.24	-0.32	0.49	0.15
0.10	-0.13	-0.26	1.00	-0.02	-0.11	-0.08	-0.18	Cl	-0.16	-0.33	-0.01	0.05	0.09
-0.09	-0.02	0.14	0.04	-0.04	0.12	-0.13	-0.18	0.02	K	0.39	-0.06	-0.10	-0.13
-0.02	0.46	-0.17	0.09	-0.16	-0.14	0.28	-0.14	0.10	-0.06	Ca	-0.18	-0.11	-0.03
-0.63	-0.01	0.57	-0.09	0.03	-0.07	0.80	-0.44	-0.11	-0.05	-0.09	Fe	-0.10	-0.10
-0.39	0.50	0.17	-0.20	-0.21	-0.22	0.72	-0.32	-0.20	-0.17	0.22	0.59	Cu	0.00
-0.46	0.24	0.30	-0.08	-0.12	-0.12	0.96	-0.39	-0.09	-0.10	0.29	0.72	0.82	Zn
AeMBR-RO													

3.3.4.3 Mass balance of deposited foulants on RO membrane surface (ROS)

In Figure 3-7, both columns display a similar structure for foulant mass composition, with Ca as the dominant foulant followed by TOC, nitrogen and phosphate. The rest of the elements only account for 4.56% and 5.78% for the AnMBR-ROS and AeMBR-ROS respectively. This agrees with the element types characterized but differ from the mass percentages obtained in the EDS analysis, suggesting the scanned areas may not fully represent the distribution of foulants on the membrane surface.

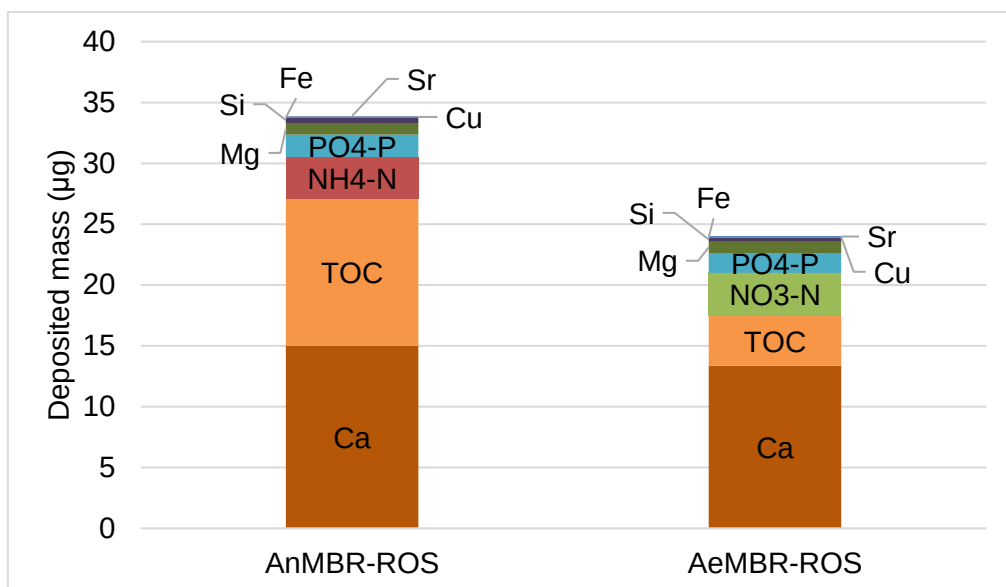


Figure 3-7 Deposited foulants mass of AnMBR-ROS and AeMBR-ROS

Deposition on the RO membrane surface was higher for the AnMBR, with a total mass of 33.895 µg compared with 24.010 µg for the AeMBR. TOC (79.9%) and Ca (16.5%) were the major components responsible for the difference with no significant difference found for PO₄-P, Mg, Si, Ca, Cu, Fe, and Sr. The difference for TOC can be explained by the initial concentration load onto the RO. However, Ca, along with other substances, constituted a similar load from both MBR effluents, resulting in a slightly higher mass deposited in the AnMBR-ROS compared to the AeMBR-ROS. This may be explained by the fact that the AnMBR-RO had more fouling and that the developed cake layer enhanced the rejection and deposition of other substances (Jiang et al., 2017).

Table 3-4 Foulants deposition rate from the AnMBR and AeMBR effluents

R _D (%)	TOC	Ca	NH ₃ -N	NO ₃ -N	PO ₄ -P	Si	Cu	Sr	Fe	Mg
AnMBR-RO	67.4	30.8	13.8		5.1	5.1	0.08	0.03	0.03	0.02
AeMBR-RO	60.7	27.1		0.02	5.3	5.9	0.06	0.03	0.01	0.02

A similar deposition rate excluding ammonia and nitrate was found for both the AnMBR-RO and AeMBR-RO (Table 3-4). TOC exhibited the highest rate of 67.4% and 60.7% for the AnMBR-RO and AeMBR-RO, respectively. This highlights organic fouling is the major fouling mechanism for RO when treating both MBR effluents, a premise supported by the low concentrations of COD and

TOC in the RO permeate, as well as a clean fluorescence intensity map. Furthermore, Ca is the major compound for inorganic fouling, where a deposition rate of 30.8% and 27.1% was found for AnMBR-RO and AeMBR-RO, respectively. Phosphate and Si respectively exhibited a 5% deposition rate in both MBR effluents, which are identified as the other major inorganic foulants during the RO filtration. Ammonia exhibited a 13.8% deposition rate for the AnMBR-RO. This indicates that ammonia is another major inorganic foulant within the AnMBR effluent but contributes less inorganic fouling than Ca. In contrast, nitrate exhibited a much lower deposition rate of 0.02% for the AeMBR-RO, which confirmed that the presence of nitrate in the AeMBR effluent has a negligible impact on RO fouling. The rest of the foulants demonstrated a much lower deposit rate. This suggested that pre-removal of Ca from the MBR effluents can be a beneficial approach to mitigate RO fouling, since Ca anti-scalants are reported as not effective against the formation and the deposition of calcium phosphate (Mangal et al., 2021, 2022).

With regard to the circular economy concept, AnMBR-RO can offer a high quality permeate while allowing a potential recovery as fertilizer the rejected nutrients in the RO concentrate, as well as an energy recovery potential to reduce the operational cost (Huang et al., 2020; Mallick et al., 2021; McCarty et al., 2011; Sendrowski & Boyer, 2013; Song et al., 2018; Stazi & Tomei, 2018; Xie et al., 2016). Treating municipal wastewater with a low nutrient concentration may be more suitable for AnMBR-RO in terms of the insufficient ammonia removal in the RO permeate as well as mitigating the RO fouling. Moreover, enhancing the organic removal performance by operating the AnMBR under optimised conditions such as mesophilic temperature, with more precise control, or possibly adding a pre-RO treatment to polish the effluent by removing organics, ammonia and phosphorus could be beneficial to mitigate the RO fouling. However, a more detailed investigation on the trade-offs between the cost of adding an extra barrier to protect the RO or optimising the AnMBR operation and the reduction in energy recovery from the AnMBR when implementing these steps is needed. In contrast, AeMBR-RO has no noticeable limitation when combined with the RO as to the final effluent quality but offers no energy recovery potential. However, adding an

anaerobic and anoxic zones to the AeMBR can remove the phosphorus and nitrogen while maintaining a high organic removal, hence reducing the RO fouling which could decrease the overall operational expenditure.

3.4 Conclusion

In this study, a direct performance comparison has been undertaken between the combination of reverse osmosis membranes with AnMBR and AeMBR processes for the treatment of real municipal wastewater for reuse. The analysis focused on a number of previously poorly reported aspects: water quality and trace elements analysis in the context of potable water reuse; the fate of DBP precursors; and characterization of RO membrane foulants - specifically the impact of ammonia and nitrate on RO fouling mechanisms. Major conclusions made were:

- The combination of RO and AeMBR was able to produce a high quality permeate for indirect potable water reuse such as groundwater or drinking water reservoir recharge, whilst the presence of residual ammonia in the permeate is a current weakness of the AnMBR-RO combination to satisfy the standard threshold. The application of an RO step efficiently removed primary halogenated DBP precursors, resulting in a low DBP formation potential for reused water distribution.
- The AnMBR-RO system experienced more membrane fouling compared to the AeMBR-RO system as evidenced by higher membrane resistance and more deposited substances observed in SEM images. The ammonium nitrogen in the AnMBR effluent may have played a role in RO fouling, while the nitrate in the AeMBR effluent may decrease the fouling due to the effect of surface charge.
- The fouling sequence during the filtration of MBR effluent is organic and colloidal fouling followed then by inorganic scaling. TOC and Ca were identified by weight as the most deposited major foulants in both effluents, and TOC provided a higher deposition rate in both effluents compared to Ca. A higher deposition rate of TOC and Ca was observed during the filtration of the AnMBR effluent in comparison to the AeMBR.

- Ca had a negative correlation with phosphate deposits and none with ammonia where the AnMBR effluent was used but was more likely to deposit with nitrate and phosphate where the AeMBR effluent was used. Ammonia had a 13.8% deposition ratio during the filtration through the AnMBR-RO, significantly higher than the value of 0.02% for nitrate when treating the AeMBR effluent.
- Selecting the combination of AnMBR-RO or AeMBR-RO should be based on the influent water matrix as well as the benefit from the recovered energy by AnMBR while also considering additional expenditure for adding treatment steps to protect the RO and control the RO permeate quality.

The RO filtration in this study was carried out with a dead-end filtration module with only one type of flat sheet membrane. Future studies carried out with cross flow filtration may provide further insights on fouling behaviour. Furthermore, continuous efforts should be made to understand the impact of foulants (specifically nitrogen species and trace elements) co-presence on RO fouling. Identification of functional groups and analytical measurement using Fourier transform infrared spectroscopy (FTIR) would be of benefit.

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3.6 Supporting information 2

3.6.1 Supporting information 2-1

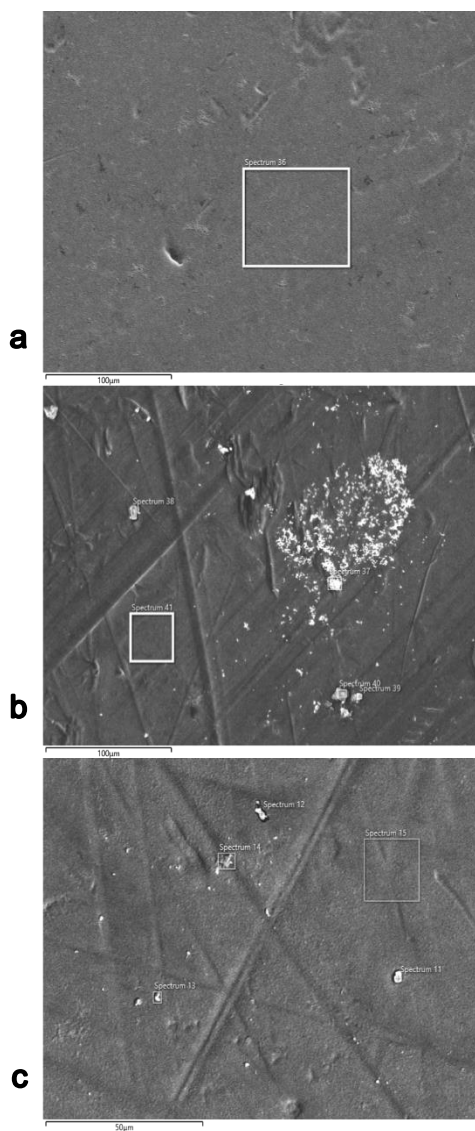


Figure 3-8 Back scatter electron EDS image of (a) virgin and tested RO membranes with the (b) AnMBR and (c) AeMBR

4 Understanding chlorine based disinfection processes and disinfection by-product formation in Anaerobic and Aerobic MBR effluents for reuse applications

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Abstract

Both anaerobic (An) and aerobic (Ae) membrane bioreactors (MBRs) are capable of purifying municipal wastewater to a quality sufficient for agriculture irrigation following disinfection. However, the disinfection performance and disinfection by-product (DBP) formation triggered by the different effluent matrices from of these two types of MBR have yet to be fully investigated. In this study, DBP (4 THMs and 9 HAAs) formation potential (FP) was investigated using chlorine based disinfection with various dilutions and chlorine doses. The disinfection of the AnMBR effluent experienced a lower DBP formation potential compared to the AeMBR effluent and surface water typically used as irrigation or drinking water source, while offering a controllable disinfection process with required residual chlorine and ammonia concentration for irrigation purposes. A fluorescence 3D excitation-emission matrix analysis suggests the organic matter DBP precursors present in the two MBR effluents were dissimilar to natural organic matter (NOM) in surface water sources. The dilution proportionally reduces the compounds concentration in the MBR effluents hence a reduced DBPFP, highlighting the potential in irrigation application of blending with natural water source. However, diluting the sample is not recommended when studying water with high organic content since the reduction impact may lead to an inaccurate detection in the low chlorine dosing scenario. Multivariate analysis between the DBPFP and each measured parameter suggests the chlorine dosage and the ammonia and DOC concentrations are the key indicators for DBPs formation in the AnMBR effluent, as opposed to only chlorine dosage and DOC for the AeMBR effluent. A mixed

process of chloramination and chlorination takes place in the AnMBR effluent due to the presence of ammonia, whilst a monophonic chlorination process happened in the AeMBR effluent. With the AnMBR effluent, three critical dosing points were identified to achieve a 1 mg/L residual chlorine concentration requirement for agriculture irrigation reuse. The corresponding chlorine dosing strategy can help maintain the targeted ammonia and chlorine concentrations for irrigation purposes or provide pre-treatment of the AnMBR effluent for biofouling control before reverse osmosis filtration in potable reuse applications.

Keywords:

Anaerobic; Aerobic; Chlorine disinfection; Disinfection by-products (DBPs); Chloramination; Chlorination; Membrane bioreactor

4.1 Introduction

Significant increases in water demand are putting a strain on water resources globally. The effect is particularly being felt in the agricultural sector which accounts for 70% of anthropogenic water consumption (Shareefdeen et al., 2015). Water reuse has been demonstrated to be a promising alternative for enhancing water availability for agricultural use (Lahlou et al., 2022) with recent studies demonstrating its comparative advantages over groundwater (Leonel & Tonetti, 2021; Tampo et al., 2022). However, the application of water reuse schemes is still limited when compared to their potential due to the lack of progressive policy, clear understanding of technical and economic feasibility, and risks for human health and the environment while reusing the processed wastewater (Rebelo et al., 2018; Vivaldi et al., 2022). In particular, concerns over microbiological contamination have attracted much attention (Delanka-Pedige et al., 2020), as reflected in their widespread inclusion in relevant water quality standards. For instance, Australia limits the *Escherichia coli* forms (*E. coli*) to below 1 CFU/100 mL when the reused water is for food crops to be consumed raw, while a non-detectable faecal and *E. coli* level is set in the standards of the USA and EU, respectively. These requirements encourage the use of disinfection processes before the final effluent can be reused. Residual chlorine concentration is normally used as the quality parameter for distributed recycled water to prevent microbial and pathogen regrowth, with 1 mg/L being a commonly applied standard for both potable and non-potable applications (CNEPA, 2021; EUPC, 2020; ISO, 2018; USEPA, 2019).

Membrane bioreactors (MBRs) can remove most pathogens and organic pollutants, producing high quality effluent sufficient for various sub-potable water reuse applications (Winward et al., 2008). Among various MBR configurations, the Anaerobic Membrane Bioreactor (AnMBR) has been extensively studied in recent years for municipal wastewater treatment (Robles et al., 2018; Shin & Bae, 2018). Indeed, while offering comparable organic pollutant removal to the Aerobic Membrane Bioreactor (AeMBR) configuration, the AnMBR has the advantage of reduced energy costs, low solids production, and inclusion of nutrients (Nitrogen and Phosphorus) in the effluent which enhances its use for fertigation purposes

(Foglia et al., 2019). This leads to improved soil fertility, crop yield, and plant growth as well as reduced usage of mineral fertilizers which ultimately lead to lower crop production costs (Lyu et al., 2016). However, adequate disinfection treatment is still compulsory to ensure there is no pathogenic concern before reusing the MBR effluent. In addition, the AnMBR effluent can be further polished by reverse osmosis membrane for potential potable application as seen for the AeMBR effluent (Cashman et al., 2018; Gu et al., 2019). The application of pre-disinfection before RO membranes is very common as a fouling control strategy (Anis et al., 2019; Jiang et al., 2017). Hence understanding the disinfection and DBPFP in MBR effluents is of benefit to unlock other options for the reuse of AnMBR effluent.

Commonly used disinfection processes include ultraviolet radiation, ozonation and various forms of chlorine dosing (Schroeder, 2003). Chlorination and chloramination are the most widely applied chlorine based disinfectants due to low operational cost and high effectiveness against pathogens (Chaukura et al., 2020). However, it has been recognised that trihalomethanes (THMs) and haloacetic acids (HAAs) – two key groups of chlorinated carbonaceous disinfection by-products (DPBs) – are formed following chlorine disinfection, posing a risk to humans and animals health (Bond et al., 2010; Marais et al., 2019). Available literature mostly focuses on drinking water supply and natural water treatment, identification and mitigation of DBP precursors, with four trihalomethanes (THMs): chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), bromoform (TBM); and nine haloacetic acids (HAAs): monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), dichloroacetic acid (DCAA), bromochloroacetic acid (BCAA), trichloroacetic acid (TCAA), dibromoacetic acid (DBAA), bromodichloroacetic acid (BDCAA), dibromochloroacetic acid (DBCAA), tribromoacetic acid (TBAA) commonly characterized to evaluate the safeness of the water in terms of disinfection by-product formation potential (DBPFP) (Bond et al., 2009; Chaukura et al., 2020; Golea et al., 2020; Goslan et al., 2017; Sfynia et al., 2020). Due to the fact that natural water sources have a much lower DOC concentration (1-2 mg/L) compared to the MBR effluents, and the chlorine weight dosing ratio varied

between each study (Cl:DOC of 1-20 : 1) (Delpla & Rodriguez, 2016; Pifer et al., 2013; Roccaro et al., 2015; Roccaro & Vagliasindi, 2009; Teksoy et al., 2008; Yang et al., 2015), this substantially restricts potential comparison between the wastewater effluents and natural water sources such as surface water. Therefore, conducting DBPFP tests with diluted and undiluted MBR effluents can reveal the efficacy of dilution/blending strategies aimed at reducing the risk of applying reused water to agricultural crops.

Literature focusing on DBP formation in the effluents from MBRs treating municipal wastewater is very limited. One influential study reported the fractionation and characterization of halogenated carbonaceous DBPs (C-DBPs), nitrogenous DBPs (N-DBPs) and N-nitrosodimethylamine (NDMA) for the biomass associated products (BAP) and utilization associated products (UAP) from the soluble organic matters in an AeMBR effluent using a lab scale single oxic stage MBR treating synthetic municipal wastewater (Ma et al., 2015). BAP was shown to have a higher THMs and HAAs formation reactivity due to higher fraction of large molecular weight materials and humic substances, whereas the UAP contained more aromatic protein-like and secondary amines with a low molecular weight (<500 Da) leading to a much higher NDMA formation. With the same setup and analytical method, Ma et al. (2016) quantified the DBPFP for AeMBR effluent disinfected by chlorination or chloramination. Chloramination resulted in a lower halogenated DBPFP compared to chlorination but a much higher NDMA DBPFP. Proteins and humic substances were identified as the primary DBP precursors for the halogenated compounds. Results from other studies using an Anaerobic-Anoxic-Oxic MBR treating actual municipal wastewater have shown that soluble microbial products (SMPs) are the major precursors for halogenated DBPs (Zhang et al., 2018). All three of these studies also identified a significant correlation between THMs and HANs yield using Fluorescence Regional Integration (FRI) data (Ma et al., 2015, 2016; Zhang et al., 2018). However, no work has yet been conducted using an AnMBR effluent to understand the influence of anaerobic effluent matrix on the performance of chlorine based disinfection process or the disinfection by-product formation.

This study aims to understand the processes and DBP formation potential of chlorine based disinfection using both an AnMBR and an AeMBR treating the same municipal wastewater. Various chlorine dosing strategies were performed with diluted samples for direct comparison to existing literature mostly focusing on drinking water production and undiluted effluent to establish the actual behaviour in practice.

4.2 Materials and methods

4.2.1 Pilot scale AnMBR and AeMBR

AnMBR and AeMBR pilot plants (Figure 3-1) were installed in Cranfield University's National Research Facility for Water and Wastewater Treatment and operated stably under ambient temperature for 55 weeks. The systems were fed with settled municipal wastewater from the Cranfield University sewage works after settling in a primary settling tank. The AnMBR was configured as a cylindrical upflow sludge blanket bioreactor (UASB - 45L) and a side stream membrane tank (30L). The AeMBR was configured with a level control tank (10L), aerobic tank (40L) and membrane tank (18L). For both systems, a PVDF hollow fibre ultra-filtration (UF) membrane with 0.93 m² surface area and 0.04 µm nominal pore size (GE ZW-10, USA) was placed in the membrane tank and operating with a flux of 10 LMH. Due to the conservative approach in the operation of the membranes, during the 55 weeks of operation of the two MBRs, no membrane cleaning was required. More details about the MBRs can be found in Chapter 3.

4.2.2 Disinfection by-products formation potential test

The DBPFP test performed in this study was adapted from Goslan et al. (2009, 2017) with diluted and undiluted effluent samples from the AnMBR and AeMBR. For the diluted samples, prior to chlorination, DOC levels were diluted to within 1 - 2 mg/L with ultrapure water (ELGA Purelab Ultra, UK) to simulate similar condition to most existing studies on natural water sources, then the pH was buffered at 7 with a phosphate buffer. For chlorination, various Cl : DOC / Cl : NH₃-N weight dosing ratios were performed (Table 4-1) with a chlorine stock

solution prepared from concentrated sodium hypochlorite (Fisher scientific, UK). Chlorinated samples were stored in the dark at 20 °C for seven days. Residual chlorine concentrations (as free chlorine) were measured immediately at the end of this period according to the DPD method (APHA, 1999). THMs (TCM, BDCM, DBCM, TBM) and HAAs (MCAA, MBAA, DCAA, BCAA, TCAA, DBAA, BDCAA, DBCAA, TBAA) were extracted according to the USEPA Method 551.1 and HAAs by the USEPA Method 552.3. Generated DBPs were measured by gas chromatography with a micro electron capture detector (Agilent 6890 GC- μ ECD, USA). The limit of detection (LOD) was 0.5 μ g/L and results below this value are reported as below the LOD. All samples were obtained in duplicate.

Table 4-1 Sampling strategy and chlorination scheme of this study

Time period	Volume	Dilution	Cl:DOC	Cl:NH ₃ -N (AnMBR)	Cl:NO ₃ -N (AeMBR)
7 days: 2 each day	14 AnMBR 14 AeMBR	yes	1-11 : 1	1-11 : 1	1-11 : 1
14 days: 1 each day	14 AnMBR 14 AeMBR	yes	5 : 1	-	-
5 days: 2 each day	10 AnMBR 10 AeMBR	yes	-	4-10 : 1	1.1-1.6 : 1
1 day: 23 samples	23 AnMBR	no	1-17 : 1	1-11 : 1	-
1 day: 11 samples	11 AeMBR	no	1-5 : 1	-	0.3-1.6 : 1

4.2.3 Analytical measurements

Chemical oxygen demand (COD), ammoniacal nitrogen (NH₃-N), nitrite (NO₂-N), nitrate (NO₃-N), phosphate (PO₄-P), and total phosphorus (TP) were analysed with cell tests and a photo spectrometer (Merck Spectroquant, Germany). Dissolved organic carbon (DOC) and total nitrogen (TN) were measured by a Shimadzu TOC analyser (Shimadzu 3201, Japan). Trace level elements were analysed with an inductively coupled plasma mass spectrometer (ICP-MS) (PerkinElmer NexION 350d, USA). Characterization of the MBRs influent was measured after filtering with a 0.45 μ m syringe membrane disc filter (Merck Millipore, Germany). Specific ultraviolet absorbance (SUVA) was measured by a UV/Vis spectrophotometer (Jenway 6715, UK) with a high performance quartz

glass cell (Hellma Analytics 100-40-40, Germany) at the UV wavelength of 254 nm. Dissolved organic composition was evaluated by a fluorescence spectrometer (HORIBA FLuoroMax+, Japan) with 3D excitation–emission matrices (EEM). Scan settings were 200–400 nm excitation and 280–500 nm emission wavelength, 1 nm entrance slit and an integration time of 0.1 seconds. The 1st and 2nd Rayleigh scatter was masked with 5 nm slit width by FluorEssence software (HORIBA, Japan). The fluorescence intensity maps were masked together by light intensity order ($n=15$). Each map uses a blue-green-yellow-red colour metric bar to quantify the detected peak intensities from low to high. Results from the fluorescence spectrometer were plotted as a 2D intensity map and the integrated composition analysis was extracted with a 5 region dissolved organic matter (DOM) categories method (aromatic protein-like (AP): tyrosine-like (APr) excitation/emission (E_x/E_m) = (200–250) nm/(280–330) nm; tryptophan-like (APy) E_x/E_m = (200–250) nm/(330–380) nm; fulvic acid-like (FA) E_x/E_m = (200–260) nm/(380–5000) nm; soluble microbial products (SMP) E_x/E_m = (250–280) nm/(310–380) nm; humic acid-like (HA) E_x/E_m = (280–380) nm/(380–500) nm) (Chen et al., 2003; Pan et al., 2021) using an R scrip modified from Lapworth and Kinniburgh (2009). Raw numerical data was input to JMP 16 pro to obtain multivariate correlation analysis and statistical analysis.

4.3 Result and discussion

4.3.1 AnMBR and AeMBR effluent

Due to the use of UF membranes in both MBRs, no TSS was found in either effluent. There was also no nitrite, organic nitrogen, or organic phosphorus present. Hence the fractions for TN and TP were ammonia (AnMBR) / nitrate (AeMBR) and phosphate, respectively (Table 4-2). The fraction of ammonia was mostly ammonium ions in the AnMBR effluent, explained by the fact that the pH was 7.2 ± 0.2 (Shin et al., 2021). The results for COD and DOC indicate that the AeMBR had a better performance in terms of organic removal compared to the AnMBR. This suggests a higher DBP precursor level in the AnMBR effluent which will lead to higher DBPFP (Ma et al., 2016). There was little difference found in the AnMBR (5.4 ± 1.2 mg/L) and AeMBR (5.6 ± 1.3 mg/L) effluent phosphate

concentrations, and phosphate has in any case been reported to have no obvious impact on DBPFP (Banihashemi et al., 2017). However, different nitrogen species present in the AnMBR (ammonia) and AeMBR (nitrate) effluent could cause different DBPFP results due to the fact that ammonia can react with chlorine to form monochloramine (NH_2Cl), dichloramine (NHCl_2), and trichloramine (NCl_3), depending on the weight ratio between chlorine and ammonia (Wahman, 2018). A previous study on natural water sources reported chlorination forming more THMs and HAAs than chloramination (Hua & Reckhow, 2007), while another study argues that where chloramination is achieved by adding ammonia first and then chlorine, less DBPs are generated than by adding chlorine followed by ammonia (Huang et al., 2017). Therefore, the ammonia present in the AnMBR effluent may have an advantage of low DBP formation over AeMBR effluent, while the dissimilar matrix of AnMBR and AeMBR effluents may have different responses when being chlorinated.

Table 4-2 Characterization of AnMBR and AeMBR effluents

	AnMBR		AeMBR	
mg/L	Mean	Std	Mean	Std
pH	7.22	±0.18	7.29	±0.28
TSS	ND		ND	
COD	47	±24	9	±9
DOC	21.12	±6.87	6.86	±1.17
NH ₃ -N	31.03	±6.40	ND	
NO ₃ -N	ND		23.51	±3.99
PO ₄ -P	5.39	±1.15	5.55	±1.29
	µg/L			
B	73.00	±9.85	64.39	±11.06
Na	50780.95	±11015.06	50647.62	±9502.61
Mg	5373.33	±329.14	5472.38	±485.74
Al	ND		ND	
Si	7028.57	±974.55	6840.00	±309.71
K	13051.43	±2610.50	12712.38	±1522.31
Ca	48657.14	±8576.75	49190.48	±9831.27
Mn	2.08	±5.90	ND	
Fe	ND		ND	

Cu	59.10	±11.41	61.80	±19.62
Zn	11.45	±13.81	16.04	±11.36
Br	ND		ND	
Sr	198.65	±44.63	200.12	±51.74
Cd	ND		ND	
Ba	37.90	±2.58	36.26	±4.40
Pb	ND		ND	

Note: ND, Non detectable.*

There were non-detectable levels of Al, Br, Cd and Pb in both effluents. The untraceable levels of Br suggest that a low proportion of brominated DBP species should be expected (Carra et al., 2021). Significant levels of Na, Mg, Si, K, and Ca were found, whereas the rest of the elements detected were at trace levels. The presence of different elements may lead to a different DBPFP. Ye et al. (2009) reported a positive relationship between THM formation and Na whilst the presence of Mn and Ca decreased THM formation. They also reported that the presence of K promoted the formation of THMs and HAAs but that Fe had the opposite effect. The effect of B, Si and Sr on THM formation is yet to be studied.

4.3.2 Fluorescence results

The influent municipal wastewater intensity map displays a different peak characteristic compared to that for natural water sources reported in the literature. The major content for settled municipal wastewater was found to be HA followed by SMP and FA (Figure 4-1.a) but for the natural water source a high content of FA and AP substances was observed (Go et al., 2021).

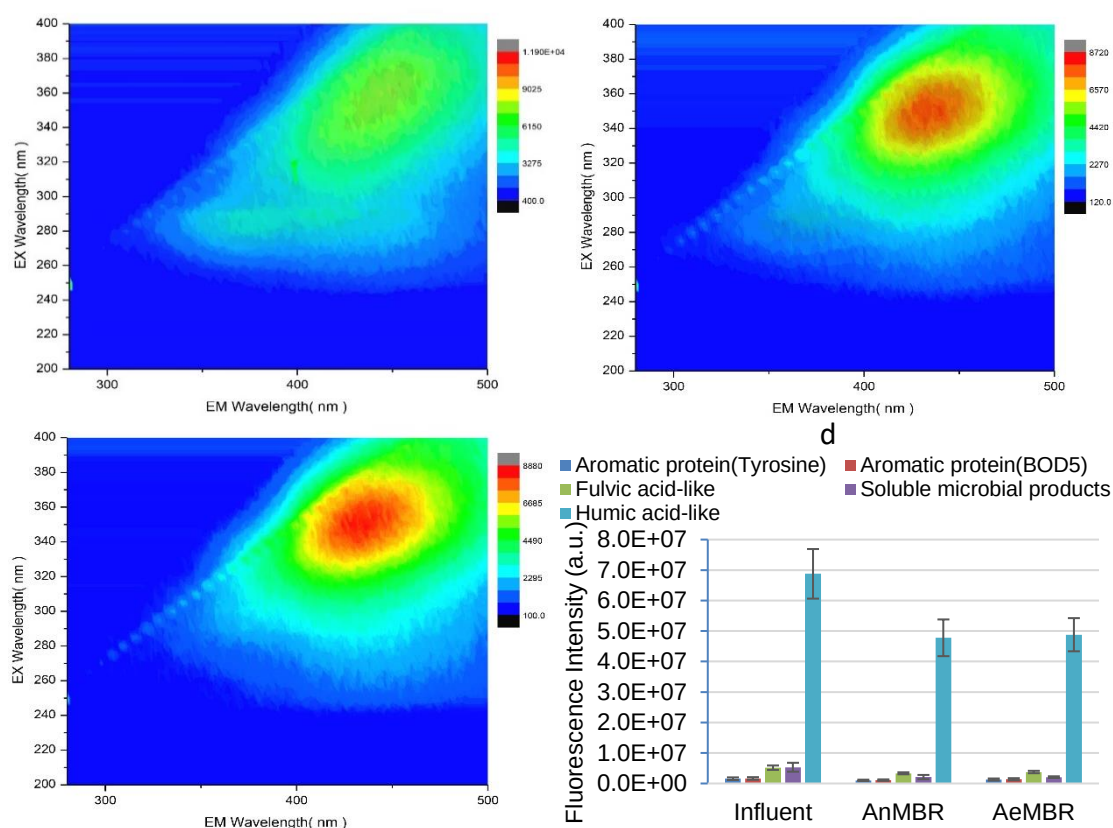


Figure 4-1 DOM characterization of influent wastewater (a), AnMBR (b) and AeMBR (c) effluent and the Fluorescence Regional Integration (FRI) analysis result (d)

Similar peak distributions were observed for the AnMBR and AeMBR effluents (Figure 4-1.b and 4-1.c); the highest peak in red indicating a high amount of HA. The peaks for AP_r, AP_y, FA and SMP were lighter than the influent in both maps, indicating that the digestion of biological stage as well as the size exclusion of the UF membrane can remove a certain amount of DOM which agreed with Zhang et al. (2018) who reported that an AeMBR coupled with an UF can remove 56.8% of C-DBP and 78.1% of N-DBP precursors. However, the major component in both MBR effluents was still humic acid-like substances (Figure 4-1.d). This finding agrees with the DOM characterization results reported by Ma et al. (2016) that different primary precursors for halogenated carbonaceous DBPs were observed in MBR effluent compared to natural water source.

4.3.3 Impact of dilution on DBPFP

Understanding the impact caused by dilution can allow a direct comparison of the DBPFP between the wastewater effluents conducted in this study with the results obtained with surface water in the documented literature for natural water sources. Simultaneously this can provide the evidence for potential blending strategies to reduce the risk of salt and ionic strength (European Commission, 2017; Zaouri et al., 2021) when applying disinfected MBR effluents to agricultural crops.

4.3.3.1 AnMBR effluent

A dilution factor of 15 was applied to adjust the DOC concentrations of AnMBR effluent samples within the target range of 1 - 2 mg/L, whereas the dosing weight ratio applied was Cl:NH₃-N from 1:1 to 11:1 (corresponding to 1.34:1 to 15.4:1 for Cl:DOC). Two selected results are displayed in Figure 4-2; the 3:1 Cl:NH₃-N (5:1 for Cl:DOC) dose is the ratio most commonly applied in the literature (Delpla & Rodriguez, 2016; Pifer et al., 2013; Roccaro et al., 2015; Roccaro & Vagliasindi, 2009; Teksoy et al., 2008; Yang et al., 2015), and the 10:1 Cl:NH₃-N (13.5:1 for Cl:DOC) dose stands for an excess chlorine dosing strategy.

In the 3:1 test, a much lower residual chlorine concentration was observed for the diluted (0.28 mg/L) compared with the undiluted (2.2 mg/L) sample, while for the 10:1 test a similar concentration in the diluted (1.62 mg/L) and undiluted (1.82 mg/L) samples was observed. However, the residual chlorine concentration and chlorine consumption percentage were not linearly correlated to the Cl: NH₃-N ratio for all 14 samples, which highlighted the significance of understanding the breakpoint chloramination of the AnMBR effluent.

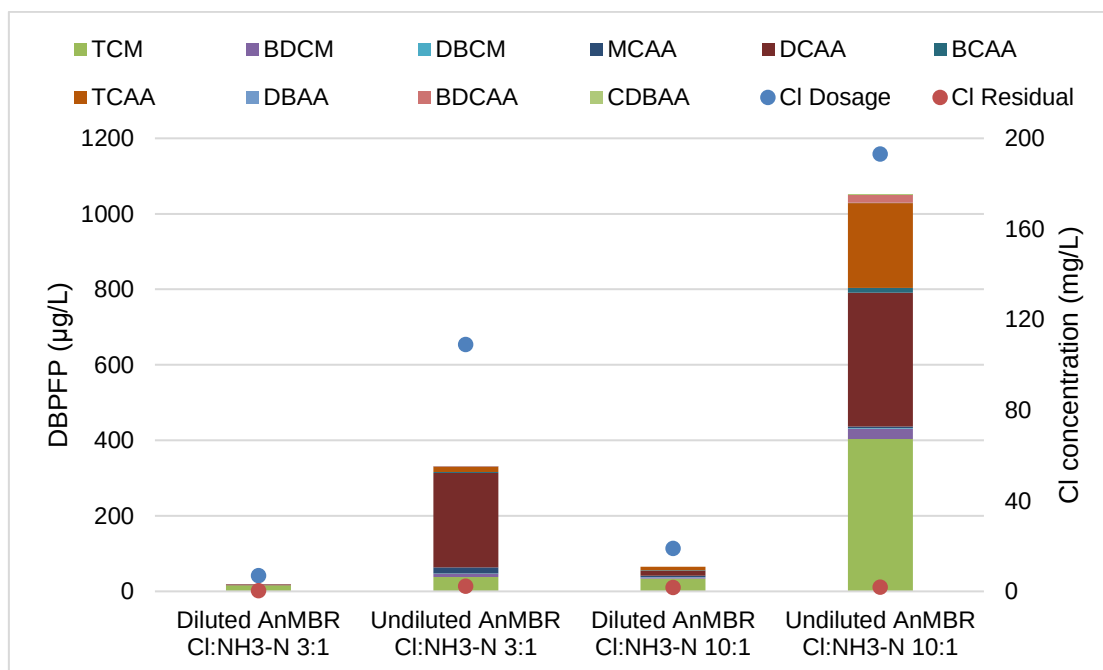


Figure 4-2 DBPFP of 15 fold diluted and undiluted AnMBR effluents, where the primary y-axis (left) is for the column and the secondary y-axis (right) is for the scatters

A clear difference between the diluted and undiluted samples was observed for the 3:1 and 10:1 dosing ratios. In the diluted 3:1 test, only 15.68 µg/L TCM was generated for measured DBPs with a 0.28 mg/L residual chlorine concentration, while the undiluted test generated a higher concentration of TCM (38.44 µg/L) and residual chlorine concentration (2.2 mg/L). The undiluted 3:1 test also generated more DBPs as BDCM for THM and MCAA, DCAA and TCAA for HAAs. Importantly, the diversity and quantity of generated DBPs were significantly increased in the undiluted sample under the 10:1 test. However, the differences observed in DBP diversity and quantity in the undiluted samples may also highlight the limit of analytical measurement. Indeed, the concentrations of formed DBPs in the diluted samples were likely to be close or below the LOD but the concentrations in the undiluted ones fitted in the range of the GC-MS calibration curve. This was supported by the DBP concentrations for the tests with 4:1 to 9:1 Cl:NH₃-N dosages and that increased chlorine dosage led to more DBPs being measured (Supporting information 3-1). This suggests that the dilution only proportionally reduced the amount of generated DBPs.

4.3.3.2 AeMBR effluent

As the AeMBR had a better performance in terms of organic removal, a lower dilution factor of 5 was applied to reach the 1 - 2 mg/L DOC range for the AeMBR effluents. The samples analysed with a Cl:DOC weight dosing ratio of 3:1 and 5:1 are displayed in Figure 4-3 since these two ratios are frequently quoted in the evidence base (Delpla & Rodriguez, 2016; Pifer et al., 2013; Roccaro et al., 2015; Roccaro & Vagliasindi, 2009; Teksoy et al., 2008; Yang et al., 2015). The chlorination process in the AeMBR effluent is expected to be monophonic (Hwang et al., 2001), since it occurred in an ammonia free environment and no nitrate consumption was observed during the chlorination process.

The diluted samples with dose ratios of 3:1 and 5:1 had 0.26 mg/L and 3.65 mg/L of residual chlorine present respectively in comparison to 1.35 mg/L and 18.9 mg/L in the associated undiluted samples. Hence a proportional reduction in residual chlorine was observed which matched the dilution factor. Noticeably, the chlorine consumption of each sample was calculated as a near-constant value, also displaying an amplified magnitude matching the dilution factor. These results for the AeMBR were observed across all the tests including the two samples shown in Supporting information 3-2, and display a different relationship than that recorded in the AnMBR tests. Moreover, the residual chlorine concentration displayed a strong linear correlation to the Cl:DOC ratio in both diluted and undiluted samples. All these findings confirmed that the monophonic chlorination process was dominant in the AeMBR effluents, which is different to that seen in chlorination of AnMBR effluent. This suggested dilution only proportionally reduces the compounds concentration in the MBR effluents, but the reduction may lead to an inaccurate detection in the low chlorine dosing scenario; that the reduced compounds concentration results in a reduced DBPFP, which is approaching the limit of current analytical measurement.

A similar impact of dilution was observed in the AeMBR effluent test compared to the AnMBR's with respect to DBPs. The diluted samples exhibited a lower quantity and diversity of generated DBPs due to the proportional reduction with the dilution and the limit of detection. This highlights possible underestimation of

DBPFP when working with diluted samples and for a better analytical measurement accuracy in relation to the application, dilution is not recommended when studying water with high organic content.

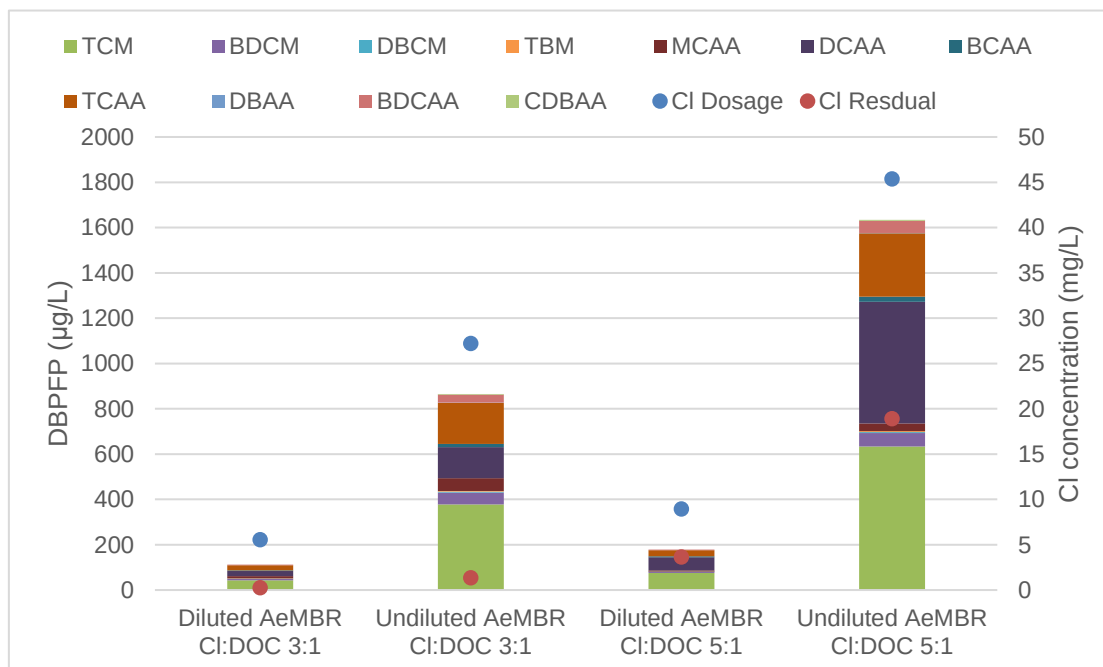


Figure 4-3 DBPFP of 5 fold diluted and undiluted AeMBR effluents

Overall, the AnMBR samples exhibited much lower DBPFP compared to the AeMBR samples regardless of dilution, even under a higher chlorine dose. For instance, the total THMFP (TTHMFP) and total HAAFP (THAAFP) concentrations in the undiluted AnMBR sample exposed to a 108.9 mg/l chlorine dose were 47.2 µg/L and 253.63 µg/L, respectively. The undiluted AeMBR sample with a 27.23 mg/L chlorine dose had TTHMFP and THAAFP values of 437.81 µg/L and 427.08 µg/L, respectively. This indicated that the AnMBR may be generating less DBPs during the chlorine based disinfection process, while satisfying the 1 mg/L residual chlorine concentration requirement with available free ammonia and phosphate for fertigation. These results highlight the need to extend our understanding of the mechanisms involved when chlorinating AnMBR effluent, specifically the factors affecting DBPFP and residual chlorine concentration as a function of different chlorine dosing regimes.

4.3.4 Identification of DBP formation indicators

In order to identify the DBP formation indicators and to check the impact of time variation of each sample, the AnMBR and AeMBR effluents were sampled twice each day for seven days then chlorinated with a fixed dose of chlorine to DOC weight ratio (5:1) regardless of the nitrogen concentration. Indeed, there was a minor variation in the substances present observed over the sampling period in the AnMBR and AeMBR effluents, which led to a very limited difference in terms of the DBPFP for both effluents.

4.3.4.1 AnMBR results

As shown in Table 4-3, under a fixed chlorine dose, Ammonia (0.79, 0.77) and DOC (0.74, 0.89) displayed strong positive correlations to THM and HAA formation potential, indicating that these two fractions are important contributors to DBP formation. Studies with natural water sources mostly report a positive trend between DBPFP with SUVA (Bond et al., 2012; Golea et al., 2017). In the current study, a negative trend was observed for both TTHMFP (-0.40) and THAAFP (-0.78), most likely driven by the dissimilar matrix of DOMs in the AnMBR effluent compared to natural waters.

A positive correlation (0.49) was only found between HA with TTHMFP in terms of Fluorescence Regional Integration (FRI) data, while weak negative correlations between TTHMFP and Apr (-0.34), APy (-0.30), FA (-0.32), and SMP (-0.17) were respectively found. SMP displayed a weak negative correlation to THAAFP (-0.22), whilst no correlation was found between THAAFP with the rest of the DOM fractions.

Na (0.73, 0.43), Si (0.78, 0.87), K (0.79, 0.81), and Mn (0.39, 0.50) respectively exhibited a strong positive correlation to TTHMFP and THAAFP, while Ca (-0.81, -0.54) and Sr (-0.84, -0.58) respectively displayed a substantial negative correlation to both DBPFP. A strong positive correlation between Mg (0.57) and Ba (0.50) with THAAFP was also observed (Table 4-4). This suggests trace elements have the potential to be used as indicators to predict the DBPFP behaviours of the AnMBR effluent.

The measured residual chlorine concentration (0.33 ± 0.06 mg/L) was inadequate to the 1 mg/L requirement in reuse standards and regulations (CNEPA, 2021; EUPC, 2020; ISO, 2018; USEPA, 2019). This can be explained by a co-metabolism effect of nitrification at lower Cl:N ratios (Wahman et al., 2016), suggests a higher Cl dosage is required for actual application. This ultimately proposed the need of obtaining a chlorine dosing response curve in terms of optimizing the chlorine dosage as well as understanding the variation of residual chlorine concentration and the DBP formation behaviour under an increasing chlorine dose.

Table 4-3 R values of multivariate correlation matrix of DBPFP with various effluent parameters for AnMBR (top) and AeMBR (bottom)

AnMBR (Ammonia nitrogen)									
Nitrogen	0.90	-0.60	-0.29	-0.29	-0.30	-0.17	0.35	0.79	0.77
0.06	DOC	-0.86	-0.11	-0.12	-0.11	-0.09	0.03	0.74	0.89
0.22	-0.07	SUVA	0.02	0.04	0.00	0.10	0.31	-0.40	-0.78
0.65	-0.36	0.08	APr	0.99	0.99	0.82	-0.78	-0.34	-0.03
0.65	-0.35	0.07	1.00	APy	0.99	0.79	-0.76	-0.30	-0.02
0.65	-0.35	-0.01	0.96	0.96	FA	0.78	-0.78	-0.32	0.01
0.66	-0.34	0.11	1.00	1.00	0.94	SMP	-0.41	-0.17	-0.22
0.65	-0.15	0.61	0.58	0.57	0.48	0.61	HA	0.49	-0.10
-0.42	-0.25	-0.45	-0.23	-0.22	-0.07	-0.27	-0.59	-0.04	-0.12
0.29	0.70	-0.21	-0.04	-0.03	0.05	-0.05	-0.19	Total THMs	0.67
-0.01	0.77	-0.38	-0.25	-0.24	-0.19	-0.27	-0.46	0.82	Total HAAs
AeMBR (Nitrate nitrogen)									

4.3.4.2 AeMBR results

The AeMBR effluent exhibited different correlation compared to the AnMBR during the fix Cl:DOC disinfection (Table 4-3). As the major different component compared to AnMBR, nitrate displayed a weak positive correlation to TTHMFP (0.29) and no correlation (-0.01) was found to THAAFP in the AeMBR samples, which indicated there was no impact caused by nitrate. However, DOC exhibited a significant positive correlation to TTHMFP (0.70) and THAAFP (0.77), respectively. This indicated that DOC can be the major indicator to predict the DBP formation in AeMBR effluent, whereas NH₃-N and DOC are the major

indicators for AnMBR effluent. The negative correlation found between SUVA with TTHMFP (-0.21) and THAAFP (-0.38) again confirmed the dissimilar matrix of DOMs in both MBR effluents compared to natural waters, demonstrating that SUVA is not suitable to quantitatively predict the DBP formation in MBR effluents.

There was no significant correlation found between each DOM fraction in FRI data with the TTHMFP, whereas the HA (-0.46) displayed a negative correlation to THAAFP and weak negative correlations for Apr (-0.25), APy (-0.24), FA (-0.19), and SMP (-0.27) were respectively found. This strong correlation displayed between HA content with DBPFP in both effluents were simply due the high content present in the initial effluent after UF filtration. However, the correlation obtained for the AeMBR effluent is different to the AnMBR effluent where the FRI data led to a stronger correlation to TTHMFP rather than THAAFP, suggesting the unique disinfection process of AnMBR and AeMBR effluent caused by dissimilar matrix performed differently on the DOM fractions present. Furthermore, this may suggest that under the same Cl:DOC weight dosing ration, chloramination formed less THMs than HAAs while chlorination forms more THMs than HAAs. Moreover, the weak correlation found between the FRI data with either TTHMFP or THAAFP regardless of the effluent matrix is different to Ma et al. (2016) who observed a significant positive correlation between TTHMFP with FA and HA during the chlorination and chloramination of an AeMBR effluent, respectively. This suggests the 3D-EEM may be used as a good tool to identify the DBP precursors but not for quantitative prediction for the application on MBR effluent disinfection process.

Positive correlations were found for Na (0.49, 0.49), Si (0.34, 0.34) and K (0.66, 0.52) and negative correlations for Ca (-0.46, -0.31) and Sr (-0.46, -0.31), with both TTHMFP and THAAFP in the AeMBR samples (Table 4-4). A weak negative correlation was observed for B (-0.32), Mg (-0.23), Zn (-0.23), and Ba (-0.23) with TTHMFP but there was no correlation found for these elements with THAAFP. The negative correlation found for Mg and Ba in the AeMBR effluent is strikingly different from the positive correlations found for the AnMBR effluent, suggesting these two elements may have different behaviours during the chloramination and

chlorination process. Our results also suggest that B and Zn have only a negligible impact regardless of the disinfection process or the water matrix, since no correlation was found between B and Zn with DBPFP during the disinfection of AnMBR and AeMBR effluents. Moreover, the correlations found for Na, Si, K, Ca, and Sr in the AeMBR effluent are similar to those performed in the AnMBR effluents. This finding agrees with values obtained with surface water by Ye et al. (2009), suggesting that these elements have a similar behaviour during the disinfection regardless of the water matrix; a conclusion which can help indicate the DBP formation potential in applications where the MBR effluent is blended with a natural water source.

The residual chlorine concentration (2.58 ± 0.48 mg/L) measured in the disinfected AeMBR effluent easily exceeds the 1 mg/L requirement specified in many reuse standards and regulations (CNEPA, 2021; EUPC, 2020; ISO, 2018; USEPA, 2019). This suggests that lower doses could be used when disinfecting the AeMBR effluent; highlighting the need to obtain accurate chlorine dose response curves for a better understanding of chlorine consumption and DBP formation behaviour, while offering a direct comparison to the disinfection of AnMBR effluent.

Table 4-4 R values of multivariate correlation matrix of DBPFP with trace element concentrations in AnMBR (top) and AeMBR (bottom) effluent

AnMBR												
B	0.14	0.25	-0.01	-0.16	0.27	-0.36	-0.55	0.09	0.29	0.34	-0.04	-0.12
-0.47	Na	-0.05	0.67	0.68	-0.64	0.29	-0.51	0.33	-0.65	-0.02	0.73	0.43
0.76	-0.81	Mg	0.43	0.28	0.22	0.36	0.05	-0.35	0.18	0.83	-0.06	0.57
-0.17	-0.26	0.27	Si	0.96	-0.74	0.54	-0.32	0.14	-0.77	0.49	0.78	0.87
-0.55	0.30	-0.33	0.74	K	-0.86	0.48	-0.21	0.29	-0.88	0.31	0.79	0.81
0.88	-0.70	0.89	-0.16	-0.68	Ca	-0.25	0.23	-0.42	0.99	0.09	-0.81	-0.54
0.00	0.00	0.00	0.00	0.00	0.00	Mn	-0.21	-0.11	-0.27	0.44	0.39	0.50
-0.34	0.62	-0.55	-0.52	-0.11	-0.37	0.00	Fe	-0.13	0.17	-0.16	-0.29	0.02
-0.10	0.18	-0.26	0.04	0.19	-0.19	0.00	-0.27	Zn	-0.41	-0.27	0.21	-0.01
0.88	-0.65	0.85	-0.22	-0.71	0.99	0.00	-0.33	-0.18	Sr	0.07	-0.84	-0.58
0.96	-0.35	0.65	-0.20	-0.51	0.80	0.00	-0.22	-0.04	0.81	Ba	0.12	0.50

-0.32	0.49	-0.23	0.34	0.66	-0.46	0.00	0.23	-0.23	-0.46	-0.23	Total THMs	0.67
-0.09	0.49	-0.10	0.34	0.52	-0.31	0.00	0.15	-0.08	-0.31	0.01	0.82	Total HAAs
AeMBR												

4.3.5 Behaviour of AnMBR and AeMBR effluent during disinfection

A chlorine dosing trial with residual chlorine concentrations from 0.01 mg/L to 10 mg/L was conducted for both the AnMBR (n=18) and AeMBR (n=11) effluents. Samples were continuously taken from the MBRs within 10 minutes to minimize the time variation and kept undiluted. Importantly, the DOC and NH₃-N for the tested AnMBR effluent was 21.39 mg/L and 36.3 mg/L, respectively, while the DOC for the AeMBR effluent was 9.08 mg/L.

4.3.5.1 Breakpoint disinfection of AnMBR effluent

In the AnMBR effluent, a complex interaction takes place between the DOM, ammonia and chlorine due to the breakpoint chloramination process which is dependent on the Cl:N weight dosing ratio. The residual chlorine concentration profile displayed a similar shape (Figure 4-4) but different Cl:N ratios and residual Cl concentrations compared to results previously reported in the literature (IXOM Watercare, 2005; Wahman, 2018). The different ratios and residuals obtained can be explained by the presence in our samples of a more complex water matrix with higher initial DOC and ammonia concentrations. As expected, three typical chloramination stages were clearly observed: Mono-chloramine; Bi-chloramination; and Chlorami-Chlorination, corresponding to the regions labelled 1, 2 and 3 in Figure 4-4. The red triangle marked in each region represent the critical dosing point needed to produce a final effluent with sufficient chlorine residual of 1 mg/L.

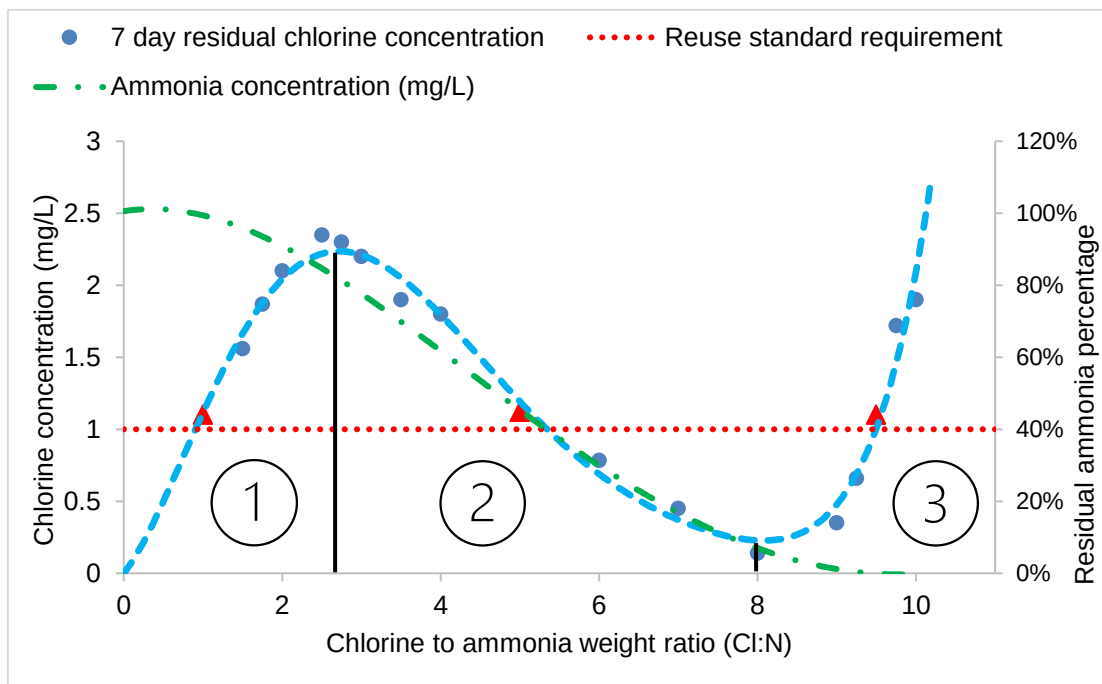


Figure 4-4 Chlorine breakpoint curve for AnMBR effluent with fixed DOC level

4.3.5.1.1 Mono-chloramine stage

In the mono-chloramine stage the first critical dosing point was observed when Cl:N was 1:1, with a negligible removal of ammonia and the lowest DBPFP amongst 18 tested samples. This was the only sample that contained no BDCAA, whilst the TTHMFP and THAAFP levels were 28.39 $\mu\text{g/L}$ and 144.7 $\mu\text{g/L}$, respectively (Figure 4-5). The rest of the samples in this stage also displayed a lower DBPFP compared to higher Cl:N ratios, with no TBM, MBAA, CDBAA and TBAA detected. This suggests that the amount of chlorine dosed into the AnMBR effluent reacted quickly with the available free ammonia to form monochloramine which ultimately led to a lower DBPFP (Hwang et al., 2001). This dosing regime offers enough ammonia and chlorine residual to ensure both fertigation and safeness of the disinfected effluent for irrigation purpose (European Commission, 2017; Xu et al., 2020; Zaouri et al., 2021).

A significant logarithmic correlation was found between TTHMFP and the Cl:N weight ratio, with all THMs increasing with rising Cl dose. However, a weak correlation between THAAFP with Cl:N was found as the main HAA component

(DCAA) varied with the residual chlorine concentration. At this stage, HAAs accounted for more than 79% of total generated DBPs.

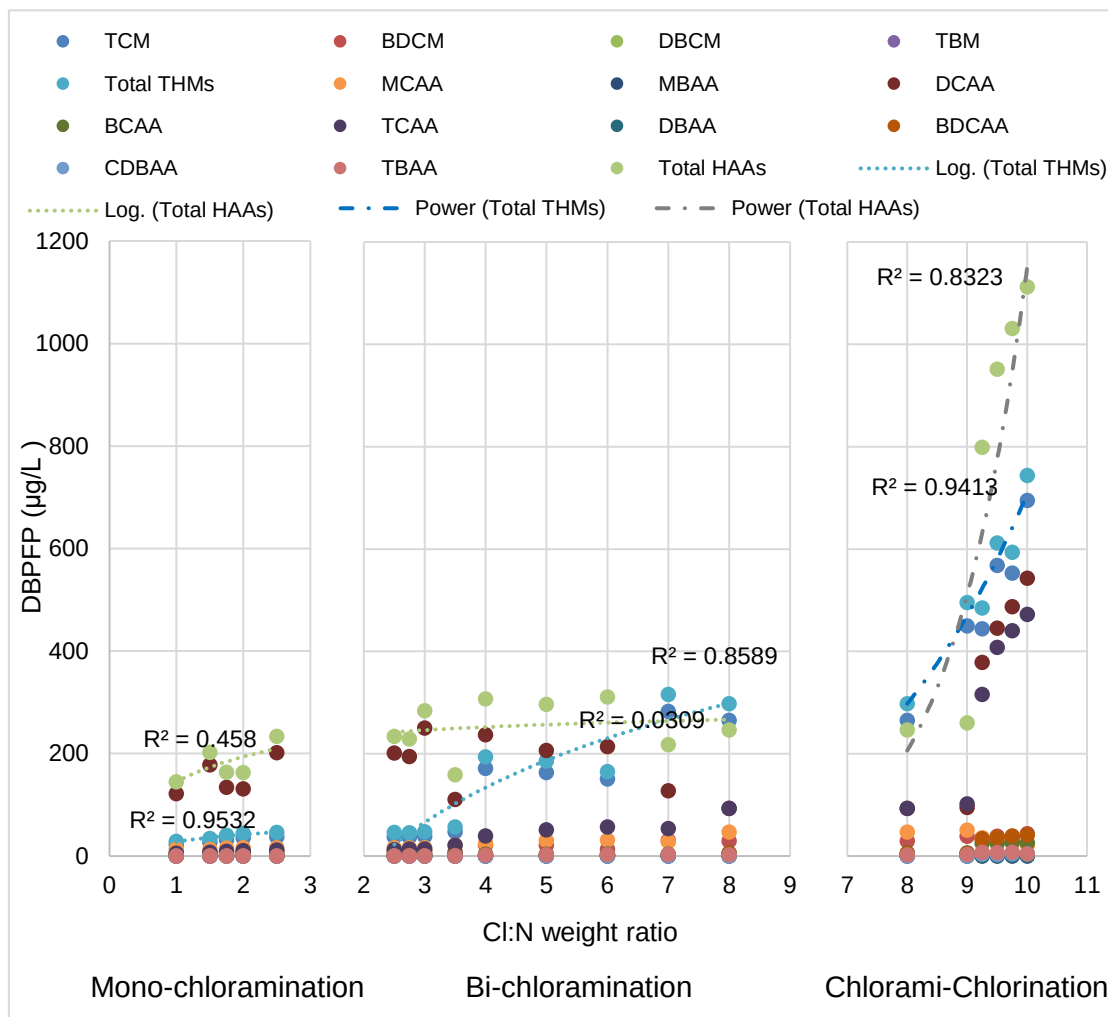


Figure 4-5 DBPFP behaviours under different chlorination stage of AnMBR effluent

4.3.5.1.2 Bi-chloramination stage

Two disinfectants are expected to be formed at this stage; monochloramine and dichloramine (Wahman, 2018), and residual chlorine concentrations decreased despite the increasing chlorine dose (Figure 4-5). A second critical dosing point was observed at a Cl:N ratio of 5:1, with 56% removal of ammonia, much higher TTHMFP and slightly higher THAAFP (185.37 µg/L and 295.99 µg/L, respectively) were formed when compared to the first point.

In comparison to the monochlorination stage, all four THMFPs exhibited a sudden increase in response to the dosing ratio increasing from Cl:N 3.5 to 4 with TBM

being first detected at the Cl:N ratio of 4. The formed dichloramines are much more unstable than monochloramines which leads to a quick release of chlorines (Coker et al., 2008), and ultimately react with DOM to generate more DBPs. However, this effect only promoted the formation of THMs but not HAAs and the Cl:N ratio exhibited a strong logarithmic correlation with TTHMFP but a poor correlation with THAAFP. The uncorrelated THAAFP was again caused by the FP of DCAA, which displayed a variable trend with the residual chlorine concentration. Apart from the DCAAFP, all other detected HAAFPs displayed a strong positive logarithmic correlation to the Cl:N ratio. This stage can be seen as a transition stage since the greatly increased THMs accounted for a higher percentage of total generated DBPs, while the percentage of HAAs decreased from 84% to 45%. Noticeably, nitrate was subsequently formed from the Cl:N ratio of 5:1, which agreed with Wahman (2018) that nitrate can form during the Bi-chloramination stage with an increasing Cl:N ratio.

4.3.5.1.3 Chlorami-Chlorination stage

In this stage the formed disinfectants are expected to be trichloramine and chlorine (Wahman, 2018). Residual chlorine concentration increased with the increasing chlorine dose. A third critical dosing point was observed at a Cl:N ratio of 9.5:1, with complete removal of ammonia and a slightly higher TTHMFP and similar THAAFP (297.39 µg/L and 246.24 µg/L, respectively) when compared to the second point.

In comparison to Bi-chloramination, the transition point of the Chlorami-chlorination stage occurred between the Cl:N ratio of 9-9.25:1. At this point the residual ammonia concentration decreased to non-detectable, which indicated the full consumption of free ammonia and the excess added chlorine starting to be present as free chlorine. This was confirmed by the increasing TTHMFP and a significantly increase in THAAFP. CDBAA was first detected after this point. Other bromide DBPs such as BCAA, DBAA, BDCAA, and TBAA also exhibited a sharp increase in concentration after the transition point. This was because chlorine can displace the heavier and less electronegative halogens from other compounds (Gregersen, 2021) - bromine in our study – and subsequently form

more bromide related DBPs. Furthermore, concentrations of both TTHMFP and THAAFP exhibited a significant positive power correlation with the Cl:N ratio. This again confirmed that the chlorination process caused by free chlorine can lead to more DBPs formation since chlorine is a much stronger disinfectant in comparison to chloramine (Hwang et al., 2001; Wahman et al., 2016). This reaction led to a sharp increase in the percentage of total HAAs seen in the samples as a function of total generated DBPs - from 34% to 60%. However, MCAAFF displayed an opposite trend compared to all other detected HAAs, with lower MCAAFF observed with the increasing Cl dose which cannot be explained at this stage.

Dosing strategies focused beyond the transition point (Cl:N ratio of 8:1) found in the Chlorami-chlorination stage can be used to prevent reverse osmosis (RO) membrane fouling while reducing the final DBPs when high grade effluent is required for potable water reuse applications. Dosing at these levels maximise ammonia removal while keeping a minimum chlorine load to the RO membranes. Previous work has demonstrated that excess chlorine after pre-treatment will reduce the life-cycle of RO system (Al-Abri et al., 2019) whilst RO is reported to effectively remove the DBPs generated by pre-RO chlorination process (Kimura et al., 2003; Zazouli & Kalankesh, 2017). Also, our previous work has suggested the unremoved ammonia in AnMBR effluent is one of the key RO foulants (Chapter 3).

4.3.5.1.4 Overview of AnMBR effluent disinfection process

Several findings from the experimental work invite further explanation. Firstly, no MBAA was detected in any of the tested samples. This may be due to the bromide in MBAA being replaced by chloride to form MCAA (Gregersen, 2021). Secondly, DCAA displayed a tight correlation to the residual chlorine concentration while all other detected DBPs correlated significantly with the increasing chlorine dose (Figure 4-6). This may suggest that DCAA has a slow reaction kinetic with chlorines compared to other DBPs. However, the reaction kinetic of free chlorine with co-presence of ammonia and DOM was not directly observable and remains unclear. Further work in this area may provide an insight into the reaction priority

which may give a better understanding of DBPs generated during this disinfection process.

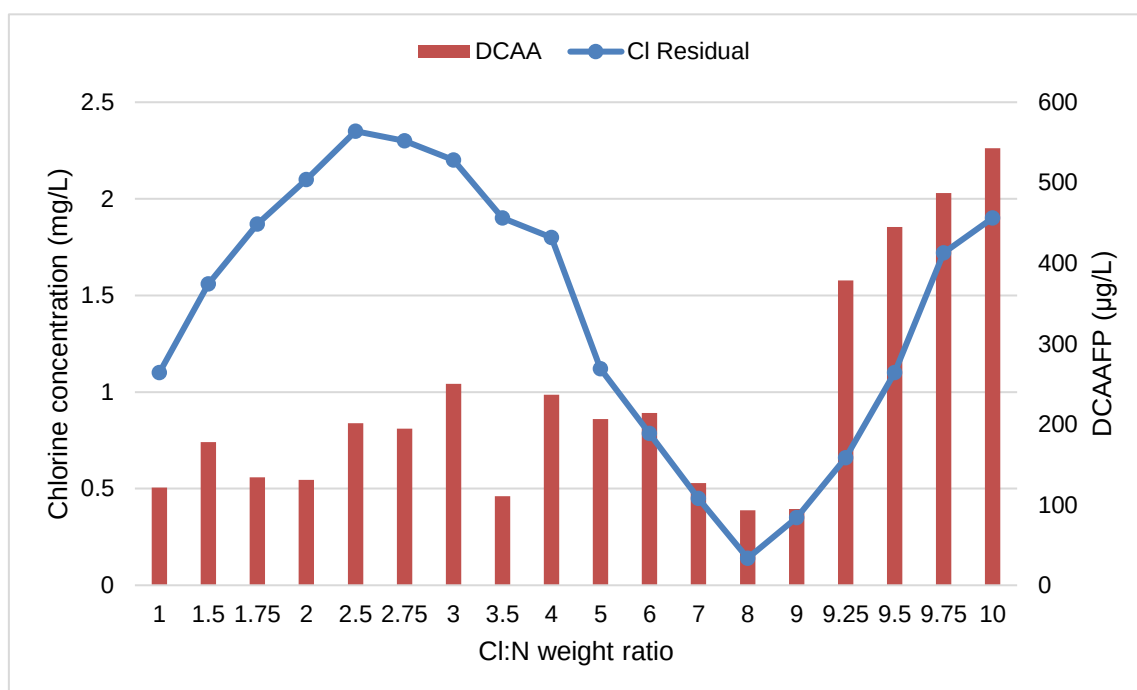


Figure 4-6 DCAA with residual chlorine concentration

The chloramination breakpoint significantly depends on the initial free ammonia concentration, DOC concentration, temperature, pH, and a specific reaction time (Wahman, 2018). Therefore, for actual reuse applications, determining a chlorine dosing response curve on a case-by-case basis is recommended since wastewater effluents may have different matrices under different treatment conditions and feed water qualities.

4.3.5.2 Chlorination of AeMBR effluent and comparison with AnMBR test

In comparison to the AnMBR effluent, the disinfectant formed in AeMBR samples was identified as free chlorine, where the Cl:DOC weight dosing ratio was showed to be the critical factor correlated to DBPFP (Figure 4-7). The increase of residual chlorine concentration with the chlorine dosing confirmed a monophasic chlorination process. The critical dosing point of 1 mg/L residual chlorine concentration was identified as occurring between a Cl:DOC ratio of 2.75-3. Both TTHMFP and THAAFP displayed a very strong positive correlation with increasing Cl dose, while overall THMs had a higher initial formation potential but

a lower increasing rate compared to HAAs (Figure 4-7). However, MCAA displayed again a different trend than all other DBPs, and kept increasing up to a Cl:DOC ratio of 3:1 then decreased with the increasing chlorine dosing, which again needs further investigation to be understood.

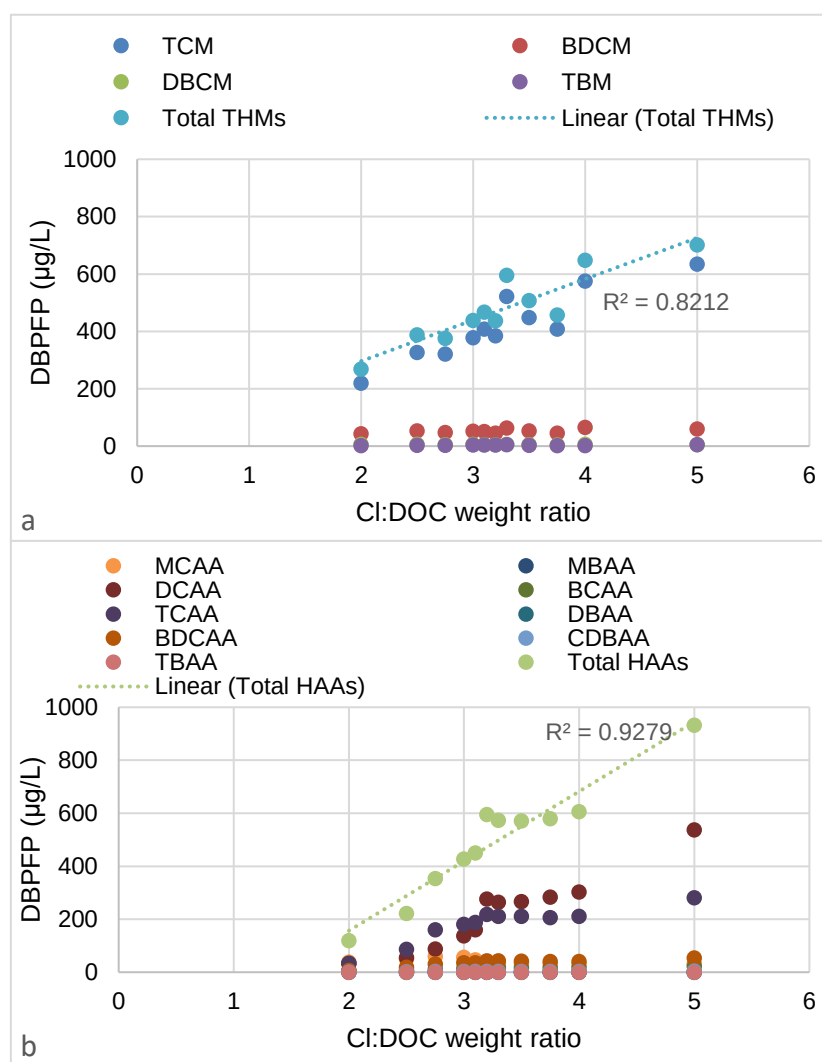


Figure 4-7 DBPFP behaviours of THMs (a) and HAAs (b) for AnMBR effluent under various Cl:DOC ratios

The detected THMs had a quantitative formation priority of $\text{TCM} > \text{BDCM} > \text{DBCM} > \text{TBM}$, which is the same as the results obtained with the AnMBR effluent. This finding agrees with Ma et al. (2016) that TCM accounts for more than 80% of THMs formed during the chlorination of AeMBR effluent. Moreover, the weight percentage of TCM in both the AnMBR and AeMBR samples rose with the increasing chlorine dose, while TCMFP of the AnMBR

samples had a sudden increase at the first transition point but displayed a constant increasing trend in AnMBR samples. This suggests that monochloramine may lead to a lower TCMFP which ultimately will lead to a lower TTHMFP under these conditions. Likewise, the sudden increase in THAAFP after the second transition point in AnMBR samples indicated that the combined chloramination may have a lower THAAFP compared to chlorination. There was no MBAA and TBAA detected in the AeMBR samples. The undetectable MBAA was similar to AnMBR results but a trace level of TBAA was found in AnMBR samples. This could be explained by the fact that the available free chlorine concentration was much higher in the AeMBR samples due the different chlorination process. Therefore, Br in the formed TBAA was replaced by Cl which may form as chlorine related HAAs. However, the AnMBR samples had a much lower bromide related DBPFP compared to the AeMBR samples. This may be because of the weaker reactivity of chloramine and the simultaneous removal of bromide with nitrate during the biological process of an AnMBR (Butler et al., 2005; Downing & Nerenberg, 2006). This suggests although the ICP-MS reported the Br as non detectable, there were still a tiny amount Br presence in both effluents, which will ultimately contribute to the bromide related DBPs.

4.3.6 DBPFP of AnMBR and AeMBR effluents compared to surface water

The DBPFP yield ($\mu\text{g}/\text{mg}$ DOC) with the corresponding chlorine dosage (mg/L) for the three AnMBR samples and one AeMBR sample which allowed to meet the 1 mg/L residual chlorine concentration standard for the potential irrigation reuse application (CNEPA, 2021; EUPC, 2020; ISO, 2018; USEPA, 2019) are displayed in Figure 4-8. An increasing trend of DBPFP yield was observed with a TTHMFP yield of 1.33, 8.67 and 28.59 $\mu\text{g}/\text{mg}$ DOC when the chlorine dose was increased from 36.3 to 181.5 and then 344.5 mg/L in the AnMBR effluent. Similarly, the THHAFP yield increased from 6.76 to 13.84 and then 44.46 for the same doses in the AnMBR effluent. Interestingly, a lower chlorine dose of 27.2 mg/L in the AeMBR effluent led to even higher TTHMFP and THAAFP yields of 48.23 and 47.05 $\mu\text{g}/\text{mg}$ DOC, respectively. At the same chlorine residual level, the AnMBR effluent displayed an increasing formation DBP variety with increasing chlorine

dosage but a lower TTHMFP and THAAFP yield compared to the AeMBR effluent even as the chlorine dose increased. This highlighted the possible advantages of reusing AnMBR effluent due to the fact that low formation of DBPs and high residual nutrients concentration can be achieved by controlling the chlorine dosage.

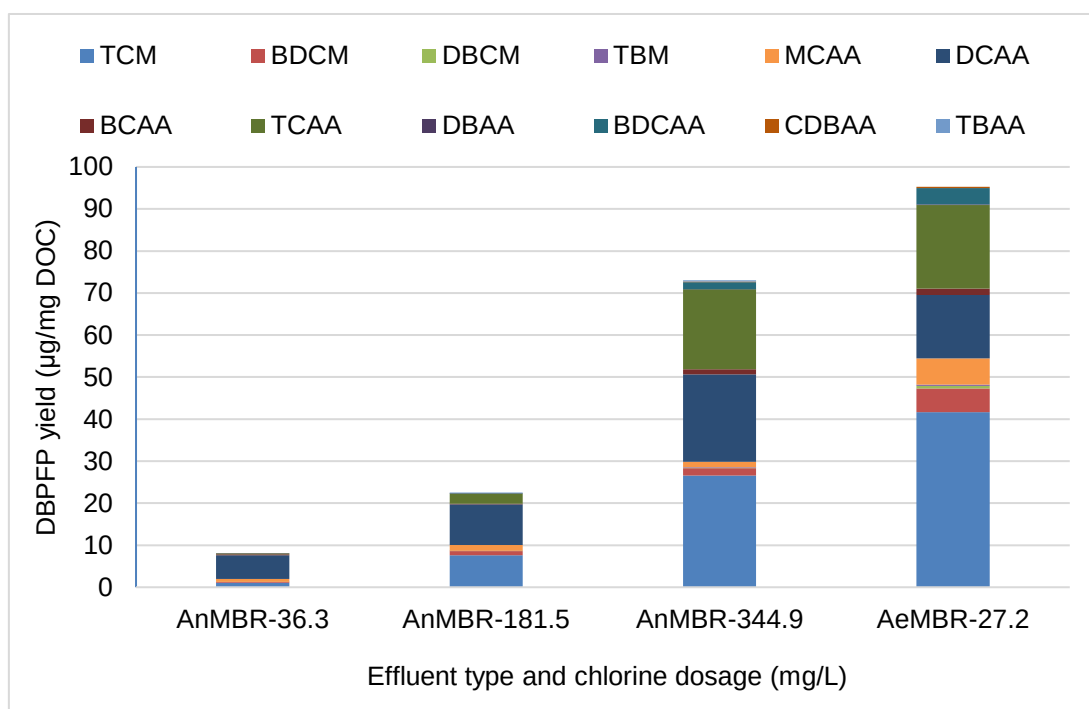


Figure 4-8 DBPFP yield vs. DOC of AnMBR and AeMBR effluent at 1 mg/L residual chlorine concentration

Table 4-5 compares the results reported above with previously published studies which investigated the disinfection of AeMBR effluents and surface waters, showing the DBPFP yield ($\mu\text{g}/\text{mg DOC}$) ranked in ascending order. The AnMBR sample with a chlorine dose of 36.3 mg/L and the AnMBR sample with 181.5 mg/L (relating to the first and second critical dosing points described above) exhibited the lowest TTHMFP (1.33 and 8.67 $\mu\text{g}/\text{mg DOC}$, respectively) and THAAFP yield (6.76 and 13.84 $\mu\text{g}/\text{mg DOC}$, respectively). These are lower than the TTHMFP (15 and 18 $\mu\text{g}/\text{mg DOC}$) and THAAFP (25 and 38 $\mu\text{g}/\text{mg DOC}$) yields of two of the treated surface waters. The AnMBR sample with a chlorine dose of 344.9 mg/L had a higher chlorine dosage and complete ammonia removal yet still performed better on DBPFP than most of the treated and raw surface

water samples and the AeMBR effluents reported in literature. The TTHMFP yield (48.23 µg/mg DOC) of the AeMBR sample with 27.2 mg/L is higher than one raw (35 µg/mg DOC) and one treated (1.33 µg/mg DOC) surface water; and one AeMBR (47.5 µg/mg DOC) but still offers a better performance compared with the rest of the surface waters. Noticeably, the TTHMFP yields of this AeMBR (Ma et al., 2016) effluent was obtained with chloramination rather than the chlorination applied in this study. This again confirms the advantage when disinfecting AnMBR effluent; that a low TTHM formation is achieved even though the AeMBR experienced a much lower chlorine dose as well as a relatively similar chloramination process. Moreover, two AeMBR effluent samples exhibited the highest two TTHMFP yields of 502 µg/mg DOC and 1402 µg/mg DOC. This was because the dosing regime conducted in these two studies had a Cl:DOC weight ratio of 20:1, which consequentially led to the high TTHMFP yields (Ma et al., 2016; Meng et al., 2017). For the THAAFP yield, the AeMBR sample with a chlorine dose of 27.2 mg/L exhibited good performance with a similar value of 47.05 µg/mg DOC compared to the AnMBR chlorine dose of 344.9 mg/L and 44.46 µg/mg of DOC. This was higher than only two of the treated surface waters.

Table 4-5 DBPFP yield (µg/mg DOC) values vs. various water source

Water type	TTHMFP yield (µg/mg DOC)	Source	Water type	THAAFP yield (µg/mg DOC)	Source
AnMBR-36.3	1.33	This study	AnMBR-36.3	6.76	This study
AnMBR-181.5	8.67	This study	AnMBR-181.5	13.84	This study
TSW	15	(Roccaro & Vagliasindi, 2009)	TSW	25	(Roccaro et al., 2015)
TSW	18	(Roccaro et al., 2015)	TSW	38	(Roccaro & Vagliasindi, 2009)
AnMBR-344.9	28.59	This study	AnMBR-344.9	44.46	This study
RSW	35	(Roccaro et al., 2015)	AeMBR-27.2	47.05	This study

TSW	43	(Delpla & Rodriguez, 2016)	RSW	65	(Roccaro et al., 2015)
AeMBR - 103.5 (Chloramination)	47.5	(Ma et al., 2016)	TSW	70	(Golea et al., 2017)
AeMBR-27.2	48.23	This study	RSW	88	(Roccaro & Vagliasindi, 2009)
RSW	55	(Roccaro & Vagliasindi, 2009)	TSW	117	(Delpla & Rodriguez, 2016)
TSW	61	(Teksoy et al., 2008)	RSW	216	(Golea et al., 2017)
TSW	71	(Golea et al., 2017)			
TSW	91	(Pifer et al., 2013)			
RSW	124	(Yang et al., 2015)			
RSW	149	(Golea et al., 2017)			
AeMBR - 88.4 (Chlorination)	502	(Meng et al., 2017)			
AeMBR - 103.5 (Chlorination)	1402	(Ma et al., 2016)			

Notes: TSW: Treated surface water; RSW: Raw surface water.

Overall, these results suggest that AnMBR effluent offers a lower DBP formation potential compared to both surface waters and AeMBR effluent. It can also achieve a controllable disinfection process with a required residual chlorine and ammonia concentration for various irrigation purposes. Furthermore, DOM in AnMBR effluent can be reduced by optimizing the operating conditions such as temperature, hydraulic retention time, and organic loading rate (Shin & Bae, 2018; Song et al., 2018), as well as blending with natural water (Comerton et al., 2006). This will ultimately lead to a further reduction of the DBPs generated during the disinfection process. Disinfection of the AeMBR effluent had a lower chlorine demand but a higher DBPFP yield compared to the AnMBR effluent. Here, further treatment might be needed with activated carbon or reverse osmosis filtration.

4.4 Conclusion

Chlorine based disinfection processes behave differently in AnMBR and AeMBR effluents due to the different water matrices. The presence of ammoniacal nitrogen in AnMBR effluent leads to a complex process of chloramination under lower chlorine dose, and a combined process of chloramination and chlorination at higher chlorine dose. The chlorine dose, ammonia and DOC concentration are the critical factors impacting DBP formation. Disinfection of AeMBR effluent is a monophonic chlorination process regardless of the chlorine dose, with the chlorine dose and DOC concentration being the critical factors. Overall, AnMBR effluent had a much lower DBPFP compared to AeMBR effluent under the same chlorine dose. Reusing the disinfected AnMBR effluent has the advantage of low DBP generation alongside a high residual of ammonia and phosphate, as well as a controllable disinfection process with a required residual chlorine and ammonia concentration. Furthermore, dosing regime after the third critical chlorine dosing point can be applied to remove the ammonia in the AnMBR effluents while mitigating the RO fouling when higher reuse water quality is needed.

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4.6 Supporting information 3

4.6.1 Supporting information 3-1

Table 4-6 DBPFP of diluted and undiluted AnMBR samples

No.\Cat.	Dilution f	Cl Residual	Cl/D OC	Cl/N H ₃	TCM	BDC M	DBC M	TBM	Total THM _s	MCA A	MBA A	DCA A	BCA A	TCA A	DBA A	BDC AA	CDB AA	TBA A	Total HAA _s
AnMBR diluted	1	15	0.2	3.0	1.9	10.5	0.0	0.0	0.0	10.5	0.0	0.0	2.1	0.0	0.0	0.0	0.0	0.0	2.1
	2	15	0.3	5.0	3.0	15.7	0.0	0.0	0.0	15.7	0.0	0.0	3.2	0.0	0.0	0.0	0.0	0.0	3.2
	3	15	0.3	5.4	4.0	10.3	0.0	0.0	0.0	10.3	0.0	0.0	4.1	0.0	0.0	0.0	0.0	0.0	4.1
	4	15	0.2	6.0	4.5	9.0	0.0	0.0	0.0	9.0	0.0	0.0	3.7	0.5	1.2	0.0	0.0	0.0	5.4
	5	15	0.7	8.3	5.0	12.2	0.0	0.0	0.0	12.2	0.0	0.0	4.0	0.0	0.0	0.0	0.0	0.0	4.0
	6	15	0.1	8.4	5.3	9.5	0.0	0.0	0.0	9.5	0.0	0.0	5.6	0.0	1.2	0.0	0.0	0.0	6.9
	7	15	0.2	8.9	5.5	10.2	0.0	0.0	0.0	10.2	0.0	0.0	5.5	0.4	1.0	0.0	0.0	0.0	7.0
	8	15	0.1	10.0	6.0	12.6	0.0	0.0	0.0	12.6	0.0	0.0	4.3	0.0	1.3	0.0	0.0	0.0	5.5
	9	15	0.1	10.2	7.6	16.3	0.0	0.0	0.0	16.3	0.0	0.0	3.4	0.0	0.0	0.0	0.0	0.0	3.4
	10	15	0.3	10.5	7.0	13.8	0.0	0.0	0.0	13.8	0.0	0.0	3.7	0.0	0.0	0.0	0.0	0.0	3.7
	11	15	0.6	11.0	9.3	28.3	0.0	0.0	0.0	28.3	0.0	0.0	7.0	0.0	0.0	0.0	0.0	0.0	7.0
	12	15	0.1	12.4	9.0	25.0	0.0	0.0	0.0	25.0	0.0	0.0	5.9	0.0	0.0	0.0	0.0	0.0	5.9
	13	15	2.1	13.0	11.0	33.0	0.0	0.0	0.0	33.0	0.0	0.0	10.8	0.0	0.0	0.0	0.0	0.0	10.8
	14	15	1.6	13.5	10.0	33.5	3.5	0.8	0.0	37.8	3.5	0.0	12.9	1.4	9.9	0.0	0.0	0.0	27.7
AnMBR undiluted	1	1	2.1	3.4	2.0	35.3	8.2	0.6	0.0	44.1	16.0	0.0	131.1	3.8	10.7	0.4	0.6	0.0	162.5
	2	1	2.3	4.2	2.5	37.0	8.4	0.5	0.0	45.9	16.1	0.0	201.4	3.2	12.0	0.4	0.5	0.0	233.2

	3	1	2.2	5.1	3.0	38.4	8.3	0.5	0.0	47.2	16.1	0.0	250. 1	3.1	13.8	0.4	0.5	0.0	0.0	283. 6
	4	1	1.9	5.9	3.5	47.1	8.6	0.7	0.0	56.5	20.8	0.0	110. 7	4.9	20.9	0.4	1.1	0.0	0.0	158. 9
	5	1	1.8	6.8	4.0	171. 3	20.8	1.0	0.4	193. 5	23.8	0.0	236. 7	3.2	39.2	0.3	1.5	0.0	1.9	306. 3
	6	1	1.1	8.5	5.0	163. 1	20.8	1.0	0.5	185. 4	30.2	0.0	206. 5	3.4	51.2	0.3	1.9	0.0	2.4	295. 7
	7	1	0.8	10.2	6.0	150. 6	12.9	0.9	0.5	164. 9	30.7	0.0	213. 9	3.9	56.9	0.3	1.9	0.0	2.9	310. 6
	8	1	0.5	11.9	7.0	282. 5	30.7	1.9	0.8	315. 8	28.2	0.0	127. 1	3.6	53.6	0.3	1.8	0.0	2.6	217. 3
	9	1	0.1	13.6	8.0	265. 0	29.1	1.8	1.5	297. 4	47.0	0.0	93.0	5.9	93.1	0.4	4.7	0.0	2.1	246. 2
	10	1	0.4	15.3	9.0	449. 6	38.7	2.7	4.0	495. 0	50.1	0.0	94.6	6.0	101. 5	0.4	4.9	0.0	2.7	260. 3
	11	1	0.7	10.7	9.3	444. 0	35.0	2.3	3.1	484. 4	35.3	0.0	378. 5	24.5	315. 8	0.9	33.4	3.5	7.0	798. 9
	12	1	1.1	11.0	9.5	567. 8	38.3	2.0	3.4	611. 5	27.5	0.0	445. 0	24.6	407. 8	0.8	35.8	2.8	6.7	951. 0
	13	1	1.7	11.3	9.8	552. 9	35.8	1.9	3.1	593. 6	26.4	0.0	487. 0	26.1	439. 9	0.9	39.0	4.3	7.0	1030 .6
	14	1	1.8	11.8	10.0	403. 4	27.1	1.7	0.0	432. 2	4.8	0.0	354. 4	12.6	224. 6	0.7	22.4	1.0	0.0	620. 5

4.6.2 Supporting information 3-2

Table 4-7 DBPFP of diluted and undiluted AeMBR samples

No.\Cat.	Dilution f	Cl Residual	Cl/DO C	TCM	BDC M	DBC M	TBM	Total THM S	MCA A	MBA A	DCA A	BCA A	TCA A	DBA A	BDCA A	CDBA A	TBA A	Total HAA S
AeMBR diluted	1	5	0.0	1.0	41.3	4.7	0.7	0.5	47.2	2.2	0.0	6.1	0.5	0.0	0.4	0.0	0.0	9.2
	2	5	0.3	3.0	42.4	6.3	1.2	1.8	51.7	10.2	0.0	22.7	2.3	22.6	0.0	2.4	0.0	60.2
	3	5	0.8	3.1	52.9	7.5	1.2	0.0	61.6	5.7	0.0	20.8	2.5	22.6	0.0	3.6	0.0	55.1
	4	5	1.2	3.2	54.0	1.1	1.2	0.0	56.3	5.4	0.0	22.0	2.6	23.2	0.0	3.7	0.0	56.9
	5	5	1.8	3.3	39.7	6.6	1.3	0.0	47.6	5.1	0.0	16.2	2.5	14.8	0.0	3.4	0.0	41.9
	6	5	1.9	3.4	53.3	8.2	1.6	0.0	63.1	5.0	0.0	16.5	2.5	15.3	0.0	3.5	0.0	42.8
	7	5	2.6	4.0	59.6	8.3	1.4	0.0	69.3	4.2	0.0	19.7	2.6	19.3	0.0	3.9	0.0	49.6
	8	5	3.0	4.4	58.1	7.7	1.2	0.0	67.0	4.0	0.0	19.6	2.5	18.3	0.0	3.5	0.0	47.9
	9	5	3.7	5.0	75.0	7.1	0.7	0.3	83.2	3.6	0.0	58.6	3.0	27.0	0.0	3.3	0.0	95.5
	10	5	3.6	5.7	51.6	10.7	2.7	0.0	65.0	8.7	0.0	19.8	3.8	16.3	0.0	5.6	0.9	55.0
	11	5	3.7	5.9	51.0	10.5	2.6	0.0	64.0	8.6	0.0	22.1	4.3	18.7	0.5	6.2	1.0	61.4
	12	5	3.5	6.0	51.1	10.6	2.6	0.0	64.3	6.9	0.0	22.8	4.3	19.5	0.5	6.5	1.1	61.7
	13	5	3.3	6.2	58.1	11.7	2.9	0.0	72.7	6.3	0.0	24.6	4.5	20.9	0.6	6.6	1.1	64.6
	14	5	4.1	7.0	38.6	10.3	3.0	0.4	52.4	11.7	0.0	27.0	3.9	18.1	0.0	4.7	0.0	65.4
AeMBR undiluted	1	1	0.0	2.0	218.8	42.8	5.4	1.0	268.0	38.2	0.0	33.4	5.4	33.3	0.7	7.0	1.0	118.9
	2	1	0.1	2.5	326.4	52.6	5.8	2.6	387.4	50.6	0.0	55.2	8.1	87.0	0.7	18.4	1.7	221.7
	3	1	0.3	2.8	320.1	47.5	4.9	2.8	375.4	60.1	0.0	87.8	11.2	160.0	0.9	30.5	2.6	353.0

	4	1	1.4	3.0	377. 9	51.7	5.3	3.0	437. 8	56.4	0.0	137. 0	13.7	180. 8	0.9	35.4	2.9	0.0	427. 1
	5	1	4.4	3.1	408. 0	50.0	4.9	4.0	466. 9	47.2	0.0	160. 1	14.5	188. 6	0.9	36.2	3.0	0.0	450. 5
	6	1	5.4	3.2	383. 5	45.3	4.1	3.6	436. 6	36.4	0.0	276. 5	17.3	217. 7	1.0	42.6	3.5	0.0	595. 0
	7	1	6.3	3.3	521. 1	62.7	5.7	5.1	594. 5	33.1	0.0	264. 2	17.7	210. 9	0.9	42.6	3.4	0.0	572. 9
	8	1	6.8	3.5	448. 0	52.3	4.7	1.9	506. 8	31.3	0.0	266. 7	16.6	210. 9	0.9	41.5	3.3	0.0	571. 1
	9	1	7.4	3.8	407. 0	44.7	3.9	1.4	457. 0	29.5	0.0	283. 5	16.1	205. 5	0.9	40.7	3.2	0.0	579. 4
	10	1	8.2	4.0	574. 7	65.2	5.6	1.7	647. 1	29.8	0.0	302. 7	17.8	211. 0	1.0	40.9	3.2	0.0	606. 4
	11	1	11.0	4.2	267. 2	46.3	6.5	1.6	321. 6	19.5	0.0	110. 9	14.9	121. 3	1.7	30.0	3.5	0.0	301. 9
	12	1	15.3	4.5	316. 2	53.1	7.3	1.5	378. 0	22.1	0.0	135. 6	17.0	146. 4	1.9	37.9	4.4	0.0	365. 3
	13	1	18.9	5.0	634. 1	58.8	4.5	4.2	701. 5	33.3	0.0	537. 1	22.5	280. 5	1.3	53.7	4.1	0.0	932. 5
	14	1	21.3	5.2	260. 5	40.3	5.2	1.3	307. 3	17.6	0.0	120. 4	13.5	114. 2	1.7	25.6	2.9	0.0	295. 9

5 UV/TiO₂ photocatalysis as a post-treatment option for anaerobic membrane bioreactor effluent in the context of reuse

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Abstract

UV/TiO₂ photocatalysis has been widely applied as a post-treatment solution to offset the insufficient removal of organic compounds in modern water reuse treatment trains. However, there has been less emphasis on its use to treat anaerobically treated effluents and in particular membrane bioreactor (AnMBR) permeate with respect to removal mechanisms for organic and nutrient compounds, as well as the impact of operational conditions. In this study, the removal of organics and nutrients (nitrogen and phosphorus) from an AnMBR effluent is evaluated during adsorption and photocatalysis processes under various conditions of TiO₂ dose and UV intensity. Results are compared to those obtained with the effluent from an aerobic membrane bioreactor (AeMBR). As a post-treatment process, UV/TiO₂ photocatalysis offers a promising solution to target the removal of organic matter or nutrients from AnMBR effluent and provides a tuneable solution to balance the catalyst dosage and energy consumption for the AeMBR effluent to manage operational cost. Results highlight several challenges emanating from the use of UV/TiO₂ as a single post-AnMBR treatment process to achieve appropriate final effluent quality for reuse. The competing effect between the organics and nutrients, along with the low UV transmission efficiency caused by the need for high doses of TiO₂, ultimately compromise the organic removal efficiency. TiO₂ dosage was observed to have a greater impact than UV intensity on improving the overall removal performance due to the fact that nutrients are competing for the limited adsorption site but not photodegraded hence reducing the removal efficacy for organic compounds

removal. Under the same operational condition, the UV/TiO₂ photocatalysis displayed a higher removal efficiency of organic matter and phosphorus in the AeMBR effluent due to a lower initial organics concentration and absence of ammonia as compared to the AnMBR effluent. The identified adsorption-photodegradation trade-off effect between the TiO₂ dosage and UV intensity suggests that pre-removal of nutrient compounds can be beneficial to reduce the overall operational expenditure.

Keywords:

UV/TiO₂; Anaerobic and aerobic membrane bioreactor; Adsorption isotherm; Organic matter and nutrient removal; Dissolved organic matter

5.1 Introduction

Global water scarcity has led to the pursuit of sustainable and safe water sourced from treated wastewater (Lazarova, 2015; Salgot & Folch, 2018). In this regard, advanced treatment technologies, such as reverse osmosis (RO) membrane filtration, ion exchange (IEX), as well as advance oxidation processes (AOPs) have been widely applied to tackle the residual fractions of organic compounds in the secondary effluent from conventional wastewater treatment trains (Rebelo et al., 2018; Salgot & Folch, 2018; Tang et al., 2018b). However, organic contaminants that present a neutral charge or have a low molecular weight (molecular size <300 Da) are poorly removed by the RO and IEX (Finkbeiner et al., 2018; Mangalgiri et al., 2019), some of which have been reported to cause endocrine and toxicological responses in human cells (Li et al., 2018). In contrast, AOPs are extensively proven as effective tools to remove the excess organic compounds in particular for water reuse applications, with numerous studies evidencing a successful elimination of organic micro pollutants (OMPs) including pesticides and pharmaceuticals (Kanakaraju et al., 2018; Suzuki et al., 2015; Villegas-Guzman et al., 2017). AOPs can be classified as photochemical or non-photochemical in form. Examples of the photochemical group are direct photolysis by UV light, UV/H₂O₂, UV/TiO₂, photo-Fenton and photo-Fenton-like processes, while non-photochemical AOPs include ozonation and the Fenton process (Collins, 2016). Heterogeneous photocatalytic systems are widely reported as more sustainable and effective over homogeneous-phase options (Atalay & Ersöz, 2020; Villegas-Guzman et al., 2017). From these, titanium dioxide (TiO₂) has been reported as a physically robust and relatively nontoxic material (Levchuk & Sillanpää, 2020), with inexpensive overall operation cost due to the regenerable catalyst and applicable solar power (Loeb et al., 2019).

Anaerobic membrane bioreactors (AnMBRs) offer the potential of treating municipal wastewater with a lower footprint, energy neutrality, and less waste sludge production (Shin & Bae, 2018; Song et al., 2018). However, AnMBRs deliver limited organic removal performance under non-favourable operating conditions, such as low hydraulic retention time (HRT) (Chen et al., 2015; Mei et al., 2017) or psychrophilic temperatures (Crone et al., 2016; Dolejš et al., 2017).

This results in an inadequate effluent quality to meet the stricter organic parameter threshold in reuse standards (Augsburger et al., 2021; Foglia et al., 2019). Furthermore, as nutrients (ammonia and phosphorus) are not removed through the anaerobic digestion (biological) and membrane filtration (physical) stages in AnMBRs, there are major concerns about their ability to deliver in the context of high grade water reuse applications such as groundwater recharge or surface water augmentation (CNEPA, 2021; Harb & Hong, 2017; USEPA, 2017). For discharge and more importantly reuse, the AnMBR effluent typically requires a post-treatment process to eliminate the residual organic fraction including trace micropollutants and possibly nutrients (CNEPA, 2021; ISO, 2018; USEPA, 2012, 2017). Photocatalytic TiO₂ offers a promising post-treatment solution for AnMBR effluent through adsorption and elimination of organic matter and ammonia (Lee et al., 2002; Maghsoodi et al., 2019; Snow et al., 2019; Zhang et al., 2021), while adsorbing the phosphorus (Lee et al., 2016), to potentially achieve a high effluent quality and resource recovery for water reuse implementations.

Existing literature has largely focused on the post-treatment of effluents from aerobic biological systems but with relatively few studies on aerobic membrane bioreactor (AeMBR) effluents. However, the combination of AeMBR + UV/TiO₂ has been shown to produce an effluent quality sufficient for recycled greywater for toilet flushing with a high removal of major pharmaceuticals (Ojobe et al., 2021) while achieving a higher oxidation rate for selected pharmaceuticals in comparison to photo-Fenton, UV/Fe and UV/H₂O₂ (Benitez et al., 2011). However, unremoved organic matter can inhibit the photocatalytic process through hindering photocatalytic formation of OH· radicals by particle aggregation (Janssens et al., 2019), or extinguish the sorption onto the TiO₂ surface hence no reaction with surface bound OH· radicals (Maghsoodi et al., 2019), as well as decreasing the affinity of DOM to the catalyst surface (Snow et al., 2019), which ultimately leads to a strong quenching effect of reactive oxygen species (ROS). Furthermore, literature sources suggest that the water matrix has a significant impact on the degradation and mineralization efficacy by affecting the compound adsorption, surface radical generation and UV transmission efficiency (Kanakaraju et al., 2018; Levchuk & Sillanpää, 2020; Snow et al., 2019; Villegas-

Guzman et al., 2017). In our previous work with AnMBR and AeMBR pilot systems (Chapter 3 and Chapter 4), a clear difference in organic removal and nitrogen species was observed between the two effluents, where the COD and DOC of the AnMBR effluent was 2-4 times higher than AeMBR effluent, while ammonia and nitrate were the unique nitrogen forms in the AnMBR and AeMBR effluents, respectively. Moreover, very limited report has been shown in applying AOP as the post-treatment step for AnMBR effluent. Augsburg et al. (2021) proposed that UV/H₂O₂ can be the final polishing and disinfection process while avoiding additional steps for the removal of nutrients, producing a high quality effluent as liquid fertilizer for crops production. However, no study has investigated the potential application of UV/TiO₂ as the post-treatment for AnMBR effluent.

The current study then reports the first investigation of the application of UV/TiO₂ to an AnMBR effluent, to understand the impact of operational conditions and nutrient content on the photocatalysis process for the post-treatment of AnMBR effluent for reuse. The removal of organic matter and nutrients (nitrogen and phosphorus) in the AnMBR effluent is evaluated during the adsorption and photocatalysis processes, under various TiO₂ doses and UV intensities with a direct comparison to an AeMBR, to evaluate the potential to achieve sufficient effluent quality and effective large scale applications for sustainable water recycling.

5.2 Materials and methods

5.2.1 Pilot scale AnMBR and AeMBR

AnMBR and AeMBR pilot plants were installed at Cranfield University's National Research Facility for Water and Wastewater Treatment and operated stably under ambient temperature for 55 weeks. The systems were fed with settled municipal wastewater from the Cranfield University sewage works after settling in a primary settling tank. The AnMBR was configured as a cylindrical upflow sludge blanket bioreactor (UASB-45L) and a side stream membrane tank (30L). The AeMBR was configured with a level control tank (10L), aerobic tank (40L) and membrane tank (18L). For both systems, a PVDF hollow fibre ultra-filtration

(UF) membrane with 0.93 m² surface area and 0.04 µm nominal pore size (GE ZW-10, USA) was placed in the membrane tank and operated with a flux of 10 LMH. More details about the MBRs can be found in Chapter 3. Effluents used in adsorption and photocatalytic tests were sampled simultaneously from the AnMBR and AeMBR to minimize the effect on the effluent matrix caused by variation of influent wastewater. Characterization of the effluents is listed in Table 5-1.

Table 5-1 Characterization of AnMBR and AeMBR effluents fed to the photocatalytic reactor

mg/L	pH	TSS	Coliforms*	COD	DOC	NH ₃ -N	NO ₃ -N	TN	PO ₄ -P	TP
AnMBR	7.22	ND	ND	68	28.02	43.96	ND	44.56	6.91	6.91
AeMBR	7.29	ND	ND	18	6.75	ND	33.02	33.09	5.22	5.22

Notes: Coliforms*: Faecal, Escherichia and total coliforms; ND: Non detectable.

5.2.2 TiO₂ adsorption tests

TiO₂ nano powder (CAS: 13463-67-7) with a 21 nm primary particle size and ≥99.5% trace metals basis was obtained from Sigma-Aldrich (Merck, USA). Under dark conditions, 500 ml samples of MBR effluent were stabilized to room temperature (20 °C) then transferred to a 1L beaker (Fisher scientific, UK) to which the TiO₂ was added. A constant 400 RPM stirring rate was maintained with a magnetic stirrer (Benchmark 3770, UK) to mix the TiO₂ powder within the MBR effluents. Prior to the adsorption test, blank tests were carried out with no TiO₂ addition to the AnMBR and AeMBR effluent. Adsorption isotherm tests were conducted with TiO₂ dosing concentrations of 100, 200, 500, 1000, 2000 and 5000 mg/L. A 15 ml sample was taken from the beaker with a syringe at regular intervals over the duration of the tests to monitor the adsorbate concentration. Adsorption isotherms were described with the Langmuir isotherm (Equation 5-1), Freundlich isotherm (Equation 5-2) and a normalised Freundlich isotherm (Li et al., 2002) (Equation 5-3).

$$\frac{1}{q} = \frac{1}{q_m} + \frac{1}{bq_m C_e} \quad (5-1)$$

$$\log q = \log K_f + \frac{1}{n} \log C_e \quad (5-2)$$

$$q = K_f' \left(\frac{C_e}{D_0} \right)^{\frac{1}{n}} \quad (5-3)$$

Where, q (mg/g) is the amount of material adsorbed per unit weight of adsorbent, C_e is the concentration of material remaining in the solution (mg/L). In equation 1, b and q_m are constants. These constants can be determined by plotting $\frac{1}{q}$ vs $\frac{1}{C_e}$; While in equation 2, K_f and n are the Freundlich parameters which can be determined by plotting $\log C_e$ against $\log q$; and in equation 3, D_0 represents the dose of adsorbent which is applied to normalize the remained concentration.

5.2.3 UV/TiO₂ tests

UV assisted photocatalysis tests were conducted with a lab-scale quasi-collimated beam apparatus (Herford, Germany) equipped with three monochromatic low pressure UV-C (254 nm) lamps placed in a dark room (Figure 5-1). The UV irradiance was determined as 30, 40 and 50 W/m² with the correspondence distance between the lamps and the petri dish of 17, 12 and 8 cm, respectively. The irradiance intensity was determined by the potassium ferrioxalate method adapted from Hatchard, Parker and Bowen (1956). The detailed system setup can be obtained from Carra et al. (2016).

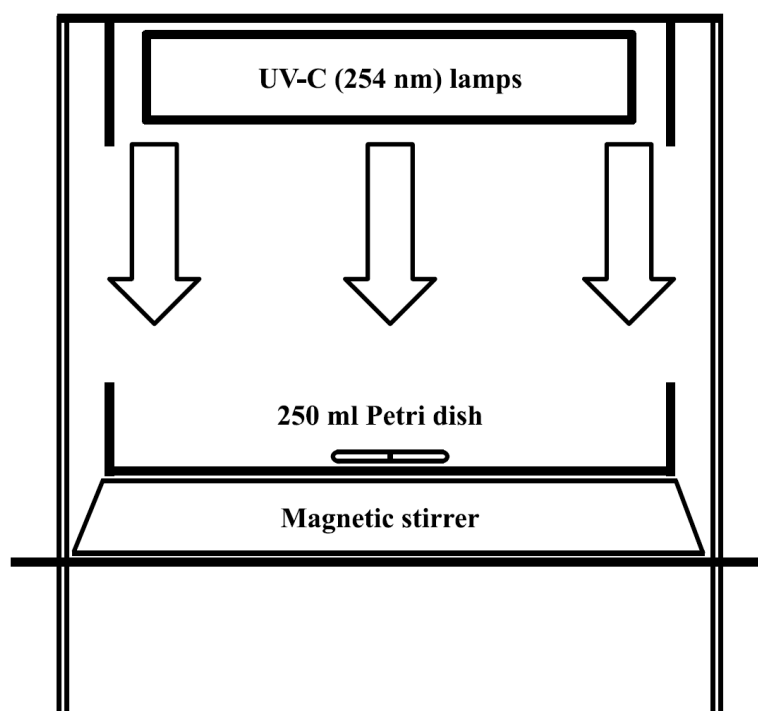


Figure 5-1 Diagram of the collimated beam

Under dark conditions, room temperature MBR effluent was transferred to a 250 ml petri dish and placed on a magnetic stirrer (Benchmark 3770, UK) under the UV lamps. The mixing of the TiO_2 powder and the solution was maintained by a 5 cm long magnetic stirring bar (Fisher scientific, UK) with a constant 400 RPM stirring rate to overcome the reduction of the photocatalysis efficiency caused by the poor mixing of the solution reported in Carra et al. (2016). Each test was done in duplicate with a total irradiance time of 300 mins, a 15 ml monitoring sample was taken from the centre of the petri dish at the reaction times of 10, 30, 60, 120, 180, 240 and 300 min. To calibrate the change in concentration caused by temperature increase from UV radiation, an evaporation coefficient was calculated with the TN concentration measured during the UV/ TiO_2 photocatalysis of the AeMBR effluent since there was no TN removal observed. The coefficient was calculated as the instant TN concentration divided by the initial TN concentration. The coefficients for 30, 40 and 50 W/m^2 were calculated as a function of the respective UV irradiation intensity, then applied to each measured parameter under the same intensity for both AnMBR and AeMBR samples. A summary of the test conditions is reported in Table 5-2.

Table 5-2 UV/TiO₂ test scheme for this study

Batch	Effluent	TiO ₂ dosage (g/L)	UV intensity (W/m ²)
1	AnMBR AeMBR	0.5	30, 40, 50
		1	30, 40, 50
		2	30, 40, 50
2	AnMBR	5	30
		10	

The photocatalytic process for organic matter and nitrogen was described with a pseudo first order expression (Equation 5-4) and the adsorption process for phosphorus was described by a pseudo second order kinetic model (Equation 5-5) which has been widely applied in documented literatures (Carra et al., 2016; Lee et al., 2016; Li et al., 2008; Murgia et al., 2005).

$$\ln \frac{C_0}{C_e} = k_1 t \quad (5-4)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5-5)$$

Where in equation 4, C_0 (mg/L) is the initial concentration of material in the solution, C_e (mg/L) is the concentration of material remaining in the solution at t time, k_1 is the rate constant of pseudo first order kinetic, and t (min) is the time consumed; while in equation 5, k_2 (g/mg·min⁻¹) is the rate constant of pseudo second order kinetic, q_t and q_e (mg/g) are the adsorption capacity at time t and at equilibrium, respectively.

5.2.4 Analytical measurements

Chemical oxygen demand (COD), ammonia nitrogen (NH₃-N), nitrite (NO₂-N), nitrate (NO₃-N), phosphate (PO₄-P), and total phosphorus (TP) were analysed with cell tests and a photo spectrometer (Merck Spectroquant, Germany). Dissolved organic carbon (DOC) and total nitrogen (TN) were measured by a Shimadzu TOC analyser (Shimadzu 3201, Japan). Particle size distribution was measured by a laser diffraction particle size analyser (Mastersizer 3000, Malvern Instruments, UK). Characterization of the samples was undertaken after filtering with 0.45 µm syringe membrane disc filter (Merck Millipore, Germany) to remove

the TiO₂ particles. Organic composition was evaluated by a fluorescence spectrometer (HORIBA FLuoroMax+, Japan) with a 3D excitation–emission matrices. Scan settings were 200–400 nm excitation and 280–500 nm emission wavelength, 1 nm entrance slit and an integration time of 0.1 second. The 1st and 2nd Rayleigh scatter was masked with 5 nm slit width by FluorEssence software (HORIBA, Japan). The fluorescence integrated composition analysis was extracted with a 5 region DOM categories method (aromatic protein-like (AP): tyrosine-like (APr) excitation/emission (E_x/E_m) = (200–250) nm/(280–330) nm; tryptophan-like (APy) E_x/E_m = (200–250) nm/(330–380) nm; fulvic acid-like (FA) E_x/E_m = (200–260) nm/(380–5000) nm; soluble microbial products (SMP) E_x/E_m = (250–280) nm/(310–380) nm; humic acid-like (HA) E_x/E_m = (280–380) nm/(380–500) nm) (Chen et al., 2003; Pan et al., 2021) using an R script modified from Lapworth and Kinniburgh (2009).

5.3 Results and discussion

5.3.1 TiO₂ adsorption isotherms

5.3.1.1 Performance on DOC removal

No DOC or TN reduction was observed in the blank test for the AnMBR and AeMBR effluents confirming that any removal observed in the presence of TiO₂ can be attributed to adsorption on the media. Limited DOC adsorption (< 20%) was found in all AnMBR (Figure 5-2.a) and AeMBR (Figure 5-2.d) effluent samples even when the TiO₂ dose was increased. Also, no clear correlation was observed between the adsorption performance and the TiO₂ dose in both MBR effluents. However, it should be noted that the dissolved organic carbon (DOC) is calculated as the difference between the total carbon (TC) and inorganic carbon (IC) and, interestingly, the removal profiles of TC (Figure 5-2.b and 5-2.e) and IC (Figure 5-2.c and 5-2.f) for the two effluents displayed a distinguishable removal linked to the TiO₂ dosing concentration. The C_e/C_0 values >1 observed for the DOC adsorption from the AnMBR effluent with a 2 g/L TiO₂ dose and under most conditions with the AeMBR effluent, are due to different adsorption rates of the TC and IC fractions by TiO₂. In comparison, the TC removal profiles in the AeMBR effluent displayed no correlation with the TiO₂ dosage, whilst IC showed

the same trend as observed in the AnMBR profile. For example, at equilibrium, C_e/C_0 ratios of 0.95, 0.90 and 0.90 were observed for TiO_2 doses of 0.2 g/L, 0.5 g/L and 0.1 g/L, respectively. This can be explained by the fact that the high molecular weight (MW) carbon substances have a higher adsorption potential by TiO_2 (Benkoula et al., 2015; Thomas & Syres, 2012). Our previous works has suggested the organic matter matrix is similar in the two effluents with a high content of humic acid-like substances (Chapter 3 and Chapter 4), while the size distribution results displayed a negligible difference in D_{v50} (size point below which 50% of the material is contained) between all samples with the same TiO_2 doses ($n=10$). Since the initial TC and IC concentrations in the AnMBR effluent were more than 4 times higher than for the AeMBR effluent, this suggests that the IC compounds in both effluents consist of high MW substances, while in contrast the other fraction of the TC has low MW. This variation leads to different adsorption rates in the isotherms and may ultimately lead to a different degradation rate during the photocatalysis process of the DOC and IC.

In terms of nutrients removal, no TN adsorption was identified in any of the AeMBR samples, while for the AnMBR, all TN adsorption profiles displayed a similar adsorption rate and capacity irrespective of the TiO_2 dose. The longer adsorption test with a 2 g/L TiO_2 dose in the AnMBR effluent did not reach equilibrium while keeping the same adsorption rate (Figure 5-2.g). Noticeably, the AnMBR effluent only exhibited a 0.15 ± 0.1 increase over the initial pH (7.22), suggesting there is no ammonia volatilization since the dominate ammonia fraction was ammonium under this pH and temperature (20 °C) (Shin et al., 2021; Venkiteswaran et al., 2019). This conclusion is further supported by the fact that no removal of ammonium was recorded in the blanks. Moreover, the unreach equilibrium observed for TN in the AnMBR effluent can explained by the triple bound adsorption of ammonium ions on the TiO_2 surface under the presence of H_2O ions (Markovits et al., 1996), which leads to the largest capacity observed over other adsorbates. For TP adsorption, the profiles of both effluents displayed a strong increasing adsorption rate and overall capacity with increasing TiO_2 dosage (Figure 5-2.h and 5-2.i), which agrees with Lee et al. (2016) that increasing the TiO_2 dosage can affect the overall adsorption capacity of

phosphorus. The adsorption of phosphate is evidenced to turn the zeta potential of the water dispersed TiO₂ surface from positive to negative (Domingos et al., 2010; Halimi et al., 2014), causing a charge neutralisation effect with the positively charged ammonium, as the adsorption of both ammonia and phosphate in the AnMBR effluent is enhanced. Therefore, this explains the slightly higher TP removal observed for AnMBR sample compared to that in the AeMBR sample conducted with the same TiO₂ dosage, but ultimately, this will reduce the adsorption capacity of the organics by taking the limited adsorption site on the TiO₂ surface. Hence the different nitrogen adsorption behaviour was caused by a different form of nitrogen in the two effluents; in the form of ammonium in the AnMBR effluent and nitrate in the AeMBR effluent. Adsorbed ammonia can potentially be oxidised by UV light (Murgia et al., 2005; Yu et al., 2021) and non-adsorbable nitrate is reported as non-degradable during the UV/TiO₂ photocatalytic process (Benkoula et al., 2015). However, the adsorption of the ammonia exhibited a completely different behaviour among all measured compounds as the adsorption capacity was not affected by the TiO₂ concentration. There is no clear explanation for this phenomenon and no evidence of a similar behaviour has been reported in the literature so further specific research may be needed for the mechanism to be fully explained.

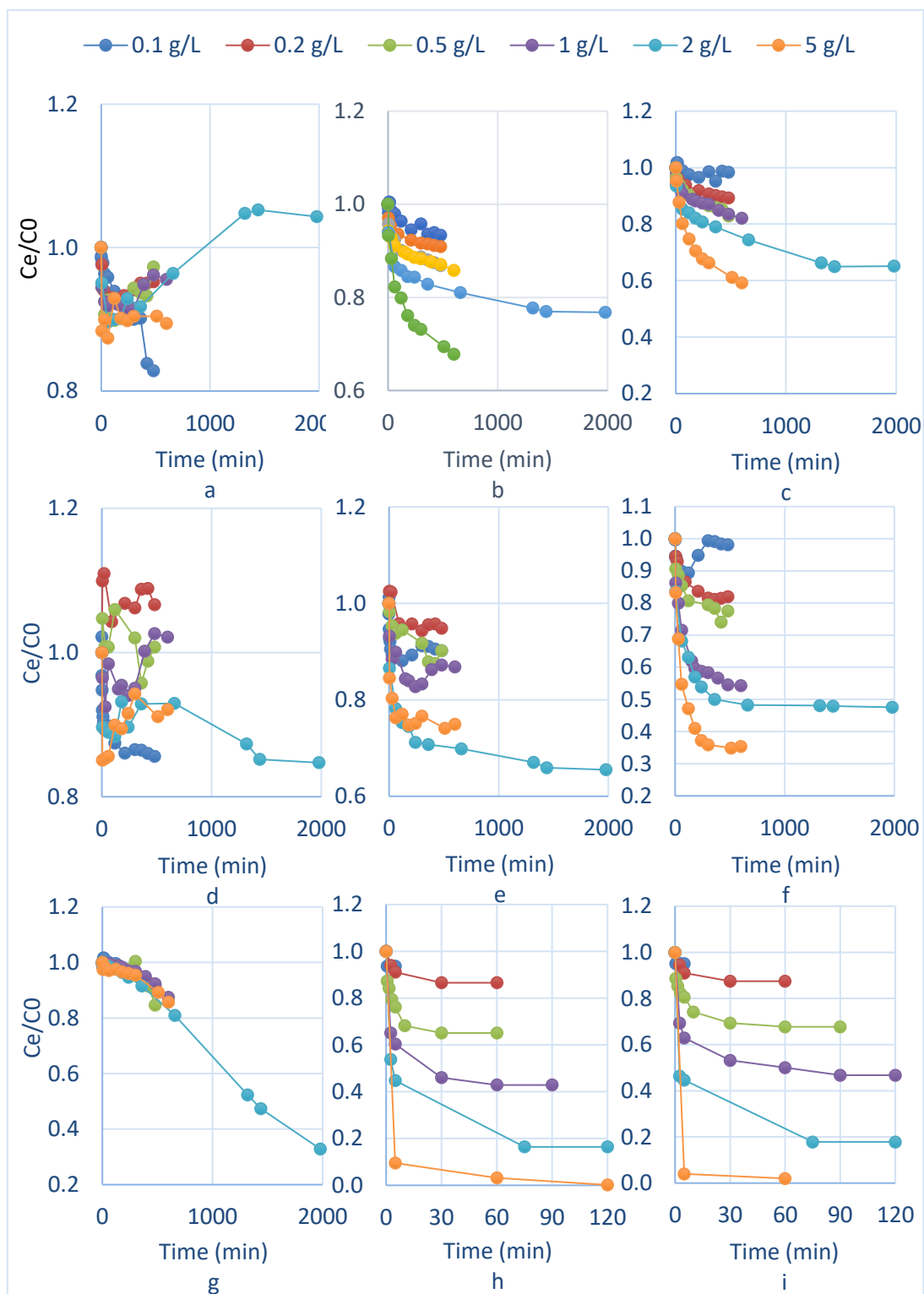


Figure 5-2 Adsorption profiles of various compounds in AnMBR (DOC-a, TC-b, IC-c, TN-g, TP-h) and AeMBR (DOC-d, TC-e, IC-f, TP-i) effluents by TiO_2 adsorption, where the value next to the legend is the correspondence TiO_2 dosing concentration

The corresponding isotherm parameter K'_f for all adsorbents displayed in Figure 5-3 were obtained with the normalised Freundlich isotherm as it presented the highest overall coefficient of determination value (R^2) among the three models applied (Supporting information 4-1). DOC was excluded due to poor correlation ($R^2 < 0.3$) observed in all fittings, therefore the displayed K'_f are for TC, IC, TP and TN (AnMBR only) with an average R^2 of 0.9 / 0.6, 0.9 / 0.9, 0.9 / 0.9 and 0.9, respectively for the AnMBR and AeMBR samples.

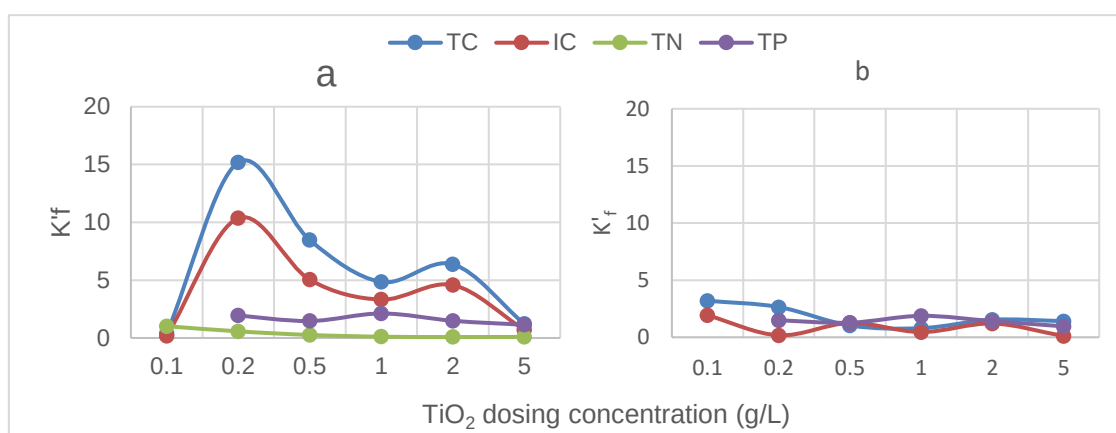


Figure 5-3 K'_f of each adsorbent from the normalised Freundlich isotherms for AnMBR (a) and AeMBR (b) effluent

As K'_f reveals the adsorption capacity for the compound per unit mass of dosed TiO_2 , this can provide an indication of adsorption efficiency at the corresponding dose. The K'_f value for TC and IC from the AnMBR effluent displayed an obvious variation at each TiO_2 dose, with the lowest K'_f observed for the lowest (0.1 g/L) and the highest (5 g/L) doses. The low K'_f value obtained for the low dose is due to the limited overall adsorption capacity, while the low K'_f value at the high dose is due to the limited increase in K'_f once the adsorbate reached equilibrium as the residual concentration in the samples remain stable. This result agrees with Li et al. (2002) who showed that the adsorption capacity is less affected by increasing the adsorbent dosage in high initial adsorbate concentration solutions, which in this case is the AnMBR effluent. In contrast, the AeMBR samples displayed much less variation with similar K'_f values regardless of the dosed amount. Indeed, the K'_f values for TC and IC for the AnMBR effluent were higher than for the AeMBR

effluent with TiO₂ doses of 0.2, 0.5, 1, and 2 g/L. This agrees with the findings reported in the literature that a higher unit adsorption capacity is expected in the more concentrated solution under the same adsorbent dose (Lim et al., 2011; Thomas & Syres, 2012). Moreover, the K'_f values for TC and IC were 15.2 and 10.4 in the AnMBR effluent with 0.2 g/L TiO₂, which is significantly higher in comparison to the corresponding values of 2.6 and 0.18 for the AeMBR effluent. Despite the different initial compounds concentration, the nitrogen species produce by the different biological stage of two MBRs also impact differently on the adsorption of organics. As discussed above, the ammonium in the AnMBR effluent could have a charge interaction effect with phosphate, competing for the available adsorption site on the TiO₂ surface, which leads to a decreased adsorption capacity for the organics. Whilst nitrate has been shown to decrease NOM adsorption on TiO₂ (Gora & Andrews, 2017), along with the competing effect from phosphate, this explains the significantly lower K'_f found for the AeMBR effluent even with a much lower initial compounds concentration compared to AnMBR effluent. Furthermore, since the performance of the photocatalysis process is influenced by the adsorption rate (Azeez et al., 2018; Levchuk & Sillanpää, 2020), this suggests a different organic removal performance should also be expected in the photocatalysis tests.

The K'_f of TN in the AnMBR effluent displayed a slightly reducing trend with increasing TiO₂ dose, while the K'_f for TP showed a slightly higher capacity at each dose. This could again be explained by the adsorption priority due to the different MW of ammonium and phosphate, which are 18.04 g/mol and 94.97 g/mol, respectively. The higher K'_f for TP compared to TN in the AnMBR effluent confirmed the charge neutralisation effect between the ammonium and phosphate. A similar K'_f of TP was observed in both MBR effluents when the same dose was applied. This confirmed these two effluent matrices had a negligible impact on the adsorption mechanism of the phosphorus compound, but may suggest there is no optimum TiO₂ dosing ratio for the MBR effluents to prioritise for phosphorus adsorption. However, phosphate reached an adsorption equilibrium within the shortest time compared to the other substances,

suggesting that phosphate has the highest adsorption rate due to a higher MW and a positive charge compared to other compounds in the effluent. As there is no evidence reported in the literature of the photodegradability of phosphate by UV/TiO₂, this may be a cause for concern for the application as the overall photocatalysis efficiency will be reduced when phosphate occupy adsorption sites on the surface of the TiO₂ but is not removed photocatalytically.

Overall, the results from the adsorption tests indicated poor removal of organics but better performance on ammonia and phosphate from adsorption alone, highlighting the need to add UV irradiation for application of photocatalysis. Furthermore, the different matrices of AnMBR and AeMBR effluents will lead to different behaviours during the photocatalysis process, whereas a more complex process should be expected for AnMBR since more organic and nutrient compounds are present and competing for adsorption sites. The adsorption priority of each compound is concluded as phosphorus - inorganic carbon - ammonia / organic carbon in the AnMBR effluent, whilst phosphorus - inorganic carbon - organic carbon in the AeMBR effluent.

5.3.2 UV/TiO₂ photocatalysis performance

It should first be noted that there was no removal of DOC, TN and TP from either MBR effluent with direct UV photolysis. Notably, the pH value in all samples showed a minor change (< 0.1) after the addition of TiO₂ but remained stable during the whole progress of the photocatalysis process, hence volatilisation was not considered as a pathway for ammonia removal from the AnMBR effluent.

5.3.2.1 Removal of bulk organic and nutrients

The DOC removal profile of the AnMBR effluent exhibited significant variations corresponding to changes in TiO₂ dose and UV intensity (Figure 5-4.a). The relative linearity of these profiles indicates a steady rate of removal over time for all tested operational conditions, whereas the general trend suggests that the TiO₂ dose has a bigger impact than the UV intensity on DOC removal. To illustrate, the DOC removal observed for the 0.5 g/L TiO₂ dose under the three tested UV intensities of 30 W/m², 40 W/m² and 50 W/m² were 18, 31 and 11%

respectively. However, at a UV intensity of 30 W/m^2 , the DOC removal increased from 18% to 34% and 44% when the dose was increased to 0.5, 1 and 2 g/L, respectively. The greater impact of dose over intensity was further confirmed by a sharper reduction gradient in the removal profile and a higher overall removal observed for the TiO_2 doses of 5 g/L (60%) and 10 g/L (79%) under the UV intensity of 30 W/m^2 .

Similarly distinctive trends were observed for the AeMBR effluent when exposed to different TiO_2 doses and UV intensities (Figure 5-4.b). At 0.5 g/L TiO_2 dose, removal percentages of 48%, 59% and 70% were observed for the corresponding UV intensities of 40, 30 and 50 W/m^2 . These three data series displayed a similar linearity to the AnMBR samples, indicating that a steady DOC removal rate was experienced over the 300 minutes experiment period. A steady removal rate was also observed in all other profiles obtained with the operational conditions of higher TiO_2 doses and UV intensities. In the samples with 1 g/L and 2 g/L TiO_2 doses, the removal profiles exhibited a rapid decrease followed by stabilisation after reaching a removal percentage of 80%. Indeed, stability was achieved with 1 g/L and 2 g/L TiO_2 doses regardless of the UV intensity, while the samples with 2 g/L dose required 120 minutes and the 1 g/L dosing samples took 180 minutes. This highlights a trade-off effect between the catalyst dosage and energy consumption during the treatment of the AeMBR effluent. However, a maximum DOC removal of 80% was observed for both MBR effluents regardless of the operational conditions, suggesting the potential presence of a recalcitrant fraction of organics or the inhibition effect that adsorption sites were all covered with unreactive compounds. This may be caused by the colloid substances in the MBR effluents (Maghsoudi et al., 2019; Snow et al., 2019), as well as the nutrient compounds (Kim & Choi, 2002; Klare et al., 2000; Lee et al., 2016).

In comparison to the trends observed with the adsorption isotherm, the TN profiles for the photocatalysis of the AnMBR effluent displayed a similar shape (constant removal rate over time) for all tested conditions but exhibited a significant improvement on the removal rate constant (Figure 5-4.c). The average TN removal rate constant increased from 0.00025 min^{-1} to 0.0046 min^{-1} , this 1.5

log enhancement clearly demonstrates the benefit of the adsorption-photocatalytic process over monophonic adsorption. The overall removal percentage varies from 56% to 85% with a TiO_2 dosing amount from 0.5 g/L to 5 g/L, whereas the UV intensity was found to have a negligible effect on the removal rate and overall removal percentage. However, literature has shown the $\text{OH}^\cdot$ radicals can oxidize the ammonium to the forms of nitrite and nitrate in the pure ammonium solution, but suggested a very limited photodegradation by pure TiO_2 suspensions in a neutral pH condition (Lati et al., 1972; Murgia et al., 2005; Zhang et al., 2021). Indeed, we did not observe the formation of nitrite or nitrate in any trials. This agrees with Kim & Choi (2002) and Klare et al. (2000) that ammonium ions are not degraded under particular conditions similar to this paper, where the ammonium tend to be photodegradable as the fraction of ammonia at a pH of 11. Hence this suggests the removal of ammonia in our experiments was only by adsorption, and ultimately experiences a competing effect as for the phosphate. This therefore explains the lowest removal percentage was observed with the highest TiO_2 dose of 10 g/L, and the second lowest was the sample dosed with 5 g/L which suggests the TiO_2 dosage in these two tests may have exceeded the optimum amount for the reactor and the overdose of TiO_2 has led to a substantial reduction in light penetration, which significantly decreases the photodegradation of organics hence much less adsorption site could be released. This ultimately resulted in a lower removal efficiency for organic and nitrogen (ammonium) compounds. However, the non degradation of ammonia compounds highlights the potential selective removal by varying the UV intensity, and no further denitrification required since no nitrite or nitrate is generated, as well as a promising insight of ammonia recovery during the regeneration of TiO_2 .

All phosphate removal profiles displayed a short critical removal period regardless of the dosage, UV intensity and water matrix, whereas more than 90% of the phosphate was removed within 60 minutes with TiO_2 dosing of 5 and 10 g/L (Figure 5-4.d). The trends after the equilibrium point displayed a steady decrease but corresponding to a very limited removal percentage. Furthermore, the overall phosphate removal percentage displayed significant correlation to the TiO_2 dose, while again the UV intensity had a negligible impact. This agreed with

the findings from the adsorption isotherm that a maximum adsorption capacity is achieved for phosphate.

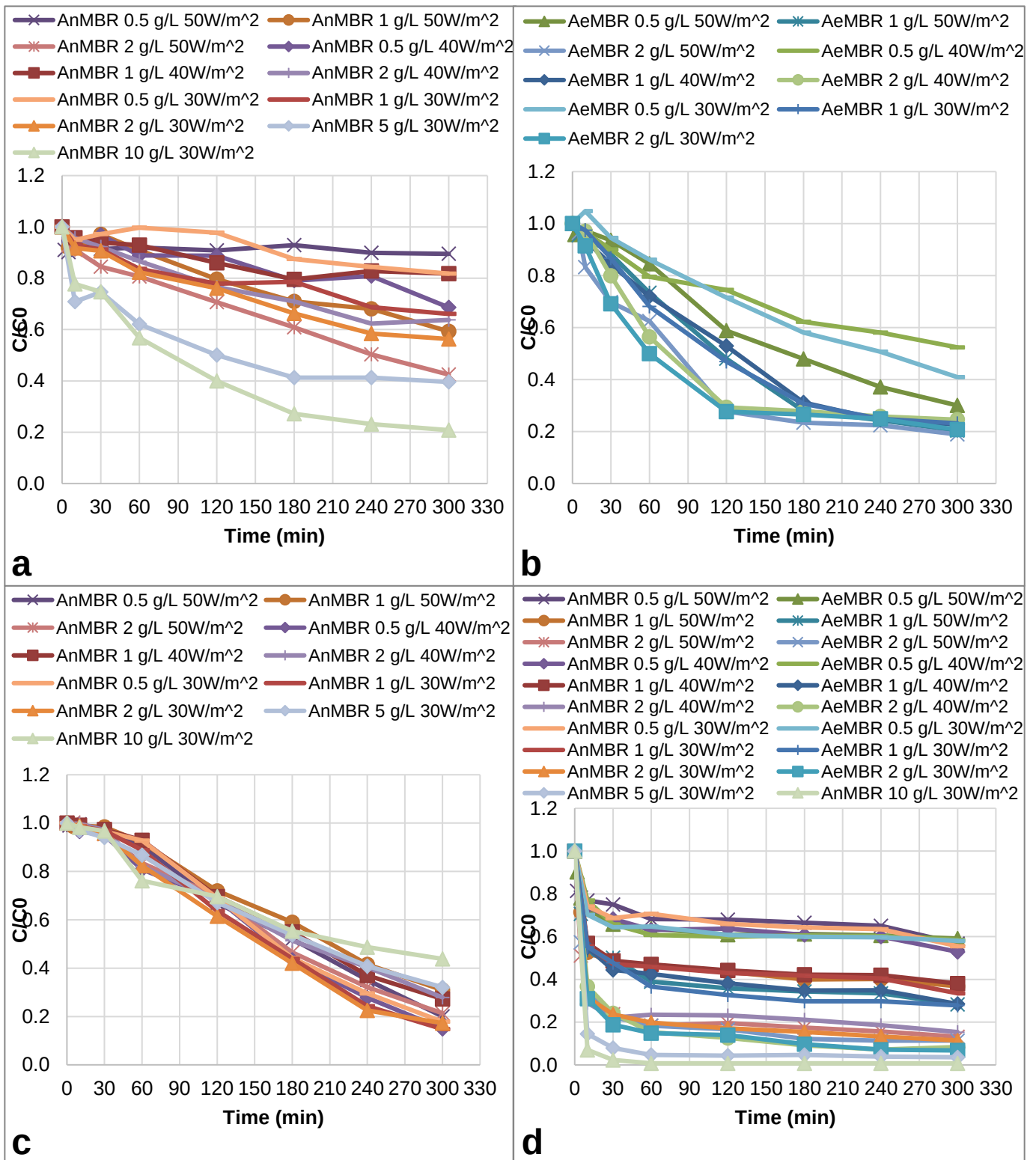


Figure 5-4 Removal profiles in AnMBR (a-DOC, c-TN, d-TP) and AeMBR (b-DOC, d-TP) effluents by UV/TiO₂, where the value next to the legend is the correspondence TiO₂ dosing concentration and UV intensity

Overall, the UV photocatalysis of the AeMBR effluent displayed a higher phosphate removal percentage in each sample compared to the percentage in AnMBR sample obtained with the same operational condition, which is different from the results observed without UV irradiation. This can be explained as a competing effect between the compounds which only adsorb and those which are photodegradable, that the mineralization of degradable compounds empty the adsorption site hence more phosphate can be adsorbed. The assistance of UV irradiation did slightly increase the overall removal rate for the 0.5, 1 and 2 g/L doses for both effluents but reduced in the AnMBR samples with 5 and 10 g/L doses. The reduced overall removal percentage observed with 5 and 10 g/L TiO₂ dose is respectively similar to the TN result in the same sample, as the overdose of TiO₂ reduces the light penetration hence less desorption of the degraded organics which in turns leads to less adsorption of the nutrients. This confirmed the finding in the adsorption tests that a high TiO₂:N/P ratio may not be the optimum dosage if the nutrients removal is set for the higher priority than organic matter. However, the identified catalyst-energy trade-off effect suggests that once the TiO₂ dose exceeds a certain TiO₂:N/P ratio to ensure there is still enough capacity for photodegradable substance after the adsorption of ammonium and phosphate, an increase in TiO₂ dose can speed up the removal of DOC regardless of the UV intensity, shortening the time required to reach the same overall removal percentage.

5.3.2.2 Removal kinetics

To better understand the removal of each compound during the photocatalytic process, the rate constant of total phosphorus displayed in Figure 5-5 was described by a pseudo second order kinetic model, while the rate constant of organic compounds and total nitrogen displayed in Figure 5-6 were both obtained using a pseudo first order expression. The average R² of each expression for the

AnMBR effluents was 0.95, 0.94, and 0.99 for DOC, TN, and TP, respectively; whilst it was 0.96 and 0.99 for DOC and TP for the AeMBR effluents.

As shown in Figure 5-5, the feed water matrix and UV intensity were found to have negligible impact on the rate constant of total phosphorus, while the increasing TiO_2 dosing concentration had a significant increase on K_{TP} . However, the K_{TP} of the AnMBR samples with 5 g/L and 10 g/L TiO_2 dosing concentrations agreed with the finding from the adsorption isotherm that a limited increase in adsorption capacity was achieved once the adsorbent reached a certain ratio (Li et al., 2002), which in this study was TiO_2 :TP weight ratio of 724:1. Moreover, the removal profiles in Figure 5-4.d exhibit a limited overall removal percentage and a limit removal after the equilibrium point, suggesting that the phosphorus present (as phosphate) can have a competing effect by adsorbing to TiO_2 at the start of the photocatalysis process but a negligible impact after reaching the equilibrium point in batch operation mode. However, if the system was operating in continuous mode, that phosphorus will not reach an equilibrium, therefore more phosphorus would be adsorbed until there is no capacity remaining on the TiO_2 . Hence phosphorus affects the UV/ TiO_2 process by proportionally reducing the adsorption capacity and the available reactive surface for radical generation, which may ultimately reduce the overall efficiency of the photocatalysis process. Therefore, pre-removal of phosphorus from the MBR effluents can reduce the required dosage of TiO_2 and simultaneously increase the efficacy of the photocatalytic process for the degradation of organic matter, which may be beneficial to reduce the overall operational expenditure.

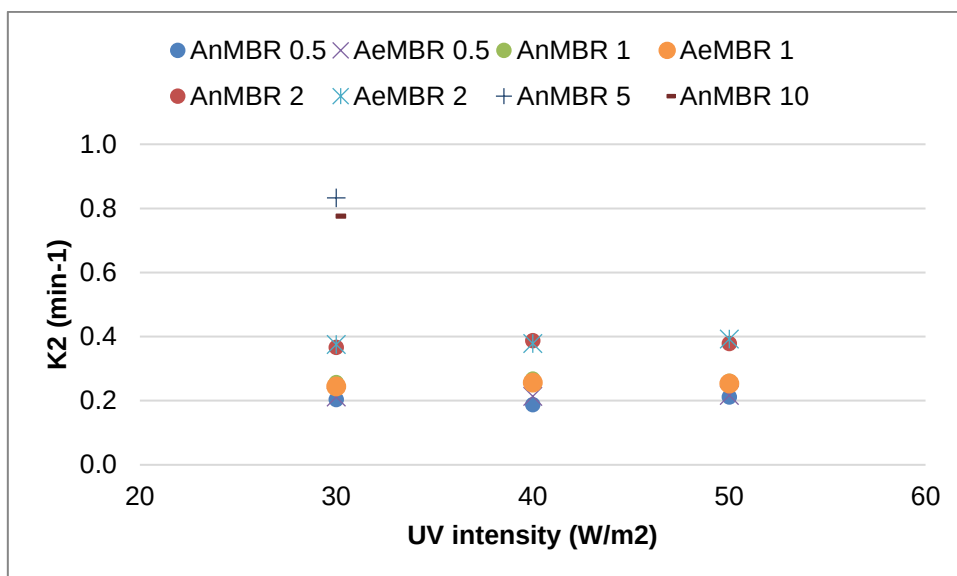


Figure 5-5 UV/TiO₂ photocatalysis rate constant for TP under various UV intensities in AnMBR and AeMBR effluent. Where, the value next to each legend entry is the corresponding TiO₂ dosing concentration in g/L.

As shown in Figure 5-6, the K_{DOC} value for the AnMBR effluent was lower than the value for the AeMBR effluent under the sample operational conditions except the samples with a 0.5 g/L TiO₂ dose and 50 W/m² UV intensity. This helps explain the low overall removal observed in Figure 5-4 for all AnMBR samples as they have much higher initial DOC concentrations compared to the AeMBR samples. This can be explained by the limited total adsorption capacity of TiO₂ under a low dosing concentration scenario. In this study the TiO₂:DOC for these samples were below 75:1. The explanatory mechanism here is that phosphate is taking the initial empty adsorption capacity, then continuously taking the released capacity after the degradation of organic compounds. Competition in the AnMBR effluent is more crucial since the ammonia was simultaneously following the pathway of adsorption, which ultimately leads to a reduction in the available photocatalysis area. Furthermore, the rate constant of K_{IC} was higher than K_{TC} in all AnMBR samples (Figure 5-6.a, 5-6.b, 5-6.c), whereas for the AeMBR samples, K_{TC} is higher than K_{IC} (Figure 5-6.d, 5-6.e, 5-6.f). Since the degradation mechanism of organic compounds by UV/TiO₂ is strongly dependent on the adsorption of the compounds (Maghsoodi et al., 2019; Snow et al., 2019), this validated the hypothesis that the other fraction of the TC, which contributes to

DOC substances in this study, consists of lower MW compounds compared to the IC compounds in both effluents. To explain, in the AnMBR effluent DOC needs to compete with IC, phosphate, as well as the ammonia, which further reduces the adsorption of DOC. Moreover, the initial DOC and IC concentrations are much higher in the AnMBR effluent compared to those in the AeMBR effluent, therefore the IC compounds with a higher MW experience a higher adsorption rate, resulting in a higher contact ratio with generated radicals (Carra et al., 2016), ultimately leading to a higher removal rate and consequently a lower K_{DOC} for the AnMBR. In comparison, in the AeMBR samples K_{TC} was consistently higher than K_{IC} , synchronously leading to a higher K_{DOC} . This additionally explains the faster DOC removal in AeMBR effluent compared to AnMBR.

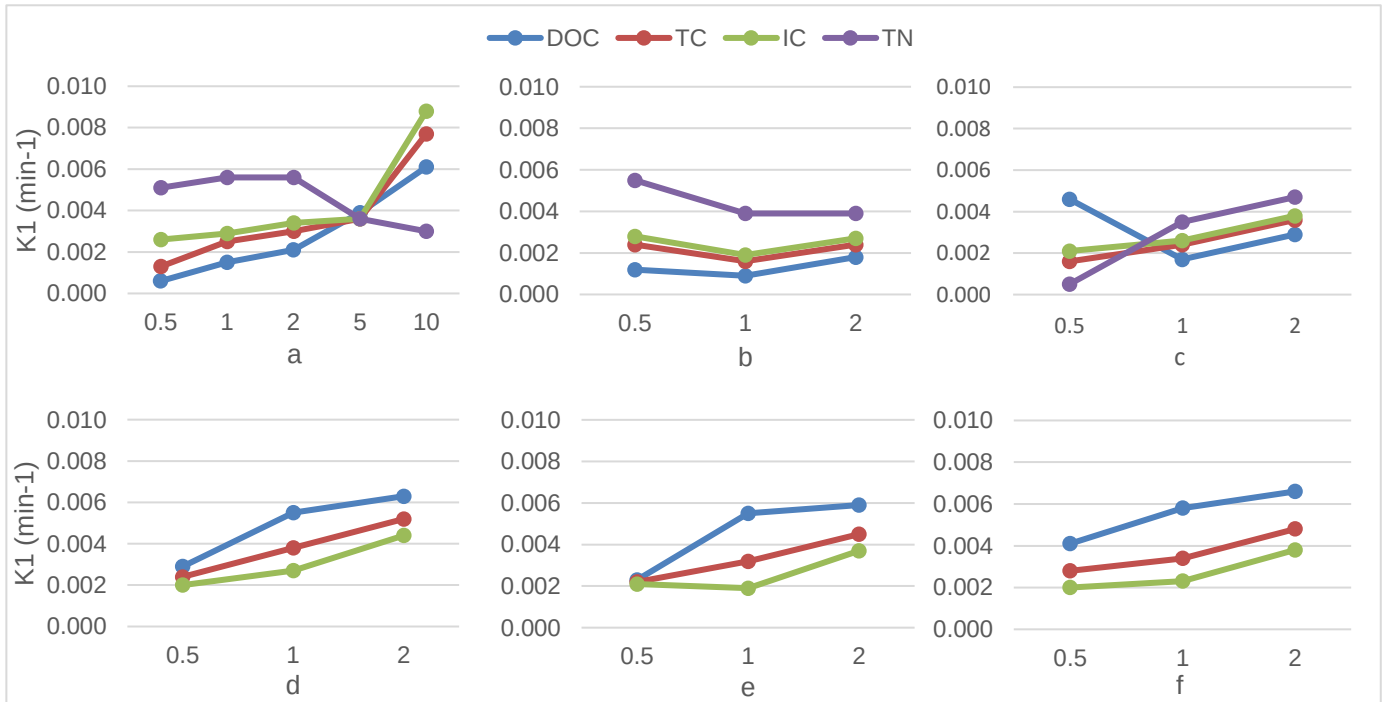


Figure 5-6 UV/TiO₂ photocatalysis rate constant under various UV intensities in AnMBR (a-30 W/m², b-40 W/m², c-50 W/m²) and AeMBR (d-30 W/m², e-40 W/m², f-50 W/m²) effluent. Where the X-axis is the TiO₂ dosing concentration in g/L.

In the sample taken at 0.5 g/L TiO₂ dose and 50 W/m² UV intensity, the K_{DOC} value (0.0046 min⁻¹) was much higher compared to two other AnMBR samples with 1 g/L (0.0017 min⁻¹) and 2 g/L (0.0029 min⁻¹) TiO₂ dosing concentration under the same UV intensity, while it is only slightly higher than the K_{DOC} value (0.0041

min⁻¹) of the AeMBR sample generated under the same operational condition. This was due to the fact observed in Figure 5-6.a, 5-6.b and 5-6.c that K_{DOC} was inversely proportional to K_{TN} in the AnMBR samples, indicating there might be a trade-off effect after the fast adsorption of phosphorus occurred between the removal of organic compounds and ammonia due to limited capacity of adsorption. Moreover, the organic-ammonia trade-off effect was observed in samples obtained both with a fixed TiO_2 dosing concentration while varying the UV intensity and with a fixed UV intensity with various TiO_2 dosing concentrations. In Figure 5-6. with a fixed UV intensity of 30 W/m², K_{TN} decreased with increasing dose, while in Figure 5-6.c the K_{DOC} decreased with increasing K_{TN} and dose at a UV intensity of 50 W/m². However, the removal mechanism is different between the samples displayed in Figure 5-6.a and Figure 5-6.c. This was due to the fact that in the particular conditions reported in this study, only DOC in the AnMBR effluent can be photocatalytically degraded. Therefore the removal rate obtained during the UV/ TiO_2 process involves the mechanisms of both adsorption and degradation for DOC, but only adsorption for the ammonia. In the samples exposed to 30 W/m² (Figure 5-6.a), the relatively lower UV intensity led to an adsorption dependent removal of DOC rather than photodegradation. To explain, at low TiO_2 doses (< 1 g/L) the UV irradiation offered a limited promotion of the degradation due to a lower adsorption capacity, therefore the competing effect between the nutrients and organics had led to a higher K_{TN} compared to K_{DOC} . Whilst in the samples treated with an increasing TiO_2 dosage from 1 g/L to 2 g/L, an increased capacity boosted the degradation of DOC and offered more adsorption capacity for both DOC and ammonia, which explains the increased K_{DOC} but constant K_{TN} . This also helps explaining the trend observed for the samples exposed to a UV intensity of 40 W/m², as the same trade-off effect between DOC and ammonia appeared as seen earlier in the samples exposed to 30 W/m². However, a further increase of the TiO_2 dosage (> 5 g/L) shifted the adsorption priority from nutrients towards the DOC, which resulted in a significant increase and decrease for K_{DOC} and K_{TN} , respectively (Figure 5-6.a).

In contrast, assuming a similar adsorption scenario of DOC and ammonia in the samples exposed to 50 W/m² (Figure 5-6.c), increasing the UV intensity from 30 W/m² to 50 W/m² significantly promoted the photodegradation of DOC, as well as restricted the adsorption of ammonia with an increasing temperature (Rouquerol et al., 1999; Striolo et al., 2005), ultimately resulting in a much higher K_{DOC} compared to other samples and the lowest K_{TN} based on the organic-ammonia trade-off effect. However, the cross point between the 0.5 g/L and 1 g/L dosing concentration in Figure 5-6.c may be the result of the competition effect discussed above, as the phosphate and ammonia took the released adsorption capacity whilst the degradation is much faster under a higher UV intensity, leading to a lower overall removal efficiency. This simultaneously explains the increased K_{DOC} and K_{TN} by increasing the TiO₂ dosage from 1 g/L to 2 g/L under the same 50 W/m² UV intensity. This performed as a co-effect of higher overall adsorption capacity and a faster degradation of DOC, creating much more adsorption capacity in comparison to samples that experienced a lower DOC degradation rate. Subsequently, this helps explain the different organic-ammonia trade-off trends observed between Figure 5-6.a and Figure 5-6.c since the adsorption rate and capacity of TN is not promoted by increasing the TiO₂ dose. Moreover, comparing the K_{DOC} and K_{TN} obtained at a fixed TiO₂ dosing concentration while varying UV intensity, increasing the UV intensity resulted in a reduction of K_{TN} but an increase of K_{DOC} . This suggests the adsorption of ammonia was less favoured than that of organic compounds by the increasing UV intensity. Therefore, this explains the similar overall DOC removal but a different removal rate constant observed for the AnMBR effluent treated by 5 g/L TiO₂ dose and 30 W/m² UV intensity (Figure 5-4.a) and with 2 g/L TiO₂ and 50 W/m² (Figure 5-4.c). This addresses the fundamental of the catalyst-energy trade-off effect which is linked to the adsorption-photodegradation trade-off effect, where similar removal efficiencies can be achieved by improving either of the adsorption and photodegradation processes. To explain, in the 5 g/L and 30 W/m² samples the large amount of dosed TiO₂ significantly enhanced the overall organics and nutrients removal by adsorption, whereas in the 0.5 g/L and 50 W/m² samples the intensive UV irradiation ensured a high level of photocatalytic degradation of

DOC then the emptied adsorption site boosted the overall removal of organic and nutrient compounds.

Due to the different nitrogen species (nitrate) present in the AeMBR effluent, the K_{DOC} obtained in the monophonic photocatalytic degradation of DOC was found to have more direct link to the operational conditions. As shown in Figure 5-6.d and Figure 5-6.f, a substantial increase in K_{DOC} was observed in the sample which used a combination of 0.5 g/L TiO_2 dosing concentration and 50 W/m^2 compared to the one with the same dose and 30 W/m^2 . However, a negligible increase of K_{DOC} was observed with a higher dosing concentration of 1 g/L and 2 g/L, while the increase of K_{DOC} by increasing the TiO_2 dosing concentration was limited. This was due to a low initial concentration of pollutants present in the AeMBR effluent; hence the increased TiO_2 dosage had limited impact in boosting the removal rate constant.

The ineffectiveness of increasing the UV intensity in the high dose samples (>1 g/L) may be caused by a limitation in the experimental setup used for this study. Degradation only occurred at the surface of the solution, where the UV irradiation contacts the TiO_2 , and the petri dish was therefore designed to have a large surface area to receive the UV irradiation from the lamps installed above (Figure 5-1). In the high dosage scenarios (where we observed that even the phosphate does not take all the absorption capacity) the overall available reacting surface area is too small due to the formed TiO_2 mask. This concomitantly leads to a limited effect through increasing the intensity of UV irradiation when a high TiO_2 dosing concentration is used. The increased removal rate found in higher TiO_2 dosing concentration samples was caused more by adsorption not degradation, which may also explain the cause of the adsorption-photodegradation trade-off effect found in AnMBR samples, as theoretically a higher K_{1c} should be expected with the operational condition of 5 g/L and 30 W/m^2 .

5.3.2.3 DOM removal

A similar DOM content was observed in the two MBR effluents, with an identical percentage of aromatic protein-like (AP) compounds with tyrosine-like (APr) compounds at 1%, tryptophan-like (APy) compounds at 1% and fulvic acid-like

(FA) compounds at 5%. The AnMBR effluent had a slightly higher fraction of soluble microbial products (SMP) of 8% caused by the less stable organic removal performance compared to 3% in the AeMBR effluent (Kunacheva et al., 2017). The fraction of humic acid-like (HA) compounds was 85% and 89% for the AnMBR and AeMBR effluents respectively. As DOC is a component of dissolved organic matter (DOM) (Moody & Worrall, 2017), monitoring the removal of DOM can also offer a comprehensive view of the overall photocatalysis process, as well as a potential view of the variation of primary halogenated DBP precursors (Bond et al., 2011; Ma et al., 2016). Therefore, the removal percentage of each measured DOM fraction was calculated with the fluorescence integrated composition analysis results (Figure 5-7).

The process of DOM fraction removal by UV/TiO₂ displayed non-selectivity with regard to each single type of DOM (Figure 5-7) regardless of the effluent type (water matrix) and operational condition (TiO₂ dose and UV intensity). The AnMBR (Figure 5-7.a) and AeMBR (Figure 5-7.b) effluents displayed a similar response in terms of the different UV intensities, whilst a relatively similar removal percentage was observed between the two effluents when conducted with the same TiO₂ dosing concentrations. Both effluents displayed a significant removal of HA (> 89%) regardless of the operational condition. The removal percentage of each DOM category confirmed the adsorption mechanisms to follow in decreasing order of the MW, whereas the higher removal is matched with the higher DOM MW (Li et al., 2002; Lim et al., 2011). Moreover, the negative removal found for AP_r and AP_y in both effluents may suggest the aromatic protein-like substances could be the mineralisation by-product of photocatalysis process. However, positive removal of AP_r and AP_y was then observed with the higher dose for AnMBR (5 mg/L, 10 g/L) and AeMBR (2 g/L) tests, suggesting the generated by-product AP could be removed by either high UV intensities with sufficient adsorption capacity, or exceed amount of adsorption capacity provided by the high TiO₂ doses.

For the AnMBR effluent, the increase of TiO₂ dosage and UV intensity were both associated with an increase in DOMs removal in the samples exposed to 0.5 - 2

g/L doses and 30 – 50 W/m² UV intensities. However, a non-significant increase in DOMs removal was then observed in the samples exposed to 5 g/L and 10 g/L TiO₂ dosage, suggesting that a further increase in the TiO₂ dose concentration resulted in no improvement in DOM removal percentage. In contrast, in the AeMBR samples, due to a lower initial organics concentration and hence lower competition for the radicals formed, the removal efficiency clearly indicated the corresponding operational condition. The highest overall removal rate was obtained with a 2 g/L TiO₂ dosage and 50 W/m². Interestingly, the combination of 1 g/L and 40 W/m² and 2 g/L and 30 W/m² achieved a similar removal performance but with a different contribution of degradation > adsorption and degradation < adsorption, respectively, suggesting a similar adsorption-photodegradation trade-off effect to that observed for the DOC and nutrients removal.

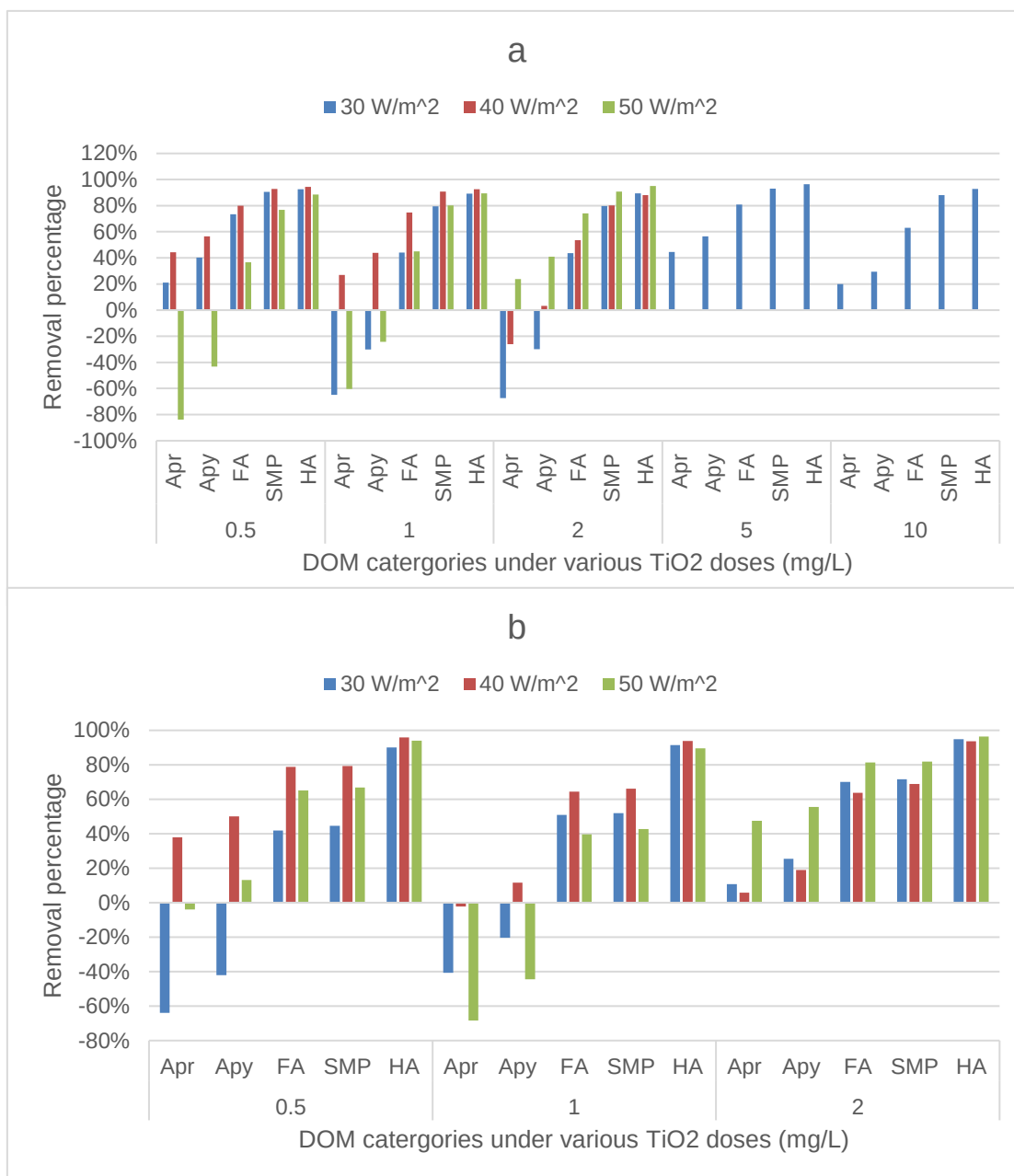


Figure 5-7 DOM removal by UV/TiO₂ in AnMBR effluent (a) and AeMBR effluent (b) under various TiO₂ doses and UV intensities. Where: Apr - tyrosine-like; Apy - tryptophan-like; FA- fulvic acid-like; SMP - soluble microbial products; HA - humic acid-like.

Under the optimal operational conditions such as 1 g/L and 40 W/m², with respect to achieving high DOM removal performance, a significant amount of HA and SMP was removed by the UV/TiO₂ photocatalysis and a very low level of AP was observed in the permeate produced by both MBRs. This has led to a significant

reduction of primary halogenated DBP precursors (Bond et al., 2011; Ma et al., 2016), which ultimately would lead to low DBP formation during the use of chlorine based pathogen control strategies for recycled water distribution (Capocelli & Piemonte, 2021). This is similar to the results reported by Gora & Andrews (2017) that TiO_2 photocatalysis can reduce the DBP formation potential of surface water with a treatment time over 30 minutes. Furthermore, AnMBR samples displayed a higher SMP removal compared to AeMBR. This was due to the non-selective removal of DOM while a high initial DOM concentration was present in the AnMBR effluent. In contrast, AeMBR samples exhibited a higher HA removal compared to the AnMBR. This agrees with Bekbölet (1996) and Wiszniowski et al. (2002) that under a fixed TiO_2 concentration, increasing the humic acid concentration drives a decrease of radical generation efficiency. This is because the HA substances can readily absorb the UV_{254} irradiation, leading to not only a lower removal of HA, but also a reduced overall photocatalysis efficiency (Carra et al., 2016; Maghsoodi et al., 2019; Snow et al., 2019). Hence the high amount of HA present in the AnMBR samples is significantly limiting the overall photodegradation efficiency.

5.3.3 Future perspective of UV/ TiO_2 as the post-treatment of MBR effluents

Due to energy consumption of the UV lights, the costs of purchasing the catalyst (TiO_2) and its replacement over time due to losses in the process, and also filtering the TiO_2 out of the solution, UV/ TiO_2 is considered an expensive and energy intensive processes although the regenerable and solar-activatable characters of TiO_2 are seen as having the potential to overcome these limitations (Chatzisyneon et al., 2013; Loeb et al., 2019; MacAdam et al., 2012). The complex matrix and high concentrations of organics and nutrients in AnMBR effluents constitute limitations for the use of UV/ TiO_2 as a single barrier to secure a water quality sufficient for many reuse applications. However, the non-photodegradation of the adsorbed ammonia leading to competition for adsorption on the media which reduces the overall removal efficiency. This raises the question if the ammonia should be removed before the UV/ TiO_2 process. If keeping the ammonia in the water, the organic-ammonia trade-off effect suggests that by

optimising the intensity of UV irradiation, UV/TiO₂ can provide a selective solution for organic or ammonia prioritised removal for the post-treatment of AnMBR effluent. However, this highlights the need for research to potentially recover the adsorbed ammonia during a regeneration of TiO₂. On the other hand, this can be offset with replacing the pure TiO₂ with metal loaded ones such as TiO₂/Pt, that with the presence of Pt the adsorbed ammonia can be oxidized to a final product of N₂H₂ (Lee et al., 2002; Yuzawa et al., 2012), ultimately forming Nitrogen gas due to the self-decomposition under ambient temperature and pressure (Zhang et al., 2021). However, by removing the phosphorus and potentially ammonia as well, this would significantly address the competing effect between the nutrients with organic compounds, which is beneficial to achieve the full photocatalytic proficiency of TiO₂. Ideally TiO₂ can be used as an eternal catalyst without adsorbing the phosphorus and ammonia. In this regard, AnMBR technologies will need an extra post-treatment step but have potential for ammonia and phosphate recovery (Lee et al., 2016; Song et al., 2018; Xie et al., 2016). With the AeMBR, anaerobic and anoxic stages can be combined with the oxic stage to remove the nitrogen (nitrate) and phosphorus by biological processes. The catalyst-energy trade-off effect then can be used to optimise the operational conditions, reducing the operational cost of implemented schemes. In general, developments in reactor design (Dai et al., 2013; Natarajan et al., 2011) are beneficial to enhance the UV transmission efficiency regardless of the effluent matrix, which can ultimately improve the overall photocatalysis performance. Therefore, by tackling the current weaknesses, UV/TiO₂ can be a promising post-treatment solution for the AnMBR effluent, specifically when facing the challenge from emerging contaminants (Carra et al., 2015, 2016; Fattahi et al., 2021; Kanakaraju et al., 2018) in modern water reuse applications.

5.4 Conclusion

In this study, a direct comparison between the UV/TiO₂ photocatalysis with AnMBR and AeMBR treating real municipal wastewater was conducted focusing on the removal performance of organics and nutrients (nitrogen and phosphorus). Results suggest that the contribution of the UV irradiation significantly improved

the removal of organic compounds in the MBR effluents, as well as the ammonia in the AnMBR effluent. The removal mechanisms in both effluents are identified as adsorption for the organic, ammonia and phosphorus, but only photodegradation for the organic compounds. The competing effect between the nutrients and organic compounds leads to fact that the TiO_2 dosing concentration has a greater impact than the UV intensity on the overall removal performance, particularly more crucial with the presence of ammonia in the AnMBR effluent. This competition led to the removal of organic and nutrients in both effluents experiencing an adsorption-photodegradation trade-off effect under different operational conditions, and ultimately led to the current boundary of implicating as a single barrier for the potential water reuse implementations. However, with the mitigation such as advancement on the reactor design, UV/ TiO_2 photocatalysis can achieve a prioritised removal of organic matter or nutrients for AnMBR effluent, as well as providing an adjustable catalyst dosage and energy consumption for AeMBR effluent. Future studies should also consider the potential of the TiO_2 media for removal and recovery of nutrients from wastewater.

5.5 References

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5.6 Supporting information 4

5.6.1 Supporting information 4-1

Table 5-3 Isotherm parameter K'_f for each adsorbent with Langmuir isotherm, Freundlich isotherm and the normalised Freundlich isotherm and correspondence coefficient of determination value (R^2)

TiO ₂ dose (g/L)	Parameter	AnMBR			AeMBR		
			1/n	R ²		1/n	R ²
0.1	DOC		1.103	0.891		0.596	0.955
	TC	0.375	2.018	0.912	3.199	0.540	0.608
	IC	0.164	1.604	0.893	1.924	0.412	0.846
	TN	1.018	1.757	0.988	N/A		
0.2	TC	15.181	0.554	0.931	2.648	0.074	0.142
	IC	10.363	0.646	0.961	0.177	0.083	0.902
	TN	0.573	1.648	0.991	N/A		
	TP	1.950	0.722	0.976	1.492	0.767	0.999
0.5	TC	8.468	0.549	0.979	1.036	0.500	0.726
	IC	5.039	0.745	0.981	1.253	0.406	0.900
	TN	0.261	1.622	0.916	N/A		
	TP	1.465	0.577	0.916	1.280	0.557	0.950
1	TC	4.845	0.380	0.986	0.798	0.289	0.412
	IC	3.323	0.487	0.972	0.458	0.535	0.902
	TN	0.110	1.405	0.849	N/A		
	TP	2.120	0.379	0.924	1.877	0.358	0.980
2	TC	6.373	0.272	0.865	1.537	0.215	0.966
	IC	4.573	0.437	0.858	1.207	0.245	0.955
	TN	0.082	2.052	0.934	N/A		
	TP	1.487	0.478	0.883	1.417	0.352	0.713
5	TC	1.199	0.704	0.981	0.302	0.210	0.809
	IC	0.698	0.942	0.957	0.108	0.616	0.895
	TN	0.093	0.713	0.620	N/A		
	TP	1.142	0.091	0.996	0.959	0.035	0.987

6 Overall discussion

Anaerobic membrane reactors (AnMBRs) offer the possibility of a sustainable municipal wastewater treatment process, producing a nutrient rich and pathogen free effluent while maintaining sufficient organic pollutant removal for discharge and some sub-potable reuse applications. However, post-treatment may be needed to polish the AnMBR effluent to meet the quality requirement for the intended purpose. This highlights the intellectual contribution of this thesis in developing the understanding of the impact of AnMBR effluent on the post-treatment technologies for various water reuse applications, and comparison of the performance with aerobic membrane reactor (AeMBR) effluent. The knowledge from the investigation of the post-treatment technologies therefore helps to better define the relative performance and practical usefulness of the treatment trains as components of water reuse applications. Figure 6-1 illustrates how the various elements of the thesis combine to create an evidence base to generate these understandings, highlighting how the following sections contextualise and assess the significance of the results from the experimental chapters.

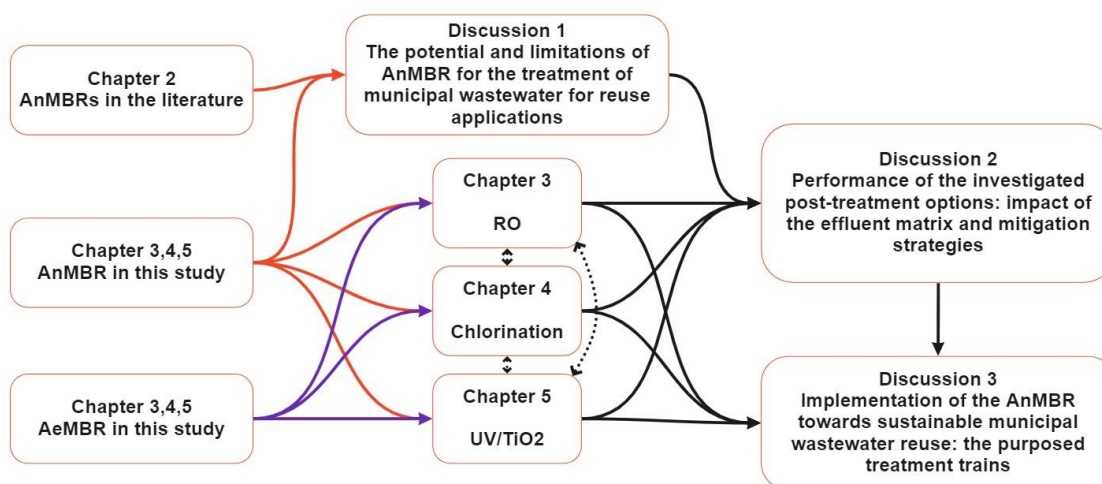


Figure 6-1 Thesis structure and related discussion

6.1 The potential and limitations of AnMBR for the treatment of municipal wastewater for reuse applications.

As highlighted in Chapter 1, previous studies have evidenced the advantage of energy recovery from the biogas produced during the anaerobic digestion of organic compounds in AnMBRs (Pretel et al., 2016; Song et al., 2018; Trzcinski & Stuckey, 2016; Wang et al., 2018). This offers a great potential towards more sustainable water reuse applications, which fulfils the target of closing the loop for a more circular economy (The European Commission, 2015). To better understand the potential and limitations of the produced AnMBR effluent, a benchmarking activity on the performance of AnMBR systems in the reviewed literatures (Chapter 2) and the AnMBR system (Chapter 3, Chapter 4, Chapter 5) in this study was carried out against available water reuse standards (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; PUB, 2020; USEPA, 2012, 2017) (Table 6-1).

Table 6-1 Performance of AnMBR systems in the reviewed literatures and the AnMBR system in this study against the threshold in various water reuse standards

Parameter	Source	Value	Non-Potable							Potable		
			ENV	RCA	LSI	TLF	SMF	FCR	FCP	FCN	IPR	DPR
TSS (mg/L)	LH	16	√	√	√	√	√	√	√	√	×	×
	LW	ND	√	√	√	√	√	√	√	√	√	√
	TH	ND	√	√	√	√	√	√	√	√	√	√
	TW	ND	√	√	√	√	√	√	√	√	√	√
FC (CFU/100 mL)	LH	90	√	√	×	×	×	×	√	√	×	×
	LW	ND	√	√	√	√	√	√	√	√	√	√
	TH	ND	√	√	√	√	√	√	√	√	√	√
	TW	ND	√	√	√	√	√	√	√	√	√	√
BOD₅ (mg/L)	LH	120	×	×	×	×	×	×	×	×	×	×
	LW	4	√	√	√	√	√	√	√	√	√	×
	TH	16	×	×	×	×	×	×	√	√	×	×
	TW	2	√	√	√	√	√	√	√	√	√	×
NH₃-N (mg/L)	LH	83.2	×	×	×	×	×	√	√	√	×	×
	LW	28	×	×	×	×	×	√	√	√	×	×

TP (mg/L)	TH	32	x	x	x	x	x	√	√	√	x	x
	TW	18	x	x	x	x	x	√	√	√	x	x
	LH	23.8	x	x	x	x	x	√	√	√	x	x
	LW	2.7	x	x	x	x	x	√	√	√	x	x
	TH	5.8	x	x	x	x	x	√	√	√	x	x
	TW	3.2	x	x	x	x	x	√	√	√	x	x

Notes: FC – Faecal Coliform; LH – Highest value reported in the literature; LW – Lowest value reported in the literature; TH – Highest value measured in this study; TL – Lowest value measured in this study; ND – Non detectable; ENV – Environmental; RCA – Recreational; LSI – Landscape irrigation; TLF – Toilet flushing; SMF – Street maintenance and firefighting; FCR – Food crops with raw consumer; FCP – Food crops with processed consumer; FCN – None food crops; IPR – Indirect potable reuse; DPR – Direct potable reuse.

The use of an ultra-filtration (UF) membrane in the anaerobic bioreactor deployed in this study successfully rejected the suspended solids and coliforms (Total, Faecal and Escherichia coliforms) (Chapter 3, Chapter 4, Chapter 5). These results agree with the non-detectable suspended solids reported by several authors (Martin Garcia et al., 2013; Petropoulos et al., 2019; Seco et al., 2018; Shin et al., 2014; Wang et al., 2019), but exhibit better performance on reported Faecal Coliforms of 42 ± 48 CFU/100 ml (Peña et al., 2019) and 49 CFU/100 ml (Martinez-Sosa et al., 2011) (Chapter 2). This confirms the advantage of hybrid membrane systems such MBRs to deliver a solids free and low pathogen contamination effluent for reuse as evidenced by the fact that most standards for reuse were met for solids and coliforms (Table 6-1).

During the 55 weeks of operation, the pilot scale AnMBR effluent exhibited an overall organic content (as BOD₅ concentration) of 9 ± 7 mg/L. This is lower than the average value of 21 mg/L in the reviewed literatures reported in Chapter 2, but is slightly higher than the 3 ± 2 mg/L BOD₅ exhibited by the AeMBR effluent treating the same influent in this study. The effluent produced by the AnMBR had the potential to exceed the 10 mg/l BOD₅ limit in Table 6-1 for recreational and toilet flushing purposes in the non-potable water reuse standard for USA, China, Israel, and Canada (CNEPA, 2021; Health Canada, 2010; ISO, 2018; USEPA,

2012), as well as exceeding the 6 mg/L BOD₅ limit for ground water recharge in the USA and Chinese indirect potable reuse standards (CNEPA, 2021; USEPA, 2017). Compared to the AnMBR systems reviewed in Chapter 2, the AnMBR exhibited a lower variability on the organic removal performance but higher than the AeMBR. This leads to a potential failure on stratifying the quality requirements for specific reuse purpose (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; USEPA, 2012). To illustrate, for agriculture irrigation purposes other than food crops with raw consumer (FCR), the AnMBR effluent can produce an effluent adequate to all standards regardless the organic removal performance. However, for reuse purposes that have a stricter organic requirement, the variability of effluent organic content has a potential to exceed the standards (CNEPA, 2021; USEPA, 2017). The observed variability of the AnMBR effluent quality could be a result of insufficient mixing in the up flow anaerobic sludge blanket (UASB) reactor before the membrane. This has led to a lower hydraulic retention time (HRT) that optimized for the organic removal, causing incomplete anaerobic biodegradation hence a higher organic content in the effluent. On the one hand, if considered the effluent is for irrigation purposes, this could be addressed with better mixing strategy such as adding multiple recirculation diffusers at the bottom of UASB reactor for the influent wastewater and the recirculate water from the membrane tank; or simply add a stirrer but comes with extra energy consumption, which highlights the trade-off between the final effluent quality with the operational cost based on the required treatment performance. On the other hand, for the higher grade reuse applications that requires both greater and more consistent organic removal performance, the variability of AnMBRs may not be offset by an optimised control strategy, highlighting the need for post-treatment to polish the final effluent to consistently meet the standards.

As noted before, nutrients (ammonia and phosphate) are not removed through anaerobic systems either in the biological anaerobic digestion or the physical membrane filtration (Chapter 3, Chapter 4, Chapter 5). This is consistent with the literature reviewed in Chapter 2 (Table 6-1). The unremoved nutrients provide the potential to reuse the AnMBR effluent as a combination of water and fertilizer

source for a wide range of irrigation applications (Song et al., 2018; Tampo et al., 2022). However, when considering reuse of the AnMBR effluent for other reuse purposes such as environmental, recreational and toilet flushing, or as a source for ground water recharge in indirect potable implementations, the unremoved nutrients become the weakness of AnMBR systems. In comparison, with the AeMBR, combining anaerobic and anoxic biological stages to the oxic stage would produce, at a significant cost, an effluent adequate for all non-potable reuse standards in terms of nutrient consents (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; USEPA, 2012). However, the AeMBR effluent still requires post-treatment if the effluent aimed at potable reuse applications (CNEPA, 2021; USEPA, 2017). Due to the requirement for 1 mg/L residual chlorine concentration set by many non-potable water reuse standards - including but not limited to the irrigation purpose (CNEPA, 2021; EUPC, 2020; ISO, 2018; USEPA, 2012) - a further disinfection process would be needed in order to use treated MBR effluent as a water source.

6.2 Performance of the investigated post-treatment options: impact of the effluent matrix and mitigation strategies

The investigated post-treatment technologies were reverse osmosis (RO) (Chapter 3), Chlorination (Chapter 4), and UV/TiO₂ photocatalysis (Chapter 5). The dissimilar matrix of AnMBR and AeMBR effluents has led to different performance in each post-treatment process, producing a final effluent to different qualities for various water reuse applications, as well as highlighting the potential to combine the post-treatment options to offset the weaknesses identified when each of them was applied as a single barrier.

The ammonia (as ammonium) present in the AnMBR effluent was not effectively removed by RO filtration during the trials conducted in this study. This agrees with the incomplete ammonia removal in a similar feed matrix observed by Qin, Oo and Coniglio (2005) and Shin et al. (2021). Inadequate ammonia removal leads to the produced permeate exceeding the Singapore's PUB NEWater standard for indirect potable reuse (PUB, 2020), as well as the threshold in the USA and Chinese potable reuse standard for ground water recharge (CNEPA,

2021; USEPA, 2017). Despite the unsatisfactory ammonia removal performance in the AnMBR, in terms of the permeate quality, RO delivered a similar removal performance of organic, trace element, and primary halogenated disinfection by-product (DBP) precursors regardless of the feed effluent matrix. This highlights that RO can be applied as a standalone post-treatment technology for the AeMBR to deliver a final permeate quality sufficient for a wide range of reuse implementations, even in fulfilment to the requirement of direct potable reuse purposes (CNEPA, 2021; USEPA, 2017) (Chapter 3). To address the ammonia concern in the AnMBR-RO permeate, a combination of RO followed by Chlorination provides a practical solution as the RO can significantly remove the DBP precursors (Chapter 3) then the post-RO chlorination can eliminate the unremoved ammonia while offering a residual disinfectant for distribution with low disinfection by-products formation potential (DBPFP) (Chapter 4). Moreover, the RO displayed a robust removal performance of all the monitored parameters in all filtration tests conducted in Chapter 3's experiment. This outcome offers a higher matrix tolerance in feed water quality variations when compared to the other post-treatment options (UV/TiO₂) investigated in this thesis (Chapter 5). This offers the opportunity to turn the identified weakness of UV/TiO₂ into an advantage when employed as a standalone post-treatment process for AnMBR effluent, by combining with RO as a following step. In this combination, UV/TiO₂ acts as an additional disinfection barrier with no chlorine related DBP concern (Chapter 5), whilst the low initial concentration of RO permeate can significantly boost the removal efficiency of photocatalysis process. Consequently, UV/TiO₂ offsets the ammonia concern in the RO permeate, also removes the phosphorus, and simultaneously effectively eliminates the DOM fractions regardless of the effluent matrix (Chapter 3, Chapter 5). Alternatively, RO followed by UV/TiO₂ would be expected to polish the MBR effluents to an adequate quality for the potable implementations (CNEPA, 2021; PUB, 2020; USEPA, 2017). However, this combination has a challenge of high operational expenditure, since both RO and UV/TiO₂ are largely evidenced as energy intensive processes (Alghoul et al., 2009; Chatzisyneon et al., 2013; Holloway et al., 2016).

For reuse for irrigation purposes, the AnMBR effluent displays a clear advantage over the AeMBR effluent, offers a controllable disinfection process with a lower DBPFP compared to both surface waters and AeMBR effluent (Chapter 4), as well as a required residual chlorine and ammonia concentration which is required for use with specific agricultural crops (Xu et al., 2020). In particular to a sufficient residual chlorine concentration (1 mg/L) is required for the irrigation reuse applications, the chlorine dosing response curve along with three identified critical dosing points identified in this thesis provides a more comprehensive understanding of the ammonia consumption and the DBP generation for the disinfection of AnMBR effluent (Chapter 4). The identified DBPFPs can be used to help regulators set appropriate reuse standards (in terms of both quality parameters and processes) where there are concerns about DBP formation in waters used for irrigation processes. Moreover, the identified critical dosing point with the minimum residual of ammonia and chlorine suggests a better pathway to mitigate the ammonia concern by removing the ammonia before the RO step, with the combination of Chlorination followed by RO. Pre-RO Chlorination offers a precise removal of ammonia by forming chloramines, simultaneously reducing the RO fouling since the unremoved ammonia was identified as one of the critical factors that led to a higher membrane fouling when comparing the two MBR effluents (Chapter 3). Moreover, a controllable chlorination process can deliver a low DBPFP yield in the disinfected effluents (Chapter 4), as well as a residual chlorine concentration to avoid the membrane damage caused by the ineffective removal of generated active chlorines (Al-Abri et al., 2019). In contrast, due to a higher DBPFP yield, as identified in Chapter 4, and the incomplete removal of trihalomethanes (THMs) and haloacetic acids (HAAs) in the pre-RO disinfected AeMBR effluent - as reported in various studies (Comerton et al., 2005; Kimura et al., 2003; Zazouli & Kalankesh, 2017) - the combination of Chlorination followed by RO is not suggested as a suitable post-treatment option for AeMBR effluent in this case.

As a post-treatment process, UV/TiO₂ photocatalysis offers a promising solution to affect the removal of organic matter or nutrient compounds from AnMBR effluent and provides a solution to balance the catalyst dosage and energy

consumption for AeMBR effluent. The combination of UV/TiO₂ followed by Chlorination can potentially purify the AnMBR and AeMBR effluent to make it suitable for a wide range of sub-potable reuse applications such as environmental, recreational and toilet flushing (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; PUB, 2020; USEPA, 2012). In this combination, the UV/TiO₂ acts as the sweeper for the unremoved organic and nutrient compounds, also degrades the DOMs, which is identified as the DBP precursor for subsequent chlorination processes (Chapter 4, Chapter 5). However, this combination was found to limit the full photocatalytic proficiency of TiO₂ due to the competing effect between nutrients with other photodegradable compounds. Ideally TiO₂ can be used as an eternal catalyst without adsorbing the nutrient, which significantly reduces the cost for catalyst consumption compared to AOPs such as UV/H₂O₂. The observed competition effect led to UV/TiO₂ photocatalysis delivering a less competitive organics and nutrients removal performance compared to RO filtration (Chapter 3), nano-filtration (NF) (Wei et al., 2015), or processes that are less prone to competition with nutrients such as activated carbon adsorption (Mai et al., 2018b). However, UV/TiO₂ is reported to effectively eliminate a broad range of emerging contaminants (ECs) (Carra et al., 2015, 2016; Fattahi et al., 2021; Kanakaraju et al., 2018). In addition to the treatment train of RO followed by UV/TiO₂ identified above, the combination of Chlorination + RO + UV/TiO₂ + Chlorination (Chapter 3, Chapter 4, Chapter 5) is expected to provide a better overall treatment performance for AnMBR effluent in potable water reuse applications by avoiding the different limitations observed for each of the technologies. In contrast, the UV/TiO₂ photocatalysis achieved a higher removal efficacy for organic matter and phosphorus in the AeMBR effluent due to a lower initial organics concentration and the absence of ammonia. Also, anaerobic and anoxic stages can be combined with the oxic stage to remove the nitrogen and phosphorus, producing an even lower initial contaminants concentration. This suggests the combination of RO + UV/TiO₂ + Chlorination (Chapter 3, Chapter 4, Chapter 5) is an attractive treatment train for the treatment of AeMBR effluent, to achieve a wide range of direct and indirect potable applications (CNEPA, 2021; USEPA, 2017). Despite the different biological

treatment stage of MBR, comparison of these two treatment trains recognises that the chlorination before RO in the treatment train for AnMBR is the other key difference. This extra step is to maximize the advantage discovered in Chapter 4 with a minimum chlorine and DBP residual hence preventing the excess ammonia problem while mitigating the RO fouling, as well as offering a minimum DBP concern in the final effluent. RO followed by UV/TiO₂ is employed as a solid defender to tackle the unremoved organics, nutrients, and the trace element in the treatment trains for both effluents. This meets the suggestion from the evidence base, that the RO can consistently provide a better permeate quality than those alternative options such as media filtration and coagulation/flocculation (Plevri et al., 2017; Shareefdeen et al., 2015), and recent reviews of DPR schemes worldwide have indeed reported that configurations do often make use of UV-based advanced oxidation as the final step before chlorination disinfection for distribution (Jeffrey et al., 2022). The combination of RO + UV/TiO₂ addresses the catalyst surface inhibition for heterogeneous photocatalysis caused by the competing effect from the nutrients (Chapter 5), thus providing the same treatment performance for ECs compared to the homogeneous AOPs for example UV/H₂O₂ and ozonation, but obviates the need to continuously supply precursor chemicals with a lower operational expenditure (Loeb et al., 2019; Villegas-Guzman et al., 2017). In terms of the energy consumption, the AnMBR based treatment train therefore delivers a potential to lower operational cost with the recovered biogas (Futselaar et al., 2013; Giménez et al., 2014; Shin et al., 2014). This offers a better application perspective than AeMBR based treatment trains in particular for the large scale operation. Moreover, the potential application of solar irradiation to drive the TiO₂ photocatalysis suggests the advantage of TiO₂ based AOPs is not limited as eternal catalyst, but also a free energy source of sunlight (Hasik et al., 2017; Klare et al., 2000; Lim et al., 2011). The application of final chlorination process is depending on the distribution pathogen control strategy, the receiving water body condition, or if applied directly as drinking water source for specific potable reuse application (CNEPA, 2021; USEPA, 2017), which is determined on a case-by-case basis.

Overall, the different removal performances and operational behaviours of the investigated post-treatment processes help identify monitoring and control priorities for water quality indicators, support regulators in the formulation of guidelines, and inform treatment train design strategies to deliver safe and sustainable water reuse implementations. Depending on the purpose of the water reuse application, the investigated post-treatment technologies can be flexibly combined to achieve a specific removal of the targeted compounds, or improve the overall performance of the combined systems, as well as offer an extra barrier in the treatment train, which ultimately produces a quality sufficient effluent for the targeted reuse implementation.

6.3 Implementation of the AnMBR towards sustainable municipal wastewater reuse: the purposed treatment trains

Classified by the threshold in available water reuse standards (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMCC, 2008; PUB, 2020; USEPA, 2012, 2017), Table 6-2 displays proposed treatment trains for water reuse implementations founded on the results from the investigated post-treatment technologies in combination with an AnMBR in descending order of water quality requirement. A benchmarking of the listed treatment trains was conducted to highlight the advantage and current limitation of purposed treatment trains (Columns 3 and 4).

Similar to the AeMBR based treatment trains that are operational or under construction for potable applications, RO remains the dominant process, regardless of the final application of the effluent for DPR or IPR. The suggested pre-treatment or post-treatment for the RO step is more based on the consideration of reducing the overall energy consumption of the system (pre-treatment) or improving the overall treatment performance with an extra barrier (post-treatment). However, the additional step may bring the need for better process control or a higher footprint. Indeed, for non-potable reuse applications, none of the options proposed involves the RO step since the quality requirement can be met with a shorter treatment train and a lower energy consumption, reducing the process complexity and footprint, but may be dependent on the

robustness of the technologies applied, due to the lack of extra barrier for quality insurance. Therefore, this highlights the need for precise real time monitoring of the final effluent, in particular for the designs with a single post-treatment step.

Operational expenditure is always considered as important when applying membrane and photocatalysis technologies (Capocelli & Piemonte, 2021). This table highlights the identified advantages and disadvantages, including the consideration on the economic aspects. Due to the fact that this thesis did not investigate the capital or operational expenditure corresponding to the treatment trains suggested in Table 6-2, the benchmarking metric on the energy consumption was obtained from the literature on treating AnMBR (Foglia et al., 2021) and AeMBR (Hasik et al., 2017; Holloway et al., 2016; Keller et al., 2022) effluent. Therefore, as the energy recovery potential is commonly considered as the benefit of the applying the AnMBR, this ultimately suggests the lifecycle economic feasibility calculation is rather significant when considering for the actual implementations.

Table 6-2 Proposed treatment trains for various water reuse implementations using the investigated post-treatment processes

Treatment train	Application(s)	Advantages	Current limitation
AnMBR + CI + RO + UV/TiO ₂ + CI	DPR	Provides highest effluent quality, low RO fouling	Requires complex process control
AnMBR + RO + UV/TiO ₂ + CI	DPR	Multiple barriers to secure the highest effluent quality	High operational cost (RO fouling + Energy)
AnMBR + CI + RO + UV/TiO ₂	IPR	Extra barrier for the ECs, low RO fouling	Requires complex process control
AnMBR + RO + UV/TiO ₂	IPR	Low footprint, straightforward configuration	High operational cost (RO fouling + Energy)
AnMBR + CI + RO	IPR	Low RO fouling, straightforward configuration	May require extra barrier for ECs
AnMBR + RO + CI	IPR	Low footprint, straightforward configuration	May require extra barrier for ECs, energy intensive

AnMBR + RO	IPR	Low footprint, straightforward configuration	May require extra barrier for NH ₃ -N and ECs
AnMBR + UV/TiO ₂ + Cl	ENV RCA LSI TLF SMF	Low footprint, straightforward configuration	High operational cost of chemicals and energy
AnMBR + UV/TiO ₂	ENV RCA LSI TLF SMF	Low footprint, straightforward configuration	High operational cost of energy
AnMBR + Cl	FCR FCP FCN	Fertigated, Low DBP formation	High chemical consumption

Notes: Cl – Chlorination; ENV – Environmental; RCA – Recreational; LSI – Landscape irrigation; TLF – Toilet flushing; SMF – Street maintenance and firefighting; FCR – Food crops with raw consumer; FCP – Food crops with processed consumer; FCN – None food crops; IPR – Indirect potable reuse; DPR – Direct potable reuse; ECs – Emerging contaminants, NH₃-N – Ammonia nitrogen.

6.4 References

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7 Conclusions and future work

7.1 Conclusions

Post-treatment offers the possibility of reusing AnMBR treated municipal wastewater for a range of applications. Due to its distinctive matrix, the AnMBR effluent has different impacts on the performance of the investigated post-treatment technologies, highlighting their potential implementation across a wide range of water reuse purposes. Overall, in tandem with targeted post-treatment processes, AnMBRs offer the potential to define more sustainable and environmentally friendly treatment trains for future water reuse applications.

Following on from the determined objectives to investigate the impact of the AnMBR effluent matrix from the treatment of municipal wastewater on selected post-treatment processes including reverse osmosis, chlorination, and UV/TiO₂ within the context of water reuse, the following conclusions made were:

- AnMBR allows a potential sustainable municipal wastewater treatment for a wide range of reuse applications with also potential for resource recovery but may need post-treatment for application needing higher qualities. Under optimum operational conditions such as mesophilic temperature and longer hydraulic retention time, AnMBR could directly produce an effluent satisfying the strictest consents for all sub-potable reuse application in terms of organic, solids and pathogen consents. The unremoved nutrients provide an opportunity for various irrigation purposes. However, for other non-potable water reuse applications with strict nutrient thresholds and potable reuse that has more comprehensive parameters as well as a low limit for organics and trace elements, the AnMBR effluent requires additional treatment to produce a water quality sufficient for reuse implementation. (Objective 1, 2; evidence in Chapter 2)
- The combination of RO and AnMBR can produce a water which meets the indirect potable reuse standard set out in Singapore's PUB NEWater regulations (PUB, 2020). The RO acted as a robust barrier for organics, nutrients, trace elements, and DBP precursors. However, the inferior organic removal performance and inclusion of ammonia by AnMBR

resulting in a higher RO fouling and a potential risk of exceeding the ammonia nitrogen limit compared to AeMBR-RO. The foulant deposition sequence during the filtration of both effluents are organic and colloidal, then followed by inorganic substances. TOC and Ca were found as the most deposited foulants in both effluents. The deposition rate of ammonia (13.8 %) during the filtration of AnMBR effluent was significantly higher than nitrate (0.02%) for the AeMBR effluent. (Objective 2, 3, 4, ; evidence in Chapter 3)

- Chlorine based disinfection processes behaved differently in AnMBR and AeMBR effluents, as a mixed process of chloramination and chlorination happened in the AnMBR effluent due to the presence of ammonia, whilst a monophonic chlorination process occurred in the AeMBR effluent. In comparison to AeMBR effluent and surface water source, chlorination of AnMBR effluent displayed a competitive advantages of low DBP formation with a controllable residual chlorine and ammonia concentration for irrigation applications. The identified critical dosing point with the minimum residual ammonia and chlorine concentration can be applied as an anti-fouling and excess ammonia control strategy for the RO filtration of AnMBR effluent. (Objective 2, 3, 4; evidence in Chapter 4)
- UV/TiO₂ photocatalysis offers a promising solution for prioritising the removal of organic matter or nutrients from AnMBR effluent to achieve the required quality, as well as providing a flexible solution to balance the catalyst dosage and energy consumption for the AeMBR effluent to optimise the operational cost. TiO₂ dosage was observed to have a greater impact than UV intensity on improving the overall removal performance due to the fact that nutrients are competing for the limited adsorption site but not photodegraded. This competition reduced the removal efficacy for organic compounds removal, led to the removal of organic and nutrients in both effluents experienced an adsorption-photodegradation trade-off effect under different operational conditions, and ultimately led to the current boundary of implicating as a single barrier for the potential water reuse implementations. However, UV/TiO₂ can be applied as a following

step for RO filtration to tackle the excess organic or nutrient compounds in the permeate to produce a high water quality sufficient for potable water reuse applications. (Objective 2, 3, 4; evidence in Chapter 5)

- The distinctive matrices of AnMBR and AeMBR effluents has led to the dissimilar performance of the investigated post-treatment technologies but also highlighted the potential combination of those processes to achieve either better final effluent quality or lower operational expenditure. In particular, the treatment train of AnMBR - Chlorination can deliver a quality sufficient for various agriculture irrigation reuse purposes (CNEPA, 2021; Health Canada, 2010; ISO, 2018; NRMMC, 2008; PUB, 2020; USEPA, 2012); and the treatment train of AnMBR-Chlorination-RO-UV/TiO₂ is expected to produce a permeate adequate for direct potable reuse applications (CNEPA, 2021; USEPA, 2017). This second option offers a sustainable potable reuse of municipal wastewater with a lower membrane fouling and DBP concern; as well as the potential resource recovery from the methane rich biogas and nutrients from the RO concentrate. (Objective 2, 3, 4; evidence in Chapter 6)

7.2 Future work

This research has explored the role of post-treatment for AnMBR based treatment trains treating municipal effluent for water reuse purposes. The performance, potential, and limitations, as well as the comparison to the role of AeMBR applications have been investigated with reverse osmosis, chlorination, and UV/TiO₂ photocatalysis, further developing the understanding of the processes and associated implications, respectively exhibited within the corresponding chapter and the overall discussion (Chapter 6, Section 2, 3). However, there are still, in this context, several avenues of research which require addressing:

- As discussed in Section 9 of Chapter 2, controlling the temperature in the AnMBR can allow to achieve better performance so a careful evaluation on the feasibility of deploying underground AnMBR systems focusing on operation risks identification and mitigation including the biogas recovery safety is needed. This would be beneficial to understand the potential

application of AnMBRs fully or semi underground in regard to optimising the operational temperature and minimizing the concern of footprint and public perception.

- The results obtained in Chapter 3 have highlighted the contribution of key components of the AnMBR effluent (e.g. ammonium) to membrane fouling so continuous efforts should be made to understand the impact of foulants (specifically nitrogen species and trace elements) co-presence on RO fouling. Other RO operation strategy such as cross flow or configuration like spiral wound will provide more comprehensive understanding and further insights in fouling behaviour. Identification of functional groups and analytical measurement using Fourier transform infrared spectroscopy (FTIR) would be of benefit.
- Due to the interruption to the research caused by COVID-19, the impact of AnMBR and AeMBR effluent on N-Nitrosodimethylamine (NDMA) formation, commonly known to occur during chloramination, was not characterised during the experiment period for Chapter 4. This work would help broaden the understanding of DBP formation behaviour caused by AnMBR effluent. In addition, the fate of organic micropollutants during the RO filtration and UV/TiO₂ photocatalysis was not addressed here. Although many studies have demonstrated the ability for these technologies to remove micro-pollutants in other contexts, it would be beneficial to carry out a specific study based on an AnMBR based treatment train for reuse.
- The work in Chapter 5 focused on TiO₂ as a catalyst because of the advantage of not having to continuously dose chemicals but further studies on other photocatalyst not based on adsorption for the polishing of the AnMBR effluent such as H₂O₂ or Fe may help further understand the removal and selectivity for the different compound investigated and provide a wider choice for technology selection in treatment train for reuse.
- As suggested in Section 2 and 3 of Chapter 6, a pilot scale trial for the identified treatment train such as AnMBR-Chlorination-RO and potentially also applying the UV based AOP as the final step could provide a more

comprehensive and precise understanding on the combination of these post-treatment technologies for water reuse schemes. This will not only help us better understand the treatment performance and generate operational knowledge but also allow us to more precisely determine the true cost of an AnMBR based direct potable reuse implementation.

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