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TREATING GREYWATER FOR BUILDING-LEVEL REUSE: CERAMIC MEMBRANE BIOREACTORS

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CONTENTS

ABBREVIATIONS

BOD_5	5-day Biochemical Oxygen Demand.
COD	Chemical Oxygen Demand.
CW	Constructed Wetland.
DO	Dissolved Oxygen.
EPS	Extracellular Polymeric Substances.
GW	Greywater.
HRT	Hydraulic Retention Time.
MB-MBR	Moving Bed Membrane Bioreactor.
MBR	Membrane Bioreactor.
ML	Mixed Liquor.
MLSS	Mixed Liquor Suspended Solids.
MLVSS	Mixed Liquor Volatile Suspended Solids.
OLR	Organic Loading Rate.
PCPs	Personal Care Products.
PVDF	Polyvinylidene fluoride.
SAOR	Specific Ammonia Oxidation Rate.
SRT	Solids Retention Time.
SS	Suspended Solids.
T	Temperature.

TMP	Transmembrane Pressure.
TN	Total Nitrogen.
TOC	Total Organic Carbon.
TP	Total Phosphorus.
TSS	Total Suspended Solids.
VSS	Volatile Suspended Solids.

ABSTRACT

English

Natural water supplies are shrinking and becoming more uncertain, while the demand for fresh water is only expected to grow. Reducing the reliability of water supply from natural resources by reusing wastewater is one of the key developments towards a more water-resilient society. Greywater has a high potential for reuse due to its relatively low pollutant load. Decentralized greywater reclamation efficiently couples greywater production to local water demands. Membrane bioreactors (MBRs) emerge as a promising technology for this purpose, offering high effluent quality and a compact footprint. Within the scope of this thesis, conventional MBRs and a moving bed MBR (MB-MBR) treating synthetic greywater were optimized in terms of hydraulic residence time, aeration flow, and organic loading rate. The primary objective was to achieve a high-quality effluent, meeting standards for non-potable greywater reuse in terms of chemical oxygen demand (COD), inorganic nitrogen compounds, and total nitrogen (TN). The synthetic greywater had an average COD concentration of 402±57 mg/L and average ammonia and TN concentrations of 15±3 and 25±5 mg N/L. While standard MBRs struggled to adequately eliminate total nitrogen, the MB-MBR demonstrated success in this aspect due to enhanced denitrification within the biofilms on the carriers. The specific ammonia oxidation rate of the sludge ranged from 1.2 to 3.2 mg NH_4^+-N/(g VSS·h). In the optimized MB-MBR, the maximum effluent concentrations for COD, ammonia, nitrite, nitrate, and TN were 43 mg/L, 3 mg N/L, 0.5 mg N/L, 4 mg N/L, and 7 mg N/L, respectively.

Nederlands

De bevoorrading van zoet water wordt steeds schaarser en onzekerder, terwijl de vraag volgens huidige voorspellingen zal blijven stijgen. Afvalwaterhergebruik is een belangrijke stap richting een duurzamere samenleving daar het de afhankelijkheid van natuurlijke waterbronnen reduceert. Grijswater in het bijzonder heeft een hoog potentieel voor hergebruik vanwege de lage verontreinigingsgraad. Gedecentraliseerde grijswaterzuivering koppelt de productie ervan efficiënt aan lokale waterbehoeften. Membraanbioreactoren (MBR's) zijn een veelbelovende technologie hiervoor door hun hoge effluentkwaliteit en compacte aard. Binnen het kader van deze scriptie werden con-

ventionele MBR's en een bewegende bed MBR (MB-MBR) voor de behandeling van synthetisch grijswater geoptimaliseerd op het gebied van hydraulische verblijftijd, beluchtingsdebiet en organische belastingsgraad. Het hoofddoel was om een effluent van hoge kwaliteit te bereiken dat voldoet aan de normen voor niet-drinkbaar hergebruik van grijswater, wat betreft de chemische zuurstofvraag (CZV), anorganische stikstofverbindingen en totale stikstof (TN). Het synthetische grijswater had een gemiddelde CZV-concentratie van 402±57 mg/L en gemiddelde ammonium- en TN-concentraties van 15±3 en 25±5 mg N/L. De conventionele MBR's waren niet in staat om totale stikstof voldoende te verwijderen. De MB-MBR slaagde hier wel in vanwege optimale denitrificatie binnenin de biofilm op het dragermateriaal. De specifieke ammoniumoxidatiesnelheid van het slib varieerde tussen 1,2 en 3,2 mg NH_4^+-N/(g VSS·h). In de geoptimaliseerde MB-MBR waren de maximale effluentconcentraties voor CZV, ammonium, nitriet, nitraat en TN respectievelijk 43 mg/L, 3 mg N/L, 0,5 mg N/L, 4 mg N/L en 7 mg N/L.

Chapter 1

INTRODUCTION

1.1 Water Scarcity: A Pressing Global Concern

Water scarcity is defined as the shortage of the available supply of freshwater towards the expressed demand for human use. It negatively impacts human health and well-being, agricultural yields, industrial productivity and environmental quality (Steduto et al., 2012; Zhang et al., 2019). Intra-annual fluctuations in water demand and availability result in significant temporal disparities of water scarcity within a given year (Damkjaer & Taylor, 2017; Mekonnen & Hoekstra, 2016). Globally, about 3.6 billion people are currently confronted with severe water scarce living conditions for at least one month a year. By 2050, this number is estimated to increase to 5 billion people (WMO, 2022). Figure 1.1 demonstrates the geographical variation in the projected percentage of the population experiencing severe water scarcity during the driest month of 2050.

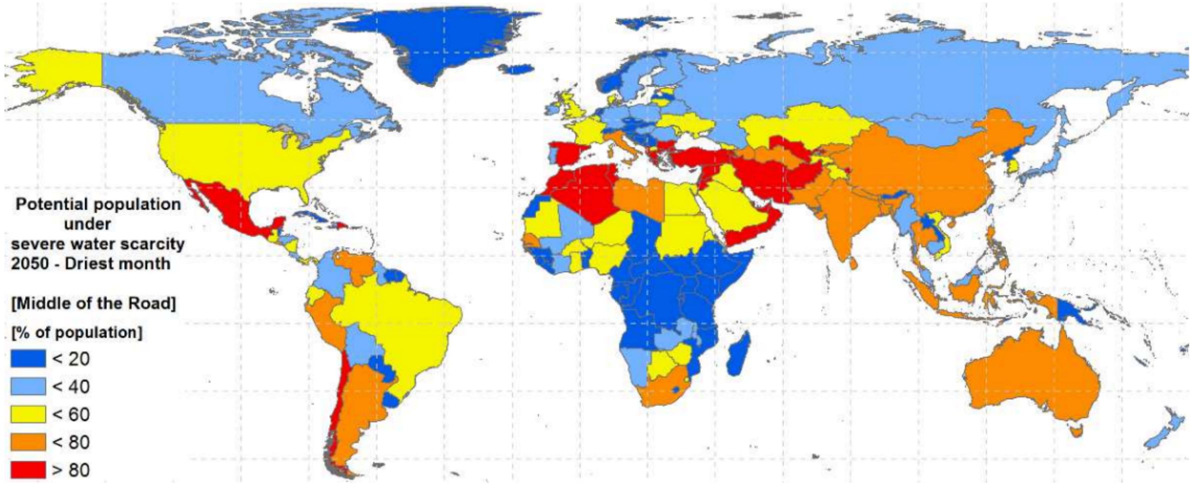

Figure 1.1: Projected percentage of the population facing severe water stress in the driest month of 2050 under a business-as-usual scenario. Retrieved from Burek et al. (2016).

Urban areas are particularly vulnerable towards water stress, because their high population densities generate a very spatially concentrated water demand. The supply of water becomes more difficult, *i.e.*, it has to be sourced from further away, and an extensive infrastructure for distribution has to be expanded and maintained (Krueger et al., 2019; Larsen & Gujer, 2013). Many of the current urban communities have not yet reached adequate resilience towards water scarcity issues (Zhang et al., 2019). The extent to which industry and agriculture are organized in or near urban areas play a very important role in this matter. Therefore, the appropriate allocation of water resources becomes very crucial in cities (Li et al., 2020; Zhang et al., 2019).

Presently, there is a worldwide trend towards urbanization, with projections that indicate an increase of the proportion of the global population living in urban areas from 55% in 2020 to 68% in 2050. It is also estimated that almost half of the world's biggest cities (*i.e.*, with more than 1 million inhabitants) are expected to endure water scarce living conditions by 2050 (He et al., 2021). This emphasizes the urgent need to develop strategies and technologies geared towards addressing urban water scarcity. The alarming state of freshwater availability has even prompted the inclusion of "water resilience" as an important element of the United Nations Climate Change Conference in 2022 (UN, 2022).

1.2 Driving Factors of Water Scarcity

1.2.1 Increasing Demand for Fresh Water

The largest water consumption originates from agriculture, it represents almost 70% of global fresh water withdrawals. Energy generation and industrial manufacturing account for 19% and domestic water consumption is responsible for the remaining 12% globally. In some developing countries, however, agricultural water demand can account for up to 95% of their total national water withdrawals (Zhongming et al., 2021).

The global human population surpassed 8 billion people in 2022. By 2050, the world's population is expected to increase by nearly 2 billion (Zeifman et al., 2022). Therefore, irrigated agricultural production will need to keep expanding to satisfy global alimentary demands. Moreover, the progression of socioeconomic factors, such as rising incomes, causes a dietary shift towards more animal-based and dairy products in many developing nations. The livestock and dairy production, necessary for such protein-rich diet, will require a significant larger amount water (Biswas & Tortajada, 2018; Jalava et al., 2014). Water consumption for agricultural purposes will increase globally due to a shift in both the quantity and quality of global food production. While agricultural water demand will keep growing, the growth rate is projected to decelerate, due to the enhanced effectiveness of irrigation systems and advancements in agricultural practices in general (FAO, 2017).

Economic growth and better living standards are expected to give rise to significant larger water requirements per capita globally (Biswas & Tortajada, 2018; Liu et al., 2017). Water demand from different industrial sectors, agricultural production and households will increase simultaneously, which inherently leads to competition between uses (Markosyan et al., 2021). This competition can become more intense during the dry and hot seasons. Water availability becomes limited, while the demand increases for different purposes, such as irrigation, cooling and recreation (House-Peters & Chang, 2011).

1.2.2 Availability of Natural Freshwater Resources

The two main natural resources for fresh water are surface water, for example rivers and lakes, and groundwater. To a lesser extent, natural springs and seawater, after desalination, are employed as freshwater resources (CDC, 2022; Murray-Playfair, 2023; Schmitt & Mendret, 2022; VMM, 2019). Surface water and groundwater are largely fed by precipitation. The inter-annual variability of rainfall consequently translates into variability in river flow and the recharge of aquifers, both temporally and spatially. High water availability can for example coincide with the period of lowest water demand and vice-versa, with water shortages as a result (Green, 2016; Steduto et al., 2012).

Timely replenishment of groundwater systems is necessary to avoid complete depletion. Excessive aquifer withdrawals are taking place globally, leading to a reduced availability of groundwater for consumptive uses. Reduced river flows and degraded groundwater quality arise from those unsustainable extractions as well, worsening natural water availability. About 30% of the world's largest groundwater systems are being exploited unsustainably (Fienen & Arshad, 2016; Zhongming et al., 2021).

Climate change is expected to alter the availability of natural freshwater resources on both a spatial and temporal scale. Temperatures are projected to increase in most part of the world, with complex effects on meteorological cycles. Extreme weather phenomena, such as droughts, will become more frequent and more severe (Cook et al., 2018; UN, 2020). Moreover, average precipitation is expected to change in numerous regions, as well as the variability of rain events (Pendergrass et al., 2017). As a result, river flows, runoff and the replenishment of aquifers will follow this variability (Kundzewicz et al., 2008; World Bank, 2016). It is also anticipated that climate change will indirectly affect the quality of surface water and groundwater. Rainfall events after long periods of drought can carry significantly augmented pollution loads (Lee et al., 2002). More intense evaporation will result in higher salinity levels of groundwater, deteriorating its quality for consumptive use (Barbieri et al., 2021).

1.2.3 Inadequate Water Management

Water scarcity is often divided in physical and economic water scarcity. Physical water scarcity refers to the lack of freshwater to meet human and environmental demands simultaneously. Economic water scarcity is caused by the human insufficiency to meet local water demands, even though water is physically available. Insufficient water infrastructure development, often in arid regions, are an important factor to this, or water distribution may be inequitable where infrastructure actually does exist. Social discrimination can also attribute to economic water scarcity, when certain groups are favored by governments. Adequate water management is, thus, essential in securing urban fresh water supplies (Chakkaravarthy & Balakrishnan, 2019; Liu et al., 2017; Molden, 2007).

A circular water economy is the sustainable water management *par excellence*, as it optimizes water resources by maximizing wastewater recycling. The reuse of wastewater builds resilience against water shortages by reducing the dependency on natural water resources (Brears, 2020; Voulvoulis, 2018; Qadir et al., 2020). Unfortunately, urban water management is generally done in a linear scheme, starting with the extraction of fresh water from natural resources, such as rivers, lakes and aquifers. First, drinking water production facilities purify natural raw water (*e.g.*, surface water and groundwater) up to drinking water quality. This potable water is then used in numerous applications, ranging from drinking and showering to flushing of toilets. Hereafter, the wastewater is either discharged directly into surface water or sent to a wastewater treatment plant. The effluent from wastewater plants is most often discharged into surface water as well. Only in some cases, this effluent is directly reused or indirectly recovered by recharging of approximate aquifers (Jones et al., 2021).

1.3 Decentralized Greywater Treatment for Reuse

Urban areas are, as mentioned before, particularly vulnerable to water shortages. The term "consumptive use of water" refers to the portion of withdrawn water that becomes unavailable for re-use, mainly due to processes like evapotranspiration, incorporation into products or direct drainage into the sea. Interestingly, domestic water usage has a minimal consumptive use, implying that the majority of the water is returned to the sewage system after being used. Thus, within the same location, wastewater is generated in comparable quantities to the demand for clean water (Steduto et al., 2012). This section discusses the potential of decentralized wastewater treatment in cities, coupled with the concept of source separation. In centralized wastewater treatment, all wastewater sources within a certain area are connected to a sewage grid an redirected to a centralized wastewater treatment plant. Decentralized wastewater treatment refers to installations that work on a smaller scale, such as a singular building or neighborhood, often with the perspective of wastewater reuse.

1.3.1 Source Separation of Domestic Wastewater

Wastewater treatment with the perspective of reuse can be optimized through source separation. Instead of bringing wastewater coming from different sources together, they are kept separate. When source separation is applied to domestic wastewater, generally separation is limited to two streams: blackwater and greywater. Blackwater refers to the wastewater coming from toilets, greywater is the remaining domestic wastewater. Greywater constitutes the largest volumetric fraction of domestic wastewater, generally around 60–70% (Lehtoranta et al., 2022; McConville et al., 2017; Shaikh & Ahammed, 2020).

By implementing source separation, the concentration of pollutants in blackwater can be effectively preserved, while the pollution levels in greywater are prevented from increasing. Water reuse efficiency is higher for slightly polluted water. Keeping greywater, which is much less polluted than blackwater, separate expands the potential for water recovery. On the other hand, keeping contamination concentrated in the blackwater stream, enlarges the efficiency of energy and nutrient recovery (Skambraks et al., 2017).

Additionally, keeping blackwater separated from greywater avoids contamination of organic pollutants, pathogens and micropollutants, such as pharmaceuticals and hormones (Kujawa-Roeleveld & Zeeman, 2006). Wastewater that has been in contact with fecal matter has a lower potential for economic reuse, due to the low acceptance of the public (Otterpohl et al., 2002).

1.3.2 Decentralized Treatment

Urban wastewater management is, generally, done in a centralized way. Domestic wastewater is collected in an extensive sewage network to be sent to a central wastewater treatment facility. The implementation of source separation onto an existing centralized system would require major investments and substantial road works. Hence, source separation is often applied simultaneously with decentralization of wastewater treatment (Jeong et al., 2018; Larsen & Gujer, 2013).

Another important advantage of treating greywater (or wastewater in general) in a decentralized manner is that reclamation of clean water occurs near the point of wastewater generation. Transport of water is minimized, which limits water and energy losses over pipelines. It is estimated that decentralized greywater reuse facilities have potentially a lower energy demand (between 0.246–0.970 kWh/m^3 wastewater) than centralized systems (between 1.224–1.914 kWh/m^3 wastewater). Greenhouse gas emissions are also estimated to be lower for decentralized (0.72–0.83 kg CO_2e/m^3) plants, compared to centralized plants (0–0.33 kg CO_2e/m^3) (Chang et al., 2017). Decentralized infrastructures are often modular systems, which can be more effective in adapting to possible needs for changes in scale. The functionality of buildings and population density change quite often in urban settings, making the flexibility of decentralized systems a supplementary appealing quality (Hoffmann et al., 2020; UN, 2017).

1.3.3 Implications of Decentralization

There are some implications of the decentralized treatment of greywater that should be taken into account during planning. As in centralized treatment facilities, personnel with a certain level of expertise is required for operation and maintenance of the system. Decentralized installations are more abundant and dispersed, thus, adequate organization is necessary to operate all (UN, 2017).

It is crucial to examine the extent of decentralization when considering a greywater system. The decision regarding the scale of the system can have a substantial impact on the financial viability of the installation, primarily due to economies of scale. The size of the pipeline network, as well as the construction and operation of the treatment plant, all contribute to determining the ultimate cost-effectiveness. Besides from the economic perspective, environmental aspects should also be considered when deliberating on the appropriate scale. A larger scale will average out fluctuations in flow and quality of the influent, which can be beneficial for the system's removal performance. The trade-off between different scales should be thoroughly evaluated (Kobayashi et al., 2020).

The implementation of decentralized greywater treatment has the potential to impact the performance of existing centralized wastewater treatment plants by altering both quality and quantity of the influent. The aeration needs of an activated sludge basin depends on the organic carbon, nitrogen and phosphorus load of the influent. Aeration may need to be adjusted in the centralized plant, depending on the degree of decentralization and source separation. The change in influent characteristics can also negatively affect denitrification within the centralized plant, due to insufficient organic carbon levels in the wastewater (Kobayashi et al., 2020; Morandi & Steinmetz, 2019; Sapkota et al., 2015).

1.4 Greywater

Domestic wastewater is divided into blackwater and greywater. As mentioned in Section 1.3.1, greywater is particularly interesting in the context of wastewater reuse, because of its quantitative and qualitative characteristics. Greywater contains only one-third of the total organic content and one-tenth of the total nitrogen content found in the total domestic wastewater stream (Kadewa et al., 2020; Oron et al., 2014). This chapter will discuss the characteristics of greywater, its possible reuse applications, the current legislation concerning greywater reuse, and the most relevant treatment technologies.

1.4.1 Sources of Greywater

All domestic wastewater streams, with the exception of toilet flushing water, are considered to be greywater. Important greywater sources are showers, washbasins, bathtubs, laundry machines, dishwashers, and kitchen sinks. Greywater is often further divided into light and dark greywater, depending on the contaminant concentration. Light greywater is less polluted and contains greywater coming from, *e.g.*, bathtubs and showers. Dark greywater has higher pollutant concentrations and includes wastewater from laundry activities and the kitchen. The attribution of greywater source streams to light or dark greywater varies between authors (Alsulaili & Hamoda, 2015; Khanam & Patidar, 2022). Some even completely exclude kitchen wastewater from the definition of greywater (Tilve, 2014). Total greywater or mixed greywater refers to all the greywater steams combined.

On average, nearly half of the mixed greywater flow in a household originates from the bathroom, including showers, baths, and washbasins. Kitchen wastewater, including the dishwasher and kitchen sink, and laundry wastewater each represent about a quarter of the total greywater flow (Ghaitidak & Yadav, 2013; Gross et al., 2015). Domestic greywater production varies considerably between households, from as low as 15 to 200 liters per day per capita. On average, the range of daily greywater generation is 30–165 L per capita for low-income countries and 88–200 L per capita for high-income countries (Khanam & Patidar, 2022; Oteng-Peprah et al., 2018a). The main factors influencing the amount of greywater produced are natural freshwater availability (see Section 1.2.2), water infrastructure services, financial resources, and water use habits (Gross et al., 2015). Greywater generation follows a diurnal-temporal pattern. During the weekdays, two main peak moments are generally observed: a morning peak between 7:00 and 9:30 and an evening peak between 17:30 and 20:00. In some cases, additional significant flows around noon were observed, suggesting that those people were home for lunch (Birks & Hills, 2007; Eriksson et al., 2009; Gurung et al., 2015; Palmquist & Hanæus, 2005). On weekends, a delay in greywater discharge is observed. One large peak in the afternoon until the evening (14:00–22:00) (Alfiya et al., 2018).

Every greywater source delivers specific constituents to the mixed flow. Bathroom greywater flows consist of soaps, toothpaste, body care products, hairs, skin cells, body fats, sand and clay particles, and microplastics (Noah, 2002; Ziemba et al., 2018). More detrimental pollution can occur in this greywater stream, due to improper user behavior like the disposal of harmful substances (*e.g.*, paints, pesticides, bleach) or the washing of domestic pets (Shaikh & Ahammed, 2020). Wastewater from the kitchen and laundry machines is generally the main contributor to organic carbon and suspended solids in the total domestic greywater flow. Greywater coming from laundry machines consists of chemicals from detergents, solvents, bleaches, paints, oils, fibers, and microplastics from clothing (Noutsopoulos et al., 2018). Kitchen wastewater contains dishwashing detergents, food residue, raw meat washing water, traces of food preservatives, and sand and clay particles (Shaikh & Ahammed, 2020). The contamination of kitchen wastewater is significantly lower when solid kitchen waste is sorted separately for composting rather than being discarded into the sink, often accompanied by a kitchen blender (Kujawa-Roeleveld & Zeeman, 2006). The type of greywater sources contributing to the total greywater flow depends on the function of a building. Different greywater source flows are present in non-residential buildings such as office buildings, schools, or public showers (Gross et al., 2015). Moreover, greywater flows can be kept separated within a building. Treating only light greywater instead of mixed greywater could increase the economic feasibility of a greywater reuse project (Leiva et al., 2021).

1.4.2 Greywater Characteristics

Physical Characteristics

Greywater is often warmer than tap water, due to the need for warm water in activities, such as laundry, washing, and cooking. Consequently, the temperature of greywater generally ranges from 18 °C to 35 °C (Oteng-Peprah et al., 2018a). Regardless of the specific source (*e.g.*, bathroom, kitchen), greywater tends to have a similar average temperature, indicating that the relative contribution of each source does not strongly affect the overall temperature of mixed greywater (Shaikh & Ahammed, 2020). The temperature of greywater exhibits a similar daily pattern as the generated volume of greywater, as fresh greywater enters at a higher temperature (Birks & Hills, 2007). The elevated temperatures of greywater encourage microbiological activity, while simultaneously reducing the solubility of calcium carbonate. This reduction in solubility can result in the undesired precipitation of calcium carbonate in either the greywater storage tank or the pipelines (Khajvand et al., 2022; Olanrewaju & Ilemobade, 2015).

The concentration of total suspended solids can become quite high in greywater (see Table 1.1). Turbidity, as well as suspended solids concentrations, are generally highest in kitchen and laundry greywater. Sources of suspended solids come from fabric fibers, zeolites from detergents, food residuals, and sand from the washing and rinsing of vegetables (Antonopoulou et al., 2013; Bakare et al., 2017; Khajvand et al., 2022; Shaikh & Ahammed, 2020).

Table 1.1: Range of physical, chemical, and biological characteristics of mixed greywater. (TSS = total suspended solids, BOD_5 = 5 day biological oxygen demand, COD = chemical oxygen demand, TN = total nitrogen, TP = total phosphorus). Adapted from Khanam & Patidar (2022) and Oteng-Peprah et al. (2018a).

Parameter		**Range in GW**
Physical characteristics		
Temperature	°C	18 – 35
Turbidity	NTU	21 – 4400
TSS	mg/L	12 – 4250
Chemical characteristics		
pH		5 – 9.7
BOD_5	mg/L	18 – 1395
COD	mg/L	24 – 6563
TN	mg N/L	9 – 58
Ammonia-N	mg N/L	0.8 – 75
Nitrate	mg NO_3^-/L	0.1 – 14
TP	mg N/L	0.4 – 20
Phosphate	mg PO_4^{3-}/L	1 – 23
Microbiological characteristics		
Fecal coliforms	CFU/100 mL	$3.57\times10^3 - 150\times10^6$

Chemical Characteristics

The pH of greywater generally ranges from 5 to 9 (Table 1.1), and is influenced by chemicals, such as fabric softeners, bleaching agents, and disinfectants. Laundry-associated detergents tend to give rise to high alkalinity. Greywater, comprising of relatively high amounts of laundry wastewater, will exhibit a higher pH (Rakesh et al., 2020).

The organic content is highly variable in greywater, as can be seen in Table 1.1. Average COD concentrations are much higher in laundry and kitchen wastewater (594 and 780 mg/L, respectively) than bathroom greywater (121 mg/L) (Shaikh & Ahammed, 2020). This is due to high detergent use in dishwashing and laundry activities and residuals of food and drinks with fat and oil in kitchen wastewater (Ghaitidak & Yadav, 2013). The COD/BOD ratio gives an idea of how biodegradable wastewater is. Greywater with a COD/BOD ratio below 2.5 is relatively easy to biodegrade (Li et al., 2009). Laundry and kitchen greywater are generally easily biodegradable, with average COD/BOD ratios of 1.79 and 1.71, respectively. The average COD/BOD ratios of bathroom and hand basin greywater are higher, 2.03 and 2.81, respectively (Shaikh & Ahammed, 2020). The concentration of surfactants, or surface-active agents, is elevated in greywater compared to mixed domestic wastewater (Oron et al., 2014). Laundry and kitchen wastewater generally have the highest surfactant concentrations (Shaikh & Ahammed, 2020). Surfactants negatively affect the biodegradability of greywater, and have toxic impacts on the environment and human health (Khalil & Liu, 2021; O'Toole et al., 2012). High oil and grease content in greywater may impair biological treatment as well (Shaikh & Ahammed, 2020).

Greywater is responsible for only 3% of the nitrogen load in domestic wastewater, where urine and feces make up 97% of the nitrogen (Otterpohl, 2002). The main source of nitrogen in greywater is food residue; other important sources are protein-containing shampoos, household products containing ammonia, and, in some cases, urine. Nitrogen in wastewater is present as either organic or inorganic nitrogen (Figure 1.2). The inorganic forms include ammonia, nitrite, and nitrate. The predominant form of nitrogen in greywater is organically bonded nitrogen (Leal et al., 2010; Pradhan et al., 2019). Forms of organic nitrogen include amino sugars and proteins and are present either in a dissolved state or as a component of particulate matter. Ammonia concentrations in greywater generally range from 0 to 16.3 mg N/L, and in some cases, up to 41.2 mg N/L (Deng et al., 2020; Khanam & Patidar, 2022). In some cases, bathroom greywater contains particularly high concentrations of ammonia. The habit of some people of urinating in the shower is the suspected reason for this, since urine is quickly converted to ammonia (do Couto et al., 2013). The levels of nitrite and nitrate in greywater are typically in the same range as those found in tap water, *i.e.*, below or around 1 mg N/L. This suggests that the nitrite and nitrate in greywater primarily come from the existing nitrite and nitrate present in the tap water itself (Bodnar et al., 2014; Eriksson et al., 2009; He et al., 2022).

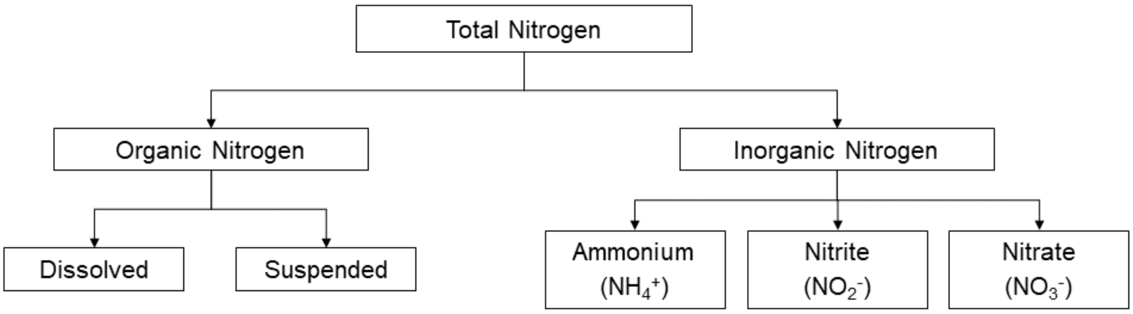

Figure 1.2: Nitrogen forms in wastewater.

Phosphorus content is highest in greywater coming from laundry and kitchen wastewater (Shaikh & Ahammed, 2020). Some countries installed bans on phosphate-containing detergents, which reduced the amount of phosphorus in domestic wastewater (Kasak et al., 2011).

Microbial Characteristics

Different microorganisms can be present in greywater, including bacteria, helminths, and protozoa. The main sources of these microorganisms are bathing and showering, childcare, hand washing after toilet use, washing diapers, and rinsing raw meat and vegetables (Khajvand et al., 2022; Oteng-Peprah et al., 2018b). Coliforms are frequently found in greywater, representing fecal contamination. Other pathogenic bacteria that have been observed in greywater include *Pseudomonas*, *Legionella*, *Escherichia coli*, and *Campylobacter* (Rakesh et al., 2020). Limiting stagnation of untreated greywater and disinfection (*e.g.*, through chlorination) before reuse are essential to minimize the spread of pathogens and human infection (Blanky et al., 2017; Dixon et al., 2000).

1.4.3 Non-Potable Reuse Applications

Reuse of treated greywater can be subdivided into three types: direct potable, indirect potable, and non-potable reuse (Capodaglio, 2021). The first type includes applications where potable fresh water is necessary, like showering (due to the high chance of oral intake) or kitchen tap water. The treated greywater is directly linked to a distribution pipe or tap. Therefore, greywater purification for direct potable reuse has the strictest quality requirements. Indirect potable reuse refers to the discharge of treated greywater into a water body or groundwater infiltration that supplies a water purification plant that produces drinking water. The intermediate water reservoir assures an extra safety margin through dilution and possible natural remediation processes. Non-potable reuse of greywater is most widely implemented. This includes domestic, urban, industrial, and agricultural applications, as well as the recharge of water bodies that are not targeted for potable water supply (Capodaglio, 2021). The success of greywater reuse applications, especially for direct or indirect potable reuse, is influenced by the public's acceptance of this (Al-Khatib et al., 2022; Thaher et al., 2020).

Greywater, used for reclamation, usually excludes the kitchen greywater, as it poses a higher pathogenic risk and contains a higher contamination level of chemical and grease.

Toilet Flushing

The use of treated greywater for toilet flushing has been done since the 1990s (Jeppesen, 1996; Nolde, 2000). It is the most common application for greywater reuse, and can reduce domestic water use by 30% (Mahmoudi et al., 2021). Toilet flushing is a very interesting application for reclaimed greywater, because quality requirements are less strict and toilets are generally located near the greywater source. Important quality parameters are surfactant content, odor, color, and factors influencing microbial growth (Zhu et al., 2018). High surfactant levels in toilet flushing water cause foaming during flushing. Toilet flushing creates an aerosol spray that can accommodate the spread of pathogens (Hamilton et al., 2018).

In the context of saving water, vacuum toilets are on the rise. They use only 0.5–1.2 L per flush, a significant difference from the 5–9 L per flush of a regular toilet (Gao et al., 2019). In certain cases, greywater production might exceed the water demand for toilet flushing by a great extent (Zadeh et al., 2013). In such situations, it becomes interesting to explore alternative applications for utilizing the excess greywater for non-potable reuse purposes.

Irrigation

Irrigation of green spaces is another common greywater reuse application. Greywater can be used for irrigation of urban parks, sports fields, green roofs, or even agricultural crops (Mahmoudi et al., 2021). In certain urban areas, irrigation of lawn and turf grass causes the largest water demands. This becomes particularly important during the summer months in arid and semi-arid regions (Capodaglio, 2021). Irrigating green spaces can reduce the potable water demand by up to 50% (Mahmoudi et al., 2021). Next to irrigation, the greywater can be a source of nutrients for the plants, depending on the degree of greywater purification. This combined effect of irrigation and fertilization is termed fertigation (Capodaglio, 2021; Ramey et al., 2017), but the fertilizing potential of greywater might be low because of the limited nutrient content (Finley et al., 2009).

Eutrophication of the environment is an important risk related to irrigation with greywater. Extensive knowledge of the greywater quality and the nutrient requirements of the plants and soil is essential to limiting this (Filali et al., 2022). Soil properties can also be negatively affected by greywater, such as an increase in the alkalinity, hardness, and salinity of the soil and making it water repellent, due to the accumulation of surfactants (Wiel-Shafran et al., 2006). The use of reclaimed greywater for irrigation of agricultural crops already takes place in some developed and developing countries around the world (Radingoana et al., 2020). Human health risks arise when greywater is used for irrigation of agricultural crops or lawns with unrestricted human access, depending on the quality of the greywater. Treatment against pathogenic microorganisms and the removal of chemical contaminants have become essential (Schoen et al., 2017; Sheikh et al., 2018). Limiting greywater reuse to crops where the water does not have to come into contact with the plant, such as vineyards and certain orchards, can help mitigate human health risks. Crops that undergo disinfection during food processing also mitigate the pathogenic hazard of irrigation with greywater (Sheikh et al., 2018).

In some countries, untreated greywater is used for garden irrigation, mainly due to financial reasons (McIlwaine & Redwood, 2010). Irrigational use of untreated greywater is not recommended, because of the risks it may pose to human health and the environment (Maimon et al., 2010).

Other Greywater Applications

Greywater reuse can be extended to various non-potable applications, including car and road washing, fire extinction, and ornamental uses (*e.g.*, in fountains, waterfalls, and ponds) (Mahmoudi et al., 2021). Local water needs should be investigated and coupled with the production of greywater *in situ*. When the demand for water is low or scarce in the area, the valorization of treated greywater can always be maximized through infiltration for groundwater recharge (Alcalde-Sanz & Gawlik, 2017).

1.4.4 Legislation & Standards for Greywater Recovery

International greywater reuse guidelines have not yet been established. Instead, many countries have relied on general wastewater reuse guidelines for the reuse of greywater (Yoonus & Al-Ghamdi, 2020). These guidelines, which were primarily designed for agricultural or urban irrigation purposes, have been implemented in both developed nations such as Australia, Germany, Japan, and the UK, as well as in developing nations like Jordan and Tunisia. During the drafting of the guidelines for greywater irrigation, the primary focus was placed on human health rather than environmental protection (Maimon et al., 2010). Some countries have established specific limit values for other types of greywater or wastewater reuse, such as toilet flushing (Filali et al., 2022). The European Union released a framework outlining quality requirements for the practice of water reuse in agricultural irrigation and aquifer recharge (Alcalde-Sanz & Gawlik, 2017). The responsibility lies with individual member states to further outline these guidelines to a finer degree. The Italian guidelines for wastewater reuse stand out as some of the most comprehensive in existing literature (Reynaert et al., 2020). For this reason, they are included as European examples in Table 1.2. In Flanders, the government is currently in the process of formulating guidelines to provide a legal framework for the utilization of wastewater for agricultural irrigation as well as other irrigation purposes (Van Gils et al., 2023).

Adherence to these standards does not necessarily guarantee the safety of reclaimed water for reuse. The existing guidelines and regulations for greywater reuse only consider a limited number of parameters. A selection of parameters, included in some guidelines for greywater reuse, are listed in Table 1.2. This raises the question of whether contaminants of emerging concern should be incorporated into these guidelines. Currently, even the most stringent drinking water and wastewater regulations often fail to address these emerging contaminants (Capodaglio, 2021; Khajvand et al., 2022).

Greywater reuse is often not encouraged and, in some cases, even discouraged, due to the complex application procedures required to obtain a permit. However, administrative mechanisms play an important role in ensuring maximal protection of human health and the environment (Vuppaladadiyam et al., 2019).

Table 1.2: Guidelines on water quality parameters for wastewater reuse in different countries. Adapted from Reynaert et al. (2020). (UR = Unrestricted Reuse)

		ISO 30500	**Italy**	**China**	**South Africa**
Reuse purpose		UR	Toilet flushing	Toilet flushing	Toilet flushing
pH		6 – 9	6 – 9.5	6 – 9	6 – 9
Turbidity	NTU			$\leqslant 5$	$\leqslant 10$ (5*)
TSS	mg/L	$\leqslant 10$	$\leqslant 10$	$\leqslant 1500$	$\leqslant 30$ (10*)
COD	mg/L	$\leqslant 50$	$\leqslant 100$		50*
BOD$_5$	mg/L		$\leqslant 20$	$\leqslant 10$	$\leqslant 20$ (5*)
TOC	mg C/L			$\leqslant 10$	$\leqslant 20$ (5*)
TN	mg N/L		$\leqslant 15$		
TN removal	%	$\geqslant 70$			
Ammonia	mg N/L		$\leqslant 1.6$	$\leqslant 7.8$	
TP	mg P/L		$\leqslant 0.2$		
TP removal	%	$\geqslant 80$			
Total coliforms	CFU/100 mL			$\leqslant 0.3$	
E. coli	CFU/100 mL	$\leqslant 1$	$\leqslant 100$**		$\leqslant 10$ (1*)

* (Additional) guideline for the average concentration; ** 80% of samples should be $\leqslant 10$ CFU/100 mL.

1.4.5 Treatment Technologies

Different treatment systems exist to purify greywater. Financial cost, environmental impact, public perception, and the desired effluent quality have to be taken into consideration during the selection of an appropriate treatment system. The required effluent quality varies depending on the type of greywater reuse and the applicable legislation. To reduce cost and energy consumption, greywater should only be treated to the necessary quality. This principle, known as fit-for-purpose treatment, plays a crucial role in sustainable water management. Overtreatment of greywater can lead to increased costs without additional benefits, thereby discouraging the implementation of greywater reuse systems (Capodaglio, 2021; Schimmoller & Kealy, 2014). On the other hand, regulations are becoming progressively stricter, requiring treatment technologies to continually meet stringent effluent quality norms. To fulfill these requirements, a combination of various technologies, along with disinfection techniques, may be necessary (He et al., 2022; Oh et al., 2018). This section provides a brief overview of some commonly employed techniques for greywater treatment.

Physical Treatment Systems

Filtration is often only used as a pre- or post-treatment step rather than as the whole greywater treatment, because it does not remove surfactants and nutrients very well. The performance of a filtration system is dependent on the particle size distribution of greywater and the porosity of the filter. A smaller porosity of the filter generally leads to better effluent quality (Albalawneh & Chang, 2015). Various filtration systems exist: mulch systems, sand bed filters, and membrane filters. The treatment performance of mulch and sand filters is limited, with only 25–30% of COD and little coliform removal (Chaillou et al., 2011; Zuma et al., 2009). Intermittent feeding of greywater on sand filters causes the formation of a biologically active top layer. These systems obtain higher COD removal (up to 80%) due to the combination of biological and physical treatment processes (Ghaitidak & Yadav, 2013).

Adsorption of organics and other components from greywater is another type of physical treatment. Frequently used sorbents are activated carbon, natural zeolite, and iron (nano zero-valent) (He et al., 2022). As with filtration, biological processes can be integrated into the physical treatment of adsorption. Biologically activated carbon promotes the growth of microorganisms on the filter bed. These microorganisms will enhance the removal and biological degradation of pollutants present in the greywater. Adsorption is a great complementary post-treatment technique in greywater treatment for greywater sources with highly variable flows and organic concentrations (Hess & Morgenroth, 2021).

Chemical Treatment Systems

Different chemical treatment technologies have been developed for the treatment of greywater, including precipitation, coagulation, (advanced) oxidation processes, electrolysis, and ion exchange techniques (Awasthi et al., 2023; He et al., 2022; Maimon & Gross, 2018). These chemical processes may be sufficient for the treatment of low-strength reuse, but are most often not suitable as standalone treatment techniques for higher-strength greywater (Li et al., 2009). Stringent guidelines for reuse are not met through chemical treatment solutions, due to the recalcitrant nature of a portion of the greywater (Pidou et al., 2008).

Biological Treatment Systems

Constructed wetlands (CWs) have been gaining attention as a treatment technique for domestic wastewater, because of their cost-effectiveness and limited environmental impact (Sijimol & Joseph, 2021). Constructed wetlands are engineered in a way to effectively utilize the natural processes of a wetland system, comprising its vegetation, soil, and associated microbial communities (Mueller et al., 2003). Appropriate selection of plant species, substrate material, hydraulic retention time, and greywater flow regime through the system are important factors in a CW, influencing the pollutant removal efficiency. A combination of physical (sedimentation and filtration), chemical (reaction and adsorption), and biological processes (microbial assimilation) takes place in the constructed wetland system and provides good removal of organics and suspended material (Vymazal, 2010). The removal of nitrogen and phosphorus in CWs remains poor. The CWs are a simple technique that requires little expertise for operation and maintenance (Singh et al., 2018). Sufficient surface area is required for constructed wetlands, especially in colder climates, which makes it a less suitable technology for urban areas where space is limited. Examples of space-saving alternatives to constructed wetlands for urban areas are green walls and green roofs.

As in CWs, nutrient removal remains an obstacle in these natural systems (Addo-Bankas et al., 2021). Rather than being used solely as a treatment technique, green walls and roofs can utilize greywater as a source of water (and to a certain extent of nutrients), as discussed in Section 1.4.3. Drainage water from these green infrastructures may not always be of high quality, but this is of less importance since the greywater has already been utilized at this point.

Physical treatment processes are usually only implemented as a post-treatment step (Shaikh & Ahammed, 2020), and chemical treatment processes are often not enough to meet guidelines for non-potable reuse, especially for high-strength greywater (Li et al., 2009). Constructed wetlands can, in combination with physical and/or chemical treatment, obtain great greywater treatment efficiencies, but lack in the removal of nutrients. Biological treatment systems can help overcome the limitations of the previously mentioned technologies. Biological greywater treatment can be carried out under either aerobic, anaerobic, or combined conditions. Anaerobic treatment of greywater is favored in certain cases, but its application is often limited, due to inadequate surfactant removal and the emission of methane. Aerobic systems are most often applied for greywater treatment. Some examples are rotating biological contactors (RBC), sequencing batch reactors (SBR), moving bed biofilm reactors (MBBR), and membrane bioreactors (MBR) (Khalil & Liu, 2021).

The most promising biological treatment systems are membrane-based bioreactors. It is a very compact technology because the need for a post-treatment filtration step is eliminated. The reduced footprint of MBRs is especially advantageous in cities, where space is limited. With the development of new membrane materials, membrane systems are becoming more and more economically viable while efficiently achieving non-potable reuse guidelines (Albalawneh & Chang, 2015; Wu, 2019; Zhu et al., 2018).

1.5 Membrane Bioreactors (MBRs)

1.5.1 Working Principle

Wastewater treatment within membrane bioreactors (MBRs), similarly to conventional activated sludge (CAS) plants, takes place in a bioreactor containing microorganisms. The difference between MBRs and CAS lays in the liquid-solid separation. In MBRs, separation of the effluent from the mixed liquor occurs via pressure-driven membrane filtration.

Biological Treatment Processes

Biological wastewater treatment is effectuated by a variety of microorganisms, the activated sludge, which ensure pollutant removal through biodegradation. The microbial community in bioreactors is strongly determined by the influent characteristics (Miura et al., 2007). Breakdown of organics, nitrogen and other pollutants is accomplished through their consumption by specific microorganisms. Organic carbon in wastewater is primarily consumed by aerobic heterotrophic bacteria that transform organic matter through their aerobic metabolism into new biomass, carbon dioxide, water and minerals (Rabaey, 2019). Nitrogen removal from wastewater mainly occurs through autotrophic nitrification, followed by heterotrophic denitrification. Autotrophic nitrification is two-step aerobic process where ammonia is converted to nitrate. First, ammonia is oxidized to nitrite by chemolithoautotrophic ammonia-oxidizing bacteria (AOB). *Nitrosomonas* and *Nitrosospira* are the dominant groups of AOB in wastewater sludge (van Loosdrecht & Jetten, 1998). Ammonia oxidation is the rate-limiting step of nitrification (Li et al., 2019). Subsequently, nitrite is oxidized to nitrate by nitrite-oxidizing bacteria (NOB). Examples of NOB in wastewater are *Nitrobacter* and *Nitrospira*. The nitrifiers are slow growers because the oxidation of inorganic material is less energy efficient than the oxidation of organic carbon sources. They are also more sensitive to toxic compounds (Liu, 2008). The range of specific ammonia oxidation rates (SAOR) found in the literature for aerated MBRs is 0.51–18.75 mg NH_4^+/g SS·h (Dytczak et al., 2008; Li et al., 2006; Scheumann & Kraume, 2009). Denitrification is the reduction of nitrate towards nitrogen gas. This conversion necessitates anoxic conditions, where the dissolved oxygen concentration is very low, and the presence of an electron donor (*e.g.*, organic carbon) (van Loosdrecht & Jetten, 1998). Different heterotrophic bacteria are involved in denitrification in wastewater, mainly facultative aerobes, like certain *Pseudomonas* and *Alcaligenes* species (Liu, 2008; Rabaey, 2019).

To obtain good removal of nitrogen, the bioreactor often consists of an anoxic zone for denitrification, followed by an aerobic zone where ammonia is oxidized. Recirculation back to the anoxic tank ensures adequate nitrification-denitrification (Phan et al., 2014). Denitrification also occurs to a limited extent within the aerated tank. The inner zones of granular flocs are anoxic due to diffusion limitations for dissolved oxygen (Layer et al., 2020). Denitrification can be further enhanced in the aerated tank by introducing a support material for biofilm attachment into the reactor (Ivanovic & Leiknes, 2012). Within well-established biofilms, anoxic zones arise in the inner regions, providing the essential environment for denitrification (Leyva-Díaz et al., 2015). This can eliminate the need for an anoxic tank for denitrification (Ivanovic & Leiknes, 2012). More detailed explanations on this will follow in Section 1.5.5.

Membrane Separation

Membrane bioreactors are operated in either a side-stream or submerged membrane configuration. In the side-stream configuration, the membrane modules are placed external to the bioreactor. Mixed liquor is pumped in cross-flow (*i.e.*, tangential to the membrane surface) and concentrated retentate is subsequently returned to the bioreactor (Figure 1.3). The tangential flow hydrodynamically promotes fouling control (Rahman et al., 2023). In submerged MBRs, the membrane module can either be integrated within the bioreactor or is placed in a separate tank containing mixed liquor. The submerged configuration corresponds with dead-end filtration, where all the feed passes through the membrane. Fouling is more problematic with dead-end filtration. Because of this, the membrane modules are aerated with (coarse) bubbles, to creates a sheer stress that displaces the accumulated fouling layer (Galinha et al., 2018). Submerged MBRs require less power per volume of wastewater treated, but need a larger membrane area. This type is MBR is compatible with lower strength wastewater with good filterability (Visvanathan, 2009), such as greywater.

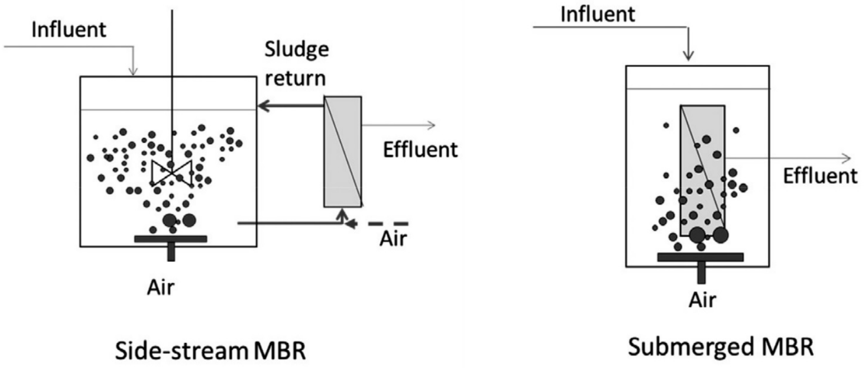

Figure 1.3: Membrane bioreactor configurations: the side-stream membrane module with cross-flow filtration (left) and the submerged membrane module, integrated within the bioreactor, with dead-end filtration (right). Retrieved from Galinha et al. (2018).

The MBRs, in the context of wastewater treatment, are commonly operated in either a hollow fiber, flat sheet or tubular membrane module (Figure 1.4) (Krzeminski et al., 2012). The hollow fiber configuration makes use of many vertically placed membrane tubes, the hollow fibers. Feed water flows across the membranes to the inside of the individual fibers. The permeate is collected from the fiber ends and brought outside the tank. The fibers are typically attached to a frame with some flexibility to enable lateral movement, in the flow of the air bubbles (Judd, 2021a). Hollow fiber membranes are able to handle high crossflow velocities, making them well-suited for cross-flow MBR wastewater treatment (Asif & Zhang, 2021). Fouling can become problematic with hollow fiber membranes in submerged MBRs, due to biofilm formation on the membranes (Shi et al., 2022). The flat sheet module consists of multiple hollow membrane panels, usually with a rectangular geometry. The panels comprise of the membrane, a thin spacer, a rigid plastic backing plate and an empty internal space. Water flow through the panel is from outside to the internal space and is collected from an outlet tube (Judd, 2023). Flat sheet membranes are mostly used in submerged MBRs, where it is relatively easy to clean (via air sparging) and replace them (Asif & Zhang, 2021). Tubular membrane modules are used for side-stream MBR configurations. A module consists of a group of tubes, housed in a cylindrical casing. The water flows from the module casing into the membrane tubes (Judd, 2021b).

Implications of MBRs for Greywater Treatment

In the context of the decentralized treatment of greywater for reuse, MBRs offer two important advantages: a high quality effluent and a low space footprint. The MBRs obtain a higher quality effluent compared to CAS plants, because of their higher mixed liquor suspended solids (MLSS) concentrations. This also promotes the development of the slow growing nitrifiers, enhancing ammonia removal (Judd, 2008). The MLSS concentrations in MBRs are generally between 10 and 20 g/L, while CAS plants operate at around 4 g/L. For greywater treatment, however, high treatment efficiencies can be achieved at lower MLSS, due to the relatively low organic load of

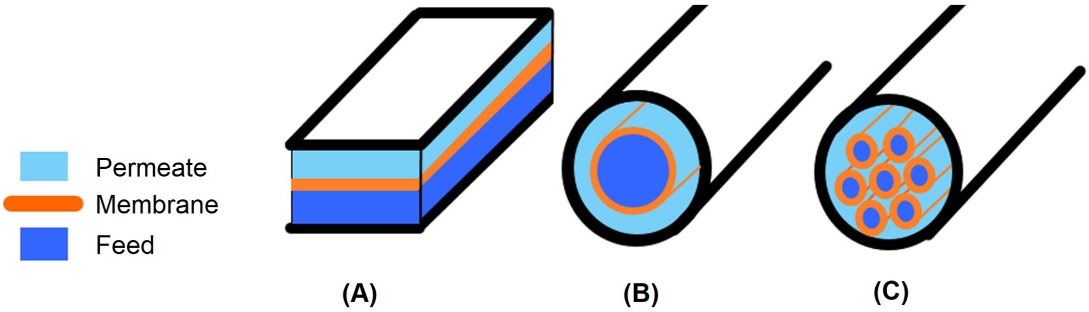

Figure 1.4: Membrane module types: flat sheet (A), tubular (B) and hollow fiber (C). Adapted from Warsinger et al. (2018).

greywater (Liberman et al., 2016). The membranes filter out the particles bigger than their pore size, such as bacteria and most suspended solids, further enhancing effluent quality (Cecconet et al., 2019; Fikri et al., 2023). Space footprint is largely reduced in MBRs compared to CAS systems, because the need for a sedimentation tank is omitted. Furthermore, the higher MLSS concentrations in MBRs enable a size reduction of the reactor itself (Judd, 2008).

1.5.2 Membrane Characteristics

Pore Size

Based on the transport mechanism through the membrane, membranes used in wastewater treatment can be classified into two groups: porous and non-porous membranes. Non-porous (or dense) membranes rely on differences in solubility and diffusivity of analytes in the membrane and require high operating pressures ($>$ 1000 kPa). These mechanisms are used in tight end nanofiltration (NF) and reverse osmosis (RO) membranes (Shirazi et al., 2010). In porous membranes, separation occurs through sieving, straining or size exclusion (Shirazi et al., 2010), resulting in much lower operating pressures (10–1000 kPa). Examples of porous membranes are microfiltration (MF), ultrafiltration (UF) and loose end NF membranes, ordered with decreasing pore size. Microfilters have a pore size larger than 50–100 nm and filter out bacteria, suspended particles and cellular materials. Nanofiltration membranes (pore size 2–50 nm) additionally retain large biomolecules, such as proteins and viruses (Nagy, 2019). Loose end NF membranes have a higher molecular cut-off weight (around 500–2000 Da) than tight end NF ($<$ 500 Da) membranes, and are used to remove divalent ions from water (Guo et al., 2021). The MBR systems typically employ MF and UF membranes, due to their ability to effectively capture bacterial sludge flocs and nearly all suspended solids from a bioreactor. By utilizing these membranes, MBRs can produce a high effluent quality, while also minimizing power consumption required for the operating pressure (Iorhemen et al., 2016).

Membrane Material

There are two main categories of membrane, based upon the membrane material: polymeric membranes and ceramic membranes. Polymeric membranes have been the predominant type in wastewater treatment applications. They offer good mechanical stability and are less expensive in purchase, partly due to easy fabrication of pores in polymeric membranes (Antony & Leslie, 2011; Mutamim et al., 2013). Polymers often used in membranes for water treatment purposes are polyvinylidene fluoride (PVDF), polyacrylonitrile (PAN), polyethylene (PE), polysulfone (PS) and polyethersulfone (PES) (Iorhemen et al., 2016). However, polymeric membranes suffer from substantial fouling, due to their hydrophobic nature. They are also more susceptible to chemical degradation induced by oxidants and chlorine, which are employed during membrane operation for disinfection and cleaning (Antony & Leslie, 2011; Iorhemen et al., 2016). In response to these challenges, significant focus has been directed toward the enhancement of polymeric membranes through various modifications. The introduction of nanomaterials into the polymer membrane structure has proven effective in enhancing many membrane properties, such as its fouling resistance, hydrophilicity, water permeability and mechanical strength (Warsinger et al., 2018). Polymeric membranes modified with nanomaterials are also better suited for treating wastewater containing oil, such as kitchen greywater (Bassyouni et al., 2019). The synthesis of nanoparticle-polymeric membranes has become well-developed, making it a cost-effective technology (Bassyouni et al., 2019).

The development of ceramic membranes for water treatment has been on the rise in the past years (Asif & Zhang, 2021). These membranes are considerably more hydrophilic than polymeric membranes, resulting in significantly reduced fouling tendencies and a largely enhanced cleaning efficiency (Alresheedi et al., 2019; Lee & Kim, 2014). Ceramic membranes possess a high chemical inertness, rendering them resistant against degradation caused by harsh chemicals used for membrane cleaning (Arumugham et al., 2021; Iorhemen et al., 2016). Being fully oxidized, ceramic membranes possess a high oxidative resistance. This offers the opportunity to incorporate catalytic ozonation processes on the membrane surface or employ ozonation as a pretreatment step, further mitigating fouling (Byun et al., 2011; Warsinger et al., 2018). The structure of ceramic membranes comprises a macroporous support layer, an intermediate layer and a selective microporous top layer. Common materials employed for the top layer of ceramic membranes used in water treatment are alumina (Al_2O_3), zirconia (ZrO_2) and titania (TiO_2). The application of other materials, such as zeolite and fly, ash are also being explored (Arumugham et al., 2021). The primary drawback of ceramic membranes has been the cost of the membranes themself, often rendering them economically unfeasible for application in MBRs (Iorhemen et al., 2016; Warsinger et al., 2018). Nonetheless, lower operating costs associated with ceramic membranes can enable the amortization of initial investment costs, thereby establishing the economic viability of such systems (Jarvis et al., 2022).

1.5.3 Membrane Fouling

Mechanisms of Membrane Fouling

Membrane fouling, the most important drawback of MBRs, is a phenomenon where the permeability of the membrane deteriorates over time, due to the accumulation of solutes and suspended particles. Soluble microbial products and extracellular polymeric substances (EPS), specifically, are considered to have a major contribution to membrane fouling (Ivanovic & Leiknes, 2012). The complete mechanisms underlying membrane fouling remain not fully understood, although three main processes can be identified: internal fouling, external fouling, and concentration polarization (Du et al., 2020; Trinh et al., 2020). Internal fouling refers to deposition and adsorption of smaller foulants (*e.g.*, solutes and colloids) withing membrane matrix (Trinh et al., 2020). External fouling refers to deposition on the membrane surface by foulants, such as macromolecules, colloids and suspended particles (Du et al., 2020). Two types of external fouling mechanisms are extensively described in literature: cake layer formation and gel layer formation. The initial development of the cake layer is primarily linked to the deposition of sludge flocs onto the membrane surface, initiating the formation of biofilms. Subsequently, the dominating process driving the cake layer's further progression is the deposition of smaller colloids (Wang & Tarabara, 2008; Wu, 2019; Yao et al., 2018). Gel layer formation can significantly diminish the membrane's permeability. Gelation of colloidal and dissolved compounds creates a thin and porous gel layer, characterized by a high specific filtration resistance (Du et al., 2020; Hong et al., 2014). The specific filtration resistance of gel layers can reach nearly 100 times higher that of cake layers (Hong et al., 2014). The paradox of such gel layers being both porous and yet possessing a substantial specific filtration resistance is not yet fully understood (Chen et al., 2016b). Concentration polarization can also increase the membrane resistance by gathering solids and ions within a narrow liquid layer, adjacent to the membrane surface (Du et al., 2020). When the concentration of solutes and/or particles near the membrane surface surpasses that within the main bulk solution, this leads to a reverse diffusion process back into the bulk solution (Guo et al., 2012).

Mitigation of Membrane Fouling

Membrane fouling can be limited in a preventive manner. One example is altering specific membrane properties. As fouling is more pronounced with hydrophobic membranes, the membrane surface can be coated with hydrophilic substances. The membrane surface can also be precoated with adsorptive particles, effectively reducing direct contact between the foulants and the membrane (Le-Clech et al., 2006). Operating conditions of the MBR system, such as the permeate flux, aeration, SRT, HRT or OLR, can be optimized to limit membrane fouling, as discussed in Section 1.5.4. The addition of coagulants, flocculants or adsorbents into the MBR feed water can also effectively temper membrane fouling (Du et al., 2020; Torretta et al., 2013; Wu et al., 2006).

Despite the existence of effective preventive strategies against membrane fouling, its occurrence is inevitable in wastewater treatment with MBRs. To maintain continuous operation, recovery of the membrane permeability following fouling is crucial. In practice, the categorization of membrane fouling is often done in terms of permeability recovery. This includes reversible fouling (removed by physical cleaning), irreversible fouling (removed by chemical cleaning) and irrecoverable fouling (not removed by any cleaning regime) (Du et al., 2020; Judd, 2008). There are different types of physical cleaning of membranes. The simplest approach is intermittent membrane operation, wherein regular relaxation intervals are introduced, during which permeate pumps are turned off. During these relaxation phases, concentrated foulants diffuse away from the membrane due to a concentration gradient, moving back into the bulk of the reactor (Hong et al., 2002). The relaxation time should be long enough, as excessively short intervals can exacerbate fouling (Chua et al., 2002). Conversely, longer relaxation times lead to a reduction in overall membrane flux. Another widely utilized physical cleaning technique is air scouring. Rising air bubbles create a strong sheer force across the membrane surface, displacing the accumulated foulant layer (Du et al., 2020). The fine bubble air flow, employed for biomass aeration, can serve this purpose. Alternatively, an additional air diffusion pipe, releasing coarser bubbles, may be installed within the reactor, offering more effective foulant removal (Rabaey, 2019). Backwashing, involving a reversal in the direction of membrane flux, is also capable of addressing reversible fouling (Cui et al., 2018). A scouring material can also be introduced within the bioreactor, with the specific purpose of scraping the deposited foulants from the membrane surface (Siembida et al., 2010). In moving bed MBRs, biomass carriers can fulfill this purpose, aside from increasing denitrification in the reactor (see Section 1.5.5). In laboratory experiments utilizing small MBR setups, a soft brush can be employed to delicately remove the accumulation from the membrane (Jiang et al., 2020). This can be upscaled to large automated brushing systems (Song et al., 2022).

Chemical cleaning is necessary in case of irreversible fouling, where fouling is so entrenched that it cannot be removed hydraulically, *i.e.*, through physical cleaning. Chemical cleaning can be executed either *in situ*, by replacing the greywater influent with a cleaning solution, or *ex situ*, where the membrane modules are removed from the MBR and submerged in a separate tank filled with a cleaning solution (Shi et al., 2014). Various types of chemicals, such as acids, bases, oxidants or chelates, are used to remove foulants from membranes. Typically, a mixture of those compounds is used for optimal results. The sequence of application of cleaning agents has an influence on the cleaning efficiency (Porcelli & Judd, 2010). The cleaning chemicals act by displacing the foulants, changing their morphology or altering the chemistry of the fouling layer (Li & Elimelech, 2004).

1.5.4 Operating Conditions

Temperature and pH

Microbial growth and activity are significantly affected by temperature (Price & Sowers, 2004). The removal of COD, nitrogen and most hydrophobic compounds was found to be stable in an MBR with temperatures between 10–35 °C (Hai et al., 2011). This corresponds with the usual temperature range of greywater (Oteng-Peprah et al., 2018a). Nitrification is sensitive to changes in the pH and is most optimal in the range of 7–8 (Tarre & Green, 2004).

Permeate Flux

The permeate flux is an important parameter in MBR operation and is expressed as the volume of permeate passing through the membrane per unit time and per unit membrane area (Quezada et al., 2021). There exists a maximal permeate flux, the critical flux, where the deposition of particles and colloids becomes significant (Diez et al., 2014). The MBRs for wastewater treatment are generally operated at a lower flux, the sustainable flux. The sustainable flux keeps membrane fouling at an economic and environmental sustainable level and prevents the need for chemical cleaning (Iorhemen et al., 2016). Centralized municipal MBR wastewater treatment plants usually have permeate fluxes between 15–30 L/m^{2}h. Greywater contains less organics than mixed domestic wastewater, resulting in lower potential for membrane fouling. The MBRs used for greywater treatment are generally operated at lower permeate fluxes in the range of 1–7 L/m^{2}h (Wu, 2019).

Solids Retention Time

The solids retention time (SRT) is the residence time of the sludge in the reactor, which affects the MLSS concentration in the reactor. Membrane bioreactors are operated at a higher SRT than conventional activated sludge plants (Rahman et al., 2023). As a consequence, there is a reduced amount of available substrate (from the greywater) relative to the microorganisms present in the reactor, resulting in a decreased generation of waste sludge (Judd, 2022). Higher biomass concentrations in MBRs, due to the longer SRTs, benefit the removal of organic matter, nitrogen, pharmaceuticals and personal care products (Maeng et al., 2013). Especially the slow-growing and sensitive microorganism (*e.g.*, nitrifiers) benefit from the longer SRTs in MBRs (Liu, 2008), as it increases their toleration towards toxic components. This is useful in greywater treatment, with regard to its high variation of pollutant concentrations (Najmi et al., 2020). Excessively high SRTs can lead to increased membrane fouling, due biomass accumulation on the membrane, whereas very low SRTs can also worsen fouling by raising EPS concentrations in the mixed liquor (Iorhemen et al., 2016). Very long SRTs can affect the aeration efficiency negatively when MLSS concentrations become too high (Van den Broeck et al., 2012).

Hydraulic Retention Time

The hydraulic retention time (HRT) refers to the residence time of the wastewater in the reactor. This corresponds to the available time the microorganisms have to biodegrade contaminants in the greywater, which should be long enough to ensure adequate pollutant removal. Greywater has is less contaminated than blackwater (or mixed domestic wastewater), the optimal HRT of MBRs for greywater treatment is therefore lower (Atasoy et al., 2007). An important advantage of MBRs compared to conventional activated sludge plants is the more straightforward independent control of SRT and HRT (Judd, 2008). In MBRs, the HRT is typically lower than in CAS plants. However, the higher biomass concentrations in MBRs still ensure adequate pollutant removal (Rahman et al., 2023). The lower HRTs in MBRs result in higher organic loading rates, reducing the required reactor volume and stimulating the growth of microorganisms (Isma et al., 2014). Large reductions of the HRT can increase membrane fouling in MBRs due to increased sludge viscosity and higher EPS concentrations (Iorhemen et al., 2016). The operating range of HRTs in MBRs treating domestic wastewater is between 2 and 24 h (Merz et al., 2007).

Aeration

Aeration has multiple functions in aerobic MBRs. It provides oxygen to the bacterial sludge, enabling nitrification and biodegradation of organics greywater, while also scouring the membrane surface, reducing deposition of sludge and suspended solids (Iorhemen et al., 2016). In moving bed MBRs, air flow ensures adequate mixing in the reactor, preventing sinking and accumulation of the biocarriers at the bottom of the reactor (Jabornig & Favero, 2013). Increasing aeration reduces membrane fouling by intensifying sheer stress across the membrane surface. Excessive air flows in the reactor may lead to breakdown of sludge flocs and increase membrane fouling (Tang et al., 2022). A thinner, but denser, fouling layer is created that has a higher extracellular polymeric substances (EPS) content, increasing the need for cleaning of the membrane (Ding et al., 2016). Intermittent aeration can prevent break up of sludge flocs and therefore mitigate membrane fouling (Shi et al., 2022). Improvement of nitrification can be obtained through the use of pure oxygen as aeration instead of air (Rodríguez et al., 2012). Sufficient air supply is crucial for the proper operation of aerobic MBRs, but it should be controlled to a necessary level, due to its significant contribution (of nearly 50%) to the total energy requirements (Gil et al., 2010). In activated sludge plants, dissolved oxygen is maintained at approximately 2 mg/L to obtain complete nitrification, while avoiding excessive aeration costs (Rieger et al., 2014).

Organic Loading

The organic loading of an MBR refers to the amount of organic matter in the incoming greywater (or any other influent). It is often quantified as the volumetric organic loading rate (OLR), expressed in kg COD per unit time and per unit volume. MBRs treating greywater are generally operated with an OLR in the range of 0.5–3 kg COD/m^{3}d (Johir et al., 2012; Sohail et al., 2020; Zhang et al., 2010; Zinatizadeh & Ghaytooli, 2015). To achieve optimal biological treatment, the organic loading of a reactor could be increased to allow the biomass to reach a higher concentrations (Rabaey, 2019). Higher OLRs carry the risk of reduced filterability of the MBR, increased membrane fouling and decreased removal efficiencies of organics and ammonia (Johir et al., 2012).

1.5.5 Moving Bed MBRs

A moving bed membrane bioreactor (MB-MBR) is a type of MBR that holds suspended biomass carriers in the bioreactor, designed to support the growth of microorganisms, initiating the formation of biofilms. These systems comprise by consequence two types of biomass: biofilms attached onto the carriers and microorganisms suspended within the mixed liquor (Ivanovic & Leiknes, 2012). Biocarriers are usually made of plastic media, although alternative materials, like polyurethane foam, activated carbon, sand, zeolite, wood chips, and ceramic carriers, are also utilized (Deng et al., 2016). There is an increasing fascination with sponge carriers or modified plastic carriers that incorporate sponge material, attributed to their affordability and remarkable biomass retention capabilities (Deng et al., 2016; Sohail et al., 2020). A critical parameter in the design of MB-MBRs is the volumetric filling fraction of these biocarriers, which significantly influences the overall surface area of the biofilm within the reactor. This fraction can typically extend to as much as 65% of the reactor's total volume. The MBRs and MB-MBRs generally exhibit comparable performance in terms of nitrification and the removal of organics (Ivanovic & Leiknes, 2012).

There are two major advantages to the addition of biocarriers in an MBR: a significantly improved total nitrogen removal and a substantial reduction in membrane fouling (Saidulu et al., 2021). The development of biofilm on the carriers induces a gradient of dissolved oxygen (DO) concentration, with lower levels in the deeper biofilm layers, due to constraints in DO diffusion from the liquid phase. Consequently, this leads to anoxic conditions within the inner biofilm, providing a suitable environment for denitrification by heterotrophic denitrifiers. This process enhances the conversion of nitrate into gaseous nitrogen, resulting in elevated removal of total nitrogen (Ivanovic & Leiknes, 2012; Leyva-Díaz et al., 2015). The heterotrophic biofilm can also accelerate the initial development of slow growing microorganisms (di Biase et al., 2019). Membrane fouling is reduced in MBBR-MBR through two mechanisms: mechanical scouring of the membrane by the carriers and through improved filterability of the mixed liquor. The mechanical effects have been identified as the major reason for reduction in fouling, consequently extending the duration of membrane filtration (Chen et al., 2016a; Sohail et al., 2023). Another advantage of MB-MBRs is that they have the potential to be a more compact technology (Leyva-Díaz et al., 2015), a crucial attribute for decentralized treatment in urban settings where spatial constraints are significant.

Chapter 2

OBJECTIVES AND HYPOTHESES

Greywater has been identified as a valuable fresh water resource to combat global water scarcity issues, especially in urban areas. Decentralized treatment of greywater can efficiently link the production of greywater with the demand for fresh water, thereby facilitating greywater reuse. Membrane bioreactors are a promising technology for decentralized greywater treatment, as they have the capability to treat greywater in a compact and efficient way, resulting in high-quality water as the end product. The MBR's compact nature is an important advantage in facilitating decentralized greywater treatment at the building level, as spatial constraints are significant in urban settings.

The goal for this thesis was to acquire a robust MBR system, with adequate COD and nitrogen removal from greywater, for non-potable reuse purposes. The operating conditions of two membrane bioreactors were adjusted (one MBR served as a control), to optimize COD and nitrogen removal. The treatment performance of the MBRs was assessed twice a week. This research was accompanied by nitrification batch tests, to elucidate the nitrification activity of the sludge present in the membrane bioreactors.

The specific objectives and hypotheses of this thesis were:

- Attaining an efficiently operating ceramic membrane MBR system that ensures sufficient removal of COD and nitrogen to meet the criteria for non-potable reuse guidelines. It is expected that a well-aerated MBR system will remove COD and nitrogen adequately.
- Elucidating the biodegradability of organics in synthetic greywater. Greywater is usually relatively biodegradable, depending on the amount of surfactants present.
- Measuring and assessing the influence of different HRTs and levels of aeration within the MBR to optimize nitrification. Higher HRTs have the potential to improve pollutant removal. Adequate aeration is essential to support the aerobic metabolism of nitrifying microorganisms.

- Comparing the total nitrogen removal between a conventional MBR and an MB-MBR containing PVDF carriers. The MB-MBR is expected to have a better nitrate, and subsequently TN, removal.

- Optimizing the organic loading rate in the MBR and MB-MBR. A higher organic loading rate (OLR) can stimulate microbial growth in the reactor, leading to higher MLSS concentrations, which improves the treatment efficiency of the MBR system.

- Investigating the effects of modified operational parameters (HRT, aeration flow, and OLR) and the introduction of biocarriers on the rate of membrane fouling in the studied MBR systems. An increased HRT has the potential to reduce sludge viscosity and concentrations of EPS, thereby decreasing the membrane fouling rate. Enhanced aeration flow rates can amplify the scouring effects on the accumulated fouling layer, also leading to a subsequent reduction in membrane fouling potential. Elevated OLR could potentially reduce sludge filterability, subsequently increasing membrane fouling.

Chapter 3

MATERIALS AND METHODS

3.1 Synthetic Greywater

The recipe for synthetic greywater, comprising a selection of personal care products (PCPs) and chemicals, was adopted from Ongena et al. (2023). A feedstock solution of a 120× concentrated synthetic greywater was prepared weekly and stored in a refrigerated room at 4–7 °C. The composition of the concentrated feedstock solution is listed in Table 3.1. The brands of the PCPs and chemicals are listed in Table A.1 of the appendix.

Table 3.1: Ingredients and their concentrations, used for the preparation of the 120× concentrated synthetic greywater. See Table A.1 for the brands and product types.

PCPs	**Conc.** (mg/L)	**Chemicals**	**Conc.** (mg/L)
Dishwashing detergent	10.44	Ammonium chloride	9.96
Liquid laundry	24.96	Calcium chloride	6.00
Shampoo	6.36	Potassium phosphate dibasic	0.36
Shaving gel	6.36	Sodium acetate	46.15
Shower gel	17.16	Sodium bicarbonate	40.32
Toothpaste	4.32	Sodium chloride	6.00
		Sodium sulfate	6.12

3.2 Experimental Set-up

Two identical membrane bioreactors, labeled MBR-1 and MBR-2, were operated in parallel (Figure 3.1) from September 2022 to February 2023. Each reactor was made of plexiglass with a working volume of 10 L. They contained two identical flat sheet ceramic (SiC) membrane panels each. One membrane had a surface area of 0.165 m^2, resulting in 0.330 m^2 membrane per reactor. The pore size of the membranes was 0.56 µm. The membranes were physically cleaned by removing accumulated sludge with a soft brush when the transmembrane pressure (TMP) exceeded 10–13 kPa. Air pumps (P-400, Paladin, China) supplied a compressed air flow through two aeration pipes at the bottom of each MBR, directly below the membrane modules. Both reactors were continuously fed from the same influent tank of 120 L. This tank was manually refilled (approximately twice a week) by adding 1 L of the concentrated greywater solution and subsequently topping it up with tap water. Prior to each refill, the influent tank was emptied and cleansed using a mop. The membrane permeate flow (*i.e.*, the effluent) was directly discarded into the drain. The influent and permeate flows of both MBRs were carried out by time-controlled peristaltic pumps (LEPP350S, LST, Korea). Following every 72-minute pumping cycle, a 4-minute relaxation period was implemented. A constant working volume of the reactors was ensured by redirecting excess mixed liquor to an overflow tank. This measure was put in place to effectively address fluctuations in the reactor's volume and operational problems related to the pumps. The overflow liquid from the MBRs was collected in the same overflow tank, which was emptied regularly, and stored in a refrigerated room for further use (*e.g.*, in nitrification activity batch tests). The supernatant of the stored overflow liquid was discarded on a regular basis to concentrate the sludge, which was used later on in the nitrification activity batch tests.

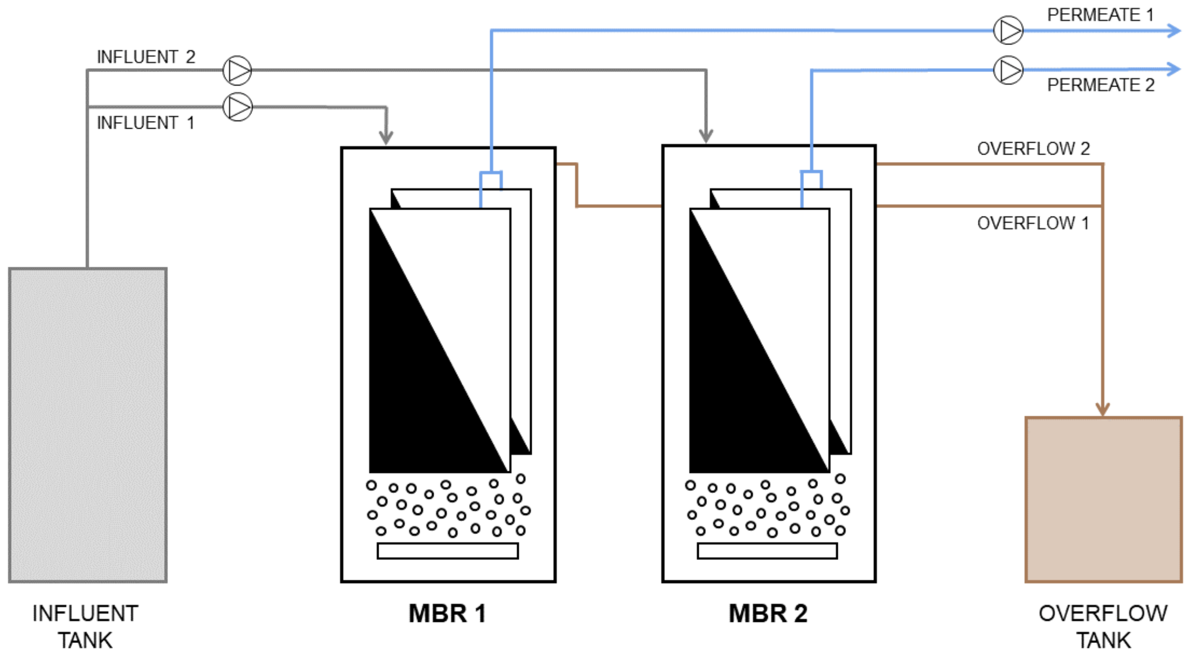

Figure 3.1: Setup of the influent tank, the aerated membrane bioreactors (MBR-1 and MBR-2), the overflow tank and the time-controlled peristaltic pumps. The effluent was directly disposed in the drain.

The influent, mixed liquor, and effluent quality of MBRs were monitored during the experimental period of 126 days (30 September 2022–3 February 2023). The membrane of MBR-1 cracked unexpectedly on day 119 (January 27, 2023) after which its monitoring was discontinued. The influent, reactor mixed liquor, and effluent were sampled twice a week (every Tuesday and Friday). The concentrations of the following analytes were measured in these samples: ammonia-N, nitrite-N, nitrate-N, TN, and total COD. The reactor mixed liquor samples were filtered using a 0.46 µm MCE syringe filter (Biofil, China) before measuring these concentrations. Hence, these concentrations solely represent the suspended TN and COD. An additional sample was taken on the same days from the reactor mixed liquor to determine the MLSS and MLVSS concentrations of the reactors. The TMP of both MBRs was recorded at the time of sampling. The temperature and pH of the reactors were continuously monitored. From day 40 onward, the DO concentration in each reactor was measured at the time of sampling. The synthetic greywater influent was sampled on days 98 and 113 for the determination of its BOD_5.

3.3 Optimization of Operating Conditions

Both MBRs were operated at a water recovery rate of 97% during the entire experimental period (*i.e.*, 3% of the influent was redirected to the overflow). Between days 1–102, MBR-1 was operated at an HRT of 15.1 h, an SRT of 21.0 days, and an OLR of 0.636 kg COD/m^{3}d. The OLR was calculated based on an anticipated average influent COD concentration of 400 mg/L. MBR-2 was operated at these same conditions between days 1–6 and 47–102. Between days 6–47, the HRT and SRT of MBR-2 were augmented to 20.0 h and 27.8 days, respectively. This resulted in an OLR of 0.480 kg COD/m^{3}d. From day 102 onward, the HRT and SRT of both MBRs were set at 8.6 h and 11.9 days, respectively to attain an anticipated OLR of 1.130 kg COD/m^{3}d. This OLR was calculated based on a slightly higher average influent COD concentration of 405 mg/L, based on the COD concentration data of the greywater up until day 102.

The aeration of both MBRs was initially set at an air supply flow rate of 0.6 L/min. The air flow was provided by one air pump per MBR. Between days 36–92, the air supply flow rate of both reactors was increased to 1.0 L/min. An exception was MBR-2 between days 85–92, where the aeration was decreased to 0.5 L/min, due to problems with the overflow grid (elaborated further in Section 5.3). From day 92 onward, an additional air pump was installed for each MBR. All pumps were operated at their maximal air flow rate from day 92 onward. This exceeded the capacity of the air flow rate meter, rendering the precise air flow rate indeterminable during this period.

Both MBRs were operated as conventional MBRs, *i.e.*, without biomass carriers, between days 1–71. From day 71 onward, MBR-2 was operated as an MB-MBR with a volumetric filling ratio of 10% of polyvinylidene difluoride (PVDF) biomass carriers. The carriers were cut up into small cylinders of 3×1 mm. They had a density of 0.24 g/mL, a surface area of 2.91 m^2/g, and a surface resistance of 600 Ω/cm^2.

Four modifications to the operational conditions were anticipated to significantly impact the treatment performance and align with the objectives of this thesis. These adjustments are summarized in Table 3.2, and their labels will serve as points of reference in the graphs depicting the treatment performance of the MBRs across various compounds in Section 4.2.

Table 3.2: Summary of modifications in the operational conditions of MBRs 1 and 2.

Label	Modification	Day
T1	Increase of HRT from 15.1 h to 20.0 h*	26
A1	Increase of aeration flow from 0.6 L/min to 1 L/min	36
T2	Decrease of HRT back to 15.1 h*	47
B	Addition of biomass carriers in 10% volume ratio*	71
A2	Increase of aeration flow from 1 L/min to the maximal capacity	92
OLR	Increase of OLR from 0.636 kg COD/m^{3}d to 1.130 kg COD/m^{3}d	102

**Modifications applied to MBR-2 only.*

3.4 Nitrification Activity Batch Tests

Sludge from both MBRs was used in nitrification activity batch tests, following the protocol by van Loosdrecht et al. (2016). Such batch test comprised of a beaker, filled with synthetic greywater and activated sludge. This beaker is continuously stirred and aerated for 4 hours. During this time, ammonia concentrations are monitored to calculate the nitrification activity. This batch test consists of a beaker containing synthetic greywater and activated sludge, which is continuously stirred and aerated for a duration of 4 hours (Figure 3.2). Throughout this period, ammonia concentrations are measured to determine the rate of nitrification activity. The tests were carried out at two different moments (days 49 and 132) in the experimental period to obtain a snapshot of the nitrification capacity of the MBR mixed liquors. Synthetic greywater was used as an ammonium and nitrite source during the batch tests. The synthetic greywater for the batch tests consisted of the ingredients listed in Table 3.1, but with the exemption of sodium acetate. Sodium acetate was omitted to minimize the aerobic heterotrophic consumption of COD. The batch tests were performed in beakers of 500 mL, filled with 5 mL of 100× concentrated synthetic greywater and 400 mL of overflow mixed liquor. The beakers were partly covered with parafilm to keep the foaming surfactants inside (Figure 3.2). A magnetic stirring bar was used to mix the batch test liquid. Dissolved oxygen was supplied by an aeration stone submerged in the liquid. The sludge used in the tests was collected from the stored MBR overflow, which was collected in the weeks leading up to the test. The obtained sludge was aerated overnight and washed three times with tap water before the start of the tests. Right before the start of the tests, the MLSS and MLVSS concentrations of every batch test beakers were measured.

Two nitrification activity batch tests (series A) were carried out on day 49. These will be referred to as batch tests A1 and A2. The MBR overflow sludge used in these batch tests was collected between days 35–48. Both tests were performed simultaneously and under the same conditions. The duration of each test was 4 hours. Sampling of mixed liquor from the beakers was done at the start of the test and at 0.5, 1, 2, 3, and 4 hours after the start of the test. Measurement of the ammonia-N, nitrite-N, nitrate-N, TN, and COD concentrations was done for all the mixed liquor samples.

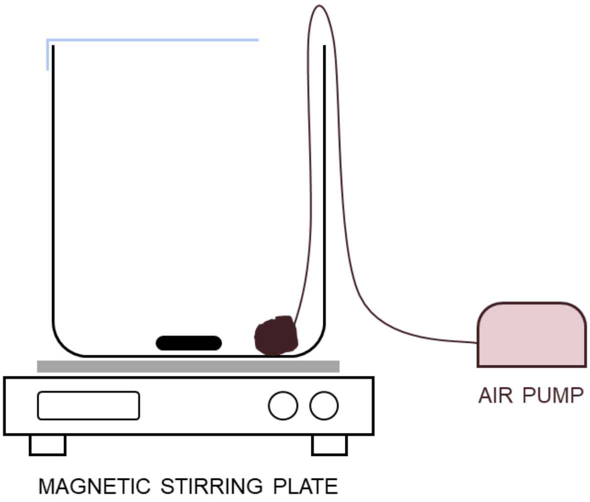

Figure 3.2: Setup of one batch test beaker.

Two additional nitrification batch tests (series B) were carried out on day 132. These batch tests were labeled as B1 and B2. The MBR overflow sludge used in these batch tests was collected between days 118–132. Batch test B2 included biomass carriers that were extracted from MBR-2. The tests were performed simultaneously, and the duration of each test was 4 hours. Mixed liquor was sampled from the test beakers at the start of the test and 15 min, 30 min, 45 min, 1 h, 1.5 h, 2 h, 2.5 h, 3 h, 3.5 h, and 4 h after the start of the test. The concentrations of ammonia-N, nitrate-N, and COD were determined for all these samples.

3.5 Water Quality Analysis Methods

The temperature and pH of the mixed liquor of both MBRs were continuously monitored with a PS-2153 temperature probe and a PS-2573 pH electrode, respectively, which were both connected to a PS-2022-220 Sparklink Air data transmitter (PASCO Scientific, USA). The temperature during the batch tests was measured with a bulb thermometer. The TMP was measured with a DPGW-09 pressure gauge (Dwyer Instruments Inc., USA). DO concentrations of the mixed liquor were determined using a DO-5519 portable DO probe (LUTRON Instruments, Taiwan).

The concentrations of COD, BOD_5, nitrogen forms, suspended solids (SS), and volatile suspended solids (VSS) were determined according to the APHA Standard Methods (Baird et al., 2017). Digestions were done using the HANA HI839800-02 digestor, and absorbance measurements were performed with a OPTIZEN POP UV-vis spectrophotometer (KLab, Republic of Korea). The COD concentrations were measured according to the *5520 D. Closed Reflux, Colorimetric Method.* For COD measurement in the MBR influent samples, the high-range digestion solution was used, and absorbance was measured at 600 nm. For COD measurement of the mixed liquor, MBR effluent, and batch test samples, the low-range digestion solution was used and absorbance was measured at 420 nm. The BOD_5 measurement was performed following the *5210 B. 5-Day*

BOD Test protocol. Six BOD_5 bottles of freshly prepared synthetic greywater were incubated at 20 °C on January 21, 2023, with the addition of 2 mL overflow mixed liquor. Measurement of ammonia-N concentrations was done according to the *4500-NH_3 F. Phenate Method*, after diluting the samples 20 times. Absorbance was measured at 640 nm. Nitrite-N concentrations were determined according to the *4500-NO_2^- B. Colorimetric Method*, with absorbance measurement at 543 nm. Nitrate-N concentrations were determined according to the *4500-NO_3^- C. Second-Derivative Ultraviolet Spectrophotometric Method*, using absorbance measurements between 200 and 250 nm. TN concentrations were determined according to the *4500-N C. Persulfate Method*, after diluting the samples 2 times. The absorbance was measured between 200 and 250 nm. The SS concentrations were determined according to the *2450 D. Total Suspended Solids Dried at 103–105°C* method. The VSS were determined according to the *2540 E. Fixed and Volatile Solids Ignited at 550°C* method.

3.6 Data Processing

Excel was used to perform linear regressions and calculate their coefficient of determination (R^2). The linear regression of the ammonia-N data in the batch tests was used to determine their ammonium oxidation rate (r_{NH4}). The specific ammonium oxidation rate (q_{NH4}) of mixed liquor was calculated with Equitation 3.1 (van Loosdrecht et al., 2016).

$$q_{NH4} = \frac{r_{NH4}}{[VSS]} \tag{3.1}$$

The organic loading rate was calculated with Equation 3.2 ($[COD]_{in}$ = influent concentration of COD).

$$OLR = \frac{HRT}{[COD]_{in}} \tag{3.2}$$

3.7 Text Processing

Throughout this thesis, the artificial intelligence tool ChatGPT was occasionally used to paraphrase sentences or passages. This function of the ChatGPT tool was utilized with the "rewrite" command on `https://chat.openai.com/`. The generated output served solely as a source of inspiration for alternative sentence structures or synonyms.

Chapter 4

RESULTS

4.1 Characterization of Synthetic Greywater

The BOD_5 tests (5 replicates) of the synthetic greywater resulted in an average BOD_5 concentration of 278±27 mg/L. The COD concentration of the greywater used in the BOD_5 test was 403 mg/L. This resulted in a BOD/COD ratio of 0.69±0.07. Mean COD and nitrogen concentrations of the synthetic greywater, used as influent for the membrane bioreactor are presented in Table 4.1.

Parameter		**Concentration**
COD	mg/L	402 ± 57
TN	mg N/L	23 ± 5
Ammonia	mg N/L	15 ± 3
Nitrite	mg N/L	0.08 ± 0.08
Nitrate	mg N/L	0.64 ± 0.56

Table 4.1: Average COD, TN, ammonia-N, nitrite-N and nitrate-N concentrations of the synthetic greywater over the operating period of the MBR reactors of 126 days (34 samples of the MBR-1 influent and 36 samples of the MBR-2 influent).

4.2 Greywater Treatment Performance of MBR and MB-MBR

4.2.1 Temperature, pH and Dissolved Oxygen

Due to an unexpected power outage, temperature and pH levels could not be monitored between days 35-74. The measured temperature patterns in both reactors were very similar, with a maximal observed temperature difference of 1.2°C between MBR-1 and MBR-2 (Figure 4.1). In September-November, the average temperature of both reactors was 24°C, which was higher than the average temperature of December-February, which was 20°C for both reactors. The pH levels for MBR-1 and MBR-2 were on average 6.5±0.7 and 6.3±0.5, respectively (Figure 4.2). In MBR-1, the DO concentration varied between 5.1–6.3 mg/L in the period of days 40–54 (Figure 4.3). For days 61–85, the DO concentration fluctuated within a range of 3.6–5.4 mg/L. From day 89 onward, the DO concentration was higher, with an average of 7.8±0.5 mg/L. The DO concentration of MBR-2 fluctuated a lot (within the range of 3.4–6.9 mg/L) between days 40–71. Between days 75–85, the DO concentration was lower, on average 1.4±0.3 mg/L. It then increased back to 5.4 on days 92–95. For days 102–123, the average DO concentration was higher, 7.3±0.4 mg/L.

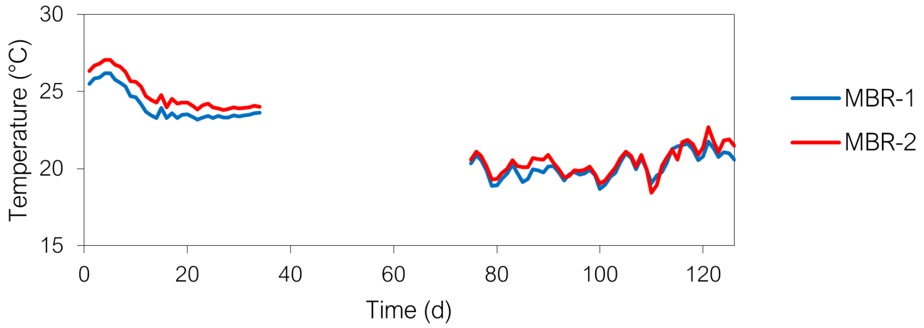

Figure 4.1: Temperature of the mixed liquor of MBR-1 and MBR-2 for days 1–34 and days 75–126.

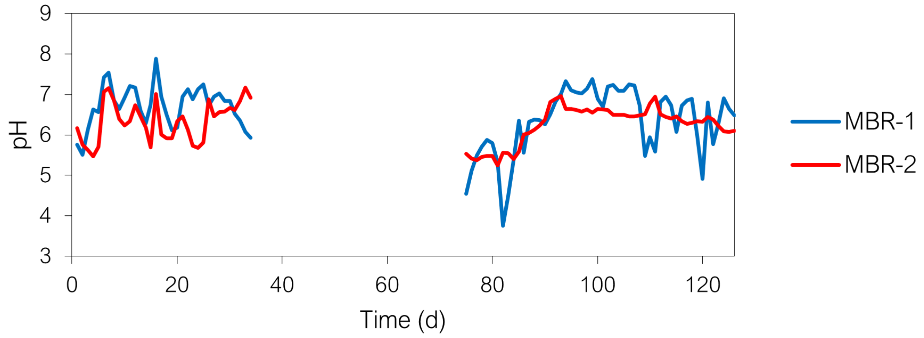

Figure 4.2: pH of the mixed liquor of MBR-1 and MBR-2 for days 1–34 and days 75–126.

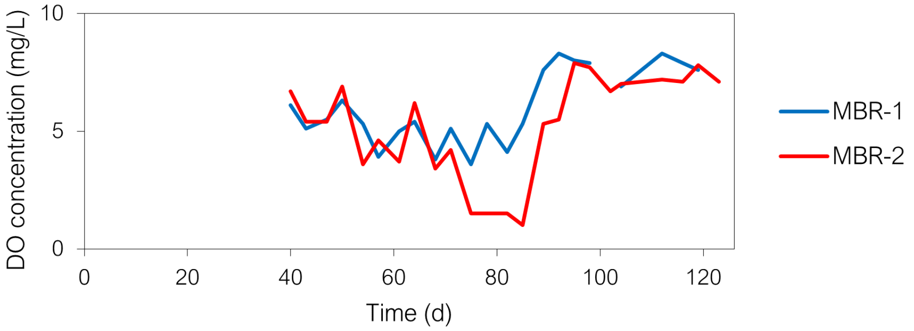

Figure 4.3: Dissolved oxygen (DO) concentrations of the mixed liquor of MBR-1 and MBR-2 for days 40–123.

4.2.2 Biomass Growth Analysis

The mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentrations of MBR-1 started at 480 mg/L on day 1 (Figure 4.4). For days 1–102, MLVSS/MLSS fluctuated between 83% and 100%. For days 5–15, the MLSS increased up to 1700 mg/L. Between days 19–22, the MLSS was lower, on average 910±92 mg/L. For days 36–54, the MLSS and MLVSS concentrations steadily increased, up to 2060 and 1920 mg/L, respectively. After day 54, they decreased to a minimum of 1305 and 1090 mg/L (on day 61). From day 64, the MLSS and MLVSS concentrations steadily increased again, up to 4050 and 3360 mg/L, respectively, on day 102. On days 104 and 112, the MLSS concentration was much higher than before, at 12660 and 10990 mg/L, respectively. The MLVSS for these days remained similar to the preceding days, resulting in MLVSS/MLSS ratios of 28% and 17% for days 104 and 112, respectively. On day 116, the MLSS and MLVSS concentrations were much lower again, at 1950 and 1910 mg/L, respectively.

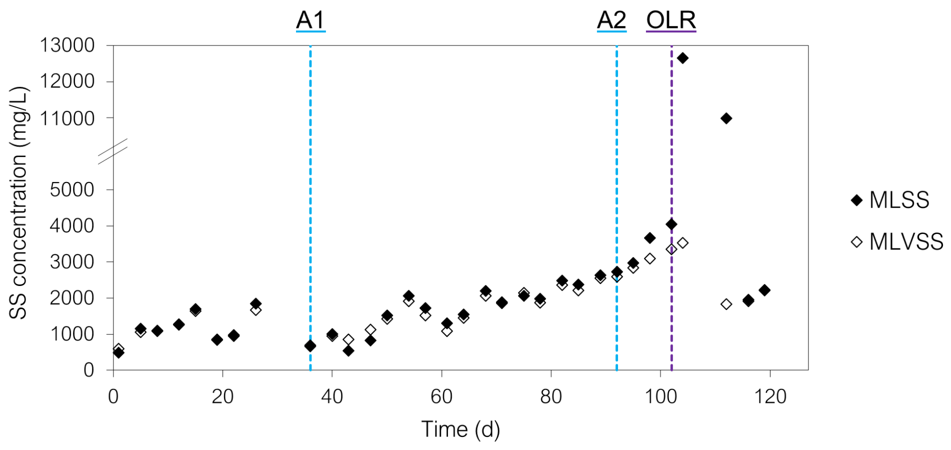

Figure 4.4: MBR-1 mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentrations for days 1–119. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

The MLSS and MLVSS concentrations in MBR-2 started off lower than in MBR-1, at 245 and 195 mg/L, respectively (Figure 4.5). For days 1–102, MLVSS/MLSS of MBR-2 fluctuated between 79% and 100%. For days 1–75, the MLSS concentration showed an increasing trend up to 3400 mg/L. Between days 75–85, the MLSS concentration remained between 3030 and 3405 mg/L. Between days 92–95, lower MLSS concentrations were measured (1945 mg/L). On days 104 and 112, the MLSS concentration was much higher, at 12580 and 11400 mg/L, respectively. The MLVSS concentration was 3350 and 2350, respectively, for these days. From day 116 onward, the MLSS concentration was lower again and fluctuated between 1940 and 2360 mg/L.

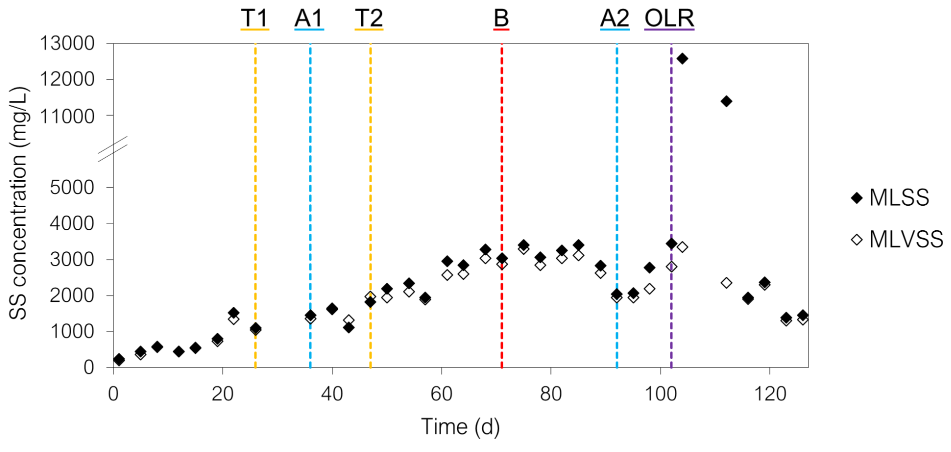

Figure 4.5: MBR-2 mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentrations for days 1–126. (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate)

4.2.3 Membrane Fouling Analysis

The transmembrane pressure (TMP) of the MBR membranes built up over time, and then they were cleaned. The course of the TMP for MBR-1 and MBR-2 is plotted in Figures 4.6 and 4.7, respectively. The membranes of MBR-1 were cleaned on days 12, 29, 85, 112, and 118. After cleaning, the TMP of MBR-1 increased already after the course of 3 to 10 days. The membranes of MBR-2 were cleaned on days 12, 47, and 123. After day 71, the cleaning frequency of MBR-2 was lower than for MBR-1. The TMP of MBR-2 also remained longer at 0 kPa after cleaning.

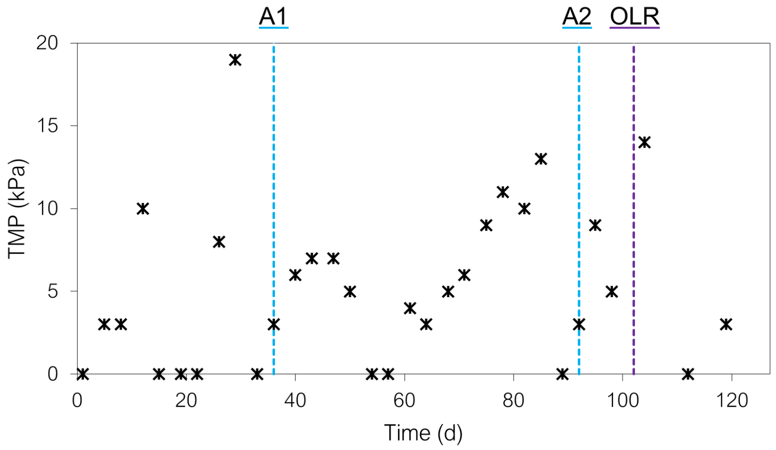

Figure 4.6: MBR-1 transmembrane pressure (TMP) for days 1–119. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

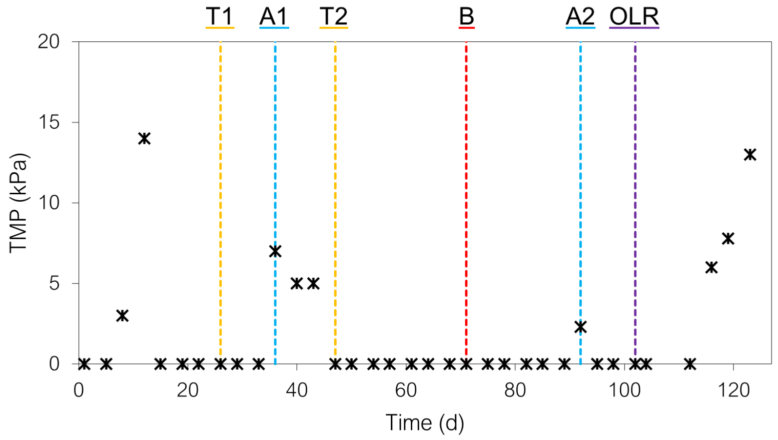

Figure 4.7: MBR-2 transmembrane pressure (TMP) for days 1–126 (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate).

4.2.4 COD Removal

The influent COD concentration of MBR-1 fluctuated the most between days 1–33 (Figure 4.8). The average concentration in this period was 381±66 mg/L. From day 36 onward, COD levels in the influent of MBR-1 were more stable. Between days 36–78, the average COD concentration was 431 mg/L, with a standard deviation of 29 mg/L. For days 82–116, the COD level in the influent was on average lower, with a mean COD concentration of 387±27 mg/L. A maximum in influent COD of 541 mg/L was measured on day 119. The effluent COD concentrations of MBR-1 had lower variability than the influent COD.

The average effluent COD concentration for the entire operational period of MBR-1 was 39±14 mg/L, and the average COD removal efficiency was (90±5)%. The effluent COD concentrations varied within a range of 14 to 70 mg/L. Elevated COD in the effluent occurred between days 12–29 and days 36–43, with an average concentration of 57±7 mg/L. The COD removal efficiency was on average (85±4)% and reached a minimum of 78% on day 22. The highest COD effluent concentration was measured on day 19, at 70 mg/L. Between days 47–61, COD removal improved in MBR-1. The average COD concentration and removal efficiency were 33±3 mg/L and (92±1)%, respectively. Between days 64–75, the effluent COD concentration was slightly higher again, with an average of 45±4 mg/L. The mean removal efficiency was (89±1)% during this period. From day 78, the effluent COD concentration and removal efficiency averaged 28±9 mg/L and (96±3)%, respectively. On day 119, a high concentration of 48 mg/L was measured for COD in the effluent. The average OLR was 0.65±0.08 kg COD/m^{3}d between days 1–98. For days 102–119, the average OLR was 1.14±0.21 kg COD/m^{3}d.

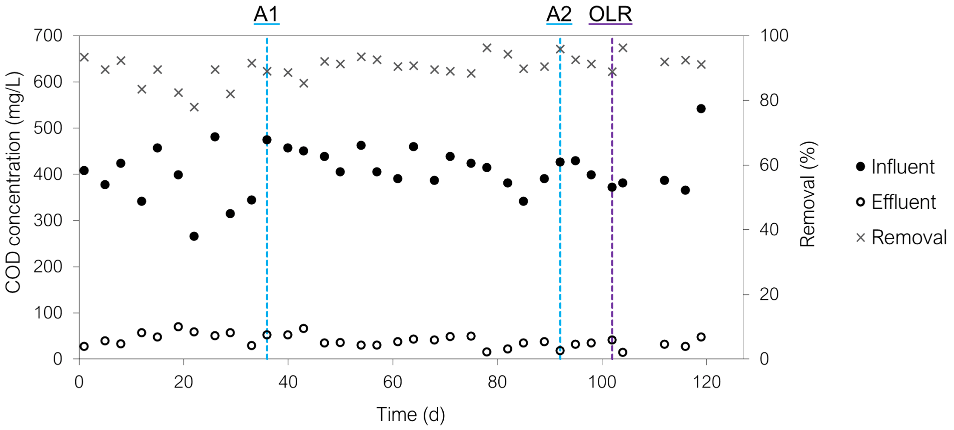

Figure 4.8: MBR-1 influent and effluent COD concentrations for days 1–119. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

For MBR-2, similarly to MBR-1, the variation in influent COD concentrations (61 mg/L) was higher than the variation in effluent COD concentrations (17 mg/L) (Figure 4.9). For the entire operational period of MBR-2, the average influent COD concentration was 398±61 mg/L. The lowest and highest COD influent concentrations were measured on days 104 (296 mg/L) and 36 (484 mg/L), respectively.

The effluent COD concentrations ranged between 18 and 108 mg/L. The average effluent COD concentration and removal efficiency were 43±17 mg/L and (89±5)%, respectively. Between days 12–43, elevated effluent concentrations were measured from MBR-2. During this period, the mean effluent COD concentration and removal efficiency were 61±19 mg/L and (84±6)%, respectively. Two higher peaks in effluent COD levels of 108 and 78 mg/L were measured on days 29 and 43, respectively. For days 47–61, the COD removal improved and was on average (93±2)%. The effluent COD concentration was 31±6 mg/L on average for this period. Between days 64–75, the average COD effluent concentration and removal efficiency were 47±6 mg/L and (89±1)%, respectively. Between days 78–82, COD effluent concentrations improved and were 20±2 on average. On day 85, a sudden increase to 42 mg/L was observed. Between days 89–104, the COD effluent concentration and removal efficiency averaged 30±4 mg/L and (92±1)%, respectively. From day 112 onward, COD levels in the effluent of MBR-2 became higher again. The average effluent concentration and removal efficiency were 45±8 mg/L and 89±2, respectively. The average OLR was 0.60±0.08 kg COD/m^{3}d between days 1–98. For days 102–119, the average OLR was 1.11±0.17 kg COD/m^{3}d.

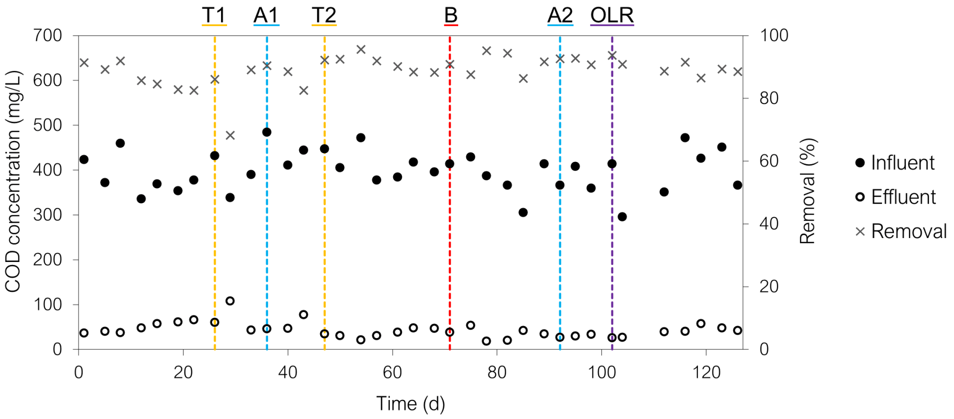

Figure 4.9: MBR-2 influent and effluent COD concentrations and removal efficiency for days 1–126. (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate)

4.2.5 Nitrogen Removal

Ammonia

The influent ammonia-N concentration of MBR-1 was on average 18.4±2.4 mg/L and fluctuated between 13.3 and 22.7 mg/L (Figure 4.10). The effluent concentration of ammonia-N from MBR-1 was initially similar to the influent concentration, and the average ammonia removal efficiency was 12%. Between days 8 and 26, the effluent concentration of ammonia-N varied from 7.30 to 10.9 mg/L. This was remarkably lower than the corresponding influent concentrations and resulted in improved ammonia-N removal efficiencies between 43 and 68%. Between days 29 and 43, effluent concentrations of ammonia-N returned to the range of the influent concentrations. Removal efficiencies decreased to 9% on day 36. The effluent ammonia-N concentration decreased to 9.3 mg/L again on day 47. From day 50 onward, two weeks after the increase in aeration of the reactor, ammonia-N concentrations remarkably decreased. The average MBR-1 effluent ammonia-N concentration from day 54 onward was 0.7±0.3 mg/L. During this period, the concentration stayed below 1.2 mg/L. From day 54 on, the average ammonia-N removal efficiency of MBR-1 was (96±2)%.

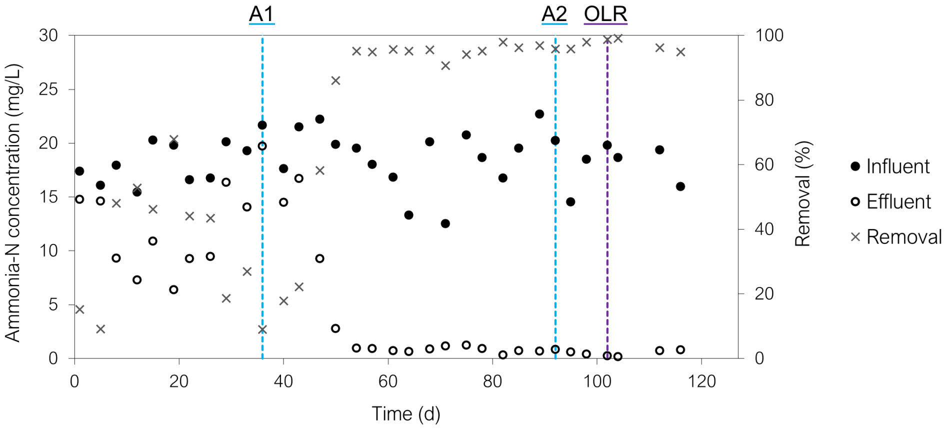

Figure 4.10: MBR-1 influent and effluent ammonia-N concentrations for days 1–116. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

The average ammonia-N concentration in the MBR-2 influent was 19.2±2.3 mg/L for the total duration of the experimental period. It generally fluctuated between 16.9 and 23.2 mg/L (Figure 4.11). The only exceptions to this are days 64 and 95, where the ammonia-N concentration was 14.2 and 11.5 mg/L, respectively. For the effluent concentration of ammonia-N from MBR-2, two clearly distinct regimes were observed. From day 1 to day 40, the average concentration was 16.5±1.9 mg/L. Effluent ammonia-N concentrations fluctuated within the range of 13.5–21.0 mg/L during this period. The ammonia-N removal efficiency of MBR-2 varied from 3 to 34% in this period. On day 43, one week after the increase in aeration of the MBR tank, the effluent ammonia-N concentration decreased to 1.4 mg/L. From day 43 to day 126, the average effluent concentration was 0.9±0.6 mg/L, and the average ammonia-N removal efficiency was (95±3)%. Between days 71–82 the average effluent ammonia-N concentration was higher, *i.e.*, 1.6±0.9 mg/L.

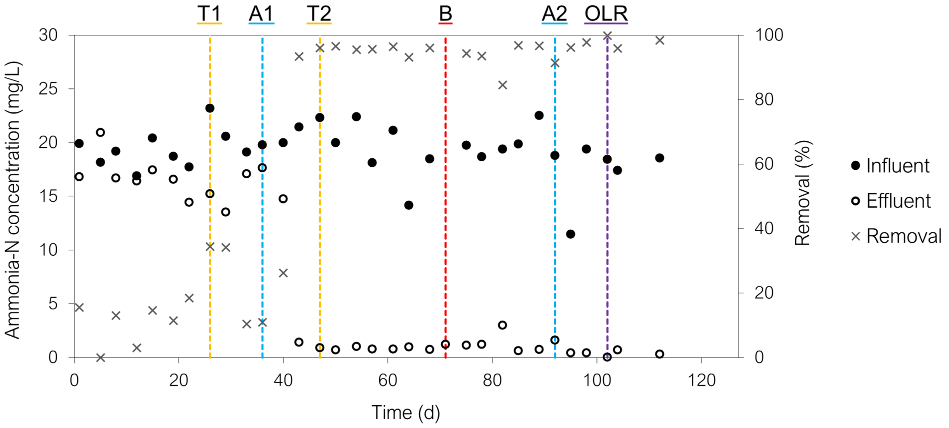

Figure 4.11: MBR-2 influent and effluent ammonia-N concentrations for days 1–126. (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate)

Nitrite

In MBR-1, nitrite was present in both the influent and effluent present at low concentrations. The nitrite-N concentration in the influent of MBR-1 fluctuated slightly but stayed between 0.01 and 0.15 mg/L (Figure 4.12). The effluent nitrite-N concentrations were higher than the influent concentrations during the first three weeks of the experimental period. The effluent concentrations between days 5–15 were remarkably higher, with an average nitrite-N concentration of 1.2±0.1 mg/L. From days 22–43, the effluent nitrite-N concentration fell within a similar range as the influent corresponding influent concentration. Between days 47–54, the effluent concentration averaged 1.2 mg/L again. From day 61 onward, the effluent nitrite-N concentrations of MBR-1 were near zero.

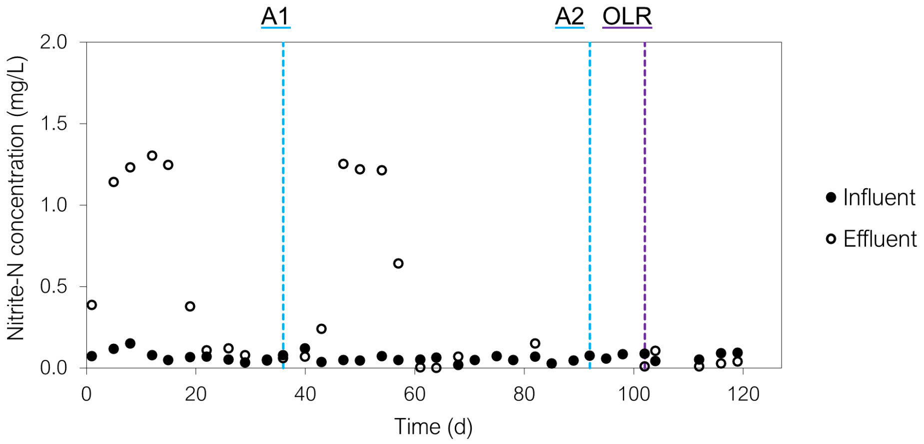

Figure 4.12: MBR-1 influent and effluent nitrite-N concentrations for days 1–119. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

The influent nitrite-N concentration of MBR-2 fluctuated within the same range as for MBR-1, between 0.01 and 0.15 mg/L (Figure 4.13). Only one higher influent nitrite-N concentration was measured, 0.80 mg/L on day 102. The effluent nitrite-N concentrations from MBR-2 were in the same order of magnitude as the influent concentrations from day 1 to day 33. From day 36 to day 47, the effluent concentration was between 0.49 and 1.25 mg/L, which was higher than the influent concentration. From day 50 onward, the effluent nitrite-N concentrations were similar to the influent concentrations, with the exception of day 61 (1.22 mg/L). Between days 75 and 85, the effluent nitrite-N concentrations of MBR-2 were slightly higher, *i.e.*, 0.23±0.15 mg/L.

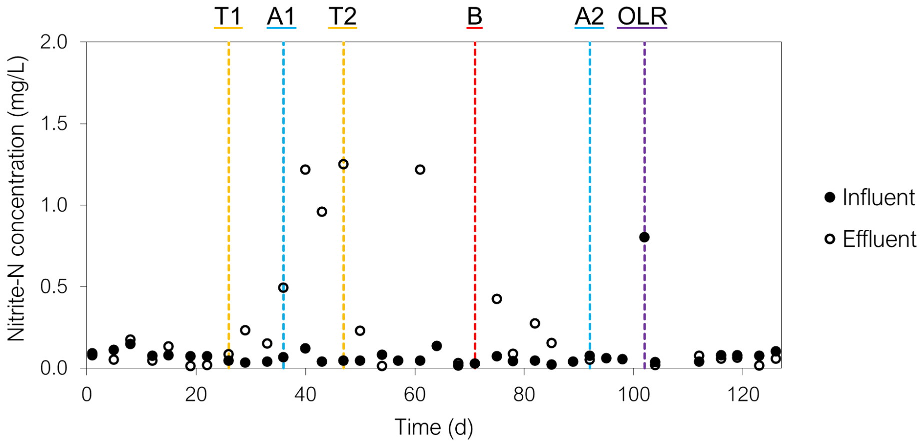

Figure 4.13: MBR-2 influent and effluent nitrite-N concentrations for days 1–126. (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate)

Nitrate

The nitrate-N concentrations in the MBR-1 influent generally remained below 1 mg/L (Figure 4.14). On days 26 and 43–54, the influent nitrate-N concentration was slightly higher, but remained between 1 and 2 mg/L. In the first week of the experimental period, the nitrate-N effluent concentration of MBR-1 was in the same order of magnitude as the influent. On day 8, a higher nitrate-N concentration of 5.0 mg/L was measured in the effluent. Between days 8–3, the effluent nitrate-N concentration decreased linearly ($R^2 = 0.93$) to 0.7 mg/L. This concentration remained constant for days 8–40. The effluent nitrate-N concentration increased linearly ($R^2 = 0.91$) from 1.6 mg/L on day 43, which corresponds to a week after the increase in aeration, up to 9.8 mg/L on day 57. Between days 57–89, this concentration fluctuated around an average value of 9.1±0.6 mg/L. The effluent nitrate-N concentration further increased from day 92 onward and averaged 12.2±0.7 mg/L between days 92–112. From day 116 onward, the effluent nitrate-N concentration decreased, averaging 7.6±0.6 mg/L.

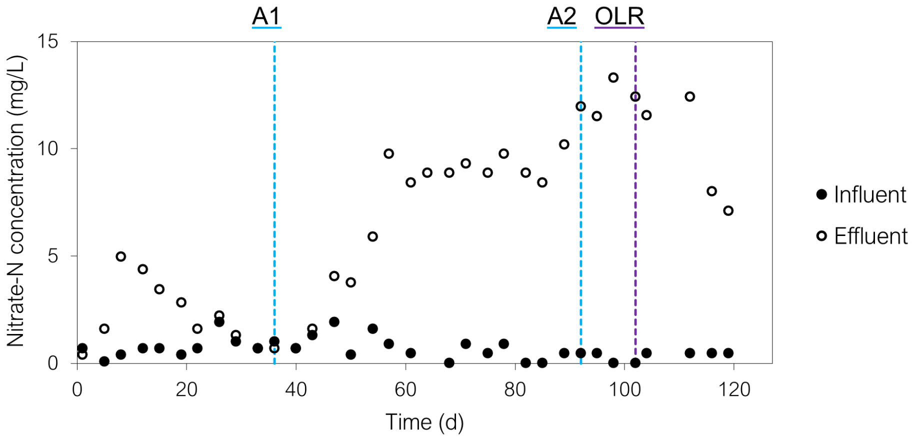

Figure 4.14: MBR-1 influent and effluent nitrate-N concentrations for days 1–119. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

For MBR-2, the influent nitrate-N concentration was generally below 1 mg/L as well (Figure 4.15). Exceptionally (on days 26, 40–54, and 102), the influent nitrate-N concentration was between 1 and 2.3 mg/L. The nitrate-N effluent concentration stayed below 0.4 mg/L between days 1–36. From day 40, which corresponds with 4 days after the increase in aeration, the nitrate-N concentration started to increase in the MBR-2 effluent. The nitrate-N concentration stayed within a range of 8.3 to 11.8 mg/L between days 47–71, with the exception of day 61 (4.5 mg/L). From day 75, the effluent nitrate-N concentration started to gradually decrease. For days 82 and 85, the concentration was 0.9 and 1.4 mg/L, respectively. A much higher concentration of nitrate-N was recorded in the MBR-2 effluent from day 89 onward. From day 89 onward, the nitrate-N effluent concentration stayed above 7 mg/L (except for day 123 with 3.1 mg/L). Between days 89–95, the mean concentration was 9.3±0.4 mg/L. On day 98, the effluent nitrate-N concentration reached a maximum of 13.3 mg/L. Hereafter, the nitrate-N effluent concentration decreased and stabilized between 7.1 and 8.5 mg/L from day 112 onward (except for day 123 with 3.1 mg/L).

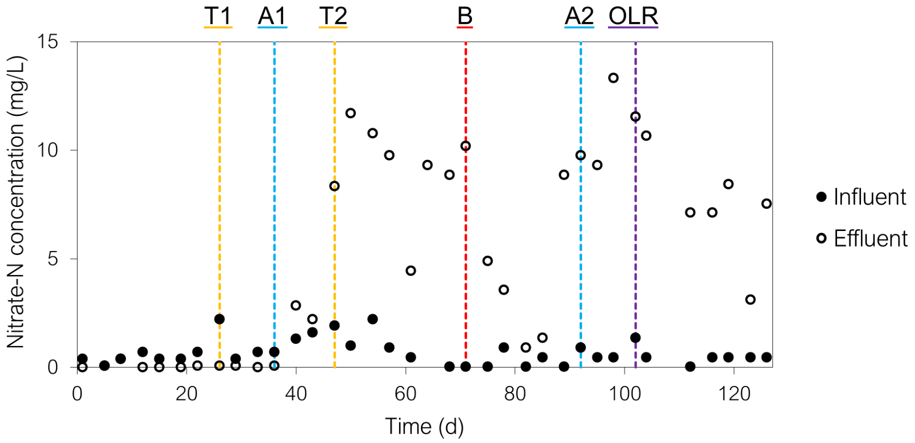

Figure 4.15: MBR-2 influent and effluent nitrate-N concentrations for days 1–126. (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate)

Total Nitrogen

The total nitrogen (TN) concentrations of the influent of MBR-1 varied between 14.3–38.2 mg/L (Figure 4.16). Two periods of different influent TN levels can be distinguished. For days 1–54, the average influent TN concentration was 17.7±2.7 mg/L. Until day 33, influent TN concentrations (= $[TN]_{inf}$) were similar to the sum of the influent ammonia-N, nitrite-N and nitrate-N concentrations (= $[NH_4^+ + NO_2^- + NO_3^-]_{inf}$). Between days 36–54, $[TN]_{inf}$ was on average 6.3±0.4 mg/L lower than $[NH_4^+ + NO_2^- + NO_3^-]_{inf}$, with the exception of day 40, where $[TN]_{inf}$ was 1.9 mg/L higher than the sum. From day 57 onward, the $[TN]_{inf}$ was always higher than $[NH_4^+ + NO_2^- + NO_3^-]_{inf}$. The effluent TN concentrations (= $[TN]_{eff}$) of MBR-1 fluctuated between 8.0–20.9 mg/L, with an average of 13.0±4.0 mg/L. Between days 50–61, the average $[TN]_{eff}$ was lower, 9.5±1.1 mg/L. Between days 78–98, the average $[TN]_{eff}$ was higher, 17.6±2.2 mg/L. From day 102 onward, $[TN]_{eff}$ highly fluctuated between 12.3–19.1 mg/L. $[TN]_{eff}$ was similar to the sum of the effluent ammonia-N, nitrite-N and nitrate-N concentrations during the entire experimental period. The removal efficiency of TN fluctuated between 14–34% for days 1–54, with the exception of day 12 (9%). The TN removal efficiency of MBR-1 was higher on days 57 and 61, with 63 and 72%, respectively. On days 64–68, the removal efficiency decreased to 45%. On day 71, a higher removal efficiency of 63% was found. Between days 75–85, the removal efficiency was on average (46±6)%. Between days 89–95, the removal efficiency was lower, on average (22±6)%. Between days 98–104, the removal efficiency increased and was on average (48±7)%.

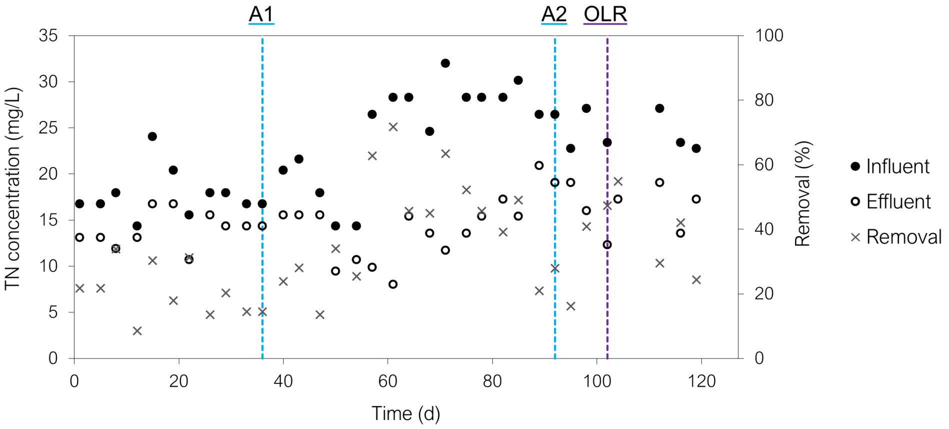

Figure 4.16: MBR-1 influent and effluent total nitrogen (TN) concentrations for days 1–119. (A1 = increase of aeration; A2 = further increase of aeration; OLR = increase of organic loading rate)

The influent of MBR-2 had an average TN concentration of 22.9±4.7 mg/L. As with MBR-1, two different periods of influent TN level were observed (Figure 4.17). For days 1–54, the average influent TN concentration was 18.4±2.0 mg/L. For days 57–126, the average influent TN concentration was 26.4±2.7 mg/L. Between days 2–40, $[TN]_{inf}$ was in the same order of magnitude as $[NH_4^+ + NO_2^- + NO_3^-]_{inf}$. Between days 43–54, $[TN]_{inf}$ was on average 6.2±2.2 mg/L lower than $[NH_4^+ + NO_2^- + NO_3^-]_{inf}$. From day 57 onward, $[TN]_{inf}$ was always higher than $[NH_4^+ + NO_2^- + NO_3^-]_{inf}$, with an average difference of 9.3±5.3 mg/L. The effluent TN concentration fluctuated between days 1–71 within 11.9 and 19.2 mg/L. Between days 75–82, lower TN concentrations were measured in the effluent (between 2.5–6.2 mg/L). On day 85, $[TN]_{eff}$ increased again to 17.2 mg/L. Between days 85–116, $[TN]_{eff}$ decreased steadily down to 9.8 mg/L. From day 119 onward, $[TN]_{eff}$ was 15.4 mg/L, except for day 123 (2.5 mg/L). Between days 1–12, $[TN]_{eff}$ was on average 4.2±0.3 mg/L lower than $[NH_4^+ + NO_2^- + NO_3^-]_{eff}$. From day 15 onward, $[TN]_{eff}$ was similar to $[NH_4^+ + NO_2^- + NO_3^-]_{eff}$, except for days 75–78, where $[TN]_{eff}$ was on average 2.3±0.2 mg/L lower than $[NH_4^+ + NO_2^- + NO_3^-]_{eff}$. The removal efficiency of TN in MBR-2 fluctuated between 16–34% during the period of days 1–26, with the exception of day 5 (with a removal efficiency of 0%). For days 29–54, the TN removal efficiency was lower and stayed between 6–14%, with the exception of days 43 and 50 (with a removal efficiency of 0%). Between days 57–68, the removal efficiency increased again (23–52%). Between days 75–82, higher TN removal efficiencies were observed, between 77 and 91%. On day 85, the removal efficiency decreased back to 43%. From day 98 onward, the TN removal efficiency increased, up to 64%. On days 119 and 126, lower TN removal efficiencies were observed, 26 and 32%, respectively. On day 123, an abrupt increase in the TN removal efficiency (90%) was found.

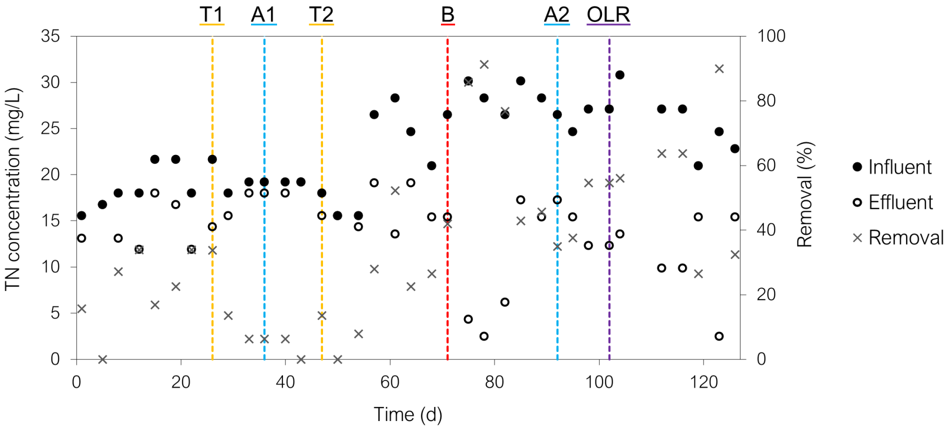

Figure 4.17: MBR-2 influent and effluent total nitrogen (TN) concentrations for days 1–126. (T1 = increase of HRT; T2 = decrease of HRT; A1 = increase of aeration; A2 = further increase of aeration; B = addition biocarriers; OLR = increase of organic loading rate)

4.3 Nitrification Activity Batch Tests

4.3.1 Series A

MLSS and MLVSS

The MLSS and MLVSS concentrations in test beaker A1 were 950 and 940 mg/L, respectively. This results in a MLVSS/MLSS ratio of 0.99. The MLSS and MLVSS contents in test beaker A2 were 2210 and 2130 mg/L, respectively, resulting in a MLVSS/MLSS ratio of 0.96.

DO and T

Dissolved oxygen was monitored during the tests by performing a measurement at the same time as sampling. The levels of dissolved oxygen stayed in the range of 6.8–7.9 mg/L in both tests. No trend in DO levels was observed during the nitrification activity tests. Temperature was measured simultaneously with DO at the time of sampling. The magnetic stirring plates had a heating option as well. During the first hour of the test, this heating function was turned on accidentally in test A1, which caused the temperature to be around 26 °C. When the heating was turned off, the beaker started to cool down to room temperature again. At t = 2 h and t = 4 h, the temperature decreased down to 24.1 °C and 22.1 °C, respectively. In test beaker A2, the temperature showed only limited variation and stayed between 22.1 and 23.7 °C.

Nitrogen and COD

The development of the concentrations of ammonia-N, nitrite-N, nitrate-N, TN, and COD during batch tests A1 and A2 is shown in Figure 4.18 and Figure 4.19, respectively. Ammonia-N concentrations steadily decreased in both test beakers. The ammonia-N concentration at the beginning of the test was about 23 mg/L. After 4 hours, ammonia-N levels were reduced by more than half of the original concentration in both test beakers. The ammonia-N concentration after 4 hours in test beakers A1 and A2 was 11.3 and 9.1 mg/L, respectively. The ammonia removal rates of tests A1 and A2 were 2.9 mg N/(L·h) ($R^2 = 0.97$) and 3.2 mg N/(L·h) ($R^2 = 0.97$), respectively. The specific ammonia oxidation rates of tests A1 and A2 were 3.1 mg N/(g VSS·h) and 1.5 mg N/(g VSS·h), respectively.

The nitrite-N concentration increased slightly in the beginning, in both batch test, but stayed constant after. In test A1, the initial nitrite-N concentration was 1.0 mg/L. In the following samples, from t = 30 min to t = 4 h, the nitrite-N concentration stayed between 1.2 and 1.3 mg/L. For test A2, the nitrite-N concentrations at the start of the test and at t = 30 min were 0.5 and 0.9 mg/L, respectively. In the following samples, from t = 1 h to t = 4 h, the nitrite-N concentration stayed between 1.2 and 1.3 mg/L.

Nitrate-N levels showed a steady increase in both tests, almost by a factor of 2. During test A1, the nitrate-N concentration increased from 2.5 to 5.0 mg/L. In test A2, the nitrate-N concentration more than doubled during the course of the test, from 1.9 mg/L initially to 4.1 mg/L at t = 4 h.

The total nitrogen concentration at the beginning of the test was 20 mg/L for both test beakers. After one hour, a remarkably low concentration of 8 mg/L was observed in beaker A1. This seems to be an outlier since the previous and following total nitrogen concentrations stayed around 17–20 mg/L. For test beaker A2, the total nitrogen concentration fluctuated during the test, but did not show any apparent increase or decrease over the testing period. In the first 4 samples of both batch tests, the sum of the inorganic nitrogen forms (ammonia, nitrite, and nitrate) was higher than the corresponding TN concentration.

The COD concentrations did not show an increasing or decreasing trend during both tests. The COD concentration in test beaker A1 fluctuated during the test, but remained within a 20% deviation from the average concentration of 97 mg/L. In test beaker A2, the COD concentration showed fewer fluctuations and was 151 mg/L on average.

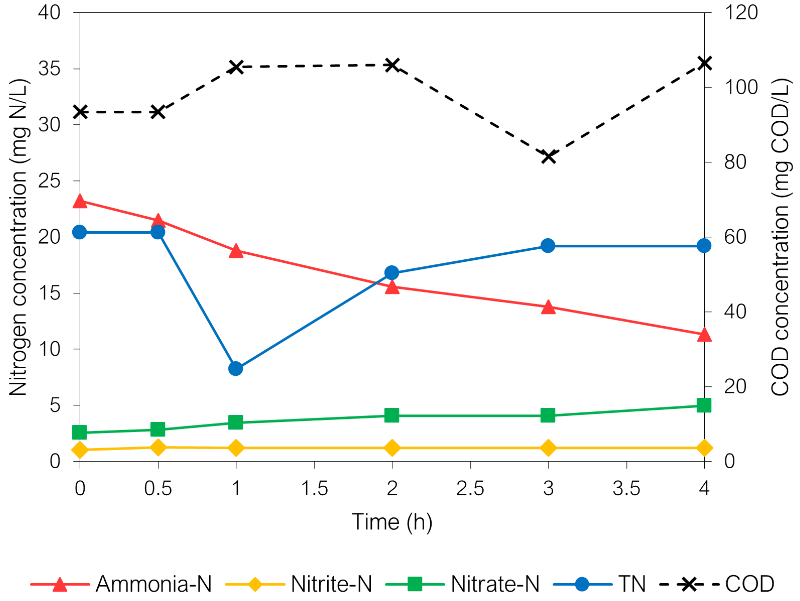

Figure 4.18: Concentrations of ammonia-N, nitrite-N, nitrate-N, TN and COD during the course of nitrification activity batch test A1.

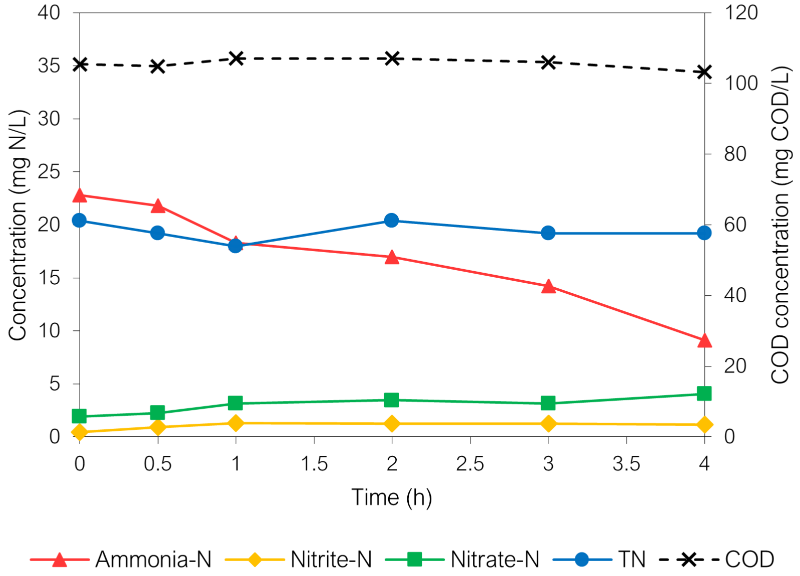

Figure 4.19: Concentrations of ammonia-N, nitrite-N, nitrate-N, TN and COD during the course of nitrification activity batch test A2.

4.3.2 Series B

MLSS and MLVSS

During the weeks leading up to the tests, the mixed liquor overflow of the MBRs was collected, so it could be used to test the nitrification activity of the sludge. The MBRs had been operated under a higher aeration regime for three months. Before the start of the tests, the concentration of MLSS and MLVSS in the batch test mixed liquor was measured. For test beaker B1, the MLSS and MLVSS concentrations were 2410 and 2280 mg/L, respectively. The MLVSS/MLSS ratio was thus 0.95. For test beaker B2, the MLSS and MLVSS concentrations were slightly higher, 2470 and 2400 mg/L, respectively. This results in a MLVSS/MLSS ratio of 0.97.

DO and T

Dissolved oxygen concentrations of the mixed liquor during the nitrification activity test were monitored at the time of sampling. In test beaker B1, the DO concentration was 7.2 mg/L on average. The DO concentration slightly increased over the course of the batch test, from 6.7 to 7.7 mg/L, but stayed within 10% of the average value. In test beaker B2, no trend was observed in DO levels. The average DO concentration in test beaker B2 was 6.3 mg/L. DO levels varied during the test between 6.1 and 7.1 mg/L and were overall slightly lower than in test beaker B1. The temperature of the mixed liquor was measured simultaneously with the DO concentration in both test beakers. The average temperature in test beakers B1 and B2 was 20.7 and 20.5 °C, respectively. The temperature showed no trend and did not fluctuate substantially in both test beakers.

Nitrogen and COD

The obtained concentrations of ammonia-N and nitrate-N of batch tests B1 and B2 are plotted in Figures 4.20 and 4.21, respectively. The measured concentrations of ammonia-N showed considerable fluctuations in test beakers B1 and B2. In test beaker B1, the initial ammonia-N concentration was 15.3 mg/L, which is lower than expected after the addition of the ammonia standard (see Section 3.4). In the first 30 minutes of test B1, the ammonia-N concentrations decreased to 4.5 mg/L. The following sample, at 45 minutes, was 13.7 mg/L. From t = 45 minutes until t = 2 h, the ammonia-N concentration decreased again, reaching 0.5 mg/L at t = 2 h. From t = 2 h, an increase in ammonia-N was observed in the next samples of the following hour. At t = 3 h, ammonia-N reached a concentration of 7.6 mg/L. The concentration of ammonia-N was very low for the last samples of test B1, with 0.1 mg/L and 0.5 mg/L at t = 3.5 h and t = 4 h, respectively. The initial ammonia-N concentration in test beaker B2 was 23.5 mg/L. A linear decrease (R^2 = 0.98) of the ammonia-N concentration was observed for the first hour of the test, with a concentration of 5.0 mg/L at t = 1. The ammonia-N concentrations fluctuated between every following sample, with a maximum of 17.4 mg/L at t = 1.5 h and a minimum in the last sample of 0.0 mg/L at t = 4 h. The ammonia removal rates of tests B1 and B2 were 2.7 mg N/(L·h) (R^2 = 0.48) and

4.0 mg N/(L·h) ($R^2 = 0.53$), respectively. The specific ammonia oxidation rates of tests B1 and B2 were 1.2 mg N/(g VSS·h) and 1.7 mg N/(g VSS·h), respectively.

The nitrate-N concentrations steadily increased during the course of both batch tests B1 and B2. In test B1, the initial nitrate-N concentration was 1.8 mg/L. During the whole test, the nitrate-N concentration increased linearly ($R^2 = 0.97$). The final nitrate-N concentration, at t = 4 h, was 4.5 mg/L. In test B2, the concentration of nitrate-N was 1.4 mg/L at the start of the test. Hereafter, the nitrate-N concentration increased linearly ($R^2 = 0.94$) for the entire duration of the batch test. At t = 4 h, the nitrate-N concentration was 7.5 mg/L in test B2, which is 3.1 mg/L higher than for test B1.

The COD concentration in test beaker B1 was, on average, 105 mg/L. During the entire batch test, the COD concentration did not show a clear trend and stayed within the 10% range of the average concentration. In test B2, the initial COD concentration was 105 mg/L. The COD concentration in the following samples was 70 mg/L on average, lower than the initial concentration. A minimum of 55 mg/L was observed at t = 2 h. The COD concentration in the last sample of test B2, at t = 4 h, gave an invalid value (-755 mg/L), and is therefore not plotted on Figure 4.21.

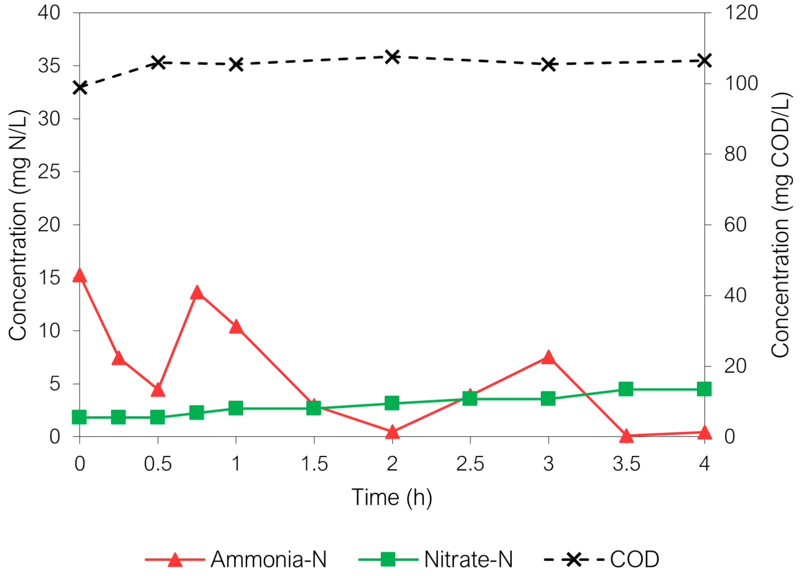

Figure 4.20: Concentrations of ammonia-N, nitrite-N, nitrate-N, TN and COD during the course of nitrification activity batch test B1.

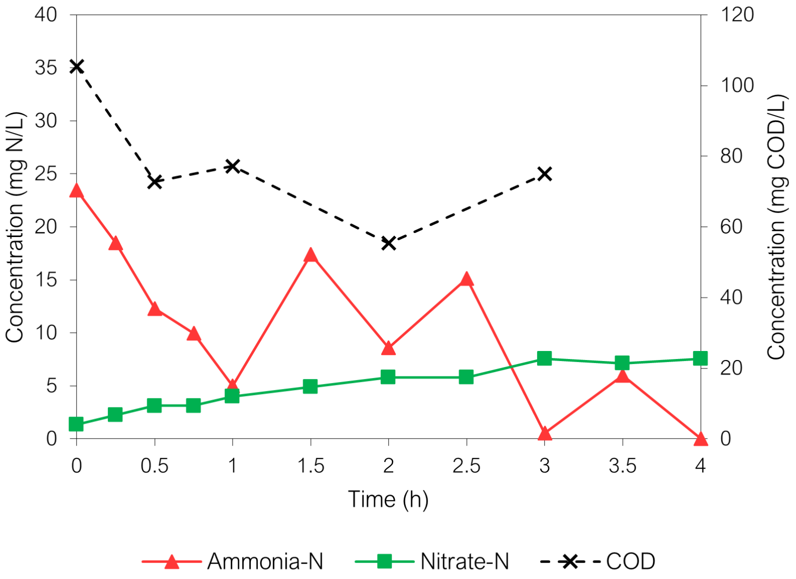

Figure 4.21: Concentrations of ammonia-N, nitrite-N, nitrate-N, TN and COD during the course of nitrification activity batch test B2.

Chapter 5

DISCUSSION

5.1 Nitrogen Balance in Samples

For some MBR influent and effluent samples, the sum of concentrations of inorganic nitrogen species (NH_4^+, NO_2^-, and NO_3^-) exceeded the concentration of total nitrogen, as reported in Section 4.2.5. This observation contradicts theoretical expectations, since TN encompasses both organic and inorganic nitrogen forms (Figure 1.2). Consequently, TN can either be equal to the inorganic nitrogen forms (in the absence of organic nitrogen) or higher than the inorganic nitrogen forms. The fact that the TN concentration was occasionally lower than the inorganic nitrogen forms indicates potential inaccuracies in the determination of TN, ammonia, nitrite, or nitrate concentrations. Firstly, small volume errors could have occurred during sample transfer and dilution. Although care was taken to exercise caution in the laboratory to minimize such human errors, they can still occur inadvertently. A second potential source of errors stems from the indirect measurement method for total nitrogen. During this method, all the organic and inorganic nitrogen forms are oxidized and converted to nitrate, which is then quantified as the indicative TN concentration (Baird et al., 2017). If this conversion does not proceed completely, the TN concentration could be underestimated. During the spectrophotometric measurement of ammonia and nitrite, they have to form a colored complex first. Similar to the TN measurement, if this chemical reaction does not proceed completely, it could lead to an underestimation of the concentrations. Lastly, inherent uncertainties associated with spectrophotometry further contribute to potential errors. These include variations in absorbance repeatability among samples and the possibility of spectrophotometer drift (Sooväli et al., 2006). The combined effect of underestimation of TN and overestimation of ammonia, nitrite, and nitrate could have led to TN concentrations lower than the sum of the concentrations of the inorganic nitrogen forms.

5.2 Membrane Fouling

The transmembrane pressure can be used as an indicator for the degree of membrane fouling (Iorhemen et al., 2016). During MBR operation, the TMP measurement data was used to organize the physical cleaning of the membrane. At the beginning of the experimental period, the membrane fouling rate of both MBRs was similar, and their membranes were cleaned after approximately 2 weeks. After T1 (*i.e.*, the increase of HRT in MBR-2 on day 26), the TMP of MBR-2 remained zero, while the TMP of MBR-1 rose up to 19 kPa. The increased HRT is a potential cause for the lower fouling rate of MBR-2 after T1. Sludge viscosity has the tendency to decrease when the HRT is increased, resulting in a lower membrane fouling rate (Iorhemen et al., 2016). On the day when the TMP of MBR-1 started to increase, its MLSS concentration was almost double that of MBR-2. As lower MLSS concentrations generally lead to a lower fouling potential (Iorhemen et al., 2016), this might be another driver for the lower fouling rate of MBR-2.

Following the increase in aeration on day 36 (A1), the TMP of both MBR-1 and MBR-2 gradually decreased over time, without any cleaning being implemented. In the literature, higher air flow rates are generally associated with lower membrane fouling, because the sheer force of the rising air bubbles intensifies (Yusuf et al., 2016). By increasing the aeration in both MBRs, the accumulated cake layer could have been gradually removed by the scouring action of the air bubbles, causing a spontaneous decrease in their TMPs. In MBR-1, the TMP started to rise again after the spontaneous decrease, but at a slower rate than before A1. The need for physical cleaning of MBR-1 was reduced from every 2 weeks to every 20 to 25 days after A1. Contrary to MBR-1, the TMP of MBR-2 stayed zero after the spontaneous decrease. Even after T2 (*i.e.*, reduction of HRT on day 47), the TMP of MBR-2 maintained zero. After A1, the aeration of both MBR-1 and MBR-2 was increased to the same extent, so a similar fouling rate was expected. From the MLSS concentration data, a higher fouling rate would be expected in MBR-2, as its MLSS concentrations were lower than those of MBR-1 in this period. A possible explanation for the absence of fouling in MBR-2 is that its sludge maintained its low viscous properties even after the decrease in HRT. After the second increase in aeration A2, on day 92, the TMP in both MBRs decreased. The higher sheer force of rising air bubbles probably in some way removed the accumulated cake layer in both MBRs, resulting in a lower TMP.

It is generally accepted in the literature that the fouling rate of MB-MBRs is lower than for MBRs, mainly due to the scouring effects of the biocarriers (Chen et al., 2016a). After the addition of biocarriers to MBR-2, the TMP stayed zero for 20 days. However, the TMP of MBR-2 had been zero for the past three weeks as well. The hypothesis of the MB-MBR having a lower fouling rate can therefore not be confirmed with great certainty from this experimental data.

After the increase in organic loading rate (OLR), the TMP of MBR-2 increased at a higher rate than before. This is in line with the reported phenomena in the literature (Johir et al., 2012). In MBR-1, only 2 data points for TMP are available after cleaning after the increase in OLR, not enough to observe an increase in fouling rate for this reactor.

5.3 Treatment Performance of MBR and MB-MBR

COD Removal in MBR and MB-MBR

The synthetic greywater had a COD/BOD$_5$ ratio of 1.45, which is relatively low compared to the average for real greywater (Shaikh & Ahammed, 2020). The COD/BOD$_5$ ratio is below 2.5, meaning that the organics in the synthetic greywater used in this thesis are relatively easily biodegradable (Li et al., 2009). The COD removal of both MBRs was generally similar to the results found in the literature during the entire experimental period. They had an overall removal efficiency of 90±4 and 89±5% for MBR-1 and MBR-2, respectively. This falls within the expected range from the literature for MBR systems treating greywater (Oteng-Peprah et al., 2018a), suggesting a well-developed community of heterotrophic microorganisms in both MBRs. The average effluent COD concentrations were 39±14 and 41±17 mg/L for MBR-1 and MBR-2, respectively. This corresponds to values reported in previous studies on greywater treatment by submerged MBRs (Bani-Melhem et al., 2015; Winward et al., 2008). However, some papers that had comparable influent COD concentrations reported lower mean effluent COD concentrations (*i.e.*, 24–26 mg/L) than observed in this thesis (Huelgas & Funamizu, 2010; Lesjean & Gnirss, 2006). The suspected cause is probably the higher MLSS concentrations of those MBRs, *i.e.*, up to 10–25 g/L. As a probable solution for further reduction of COD levels in the effluent, the reactor could operate at a higher MLSS content. One study operated an MBR with comparable influent COD and MLSS concentrations, but with much lower COD levels in the effluent (9 mg/L on average). This was probably possible due to the relatively high HRT of 36 hours (Liberman et al., 2016). Higher HRTs can indeed improve COD removal, but may deteriorate ammonia removal at excessively high HRT levels (Najmi et al., 2020), as will be discussed later.

Effluent COD concentrations decreased in both MBRs 11 days after the increase in aeration from 0.6 L/min to 1 L/min. This decline can likely be attributed to the increased oxygen availability in the mixed liquor, which stimulated the aerobic metabolism of heterotrophic COD-consuming microorganisms. Literature on COD removal by MB-MBR systems generally does not report significant improvements with respect to standard MBRs (Ivanovic & Leiknes, 2012). After the addition of biocarriers to the standard MBR-2, a decrease in COD was observed in the effluent. However, this cannot certainly be attributed to the addition of biocarriers, since MBR-1 (without biocarriers) showed the same decreasing trend during that period. The observed lower COD concentrations in both MBR-1 and MBR-2, approximately at 20 mg/L, only applied to days 78 and 81. Considering that these samples were collectively analyzed, possible analytical errors could have contributed to the underestimation of COD concentrations across all these samples. Overall, this change still falls within the range of variation in effluent COD concentrations throughout the entire experimental period.

Increasing the MLSS content of both MBRs was attempted by increasing the OLR, which was accompanied by an increase in the food-to-microorganism ratio (F/M) on day 102. This was supposed to allow the microorganisms to grow, eventually resulting in increased MLSS concentrations.

Activated sludge systems need some time to adjust to their new conditions and develop a higher MLSS. The operation of MBR-1 was discontinued 17 days after the increase in OLR, due to a damaged membrane. MBR-2 was still operated for 7 days more, but had experienced a loss of biomass, due to operational problems two weeks prior to the increase in OLR. Moreover, during the 3 weeks after the increase in OLR, the mixed liquor of MBR-1 and MBR-2 was perturbed at several moments (on days 95, 112, and 123). This probably made it difficult for the biomass to further develop. The mentioned technical issues with both MBRs and the limited monitoring time after the increase in OLR limit accurate interpretation of the data.

Nitrogen Removal in the MBRs

Ammonia removal improved over the course of the first two weeks of the experimental period, from 9 to 68%. The simultaneous increase of nitrite and nitrate in the MBR-1 effluent indicated that nitrification took place as an ammonia removal mechanism, since nitrite and nitrate are reaction products of the nitrification process. During that time, MLSS and MLVSS concentrations were 2–3 times higher in MBR-1 compared to MBR-2. The MLVSS content of activated sludge closely relates to the amount of active biomass present, such as, among others, nitrifying microorganisms. This could explain why there was no improved ammonia removal in MBR-2 during that time. The nitrifying microbial community of MBR-1 did, however, not develop sufficiently enough to completely remove the incoming nitrogen load, as effluent ammonia-N levels remained above 7.30 mg/L. After day 29, ammonia concentrations in the effluent increased back to the range of the influent, deteriorating the ammonia removal. Unfortunately, the MLSS data between days 29 and 33 was lost. On the subsequent sampling day (day 36), the MLSS concentration was noted to be more than half of its value before day 29. This observation raises the suspicion that the MLSS concentration has begun to decrease from day 29 onward, possibly explaining the decline in ammonia removal. The abrupt decrease in biomass concentrations within MBR-1 may have stemmed from the clogging of the effluent tube of MBR-1. With organic matter and a certain degree of nitrogen present in the effluent, microbial growth initiated on the inner surfaces of the effluent tubes. As the tubes gradually became clogged, the overflow rate increased, leading to a loss of biomass from the reactor's mixed liquor.

In activated sludge, a minimal DO concentration of 2 mg/L is aimed at for good nitrification (Rieger et al., 2014). It is important that the whole reactor is sufficiently aerated. If the aeration system is not capable of maintaining sufficient DO concentrations throughout the entire bioreactor, zones of low oxygen levels could arise, which might lead to increased ammonia concentrations in the effluent (Rieger et al., 2014). To ensure homogeneous and adequate DO concentrations, the aeration of both MBR-1 and MBR-2 was increased on day 36, from 0.6 L/min to 1 L/min. Two weeks after the increase in aeration in MBR-1, ammonia effluent levels dropped to around 1 mg N/L. In MBR-2, which had MLVSS concentrations twice as high as MBR-1, this decline was already realized within one week. The obvious difference in ammonia removal after the increase in aeration suggests that DO levels in the bioreactor were indeed insufficient in certain zones of the MBR reactors. The increased ammonia removal resulted from the higher activity of the ammonia-oxidizing bacteria.

Simultaneously, nitrite increased in the effluent of both reactors, to a range of 1–1.5 mg N/L. This observation confirms that the increased activity of AOB is responsible for the improved ammonia removal. The nitrite buildup is much lower than the decrease in ammonia concentrations in the effluent, due to nitrite oxidation by the NOB, the second step in the nitrification process. Two weeks after the nitrite breakthrough in the reactor effluents, nitrite levels returned to concentrations near zero, alluding to a well-developed community of NOB. The development of the NOB became possible after receiving nitrite, supplied by the increased activity of the AOB. The NOB probably needed some time to fully develop, as nitrite oxidation is energetically less favorable than ammonia oxidation. The interpretation of increased nitrification, *i.e.*, AOB and NOB activity, is supported by the simultaneous gradual breakthrough of nitrate in both reactor effluents. Nitrate levels in both reactor effluents increased up to around 10 mg N/L. This suggests nitrification as the main mechanism of ammonia removal in both MBRs. There was also a part of the removed ammonia that did not accumulate as nitrate in the effluent. This can be attributed to denitrification in the inner part of sludge flocs and the assimilation of nitrogen in new biomass.

Nitrogen Removal in the MB-MBR

Biomass carriers were introduced into MBR-2 on day 71, transforming it into a MB-MBR. Microorganisms started to form biofilms on the carriers. Four days after the addition of biocarriers, the effluent nitrate concentration of MBR-2 decreased by nearly 50%. In less than two weeks, the nitrate effluent concentration of the MB-MBR stabilized around 1 mg N/L. During this period, ammonia removal stayed constant, implying that denitrification improved in the reactor, subsequently reducing nitrate in the effluent. The simultaneous improved TN removal efficiency further confirms this hypothesis. In MBR-1, nitrate in the effluent remained around 10 mg N/L after day 71. The biocarriers in MBR-2 are, thus, most probably responsible for the increased denitrification during that period of time. This observation complies with the existing literature on MB-MBRs, where it has been established that anoxic zones in the inner part of well-developed biofilms create a niche for the denitrification process and therefore improve nitrate removal in MBRs. After the introduction of biocarriers, ammonia and nitrite concentrations increased in the MBR-2 effluent for a period of approximately 10 days. This could be due to the beginning of the development of nitrifying communities on the biocarriers. The ammonia and nitrite levels dropped back to previous values in the effluent in less than 2 weeks, possibly due to the stabilization of the AOB and NOB communities in the attached growth. It has to be noted that the differences in ammonia and nitrite concentrations are minimal and could have arisen within the uncertainty of the measurements.

A small metal grid was positioned in front of the overflow tube to prevent the biocarriers from escaping the reactor via the overflow. However, by day 85, an issue emerged as certain biocarriers became entangled with the grid, obstructing the overflow mechanism. Due to the grid lacking a top cover, the liquid level within the reactor rose, causing mixed liquor and biocarriers to spill over the grid. This situation led to concerns, such as the loss of attached growth from the biocarriers and the clogging of the overflow tube. The clogged overflow tube posed a potential risk of the liquid level exceeding the reactor's upper limit. To prevent the previously mentioned issues, the aeration of MBR-2 was temporarily decreased so that the carriers stayed in the lower part of the reactor. In the meantime, a new grid with a smaller grid size and an added top part was constructed. The new grid effectively prevented carriers from entering the overflow tube. Once a week, accumulated carriers were removed from the grid surface with a brush. From day 85, aeration was decreased in MBR-2, and nitrate increased back to the range from before the introduction of the biocarriers (approximately 9 mg/L). The biocarriers were not homogeneously distributed anymore in the reactor since the majority accumulated at the bottom. Nitrate was still produced at the same rate by the nitrifying community. Due to the flow pattern and sunken biocarriers after day 85, most of the nitrate did not reach the anoxic denitrifying zones efficiently, resulting in the breakthrough of nitrate in the effluent of MBR-2. Ammonia and nitrite in the effluent stayed the same as before day 85. Nitrification was, unlike denitrification, performed by the suspended biomass. The reduced aeration was thus still sufficient to evenly distribute the MLSS and provide adequate DO levels throughout the reactor.

Aeration was increased again in both MBRs on day 92. The biocarriers in MBR-2 were much more homogeneously distributed due to improved mixing by the rising air bubbles. Nitrate removal was expected to improve hereafter. However, nitrate in the effluent did not decrease. The additional operational issues on days 95, 112, and 123 probably obstructed the re-development of biofilms on the carriers.

5.4 Nitrification Activity of the MBR Mixed Liquor

The MLSS concentration of test A1 was 950 mg/L, which corresponds to the lower MLSS concentration range in the MBRs of the first two weeks of the operational period. Tests A2, B1, and B2 had higher MLSS concentrations, between 2200 and 2500 mg/L. The COD content of the batch test synthetic greywater was reduced to minimize the activity of heterotrophic microorganisms. Because the autotrophic nitrifiers exhibit a slow growth rate, it was assumed that the biomass concentration remained constant throughout the batch tests (Li et al., 2006). The MLVSS/MLSS ratios of the batch tests were between 0.95 and 0.99, suggesting that almost all the suspended solids from the sludge were organic. Some other papers that worked with membrane bioreactors for wastewater treatment documented MLVSS/MLSS ratios of up to 0.90 (He et al., 2006; Kim et al., 2021; Sabouhi et al., 2020). The synthetic greywater used for the batch tests in this thesis was probably characterized by minimal inorganic content. This assumption is supported by the high biodegradability derived from the BOD_5 tests. Additionally, potential laboratory inaccuracies during the determination of MLSS and MLVSS contents might have contributed to an overestimation of the MLVSS/MLSS ratios.

The specific ammonia oxidation rate (SAOR) from test A1 was 3.13 mg NH_4^+-N/g VSS·h, which was more than double that from tests A2, B1, and B2. The SAOR of all tests falls within the range reported in the literature (Dytczak et al., 2008; Scheumann & Kraume, 2009). A possible explanation for the lower SAOR of tests A2, B1, and B2 is that they coincidentally might have contained more dead and old biomass than test A1. The sludge had been stored for some time (up to two weeks) before the batch test, which probably resulted in the die-off of part of the nitrifiers. As the MLVSS concentration represents both living and dead biomass (Hasar & Kinaci, 2004), this hypothesis cannot therefore be verified with the available information.

The measured ammonia concentrations from tests B1 and B2 did show a linearly decreasing trend. The R^2 of the linear regressions of the ammonia concentrations in tests B1 and B2 were 0.48 and 0.53, respectively. The ammonia concentrations even increased several times during the tests. The suspected reason for this is the poor mixing and the subsequent limited oxygen transport in the beakers. The mixed liquor of tests B1 and B2 was sampled more frequently than tests A1 and A2, causing the liquid level to decrease faster during the course of the tests. Due to the low liquid level, the aeration stone did not stay completely submerged at all times during the tests. Mixing was also not optimal during tests B1 and B2, due to the continuous presence of a thermometer and pH probe combined with the low liquid level. The more limited mixing and oxygen transport could have created zones of low DO levels in the batch test mixed liquor, where nitrification could have been limited. Sampling from such a zone in the mixed liquor could have led to higher ammonia concentrations than the previous sample. Moreover, errors due to the spectrophotometric analysis, as discussed in Section 5.1, could have exacerbated this.

Ammonia removal in tests A1 and A2 occurred linearly, each with an R^2 of 0.97. This indicates zero-order kinetics, suggesting that ammonia oxidation was not inhibited by the concentrations of greywater components or formed products (Li et al., 2006). After 4 hours, *i.e.*, the duration of the tests, ammonia concentrations were reduced to approximately 10 mg N/L. In tests B1 and B2, ammonia levels were near 0 mg/L at the end of the tests. The MLSS contents of tests B1 and B2 were relatively similar to those of tests A1 and A2. It was expected that the microbial communities in the sludge from the series B tests could have been changed towards a higher nitrification capacity in order to almost completely remove ammonia. The sludge from test series B was obtained from the MBRs after they had been operated for three months under a higher aeration regime, which might have caused a shift in the nitrifying populations (Kraume et al., 2005). However, the calculated SAORs of tests B1 and B2 are similar to those of test A2, indicating that they have similar nitrification activities. The uncertainty of the ammonia measurements and the low R^2 of the SAORs of test series B make it impossible to confirm any hypothesis explaining differences in ammonia removal between test series A and B.

The nitrogen originating from oxidized ammonia undergoes three potential fates: it is incorporated into new biomass as organic nitrogen by the nitrifiers, converted into nitrite and nitrate through nitrification, or subjected to denitrification, leading to its conversion into gaseous nitrogen. Nitrifiers are slow growers, so the fraction of nitrogen incorporated into new biomass is typically limited during short batch tests like these. Tests A1 and A2 demonstrated minimal nitrite accumulation, suggesting an equilibrium between the two nitrification phases: nitrite production (through ammonia oxidation) and nitrite removal (via oxidation to nitrate). In all the batch tests (A1, A2, B1, and B2), the accumulation of nitrate was lower than the oxidation of ammonia, indicating that a portion of the nitrified ammonia underwent subsequent denitrification. Denitrification could have taken place inside granular sludge flocs. Test B2 did not exhibit a decreased nitrate buildup compared to B1, implying a comparable level of denitrification in both tests. Therefore, the biocarriers used in B2 probably did not effectively enhance denitrification in this scenario, possibly due to the limited extent of microbial growth attached to the introduced biocarriers.

The total nitrogen concentrations from the samples during the first two hours of tests A1 and A2 were lower than the corresponding inorganic nitrogen (NH_4^+, NO_2^-, and NO_3^-) concentrations. This is not possible and probably originated from incorrect measurements, as discussed in Section 5.1. It is interesting to note that this was only the case for the first half of the batch tests. Over the course of the batch tests, the dissolved inorganic nitrogen declined, due to bioaccumulation and denitrification. The samples of mixed liquor were filtered before analysis, which excluded the assimilated nitrogen from TN measurements. Simultaneously, the dissolved organic nitrogen content increased during the batch tests, due to the breakdown of microorganisms through cell lysis (Xiao et al., 2020). As the batch tests progressed, the rise in organic nitrogen content and the decline in inorganic nitrogen content led to an increasing discrepancy between total nitrogen and inorganic nitrogen concentrations. This might explain why TN surpassed inorganic nitrogen concentrations during the later stages of the tests.

The batch tests had some limitations that could be improved in the future. To characterize the nitrification activity more specifically per MBR, the sludge of only one MBR should be used per batch test. Differences in HRT, for example, can influence the nitrification activity. These effects could not be investigated in the batch tests, because they used mixed overflow sludge from both reactors. A second point of improvement is the batch test volume. Lager beakers should be used to optimize mixing and aeration. Moreover, a higher number of replicates can always help increase the reliability of the obtained results.

5.5 Comparison of MBR Effluent with Reuse Guidelines

In this thesis, the greywater treatment performance of MBRs under different operational conditions was monitored. Table 5.1 lists the average effluent quality of the MBRs once they stabilized to their operational conditions, together with the ISO 30500 and Italian guidelines on the reuse of greywater. An overview of more guidelines can be found in Table 1.2.

The pH of the mixed liquor in both MBRs, which was assumed to be representative for the effluent pH, was continuously measured. In all three considered MBR phases, the pH dropped below the minimal pH criterion for non-potable reuse. A pH below 4 was recorded in MBR-1 when it was operated under the optimized aeration flow rate of 1 L/min. The measured pH of the MB-MBR system was at all times below the minimum required pH level of 6. However, the pH sensors of both MBRs were often not completely submerged in the mixed liquor, which reduces the reliability of their measurements. If these low pH levels were in fact prevailing in the MBRs, nitrification would have been detrimentally affected (Tarre & Green, 2004). No deteriorated nitrification was observed during the experimental period, questioning the reliability of the pH measurements.

Table 5.1: The effluent quality of the MBRs operating under low aeration conditions ($Q_{air,low}$), MBRs operating under optimized aeration conditions ($Q_{air,opt}$), and the MB-MBR system, along with the corresponding water quality criteria outlined in the ISO 30500 and Italian guidelines for wastewater reuse. The aeration flow rates of $Q_{air,opt}$ and $Q_{air,low}$ were 0.6 and 1 L/min, respectively. Guideline criteria are retrieved from Reynaert et al. (2020).

Parameter		**MBR**$^{(1)}$ $Q_{air,low}$	**MBR**$^{(2)}$ $Q_{air,opt}$	**MB-MBR**$^{(3)}$	**Italy**	**ISO 30500**
pH		5.5 – 7.9	3.8 – 6.8	5.2 – 5.6	6.0 – 9.5	6 – 9
COD	mg/L	27 – 108 (51)	16 – 78 (38)	18 – 42 (27)	⩽ 100	⩽ 50
TN	mg N/L	11 – 18 (15)	8 – 21 (15)	2 – 7 (4)	⩽ 15	
TN removal	%	0 – 34 (20)	0 – 72 (34)	77 – 91 (85)		⩾ 70
Ammonia	mg N/L	6 – 21 (14)	< 1.5	1 – 3 (2)	⩽ 1.6	
Nitrite	mg N/L	< 1.5	< 1.5	< 0.5		
Nitrate	mg N/L	0 – 5 (1)	2 – 12 (9)	1 – 4 (2)		

(1) Average effluent quality of MBR-1 and MBR-2 between days 1 and 36.
(2) Average effluent quality of MBR-1 (between days 54 and 92) and MBR-2 (between days 43 and 71).
(3) Average effluent quality of MBR-2 between days 78 and 85.

The MBRs operated at low aeration (*i.e.*, an air flow rate of 0.6 L/min) and did not comply with either of the guidelines, which had a COD criterium (ISO 30500 and Italy). It might be noteworthy that only one sample during this phase exceeded the COD limit of Italy of 100 mg/L, while all the other samples had a concentration below 70 mg/L. The exceeded COD sample may, however, not be ignored, as guidelines or regulations should be fulfilled at all times. Sampling of the MBR effluent only took place every 4 days, leaving the complete image of the effluent quality missing. The single exceeding COD sample could have been an indication of a recurring phenomenon. The ammonia and total nitrogen guidelines were not met by the MBR system at low aeration. Increasing the aeration flow rate from 0.6 L/min to 1 L/min caused an improvement in the removal of COD, TN, and ammonia. All the measured effluent concentrations of COD were below the criterium of 100 mg/L of the Italian guidelines but did not fulfill the required criterium of the ISO 30500 guidelines. Ammonia removal was adequately improved, resulting in compliance with all the selected guidelines that specify maximal ammonia concentrations (Italy and China in Table 1.2). Total nitrogen, however, did not meet either the ISO 30500 or the Italian criteria for reuse. This is due to the nitrate buildup as a result of the improved nitrification.

During the selected stabilized state, the MB-MBR removed COD below the maximum concentrations presented by the ISO 30500 and Italian guidelines. The ammonia effluent concentrations were below the required effluent concentrations of the North Carolinian and Chinese guidelines (7.8 and 6 mg N/L, respectively), but not for the Italian guidelines (1.6 mg N/L). The total nitrogen removal efficiency and effluent concentrations were adequate at all times in accordance with ISO 30500 and Italian guidelines, thanks to the improved nitrate removal by the biofilms. The treatment performance of the MB-MBR was assessed for a time series of only 8 days. Operational problems occurred after this period, causing an immediate deterioration of the nitrate removal. Therefore, only the time series of days 78 to 85 is used in this chapter to assess compliance with the guidelines, as it is perceived as the stabilized treatment performance of the MB-MBR system. Even though the effluent quality from this period complies with the literature () it remains a limitation in this thesis research. The effluent quality values presented in Table 5.1 of the MB-MBR give a hint of the potential of this technology for the removal of nitrate.

Most guidelines include constraints for turbidity, suspended solids, BOD_5, and microbial parameters, as can be seen in Table 1.2. In this thesis, however, these parameters were not monitored in the MBR effluent. Investigating compliance of the MBR effluent with the complete criteria of existing guidelines for non-potable reuse was therefore not possible. Sampling of the MBR effluent was carried out only twice a week, resulting in a less complete insight into the MBR treatment performance and any possible contaminant breakthroughs. This is especially important when effluent contaminant concentrations approach the limits posed by the guidelines. More frequent sampling becomes necessary to validate the continuous compliance of the MBR treatment with the guidelines.

Chapter 6

CONCLUSIONS

6.1 General Conclusions

The main objective of this thesis was to obtain sufficient removal of COD and nitrogen in an aerobic MBR system to meet their criteria in existing guidelines for non-potable greywater reuse. This was realized by optimizing the aeration flow rate and adding biocarriers into the submerged MBR. An aeration flow rate of 0.6 L/min to the reactor of 10 L was insufficient for good nitrification. When aeration was supplied at a flow rate of 1 L/min, adequate nitrification ($[NH_4^+]_{eff}$ < 3 mg N/L) was obtained. Biocarriers were added at a 10% (v/v) filling ratio, ensuring adequate denitrification ($[NO_3^-]_{eff}$ < 4 mg/L). The HRT and SRT of the MB-MBR were 15 hours and 21 days, respectively. As a result of these optimized operating conditions, the permeate quality of the MBR system was considerably improved, containing maximal concentrations of COD, total nitrogen, and ammonia of 42 mg/L, 6 mg N/L, and 3 mg N/L, respectively. Therefore, the moving bed MBR met the criteria for COD and total nitrogen removal of the international (ISO 30500) and the Italian guidelines. The maximal ammonia limit of the Italian guidelines was surpassed, but the maximal ammonia limit of the Chinese guidelines was always respected.

The second main objective of this thesis was to elucidate the effects of the optimized operating conditions on the potential for membrane fouling. The membrane fouling rate was found to be much lower in the MBR with an HRT of 20 h, compared to the MBR with an HRT of 15 h. When the MLSS increased, higher membrane fouling rates were observed. Increased aeration also had an impact on membrane fouling, due to the intensifying scouring effect of the rising air bubbles. Every time the air flow rate was increased, the accumulated fouling layer was partly removed, and the subsequent rate of fouling became lower.

The use of aerated MBRs had previously been identified in several research papers as a suitable technology for the treatment of greywater. Its practical implementation in building level greywater reclamation is unfortunately not widespread yet. However, in the past few years, wastewater recovery has gained momentum in Belgium, probably due to the increasing pressure of water scarcity issues. In Ghent, two innovative neighborhood construction projects have included aerated MBRs for greywater reclamation. In the *Nieuwe Dokken* project, the treated greywater is recuperated as process water for nearby industry (Demolder, 2020), while in the project *Cohousing Jean*, the treated greywater is reused as toilet flushing water (VLAIO, 2022). The research presented in this thesis demonstrated the appropriateness of MBRs, and MB-MBRs specifically, on a lab scale installation, for the treatment of greywater with the intention of non-potable reuse. This supplementary scientific evidence can play a significant role in persuading policymakers and facilitating the broader adoption of MBR systems for small scale greywater reuse.

6.2 Future perspectives

High denitrification was initially obtained in the MB-MBR. Nevertheless, the MB-MBR overflow grid faced rapid obstruction due to the accumulation of small biocarriers. This obstruction resulted in sludge loss from the reactor, necessitating a reduction in aeration to facilitate the repair of the grid. Nitrate removal was subsequently deteriorated, resulting in only a short timeframe of effective treatment performance of the MB-MBR. In future research involving overflow implementation, the potential issue of biocarriers passing through or becoming trapped on the overflow grid should be meticulously considered. To mitigate this, it is advisable to design the grid with a sufficiently small aperture size. Additionally, establishing a regular maintenance routine for frequent inspection and cleaning is crucial, especially when carriers and sludge accumulation onto the grid becomes evident. An additional problem with the biocarriers in the MB-MBR was their tendency to sink and accumulate at the bottom of the reactor. To address this, a strategy should be developed to maintain effective suspension and mixing of the biocarriers throughout the bioreactor.

The primary focus of this thesis was the efficient removal of COD, inorganic nitrogen forms, and total nitrogen from greywater. However, many guidelines for greywater reclamation include a broader range of water quality criteria, including BOD_5, total suspended solids, total phosphorus, and microbial parameters such as total coliforms or *E. coli*. To comprehensively assess the adherence of the treated effluent of the MBR system to existing guidelines, it is essential to monitor all parameters specified in these guidelines. In future research, an increased sampling frequency of the effluent could be adopted to gain a more complete insight of the MBR system's treatment performance robustness.

Synthetic greywater was produced in the laboratory to simulate greywater, maintaining relatively consistent characteristics. Nonetheless, testing with actual greywater is crucial due to its wider range of pollutants and pathogens. Real greywater production typically exhibits a diurnal contamination load variation. Although this temporal pattern was not replicated in this thesis, exploring the impact of this temporal pattern on the treatment efficiency of the MBR system could bring valuable insights.

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APPENDIX A

ADDITIONAL TABLE

Table A.1: Brands of the personal care products and chemicals used in the preparation of synthetic greywater.

Component	**Brand**
Personal care products	
Dishwashing detergent	Trio Tumyeonghan Saenggak
Liquid laundry	LiQ Baking Soda Concentrated Gel
Shampoo	Elastine Propolithera Anti Aging Care
Shaving gel	Gillette Fusion Hydra Gel Sensitive Skin
Shower gel	ON:THEBODY Ultra Moisture Cotton Flower Scent
Toothpaste	2080 Ginghivalis Original
Chemicals	
Ammonium chloride	Daejung (reagent grade)
Calcium chloride	
Potassium phosphate dibasic	
Sodium acetate	
Sodium bicarbonate	
Sodium chloride	
Sodium sulfate	