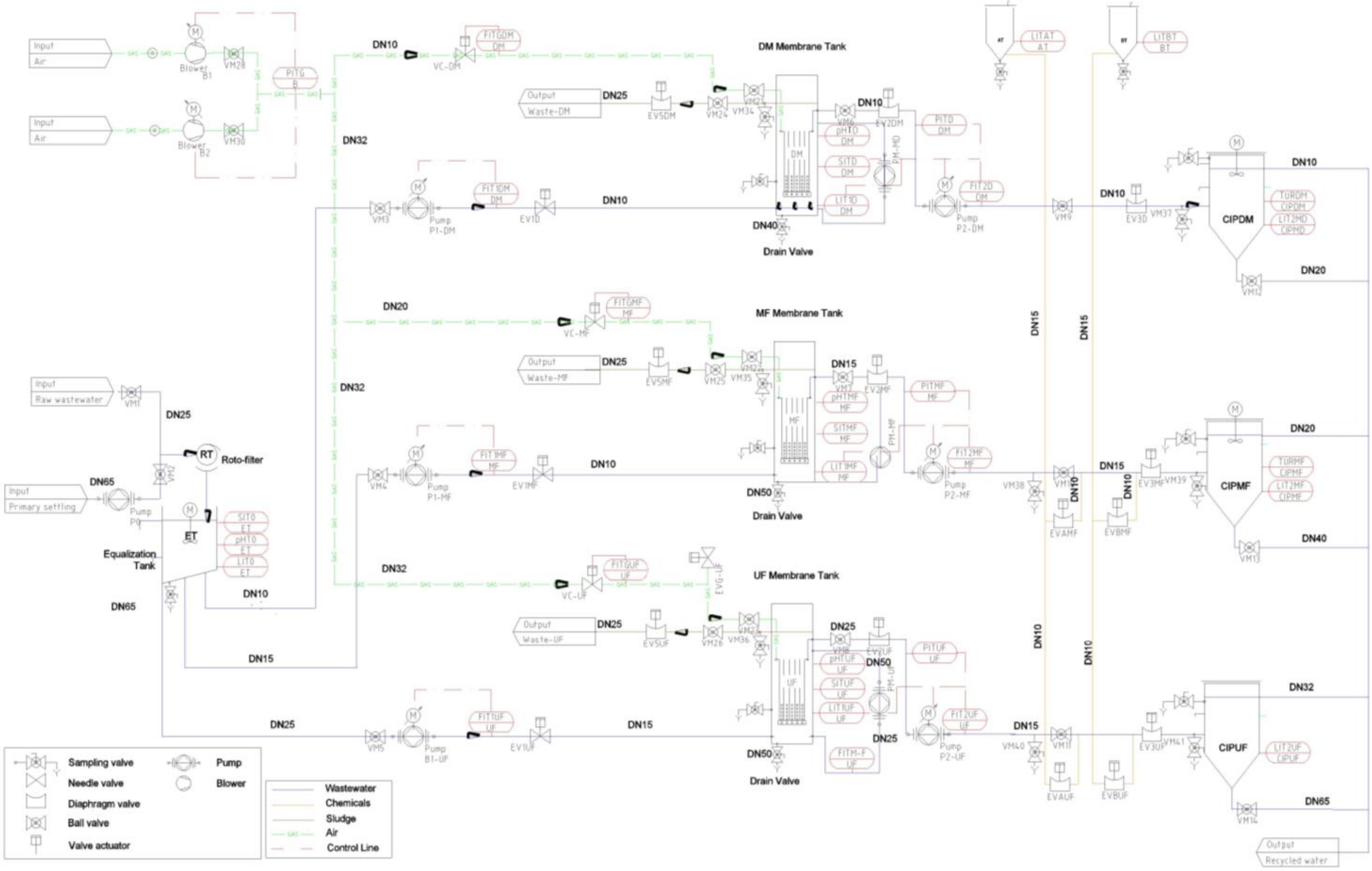
FILTRACIÓN DIRECTA POR MEMBRANAS PARA POTENCIAR LA RECUPERACIÓN DE RECURSOS DE LAS AGUAS RESIDUALES

Programa de doctorado en ingeniería química, ambiental y de procesos



TESIS DOCTORAL
Pau Sanchis Perucho
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DIRECT MEMBRANE FILTRATION TO BOOST RESOURCE RECOVERY FROM MUNICIPAL WASTEWATER

Doctoral Program in Chemical, Environmental and Process Engineering

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CERTIFICAN:

que la presente memoria, titulada "**FILTRACIÓN DIRECTA POR MEMBRANAS PARA POTENCIAR LA RECUPERACIÓN DE RECURSOS DE LAS AGUAS RESIDUALES**", corresponde al trabajo realizado bajo su dirección por **D. PAU SANCHIS PERUCHO** (DNI 20455928G), para su presentación como tesis doctoral en el programa de doctorado en Ingeniería Química, Ambiental y de Procesos de la Universitat de València.

Los artículos presentados en esta tesis doctoral no se han utilizado ni se utilizarán en ninguna otra tesis doctoral.

Y para que conste a los efectos oportuns, firman el presente certificado en Valencia, a la fecha de la firma.

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in memoriam

"Quienes creen en la física, saben que la distinción entre el pasado, el presente y el futuro es sólo una ilusión obstinadamente persistente"

Albert Einstein

Abstract

Direct membrane filtration (DMF) is a potential alternative to boost resource recovery within municipal wastewater (MWW) treatment field. This technology aims to enhance particulate matter recovery from MWW by implementing a membrane filtration unit as a previous step to the biological secondary treatment, thereby reducing the energy requirements associated to the aerobic oxidation of organic matter in activated sludge systems. Additionally, the particulate organics retained and concentrated in the DMF tank can be used to increase methane production via anaerobic digestion, enhancing the energy balance of the overall wastewater treatment.

This PhD thesis aimed to determine the most suitable membrane technology (microfiltration, ultrafiltration, or dynamic membranes) to perform the direct filtration of municipal wastewater. Additionally, different influent types (*i.e.* raw wastewater and primary settler supernatant) and operating conditions (*i.e.* solids concentration, transmembrane flux, and supporting material pore size for dynamic membranes) were studied in order to determine effective design and operational strategies to minimize fouling while maximizing resource recovery in the long-term operation. To this aim, a membrane-based pilot plant equipped with commercial membrane modules was operated for more than 3 years, which was fed with MWW coming from a full-scale municipal wastewater treatment facility.

The relationship between membrane pore size and influent particles size distribution was found to be one of the major fouling controllers, identifying UF technology much more suitable than MF for DMF of MWW. Operating at relatively high suspended solid concentrations in the membrane tank (between 6 – 11 g L^{-1}) allowed minimizing membrane fouling. Applying low/middle transmembrane fluxes (around 10 LMH) also kept fouling propensity in low levels, *i.e.* severe fouling intensities were observed when the transmembrane flux was increased. Both tested influent sources (*i.e.* raw wastewater and primary settler supernatant) showed similar fouling propensities when operating at adequate suspended solids concentrations in the membrane tank. Sludge retention times below 3 days resulted in negligible organic matter losses due to biodegradation in the membrane tank. Organic matter was found to be the major fouling promotor during filtration (permeability losses from 75 to 99% depending on the porous membrane

employed). UF treatment scheme generated a high quality permeate meeting European discharge requirements for non-sensible discharge environments. Moreover, promising results were obtained from preliminary energy, economic, and carbon footprint analysis.

Sludge filterability tests identified the total solid concentration as the dominant parameter affecting sludge resistance to filtration when treating non-biological sludge. The time to filter filterability methodology was identified as the most convenient one compared to the capillary suction time and specific resistance to filtration due to its higher sensibility when treating non-biological sludge.

A simple and generic model was proposed to predict/capture transmembrane pressure dynamics during UF operation treating raw MWW and primary settler supernatant. This mathematical model was able to properly predict membrane fouling within the range of operating conditions evaluated.

Dynamic membranes were identified as a potential alternative for DMF of MWW, mainly due to reduced investment, replacement, and maintenance cost compared to porous membranes. Raw MWW treatment allowed to self-form a dynamic membrane in the short-term. Since the permeate quality was not significantly improved by using more restrictive supporting materials, the use of a unique supporting material layer with 5 µm of pore size was established in this study as the most convenient alternative. Unfortunately, the permeate quality produced was far from meeting the European discharge standards, while the energy and carbon footprint improvements that this membrane technology could achieve compared to primary settling where not high enough to justify the replacement of the later in existing municipal facilities.

Resumen

La filtración directa mediante membranas (FDM) es una atractiva alternativa para potenciar la recuperación de recursos en el ámbito del tratamiento de aguas residuales municipales (ARM). El objetivo de esta tecnología es mejorar la recuperación del material particulado afluente de las ARM mediante la implementación de una unidad de filtración por membrana como paso previo al tratamiento biológico secundario, reduciendo así las necesidades energéticas asociadas a la oxidación aeróbica de la materia orgánica en los sistemas de fangos activos. Asimismo, el fango concentrado en el tanque de membranas puede emplearse para incrementar la producción de metano durante el proceso de digestión anaerobia, mejorando el balance energético global del tratamiento de ARM.

Esta tesis doctoral tuvo como objetivo determinar la tecnología de membranas más adecuada (microfiltración, ultrafiltración o membranas dinámicas) para realizar la FDM de ARM. Además, se estudiaron diferentes tipos de influentes (agua residual bruta y sobrenadante del decantador primario) y condiciones de operación (concentración de sólidos en el tanque de membranas, flujo transmembrana y tamaño de poro del material de soporte en el caso de la membrana dinámica) con el fin de identificar eficaces estrategias operativas y de diseño para minimizar el ensuciamiento de la membrana y maximizar la recuperación de recursos durante la operación a largo plazo. Dicho estudio se realizó mediante la operación de una planta piloto basada en membranas (equipada con módulos de membrana comerciales) durante un periodo de más de 3 años, siendo esta alimentada con ARM procedente de una instalación para el tratamiento de ARM a escala industrial.

La relación entre el tamaño de poro de la membrana y la distribución de tamaños de partícula en el afluente tratado se identificó como una de las principales variables involucradas en el ensuciamiento de la membrana. En este caso, la tecnología de UF se determinó como mucho más adecuada en comparación a la MF debido a su menor propensión al ensuciamiento durante la FDM de ARM. Por otro lado, operar a concentraciones de sólidos suspendidos relativamente elevadas en el tanque de membranas (entre 6 – 11 g L^{-1}) permitió minimizar la propensión al ensuciamiento. La aplicación de flujos transmembrana bajos/medios (alrededor de 10 LMH) también fue necesaria para mantener el ensuciamiento de la membrana en niveles bajos, observado un

severo incremento del encaminamiento al aumentar dicho flujo. Los dos afluentes tratados (agua residual bruta y sobrenadante de la decantación primaria) presentaron similares propensiones al ensuciamiento al operar bajo adecuadas concentraciones de sólidos suspendidos en el tanque de membrana. Un tiempo de retención del fango de 3 días fue identificado como límite superior para evitar pérdidas significativas de la materia orgánica recuperada en el tanque de membranas debido a su biodegradación. La materia orgánica se determinó como el principal promotor del ensuciamiento durante la filtración de ARM, registrándose perdidas en la permeabilidad de la membrana del 75 al 99% en función del tamaño de poro de la membrana empleada. La FDM de ARM fue capaz de producir un permeado de alta calidad, capaz de cumplir con los estándares impuestos por la normativa europea en cuanto a vertidos en ambientes no sensibles. Asimismo, se obtuvieron resultados prometedores respecto a los análisis preliminares de impacto energético, económico y de huella de carbono del proceso.

Los ensayos de filtrabilidad del fango identificaron la concentración de solidos suspendidos como variable dominante respecto a la resistencia a la filtración de fangos no biológicos. El método de tiempo de filtración (del inglés, *time to filter*) se determinó como la más adecuada en comparación con el tiempo de succión capilar (del inglés, *capillary suction time*) y la resistencia específica a la filtración (del inglés, *specific resistance to filtration*) debido a su mayor sensibilidad al tratar fangos sin actividad biológica.

Un modelo simple y genérico fue propuesto para predecir la dinámica de la presión transmembrana durante la filtración de ARM (agua residual bruta y sobrenadante de la decantación primaria) mediante membranas de UF. Dicho modelo fue capaz de predecir adecuadamente el ensuciamiento de la membrana dentro del rango de condiciones evaluadas.

Las membranas dinámicas se identificaron como una atractiva alternativa para la FDM de ARM debido sus extremadamente inferiores coste de inversión y sustitución de las membranas, así como su significativo menor coste de mantenimiento y operación en comparación con las membranas porosas. La auto-formación de una membrana dinámica a corto plazo fue posible al tratar agua residual bruta. Dado que la calidad del permeado no mejoró significativamente al emplear materiales de soporte más restrictivos, se estableció el uso de una única capa de material de soporte con 5 µm de tamaño de poro como la alternativa más conveniente dentro de las condiciones estudiadas en este trabajo. Lamentablemente, la calidad del permeado producido por esta tecnología no fue capaz de cumplir con la normativa europea de vertido. Además, las mejoras energéticas y de huella de carbono obtenidas por la misma en comparación con la decantación primaria no fueron lo suficientemente elevadas como para justificar su uso en instalaciones ya en funcionamiento.

Resum

La filtració directa mitjançant membranes (FDM) és una atractiva alternativa per a potenciar la recuperació de recursos en l'àmbit del tractament d'aigües residuals municipals (ARM). L'objectiu d'aquesta tecnologia és millorar la recuperació del material particulat afluent de les ARM mitjançant la implementació d'una unitat de filtració per membrana com a pas previ al tractament biològic secundari, reduint així les necessitats energètiques associades a l'oxidació aeròbica de la matèria orgànica en els sistemes de fangs actius. Així mateix, el fang concentrat en el tanc de membranes pot emprar-se per a incrementar la producció de metà durant el procés de digestió anaeròbia, millorant el balanç energètic global del tractament de ARM.

La present tesi doctoral tingué per objectiu el determinar la tecnologia de membranes més adequada (microfiltració, ultrafiltració o membranes dinàmiques) per a realitzar la FDM de ARM. Així mateix, es van estudiar diferents tipus d'afluent (aigua residual bruta i sobrenedant de la decantació primària) i condicions d'operació (concentració de sòlids en el tanc de membranes, flux transmembrana i grandària del porus del material de suport en el cas de la membrana dinàmica) amb la finalitat d'identificar eficaces estratègies operatives i de disseny per a minimitzar l'embrutiment de la membrana i maximitzar la recuperació de recursos durant l'operació a llarg termini. Aquest estudi es va realitzar mitjançant l'operació d'una planta pilot basada en membranes (equipada amb mòduls de membrana comercials) durant un període de més de 3 anys, sent aquesta alimentada amb ARM procedent d'una instal·lació per al tractament de ARM a escala industrial.

La relació entre la grandària de porus de la membrana i la distribució de grandàries de partícula en l'afluent tractat s'identificà com una de les principals variables involucrades en l'embrutiment de la membrana. En aquest cas, la tecnologia de UF es va determinar com a molt més adequada en comparació a la MF degut a de la seva menor propensió a l'embrutiment durant la FDM de ARM. D'altra banda, operar a concentracions de sòlids suspesos relativament elevades en el tanc de membranes (entre 6 – 11 g L^{-1}) va permetre minimitzar la propensió a l'embrutiment. L'aplicació de fluxos transmembrana baixos/moderats (al voltant de 10 LMH) també va ser necessari per a mantenir l'embrutiment de la membrana en nivells baixos, observat un sever increment de l'encaminament en augmentar aquest flux. Els dos afluents tractats (aigua residual bruta i sobrenedant de la decantació primària) presentaren similars propensions a l'embrutiment

en operar sota adequades concentracions de sòlids suspesos en el tanc de membrana. Un temps de retenció del fang de 3 dies s'identificà com a límit superior per a evitar pèrdues significatives de la matèria orgànica recuperada en el tanc de membranes a causa de la seva biodegradació. La matèria orgànica es va determinar com el principal promotor de l'embrutiment durant la filtració de ARM, registrant-se perdudes en la permeabilitat de la membrana del 75 al 99% en funció de la grandària de porus de la membrana emprada. La FDM de ARM va ser capaç de produir un permeat d'alta qualitat, complint amb els estàndards imposats per la normativa europea respecte al seu abocament en ambients no sensibles. Així mateix, s'obtingueren resultats prometedors respecte als anàlisis preliminars d'impacte energètic, econòmic i de petjada de carboni del procés.

Els assajos de filtrabilitat del fang identificaren la concentració de sòlids suspesos com a variable dominant respecte a la resistència a la filtració de fangs no biològics. El mètode de temps de filtració (de l'anglès, *time to filter*) es determinà com el més adequat en comparació amb el temps de succió capil·lar (de l'anglès, *capillary suction time*) i la resistència específica a la filtració (de l'anglès, *specific resistance to filtration*) a causa de la seva major sensibilitat en tractar fangs sense activitat biològica.

Es proposà un model simple i genèric per tal de predir la dinàmica sobre la pressió transmembrana durant la filtració de ARM (aigua residual bruta i sobrenedant de la decantació primària) mitjançant membranes de UF. Aquest model va ser capaç de predir adequadament l'embrutiment de la membrana dins del rang de condicions avaluades.

Les membranes dinàmiques s'identificaren com una atractiva alternativa per a realitzar la FDM de ARM degut als seus extremadament inferiors costos d'inversió i substitució de les membranes, així com el seu significatiu menor cost de manteniment i operació en comparació amb les membranes poroses. L'auto-formació d'una membrana dinàmica a curt termini va ser possible al tractar aigua residual bruta. Atès que la qualitat del permeat no millorà significativament en emprar materials de suport més restrictius, s'establí l'ús d'una única capa de material de suport amb 5 µm de grandària de porus com l'alternativa més convenient dins de les condicions estudiades en aquest treball. Lamentablement, la qualitat del permeat produït per aquesta tecnologia no va ser capaç de complir amb la normativa europea d'abocament. A més, les millores energètiques i de petjada de carboni obtingudes per la mateixa en comparació amb la decantació primària no van ser prou elevades com per a justificar-ne el seu ús en instal·lacions ja en funcionament.

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CHAPTER 1. Introduction: Application of Membrane Technology for the Direct Filtration of Municipal Wastewater

Fresh water stress and climate change are two of most important worldwide problems to address over next decades [1.1; 1.2]. Global water scarcity has been worryingly raising in the last years [1.3] even in regions with a consistent pluvial regime [1.4], turning the search of high quality water sources a prominent policy around different countries [1.5]. On the other hand, greenhouse gas (GHG) emissions forecasted for the next years predict trends significantly higher than those necessary for the 2 ºC-target for global temperature stabilization [1.6], not being in track for meeting the established Paris agreement objectives. In addition to these issues, energy crisis is also a prominent matter to solve in the coming years [1.7]. Other essential resources, such as phosphorous for agriculture, are also approximating to their exhaustion due to the increasing population growth and food demand, auguring possible scarcities in a not distant future [1.2]. All these problems, which are in several cases interconnected with each other, are exposing the limitations of current economic models and worldwide policies, demanding a new approach on human interaction with the environment. Indeed, the non-renewable resource exploitation and linear economy approaches used during the last century have showed its unsuitability to sustain (developed) societies in the long-term, strongly contributing and aggravating all cited issues. Consequently, numerous experts are exposing the urgent necessity of adapting our consuming system to circular economy (CE) models in order to achieve sustainability on future human activity development [1.8], as well as to develop greener and more energy-efficient technologies to minimize our impact on the environment.

1.1 Current municipal wastewater treatment

In the context of municipal wastewater (MWW) treatment, the CE approach has gained especial interest in the last years due to all the potential resources that they contain. MWW is now regarded as a source of reclaimed water, carbon, energy, and essential nutrients (mainly nitrogen and phosphorus) [1.9]. Unfortunately, most MWW treatment processes fail on recovering a notorious fraction of these resources. Historically, municipal wastewater treatments plants (WWTP) were designed only to protect human health and to avoid the important environmental impact that the direct MWW discharge produce to receiving ecosystems and water bodies. Consequently, WWTP were composed of different physical, chemical, and biological steps aimed to reach acceptable depuration levels. Despite some modifications have been implemented for process enhancement, the same basic scheme has been mostly implemented so far worldwide (see Fig. 1.1a), even though its limitations have been consistently reported by the scientific community during the last decades.

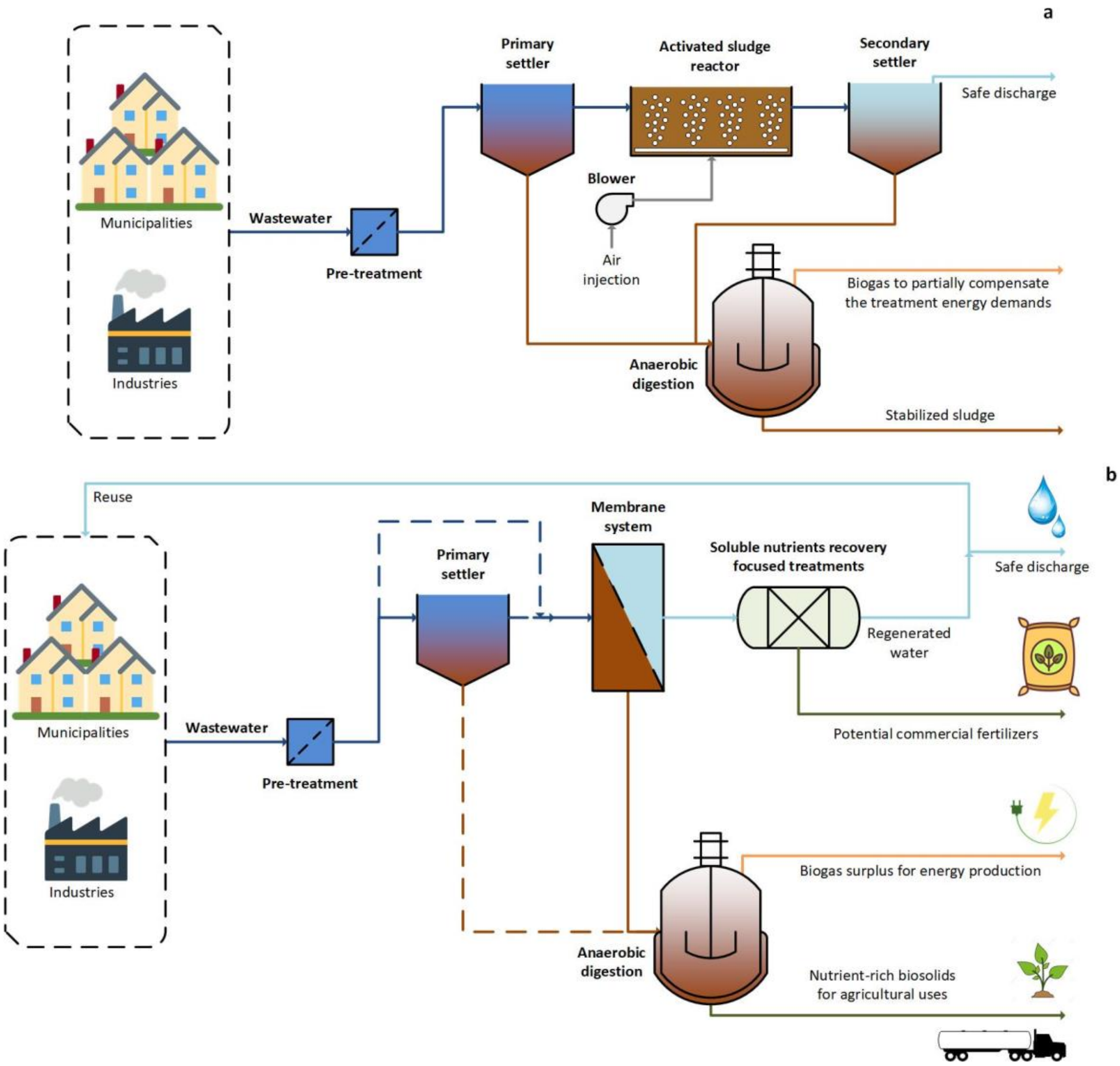

Figure 1.1. MWW treatment scheme: (a) classic management and (b) direct filtration concept.

MWW treatment core generally depends on an aerobic biological treatment, commonly known as activated sludge (AS) process, aimed to remove organic matter (OM) through biological oxidation, losing the recovery potential of carbon and energy from OM while requiring significant energy inputs in the aerobic reactor. Indeed, AS is recognized as an energy-intensive process with energy demands that can reach up to the 50% of the total energy requirements of a full-scale WWTP [1.10]. In addition, associated nitrification-denitrification and phosphorous chemical precipitation processes fail to recover valuable nutrients from MWW, representing even a negative impact due to possible emissions of N_2O gas to the atmosphere [1.11] and chemicals expenses.

Instead, anaerobic technology is extremely more convenient for MWW management since it does not require aeration while energy can be obtained from the OM valorization via methane production (see Fig. 1.2). Additionally, influent nutrients can be mineralized during the treatment, allowing their later recovery, while lower sludge production rates are expected compared to aerobic systems, reducing the associated management expenses [1.12]. Unfortunately, the slower growth rate of anaerobic biomass compared to aerobic microorganisms has historically forced the application of aerobic technologies for MWW treatment, only allowing the use of anaerobic systems to valorize the primary and biological sludge generated in the treatment mainline. Nevertheless, different technological solutions have been proposed to boost the potential of anaerobic technology for resource recovery, transforming the former WWTPs into new water resource recovery facilities (WRRF) where not only reclaimed water but also different valuable resources can be obtained from MWW, such as bio-products and bio-energy [1.9; 1.13].

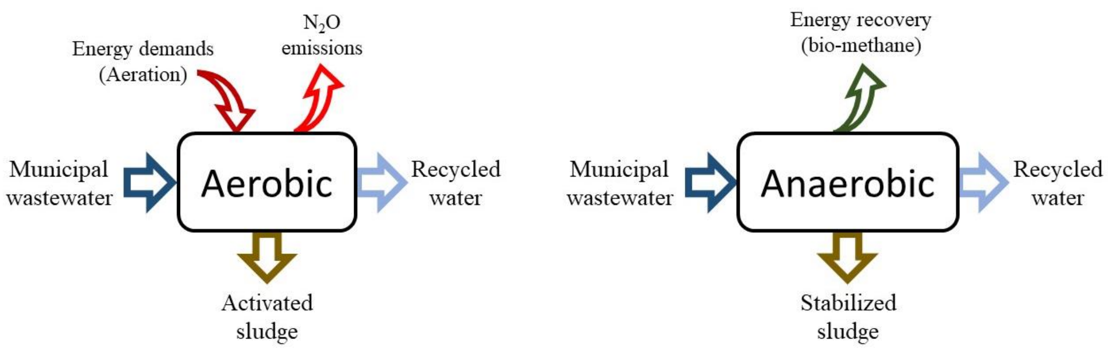

Figure 1.2. Aerobic and anaerobic system features.

1.2 Direct filtration of municipal wastewater

The direct filtration of MWW by using membranes – commonly known within the scientific community as direct membrane filtration (DMF) – has recently emerged as an interesting alternative to enhance current MWW treatment [1.14]. DMF consists in the installation of a membrane filtration system as a previous step to the secondary treatment (see Fig. 1.1b), allowing the capture of all the MWW particulate fraction before the conventional AS process. Thanks to this separation process, only soluble compounds would be fed to a secondary treatment step, thus dramatically reducing energy demands in the case of an aerobic biological system. Indeed, when applying DMF technology, the biological permeate treatment could even be unnecessary [1.15], being able to focus the secondary treatment on other nutrients recovery systems. On the other hand, the sludge retained in the membrane module during DMF could be valorized via anaerobic digestion (AD) jointly with other WWTP produced sludge, increasing the energy recovery of MWW treatment. In addition, the application of this alternative to existing WWTPs would only require the acquisition of compact membrane modules, minimizing the space requirements for its implementation. Therefore, DMF technology represents an interesting alternative for up-grading the currently-in-operation WWTPs without representing important structural or operational modifications.

1.3 Membrane filtration basics

Membrane filtration is a basic operation consisting in separate some components from a fluid solution. A membrane can therefore be considered as a physical barrier whose main aim is to retain some specific components and let pass through the rest of the treating solution [1.16]. Membranes are fed with an influent solution by a side, recovering the filtered solution or permeate in the other side, while concentrating the influent solution in the feeding side obtaining a retentate or waste (see Fig. 1.3). To force the fluid to overcome this physical resistance and perform the separation, two main driving forces can be applied: pressure drop and concentration gradients. Pressure driving filtration consists on pressurizing the feed side or performing vacuum in the permeate side for generating a fluid flux. A classic example of this driving force application is the desalination of sea water by reverse osmosis. On the other hand, concentration driving filtration consists in creating a solute concentration gradient between two fluid solutions separated by a selective permeable membrane. As result, a fluid flux will occur from the lowest to the highest solute concentration solution [1.17]. The high solute solution employed in this configuration is commonly called draw solution (DS). An example of this driving force is the water exchange through the cellular wall between microorganisms and the medium by forward osmosis.

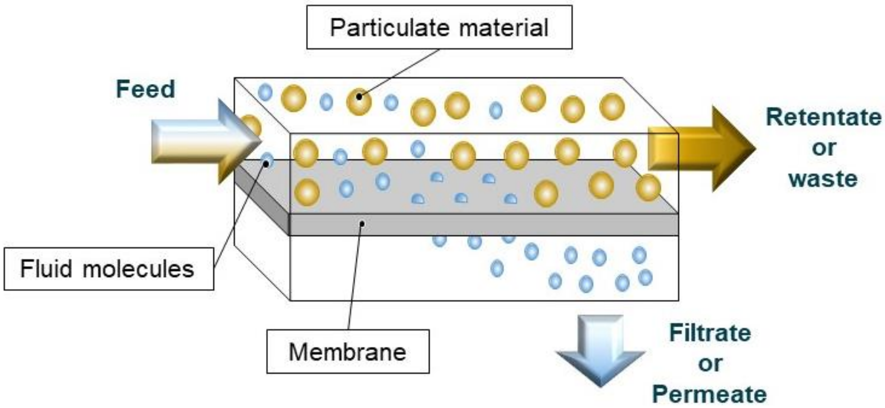

Figure 1.3. Filtration scheme.

The permeate flux (J) obtained during filtration is directly proportional to the driving force applied (ΔD) and inversely proportional to the filtering resistance (R). However, the filtering capacity can also be represented as membrane permeability (K) being inverse each other (see Eq. 1.1).

$$J = \frac{\Delta D}{R} = \Delta D\ K \qquad 1.1$$

As above explained, the driving force can be a pressure gradient, commonly referred as transmembrane pressure (TMP) or a concentration gradient ($\Delta\pi$). The filtering resistance, on the other hand, may cover different terms depending on the process. An initial resistance will be always present during filtration, which corresponds to the intrinsic resistance of the membrane used. This resistance will depend on membrane material, thickness, average pore size, etc. Moreover, additional resistances to filtration appear as the filtration process advances as a consequence of the deposition of different influent substances onto the membrane surface, what represent the so-called phenomenon of membrane fouling.

Fouling is one of most challenging issues in any membrane-based system [1.18], which may limit the applicability of membrane technology for treating certain influents. The higher the membrane fouling (*i.e.* the higher the total filtering resistance), the higher the required driving force for a constant permeate flux (see Eq. 1.1). Thus, membrane fouling directly affects the energy balance during filtration.

Since filtration resistance is a dynamic term that will increase according to membrane fouling propensity, permeate flux and/or applied driving force need to be modified accordingly as filtration progresses. In consequence, filtration is usually performed by setting one variable to a constant/controlled value (*i.e.* permeate flux or operating driving force) and monitoring the other to achieve stable processes. Permeate flux is commonly measured in liters per membrane area and hour (L m^{-2} h^{-1}), which its typical acronym is LMH. Moreover, to control membrane fouling development during filtration, membranes are usually operated by cycles, performing filtration during a period of time and subsequently ceasing permeate production from some seconds to minutes before starting filtration again. This step is typically defined as membrane relaxation. Other additional steps can also be included during membranes operation depending on the process for improving filtration performance such as backwashing [1.19].

1.4 Application of DMF technology to MWW treatment

1.4.1 Pre-treatment

Although in the DMF scheme a classic pre-treatment is included when treating raw MWW (*i.e.* screening and sieving, desanding and degreasing), several authors have recommended the use of a low size solids screening before the membrane tanks in order to achieve better filtration performances [1.20; 1.21]. Thanks to the solids screening, higher foulants can be removed from the treated wastewater, decreasing the fouling formation propensity [1.20] and reducing the possibility of membrane damage. Also, higher flux stabilities have been reported when low size solids screening is included before the membrane filtration due to the reduction in the matter size range [1.22]. Strainer or micro-sieving ranging from 500 to 100 µm are usually proposed [1.22–1.25]. Alternatively, a two-stage high size screening (1190 and 600 mm) [1.26] or sand-filters [1.27] have been also recommended.

1.4.2 System configuration

Emulating membrane bioreactors (MBRs), two main configurations have been defined for DMF technology based on the integration between the membrane and treated influent: side-stream and submerged systems (see Fig. 1.4). In side-stream configuration, the membrane and concentration tanks are placed separately, continuously recycling the treated sludge into the membrane tank at relatively high flow rates and pressures (see Fig.

1.4a). The main advantage of this configuration is the possibility of applying high cross-flow velocities on the membrane, thereby providing a membrane scouring by tangential liquid forces during filtration [1.28]. However, due to the high pumping demands, important energy requirements are expected when using this configuration [1.29]. Side-stream configuration is generally employed when treating high-strength sewage [1.29], having been evaluated in different studies (see for instance [1.30–1.32]). In the submerged configuration, the membrane is directly immersed in the concentration tank (see Fig. 1.4b), avoiding continuous pumping recycling requirements thereby involving lower energy demands [1.33]. However, additional membrane cleaning strategies are required for fouling control, which usually represents the main energy input of the system (*e.g.* air-assisted membrane scouring). In conventional MBRs, the membrane can be immersed directly in the bioreactor or placed in an auxiliary membrane tank focused on sludge filtration [1.29]. However, since negligible biological activity is expected in DMF technology, reactor and membrane tank can be integrated, thus simplifying the configuration scheme. Submerged DMF configuration is generally employed when treating low-strength wastewater (such as MWW), being the predominant configuration when applying DMF technology to wastewater treatment (see for instance [1.34–1.36]).

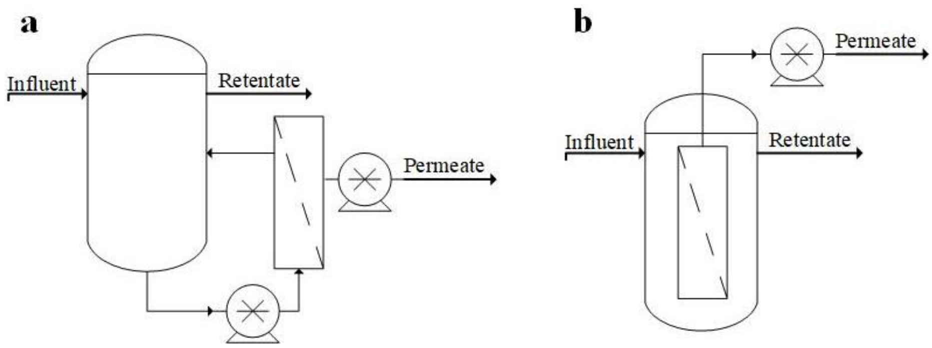

Figure 1.4. DMF main operating configurations.

1.4.3 Commercial membrane modules

Four major membrane configurations are available in the market: plate-and-frame, spiral wound, tubular, and hollow fiber [1.37]. Fig. 1.5 illustrates an example for each membrane configuration.

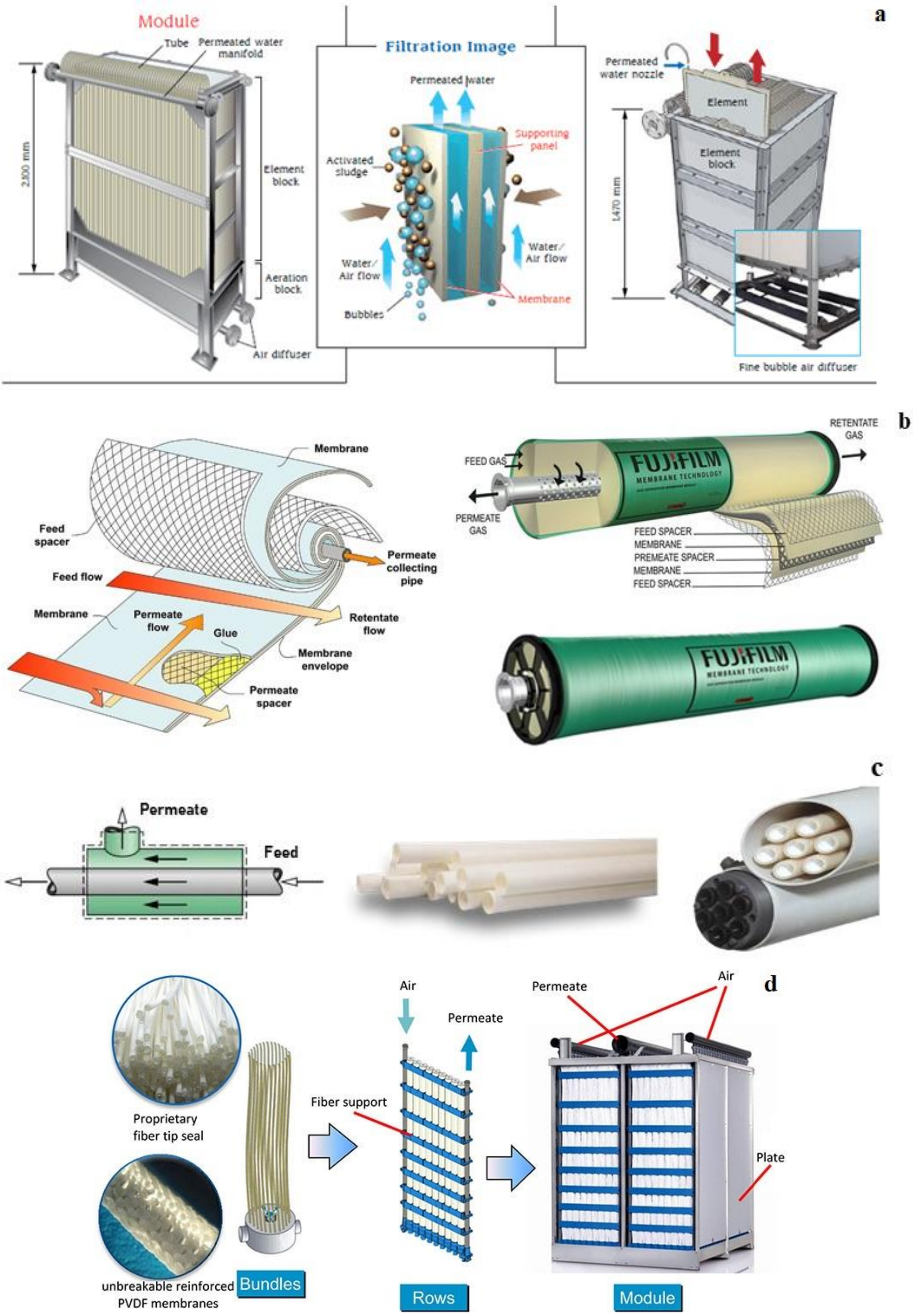

Figure 1.5. Examples of commercial membrane configurations: (a) submerged plate-and-frame module (source: Toray Industries, INC), (b) cross-flow spiral wound module (source: Fujifilm membrane technology), (c) cross-flow tubular module (source: PCI membranes), and (d) submerged hollow fiber module (source: Koch Membrane Systems).

Plate-and-frame configuration consists in a plain square attached to an exterior frame. Individual membranes can be arranged in vertical or horizontal stacks to conform modules. In general, not high operating pressures can be applied with this configuration, while the surface area to volume ratio is not very high compared to other module configurations [1.38]. Consequently, this configuration is not very employed at industrial scale [1.37].

Spiral wound membranes are built by sealing on three sides two large membrane sheets, forming a bag. The formed bag is winded around a collector tube, while a flexible porous material is inserted between each two membrane layers for alternatively feeding the module or collecting the permeate in the central tube (see Fig. 1.5b). Modules can be feed axially or radially and can provide a high surface area per volume ratio [1.38].

Tubular configuration resembles a classic double pipe heat exchanger. Treating solution is feed though the inner tube, collecting permeate in the annular space or vice versa. However, to increase packaging, membrane tubes can be arranged in parallel for forming bundles and placed inside a shell for assemble a module (see Fig. 1.5c). Tubes diameter generally ranges between 10 – 25 mm, which entails easier cleaning and inspection. Consequently, this membrane configuration is particularly suitable for treating solutions with an elevated solids charge or operating at high retentate concentrations [1.38]. Nevertheless, not high packing ratios (membrane area per volume) can be achieved with this configuration [1.38].

Hollow fiber (HF) configuration consist in the union of several tubular fibers connected to a collector plate for building bundles. The fibers diameter is extremely reduced in this case (from 1 mm down to capillary sizes) which allows to assemble numerous fiber (or lumens) per membrane module thereby achieving high packing ratios (see Fig. 1.5d). In consequence to this high packing density, this configuration has been extensively used for numerous applications. However, due to its high compactness, it shows significant drawbacks when treating solutions with elevate viscosities, being more prone to clogging issues [1.38]. Permeate is usually collected inside the fibers to reduce clogging issues and facilitate membranes cleaning.

Emulating MBR technology, HF is the most extended configuration in DMF of MWW (see for instance [1.36; 1.39; 1.40]). Nevertheless, plate-and-frame configuration is typically employed when using dynamic membranes, which are gaining an increasing interest for developing this alternative.

1.5 Membrane technology in DMF

Different membrane technologies have been tested for DMF of MWW: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and forward/reverse osmosis (FO/RO) membranes. The lower the membrane pore size, the higher the amount of substances retained (see Fig. 1.6), increasing both filtration resistance and required driving force. Then, based on this increasing filtration demand, MF and UF membranes have been usually used for primary and secondary sewage treatment while NF and RO membranes have been more focused on tertiary treatments. Nonetheless, FO membranes have been shown as a technically feasible alternative for treating primary MWW effluents [1.41], which could be used for producing a high quality clean water, whilst secondary and tertiary effluents can be filtered for water production [1.42]. Alternatively to classic membrane technology, dynamic membranes (DM) have attracted great interest for performing DMF of MWW, which accumulate so far promising results thanks to the low prices of the available supporting materials and relatively easy fouling control. Table 1.1 summarizes the different operating conditions (membrane technology, influent, and fouling control methodology) under which several authors have studied the DMF of MWW.

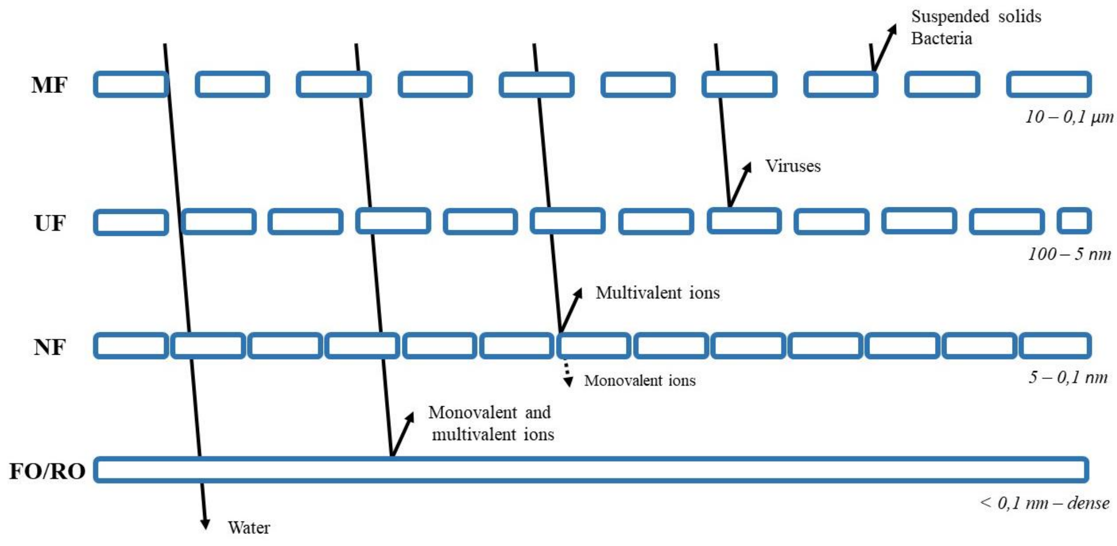

Figure 1.6. Membrane technology pore size and pollutants retention.

Table 1.1. Application of DMF technology for the treatment of MWW in the middle-/long-term.

Membrane type	Treated influent	Fouling control strategy applied	Operation	Operating time	Ref.
Microfiltration	Raw MWW	Coagulant dosing combined with permeate backwashing every filtration-relaxation cycle. Air scouring applied during backwashing and relaxation periods.	Average operating flux of 41.7 LMH. Off-line chemical cleaning every 3 – 5 days once reached a TMP of 35 kPa.	700 hours	[1.14]
		Coagulant dosing combined with micro-sieving and membrane air scouring.	Constant TMP of 3 kPa achieving a permeate flux of 6.1 LMH.	159 hours	[1.22]
		Mechanical agitation.	Filtration of low-strength sewage. The filtration resistance raised from $8{\cdot}10^9$ to $4{\cdot}10^{10}$ m^{-1} for an operating flux of 20 LMH.	120 days	[1.23]
		Continuous coagulant dosing combined with air backwashing.	Average operating flux of 10 – 15 LMH achieving a pseudo-steady operation with average TMPs of 20 – 40 kPa.	1600 hours	[1.39]
		Air-assisted membrane scouring combined with intermittent CEBs every 12 hours.	Concentration of the influent COD in two different tanks: Tank 1: concentration factor of 21 at an operating flux of 20.8 LMH. Tank 2: concentration factor from 21 to 50 at an operating flux of 16.7 LMH.	200 hours	[1.43]
		Coagulant dosing combined with air backflushing.	Operating flux of 20 LMH.	93 hours	[1.44]
		Coagulant + adsorbent (activated carbon) dosing.	Operating flux of 5 LMH.	144 hours	[1.45]
		Continuous coagulant dosing combined with intermittent air scouring.	Operating flux of 13.3 LMH.	295 hours	[1.46]
		Backwashing with ozonized water.	Operating flux about 8 LMH.	180 days	[1.47]
		Membrane vibrations combined with air scouring.	Operating flux of 12.8 LMH.	20 hours	[1.48]
	Primary settler effluent	Intermittent membrane vibrations combined with mechanical agitation and periodical CEBs.	TMPs under 10 kPa for an operating flux of 6.5 – 4.2 LMH.	600 hours	[1.40]

Table 1.1. Cont.

Membrane type	Treated influent	Fouling control strategy applied	Operation	Operating time	Ref.
Ultrafiltration	Raw MWW	Cross-flow operation and demineralized water backwashing.	Operating at a constant TMP of 0.3 bar achieved an average flux of 120 LMH.	120 min	[1.25]
Ultrafiltration	Raw MWW	Cross-flow operation.	For an operating TMP of 1.8 bar the flux decreased from 262 to 143 during continuous filtration.	7 hours	[1.27]
Ultrafiltration	Raw MWW	Cross-flow operation.	TMPs about 0.2 – 0.4 bar achieved for an average operating flux of 300 LMH.	7 hours	[1.49]
Ultrafiltration	Primary settler effluent	Cross-flow operation and demineralized water backwashing.	Operating at a constant TMP of 0.3 bar achieved an average flux of 160 LMH.	100 min	[1.25]
Ultrafiltration	Primary settler effluent	Coagulant + adsorbent (activated carbon) dosing combined with cross-flow operation.	Operating flux of 59.8 LMH.	180 min	[1.30]
Ultrafiltration	Primary settler effluent	Air-assisted membrane scouring.	Operating flux of 20 LMH.	72 hours	[1.34]
Ultrafiltration	Primary settler effluent	Coagulant + flocculent dosing combined with permeate backwashing.	Operating flux of 23 LMH.	Few hours	[1.35]
Ultrafiltration	Primary settler effluent	Membrane air scouring and permeate backwashing.	Operating flux of 10 LMH.	54 days	[1.36]
Forward osmosis	Raw MWW	Cross-flow operation.	Asymmetric CTA membrane. DS: seawater (osmotic pressure of 26.45 bar).	70 days	[1.50]
Forward osmosis	Raw MWW	Cross-flow operation (0.28 m s^{-1}).	CTA membrane. DS: NaCl solution (35 g L^{-1}). Continuous DS regeneration by membrane distillation. Stable flux of 17.6 LMH during filtration.	120 hours	[1.51]
Forward osmosis	Raw MWW	Cross-flow operation (from 0.29 to 0.48 cm s^{-1}).	TFC membrane. DS: $MgCl_2 \cdot 6H_2O$ (osmotic pressure of 150 bar). Average flux of 5.3 LMH (53% water recovery). Membrane cleaning required every 24 hours.	24 hours	[1.52]
Forward osmosis	Raw MWW	-	Pilot-scale spiral wound membrane. DS: NaCl (0.5 M). Average flux of 6 LMH.	450 hours	[1.53]
Forward osmosis	Raw MWW	Cross-flow operation (from 0.5 to 3 L min^{-1}).	CTA membrane. DS: NaCl (from 0.2 to 4 M).	1500 min	[1.54]
Forward osmosis	Raw MWW	Cross-flow operation (10 cm s^{-1}).	CTA membrane. DS: synthetic seawater (osmotic pressure of 26.45 bar).	30 days	[1.55]
Forward osmosis	Raw MWW	Cross-flow operation.	TFC membrane. DS: synthetic seawater.	24 – 46 hours	[1.56]

Table 1.1. Cont.

Membrane type	Treated influent	Fouling control strategy applied	Operation	Operating time	Ref.
Dynamic membrane	Raw MWW	External physical cleaning with air backwashing and surface brushing each 48 h.	Three-layer stainless steel mesh of 25 µm as supportive material. Continuous filtration without relaxation at a flux between 33 – 54 LMH for 48 h.	200 hours	[1.57]
		Continuous coagulant dosing. External physical cleaning with tap water after reaching a TMP of 35 kPa.	Dacron mesh of 61 µm as supportive material. Operation flux of 60 LMH. External physical cleaning required every 25 – 50 days.	300 days	[1.58]

CTA: cellulose triacetate.
TFC: thin film composite.
DS: draw solution.

1.5.1 Micro- and ultra-filtration membranes

MF and UF membranes are classified as porous membranes with a pore size range around 10 – 0.1 µm and 100 – 1 nm, respectively [1.59]. Given their average pore sizes, these membranes are able to retain suspended solids and bacteria during filtration, also being able to reject viruses in the case of UF membranes (see Fig. 1.6). Pressure gradient is generally used as driving force to perform filtration while the concentration gradient usually has negligible effects. Lower filtration resistances compared to other more restrictive membranes (*i.e.* NF and RO) are expected in MF and UF technology, which significantly reduce their filtration energy demands. Accordingly, their use for wastewater treatment (municipal, industrial or any kind of biological sludge) has been extended intensely in the last two decades, finding in bibliography numerous membrane-based configurations focused on improving classic wastewater treatment systems (*e.g.* aerobic MBR (AeMBR), anaerobic MBR (AnMBR), etc.).

1.5.1.1 Filtration performance

MF and UF membranes are mostly applied to DMF of MWW [1.20]. Unfortunately, severe fouling intensities have been noticed when filtering untreated MWW [1.40; 1.44–1.46; 1.60], which considerably overcomes the fouling propensities reported in other MBR systems [1.36; 1.46]. For example, Jin *et al.* [1.46] showed sharply membrane permeability decreases (from 1000 to 20 LMH bar^{-1}) in the first 5 h of operation when treating raw MWW by MF membranes, further decreasing until about 10 LMH bar^{-1} after 20 h of operation. Similarly, Jin *et al.* [1.44] reported a quick decline in membrane permeability (from 2000 to 20 LMH bar^{-1}) in the first 4 hours of operation, reaching values lower than 10 LMH bar^{-1} in less than 7 hours, while Kimura *et al.* [1.40] showed a drastic increase in the operating TMP (about 45 kPa) within only 5 min of operation. Additionally, Gong *et al.* [1.45] studied the evolution in the membrane resistance of a MF membrane when treating raw MWW, noticing a similar multi-stage performance than those reported when operating other MBRs but with quicker and higher resistance increasing stages. Thus, major issues to attend in order to boost the applicability of DMF technology to MWW treatment are the study of the source and main mechanisms of fouling and the development of efficient fouling control strategies.

1.5.1.2 Membrane materials

Different membrane materials have been used within DMF technology for MWW treatment, being polyvinylidene fluoride (PVDF) the most common material used (see for instance [1.20; 1.40; 1.44; 1.46]). However, other membrane materials such as polypropylene (PP) [1.22], polyethylene (PE) [1.23; 1.48], polyamide (PA) [1.32], polyethersulfone (PS) [1.34], and ceramics [1.30; 1.31; 1.60] have been also employed within this field. Despite the numerous membrane materials used, few studies are available evaluating its effects on filtration performance, being mainly focused on MF technology. For instance, Mezohegyi *et al.* [1.48] reported similar fouling propensities in the short-term (20 h of operation) when using PE and PVDF membranes, whilst Fujioka *et al.* [1.60] observed similar abrupt TMP increases (from 49 to 143 kPa in the first 40 min of filtration) when using ceramic membranes. Therefore, despite the little research performed so far in this field, it could be concluded that the membrane material used is not a key variable for DMF of MWW, expecting therefore similar filtration performances regardless the material used.

1.5.1.3 Membrane pore size

The selection of an adequate membrane pore size is a crucial issue in any membrane-based process. Different fouling mechanisms and propensities are triggered depending on the interaction between the applied membrane pore size and treated particles suspension [1.61]. Thus, the effective pore size may control membrane fouling during filtration, being a key aspect to take into account for achieving feasible processes. Unfortunately, as far as the authors know, a properly comparison between MF and UF performance filtering untreated MWW has not been performed yet, while results from different studies cannot be directly compared due to the extremely different flux, fouling control strategy, and influent source used in each case (see Table 1.1).

MF membranes may present better performances due to their high pore size which should reduce membrane filtration resistance. Nevertheless, due to the large particles size distribution (PSD) range and sticky substances expected in untreated MWW, high pore sizes might enable the deposition of particles of different nature into the pores, favoring pore narrowing which would finally result in a subsequent pore blocking. On the other hand, UF membranes may require higher energy inputs to perform filtration due to their inherent higher filtration resistances consequence of their lower pore sizes. However, a reduction of the effective pore size may result in better filtration performances thanks to reducing the number of particles that can interact with the membrane pores. In this respect, Kramer *et al.* [1.62] operated a UF ceramic membrane (3 kDa of pore size) for the treatment of raw MWW during 22 h, observing a reduction in the membrane permeability of about 16%, which is clearly lower than that reported in numerous studies using MF membranes. Nonetheless, it is important to consider that reducing membrane pore size not just increases the inherent resistance of membranes to conduct filtration but also favors the development of thicker cake layers due to the accumulation of more type of particles and substances on the membrane surface. Indeed, when the cited authors [1.62] compared the filtration performance of raw MWW by using UF and NF membranes (3000 and 450 Da of pore size, respectively), a higher permeability decrease was reported when using the NF membrane (from 5.9 to 2.5 LMH bar^{-1} and from 5.8 to 4.8 LMH bar^{-1} for the NF and UF membranes in the first 22 – 24 h, respectively), showing that reducing the membrane pore size not always reduces membrane fouling propensity. A similar experience was reported by Ahn *et al.* [1.63], whom observed a significant resistance increase when a lower pore size UF membrane was used (pore size used from 300 to 15 kDa) for treating hotel building wastewater in the short-term.

As these studies illustrate, the most suitable membrane pore size to employ is strongly related with the characteristics of the treated influent and, therefore, further studies are required for properly select the most adequate membrane in each case.

1.5.1.4 Treated influent

Similar to membrane pore size, influent MWW may significantly impact filtration performance in DMF of MWW. Indeed, the PSD of treated influent have showed a strong influence onto the membrane fouling in different membrane systems [1.30; 1.64]. As some authors suggest, the large PSD displayed in raw MWW may promote the formation of quick and thick cake layers on the membrane surface [1.61]. In addition to this large PSD, the irregular shape and diverse material of the particles present in MWW could favor the development of simultaneous fouling mechanisms during filtration, increasing therefore the filtration resistance and reducing the effectiveness of the on-line cleaning strategies.

To reduce fouling impacts during DMF, the treatment of more depurated influents has been recommended [1.25; 1.40]. Among possible MWW pre-treatments, some authors have proposed the use of coagulants and micro-sieving for reducing the amount of particulate material send to filtration membrane modules [1.22; 1.65]. However, the use of a conventional primary settler is the most used option [1.25; 1.40]. Primary settling allows recovering and concentrating an important fraction of the influent total suspended solids (TSS) by sedimentation. Thus, only the smaller particles would be feed to the DMF modules. Additionally, since primary settling is a fundamental part of almost all currently in use WWTP, the use of the primary settler effluent (PSE, *i.e.* supernatant from primary settling) for feeding the DMF unit would not require of any additional WWTP up-grading, thereby representing an attractive alternative. Unfortunately, the impact of using PSE during DMF of MWW has not been clearly reported yet.

Kimura *et al.* [1.40] observed a quick TMP increase (up to 50 kPa in less than 5 h) when treating PSE MWW by a MF HF membrane. Since similar results have been reported when treating raw MWW by this type of membranes, similar filtration performances could be anticipated regardless the influent used. In fact, Ravazzini *et al.* [1.25; 1.61] concluded that during a continuous filtration process, similar fouling developments and fouling mechanisms can be expected regardless the treated influent (i.e. raw or PSE). On the other hand, the cited authors [1.25; 1.61] also reported that higher permeate production rates can be obtained during short-term filtrations when treating PSE (around 20 – 70% higher compared with raw MWW) depending on operating conditions. Furthermore, these authors also reported that a more intense improvement in membrane permeability could be achieved by physical cleaning when treating PSE, especially when high TMPs and low cross-flow velocities are applied. These results were attributed to both the reduction in the amount of particulate material fed to the filtration modules and a decrease in its average particle size when treating PSE compared to raw MWW, promoting the formation of thinner but denser cake layers in the former than in the latter [1.61].

Therefore, although the severe fouling development detected during DMF of MWW could not be directly mitigated by the implementation of a primary settler as influent pre-treatment, this alternative could be an interesting option to improve the effects of the fouling control strategies implemented during filtration, thereby enhancing DMF feasibility.

1.5.1.5 Operating conditions

Operating conditions when treating MWW seem to play a more important role on filtration performance than changes on feed characteristics [1.61]. Therefore, the selection of a suitable permeate flux, TMP or filtration/relaxation ratio is essential to achieve energy-efficiency and cost-effective processes. In this respect, Ravazzini [1.61] and Ahn *et al.* [1.63] studied the effect of applying different TMPs during raw MWW filtration using UF membranes. These authors reported that raising TMP may increase permeate productivity only in the first moments of operation, reaching later similar low fluxes than those obtained when operating at lower TMPs. Nonetheless, operating at high TMPs may increase the fouling development in the membrane probably by a growth in the material transfer forward the membrane surface [1.61]. Additionally, since cake layers formed in different MF and UF MBRs are often found to be compressible [1.61], high TMPs could favor the formation of denser and more compact cake layers during DMF of MWW thus raising the filtration resistance. In fact, Ravazzini *et al.* [1.25] and Fujioka and Nghiem [1.60] suggested the development of compressible cake layers during DMF and Jin *et al.* [1.46] reported that relatively dense cake layers could be formed in these systems. Moreover, Ravazzini [1.61] showed that compressible cake layers can be expected during the DMF of MWW by means of a mathematical approach, concluding that the TMP is the main factor affecting filtration resistance. Consequently, the use of middle to low TMPs during filtration (about 0.3 – 0.5 bar) have been recommended to minimize the substantial fouling developed in DMF technology [1.25; 1.61]. Additionally, these authors [1.25; 1.61] also recommended operating at short filtrations times (only a few minutes long) to reduce backwashing frequency and achieve higher net permeate productions.

Regarding permeate flux, the most convenient values have not been clarified. Several authors operating different MBRs have reported a negative effect in the filtration performance when using high initial permeate fluxes. Madaeni [1.66] reported those hindering effects when treating inorganic colloids solutions, Ho and Zydney [1.67] with proteins solutions, and Ahn *et al.* [1.63] when filtering MWW. However, high permeate fluxes may overcome the electrostatic rejection between the treated mixed liquor and surface membrane [1.61] and accelerate the transition from an initial pore blocking fouling forward a cake layer filtration [1.68], thereby preventing the development of irreversible fouling. In any case, when Nascimento *et al.* [1.36] studied the effects of starting-up with high fluxes or applying a step-flux-increase method for filtering PSE with UF membranes, similar fouling growth rates were obtained regardless the operating flux. Thus, these authors concluded that both strategies are a valid approach when filtering MWW.

1.5.2 Osmosis membranes

Osmosis membranes are built on semipermeable materials being able to retain from colloidal material to even different monovalent ions depending on their molecular size [1.59]. Polymeric materials dominate the market due to their excellent performance and low cost [1.69], mainly finding two commercial alternatives: thin-film composite (TFC) and cellulose base (CA) membranes. TFC membranes are clearly predominant due to their higher permeability, lifespan, pH stability and resistance to hydrolysis, and biological degradation [1.70]. However, CA membranes, especially cellulose triacetate (CTA) membranes, are still available due to their exceptional chorine resistance [1.69]. Since membrane cleaning and disinfection is usually conducted by using NaOCl, these membranes can show higher lifespans depending on the treated solution.

Osmosis membranes can be operated by pressure or concentration gradients, classically differentiating between RO and FO processes. Concerning their applicability to DMF, FO membranes have been proposed for directly treating MWW [1.71] since these membranes generally entail lower fouling propensities that other pressure-driven membranes [1.53]. Instead, RO membranes require of an additional pre-treatment for filtering MWW since their higher sensitivity to membrane fouling [1.53] is translated into elevated energy costs due to the high operation pressures required. Hence, RO membranes are not usually applied to DMF of MWW. However, the application of these membranes can be especially interesting for treating the effluent produced from DMF since it could be used for boost nutrients recovery and enhance the quality of the produced permeate.

The viability of using FO membranes for DMF of MWW has been evaluated by several authors. In lab-scale systems, Zhang *et al.* [1.72] were able to multiple by 6 the influent MWW concentration when employing a lab-scale CTA cross-flow membrane module.

Similarly, Ansari *et al.* [1.73] demonstrated the feasibility of using a CTA membrane for concentrating raw MWW (concentration factor of 10), reaching COD concentrations in the membrane retentate high enough for directly feeding an AD. Moving to pilot-scale studies, Wang *et al.* [1.53] reported a stable long-term filtration (average flux of 6 LMH during 51 days) when treating MWW with a CTA spiral-wound membrane. In this last study a concentration factor of 5 was achieved [1.53].

Unfortunately, membrane fouling is also a relevant issue when filtering MWW with FO membranes. In this respect, Ansari *et al.* [1.73] reported a 50% flux decline after 70 h of filtration due to membrane fouling, being their development especially important after the first 40 h of filtration. Similarly, Gao *et al.* [1.54] reported significant membrane permeability reductions (about the 20 – 40%) after filtering MWW for 1500 min, whilst Ferrari [1.70] reported that the flux decrease observed during the filtration of MWW (39.9 hours) was associated to both fouling development propensity and concentration polarization. Other limitation that need to be taken into account when concentrating MWW with FO membranes is the accumulation of toxic substances. NH_3 and different salts can be easily retained and concentrated by FO membranes, which are known as strong AD inhibitors when achieving high enough concentrations in the feed [1.70]. Additionally, other classical or emerging contaminants (SO_4^{2-}, heavy metals, pharmaceutical products, hormones, microplastics, etc.) are also susceptible to be retained to a high extent by FO membranes [1.74], which may hinder the AD process and cause negative effects on the methane production. Thus, depending on the concentration of these substances in the influent MWW, moderate concentration factors (around 1 – 10) may be recommended to avoid possible problems with the posterior AD system [1.70].

Apart from limiting the operating FO retentate concentration, the employed DS is also a relevant matter to consider. Different artificial saline solutions can be efficiently employed to conduct filtration, such as NaCl, KCl, or $MgSO_4$, among other [1.75]. However, the use of seawater emerges as an attractive alternative in the DMF concept. The use of seawater as DS do not just would eliminate chemicals demand and DS regeneration requirements, but the final salt-diluted solution obtained after the process could be employed for potable water production by RO [1.76]. This combination has already demonstrated several advantages like lower RO fouling tendencies, achieving significant energy and economic benefits compared to classic RO desalination [1.76].

Unfortunately, due to the high flux of salts that may travel from the DS towards the concentrated MWW during filtration, an elevated salt content may reach the AD, inhibiting the process [1.54; 1.73]. Consequently, the use of organic DSs, such as sodium acetate, or EDTA-2Na, has been recommended by some authors when treating MWW [1.73]. Furthermore, the use of moderate DS concentrations when directly filtering MWW has also being recommended [1.54]. This is due to the higher and quicker fouling formations noticed when operating at relatively elevated permeate fluxes (20 – 25 LMH).

1.5.3 Dynamic membranes

In addition to conventional membrane technology, DMs have attracted great interest for DMF of MWW. DMs consist of two different layers: (1) a low filtration-resistance supporting material, and (2) a developed cake layer. The supporting material acts as a base onto which the particles present in the treated influent can add while the increasing particles deposition form aggregates until achieving a (stable) cake layer which will act as the main filtration actor [1.77]. Thanks to removing the intrinsically filtration resistance that conventional membranes represent, a higher permeability can be achieved during filtration, being able to control the filtration performance by acting onto the thickness and density of the formed cake layer [1.57]. Thus, membrane fouling paradigm changes in DM technology, playing in this case a partially beneficial role during filtration, which can usually be easily controlled by low energy-demanding physical cleaning methodologies [1.57]. In addition, the employed supporting structures are generally low-cost materials, such as woven meshes or filter-cloths, thereby representing a significant lower acquisition and/or replacement cost than conventional membrane modules [1.78].

Despite all the potential benefits that DMs may bring, influent characteristics (*e.g.* particles size, pH, temperature, etc.) and operating conditions (*e.g.* supporting material, flux, filtration time, cleaning, etc.) have a strong influence on filtration resistance and permeate quality [1.79; 1.80], hindering their applicability in some cases. Indeed, significantly worse permeate qualities can be expected when using DMs instead of other membrane technologies since the formed cake layer would present a less homogeneous structure with a higher porosity than commercial membranes. In addition, since the formed cake layer mainly controls the filtration performance, the generated permeate in DM filtration may be unstable to some degree, changing regarding the characteristics of the formed DM during filtration. On the other hand, the formation of a stable DM can also represent a difficulty in some cases since its formation strongly depends on the characteristics and concentration of the influent suspended material and its interaction with the employed supporting structure. Thus, the selection of a suitable supporting material regarding the treated influent is a key matter to achieve proper DM performances.

1.5.3.1 Dynamic cake layer formation

Two different DMs can be differenced depending on how the filtering cake layer is developed onto the supporting structure, namely self-forming and pre-coated [1.77; 1.81]. Self-forming DMs are formed when just developing the filtering cake layer from the direct deposition of the particulate material onto the supporting structure during filtration. Instead, pre-coated DMs consist on developing a previous stable structure onto which the influent particulate material can be deposited for forming the filtering cake layer. A pre-treatment with external solutions containing additional particulate material (*e.g.* powdered carbon or kaolite) can be used for pre-coating the employed supporting structure before starting the real influent filtration. Pre-coating DMs are *a priori* less advantageous since they require an auxiliary chemical dosing which increases its operating cost [1.81]. However, due to the high PSD and lower sticky substances presents in MWW compared with other membrane-based bio-reactors, DM formation improving strategies may be required to properly apply this technology in the DMF alternative.

Instead of previously pre-coating the supporting material, chemicals addition during filtration has been also a commonly proposed solution [1.82]. Thanks to increasing the average particles size by flocculation, quicker and more stable DM developments can be achieved, allowing consequently the use of higher supporting material pore sizes. Indeed, Ma *et al.* [1.58] reported stable permeate quality in 1 – 2 days of operation when dosing polyferric sulfate (PFS) in a 61 μm Dacron mesh, while Gong *et al.* [1.80] achieved a stable DM in just 2 hours when dosing diatomite in a 100 μm propane polymer mesh, both treating raw MWW. Coagulant dosing could be used during the first DM development step (as some kind of pre-coating), favoring just the DM formation, or during all the operating period, controlling also membrane fouling development and permeate quality.

Other alternative to enhance DM development and permeate quality is to modify supporting material sewing or thickness [1.83]. Xiong *et al.* [1.57] studied the effect of using multi-layer supporting materials for raw MWW treatment, reporting significant increments on the OM recovery and operating TMP when increasing the added layers. Despite the membrane resistance raise, the authors selected the three-layer structure due to the significant OM recovery improvement and rapid DM layer formation. This conclusion would be in disagreement with results reported in AnMBRs operated with DMs (AnDMBR), which usually recommend the use of simple mono-filament mesh supporting materials due to their lower filtration resistances [1.79; 1.83].

1.5.3.2 Supporting materials and pore size

Metallic, ceramic, and polymeric materials are supporting layers commonly used in AnDMBRs [1.79; 1.82]. When treating raw MWW, Xiong *et al.* [1.57] showed that nylon and stainless steel meshes can be used to quickly develop a dynamic membrane, observing at same pore size, better performances by the stainless steel mesh when considering OM recovery and filtration resistance. Dacron and propane polymer meshes have been also showed as suitable supporting materials when treating raw MWW [1.58; 1.80].

Regarding the supporting material pore size, a range between 1 and 25 µm has been used for raw MWW treatment [1.57; 1.80], achieving stable DM formations between 10 – 24 hours [1.57]. In this respect, Xiong *et al.* [1.57] identified 25 µm as the more suitable pore size when considering permeate quality and TMP operating raise, while Gong *et al.* [1.80] suggested 1 µm due to its higher OM recovery efficiency. Additionally, when treating raw MWW, Gong *et al.* [1.80] indicated that when 1 and 10 µm pore sizes where used, cake layer filtration mechanism was developed, identifying a complete blocking for 50 µm and a standard blocking for 100 µm. As these authors suggests, DM filtration mechanism change was probably due to the PSD in the influent (particles size concentrations between 20 and 50 µm). Due to the dramatic membrane resistance raise observed for the 50 and 100 µm pore sizes, small pore sizes may be suggested to avoid membrane pores complete blocking [1.80]. Proposed supporting material pore sizes are similar to those reported by AnDMBR [1.83; 1.84], which may indicate that similar DM formation mechanisms take place despite the different characteristics of the wastewater treated.

1.5.3.3 Operating conditions

Proper filtration performances have been informed during DM operation for raw MWW treatment when fluxes around 60 LHM were applied [1.57; 1.58], which clearly overcomes regular MF and UF fluxes. Also, stable operating TMPs between 20 and 40 kPa have been reported in the middle-term (200 h of operation) [1.57] and long-term (300 days of operation) [1.58]. Regarding fouling development, Gong *et al.* [1.80] informed that during more than 60 h of operation, all developed fouling could be considered reversible, completely recovering initial flux when mechanical cleaning was conducted. However, Xiong *et al.* [1.57] observed a slight descend in the operating flux around the first 100 h of operation despite the physical cleanings performed, which was attributed to a significant irreversible fouling development as noted in other DM systems [1.85; 1.86]. Additionally, Ma *et al.* [1.58] determined that seasonal temperature changes did not affected fouling propensity. Air backwashing and surface brushing has commonly been employed as cleaning methods when operating DMs [1.87]. However, same strategies than those used in MF and UF membranes can be proposed to control fouling development in the long-term [1.57].

1.6 Membrane fouling in MWW treatment

Membrane fouling refers to the adsorption and/or deposition of any material on the membrane (superficially or internally), which interfere during filtration hindering membrane permeability [1.16]. Depending on its nature, fouling can be classified as reversible, irreversible, and irrecoverable fouling. Reversible fouling refers to any type of membrane permeability reduction that can be recovered by applying any kind of physical cleaning method (*i.e.* relaxation, air sparging, backwashing, etc.). Irreversible fouling instead remains regardless the application of physical cleaning, being necessary to apply chemical cleanings to recover the original membrane permeability. This kind of fouling is more associated to internal pores obstructions, hindering the possibility of removing the attached foulants, although it can be also related to the consolidation of biofilms or irreversible fouling layers on the membrane surface. Finally, irrecoverable fouling refers to any type of fouling that cannot be removed either physically or chemically, thus representing the eventual permanent loss of the membrane permeability.

Considering its source, fouling can be classified in particulate/colloidal, inorganic, biological, and organic fouling [1.88]. Particulate or colloidal fouling is caused by the bulk particles carried by the treating solution which can occupy the membrane pores or accumulate onto the membrane surface forming a cake layer. This kind of fouling is extremely usual in numerous membrane-based systems when treating solutions with particulate material, representing one of majors fouling contributors in MBR technology [1.89]. Inorganic fouling is mainly caused by the precipitation of diverse inorganic ions when surpassing their solubility in the membrane retentate [1.90]. Calcium and carbonate based salts such as struvite, $CaCO_3$, $CaSO_4$ and $MgCO_3$ are the most common source of inorganic fouling when filtering MWW. Biological fouling or bio-fouling results from the interaction between the membrane and microbial activity which can attach to the membrane surface and form biofilms or excrete different substances that affect the membrane. Finally, organic fouling is associated to the accumulation and/or adsorption of different organics, such as proteins, carbohydrates, polysaccharides, etc., on the membrane surface. Among most employed classifications for organic fouling, the division between soluble microbial products (SMP) and extracellular polymeric substances (EPS) is extensively used in bibliography, which combines bio-fouling and organic fouling [1.91]. SMP strictly refers to any soluble organic (mainly proteins, carbohydrates and humic substances) produced by microbial activity, whilst EPS similarly refers to any substance (soluble and particulate) resulting from biomass. Nevertheless, in practice, any organic which may hinder filtration can be included in this general classification.

In the case of untreated MWW, a low bio-fouling influence could be expected since any biological activity is involved or desired. However, it contains a large PSD, colloids and dissolved organic and inorganic compounds which can interact each other and/or cause different problems on the membrane during filtration [1.61; 1.92]. Given this diversity of contaminants, pretty complex, diverse, and changing fouling mechanisms can be anticipated when filtering MWW.

1.6.1 Fouling in porous membranes

Fouling in porous membranes can be generally classified into two categories: cake layer and pore blocking [1.60]. Cake layer fouling describes the accumulation of particulate material onto the membrane surface and it is usually associated to reversible fouling. Conversely, pore blocking describes the partial/complete obstruction of the membrane pores by colloidal particles deposition or sticky gel formation, rapidly hindering membrane permeability. This kind of fouling can be reversible or irreversible depending on the difficulty for removing the stuck substances. Additionally, pore blocking can be divided into different sub-categories depending on the nature of the utile membrane area affected: i) intermediate pore blocking, where some particles can block the membrane pores or be accumulated onto other particles forming a pseudo cake layer; ii) standard pore blocking, where a reduction of the membrane pore size due to the accumulation of materials inside the membrane pores; and iii) complete pore blocking, where the particles accumulates only on the membrane surface, completely blocking the membrane pores). Fig. 1.7 shows a graphic scheme for each kind of fouling described.

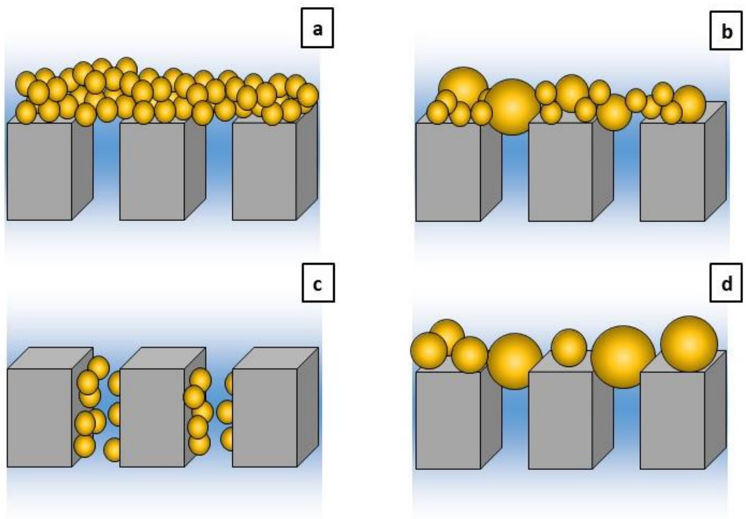

Figure 1.7. Main fouling mechanisms in membrane systems: (a) cake layer, (b) intermediate pore blocking, (c) standard pore blocking or pore narrowing and (d) complete pore blocking.

As introduced previously, severe fouling has been generally reported when employing porous membranes for filtering MWW. Membrane pore blocking is characterized by a quickly development during the initial minutes of each filtration stage [1.93] thereby dramatically reducing membrane permeability in the short-term. Due to the performance observed during raw MWW filtration, a large part of the developed fouling could be related with this mechanism. Nevertheless, as several authors conclude, other fouling mechanism could also produce similar outputs. For instance, the wide PSD in MWW may foment the quick formation of a consistent thick cake layer onto the membrane surface. In this respect, Fujioka and Nghiem [1.60] and Ravazzini *et al.* [1.25; 1.61] studied the fouling mechanisms affecting MF/UF membranes by mathematical approaches, concluding that although pore blocking phenomenon could occur at the very beginning of the filtration stage, the filtration process was predominantly governed by cake layer formation. In this case, the increase in the fouling growth rate detected in the following filtrations cycles would be related to a compression of the formed cake layer, reducing the medium porosity [1.60].

Despite having been stated that cake layer filtration is the main fouling mechanism, other authors have also indicated that fouling could be significantly affected by EPS and SMP substances [1.46; 1.61]. In this respect, Ravazzini [1.61] showed that when employing UF membranes for the treatment of both raw and PSE MWW, a significant fraction of the total influent EPSs was captured by the membrane (around 7 – 22% and 33 – 38% for proteins and polysaccharides, respectively). These substances could form aggregates, being absorbed and accumulated on the membrane surface or penetrate into the pores [1.61]. Moreover, Jin *et al.* [1.46] suggested that these substances may induce the formation of a sticky gel layer on the membrane, which could strongly attach deposited particles, especially when employing air scouring. Therefore, other fouling mechanism more related with EPS and SMP concentrations (*e.g.* gel formation on the membrane surface) may also importantly contribute to the severe fouling noticed during the DMF. Regarding the continuous filtration of raw MWW using MF membranes, Lateef *et al.* [1.43], Jin *et al.* [1.46] and Mezohegyi *et al.* [1.48] reported that despite raising the concentration of the bulk to be filtered (sludge concentration factors referred to influent concentration from 5 to 25, 21 to 50, and around 11 to 46, respectively), similar fouling propensities were achieved, detecting in some cases even less fouling developments compared to fresh raw MWW filtration. As similar fouling propensities were reported at different sludge concentrations, it could be stated that particulate material was not the main fouling promotor but the colloidal and/or soluble fraction. Similarly to other MBR studies (see for instance [1.89; 1.94]), the concentration of SMPs or colloidal EPSs could be determinant on the fouling propensity observed during DMF of MWW [1.48].

Regarding fouling nature, different studies using oxidant chemicals [1.36; 1.40; 1.43; 1.62] and ozone [1.47; 1.60] as cleaning agents have identified OM as the main fouling precursor, representing between the 68 – 89% of total membrane resistance. Furthermore, Kramer *et al.* [1.62] evaluated the composition of the cake layer developed on a ceramic

UF membrane, attributing about 98% in mass to organic materials. Since only a 2% in mass of the cake layer was attributed to inorganic elements (Na, Al, Si, S and Cl), the authors concluded that not important salt precipitation occurred. Nevertheless, Nascimento *et al.* [1.36] and Lateef *et al.* [1.43] defended that a significant inorganic fouling can be developed during DMF of MWW, showing the effectivity of acid chemicals (as citric acid) for membrane fouling control. Therefore, inorganic compounds deposition should be also considered as a fouling precursor in DMF processes, although to a lower extent.

It is important to highlight that, despite the massive and quick fouling development exhibited during DMF, different studies have showed that the main fouling generated in the short-term (7 – 20 hours) is apparently reversible [1.25; 1.36; 1.46; 1.48; 1.61] and, therefore, it could be effectively controlled by physical cleaning strategies. In this respect, Ravazzini *et al.* [1.25] suggested that, regardless the influent sewage treated (raw or PSE), the long-term DMF process could be possible just by applying middle cross-flow velocities and operating at moderate TMPs. Thus, better filtration performances for the DMF of MWW can be expected by applying proper operating conditions and fouling control strategies.

1.6.2 Fouling in semipermeable membranes

Osmosis membranes can be affected by the same fouling mechanisms that classic porous membranes. However, since they present more compact structures, surface fouling is comparatively more frequent [1.88], expecting higher proportions of reversible fouling when operating this type of membranes. Unfortunately, since fouling mechanisms may be extremely complex, especially in semipermeable membranes, fouling propensity can hardly be predicted [1.90]. Osmosis membranes generally entail lower fouling propensities than other membrane technologies [1.53]. Nevertheless, the contrary can also be found depending on the influent treated [1.95]. Similar outcomes have been reported when comparing FO and RO membranes.

FO membranes present lower fouling propensities than RO due to avoiding pressure in the feed side during operation which promotes the deposition and compression of foulants onto the membrane surface [1.96]. Indeed, numerous authors have shown the benefits on fouling control of using FO instead of RO [1.97]. Nonetheless, opposite results have been also reported [1.98], resulting in unclear conclusions. Consequently, although semipermeable membranes are generally identified as less prone to fouling processes, specifically in the case of FO filtration, the developed fouling may be more strongly dependent of both treated influent characteristics and interaction with the membrane material [1.88].

Regarding DMF technology, a significant fouling propensity has been mostly observed when filtering MWW with FO membranes, especially when operating at elevated feed concentration factors [1.54; 1.73]. In fact, a severe organic, inorganic and even biological fouling has been reported in DMF compared to activated sludge filtration at similar operating conditions [1.55]. Gao *et al.* [1.54] reported flux declines from 25 – 20 to 10 – 5 LMH in a TCA FO membrane when filtering MWW for 1500 min due to both DS dilution and fouling development. In this case, the authors determined that a significant fouling cake layer is formed after 500 – 700 min of operation, which development was intensified as the initial flux was increased. Consequently, moderate osmotic pressures could be recommended for fouling mitigation [1.54]. On the other hand, Ansari *et al.* [1.73] reported a 50% flux decline after 70 h of filtration, starting it after the first 40 h and evolving consistently (linear trend) as the filtration continued, whilst Zhang *et al.* [1.72] also reported flux reductions (from 8 – 6 to 4 – 3 LMH after 17 h of filtration) when operating with a CTA FO membrane. In this last study, however, two different membrane orientations were tested (active layer in contact with the DS or feed), observing a clear benefit during filtration when facing the active layer to the feed solution. As the authors concluded, a less thick fouling cake layer was formed in the latter orientation, which was related with the smoothness of the membrane active layer [1.72].

Concerning the nature of the formed fouling, some authors reported that the accumulated fouling layer when filtering MWW by FO membranes is loose and not tight, being easily removed [1.41; 1.54; 1.73] which will make it mainly reversible. In this respect, Gao *et al.* [1.54] reported that although about the 20 – 40% of the original membrane permeability was lost after 1500 min of MWW filtration, a permeability recovery of the 90% was achieved after a physical cleaning. Similarly, Ansari *et al.* [1.73] reported a complete membrane permeability recovery after a simple membrane flushing despite having lost the 50% of the permeate flux during filtration (70 h of continuous filtration). Nevertheless, a significant irreversible fouling has also been reported when treating

MWW. Gao *et al.* [1.54] informed that a chemical cleaning (1% NaOCl for 30 min) was necessary for increasing from 90 to 96% the original membrane permeability after 1500 min of operation. In consequence, these authors assumed that 90% of the developed fouling was reversible, 6 % could be considered as easy to remove (practically reversible) and the rest 4% irreversible. Less optimistic results have been reported by Ferrari [1.70], who just recovered the 80% of the original operating flux after performing an osmotic backwash (OBW). This fouling was besides intensified during the posterior filtration cycles, reporting average flux declines after every filtration cycle (4 in total) of 14.6, 24.4 and 26.8%, respectively, regarding the first filtration cycle one (filtration times of each cycle between 24.5 – 37.2 hours).

Regarding the fouling source, Gao *et al.* [1.54] determined that the developed cake layer was conformed mainly by carbon, calcium, magnesium, silicon, aluminum, and phosphorus. The constituents of this cake layer were mostly identified as polysaccharides, humic acids, and proteins present in the treated MWW [1.54]. A light chemical cleaning (1% NaOCl for 30 min) was able to easily remove aluminum and phosphorus which were considered as reversible foulants. Instead, calcium and silicon were more adhesive and not easy to remove, being considered as irreversible fouling promotors [1.54].

In addition to membrane fouling, other phenomenon gains an important relevance when operating osmosis membranes such as concentration polarization and reverse solute flux (also known as negative rejection) [1.99]. Concentration polarization consists in the formation of a fluid flux contrary to the permeate flux induced by several concentration gradients between the solutes present in feed and permeate solutions. This effect can take place in the proximity of the membrane, concerning the feed and permeate solutions themselves, which is then defined as external concentration polarization or take place in the pores or dense material of the membrane, being known then as internal concentration polarization [1.100]. External concentration polarization can be mitigated by any fouling control strategy that affects hydrodynamics such as gas sparging or crossflow operation, while internal concentration polarization is difficult to affect. Concentration polarization has showed a significant impact when filtering MWW, being determined as the most important filtration resistance in several studies. Wang *et al.* [1.53] identified the formation of an enhanced concentration polarization cake as the major contributor to flux decline during MWW filtration, while Ferrari [1.70] recognized external concentration polarization as an important flux decliner which effects were mitigated by gas scouring. In this respect, Zhang *et al.* [1.72] informed that orientating the active membrane layer to the feed solution could be an interesting option to reduce the negative effects of internal concentration polarization since it could promote a reduction of the fouling cake layer thickness and pore plugging, thereby improving the contact between the membrane surface and operating solutions. On the other hand, reverse solute flux is exclusive of FO filtration and refers to the flux of salts from the DS to the feed during filtration. This salts flux is contrary to permeate flux and entails a reduction of the effective system's osmotic pressure, negatively affecting the filtration cost [1.101]. Furthermore, the salts accumulated in the feed solution may inhibit the posterior AD process, compromising the main objective of this alternative treatment [1.54; 1.73]. Several authors have reported the significant reverse solute flux of salts when filtering MWW with FO membranes [1.54; 1.101], which could force the use of moderate DS salt concentrations or limit the concentration of MWW to moderate values to prevent AD inhibiting effects.

1.7 Membrane fouling control strategies

Membrane fouling is one of most relevant issues to address in any membrane-based system. The development of fouling during filtration entails significant increases in the process energy demands that may affect its technical/economic viability. Once membrane permeability is hindered by irreversible fouling, the use of chemicals (basic and acid solutions) to perform a membrane chemical cleaning is the only effective methodology to conduct. However, proper membrane chemical cleaning is an off-line methodology which require the complete stop of the process for several hours (membrane suppliers may recommend a direct contact between the membrane and each cleaning solution of 14 – 20 hours) while the use of chemicals (mainly basic ones) strongly decreases membranes lifespan. To prevent premature membrane chemical cleanings and enhance the filtration feasibility when filtering solution with high propensities to form fouling (such as MWW), multiple on-line cleaning strategies have been developed so far.

1.7.1 Physical methodologies

Physical cleaning strategies for fouling control have been extensively used in different MBRs [1.102]. These kind of strategies are mainly based on mechanical forces to continuously detach fouling formation from the membrane surface. Consequently, due to the nature of the applied cleaning, physical methodologies mainly act on reversible fouling mitigation, losing effectivity over time when irreversible fouling is developed [1.102]. Since significant energy inputs can be required depending on the employed fouling control strategy, their proper selection, application, and optimization is an essential matter in any energy-efficient and cost-effective filtration process.

1.7.1.1 Air Scouring

Air-assisted membrane-scouring is one of most extended methodologies for membrane fouling control, being successfully applied in numerous membrane systems [1.102]. Generally, the continuous air sparging is able to prevent fast particle deposition over the membrane surface by the induction of local shear stress and liquid flow fluctuations nearby the membrane surface, thus improving membrane permeability especially in the early filtration stages. Besides, in HF membranes, air sparging also favor cake layer detachment by the sway of membrane fibers in module package, reducing in addition membrane clogging problems which are especially important in HF high packing densities modules [1.103].

In the case of DMF of MWW, different studies have shown the effectiveness of air scouring for reversible fouling control in MF and UF membranes [1.36; 1.46; 1.48]. Jin *et al.* [1.46] showed that after 5 h of operation, membrane permeability could be hold around 10 times higher when high specific air demands (SAD) (about 0.6 $Nm^3\ m^{-2}\ h^{-1}$) are applied, whilst Nascimento *et al.* [1.36] reported that the application of intensive aeration (56 m^3 of air per m^3 of permeate) enabled the filtration process at low TMPs (around 150 mbar) and moderate fouling growth rates for a period of 40 days. Air/gas sparging have also shown great effectivity for improving FO membranes filtration. For instance, Ferrari [1.70] reported that air/gas sparging allowed to significantly reduce FO filtration flux decline thanks to reducing membrane fouling and concentration polarization. Specifically, a 70% water recovery in 40.1% less time was achieved when employing continuous air sparging while 17.4% less time was reached when intermittent N_2 sparging was used (both compared with a FO test without air-assisted fouling control). Additionally, a flux test was performed by the cited author after around 24 – 40 hours of filtration, highlighting that the flux declined compared to the original clean membrane one was about the 5.7, 12.4 and 31.3% for continuous air sparging, intermittent N_2

sparging and no gas injection, respectively [1.70]. According to these results, the author concluded that gas sparging (even intermittent) has a strong fouling mitigation effect, being able to control both, reversible and irreversible fouling.

Despite the effectivity showed by air sparging to control membrane fouling, several drawbacks are involved when applying this strategy to DMF. First of all, intensive SADs are usually required in DMF to get a suitable fouling mitigation, which are significantly higher than those reported in MBRs [1.36]. As some authors point (see *e.g.* [1.36]), high SADs requirements during DMF of MWW could be related with a specific resistance raise of membrane surface solids deposition during the filtration of untreated MWW compared to that presented in activated sludge in MBRs. Since air scouring commonly represents the most energy demanding instrument in MBRs [1.102], other cleaning strategies should be considered in order to improve the energy balance of DMF [1.36; 1.40]. On the other hand, the combination of small particles present in untreated MWW (*e.g.* sand or crystalline precipitates) and air sparging may promote abrasive effects on the membrane surface resulting in reductions of membrane lifespan [1.21]. Moreover, the continuous membrane cleaning by air scouring could result in a more vulnerable membrane surface against EPS and SMP [1.46], being these compounds identified as main membrane fouling promotors on MBRs [1.94]. In this respect, Jin *et al.* [1.46] reported that the use of air sparging was ineffective in the last moments of filtration which the authors justified as a possible faster gel layer formation in these systems than that observed in other MBR. This gel layer could strongly attach deposited particles onto the membrane surface due to their sticky proprieties, drastically reducing the effectivity of air sparging to control membrane fouling [1.46; 1.104]. On the other hand, the continuous injection of air in the system can result in a reduction of DMF energy recovery potential by promoting the development of heterotrophic organisms, which consumes OM retained in the system [1.44–1.46]. In addition, OM degradation by heterotrophs could induce the raise of EPSs and SMPs in the membrane tank, contributing to membrane fouling. Indeed, significant heterotrophic activity has been reported even when low air flow rates were introduced in the system [1.43; 1.44; 1.70]. Then, even for mixing purposes, mechanical systems have been recommended instead of aeration [1.40; 1.43; 1.46].

1.7.1.2 Cross-flow operation

Based on side-stream configuration, cross-flow operation has also been applied in numerous membrane systems for fouling mitigation. Thanks to introducing a flux perpendicular to the permeate flow with significant tangential velocity, fast particles accumulation on membrane surface can be mitigated, reducing the thickness of the formed cake layer [1.61]. Also, due to the turbulence and scouring effects promoted on the membrane surface, deposited solids back-transport is favored [1.61], especially during relaxation periods when the formed cake layer can be detached from the membrane surface.

In DMF technology, the effectivity of applying cross-flow for fouling mitigation has been severally proven [1.25; 1.61; 1.63; 1.73; 1.105]. Ravazzini [1.61] showed the effectivity of this strategy for fouling control when treating both raw and PSE MWW using UF membranes. The filtration resistance was considerably reduced in this study by increasing the cross-flow velocity from 1 to 2 m s^{-1}, allowing to operate for 7 h without a relevant fouling appearance. Ahn *et al.* [1.63] informed similar results when operating MF and UF ceramic membranes for hotel building wastewater filtration, achieving stables TMPs during more than 12 h of continuous filtration when using cross-flow (2.5 – 4 m s^{-1}). Moreover, Ansari *et al.* [1.73] also showed the effectivity of cross-flow operation when employing a CTA FO membrane for filtering MWW. Water recoveries of only the 70% were able to be achieved in this last study when the cross-flow velocity was set in 9 cm s^{-1}, reporting a considerable flux decline during filtration due to a severe fouling formation. However, this fouling was significantly reduced when the cross-flow velocity was almost doubled (17 cm s^{-1}), achieving in this case a water recovery efficiency of 90%. This strategy showed therefore its effectivity regardless the membrane pore size used (MF, UF and FO membranes) and treated influent (raw or PSE MWW). Nevertheless, since providing high cross-flow velocities during filtration represents an important energy cost by intensive pumping, the optimization of this operation is imperative. According to Hao *et al.* [1.105], who used MF membranes for raw MWW filtration, increasing cross-flow velocity (ranging from 2 to 3.7 m s^{-1}) always resulted in significant flux improvements. Similar improvements on the permeate flux regarding applied cross-flow velocity (ranging from 1 to 4 m s^{-1}) were reported by Ahn *et al.* [1.63] operating ceramic MF and UF membranes for hotel building wastewater treatment. However, when operating a UF membrane for raw and PSE MWW treatment, Ravazzini [1.61] showed that cross-flow velocities above 2 m s^{-1} did not entail significant improvements in the process stability or permeate production thereby they recommended to operate around 1.5 m s^{-1}.

Besides controlling fouling growth rate during filtration, enhanced shear conditions provided by cross-flow operation could affect developed fouling by changes on the membrane material transportation. Indeed, shear conditions could affect protein aggregates in the treated solution or induce the aggregation of inorganic colloids around the membrane pores [1.61]. Moreover, high cross-flow velocities can also boost the formation of irreversible fouling due to the higher permeation drag achieved under these conditions [1.106]. In this respect, Ravazzini [1.61] detected that, although fouling was mainly reversible when filtering raw and PSE MWW with a UF membrane in cross-flow mode, a significant irreversible fouling development always appeared at the beginning of the operation regardless of the cross-flow velocity applied. On the other hand, in cross-flow UF, the main interacting forces among particles are related with particle size [1.25]. Thus, the use of more depurated influents (*e.g.* PSE) may affect cross-flow cleaning effectivity. In this regard, when treating PSE by UF membranes, Ravazzini *et al.* [1.25] detected that a denser cake layer could be formed on the membrane surface due to a reduction in the average pore size of the deposited particles. This phenomenon was highlighted by the authors since the formed cake layer under these circumstances was more effectively removed by the applied cross-flow, thereby reducing the membrane resistance.

Concerning the operating permeate flux, the use of middle/low fluxes could be recommended. This is due to the reduction of the transporting material forward the membrane surface during filtration which have been reported as a beneficial action for enhancing the scouring effects of cross-flow [1.25]. However, the proper permeate flux need to be optimized in each case for maximizing wastewater treatment without compromising membrane fouling.

1.7.1.3 Backwashing

Together with air scouring, the use of backwashing with tap water or generated permeate is the most common practice for cake layer removal in MF and UF membranes [1.62].

Thanks to periodic backwashing, the formed cake layer can be detached from the membrane surface, recovering membrane permeability and relieving cake layer compression. Unfortunately, the use of backwash has been showed as an inefficient strategy for fouling mitigation in DMF [1.43]. Kramer *et al.* [1.62] observed no differences in the filtration performance when intensive backwash was applied hourly when operating a ceramic UF membrane. Similarly, Fujioka and Nghiem [1.60] showed that backwashing was not enough to control membrane fouling, even when it was performed every filtration cycle. The ineffectively of backwashing for fouling growth control could confirm the different hypothesis performed by some authors regarding the main fouling mechanism during DMF of MWW, *i.e.* pore blocking (by colloidal particles or by gel layer formations) or consolidated cake layer formation that cannot be removed when the flux was inverted [1.46; 1.61]. Despite these results, the use of backwashing could be useful as complementary cleaning, alleviating a possible OM concentration polarization on the membrane surface and the compression of the formed cake layer [1.44]. Indeed, some authors have reported that when using cross-flow operation for fouling control, backwashing is strictly necessary and raising its frequency can help to mitigate fouling [1.61].

When operating FO membranes, backwashing is usually carried out by using osmotic pressure as driving force. In this case, after some conventional filtration, the membrane permeate side is replaced by tap water or any other cleaning solution, feeding the membrane waste side with the same DS employed in the permeate side during filtration. Since the driving force changes directions towards the membrane waste side, a permeate flux appears which is used as other pressure-driven membranes backwashing for detaching any foulant remaining in the membrane surface. This methodology has showed good efficiencies for mitigating fouling, even when its source is biological [1.107]. Unfortunately, likewise in porous membranes, this strategy seems to not be enough for controlling fouling by itself in the case of MWW direct filtration. At this respect, Ferrari [1.70] reported that after around 23.9 – 39.9 hours of filtration, a OBW was just able to recover the 80% of the original membrane flux, indicating that an important fouling still remained despite the cleaning. This membrane permeability recovery was able to be enhanced only when other cleaning methodologies (continuous or intermittent gas sparging) were used together with OBW, achieving in this case nearly full recoveries (95.1% of the original membrane flux).

1.7.1.4 Use of granular materials

The introduction of granular materials in the membrane tanks during filtration have been proposed by some authors to provide a direct membrane surface mechanical cleaning [1.40]. Sparging this material allows to detach fouling depositions by hitting and scouring the membrane surface, having displayed good results in different flat-sheet MBRs [1.108; 1.109]. This alternative has captured some interest for being applied during DMF of MWW since the required sparging can be easily provided by mixing mechanism, such as vibrations or paddle stirring, avoiding air injection in the system and significantly reducing the cleaning energy input [1.40]. Nevertheless, the use of this strategy when employing HF membranes seems to result inadequate. For instance, Kimura *et al.* [1.40] studied the efficiency of using granular materials in the operation of a HF MF membrane treating PSE MWW, reporting similar behaviors regardless the use of the granular material. As these authors reported, this effect was due to the capture of the used granules among the membrane fibers, inhibiting the physical cleaning. To avoid the granules to be captured by the membrane, Kimura *et al.* [1.40] combined this strategy with a vibrating mechanism. Unfortunately, not significant enhancements in membrane cleaning were observed either in this case, reporting even negative effects when the sludge was concentrated to around 8 g L^{-1} of COD. In this case, the increase in the medium viscosity could have affected negatively the granules dynamics, deteriorating the medium filterability. This phenomenon has been also reported by other studies [1.108], resulting in alterations on the foulants characteristics because of the granules performance that revealed increments in the irreversible fouling development when this strategy was used. Since one of the main objectives of DMF is to increase as much as possible the sludge concentration in order to reduce pumping energy and AD volume requirements, this alternative seems not to be a proper approach when filtering untreated MWW, especially when operating high density packing membranes.

1.7.1.5 Vibrations

Membrane-movement-based strategies have also been proposed for controlling fouling development in MBRs. This strategy focuses on providing an intensive movement to the membrane materials for raising their liquid-membrane interface shear and favors membrane fouling detachment. This membrane movement is generally produced by fibers vibrations and has shown a relevant effectivity in diverse submerged MBRs [1.110;

1.111]. Concerning DMF, Kimura *et al.* [1.40] showed that operating a HF MF membrane for PSE MWW treatment it was possible to significantly enhance the membrane permeability by applying intermittent vibrations. As these authors reported, filtration was able to be extended for 100 h at low TMPs (under 30 kPa) when intermittent vibrations were employed, reaching about 45 kPa in less than 5 h when this mechanism was not used. On the other hand, Mezohegyi *et al.* [1.48] reported that membrane cleaning by vibrations allowed a better control of fouling than aeration, achieving critical fluxes of 10, 15, and 25 LMH when any control strategy, air-assisted membrane scouring, and vibrations were used, respectively. Due to the significantly lower energy demand of vibrations compared to aeration, this cleaning strategy has been considered as an attractive alternative for the DMF fouling control [1.48].

Despite the promising results reported on membrane vibration for fouling control during DMF of MWW, some undesirable effects have also been observed when applying this alternative. Indeed, although fouling removal may be improved by increasing vibrations intensity and/or frequency, intensive vibrations could negatively affect OM recovery [1.48]. Raising vibration intensity can generate a strong perturbation on the medium, enhancing the air capture and gas transfer in the liquid phase [1.112] and ultimately improving the aerobic bacteria development. Additionally, intensive vibrations can cause perturbations on the particulate material, inducing the partial solubilization of the low size suspended solids fraction [1.48], thereby negatively affecting the resource recovery efficiency and permeate quality. Hence, a deep study and optimization of this alternative is required to determine its suitability for MWW using DMF technology.

1.7.2 Chemical methodologies

In contrast to physical methodologies, chemical cleaning strategies are mainly focused in mitigate/remove irreversible membrane fouling [1.102]. Although irreversible fouling is commonly associated to long-term operation, several authors have informed of the short-term physical methodologies ineffectiveness when operating with untreated MWW (see for instance [1.36]), denoting the necessity of implementing other more effective methodologies to achieve viable systems. Since these strategies involve important reagents dosing (*e.g.* chemical membrane cleaner reagents) or energy-intensive associated processes (*e.g.* ozone generation) their proper selection, utilization, and optimization regarding treated influent is essential in order to achieve feasible processes.

1.7.2.1 Coagulation/flocculation

Coagulation/flocculation is generally consolidated as an efficient methodology to reduce fouling development in membrane processes. Coagulant addition promotes the aggregation of small particles to produce more voluminous flocs which are able to prevent the membrane pore occupation by colloidal materials [1.44]. On the other hand, soluble compounds can also be captured based on other mechanism, such as adsorption or precipitation, resulting in a reduction of these components hindering effects on the membrane surface or pores. Additionally, the formed voluminous flocs during this treatment can quickly cover the membrane surface during filtration, protecting the membrane from other fouling promotors [1.61] and reducing the interaction between the new influent particulate material and the membrane surface [1.46]. Increasing the average particles size can also bring some other beneficial consequences such as the generation of thinner and more porous cake layers [1.61] that would be more susceptible to be removed by back-transport mechanism such as gravitational and/or shear forces [1.44; 1.46; 1.61]. These more easily-to-remove cake layers would therefore allow to operate at higher permeate fluxes [1.44], representing an important benefit for the process viability by reducing the membrane area requirements for MWW filtration. Nonetheless, coagulation cost can represent a determinant issue when requiring a continuous dosage. Moreover, coagulant dosing results in increased sludge productions and may affect its biodegradability due to chemical contamination [1.61], compromising the main aim of DMF technology. Therefore, this strategy needs to be deeply studied in each case in order to select a suitable reagent and dosing concentration.

Concerning DMF of MWW, numerous studies have reported the beneficial effects of coagulation on wastewater filterability [1.21; 1.44; 1.46; 1.61]. Jin *et al.* [1.46] reported high filtration improvements when operating a MF membrane for raw MWW treatment, enhancing the membrane permeability after 5 hours of operation from 20 to 300 LMH bar^{-1} when coagulation was used. Similarly, Jin *et al.* [1.44] reported a permeability more than 5 times higher after 1 hour of operation under similar conditions, whilst Ravazzini [1.61] showed filtration resistance reductions up to 35% when operating an UF membrane for raw MWW treatment. Moreover, average fouling rates of around 5.5 – 6.7 mbar h^{-1}

have been displayed when employing coagulation in DMF [1.46], being closely to values achieved in MBR systems [1.46]. On the other hand, thanks to the promoted capture of colloidal and soluble compounds, coagulation seems to significantly reduce the development of irreversible fouling in the middle-term operation of DMF (about 80 – 300 h) [1.46; 1.61]. Indeed, thinner and most loosely attached cake layers have been observed when employing coagulation [1.44]. Therefore, frequent chemical cleanings could be prevented by using coagulation [1.46], resulting in higher membrane lifespans. However, despite all these reported beneficial aspects, the effect of coagulation itself could be not enough for fouling mitigation in the long-term operation. In fact, despite the coagulation dose used, a consistent permeability decrease has been reported during filtration in the middle-term operation (between 7 and 70 h), reaching values similar to those obtained when no chemicals were used [1.44–1.46]. This is due to an incapacity to remove the suspended material accumulated onto the membrane when only dosing chemicals. Therefore, this strategy should be always implemented together with other assistant methodology for effectively controlling membrane fouling in the long-term, such as air sparging, cross-flow, or backwashing.

1.7.2.1.1 Coagulant reagents

Numerous reagents have been proposed to improve the feasibility of MMW DMF. In fact, different inorganic/metallic coagulants (*e.g.* iron chloride ($FeCl_3$) [1.21; 1.61; 1.113], alumina salts (*e.g.* $Al_2(SO_4)_3$) [1.61] or polyaluminum chloride (PACl) [1.21; 1.35; 1.44; 1.61; 1.113]) and organic/polymeric coagulants (*e.g.* C-492 [1.61; 1.114], C-581 [1.61] or C-592 [1.61]) have been employed for both fouling mitigation and OM recovery.

Among the proposed coagulants, PACl has been usually selected by its aptitude to form larger aggregates and its higher OM capture capacity regarding other coagulants, achieving generally better results in jar tests [1.45; 1.46; 1.65; 1.115]. Additionally, stronger produced flocs and better performances regarding membrane filtration have been also reported when using PACl [1.61; 1.116]. However, $FeCl_3$ is the cheapest and more extended chemical reactive in MWW treatment, while other aluminum based salts also display high coagulation performances at low prices [1.117]. Thus, these alternative reagents can also be considered as an attractive option for reducing the coagulant-assisted DMF cost.

Regarding to polymeric coagulants suitability for the DMF fouling mitigation, their benefits are unclear. Rulkens *et al.* [1.114] reported negative effects on membrane fouling rate when using C-492, and Hey *et al.* [1.21] informed of low permeate fluxes when employing cationic polymeric coagulants thereby concluding that the filtration performance is negatively affected by these reagents. Nevertheless, in the short term (*e.g.* 40 min), Ravazzini [1.61] reported reductions in the membrane resistance of up to 18, 32 and 61% when using $FeCl_3$, PACl and C-592, respectively, showing the better performance of organic coagulants when treating MWW. Based on these results, it could be assumed that organic coagulants could be a competitive alternative under certain circumstances although its suitability should be studied for each specific scenario. Regarding polymeric coagulants, C-592 seems to present higher performances than C-581 at low dosing [1.61] and, although C-492 shows the better particles capture at low dosages, its use is not recommended by the sticky flocs generated which may negatively affect filtration [1.61].

1.7.2.1.2 Dosing protocol

The coagulant dosing protocol is one of most important issues to consider to achieve proper DMF performance. Indeed, as several studies show [1.45; 1.61], coagulant performance is more strongly related with dosing methodology, concentration, and medium interaction than with the reagent itself. Since coagulant addition represents an extra operating cost, this reagent should be limited to small dosages [1.61]. However, both coagulant underdose and overdose have shown negative effects on DMF technology [1.45; 1.61]. Low coagulant concentrations are ineffective and even prejudicial to filtration performance since they produce a raise on the TSS concentration in the system without forming flocs [1.61]. On the other hand, overdosing produce an excess of available cations, positively charging the suspended particles which results in stable suspensions instead of flocculation [1.118]. Hence, the proper determination of the most convenient dosage considering the selected coagulant and its interaction with the influent wastewater characteristics is an imperative task. In the case of PACl, concentrations of about 15 – 30 mg L^{-1} have been generally reported as optimum dosages when filtering untreated MWW [1.21; 1.44; 1.46; 1.115].

Coagulant addition methodology also plays an important role in fouling control. As Gong *et al.* [1.45] reported, the punctual addition of PACl produced more severe fouling problems than direct filtration itself, while continuous dosage achieved opposite results. Rulkens *et al.* [1.114] reported similar results when applying cationic electrolyte (C-492) punctually, noticing a negative effect on the membrane fouling rate. This phenomenon could be attributed to the sudden increase of the TSS concentration due to particles flocculation in the one-time coagulant addition together with a raise on the colloidal material deposition onto the membrane surface due to the more porous initial cake layer formed [1.45]. Then, one-time coagulant addition may promote the formation of thicker and tighter cake layers, increasing the filtration resistance [1.119]. To avoid these unfavorable effects, Gong *et al.* [1.45] recommended the use of continuous low dosing instead of high punctual coagulant additions, controlling in this way continuously the amount of soluble and colloidal particles that reach the membrane surface.

1.7.2.1.3 Strategies to improve coagulant effectivity

As Jin *et al.* [1.44] reported, the use of a coagulant by itself could not be a suitable solution by the lack of a direct removal of the formed cake layer. Thus, this strategy should be always implemented together with an additional cleaning strategy, such as permeate and/or air backwashing. Thanks to an appropriate back-transport force, the cake layer formed can become thinner while raising their prone to be flushed back into the mixed liquor [1.44]. Cross-flow operation has also been proposed as assistant cleaning strategy when dosing coagulant. However, temporal effectivities (reduction of the membrane resistance for just several hours) have been reported when applying this option [1.61]. This phenomenon could be related to changes in the cake layer composition and structure during filtration [1.45] or to flocks break by the provided shear forces [1.61]. Hence, when employing cross-flow, Ravazzini [1.61] recommended the use of PACl by their stronger produced flocs.

On the other hand, Gong *et al.* [1.45] studied the effect of increasing coagulant dosing to improve this alternative effectivity on fouling control. Unfortunately, even after raising PACl dosing concentration 10 times (from 60 to 600 mg L^{-1}), slight enhancements in the membrane resistance (from $4.5 \cdot 10^{13}$ m^{-1} to $4.0 \cdot 10^{13}$ m^{-1}) were observed in an operating time of 30 h. Given the few benefits that rising chemicals costs would provide regarding filtration resistance enhancement, this option was strongly not recommended. As an alternative, Hey *et al.* [1.21] studied the use of both, coagulant and flocculants addition for controlling membrane fouling. However, as these authors reported, the use of flocculants only favorably affected resources recovery, not influencing the filtration performance in any of the tested coagulant/flocculants concentrations. Finally, Gong *et al.* [1.45] determined that the combination of a continuous PACl dosage with a punctual addition of powered activated carbon (PAC) may be a suitable alternative for fouling mitigation. Indeed, thanks to PAC addition, the filtration resistance was dramatically reduced (from $5.0 \cdot 10^{13}$ m^{-1} to $1.5 \cdot 10^{13}$ m^{-1}) during 120 h of operation under the conditions described in this study. This improvement was attributed to the adsorbent capacity of PAC, reducing the presence of dissolved OM, such as proteinaceous matter and some humic-type substances [1.120], hence preventing their deposition on the membrane surface. The results reported could denote both, that the membrane fouling was significantly controlled by SMPs and EPSs concentration and that PAC can be used to effectively control this fouling focus.

1.7.2.2 Chemical enhanced backwash

Membrane irreversible fouling is commonly removed by periodical chemical cleaning using basic and/or acid solutions during several hours to favor membrane external and internal cleaning. Due to the high effectivity of this methodology to recover membrane permeability, the same reagents can be used to minimize irreversible fouling development during continuous filtration operation [1.121]. Chemical enhanced backwashing (CEB) consists in the application of low basic and/or acid reagents concentrations during backwashing to improve backwash effectivity thanks to reagents action. In DMF, Lateef *et al.* [1.43] studied the effect of CEB on fouling control of a MF membrane treating raw MWW, reporting significant permeability improvements when using low frequency CEBs. Indeed, a 0.1% NaOCl CEB for 30 s every 12 h at a flux of 210 LMH allowed the continuous membrane operation (for more than 200 h) at a TMP below of 30 kPa in comparison to the 45 kPa reached at 96 h of operation when only pure water was used. Similarly, Kramer *et al.* [1.62] reported permeability recoveries about 93% when applying every 22 h a consecutive backwashing by 0.1% NaOCl and 0.1 mol L^{-1} HCl solutions (15 min each one) on a ceramic UF membrane. Unfortunately, as Fujioka *et al.* [1.60] and Gong *et al.* [1.45] suggested, the chemical cleaning effects may be temporal, quickly recovering the poor membrane permeability conditions, thus requiring of high CEB frequencies to efficiently improve the filtration process in the long-term. Consequently, the nature of the developed fouling as well as the CEB applied reagent, concentration, intensity and frequency need to be thoroughly studied and optimized in each case. Additionally, due to the structural damage that chemicals cause on polymeric membranes [1.40], more robust membrane materials (*e.g.* ceramic membranes) could be recommended in order to improve membrane lifespan [1.62].

Since OM is identified as the main fouling precursor in DMF of MWW [1.36; 1.40; 1.43; 1.62], oxidant chemicals can be suggested to efficiently mitigate fouling, being NaOCl the most extended one [1.43; 1.45; 1.60; 1.62]. In fact, compared to NaOH, NaOCl has reported higher reversible and irreversible fouling cleaning efficiencies [1.43], achieving effective fouling mitigations even at high sludge concentrations [1.43]. However, other specific cleaning reagents (*e.g.* Divos109) can present higher cleaning performances in DMF systems for MWW treatment [1.25]. On the other hand, inorganic matter can also represent an important fouling precursor in DMF [1.36; 1.43]. As Lateef *et al.* [1.43] reported, although NaOCl presented the best cleaning performance, citric acid showed more effective cleaning effects than NaOH. Nevertheless, the use of citric acid was shown to be not recommendable for irreversible fouling control by itself [1.43]. Moreover, Nascimento *et al.* [1.36] determined that both citric acid and EDTA can be used for inorganic fouling removal, achieving similar performances. Due to the significant organic and inorganic fouling developed reached during DMF of MWW, alternative use of NaOCl and citric acid CEBs could be proposed as a suitable option for fouling mitigation [1.43; 1.62].

1.7.2.3 Ozone

Alternatively to chemicals, ozone can be used to remove OM-based fouling from the membrane surface and/or membrane pores [1.15; 1.122]. Fujioka and Nghiem [1.60] studied the effect of adding ozone during backwashing for organic foulants oxidation in a ceramic MF membrane treating raw MWW. These authors observed that when ozone concentrations of 2 mg O_3 L^{-1} were employed after each filtration cycle (10 min of continuous filtration), the membrane recovered the initial permeability with only 2.5 min of backwashing, being used effectively as fouling control strategy in the short-term operation (60 min). Additionally, the authors determined that this complete membrane cleaning was able to be achieved even when reducing the periodicity of ozonized water backwashing to 1:4 (backwash:filtration cycle ratio), although applying longer backwashing periods (3.5 min). Thus, the effectivity of this methodology could be optimized adjusting the backwash time according to the fouling development. Similarly, Kim *et al.* [1.47] showed that ozone enhanced backwashing (ozone concentrations between 0.2 – 0.8 mg O_3 L^{-1}) resulted in significant fouling removal improvements when treating raw MWW by a metallic MF membrane, achieving permeability recoveries of around 47% when simple backwashing was used, compared to the recovery of 92% displayed when ozone was employed. In addition, the metallic membranes used did not suffer any integrity damage during the study. Despite the potential beneficial effects of this alternative, the in-situ generation of ozone is cost-intensive [1.15] and requires advanced control systems to avoid dangerous issues, compromising the DMF process. Moreover, robust membranes (*e.g.* ceramic or metallic) need to be used when employing this methodology to avoid any integrity membrane damage during filtration, thus excluding the use of the cheaper and more studied/applied type of membranes (*i.e.* polymeric membranes).

1.7.3 Combination of different strategies

As Kimura *et al.* [1.40] suggested, since cleaning efficiency of a single method may not be enough for the DMF long-term operation, a combination of different physicochemical cleaning strategies could improve significantly the filtration performance. Indeed, when intermittent membrane vibrations, mechanical agitation, and periodical CEBs cleaning strategies were combined during the operation of a HF MF membrane treating PSE MWW, the TMP was perfectly controlled [1.40]. Indeed, TMPs lower than 10 kPa were achieved for more than 600 h of operation, even when the concentration in the treated mixed liquor reached about 8 g L^{-1} of COD. On the other hand, Jin *et al.* [1.44] used coagulation combined with intermittent air backwashing for raw MWW treatment by MF membranes, reporting sharply membrane permeability enhancements after 7 h of operation (300 LMH bar^{-1} in comparison with the 20 LMH bar^{-1} observed when only coagulant was employed). Similarly, Jin *et al.* [1.46] compared this same strategy with intensive air sparging for fouling control in a MF membrane treating raw MWW, reporting a membrane permeability of about 100 and 45 LMH bar^{-1}, respectively, after 20 h of operation. Thus, these authors appointed this combination as an effective alternative for both membrane fouling mitigation and process energy demand minimization. Furthermore, fouling growth rate was not only significantly reduced (from 80 kPa h^{-1} when direct filtration was performed to 5 kPa h^{-1} when this strategy was employed), but also the maximum fouling increase was reached at high solids operating concentrations (from a concentration factor of 5 when direct filtration was performed up to a concentration factor of 12 when this strategy was employed) [1.44]. Hence, this strategy showed to be effective for membrane fouling mitigation while allowing to filtrate at high solids concentrations in membrane module. Finally, when this strategy was compared to CEB application, the authors concluded that a more effective filtration performance can be obtained with the presented alternative, improving additionally the membrane productivity and reducing the OM mineralization.

1.8 Resources recovery potential of DMF

DMF technology main aim is to enhance resource recovery from MWW. Thus, the most suitable membrane technology to its implement would be decided not only based on filtration performance but also on membrane capacity to capture influent resources.

1.8.1 Micro- and ultra-filtration membranes

As it has been reported in numerous MBR studies, high permeate qualities can be achieved when employing MF or UF membranes. The pore size of these membrane technologies allows all particulate material to be captured, only allowing the pass of soluble compounds and a fraction of colloids. Regarding their application to direct filtration of MWW, turbidity reductions of around 97 – 99 % have been reported when using this type of membranes [1.26; 1.60], achieving NTU values in the permeate below 1.0 [1.23; 1.25; 1.26; 1.61; 1.123]. However, due to the significant amount of colloidal material that may be present in untreated MWW, significant amounts of total solids can be reached in the permeate, reporting in some cases poor influent capture efficiencies, ranging between 20 – 28% and 35 – 41% of total solids (TS) and volatile solids (VS) concentrations, respectively [1.36].

Concerning OM, high fractions of influent COD can be recovered during the direct filtration of MWW, generally achieving COD recovery efficiencies of up to 70 – 80% [1.22; 1.40; 1.43; 1.62; 1.64] and BOD recovery efficiencies about 47 – 95% [1.21; 1.22; 1.24; 1.26]. However, changes in the membrane pore size and influent wastewater characteristics could strongly affect both OM recovery efficiency and permeate quality.

Indeed, the results reported by numerous studies are not consistent each other, having been reported a wide range of COD recovery efficiency (from 35 to 80%) regardless the pore size of the membrane employed [1.21; 1.25; 1.36; 1.40; 1.43; 1.124].

To unveil the real relevance of membrane pore size on OM recovery, Ahn *et al.* [1.64] studied the effect of using different UF membranes pore sizes for filtering MWW, observing limited improvements on COD recovery (around 80, 83 and 84%) as the membrane pore size was reduced (300,000, 150,000 and 30,000 Da, respectively). In addition, Kramer *et al.* [1.62] showed not significant differences in COD recovery efficiency when comparing UF and NF membranes for raw MWW treatment. Similarly, other authors reported that no significant OM recovery efficiency differences are expected by filtrating raw or PSE MWW [1.21; 1.25], detecting in some cases slight reductions in OM recovery when treating PSE (lower than 5%), probably related with the previous reduction on the particulate material content in the influent [1.21]. Additionally, Lateef *et al.* [1.43] also concluded that the increase in the operating sludge concentration do not affect the permeate quality. Thus, as Ravazzini *et al.* [1.25] concluded, the OM recovery would be more related to the characteristics of the influent to be treated (*e.g.* OM content, particulate fraction, PSD, etc.) than to the membrane or influent pre-treatment employed.

On the other hand, different authors have proven the feasibility of OM concentration by DMF, achieving solids concentrations between 5 and 15 gDQO L^{-1} [1.36; 1.43–1.45] which could be directly valorized in AD processes [1.36]. Additionally, the recovered OM have shown high biodegradability rates [1.36; 1.46], similar to those reported from primary and activated sludge digestion [1.36], being the low size particles the most biodegradable ones [1.46]. Unfortunately, operating at elevate sludge concentrations during DMF of MWW could not only hinder filtration performance but also reduce the process feasibility by the mineralization of retained OM. In this respect, Rulkens *et al.* [1.114] reported OM mineralization degrees of about 50% when operating in batch mode for 8 days, also detecting a partial conversion of soluble COD into colloids, probably due to the generation of bio-flocs, increasing the average particle size. In accordance to this, Faust *et al.* [1.24–1.26] reported important OM mineralization degrees (up to 32%) when operating a membrane module at solids retention times (SRT) of 5 days, concluding that this negative effect can be effectively mitigated by operating at short SRTs. Indeed, small

COD losses (around 1 – 2%) were reported when operating at SRTs of 0.125 – 0.250 days [1.124; 1.125].

Regarding nutrients capture, total nitrogen (TN) and total phosphorous (TP) influent recoveries ranging between 9 – 36% and 15 – 75%, respectively, have been reported when operating MF/UF membranes [1.21; 1.22; 1.25; 1.114; 1.123]. Unfortunately, since important nutrients concentrations can be found in the soluble fraction of MWW, low nutrient recovery efficiencies could be reached in DMF [1.43; 1.48; 1.61], being the recovery efficiency strongly related to the amount of nutrients present in the particulate material of the influent wastewater. Indeed, the influence of UF and even NF membranes on untreated MWW ions concentration is extremely limited, reporting recoveries below 10% for NH_4^+, SO_4^{2-}, Mg^{2+}, and Ca^{2+} [1.62]. In the case of PO_4^{3-}, high recoveries (up to 97%) have been reported when using NF membranes [1.62], but descending until values comparable to those reached in UF (14 – 9%) as membrane fouling develops [1.62]. These results would be in accordance with those reported by Fujioka and Nghiem [1.60], who reported that low conductivity reductions (around 9%) can be expected during raw MWW filtration by MF membranes.

In addition to particulate resources capture, membrane-based systems usually act as a physical barrier for microorganism, including pathogens and virus, which can be later eliminated during AD. Hence, membrane effluent can be partially-disinfected, promoting its reclamation [1.127]. Concerning DMF, Sethi and Juby [1.26] reported excellent removal efficiencies of coliforms when operating a MF membrane for PSE treatment. Indeed, average total coliforms and fecal coliform bacteria levels were reduced from $9.4 \cdot 10^6$ and $2.7 \cdot 10^6$ MPN to 204 and 25 MPN in 100 mL, respectively. Meaningful removals of coliphage viruses were also reported, achieving an average influent-to-permeate removal from $2.6 \cdot 10^5$ to 838 PFU in 100 mL. Moreover, other important pollutants (*e.g.* heavy metals and micro-plastics) can be also removed by MF and UF membranes [1.21]. Thus, DMF technology shows an elevated potential to solve some relevant issues of MWW treatment, providing high quality permeates.

Despite all the above-mentioned benefits, the high soluble compounds content, specially nutrients, may not allow the direct discharge of the produced permeate into natural water bodies [1.22; 1.36; 1.43]. In fact, effluent COD, BOD, TN and TP concentrations ranging between 7 – 550, 1 – 65, 11 – 48, and 6.4 – 0.5 mg L^{-1}, respectively, have been reported in DMF [1.22; 1.23; 1.26; 1.36; 1.46; 1.48], which may not meet the standards established by the European Union regulation on discharged water quality. Therefore, different alternatives have been proposed to recover and valorize the valuable soluble resources presents in the DMF permeate, such as reverse osmosis [1.62; 1.114], packed biological filters [1.40], electrodialysis [1.47], or anaerobic ammonium oxidation (Anammox) process [1.46], have been proposed in order to. On the one hand, as some authors suggested [1.21; 1.46], the produced permeate might be applied for irrigation purposes when possible, suppling crops with viable nutrients.

1.8.2 Osmosis membranes

Given the nature of osmosis membranes, high resource recovery potentials can be expected when filtering MWW. Complete retention of influent particles can be assumed, while microorganism (virus and bacteria) and other contaminants (heavy metals, pharmaceutical products, hormones, microplastics, etc.) can also be removed [1.74]. Almost complete COD removals (between 95 – 99.8%) have been reported when using FO membranes for DMF of MWW [1.51; 1.52; 1.128]. However, as reported in some studies [1.53], a significant fraction of the influent COD (around 19%) can be lost due to degradation or attachment on the membrane surface. These COD losses should be properly identified for preventing fouling and enhance process energy efficiency. Regarding retentate characteristics, different authors have reported the viability of concentrate the MWW influent material in 5 – 10 fold by FO membranes [1.53 1.72; 1.73]. Thus, the FO retentate could be directly valorized via AD [1.73]. In addition to enhance energy recovery via AD, DMF of raw MWW through FO would also bring benefits to other treatment technologies, such as AnMBR. For instance, Vinardell *et al.* [1.101] reported that a methane yield increase from 214 to 322 mL CH_4 g^{-1}COD can be achieved when concentrating the AnMBR influent MWW from 1 to 10 by using FO membranes. This phenomenon was mainly attributed to lower methane losses achieved in the permeate when operating at higher organic load rates. In base of these results, any membrane technology contemplated to develop the DMF could also produce a similar beneficial effect on a posterior AnMBR system. However, since FO membranes show the higher COD recovery effectivity, coupling these membranes with AnMBR technology can be an attractive option for increasing the energy recovery and minimize the environmental impact of MWW treatment. In this scenery, MWW concentrations over 10 fold in the FO system are recommended for achieving economically-self-sufficient processes [1.101].

Regarding nutrients recovery, high TP captures (about 93 – 99%) are easily achieve when employing FO membranes for DMF of MWW, especially when using CTA ones [1.50; 1.51; 1.128]. Unfortunately, relatively low TN recoveries (up to 50 – 60%) have generally been reported [1.53; 1.70; 1.72; 1.129]. This is due to the extensive use of TFC membranes which show poor cations capture capacity. While CTA membranes have negligible charge, TFC membranes surface has carboxyl groups which serve as fixed ionic groups [1.130]. Consequently, these membranes confer a cation exchange feature dramatically reducing their capacity to retain cations [1.131]. In this regard, Ferrari [1.70] illustrated how high anions retentions (captures of SO_4^{2-} and PO_4^{3-} over 90%) can be achieved when operating a TFC membrane, while less than 20% of the cations (NH_4^+, K^+, Mg^{2+} and Ca^{2+}) where retained. On the other hand, Gao *et al.* [1.54] reported an almost complete capture of anions (SO_4^{2-} and PO_4^{3-}) when operating a CTA membrane, while K^+, Mg^{2+}, and Ca^{2+} rejections were about 60, 90 and 80%, respectively. Therefore, the low TN captures reported when filtering MWW by FO technology can be fixed by using CTA membranes, where NT retentions up to 96% have been reported [1.54; 1.128], although reductions in membrane permeability were also observed.

Other alternative would be modifying/functionalizing TFC membranes for improving cations capture efficiency. Indeed, several studies have proven that some modification on the membrane active layer with polyethylenimine can improve NH_4^+ recovery by 15 – 25% [1.129], while aquaporin modules incorporates a selective barrier which allows NH_4^+ recoveries comparable to CTA membranes without compromising the elevated water fluxes that characterize TFC membranes [1.128].

Concerning endocrine disrupting chemicals, Gao *et al.* [1.54] reported rejections between 60 – 100% for Bisphenol A, Estrone, 17β-estradiol and Estriol when filtering MWW whit a CTA FO membrane. These results coincide with the information reported by Cartinella *et al.* [1.132], who also achieved similar endocrine disruptors rejections (from 77 to 99.9%) when using an analogous FO membrane for the treatment of diverse influents. Nevertheless, significant concentrations of these substances can still be found in the effluent despite FO filtration, reporting Bisphenol A and 17β-estradiol contents in the permeate of 1388 and 30.3 ng L^{-1}, respectively [1.54]. Consequently, as some authors recognize [1.54], FO could not be enough for removing substances of this type.

1.8.3 Dynamic membranes

DM resource recovery potential is based on the characteristics of the cake layer developed onto the supporting material. Thus, since this cake layer can be extremely heterogeneous and dynamic, lower and more variable particulate material captures than those reported in MF or UF membranes should be expected [1.57].

Xiong *et al.* [1.57] reported effluent turbidities between 20 – 50 NTU when operating a DM for raw MWW treatment, which are in agreement with other DM studies treating wastewater [1.133]. Regarding OM and nutrients recovery, COD, TN, and TP recoveries of 51, 22.9, and 14.5%, respectively, were obtained by Xiong *et al*. [1.57]. Additionally, a limited but significant capture capacity of soluble compounds was also reported. SCOD, SN, and SP captures of 18.7, 8.7, and 5.8%, respectively, were reported in the cited study [1.57]. This soluble capture capacity was related to a mineralization or compounds adsorption on the formed cake layer. Other authors have reported better recoveries when dosing coagulants during raw MWW treatment, *e.g.* COD, NH_3-N, and TP captures around 71 – 81, 30.3, and 61.9%, respectively [1.58; 1.80]. Similarly to MF and UF membranes, the quality of the DM permeate is unable to meet the standards established by the European Union regulation on discharged water quality, reaching COD, SCOD, TN, NH_3-N, TP, and PO_4^{3-}-P concentrations in the permeate of about 76 – 113, 74, 36, 21 – 34, 2.2 – 5.3, and 4.9 mg L^{-1}, respectively [1.58; 1.80]. Therefore, similar alternatives than those cited for MF and UF membranes effluent treatment can also be proposed in this case to valorize the generated permeate.

Regarding OM capture, Xiong *et al.* [1.57] reported low OM concentrations (up to 1.2 gCOD L^{-1}) when directly self-forming a DM by raw MWW due to the significant loss of suspended material in the permeate. However, these authors noticed a significant increase towards higher particle sizes in the concentrated sludge not only by the capture of higher size particles but also to the aggregates formation during OM concentration [1.57]. Thus, sludge sedimentability may be improved enough to allow the AD treatment after a conventional sludge thickening. On the other hand, thanks to coagulant dosing, high COD retentate concentrations (from 4.5 up to 27 g L^{-1}) can be reached [1.58; 1.80], allowing their direct valorization via AD. Additionally, the reported bio-methane potential of the produced concentrate is similar or even higher than that reported for activated sludge [1.57; 1.58]. This phenomenon was related to the C:N ratio increase during DM filtration, which would enhance organics fermentation [1.134].

1.8.4 Side-effects of cleaning methodologies on resource recovery and permeate quality

Physical/chemical cleaning strategies used during filtration have shown to strongly influence the process, involving favorable and/or unfavorable side-effects on resources recovery and permeate quality. Hence, wastewater characteristics should also be considered when selecting the fouling control strategy employed in order to not only achieve energy-efficiency filtration processes but also to maximize resources recovery and achieve suitable permeate qualities.

1.8.4.1 Air-assisted membrane cleaning

Air injection during DMF has been identified as an unsuitable strategy due to favoring the development of heterotrophic microorganism. Lateef *et al.* [1.43] observed losses in OM of about 10 – 20% of total influent COD due to aerobic biodegradation when air sparging was employed for membrane scouring and/or tank mixing, even when operating at short SRTs (around 9 – 75 h). Similarly, Jin *et al.* [1.46] reported OM aerobic mineralization of about 19% of influent OM when employing intermittent aeration. On the other hand, alternative cleaning strategies, such as membrane vibration, may promote the re-aeration of the mixed liquor, resulting in unintentional biodegradation of the concentrated COD of up to 50% in only 22.5 h of operation [1.48], compared with the 8 days reported in batch mode when the system is not aerated [1.114]. Due to these results, the use of air sparging in DMF has been usually avoided [1.40; 1.43; 1.46; 1.57].

1.8.4.2 Coagulant dosing

In addition to improve filtration performance, the application of coagulant materials has shown important beneficial effects on resource recovery and permeate quality in DMF. COD recovery improvements up to 89 – 98% in MF/UF membranes have been reported when coagulant chemicals were used [1.21; 1.35; 1.46; 1.124]. This raise in OM recovery has been attributed to the capture of low size particulate materials in the formed flocs [1.44], including colloids and supra-dissolved solids [1.61]. Moreover, a portion of the soluble OM fraction can be adsorbed on the developed cake layer thanks to chemicals addition [1.46].

Coagulant dosing has also showed favorable operating effects during OM retention, allowing to achieve higher OM concentrations in less time [1.45; 1.46; 1.124]. Coagulants use would therefore allow operating the membrane tanks at lower SRTs, preventing OM mineralization. Furthermore, Hafuka *et al.* [1.135] studied the potential energy recovery of the captured OM when dosing PACl during DMF, reporting that this coagulant seems not to present any unfavorable effect on sludge biodegradability by AD, whenever keeping the Al^{3+} sludge concentration below 4.3 mg L^{-1}.

Important phosphorous recoveries can also be achieved when using chemicals. Indeed, coagulant dosing has been conventionally used in WWTP for phosphate precipitation [1.21]. Since membrane filtration presents higher particulate material retention efficiency than sedimentation, high phosphorous captures could be expected when combining coagulation and DMF, reporting TP recovery efficiencies up to 95 – 99% when using membranes or micro-sieving [1.21; 1.115; 1.136]. Unfortunately, TN capture is insensitive to coagulant application, reaching similar recovery efficiencies regardless using these chemicals [1.21; 1.61].

Organic coagulants have showed high OM recoveries with TP capture efficiencies around 75% [1.21], not affecting other chemical wastewater features (*e.g.* pH or conductivity) [1.61]. However, although modest pH reductions and conductivity increases have been reported by inorganic coagulants [1.61], similar or slightly higher OM recoveries have been reported by them [1.21; 1.61], strongly improving TP capture efficiency until achieving an almost complete recovery [1.21]. This difference in the phosphorous capture efficiency is due to the lack of metallic ions in organic coagulants [1.61; 1.136]. Thus, it could be recommended the application of inorganic coagulants from a nutrients recovery point of view. Among the inorganic coagulants, $FeCl_3$ and $AlCl_3$ salts seems to present similar performances [1.21; 1.30; 1.35], while slightly higher OM recoveries have been reported when using PACl [1.21; 1.35].

To further improving resource recovery, Hey *et al.* [1.21] proposed the use of flocculants together with coagulant dosing. In this study, anionic and cationic flocculants were tested, showing similar behaviors on OM capture efficiency, but improving TP capture efficiency up to 99%. Furthermore, since better filtration performances were observed when using PACl together with anionic flocculants, this combination was selected as the most suitable for DMF applicability. Thanks to coagulant or coagulant + flocculent dosing, COD, BOD, TN, and TP concentrations of 25 – 73, 11 – 17, 16 – 42, and 0.02 – 2.5 mg L^{-1}, respectively, can be achieved in the produced permeate [1.14; 1.21; 1.46]. Thus, the application of these chemicals could be considered alternatively to additional permeate post-treatments due to the achieved improvements on filtration performance. However, traces of metals or cations can be expected in the permeate depending on the dose used, which need to be taken into account according to the expected water fate [1.61].

1.8.4.3 Membrane cleaning chemicals

Similar to coagulants addition, other factor that may significantly affect the sludge and permeate quality is the use of cleaning reagents for membrane cleaning. Indeed, the use of CEBs during DMF can strongly limit the biological activity development during filtration, thereby avoiding OM losses and preventing biological fouling related problems [1.40; 1.43]. However, not all cleaning chemicals present the same effects.

Lateef *et al.* [1.43] showed that the use of NaOH and citric acid lightly influenced heterotrophs activity, while NaOCl resulted in an almost complete activity inhibition. On the other hand, the use of NaOCl in DMF can favor the solubilization of part of the particulate/colloidal material present in MWW, resulting in significant losses of soluble COD with the permeate [1.43]. Conversely, the use of NaOH and citric acid seems not to affect permeate quality, recording similar permeate COD concentrations after the use of these reagents than that obtained when pure water was employed for backwashing [1.43].

In addition to these results, the effect of CEBs on the biological permeate quality (*i.e.* pathogenic bacteria and virus removal) should also be considered since the use of chemicals may enhance the disinfecting capacity of the system, thereby improving the potential reuse of the recovered water. Thus, a suitable selection of chemicals and optimum adjustment of CEBs concentration and frequency is imperative to enhance both OM recovery and permeate quality [1.43].

1.9 Feasibility of DMF technology for MWW treatment

The feasibility of an emerging technology could be determined by its energy balance, investment/implementation and operation costs, required area for its application, and environmental impact, among others. Table 1.2 summarizes the results obtained by different authors when studying the energy balance of DMF of MWW using different membrane technologies.

Focusing on MF and UF membranes, DMF energy demands are usually estimated taking into account other similar MBRs. In this case, energy inputs between 0.30 – 0.60 kWh per m^3 of treated MWW have been reported [1.40; 1.43]. However, aeration may represent up to 60 – 70% of the total energy demand on MBRs operation [1.22; 1.40; 1.48]. Thus, since air-assisted membrane scouring could be replaced by other less energy-intensive cleaning strategies in DMF, different authors suggested that the energy cost of DMF could be significantly reduced [1.40; 1.43; 1.46]. Some authors have also estimated the DMF energy input from lab/pilot-scale systems, reporting around 0.08 – 0.09 kWh per m^3 of treated MWW when using coagulant and intermittent aeration [1.39; 1.46], while values around 0.40 – 0.41 kWh per m^3 of treated MWW have been reported when using coagulant, micro-sieving, and air sparging [1.22; 1.65]. On the other hand, thanks to OM recovery, energy recoveries between 0.19 – 1.40 kWh per m^3 of treated MWW have been estimated in different studies [1.22; 1.36; 1.43; 1.45; 1.65; 1.124]. Additionally, this energy output can be significantly improved if considering nutrient recovery and the equivalent cost for inorganic fertilizers production [1.124]. Heat requirements during AD would also be completely covered according to some studies [1.22; 1.65], achieving even heat energy surplus. These promising results make numerous authors agree in that neutral energy processes or even net energy producer processes can be achieved through DMF of MWW when considering the overall process energy balance [1.22; 1.39; 1.46; 1.65; 1.124]. These results clearly overcome the energy input of current aerobic treatments which may represent from around 0.3 to 1.9 kWh per m^3 of treated MWW [1.22; 1.45]. Thus, from an energy point of view, DMF could be considered as an attractive alternative for MWW treatment.

Table 1.2. Energy feasibility of DMF for MWW.

Membrane Type	Treated influent	Operation	Energy input (kWh per m³ of treated water)	Energy output (kWh per m³ of treated water)	Total energy demand (kWh per m³ of treated water)	Ref.
Microfiltration	Raw MWW	Coagulant dosing combined with micro-sieving and membrane air scouring.	0.40			[1.22]
		Coagulant dosing and intermittent air-backwashing.	0.08	0.11	-0.03	[1.39]
		Air mixing combined with CEBs.	< 0.40[a]	0.14 – 0.50		[1.43]
		Coagulant and powdered activated carbon dosing.		0.19		[1.45]
		Coagulant dosing and intermittent aeration.	0.09	0.10	-0.01	[1.46]
		Coagulant dosing combined with micro-sieving and membrane air scouring.	0.41	1.3		[1.65]
		Coagulant and powdered activated carbon dosing.		0.26 – 0.39		[1.124]
	PSE	Vibrations combined with mechanical mixing and periodical CEBs.	< 0.40[a]			[1.40]
Ultrafiltration	PSE	Air sparging and permeate backwashing.	0.34	0.50		[1.36]
Forward osmosis	Raw MWW	Coagulant dosing combined with micro-sieving. Seawater as draw solution.	0.41	1.5		[1.65]
		Synthetic MWW. Water recovery in the FO dispositive from 50 to 90%.		1.0 – 6.9		[1.101]
Dynamic membrane	Raw MWW	Three-layer stainless steel mesh of 25 µm as supportive material. External physical cleaning with air backwashing and surface brushing each 48 h.	0.01	0.10	0.09	[1.57]
		Dacron mesh of 61 µm as supportive material. Continuous coagulant dosing to perform the DM auto-formation and control fouling development. External physical cleaning with tap water after reach a TMP of 35 kPa.	0.24 – 4.57[b]	0.13 – 4.33[b]	0.11 – 0.24[b]	[1.58]

[a] Estimated from MBRs.
[b] Including AD and nutrients recovery equipment energy impacts.
MWW: Municipal wastewater.
PSE: Primary settler effluent.

In addition to the energy balance, other important aspects need to be considered to assess DMF feasibility, such as area requirements or economic cost. Regarding footprint, Hey *et al.* [1.21; 1.65] estimated that the implementation of a MF system for MWW treatment would reach about 0.0075 m^2 per population equivalent (PE), ascending up to 0.046 m^2 per PE when considering all additional equipment (*i.e.* screening, sand-trap, micro-sieving, coagulant and equalization tanks, AD, etc.). Since lower area requirements than those reported by conventional WWTP facilities (about 0.107 and 0.228 m^2 per PE) have been reported [1.21], the up-grading of existing WWTP without increasing footprint requirements could be achieved. On the other hand, according to theoretical calculations performed by Mezohegyi *et al.* [1.48], economic improvements can be expected from a realistic semi-continuous OM concentration by membrane filtration. Considering the capital expenses, Ravazzini [1.61] estimated a DMF investment implementation cost of 0.05 – 0.06 € per m^3 of influent water, which agrees with the range reported by Baker [1.137] considering different UF membrane plants. Regarding operational expenses, Rulkens *et al.* [1.114] estimated a MWW treatment cost of 0.24 € per m^3 of treated water, whilst Ravazzini [1.61] reported values between 0.20 and 0.40 € per m^3 of treated water, both studies considering UF membranes. Since the overall current MWW treatment cost could be estimated between 0.57 – 1.02 € per m^3 of treated water [1.61], the DMF concept may be considered as an attractive alternative to reduce MWW treatment cost. It is also important to highlight that all the equipment required for implementing a DMF process is commercially available [1.65]. Nonetheless, further studies focused on environmental and economic impacts of used equipment and materials (*i.e.* LCA and LCC analysis) are required in order to realistically evaluate the feasibility of DMF from an environmental and economic point of view.

Regarding the use of FO and DMs for MWW treatment, to the extent of authors knowledge, few studies have been performed so far focused on energy or economic feasibility. FO involves low energy demands since the DS is the main responsible driving force [1.17; 1.73]. However, the regeneration of this DS can be an energy intensive process, which is generally conducted by RO or membrane distillation [1.73; 1.138]. In this case, energy requirements and environmental impacts similar (or even higher) to those reported in water desalination have been estimated for MWW concentration by FO-RO membranes [1.73], although recognizing the limitations in this comparison. Regarding FO-RO treatment costs, Vinardell *et al.* [1.139] reported values around 0.81 €

per m^3 of treated water when water recovery was restricted to 50%, raising until 1.01 and 1.27 € per m^3 of treated water when increasing water recovery to 80 and 90%, respectively. Using seawater or concentrated fertilizers as DS have been considered potential alternatives to avoid regeneration [1.73]. For instance, Hey *et al.* [1.65] reported energy demands about 0.41 kWh per m^3 of treated MWW when combining FO filtration with a micro-sieving pre-treatment, thus resulting in competitive results compared to MF/UF. Moreover, slight higher energy recoveries were reported when comparing with MF thanks to an increased COD capture efficiency [1.65], achieving an energy output potential of 1.5 kWh per m^3 of treated MWW. The area requirements for implementing DMF with FO membranes are similar to those reported for MF or UF, since analogous membrane areas are required. Specifically, footprints between 0.039 – 0.050 m^2 per PE have been estimated for FO technology [1.65].

Finally, promising results have been reported when considering DMs for direct filtration of MWW. In this respect, when considering OM recovery and filtration energy requirements, total energy costs of about 0.24 kWh m^{-3} of treated water have been reported for raw MWW treatment by DMs [1.58], achieving even an energy surplus of around 0.09 kWh m^{-3} when aeration was avoided and chemicals dosed [1.57]. Additionally, although not proper economic analysis has been performed yet, promising results can be expected due to the lower cost and maintenance requirements of DMs compared to MF and UF [1.57; 1.85; 1.86]. Thus, DMs can be considered as an attractive alternative to implement DMF technology, although further studies are needed to determine their potential applications.

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CHAPTER 2. Thesis Scope and Outline

The direct membrane filtration (DMF) of municipal wastewater (MWW) can be considered as an interesting alternative to enhance resource recovery within the MWW treatment field, bringing the sector closer to circular economy standards. This alternative treatment approach can be especially remarkable when aiming to up-grade facilities in operation since not important adjustments are required neither in the treatment scheme nor in the existing infrastructure. However, membrane fouling remains a major issue to tackle in order to achieve feasible long-term membrane operation.

The main aim of this PhD thesis was to determine the most suitable membrane technology (microfiltration, ultrafiltration or dynamic membranes) for direct filtration of municipal wastewater. Additionally, the type of influent entering the system (raw wastewater or primary settling effluent) and the operating conditions (solids concentration range, transmembrane flux, and dynamic membrane supporting material pore size, depending on the operated membrane) was studied in order to determine effective design and operational strategies to minimize fouling while maximizing resource recovery in long-term operation.

Besides conducting different batch experiments at lab-scale, a membrane-based pilot plant equipped with commercial membrane modules treating MWW from a full-scale MWW facility was operated for more than 3 years. Therefore, this work provides of realistic and useful information on the performance of DMF for MWW treatment, as well as delivering guidelines for full-scale implementation of the evaluated technology.

This PhD thesis has been developed in the framework of a national project entitled *'Aplicación de la tecnología de membranas para potenciar la transformación de las EDAR actuales en estaciones de recuperación de recursos / Application of membrane technology to enhance the transformation of current WWTPs into Wastewater Resource Recovery Facilities'* (project code: CTM2017-86751-C2-1-R) which was founded by the Spanish ministry of economy, industry and competitiveness (MINECO). The document is presented as a compendium of papers (a total of six, three already published and three prepared for submission), each one conforming an individual chapter. According to the above cited main objective, each chapter focuses on the following specific aspects:

Chapter 4. *Assessing the most suitable methodology to determine sludge filterability from different municipal wastewater treatment systems*

Sanchis-Perucho, P., Moyano Torres, K.M., Ferrer, J., Seco, A., Robles, Á. Assessing the Most Suitable Methodology to Determine Sludge Filterability from Different Municipal Wastewater Treatment Systems. Prepared for submission.

The capability of different methodologies to determine sludge filterability and estimate sludge filtration resistance from different membrane-based MWW treatment systems was evaluated. Capillary suction time, time to filter, and specific resistance to filtration methods were studied using three different sludge sources: aerobic activated sludge, supernatant from a primary settler further concentrated by ultrafiltration membranes, and digestate from the anaerobic co-digestion of microalgae and primary sludge.

Chapter 5. *Direct membrane filtration of municipal wastewater: studying the most suitable conditions for minimizing fouling rate in commercial porous membranes at demonstration scale*

*Sanchis-Perucho, P., Aguado, D., Ferrer, J., Seco, A., Robles, Á. Direct Membrane Filtration of Municipal Wastewater: Studying the Most Suitable Conditions for Minimizing Fouling Rate in Commercial Porous Membranes at Demonstration Scale. Membranes **2023**, 13, 99. https://doi.org/10.3390/membranes 13010099.*

Ultra- and micro-filtration membrane performance was compared when treating two influent sources (raw wastewater and primary settling effluent) at two operating suspended solids concentrations (around 1 and 2.6 g L^{-1}). The effectivity of two physical fouling control strategies (continuous air-assisted membrane scouring and periodical permeate backwashing) for membrane fouling mitigation/removal was evaluated in the short- and middle-term operation. The origin of fouling (organic or inorganic) was characterized by analyzing membrane permeability recovery after acid and basic chemical cleaning, while fouling mechanisms were studied using theoretical literature mathematical models.

Chapter 6. ***Evaluating resource recovery potential and process feasibility of direct membrane ultrafiltration of municipal wastewater at demonstration scale***

Sanchis-Perucho, P., Aguado, D., Ferrer, J., Seco, A., Robles, Á. Evaluating Resource Recovery Potential and Process Feasibility of Direct Membrane Ultrafiltration of Municipal Wastewater at Demonstration Scale. Prepared for submission.

Ultrafiltration membrane performance was evaluated at higher solids concentrations (about 6 and 11 g L^{-1}) for both raw MWW and primary settling effluent (supernatant) treatment. Resource recovery potential was assessed for the experimental conditions evaluated, conducting preliminary energy, economic, and carbon footprint analysis.

Chapter 7. ***Building a simple and generic filtration model to predict membrane fouling in the long-term when treating municipal wastewater***

Sanchis-Perucho, P., Harmand, J., Feddaoui-papin, A., Aguado, D., Robles, Á. Building a Simple and Generic Filtration Model to Predict Membrane Fouling in the Long-Term when Treating Municipal Wastewater. Prepared for submission.

A qualitative filtration model was developed to dynamically predict membrane fouling in DMF technology. Filtration results from chapters 4 and 5 were used to calibrate the model. The capacity of the model to capture the dynamics on transmembrane pressure was evaluated within the whole range of operating conditions evaluated. Moreover, a global sensitivity analysis was conducted to identify the most influential parameters on model outputs. Model uncertainty was also assessed.

Chapter 8. ***Dynamic membranes for enhancing resources recovery from municipal wastewater***

*Sanchis-Perucho, P., Aguado, D., Ferrer, J., Seco, A., Robles, Á. Dynamic Membranes for Enhancing Resources Recovery from Municipal Wastewater. Membranes **2022**, 12, 214. https://doi.org/10.3390/membranes12020214.*

The use of dynamics membranes for treating primary settling effluent (supernatant) from a full-scale WWTP was studied. Dynamic membrane self-forming capacity, resource recovery potential, and membrane fouling was evaluated under different operating conditions: i) two supporting material alternatives (one or two layers with 1 µm of pore size), ii) two transmembrane filtration fluxes (15 and 45 LMH), iii) three suspended solids concentrations (above 2, 5 and 9 g L^{-1}), and iv) dosing coagulant. Preliminary energy, economic and carbon foot-print analysis were performed to assess process feasibility.

Chapter 9. *Evaluating the Feasibility of Employing Dynamic Membranes for the Direct Filtration of Municipal Wastewater*

*Sanchis-Perucho, P., Aguado, D., Ferrer, J., Seco, A., Robles, Á. Evaluating the Feasibility of Employing Dynamic Membranes for the Direct Filtration of Municipal Wastewater. Membranes **2022**, 12, 1013. https://doi.org/10.3390/membranes12101013.*

Dynamic membrane performance was studied when treating raw MWW. The influence of four supporting material alternatives (1 and 5 µm pore size with one or two layers each) on dynamic membrane self-forming capacity, resource recovery, and membrane fouling propensity was evaluated. Preliminary energy, economic and carbon foot-print analysis were performed to assess process feasibility. The results were compared with those obtained in chapter 7.

Finally, chapter 10 synthetize and provide an overall discussion of the key findings from this PhD research, while chapter 11 compiles and summarizes the most relevant conclusions. A supplementary abstract section written in Spanish (Appendix) is included at the end of this document according to Valencia university regulations.

*Journal Editor has authorized the use of cited articles in this dissertation document.

CHAPTER 3. Materials and Methods: Description of the Demo-scale Membrane-based Plant Operated in this Thesis

3.1 Membrane based pilot plant

This PhD work was mainly conducted using a membrane-based pilot plant within the framework of the national project CTM2017-86751-C2-1-R. It consisted of three independent membrane tanks each one fitted with a different membrane system: microfiltration (MF) (TERAPORETM 5000, commercial membrane module, 0.4-µm pore size, filtration area 18 m^2), ultrafiltration (UF) (PULSION® Koch Membrane Systems, commercial membrane module, 0.03-µm pore size, filtration area 43.5 m^2), and dynamic membrane (DM) (NITEX® SEFAR, two flat open monofilament polyamide woven meshes with 1 and 5 µm of average pore size tested as supporting material, filtration area 2 m^2). Each membrane module was equipped with one screw pump (PCM, EcoMoineau™) for vacuum filtration and influent feeding. Each membrane tank had a clean-in-place (CIP) tank (100, 400 and 100 L for the MF, UF and DM CIP tanks, respectively), to store the generated permeate and allow integrated sampling and membrane backwashing during continuous operation when necessary. Air-assisted membrane scouring was used to mitigate fouling during filtration, using a blower (G-BH7, Elmo Rietschle) to inject air to the bottom of each membrane tank. Three additional mixing pumps (PCM, EcoMoineau™) continuously recirculated the content of each membrane tank to ensure complete mixing thus avoiding solids stratification. Finally, two storage tanks (80 L each) were deployed for membrane chemical cleaning purposes: basic tank (BT) which contained the basic reagent ($NaOCl$ in this case), and acid tank (AT) which contained the acid reagent (citric acid in this case). Both tanks were connected to the MF and UF permeate pumps to allow the backwashing with these chemicals when necessary. Fig. 3.1 shows a schematic flow diagram of the pilot plant and Fig. 3.2 shows a full frontal and side picture of it.

The pilot plant was continuously fed with municipal wastewater (MWW) coming from the “Conca del Carraixet” full-scale MWW treatment facility (Alboraya, Spain). Two different influent sources were used in this work: (1) raw MWW coming from a pre-treatment step consisting of screening, sieving, desanding, and degreasing); and (2) effluent (supernatant) from the primary settler (PSE). Regardless of the influent used, a 0.5 mm screen size roto-filter was installed as additional pre-treatment step to protect the membrane modules. The pre-treated influent was collected in an equalization tank (745 L) to feed each membrane tank with the same MWW.

3.2 Automation and control

The pilot plant was designed with a high level of automation. Numerous on-line sensors and automatic equipment were installed to allow full control of the process (see Fig. 3.1). Main on-line sensors were: four pH-temperature sensors (InPro3100/120/PT100, Endress+Hauser) installed in the ET, MF, UF, and DM tanks; nine level sensors (Cerabar PMP11, Endress+Hauser), one for each tank (ET, MF, UF, DM, CIP-MF, CIP-UF, CIP-DM, BT, and AT); three liquid pressure sensors (IP65, Druck) to control the transmembrane pressure (TMP) in each membrane tank; six liquid flow meters (Picomag, Endress+Hauser), each one associated with a feeding and permeate screw pump; three air flow meters, one for each membrane tank (MF, UF and DM); four solids sensors (LXV424.99.00100, Hach) to control the solids concentration in the ET, MF, UF and DM tanks; and two turbidity sensors (LXV424.99.00100, Hach) to control the turbidity level in the CIP-MF and CIP-DM tanks. On the other hand, the pilot plant was equipped with the following actuators: seven frequency converters (SINAMICS G120C, Siemens), six to control the liquid flowrate of each liquid pump and one to control the air flowrate supplied by the blower; three automatic needle valves to accurately control the air flow-rate distributed to each membrane tank; and eleven on-off control valves to avoid liquid fluxes during relaxation and backwashing stages or when cleaning any membrane tank.

All these instruments were connected to a programmable logic controller (PLC) for proper multi-parametric control and data acquisition. The PLC was also connected via Ethernet network to a PC from which a SCADA system centralized all the information collected by the instrumentation and facilitated their supervision and control. Due the significant number of sensors and actuators installed, the control script was based on multiple control loops which consisted of classic PID and on-off controllers designed to act on the main operational variables (*e.g.* liquid and air flow rates, TMP control, level measures in each tank, etc.) to reach the established set-points. Further specifications concerning operating conditions used for each membrane study are described in the corresponding chapter.

3.3 Analytical methods and calculations

The feasibility of the above described pilot plant was studied by determining resource recovery capacity, and energy, economic and carbon footprint impacts. Solids, total and soluble chemical oxygen demand (COD and SCOD), total nitrogen (TN) and total phosphorus (TP) were determined according to standard methods. 0.45-mm pore size glass fibre membrane filters (Millipore, Merck) were used to produce the soluble fraction of collected samples. The particle size distribution of treated influents and concentrated sludge were determined by a laser granularity distribution analyser (Malvern Mastersizer 2000; detector range of 0.01 to 1000 μm).

Filtration energy balance was assessed considering the energy demands of the equipment used (permeate pump, mixing pump and blower) and the potential energy production of the recovered organic matter. Since other energy requirements would also be necessary when operating conventional systems (feeding pumping, sludge to anaerobic digestion pumping, etc.), only the specific equipment required in the DMF process was considered. Further specifications concerning each membrane performance assessment are described in its corresponding chapter.

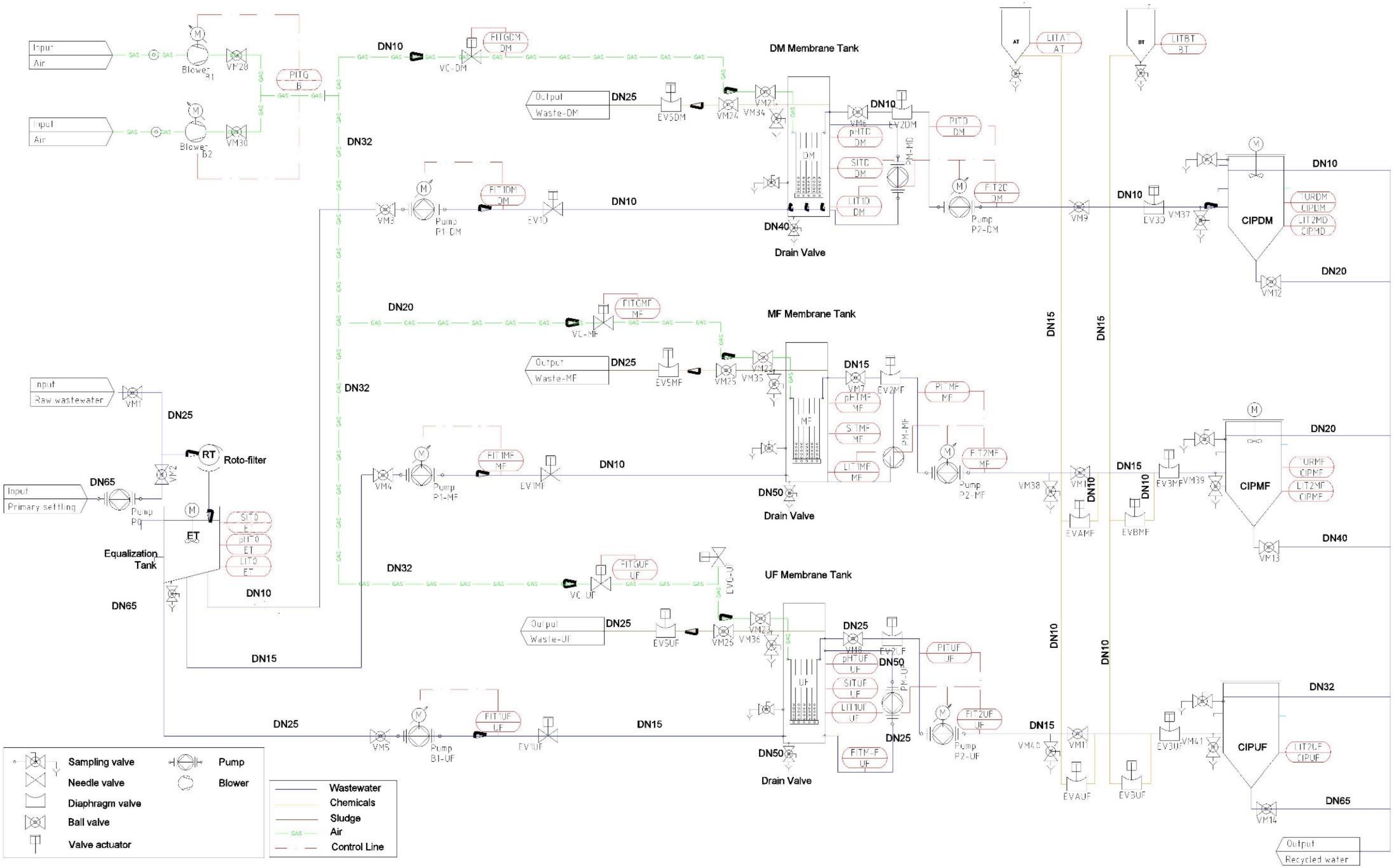


Figure 3.1. Flow diagram of the membrane pilot plant.

Figure. 3.2. Complete picture of the membrane pilot plant: (a) side face and (b) frontal face.

CHAPTER 4. Assessing the Most Suitable Methodology to Determine Sludge Filterability from Different Municipal Wastewater Treatment Systems

Abstract

This study aimed to evaluate the capability of different methodologies for determining sludge filterability and estimate filtration resistance during real filtration processes. Three filterability methods were applied during this study: capillary suction time (CST), time to filter (TTF), and specific resistance to filtration (SRF). These methods were evaluated using three different sludge sources: aerobic activated sludge, supernatant from a primary settler further concentrated by ultrafiltration membranes (PSE), and digestate from the anaerobic co-digestion of microalgae and primary sludge. These sludge sources were taken from systems treating municipal wastewater entering to a full-scale wastewater treatment facility. The capability of CST, TTF and SRF to estimate total suspended solids (TSS) and soluble microbial products (SMP) concentrations was also assessed, while validating the results obtained with the real filtration process of the operated membrane-based systems. The results suggested that both TSS and SMP concentrations significantly affect filterability. However, each sludge filterability was mainly dominated by one of these parameters (TSS or SMP), being the biological sludge more influenced by the SMP content, while the PSE filterability was mainly controlled by the TSS concentration. SRF method resulted in poor correlations between filterability and TSS and SMP concentrations, especially regarding SMP. CST method resulted in good correlations for all treated sludge regardless the TSS and SMP concentration. However, when treating sludge without an important biological activity (*e.g.* PSE), TTF method was identified as the best option due to its better correlation with the experimentally determined sludge filtration resistance.

Keywords

Aerobic activated sludge; Anaerobic digestate; Direct membrane filtration; Filterability; Microalgae; Municipal wastewater.

Sanchis-Perucho, P., Moyano Torres, K.M., Ferrer, J., Seco, A., Robles, Á.

Prepared for submission

4.1 Introduction

Municipal wastewater (MWW) treatment paradigm has changed in the last years since MWW is nowadays considered as a potential source of essential resources (reclaimed water, energy, nutrients, etc.) [4.1]. Thus, to recover MWW resources, numerous treatment alternatives are being proposed to transform current municipal wastewater treatment plants (WWTP) in new resource recovery facilities. For instance, numerous membrane-based systems aiming to improve MWW treatment field can be found in recent literature, such as aerobic membrane bioreactors [4.2], anaerobic membrane bioreactors [4.3] also including alternative treatment platforms and configurations [4.4], membrane photo-bioreactors [4.5; 4.6], direct membrane filtration configurations using porous [4.7] or dynamic membranes [4.8; 4.9], and other advanced systems for water and nutrient recovery [4.10–4.12]. Unfortunately, membrane fouling represents one key issue in any membrane-based system. Indeed, membrane fouling control usually represent a major cost in these systems, thus their optimization is one of most relevant aspects to consider in order to achieve feasible treatments [4.13; 4.14]. In this respect, adequately estimating fouling propensity and filtration resistance depending on operating conditions and treated mixed liquor can provide of valuable information to properly select and optimize fouling control for each membrane-based treatment system.

The filterability of a sludge sample is defined as its capacity to release its water content from its sludge matrix in a temporal lap [4.15], providing therefore of a numerical estimation of the resistance to filtration of the treated sludge. The higher the filterability value (*i.e.* more fluid separated from the initial matrix at same driving force and time), the lower the resistance of the treated sludge to be filtered, then presumably requiring of lower driving force intensities to perform the process. Different methodologies can be found in literature to determine sludge filterability, such as capillary suction time (CST) [4.16; 4.17], time to filter (TTF) [4.18; 4.19] or specific resistance to filtration (SRF) [4.16; 4.20]. However, the results obtained from applying these methods could not provide of useful information depending on the sludge to be evaluated, mainly due to: i) the large amount of possible fouling pollutants present in MWW, ii) the extremely complex and changing fouling mechanisms that may occur in the system, and iii) the different configuration and/or driving force applied in the system. Generally, membrane fouling can be correlated to the solids concentration of the sludge treated, playing a dominant role in several systems [4.21; 4.22]. Other pollutants, such as soluble microbial products (SMPs), have also been extensively reported in several systems as important fouling promotors [4.22; 4.23]. In consequence, further studies are required to determine the most convenient filterability methodology to be applied regarding the sludge to be treated, identifying their capacity to predict sludge fouling propensity regarding its characteristics.

The aim of this study was to identify main constrains and benefits of applying different filterability methodologies when treating different MWW sludge, proposing the best alternative regarding the operating conditions and sludge sources evaluated in this work. Three filterability methods (*i.e.* CST, TTF and SRF) were applied to three different sludge sources: aerobic activated sludge, supernatant from a primary settler further concentrated by ultrafiltration membranes (PSE), and digestate from the anaerobic co-digestion of microalgae and primary sludge. Sludge filterability was correlated with TSS and SMP concentrations in the evaluated sludge. Finally, in order to validate the applicability of the evaluated filterability methods in real filtration conditions regarding changes on TSS and SMP concentrations, the results from PSE and anaerobic digestate were correlated with the transmembrane pressure (TMP) monitored in the corresponding membrane-based systems: direct membrane filtration of primary settler effluent and anaerobic co-digestion of microalgae and primary sludge, both equipped with UF membranes.

4.2 Material and Methods

4.2.1 Sludge source

Sludge sampled were taken from three systems treating MWW coming from the full-scale “Conca del Carraixet” WWTP (Alboraya, Spain).

Aerobic activated sludge was obtained from the activated sludge system of the full-scale plant. To increase TSS concentration when necessary, some samples were introduced to a bench-scale aerobic reactor fed with primary settler supernatant from the full-scale process. This also helped to increase SMP secretion in some samples applying famine regime.

PSE samples were taken from a pilot plant equipped with a commercial UF membrane module (PULSION® Koch Membrane Systems, 0.03-μm pore size, filtration area 43.5 m^2) that treated the supernatant from the primary settler of the full-scale system, concentrating its particulate faction. This pilot plant used air scouring as fouling control strategy during filtration. Further information regarding this pilot plant can be found in [4.7].

Anaerobic digestate was taken from an anaerobic co-digestion membrane bioreactor pilot plant treating primary sludge from the full-scale primary settler and microalgae biomass. The anaerobic co-digester was also equipped with a UF membrane module (PURON® Koch Membrane Systems, 0.03-μm pore size, filtration area 0.44 m^2) assisted by air scouring for membrane scouring. Further information regarding described pilot-plant can be found in [4.24].

4.2.2 Filterability methods

Three methods were evaluated in this work to determine sludge filterability: CST, TTF, and SRF.

CST and TTF methods evaluate filterability directly from the time required to filter a specific volume of sludge (value expressed as time units). CST was performed according to standard methods [4.15], employing a 7x9-cm flat Whatman™ 17-grade chromatography papers (thickness of 0.92-mm). TTF was also performed according to standard methods [4.15], using a circular 90-mm diameter Whatman® 2-grade filters (thickness of 0.19-mm with a pore size of 8 μm).

To determine sludge filterability through SRF method some calculations are required. According to Darcy's law, the volume of fluid filtered in a porous medium can be expressed as follows [4.25]:

$$\frac{dV}{dt} = \frac{PA^2}{\mu(rcV + R_M A)} \tag{4.1}$$

Where P is the pressure drop, A is the filter area, μ is the medium viscosity, r is the specific resistance to filtration (SRF value), c the sludge solids concentration, and R_M the filter resistance (initial resistance to filtration).

Eq. 4.1 can be arranged as a linear equation (see Eq. 4.2). Thus, SRF and R_M values can be determined from the slope and intercept, respectively, of a linearly correlation of t/V as a function of the volume filtered (V) (see Eq. 4.3 and Eq. 4.4).

$$\frac{t}{V} = \frac{\mu r c}{2\,PA^2}\,V + \frac{\mu R_M}{PA} \tag{4.2}$$

Thus, defining the linear equation slope as φ and the intercept as λ, it can be concluded that:

$$SRF = \varphi\frac{2\,PA^2}{\mu c} \tag{4.3}$$

$$R_M = \lambda\frac{PA}{\mu} \tag{4.4}$$

A sludge volume of 100 ml was filtered to estimate SRF and R_M, recording the time necessary to filter a volume from 10 to 50 ml with an increasing step of 10 ml (five points in total). Same filters used in the TTF method were employed in this case.

4.2.3 Sampling and complementary analysis

Sludge samples from all three systems were taken twice a week, performing three replicas of each analysis. In addition to filterability methods (CST, TTF and SRF), TSS concentration, SMP content, and medium viscosity were determined. TSS concentration was determined according to standard methods [4.15]. Sludge viscosity was obtained by a Cannon-Fenske viscometer (Series 50, COMECTA®). SMP effective content was attributed to protein and carbohydrate concentrations only. A commercial total protein kit (Micro Lowry Peterson's Modification, Sigma-Aldrich) was used to determine the protein content, while carbohydrates were determined by Dubois method [4.26]. Soluble fraction of treated sludge was obtained by filtering each sample with 0.45-mm pore size glass fibre membrane filters (Millipore, Merck). Depending on the sludge source, the correlation coefficient (R^2) and/or the mean absolute percentage error (MAPE) were employed to determine the accuracy of each methodology to predict TSS and SMP concentrations.

4.3 Results and Discussion

4.3.1 Sludge filterability determination

Fig. 4.1 and Fig. 4.2 show the resulting filterability obtained from the application of each method to each sludge source. In general, both TSS and SMP concentrations seem to have a significant impact on sludge filterability. However, significant differences were obtained when raising these two variables depending on sludge source and methodology employed. Overall, the aerobic sludge resulted in a higher filterability. This was related to the presence high size biomass floccules in activated sludge due to the adhesion of filamentous bacteria and other microorganisms [4.27]. Thus, high size paths can be created among particulate material, allowing water to easily flow across the sludge matrix. On the other hand, high filtration resistances were obtained for the anaerobic digestate (mixture of microalgae and primary sludge). Microalgae cultures are identified in literature as difficult-to-filter liquors due to the secretion of a large amount of polymeric substances under stress conditions [4.24; 4.28]. Thus, poorer filterability even than conventional anaerobic digestate may be expected in co-digestion systems fed with microalgae. Similarly, PSE sludge is reported as a high-fouling promotor in porous membrane systems [4.7; 4.29], thus low filterability values are also expected in this case.

Sludge viscosity was also overall influenced by TSS and SMP concentrations (see Fig. 4.3). However, TSS concentration seemed to mainly control medium viscosity, showing a stronger and more consistent effect than SMP. Indeed, the SMP content displayed slight influences on the medium viscosity, especially for the anaerobic digestate, where even a null correlation could be concluded. Additionally, similar viscosity values were obtained in this study as raising the TSS and SMP concentration regardless the sludge source (see Fig. 4.3). These results suggested that the medium viscosity by itself may not provide of sufficient information to predict the resistance to filtration of the evaluated sludge samples, although it could be used to estimate the medium filterability when filtering the same sludge source.

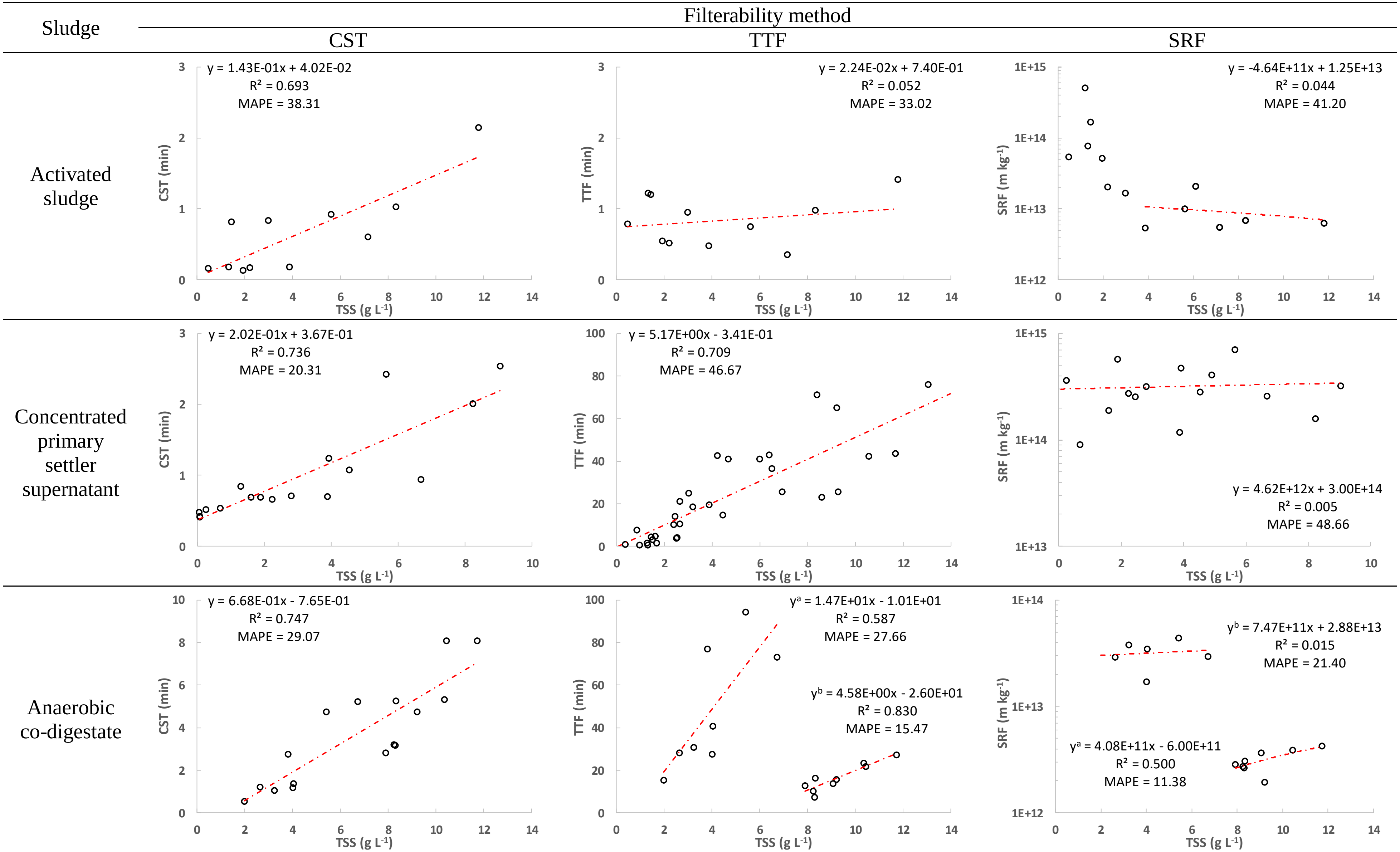


Figure 4.1. Effect of TSS concentration on sludge filterability determined by CST, TTF, and SRF methods. Operating SMP range between [113 – 302]^a and [80 – 190]^b mgDQO L^{-1}.

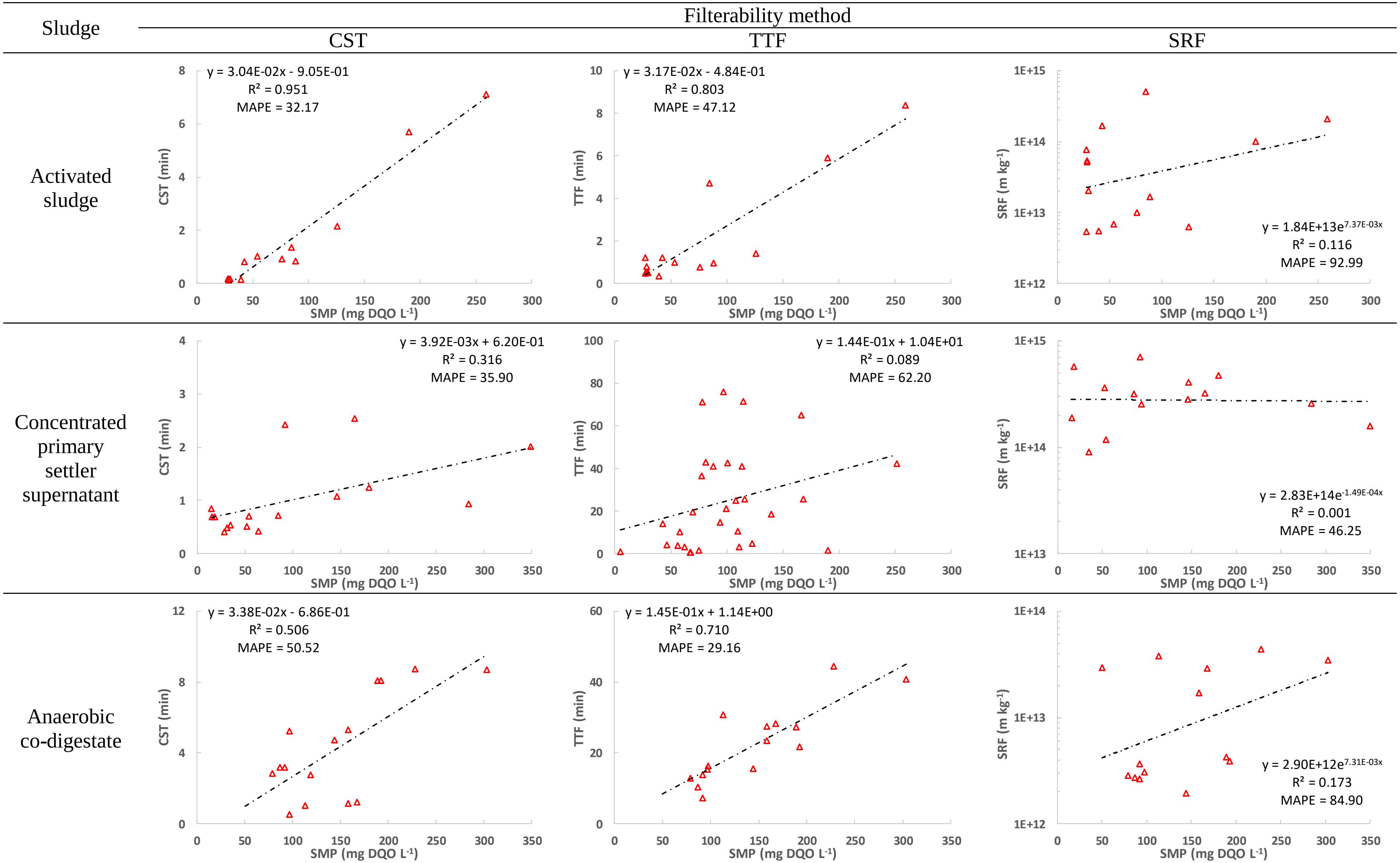


Figure 4.2. Effect of SMP concentration on sludge filterability determined by CST, TTF, and SRF methods.

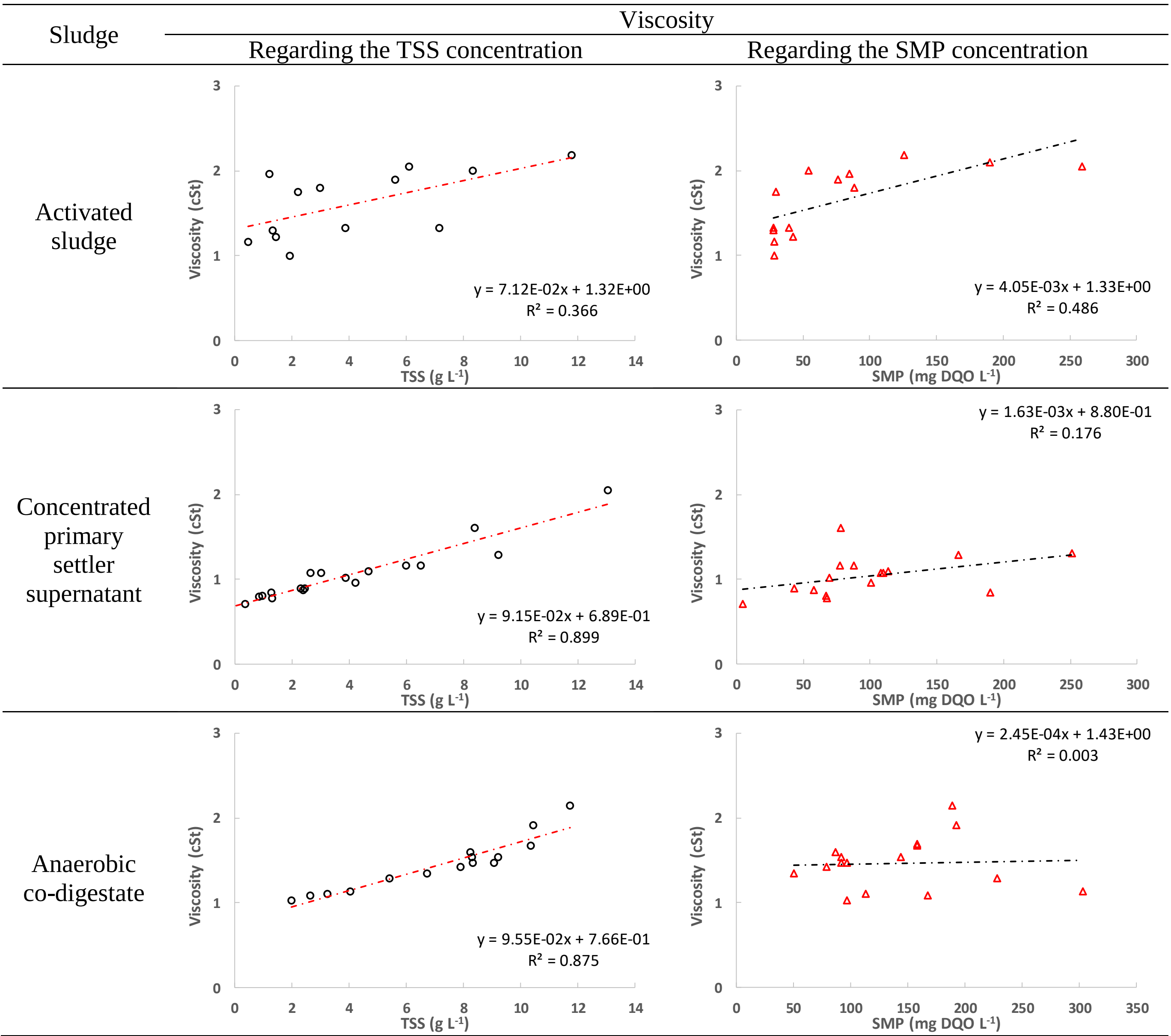


Figure 4.3. Effect of TSS and SMP concentrations on sludge viscosity.

4.3.1.1 Effect of TSS concentration on sludge filterability

In general, a direct relationship was observed between TSS concentration and filterability, although it was differently affected by the sludge source (see Fig. 4.1). Regarding aerobic sludge, a slight influence of TSS on filterability could be concluded within the TSS range tested. As previously commented, this could be related to the presence of high size biomass floccules in this kind of sludge, creating high apertures for the water to flow between particulate material regardless solids concentration. In this respect, the CST method showed a slight slope, indicating that high changes in the TSS are required to affect sludge filterability. On the other hand, TTF and SRF methods resulted in negligible relationships between TSS concentration and calculated filterability. In addition, the SRF results, which includes TSS concentration as divisor in its calculation normalizing therefore the result (see Eq. 4.3), seems also to indicate that there is not a good relation between sludge filterability and TSS concentration when operating at low TSS concentrations (under about 3 g L^{-1}). However, a SRF stabilized value was reached above 3 g L^{-1}, indicating that a proper relation can be obtained as increasing the TSS concentration. Considering the method's accuracy (lower MAPE values) and sensibility (higher slopes), CST methodology was identified as the most recommended method to evaluate aerobic sludge filterability, within the conditions evaluated in this study.

Regarding PSE sludge, a clearer filterability increasing trend was appreciated as TSS concentration raised when applying both CST and TTF methods. SRF method also showed relatively stable values for all the tested TSS range, denoting a proper relationship between TSS concentration and sludge filterability. Similarly to activated sludge, the CST method resulted in the best precisions when determining the sludge filterability (R^2 and MAPE values of 0.736 and 20.31%, respectively), although acceptable results were also observed for TTF (R^2 and MAPE values of 0.709 and 46.67%, respectively). Since TTF method showed the highest sensibility (*i.e.* highest slope value), it was considered as a strong alternative to evaluate filterability for this kind of sludge.

Finally, a more complex trend was obtained when treating the anaerobic co-digested sludge. As Fig. 4.1 shows, results from both TTF and SRF methods split in two differenced clusters. In this respect, during the anaerobic co-digester operation, the low TSS concentration period coincided with a biomass stress period, reaching the highest SMP concentrations in the system. Consequently, the clear two clusters observed when representing sludge filterability vs TSS concentration would be caused by this significant SMP concentration difference which would also denote that SMP content strongly affects sludge filterability. Despite this important SMP influence on TTF and SRF results, the CST method revealed a continuous direct lineal trend involving all collected data. Since relatively good precisions were displayed by the CST method (regression index and MAPE values of 0.747 and 29.07%, respectively), this methodology could be recommended for estimating sludge filterability from TSS concentration, and vice versa. However, for stable operating periods where the biomass does not suffer stress that force the segregation of more SMP, TTF methodology could be a solid alternative for determining medium filterability with also higher precisions (R^2 and MAPE values of 0.830 and 15.47%, respectively).

4.3.1.2 Effect of SMP concentration on sludge filterability

Results obtained from each filterability method and treated sludge were also correlated with the SMP content (see Fig. 4.2).

According to results obtained by the TTF and CST methods, the aerobic sludge filterability would be more influenced by the SMP content than by the TSS concentration, within the ranges contemplated in this study. Indeed, higher correlations between CST and TTF results and SMP content were observed than when evaluating the TSS concentration (see Fig. 4.1). However, the SRF method seems to be not able to capture the influence of SMP on sludge filterability neither. Although, a potential equation was assumed in this case for representing the relationship between sludge filterability and SMP content since it showed the best results, a low R^2 (0.116) and high MAPE (92.99%) were obtained still. Hence, SRF method was identified as a methodology no recommended for activated sludge filterability determination. Since both TTF and CST methods showed a similar sensibility for estimating the activated sludge filterability in this study, CST method would be recommended due to its better precision, achieving R^2 and MAPE values of 0.951 and 32.17%, respectively.

The anaerobic digestate also showed a significant relationship between its filterability and SMP concentration. The SRF method was found once again as the least suitable method, showing the lowest R^2 and higher MAPE. On the other hand, the CST method showed a lower precision and sensibility than TTF in this case (TTF method R^2 and MAPE values of 0.710 and 29.16%, respectively). As commented before, the TTF method seemed to strongly depend on SMP content, identifying the SMP concentration as the dominant variable onto the anaerobic co-digestion system. Instead, the CST method seems not to be so influenced by this variable. In consequence, although the TTF method may be recommended to determine this kind-of-sludge filterability when considering changes on the SMP content, the CST could be considered as a more convenient option due to its more overall robust filterability determination.

Unlike the important effects of SMP concentration on filterability of activated sludge and anaerobic digestate, the PSE sludge showed a completely different behavior. In this case, all three tested methodologies seem to agree that the SMP content did not significantly influenced the PSE sludge filterability. Low R^2 (0.001 and 0.089) and high MAPEs (46.25 and 62.20) were obtained for both SRF and TTF methods, respectively, displaying the insignificant effects of the SMP content on sludge filterability. CST method also captured with a poor precision SMP influence in sludge filterability, with R^2 and MAPE values of 0.316 and 35.90, respectively. Hence, slight relationships between SMP content and PSE sludge filterability could be expected within the concentration range considered in this study. This phenomenon may to be related with the source of the treated sludge SMP. The SMP terminology is an extremely general definition, gathering multiple compounds such as humic substances, proteins, DNA, lipids, polysaccharides, and other carbohydrates and small molecules [4.30]. Consequently, although important SMP concentrations were found in the PSE during its filtration, the kind of substances involved in this sludge and the aerobic sludge and anaerobic co-digestate may be strictly different. The biological sludge could be producing SMPs with higher molecular weights or with higher viscosity or sticky proprieties, hindering the filtration interphase between the formed cake layer and the membrane. Indeed, the active organisms produce SMPs for capture the near substrate or hydrolyze and metabolize it [4.30]. Unlike them, the SMPs obtained in the PSE sludge could be more related with some organism debris or other inorganic sources. Thus, although the SMP determination could be useful for estimating the medium filterability, a more specific analysis is required when wanting to properly determine the effects of the SMP on each filtration process.

4.3.2 Filterability estimation and 3D representations

Fig. 4.4 shows a 3D representation of the estimated effect of TSS and SMP concentrations on sludge filterability. This figure allows to identify overlapping effects, as well as to evaluate synergistic interactions between these two variables. Since the influence of both TSS and SMP concentrations was found to be roughly linear when applying the TTF and CST methodologies, a simple plane equation (*i.e.* z=ax+by+c) was used to adjust each sludge filterability to changes on these two variables (see Fig. 4.4). Least squares method was used to adjust the proposed model to the experimental data. Table 4.1 shows the results obtained for each plane parameter together with the model MAPE for every case study. In general, the sludge filterability was found to mainly depend only on one variable. As previously commented, the activated sludge filterability depended only on the SMP content, reaching same conclusion for both, TTF and CST methods. Similarly, the PSE filterability was only influenced by the TSS concentration. However, the anaerobic digestate was found to be influenced by both SMP and TSS concentrations, although one of them showed a higher influence than the other depending on the filterability method employed. In this case, the CST method indicated that the sludge filterability was more dependent on TSS concentration, while the TTF method displayed instead a more dependence on SMP content. This discrepancy was also observed when representing the sludge filterability as a function of only one variable (see Fig. 4.1 and Fig. 4.2). This discrepancy was associated to the different mechanism used by each method to determine sludge filterability (*i.e.* CST is based on the water movement through a capillary medium, while TTF is based in the amount of water filtered at a constant pressure driven force). Thus, an evaluation of the effect of the TSS and SMP concentration in the real filtration process is required to determine the method which provides of most accurate and useful information.

Table 4.1. Equation parameters and mean absolute percentage error (MAPE) of each filterability model

Sludge	**Method**	**Equation parameters***			**MAPE (%)**
		a (min L $gSST^{-1}$)	b (min L $mgDQO^{-1}$)	c (min)	
Activated sludge	CST	0	$2.38 \cdot 10^{-2}$	0	50.89
	TTF	0	$2.81 \cdot 10^{-2}$	0	41.38
Concentrated primary settler supernatant	CST	0.253	0	0.483	36.94
	TTF	5.642	0	0	46.85
Anaerobic co-digestate	CST	0.547	$1.02 \cdot 10^{-3}$	0.365	40.69
	TTF	0.356	0.106	0	47.67

*Simple plane equation: *Filterability = a SST + b SMP + c*

Overall, similar MAPE values (about 35 – 50%) were found by all presented plane models and simple linear regressions obtained in this study (see Fig. 4.1, Fig. 4.2 and Table 4.1). Therefore, similar limitations were assumed by all adjusted equations, the 3D representations considered as a most convenient model to estimate sludge filterability, or vice versa, since the effect of both variable is considered simultaneously.

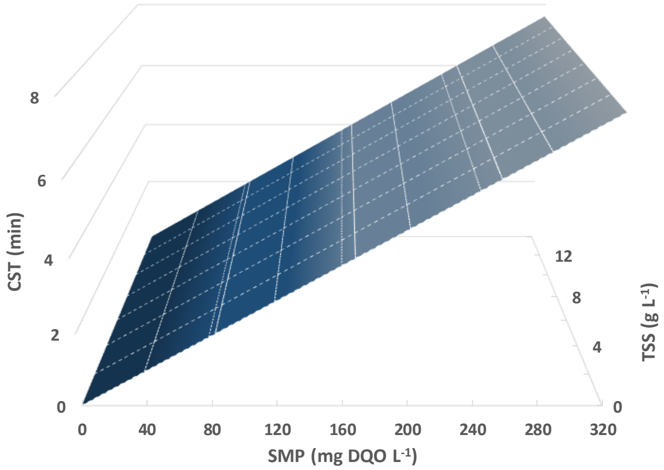
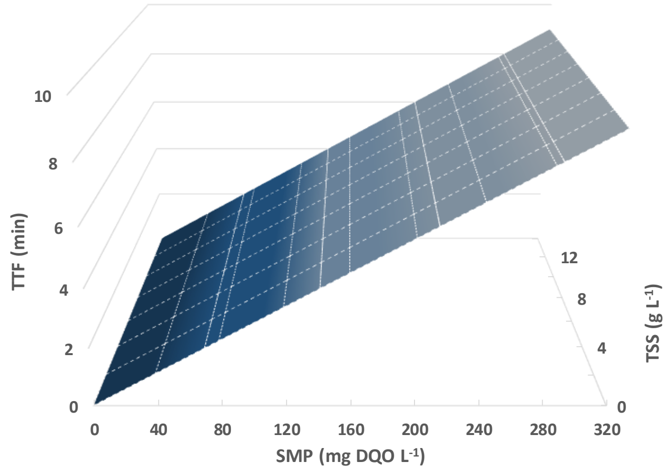
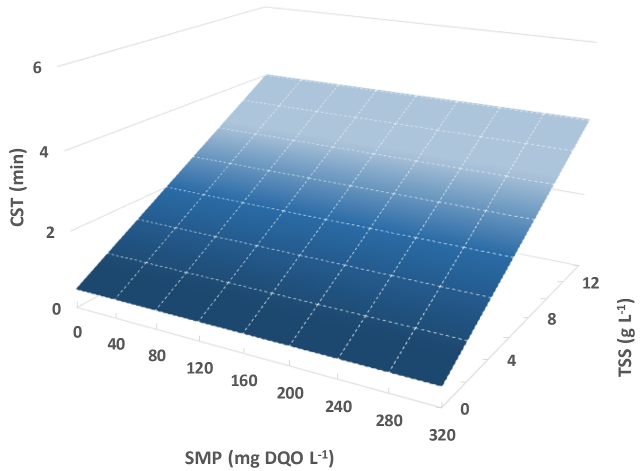
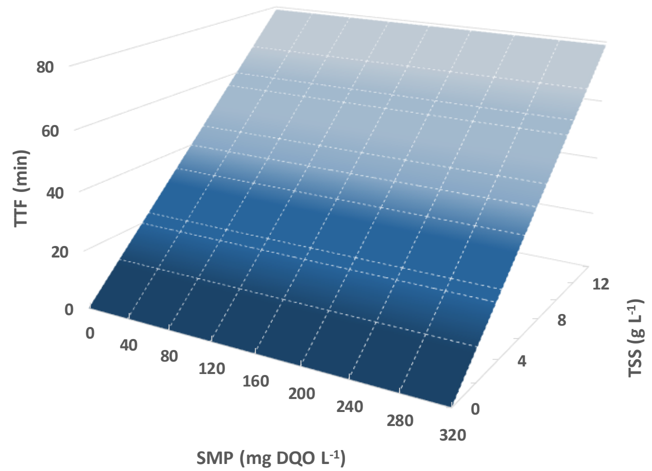
Sludge	Filterability method: CST	Filterability method: TTF
Activated sludge		
Concentrated primary settler supernatant		

Figure 4.4. Cont.

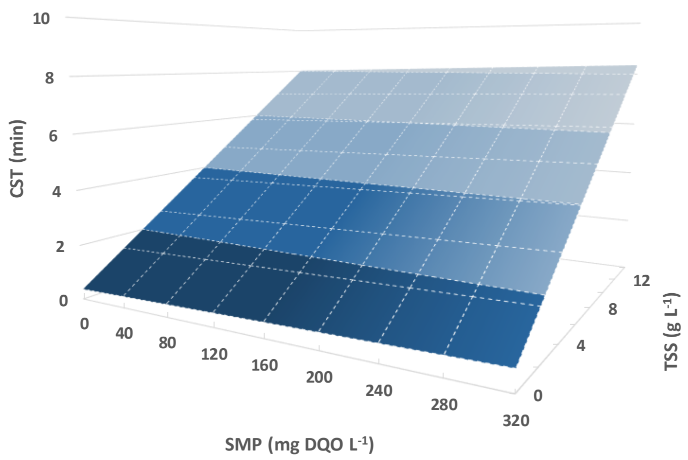
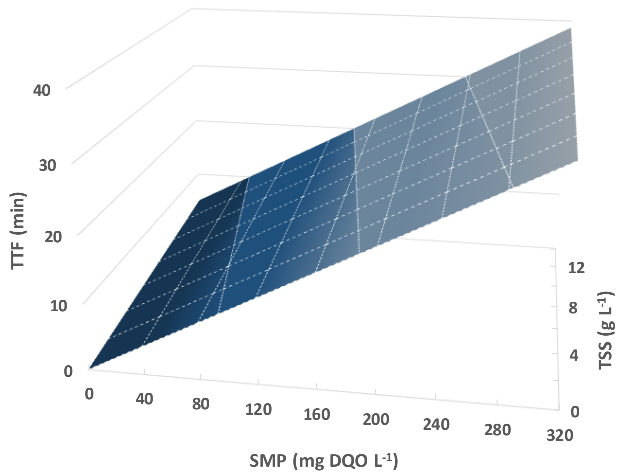
<table>
<tr><th rowspan="2">Sludge</th><th colspan="2">Filterability method</th></tr>
<tr><th>CST</th><th>TTF</th></tr>
<tr><td>Activated sludge</td><td>
</td><td>
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Figure 4.4. 3D representation of modeled sludge filterability from TSS and SMP concentration effects.

4.3.3 Validation of filterability results at pilot scale

Both PSE and anaerobic sludge were obtained from pilot plants including membrane modules. Since this filterability study was performed during the operation of these membranes, the filtration data monitored in the pilot plants when taking the sludge samples was used to validate the filterability results from each method. In this study, since both processes operated at a constant flux, the recorded filtration transmembrane pressure (TMP) was used as indirect indicator of the filtration resistance. Fig. 4.5 shows the TMP recorded in the pilot plants and the experimentally determined filterability by TTF and CST methodologies. Acceptable regression results were found for both sludge systems, indicating that the sludge filterability determined by these methods was in fact representing the real filtration resistance of the treated sludge. The PSE sludge showed relatively low filtration resistances within the operating conditions tested, displaying a slight TMP variation (between 40 – 80 mbar) which hindered the proper correlation between filtration resistance and sludge filterability. On the other hand, the anaerobic sludge filtration showed a slight higher resistance during filtration (TMP variation between 40 – 120 mbar), achieving in this case a higher correlation between filtration resistance and CST results. The TTF method showed a lower correlation, indicating that the high sensibility that this method showed on the SMP content was not representative of the real filtration process when filtering anaerobic sludge. In fact, when representing the modeled filterability values regarding the real filtration resistance (see Fig. 4.6), an extremely low R^2 (0.122) was obtained between the modeled TTF and the process TMP, showing that this model and method were not suitable for determining the filtration resistance of this kind of sludge/system. Instead, the CST method achieved an acceptable R^2 (0.647) between the modeled filterability and the process TMP. In the case of PSE sludge, acceptable regressions were obtained for the modeled filterability and the process TMP for both CST and TTF methods, showing the later a slight better correlation (R^2 values of 0.718 and 0.612 for the modelled TTF and CST, respectively).

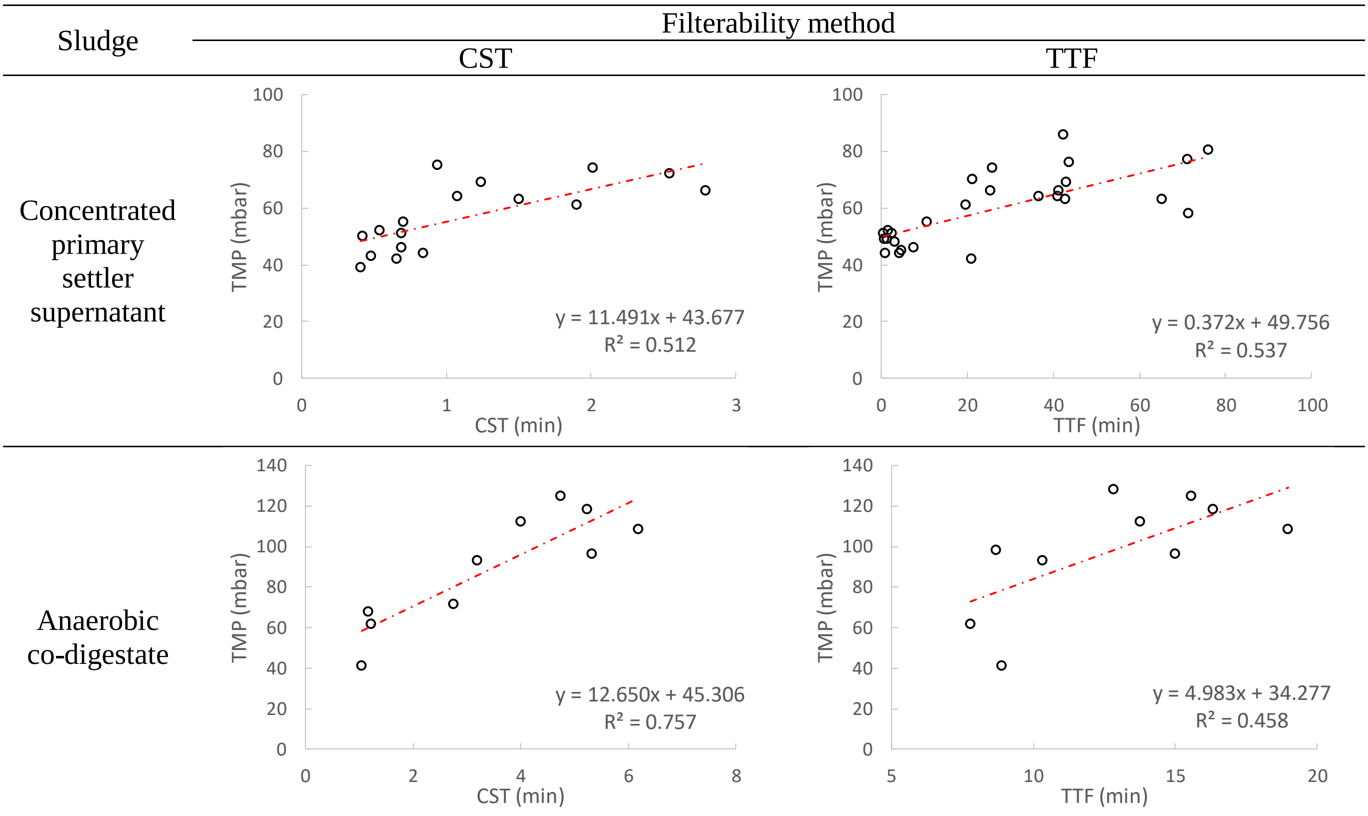

Figure 4.5. Correlation between the transmembrane pressure (TMP) from pilot-scale systems and the experimentally determined sludge filterability.

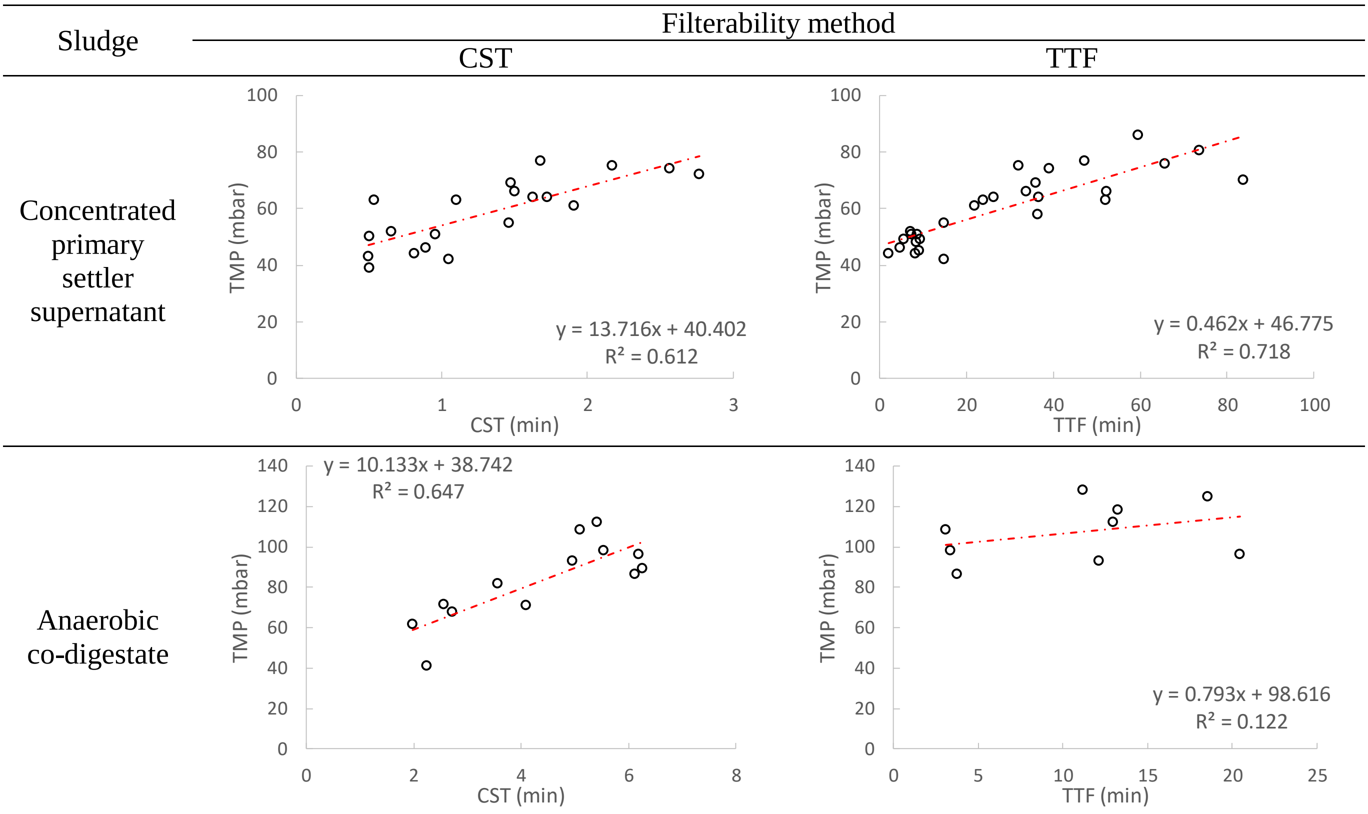

Figure 4.6. Correlation between the transmembrane pressure (TMP) from pilot-scale systems and the modelled sludge filterability.

4.3.4 Guidelines for method selection depending on sludge source

CST method was identified as a suitable alternative for determining sludge filterability regardless of sludge source, within the operating conditions evaluated in this study. However, TTF method was determined as a solid alternative when treating PSE and anaerobic sludge, achieving correlations similar and even superior in some cases than those obtained by CST method. On the other hand, overall, SRF method resulted in the poorest MAPE values within the methods evaluated, especially displaying bad correlations regarding SMP content. Moreover, this method presented several drawbacks: (1) SRF requires of determining the treated sludge TSS and viscosity, thus preventing filterability results to be used as indirect indicator of these sludge characteristics; (2) higher errors could be expected by this method since it accumulates all the errors of each experimental determination; and (3) SRF could be considered as an extension of the TTF method, requiring TTF to be calculated. Thus, all potential limitation observed when applying the TTF method to a given system will be transferred to SRF results. Consequently, the SRF method would be not recommended for monitoring sludge filterability, within the operating conditions evaluated in this work.

Concerning correlations between determined filterability (experimental and modeled) and filtration resistance during the ultrafiltration of PSE and anaerobic sludge, results obtained showed that both, CST and TTF methods, could be employed to predict the sludge filtration resistance or estimate its TSS and SMP content. TTF method presented better results when treating PSE sludge while CST method was the only option when treating anaerobic sludge since TTF method was not able to reach proper correlations. This different behavior regarding filterability method employed and sludge treated may to be related with both, the source of the treated sludge SMP and the basics of the used experimental technique. As above commented (see Section 4.3.1.2), the effect of SMP on filtration may be extremely different depending on the nature of soluble substances involved. Thus, the poor correlations obtained between the anaerobic sludge TMP and TTF method may indicate that this methodology is not suitable to estimate the effects of biological-origin SMPs on sludge filtration resistance. On the other hand, the basics of each filterability method used (filtration with a porous filter or capillarity through a porous medium) could also play a crucial role in the filterability determination and on its correlation onto the real filtration process. *A priori*, more accordantly results to those observed during the real filtration sludge could be expected from TTF methodology since both operate under the same physical laws, *i.e.* filtration using a filter/membrane onto which it will be formed a cake layer from the particulate material accumulation due to the application of a pressure driving force. However, due to the significant difference in the average pore size of used filters compared to the ultrafiltration membranes from the pilot systems (8 and 0.03 µm, respectively), the effects of SMPs, specially their capacity to block the membrane pores, could vary dramatically in each case. Additionally, the real filtration process used air sparging for reducing the hindering effects of the formed cake layer during filtration, which differentiated the expected resistance even more.

Thus, based on the results obtained in this study, the CST would be the most convenient method to employ when treating biological sludge. On the other hand, when filtering sludge with low/negligible biological activity (*e.g.* PSE sludge), the TTF method could be considered as an interesting alternative since it showed better correlations regarding medium filterability and sludge filtration resistance.

4.4 Conclusions

From the results obtained in this study, it can be concluded that biological sludge is more influenced by SMP content, while PSE sludge was mainly controlled by TSS concentration. Among the methods and sludge sources evaluated, SRF was found to be not recommendable due to its worse filterability – studied variables correlation, especially regarding the SMP concentration. CST method was identified as the most solid method to be used, displaying good correlation for all treated sludge (aerobic sludge, anaerobic co-digestate and concentrated PSE), regardless of TSS and SMP concentration. However, when treating sludge without an important biological charge (*e.g.* PSE), the TTF represented an attractive alternative due to its better correlation with the experimentally determined sludge filtration resistance.

4.5 Acknowledgements

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CHAPTER 5. Direct Membrane Filtration of Municipal Wastewater: Studying the Most Suitable Conditions for Minimizing Fouling Rate in Commercial Porous Membranes at Demonstration Scale

Abstract

This study aimed to evaluate the feasibility of applying commercial porous membranes to the direct filtration of municipal wastewater. The effect of membrane pore size (MF and UF), treated influent (raw wastewater and the primary settler effluent of a municipal wastewater treatment plant) and operating solids concentration (about 1 and 2.6 g L^{-1}) were evaluated on a demonstration plant. Filtration periods of 2 – 8 hours were achieved when using the MF membrane, while these increased to 34 – 69 days with the UF membrane. This wide difference was due to the severe fouling when operating the MF membrane, which was dramatically reduced by the UF membrane. The use of raw wastewater and higher solids concentration showed a significant benefit in the filtration performance when using the UF module. The physical fouling control strategies tested (air sparging and backwashing) proved to be ineffective in controlling UF membrane fouling, although these strategies had a significant impact on MF membrane fouling, extending the operating period from some hours to 5 - 6 days. The fouling evaluation showed that cake layer seemed to be the predominant reversible fouling mechanism during each independent filtration cycle. However, as continuous filtration advanced, a large accumulation of irreversible fouling appeared, which could have been related to intermediate/complete pore blocking in the case of the MF membrane, while it could have been produced by standard pore blocking in the case of the UF membrane. Organic matter represented more than 70% of this irreversible fouling in all the experimental conditions evaluated.

Keywords

Membrane fouling control; Microfiltration; Primary settled wastewater; Raw sewage; Ultrafiltration.

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5.1 Introduction

Global water stress and climate change have been identified as two of the most important worldwide problems to be addressed in the next decades [5.1; 5.2]. The continuous use of non-renewable resources in the current economic models have shown their unsustainability in the long-term, boosting the need to find new water, energy and nutrient sources to cope with the increasing demands for these essential resources. Due to this, new economic models based on the Circular Economy (CE) need to be adopted [5.3] while developing more energy-efficient and greener technologies to achieve self-sufficient sustainable systems. In the sewage treatment sector, the municipal wastewater (MWW) paradigm has changed dramatically in recent years and is starting to be considered not as a waste but as a source of vital resources, including reclaimed water, energy and nutrients [5.4]. New membrane-based technologies and alternative treatment schemes are being proposed to transform the former concept of wastewater treatment plants (WWTP) into new resource recovery facilities, such as membrane photobioreactors [5.5; 5.6], anaerobic membrane bioreactors [5.7] also including alternative treatment platforms and configurations [5.8], and other advanced systems for water and nutrient recovery [5.9–5.11]. Moreover, produced water quality is being improved by emerging MWW tertiary treatment for effectively removing antibiotics and other emerging contaminants, such as photodriven advanced oxidation and photocatalysis [5.12–5.14]. Among the new MWW treatment alternatives, the direct filtration of MWW, commonly referenced by the scientific community as direct membrane filtration (DMF), has recently gained increasing interest [5.15]. This alternative consists of using membrane technology to capture and concentrate the organic particulate fraction of the influent MWW to reduce/avoid the energy demands of secondary treatments (reduction of oxygen requirements of aerobic microorganism included in classic sewage treatment scheme) while boosting the amount of energy that can be recovered from anaerobic digestion (AD) by increasing the organic matter intake. Numerous studies have been carried out to date aimed at determining the feasibility of applying the DMF concept to MWWs [5.16]. Unfortunately, severe fouling is usually reported when directly filtering sewage by porous membranes [5.17; 5.18], so that the implementation of effective strategies for minimizing membrane fouling is an imperative matter to enable the application of the DMF alternative [5.19].

Fouling in porous membranes can be generally classified into two categories: cake layer and pore blocking [5.20]. The former describes the accumulation of particulate material on the membrane surface as filtration advances and is usually associated with reversible fouling in proportion to the applied flux. Conversely, the latter describes the partial/complete obstruction of the membrane pores by the deposition of colloidal particles or sticky gel formations, which rapidly hinders membrane permeability. This type of fouling can be either reversible or irreversible, depending on the difficulty of removing the adhering substances. Pore blocking can be divided into different sub-categories depending on the nature of the useful membrane area affected, differentiating between intermediate pore blocking (some particles can block the membrane pores or be accumulated onto other particles forming a pseudo cake layer), standard pore blocking (a reduction of the membrane pore size due to the accumulation of materials inside the membrane pores) and complete pore blocking (the particles accumulate only on the membrane surface, completely blocking the membrane pores). Fig. 5.1 shows a graphic scheme of the different types of fouling.

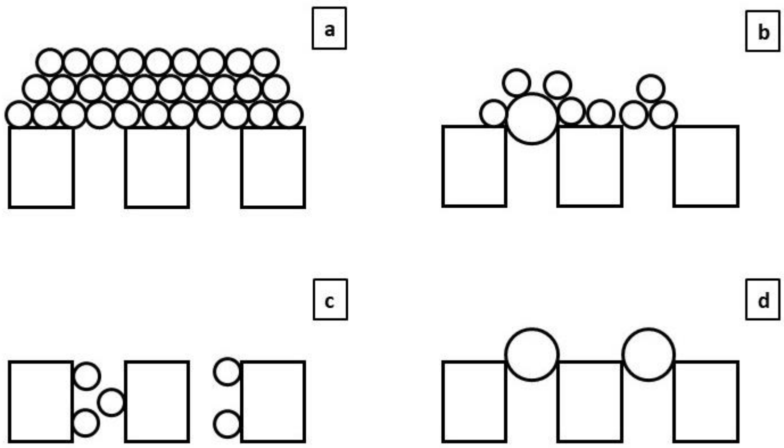


Figure 5.1. Fouling mechanism graphical scheme: (a) cake layer, (b) intermediate pore blocking (c) standard pore blocking and (d) complete pore blocking.

In the case of the DMF of MWW, the main source of the reported severe fouling has not yet been clearly described and so further studies are required to minimize fouling during filtration. First of all, the appropriate operating conditions need to be determined for passively minimizing fouling when directly filtering MWW. In this regard, several authors have studied different membrane technologies to apply the DMF alternative, generally using MF and UF membranes due to their larger pore sizes [5.19]. Nonetheless, the different operating conditions used in the different studies (*i.e.* filtering flux, filtration-relaxation periods, influent characteristics, physical fouling control strategies, etc.) hinder the attainment of proper conclusions, so that further studies are needed to compare the real potential of each membrane technology. The use of more refined influent for applying the DMF, such as primary settler effluent (PSE), has also been suggested for reducing membrane fouling [5.17; 5.21]. This strategy could be an interesting approach, since it takes advantage of the existing facilities and a large amount of the influent MWW particulate material would be recovered in a previous step, reducing the amount of solids in contact with the membrane and presumably reducing the treated MWW fouling potential. Additionally, the sludge recovered during filtration can be mixed with sludge from the primary settler for direct use via AD and would not affect the energy recovery potential. This alternative scheme would provide important potential benefits and needs to be studied in depth to properly compare the overall energy and resource recovery that can be obtained regarding the influent used (*i.e.* raw and PSE MWW).

Diverse physical and chemical fouling control strategies have been proposed by numerous authors for minimizing fouling growth, thereby avoiding short-term chemical cleaning [5.16; 5.17]. Nonetheless, some of the proposed on-line methods, although effective, are energy-intensive applications which would dramatically reduce the resource recovery potential of the DMF alternative. Different fouling control strategies thus need to be tested under diverse operating conditions to determine the best methods of minimizing fouling while reducing the filtration energy input. Other crucial operating conditions, such as the effective operating solids concentration, need to be considered and evaluated to boost resource recovery and minimize energy demands. In this regard, low operating solids concentrations would presumably involve lower filtration energy inputs, since less filtering resistance is expected. However, the recovered sludge would require post-treatment for valorisation, increasing the resources required for feeding the AD process (*e.g.* pumping demands, space requirements, etc.), while high operating concentrations may require higher energy filtration requirements but could reduce or even eliminate any additional steps to recover the sludge. Thus, the overall process should be considered to properly determine the most appropriate treatment scheme.

Several studies have explored the best/optimal operating conditions to minimize fouling when filtering MWW using porous membranes. However, most of them have been conducted at lab-/bench-scale, thus further studies are needed in order to assess the feasibility of the technology on a semi-industrial scale. Moreover, the effect of the main operating conditions on membrane fouling cannot be directly extrapolated from lab- to industrial-scale because it depends heavily on the membrane configuration and size/length. For instance, the effect and efficiency of air sparging upon membrane fouling depends considerably on fibre length and hydrodynamic conditions in the membrane tank. On the other hand, a proper comparison between MF and UF technology using comparable influent and operating conditions (operating flux, fouling control strategies, etc.) has not been performed yet.

In this work, a demonstration prototype was operated to determine adequate operating conditions to minimize fouling when filtering MWW using porous membranes. The influence of the membrane pore size (MF and UF membranes), the treated influent (raw and PSE MWW), and operating total suspended solids (TSS) (about 1 and 2.6 g L^{-1}) was evaluated. Additionally, the efficiency of two physical fouling control strategies (air scouring and permeate backwashing) was assessed for each operating scenario tested. On the other hand, chemical cleaning analysis and theoretical filtration modelling exercises were conducted to determine/elucidate the origin (inorganic, organic) and mechanism (cake layer, intermediate/complete/standard pore blocking) of fouling occurring in each of the tested alternatives, thus providing of data for developing future fouling control strategies aimed to optimize systems of this type. This plant was fed with wastewater coming from the "Conca del Carraixet" WWTP (Valencia, Spain). The design characteristics and scale of this plant is considered adequate to give good performance data for scaling-up the evaluated technology to full-scale UWW treatment, since it incorporates commercial full-scale hollow-fibre membrane modules and equipment.

5.2 Materials and Methods

5.2.1 DMF plant

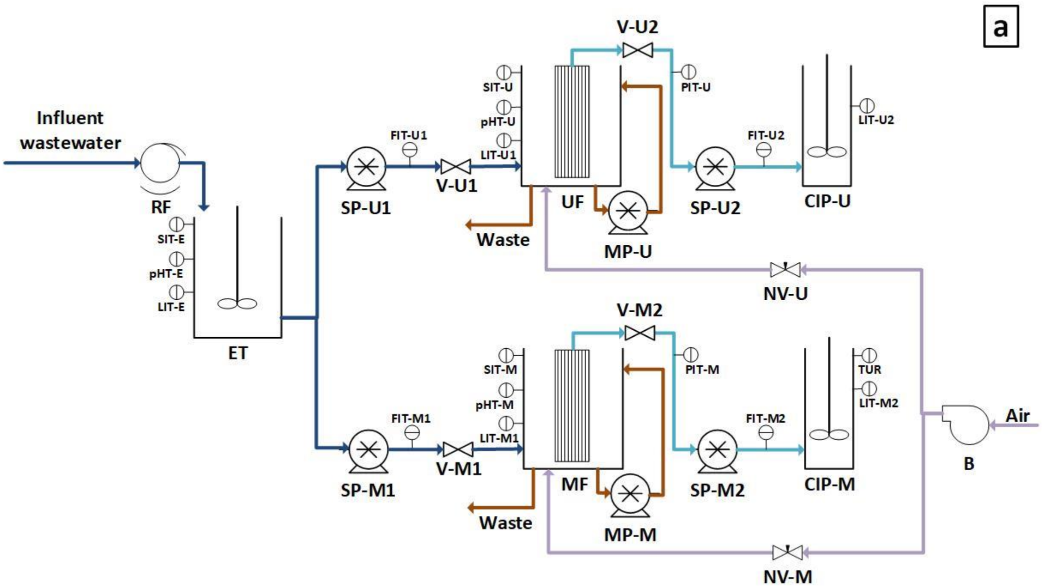
Fig. 5.2a shows a flow diagram of the DMF plant used in this study, which consisted of two independent membrane tanks fitted with two different industrial membrane systems: MF (TERAPORETM 5000, 0.4-µm pore size, filtration area 18 m^2, initial tap water permeability of 455 L per bar m^2 h) and UF (PULSION® Koch Membrane Systems, 0.03-µm pore size, filtration area 43.5 m^2, initial tap water permeability of 294 L per bar m^2

h), each equipped with a screw pump (PCM M series, EcoMoineau™) for vacuum filtration. The membrane tanks had a clean-in-place (CIP) tank (100 and 400 L for the MF and UF CIP tanks, respectively), to store the generated permeate and allow membrane backwashing during the continuous operation. Air scouring was used to mitigate fouling during filtration, using a blower (G-BH7, Elmo Rietschle) to inject air from below each membrane tank. Two additional mixing pumps (PCM M series, EcoMoineau™) continuously recirculated the content of each module to ensure the complete mixing of the membrane tanks and avoid solids stratification.

The plant was continuously fed by MWW from the "Conca del Carraixet" full-scale WWTP (Alboraya, Spain). Two different influents were used during this study: (1) raw MWW (after the typical screening and sieving, desanding and degreasing pre-treatment) and (2) PSE from the WWTP. Table 5.1 shows a characterization of both influents. Regardless of the influent used, a 0.5 mm screen size roto-filter was installed as pre-treatment to protect the membrane modules, collecting the pre-treated influent in an equalization tank (745 L) to feed each membrane tank with the same MWW. Two additional screw pumps (PCM M Series, EcoMoineau™) were used in each membrane tank to continuously replenish the permeated volume with fresh MWW according to filtration requirements. Figs. 5.2b and c show views of the system.

Table 5.1. Influent municipal wastewater characteristics

Treated sewage		**Raw**	**PSE**
Parameter	**Units**	**Mean ± SD**	**Mean ± SD**
TSS	mg TSS L^{-1}	321 ± 98	132 ± 43
COD	mg COD L^{-1}	512 ± 118	195 ± 59
SCOD	mg COD L^{-1}	63 ± 28	57 ± 21
SMP	mg COD L^{-1}	58.4 ± 11.3	60.9 ± 13.1
TN	mg N L^{-1}	56.7 ± 10.8	45.5 ± 8.5
TP	mg P L^{-1}	6.4 ± 1.6	5.9 ± 1.1
Alk	mg $CaCO_3$ L^{-1}	342 ± 73	335 ± 67
pH	-	7.4 ± 0.7	7.6 ± 0.5



RF:	Roto-filter	**SP:**	Screw pump	**SIT:**	Solids sensor
ET:	Equalization tank	**MP:**	Mixing pump	**LIT:**	Level sensor
UF:	Ultrafiltration membrane tank	**B:**	Blower	**FIT:**	Flow-rate sensor
		V:	Automatic valve	**PIT:**	Pressure sensor
MF:	Microfiltration membrane tank	**NV:**	Automatic needle valve	**TUR:**	Turbidity sensor
				pHT:	pH-temperature sensor
CIP:	Clean-in-place tank				

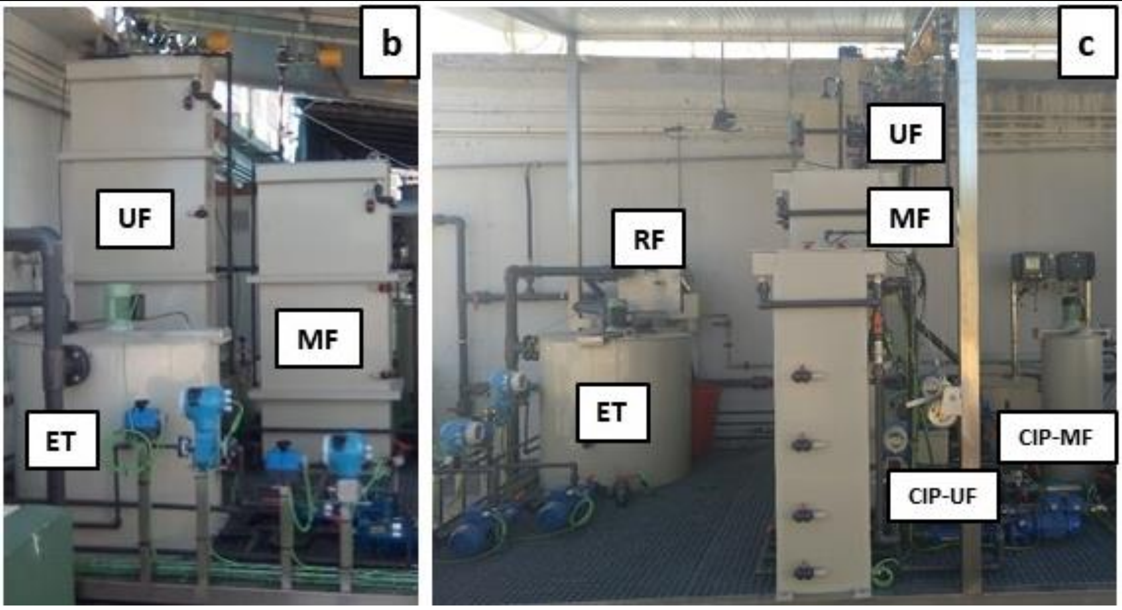


Figure 5.2. DMF demo-scale plant: (a) diagram scheme, (b) front view and (c) side view.

5.2.2 Instrumentation, automation and control

The DMF plant was designed with a high level of automation. Numerous on-line sensors and automatic equipment were installed to allow full control of all its functions (see Fig. 5.2a). The on-line sensors were: three pH-temperature sensors (InPro3100/120/PT100, Endress+Hauser) in the ET, MF and UF tanks; five level sensors (Cerabar PMP11, Endress+Hauser), one for each tank: ET, MF, UF, CIP-MF and CIP-UF; two liquid pressure sensors (IP65, Druck) to control the TMP in each membrane tank during filtration; four liquid flow meters (Picomag, Endress+Hauser), each one associated with an individual screw pump; two air flow meters, one for each membrane module: MF and UF; three solids sensors (LXV424.99.00100, Hach) to control the solids concentration in the ET, MF and UF tanks; and one turbidity sensor (LXV424.99.00100, Hach) to control the turbidity level in the CIP-MF tank. On the other hand, the plat was equipped with the following actuators: five frequency converters (SINAMICS G120C, Siemens), four to control the liquid flowrate of each liquid pump and one to control the air flowrate supplied by the blower; two automatic needle valves to accurately control the air flow-rate distributed to each membrane tank; and four on-off control valves to avoid liquid fluxes during relaxation stages. All these instruments were connected to a programmable logic controller (PLC) for proper multi-parametric control and data acquisition. The PLC was also connected via Ethernet network to a PC from which a SCADA system centralized all the information collected by the instruments and facilitated their supervision and control.

Due to the significant number of sensors and actuators installed, the control script was based on multiple control loops which consisted of classic PID and on-off controllers designed to act on the main operational variables (*e.g.* liquid and air flow rates, TMP control, level measures in each tank, etc.) to reach the established set-points.

5.2.3 Plant operation and experimental plan

The system was operated continuously, performing consecutive filtration:relaxation cycles. The filtration cycle duration was set to 300 s and relaxation duration was set to 60 s, operating at a filtration-relaxation ratio of 5:1. The operation flux was set to 10 LMH (L per m^2 and h) for both membranes during all the experiments performed. Air sparging and permeate backwashing were used as fouling control strategies during filtration for both membrane technologies studied. Air sparging intensity was set at a low specific air demand (SAD) of 0.1 $m^3\ m^{-2}\ h^{-1}$ for minimizing the filtration energy demands, while backwashing was also set to a relatively low periodicity (2 min of backwashing every 10 cycles of filtration:relaxation, therefore operating at a filtration:relaxation:backwashing ratio of 50:10:2) to maintain low physical cleaning downtimes of total operating time. Both air sparging and backwashing were kept constant during all the experiments and were only increased at the end of every experimental filtration period to test their effectiveness for reducing fouling. Each membrane module was operated at a constant TSS concentration, depending on the experiment performed. Since the TSS in the waste increases during filtration due to the retention of particulate material, part of the sludge produced was continually purged from the membrane tanks, adjusting this purge as necessary to operate at the required TSS concentration.

Different operating/design conditions were tested to determine the most suitable ones for minimizing fouling when filtering MWW: membrane pore size (MF and UF membranes), treated influent (raw and PSE MWW) and operating TSS concentration (about 1 and 2.6 g L^{-1}). Table 5.2 summarize all the experiments performed. Regarding the different TSS concentrations, the membrane tanks were operated without waste production until the desired concentration was reached (from some hours to a maximum of 1.2 days depending on the concentration desired), later performing continuous waste purge to maintain a constant TSS concentration during filtration. However, due to the severe fouling growth rate when operating the MF membrane, this module was initially fed with the UF membrane waste when necessary (2.6 g L^{-1} TSS concentration experiments) to achieve the required TSS concentration in this membrane tank without losing a significant fraction of its permeability during the concentrating time.

Table 5.2. Experimental plan

Exp. nomenclature*	**Membrane**	**Sewage treated**	**Waste concentration (g L^{-1})**
MF-Raw-1.0	MF	Raw	0.98 ± 0.39
MF-PSE-1.1	MF	PSE	1.09 ± 0.47
MF-Raw-2.7	MF	Raw	2.72 ± 0.68
MF-PSE-2.8	MF	PSE	2.83 ± 0.70
UF-Raw-1.1	UF	Raw	1.11 ± 0.51
UF-PSE-1.2	UF	PSE	1.23 ± 0.43
UF-Raw-2.6	UF	Raw	2.56 ± 0.42
UF-PSE-2.6	UF	PSE	2.63 ± 0.48

*Nomenclature was build quoting in order the membrane system used, treated influent and operating solids concentration in the module.

5.2.4 Analytical methods and calculations

Each treated influent was sampled once a week to characterize the main pollutants. The UF membrane concentrated sludge and generated permeate were sampled twice a week, while sampling was performed every day in the MF membrane due to severe fouling. Solids, total and soluble chemical oxygen demand (COD and SCOD), total nitrogen (TN) and total phosphorus (TP) were determined according to standard methods [5.22]. 0.45-mm pore size glass fibre membrane filters (Millipore, Merck) were used to produce the soluble fraction of collected samples.

To assess the filterability of the concentrated sludge in the UF membrane, soluble microbial products (SMP) concentration, viscosity and time to filter (TTF) were determined twice a week. The effective SMP concentration was attributed to protein and carbohydrate concentrations only. A commercial total protein kit (Micro Lowry Peterson's Modification, Sigma-Aldrich) was used to determine the protein content, while carbohydrates were determined by the Dubois method [5.23]. The treated sludge viscosity was determined by a Cannon-Fenske viscometer (Series 50, COMECTA®) while the TTF was performed according to standard methods [5.22]. The particle size distribution of the treated influents and TSS concentrations tested in the UF membrane were determined by a laser granularity distribution analyser (Malvern Mastersizer 2000; detector range of 0.01 to 1000 µm).

Different theoretical equations were used to study the predominant fouling mechanism involving the direct filtration of MWW. As proposed by Fujioka and Nghiem [5.20] and Ho and Zydney [5.24], the general fouling mechanism (*i.e.* cake layer, intermediate pore blocking, standard pore blocking and complete pore blocking), can be mathematically modelled by the following expression:

$$\frac{d\,TMP_t}{d\,t} = K(TMP_t)^{\omega} \qquad 5.1$$

Where t represents the filtration time, TMP_t is the transmembrane pressure at each instant, K is a fouling law constant and ω represents the fouling index which can take different values depending on the dominant fouling mechanism (ω = 0 for cake layer, ω = 1 for intermediate blocking, ω = 1.5 for standard blocking and ω = 2 for complete pore blocking). The n value in Eq. 5.1 can thus be determined from the slope based on a linear correlation between the log(TMP_t/dt) and log(TMP_t). More specific models can be deduced for each fouling mechanism when operating at a constant flux crossflow filtration. In this case, as proposed by Kirschner *et al.* [5.25], each specific model can be expressed as follows:

Cake layer
$$\Delta TMP = TMP_0(1 + K_C Jt) \tag{5.2}$$

Intermediate blocking
$$\Delta TMP = \frac{TMP_0}{\frac{1}{K_I} + \left(1 - \frac{1}{K_I}\right)\exp(-K_I B_S t)} \tag{5.3}$$

Standard blocking
$$\Delta TMP = \frac{TMP_0}{(1 - K_S a_0 Jt)^2} \tag{5.4}$$

Complete blocking
$$\Delta TMP = \frac{TMP_0}{1 - \frac{\alpha J}{B}(1 - \exp(-Bt))} \tag{5.5}$$

Where TMP_0 is the initial pressure drop when performing the filtration associated with the intrinsic membrane resistance, J represents the flux, a_0 is the unobstructed membrane surface, α is the cake specific resistance, B_S is the particle resuspension rate and K represents a filtration constant for cake layer (K_C), intermediate blocking (K_I) and standard blocking (K_S). The least squares method was used to adjust the described specific models (Eq. 5.2–5.5) to the experimental TMP evolution, thus deducing the theoretical parameters from the best fit, while the square of the Pearson correlation (R^2) and the root-mean-square error (RMSE) were used to identify the model that bests fits the experimental data.

The original permeability of each (virgin) membrane was determined before conducting the experiments. Filtration with tap water was performed, determining the increase in TMP when raising the operating permeate flux from 5 to 25 LMH, with an increasing step of 4 LMH. Membrane permeability was calculated from Eq. 5.6, estimating it as the average value. Fig. 5.S1 shows the curve obtained for each membrane (figure attached as supplementary material at the end of this chapter).

$$K_{exp} = \frac{J}{TMP} \tag{5.6}$$

NaOCl (2000 ppm) and citric acid (2000 ppm) were used for the membrane chemical cleanings. A cleaning protocol was developed to determine the main fouling source and the chemical cleaning effectiveness of each reagent used which consisted in the following; after the filtration period, the concentrated sludge was removed from the membrane tanks and substituted by tap water. Different filtering fluxes were applied for at least 60 s (from 5 to 25 LMH with an increasing step of 4 LMH) to determine its influence on the TMP and consequently the remaining irreversible membrane fouling. After this process, the membrane modules were cleaned with the basic reagent for at least 8 hours, again filling the membrane tank with tap water at the end and repeating the increasing flux-TMP study to assess the membrane permeability recovery. Finally, a similar procedure was performed with the acid reagent, comparing the membrane permeability with the original membranes to assess the effectiveness of the membrane chemical cleaning.

5.3 Results and Discussion

5.3.1 Effect of membrane pore size

Fig. 5.3 shows the results obtained during the continuous operation of the plant. In this case study, the MF membrane was only able to operate for few hours before reaching high TMPs. In this respect, regardless the different operating conditions tested, a severe fouling evolution was observed when using the MF membrane for filtering MWW. A similar performance has been reported in several studies when operating submerged membranes (see for instance [5.18; 5.26; 5.27]), requiring of specific fouling control strategies in cited works to dilate filtration (such as coagulant dosing, intensive air sparging or enhanced backwashing). Nonetheless, several studies have demonstrated the potential of MF membranes for MWW treatment when operating in side-stream configuration, where longer filtrations are possible by controlling membrane fouling through cross-flow physical cleaning [5.28]. As different authors argue, this phenomenon could be due to a thick cake layer formation on the membrane surface [5.20; 5.21; 5.28] or due to a sticky gel layer on the membrane surface induced by the SMP and EPS content [5.18; 5.28]. Membrane pore blocking could also occur during the early stage of every filtration cycle before the development of the filtration cake layer [5.20; 5.21].

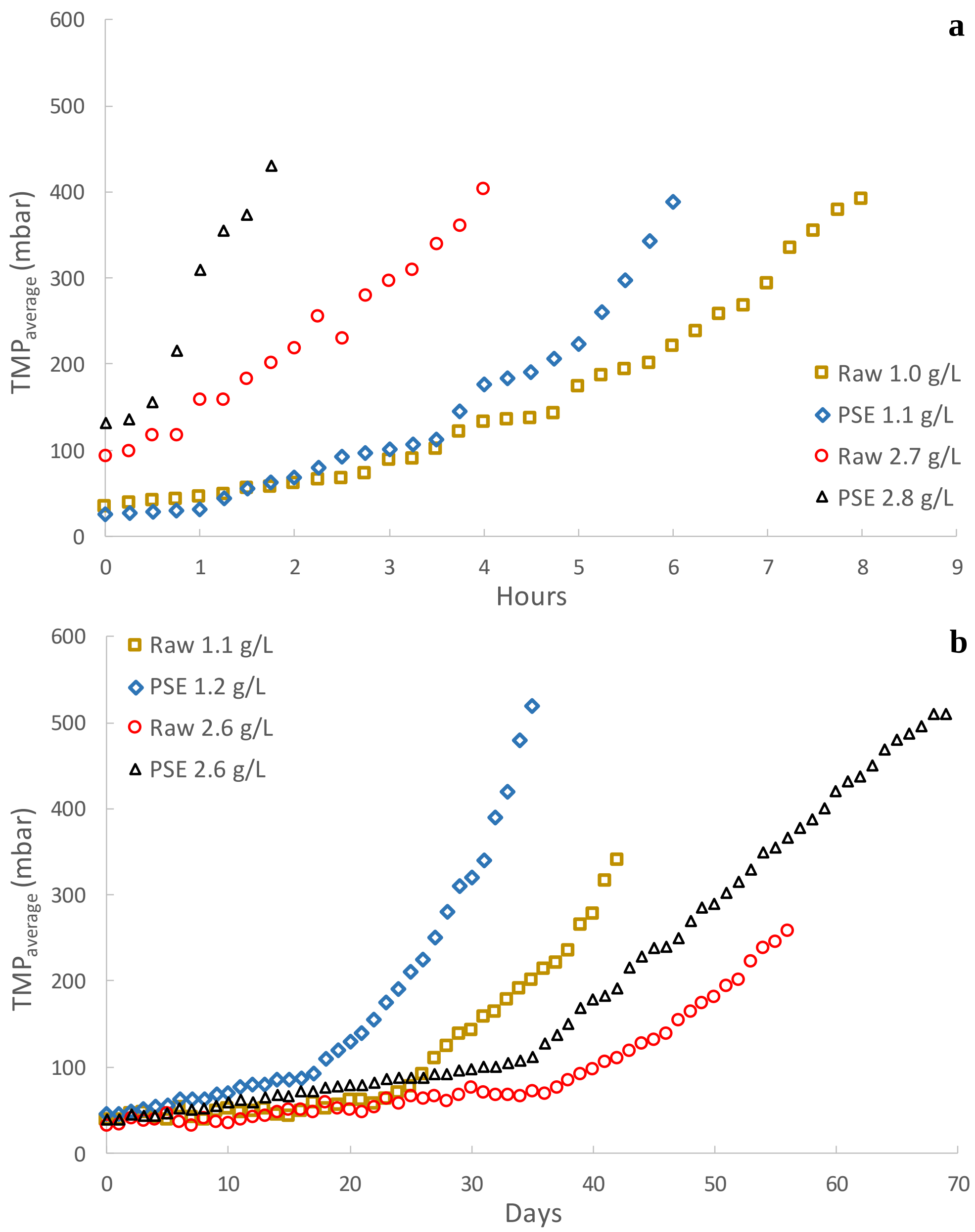


Figure 5.3. Direct municipal wastewater filtration performance: (a) microfiltration membrane and (b) ultrafiltration membrane. Note that the legend shows the influent treated together with the average operating total suspended solids concentration. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

However, despite the unfavourable results obtained when using the MF membrane, the performance was significantly better when using the UF membrane in similar operating conditions, in this case achieving longer filtering periods thanks to the lower fouling growth rate (see Fig. 5.3b). In fact, two different stages can be appreciated during filtration, a first stable period with no significant irreversible fouling in the first days of operation, followed by a second stage in which irreversible fouling consistently increased (see Fig 5.3b and Table 5.3). It is important to highlight that the desired TSS concentration was reached in just some hours in all cases so that the concentrating period did not significantly influenced the first fouling phase. This dramatic fouling growth difference comparing MF and UF could suggest that the main fouling promotor was related to membrane pore size. Fewer fouling issues could be expected in theory when using higher pore size membranes, since less small particulate and colloidal material will be retained on the membrane, reducing the amount of components that could affect the membrane surface or contribute to the cake layer. However, due to the wide particles size range and sticky substances that can be found in untreated MWW, larger pore size membranes would enable more types of material to be deposited in the membrane pores, first promoting pore narrowing, which would evolve until the complete blocking. Therefore, the use of smaller pore size membrane could be an interesting alternative when filtering untreated MWW, since fewer materials would interact with the membrane pores, reducing their blocking propensity.

Table 5.3. Irreversible fouling growth rate during the ultrafiltration membrane operation

Sewage treated	**Waste concentration ($g\ L^{-1}$)**	**Stable period (days)**	**1st slope* ($mbar\ day^{-1}$)**	**2nd slope* ($mbar\ day^{-1}$)**
Raw	1.1	22	1.81	13.22
PSE	1.2	16	2.87	21.58
Raw	2.6	36	1.13	9.31
PSE	2.6	34	1.99	12.01

*Filtration slopes calculated from a linear regression of experimental data.

To study the influence of the influent pollutants on the membrane, particle size distribution, SMP content, viscosity and filterability of the influents was determined during the UF membrane operation. As can be seen in Fig. 5.4, the particle size distribution of both influents (*i.e.* raw and PSE) showed that no important particle concentrations formed around the UF membrane average pore size (0.03 µm), barely observing a small percentage around the MF membrane average pore size (0.4 µm). According to these results, the particle size of the treated influents should not be able to directly block the pores of the membranes used. On the other hand, despite being soluble compounds, an important retention of the influent SMP content occurred during UF membrane operation (see Table 5.4). In this regard, Ravazzini [5.28] reported a similar external polymeric substances (EPS) retention capacity when filtering both raw and PSE MWW with an ultrafiltration membrane. These SMP substances could be responsible for a significant fraction of the fouling during filtration, inducing the generation of gel layers on the membrane surface, blocking the membrane pores or promoting the attachment of other materials to the membrane. Additionally, since both the SMP concentration and filterability of the treated sludge remained constant during each operating period, no important biological effects should be expected. In any case, considering the results obtained from the comparison between the MF and UF membrane technology, the use of UF membranes can be strongly recommended for the application of the DMF concept. Further studies using smaller pore size membranes (*i.e.* NF and FO membranes) could thus be suggested for an in-depth study of the influence of membrane pore size on the fouling growth rate.

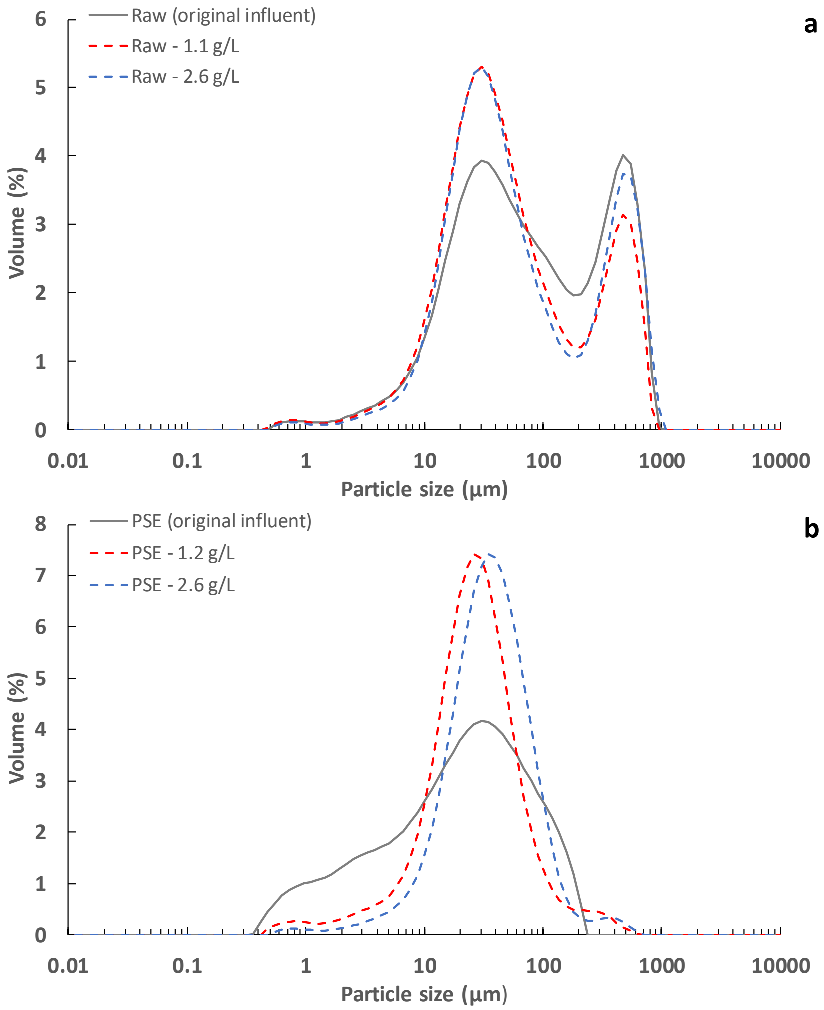


Figure 5.4. Particle size distribution of the different influents treated during the ultrafiltration membrane operation: (a) Raw and (b) PSE. Note that the legend shows the influent treated together with the average operating total suspended solids concentration. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

Table 5.4. SMP concentration, viscosity and filterability of the treated sludge during the ultrafiltration membrane operation

Sewage treated	**Waste concentration ($g\ L^{-1}$)**	**SMP concentration ($mg\ COD\ L^{-1}$)**	**SMP retention (%)**	**SMP:TSS ratio**	**Viscosity (cSt)**	**TTF (s)**
Raw	1.11 ± 0.51	96.0 ± 14.1	10.6	86.5	0.83 ± 0.04	103 ± 57
PSE	1.23 ± 0.43	78.3 ± 17.8	11.5	65.0	0.82 ± 0.03	79 ± 46
Raw	2.56 ± 0.42	109.1 ± 20.1	16.3	42.6	0.89 ± 0.08	566 ± 178
PSE	2.63 ± 0.48	102.8 ± 22.7	13.8	39.1	0.93 ± 0.08	612 ± 237

5.3.2 Effect of the influent used

As Fig. 5.3 shows, lower fouling growth rates were obtained when raw MWW was filtered, regardless of the used membrane or the operating TSS concentration. Since only the higher particles (mainly sedimentable solids) were removed from the treated influent when using PSE instead of raw MWW (see Fig. 5.4), these results suggest that although they are not around the membrane average pore sizes, the smaller size particles are closely related to the fouling mechanisms during filtration. In this case, the treatment of influents with larger size particles could contribute to lower fouling growth by (1) reducing the amount of small size particles in the mixed liquor and (2) promoting the formation of a low-density protective cake layer on the membrane surface. In this regard, when concentrating the particulate fraction of raw MWW, a lower amount of small size particles would be interacting with the membrane surface, since a higher mass percentage would be covered by the larger size particles, thus reducing the pore blocking propensity, especially as the TSS concentration is increased. The presence of larger size particles may also promote the formation of more porous cake layers, which could not present a strong resistance to filtration while protecting the membrane surface from direct interaction with colloidal particles or sticky substances. Unfortunately, since a similar sludge filterability was obtained regardless of the influent used for each TSS concentration tested (see Table 5.4), it was impossible to confirm these previous hypotheses. However, since the filterability determination use filters with a completely different material and pore size in respect of the employed membranes ones, these results should not be directly compared.

According to the results obtained, the use of raw MWW can be recommended for applying the DMF process due to the lower expected irreversible fouling growth rates, at least under the conditions studied in this work. However, further studies considering all the important variables (energy recovery from saved COD, operation and membrane fouling control energy demands, membrane area requirements, chemical cleaning necessities, etc.) need to be performed to properly determine the most favourable conditions for applying this technology, taking into account both energy and economic point of view as well as environmental aspects.

5.3.3 Effect of solids concentration

Different effects were found as the operating TSS concentration was increased depending on the membrane used (see Fig. 5.3). With the UF membrane, a significant improvement was obtained in the filtration performance as the TSS concentration was raised, providing both an extension of the unimportant irreversible fouling evolution period and a reduction in the subsequent irreversible fouling growth rate (see Fig. 5.3 and Table 5.3). This effect was appreciated regardless of the influent filtered, although the benefits of using raw wastewater seemed to be less marked as the TSS concentration increases. In this case, raising the TSS concentration could promote the sporadic flocculation of the small particles by increasing the probability of contact and particle collisions at higher TSS concentrations, thereby reducing the amount of particles able to cause pore blocking or forming a dense cake layer. In this regard, the particle size distribution of the concentrated sludge showed that slightly larger size particles can be expected as the TSS concentration is raised for the PSE (see Fig. 5.4b). However, there was a significant reduction in the average particle size when filtering raw wastewater (see Fig. 5.4a). In this respect, it is possible that due to the compression and shear forces during filtration a certain aggregates size was promoted. Indeed, a similar predominant particle size was obtained after the filtration process regardless of the original influent. The beneficial effects of increasing the TSS concentration could thus be more related to the formation of a protective low-density cake layer on the membrane surface than with the average size of the particles in the treated sludge. Moreover, a slightly increased SMP concentration was found in the treated sludge as TSS concentration was raised (see Table 5.4). However, this higher SMP concentration did not hinder filtration. According to these results, it can be concluded that either the SMP substances are not as strongly related to the observed fouling as anticipated, or that the cake layer formed at higher TSS concentrations is able to protect the membrane from the harmful effects of the SMP compounds [5.29–5.31]. In fact, the SMP:TSS ratio was importantly reduced as the TSS concentration was raised in the membrane tank (see Table 5.4) which supports this hypothesis. On the other hand, since using raw MWW showed a similar benefit in the fouling control due to the high content of large particles in the treated sludge, increasing the TSS concentration could reduce the influence of the influent fed, reaching a TSS concentration for which the effect of the influent used would be negligible. In this regard, Ravazzini [5.28] found similar filtration performances regardless of the influent used (raw or PSE wastewater) when operating a UF membrane, concluding that the influent treated is irrelevant. Since increasing the operating TSS showed a much better improvement regarding fouling control than the influent used, this strategy can be strongly recommended for the direct filtration of MWW. Further studies are required to determine the most favourable TSS concentration, considering both the membrane fouling rate and the energy/space requirements for the recovery of organic matter. Indeed, since increasing the TSS concentration in the membrane tank would reduce/avoid a later sludge concentration step before injection into the anaerobic reactor, this strategy is recommended for reducing the overall scheme energy and space demands.

A completely different trend was found when increasing the TSS concentration in the MF membrane tank. In this case, a higher irreversible fouling rate was obtained, reducing the filtration time from 8 – 6 hours to just 4 – 2 hours, with a clear benefit when using raw wastewater. As in the UF tank, a slight increase in the average particle size of the filtered mixed liquor could be expected as the TSS rose. However, in this case, the slight increase in the particle size would not be enough to protect the membrane from pore blocking or high density cake layers, being able even to promote it due to the greater amount of particles with a similar size to the MF membrane pores.

5.3.4 Fouling control strategies effectiveness

Air sparging and permeate backwashing were used as fouling control strategies. As can be seen in Fig. 5.5, the results obtained when using these strategies depended on the membrane studied. With the UF membrane, neither air sparging intensity nor backwashing frequency were able to significantly reduce the fouling growth rate in any of the operating conditions tested (see Fig. 5.5). These results seem to indicate that the main fouling source when operating the UF membrane was related to irreversible fouling and not to the formation of an important reversible cake layer, for which air sparging or backwashing would be effective [5.32]. It could be assumed then that the operating conditions stablished during continuous filtration (0.1 $m^3 m^{-2} h^{-1}$ of SAD and 2 min of backwashing every 10 filtration:relaxation cycles) were enough to control the thickness of the developed cake layer as the filtration advanced.

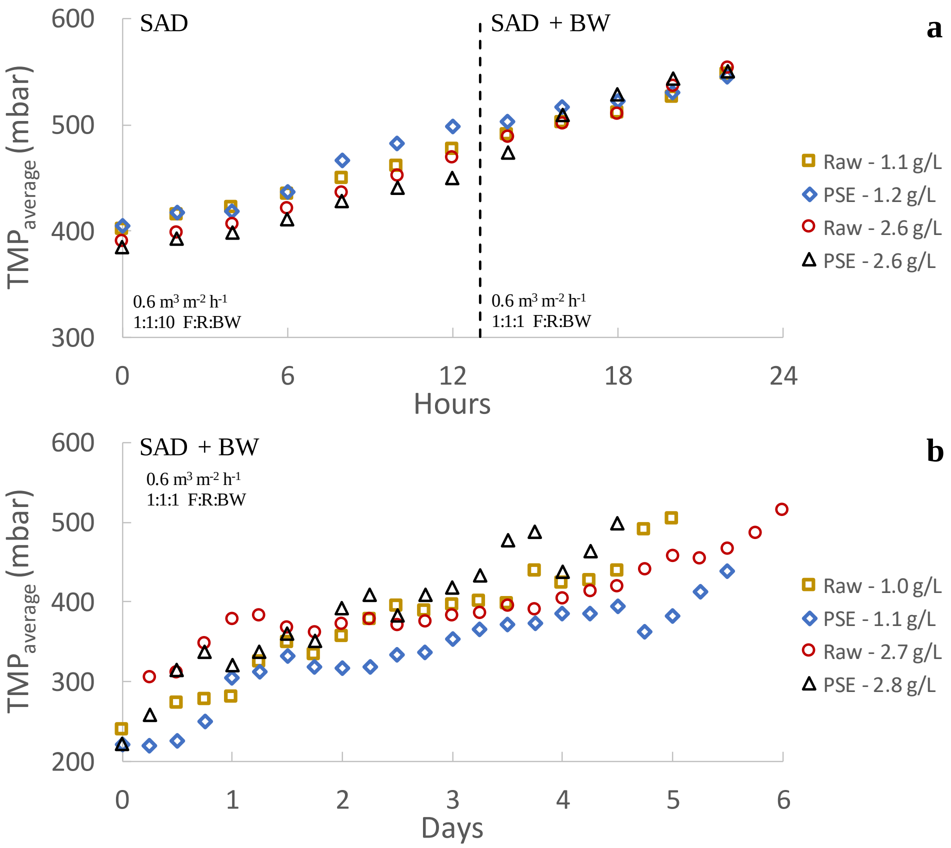
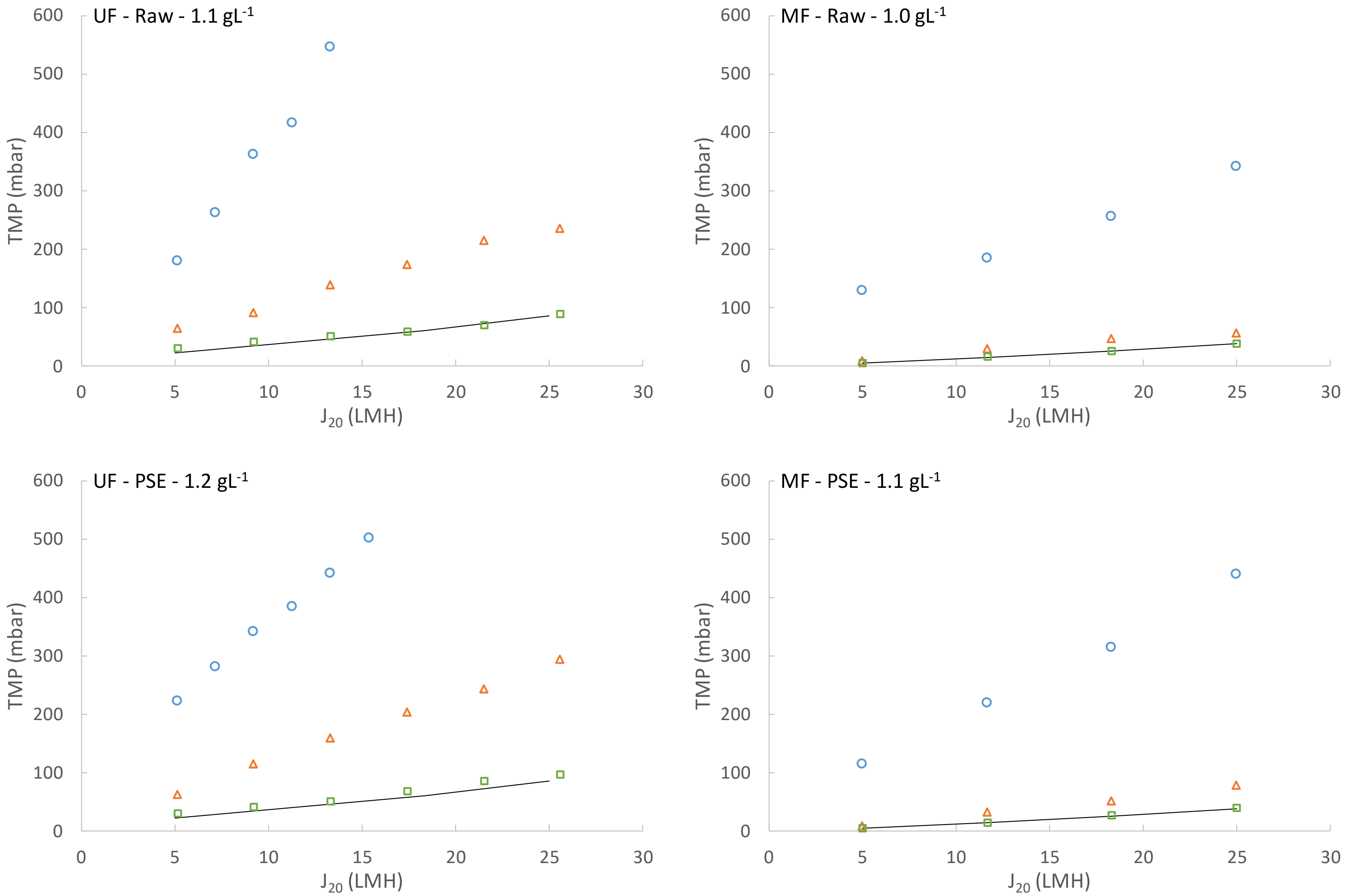


Figure 5.5. Influence of the applied reversible fouling control strategies (increase of the sparging air intensity and backwashing periodicity) on the membrane fouling development: (a) ultrafiltration membrane and (b) microfiltration membrane. SAD: Specific air demand. BW: Backwashing. F:R:BW: backwashing periodicity, *i.e.* filtration:relaxation:backwashing ratio. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

However, when same conditions were applied to the MF membrane, there was a significant improvement during filtration, allowing the process to last for at least several days (see Fig. 5.5). This indicated that an important part of the extremely fast membrane fouling developed with the MF module was reversible fouling, which can be minimized by using similar physical cleaning strategies as those used in this study. In fact, other authors have also reported the beneficial effects of using air sparging on filtration performance when filtering MWW with MF membranes [5.18]. The different influence of the fouling control strategies used on the MF and UF membranes may indicate that different fouling mechanisms were involved. Nonetheless, despite the significant enhancement of the MF membrane filtration performance when these strategies were employed, its overall process remained dramatically inferior to the one achieved by the UF membrane. Thus, although physical fouling control strategies would be an interesting option for boosting MF membrane performance when filtering MWW, UF membranes are a more attractive option for applying the DMF of MWW.

5.3.5 Fouling study

Chemical cleaning was performed on both membranes after each set of experiments, studying permeability recovery of each chemical solution used (*i.e.* basic and acid solution cleaning). As Fig. 5.6 shows, high fouling removals were obtained after applying the basic solution (2000 ppm of NaOCl), achieving a permeability recovery of about 70 – 85 % and about 92 – 99 % for the UF and MF membranes, respectively. According to these results, organic matter seems to be the main fouling promotor during filtration for both membranes, showing a special relevance for the MF, since it was operated during relatively short time periods (around 5 – 7 days) (see Fig. 5.5). In this regard, other studies have also reported similar results when applying basic solutions for membrane cleaning [5.17; 5.33; 5.34], also concluding that organic matter was the major fouling promotor during direct MWW filtration. Nonetheless, inorganic fouling (mainly related to inorganic compound precipitation on the membrane) also had a significant effect on UF membrane filtration, which operated for a middle-term (around 1 – 2 months) (see Fig. 5.3). Although the fouling related to organic matter should be the main issue to control during filtration (*e.g.* by applying chemical-enhanced backwashes with basic solutions) inorganic fouling should also be taken into account to prolong higher membrane permeability during long-term MWW filtration.



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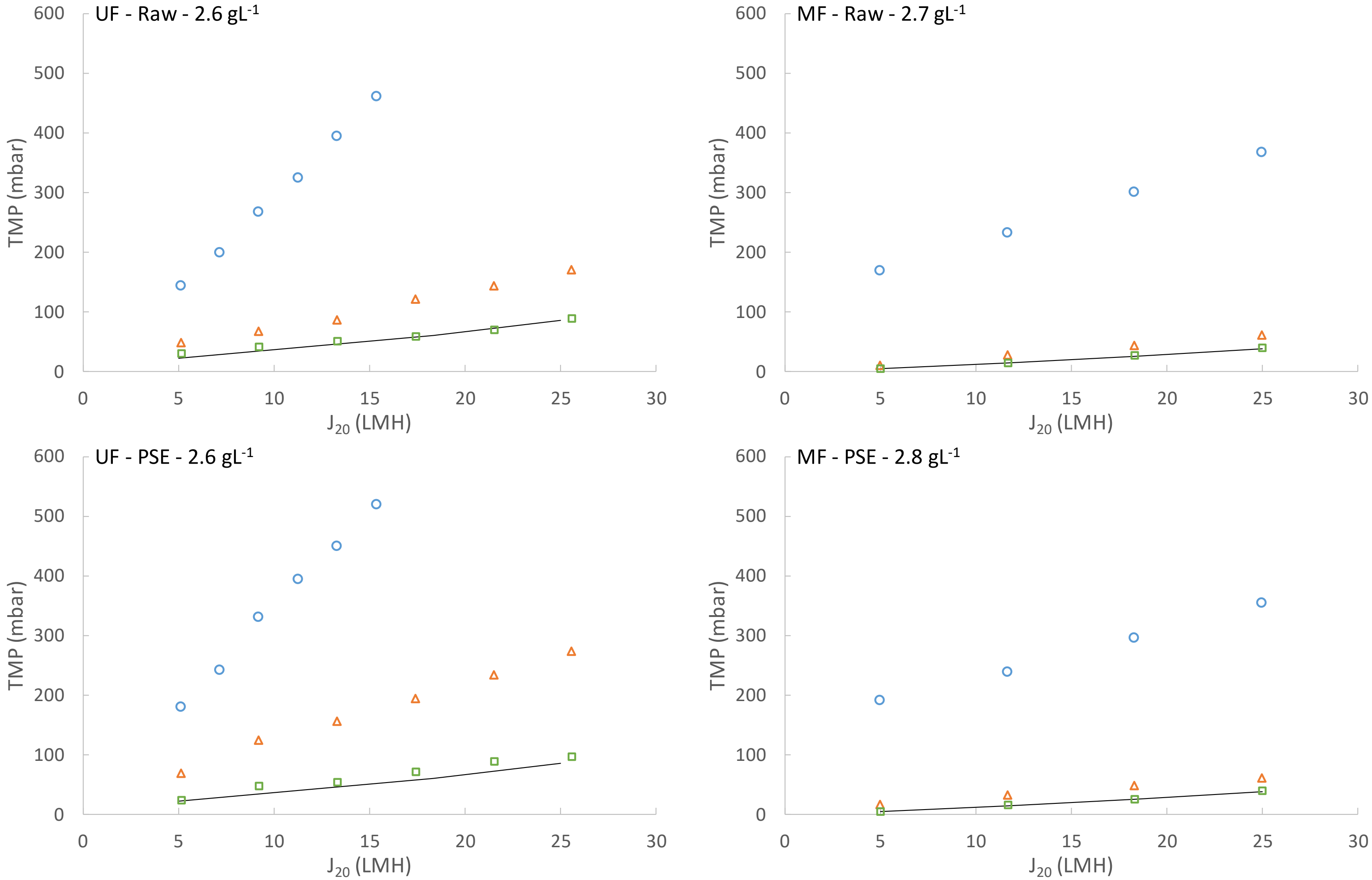

Figure 5.6. Membrane permeability recovery after the chemical cleaning. Note that the legend shows the membrane used, influent treated and average total suspended solids concentration of each operating period before the chemical cleaning. Tap water was used in all cases for determining the membrane resistance. — original membrane resistance; ○ Membrane resistance after the operating period; △ Membrane resistance after the basic cleaning; □ Membrane resistance after the acid cleaning. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

All the data concerning TMP evolution during the operation of the plant were adjusted to different theoretical membrane fouling models (Eq. 5.1–5.5) to study the predominant fouling mechanisms during direct MWW filtration. Since the filtration pump was turned on and the liquid flow rate was adjusted to the set point in the first seconds of every filtration cycle, the first 10 seconds of data were not considered in the fitting. As can be seen in Fig. 5.7, despite the completely different filtration performances obtained from the MF and UF membranes, there were similar trends when focusing on membrane fouling during each filtration cycle. During the first cycles of operation, both membranes showed low filtration resistance with no significant increase of the TMP as the filtration cycle evolved. Indeed, extremely low R^2 were obtained when fitting all the mathematical models, since an inconsistent evolution of the experimental TMP was found due to some white noise in the sensor signal, although low RMSE values were obtained, indicating a correct fit of the data to all proposed models. Regarding the general model, close to zero values were obtained, which indicates that cake layer was the predominant fouling mechanism. Since not a significant fouling growth was found during filtration, the resistance to filtration would be controlled by the filterability of the treated sludge and the light cake layer formed in the first seconds of each filtration cycle. Regardless of the results obtained during the first filtration cycles, significant irreversible and reversible fouling increases occurred as the operating filtration advanced (see Fig. 5.7, Section 2) which can be divided into two different phases. In the early phase, a significantly higher initial resistance was obtained in respect of the first filtration cycles, although no sludge filterability changes were appreciated (see Table 5.4). This dramatic increase of filtration resistance was presumably identified as an increment of irreversible fouling, due to its quick effect on filtration resistance during the first seconds of every filtration cycle and the inability of air sparging and backwashing to reduce it. This resistance could thus be due to a loss of a significant fraction of the useful membrane area caused by pore blocking, gel layer and/or an increment in the compression and robustness of the early formed cake layer as the filtration advanced. After the first seconds of operation, a slight increase of filtration resistance was also found as the filtration cycle advanced. In this second phase, the slow reversible fouling was associated with an increment of the formed cake layer thickness on the membrane surface. Indeed, the general model fit produced near to zero values, indicating that the cake layer mechanism was the predominant fouling promotor (see Fig. 5.7, a2 and c2). Additionally, both the cake layer and the standard pore blocking models showed better correlation with the experimental data (see Fig. 5.7, b2 and d2). However, due to the gradual increase of the TMP as the filtration cycle advanced, all the models could fit reasonably well with the experimental data, the low correlations obtained being more related to the extremely low slope produced by the intermediate and complete pore blocking models than due to the error between the models and the experimental data as the low RMSE values obtained confirms. Finally, continuing with the fouling evolution, similar performances were observed as filtration reached high TMPs (see Fig. 5.7, Sections 3 and 4), accompanied again by two clear fouling phases. In the first phase, there was an important increase in the initial filtration resistance, regardless of unchanging sludge filterability, denoting the irreversible fouling increase. In this case, the general model produced ω values in the range 0.4 – 0.9 for this phase, indicating that some intermediate blocking could be contributing to fouling (see Fig. 5.7, a3, a4, c3 and c4). However, as commented above, the fouling in this case would be an increment of irreversible fouling during continuous filtration. Indeed, pore blocking generally produces a severe exponential TMP increase as filtration advances without the attenuation after reaching a certain point that was obtained during this study. The results produced by the general model could thus be contaminated by this additional accumulated resistance to some degree and should be treated with caution. As in the former case, the second fouling phase seemed completely controlled by cake layer formation, as the ω values were close to zero. However, it is important to highlight that a significant increase of this reversible fouling was obtained as the overall filtration advanced, obtaining higher slopes each time. This phenomenon was probably related to the loss of usable membrane area due to irreversible fouling thereby promoting the accumulation of more thick cake layers in the not blocked zones and/or favouring their compression. Concerning the specific models, only the intermediate and complete pore blocking models were able to properly fit the experimental data during these operating sections (see Fig. 5.7, b3, b4, d3 and d4). Nonetheless, as already commented, the evolution of these models is not the classic one, being forced to take this trend due to the additional resistance produced by the accumulated irreversible fouling. In fact, the cake layer and standard pore blocking models would fit the experimental data perfectly if the first 30 seconds of the filtration cycle were not considered (data not shown). Finally, there were no significant differences of the model fitting in every operating section when applying this analysis to the data obtained when a different influent (raw or PSE) or TSS concentration (about 1.1 and 2.6 g L^{-1}) was employed (data not shown). No relevant differences of the predominant fouling mechanisms during the filtration cycles could thus be expected when operating under the conditions tested in this study.

Due to the dramatic difference between the MF and UF membrane performance during continuous filtration, despite the similar TMP evolution when only considering independent filtration cycles, all the theoretical models were also applied to the daily (UF membrane) and hourly (MF membrane) TMP evolution exposed in Fig. 5.3 to study/elucubrate the fouling mechanisms involved in the irreversible fouling of these two membranes. As can be seen in Table 5.5 and Fig. 5.8, the predominant fouling mechanism for both membranes could be considered as intermediate pore blocking, achieving ω values in the general model around 0.7 – 1.5 in all cases. The irreversible fouling growth during continuous filtration could thus be due to the same source for both membranes, the different performance between them being exclusively due to the UF membrane capacity to reduce the propensity to fouling. However, the MF membrane data seem to better fit the intermediate and complete pore blocking models, while the UF membrane data in general fits better with the complete and standard pore blocking models. These results could indicate that the dramatic difference between the MF and UF membrane performance could be partially due to the different-acting fouling mechanism combination. An intermediate/complete blocking of the pores may occur in the MF membrane during filtration, rapidly hindering its permeability. Instead, UF membrane fouling could be subjected to an initial pore narrowing, which could evolve into a complete block as filtration advances, which would explain why the fouling of the UF membrane showed two different stages (the first with an insignificant irreversible fouling generation related to a slight pore narrowing, while the second with a consistent permeability lose due to complete pore blocking). Nonetheless it is important to consider that although one mechanism can govern membrane fouling, they all can occur simultaneously and even change in importance during filtration. Indeed, the sharper manifestation of irreversible fouling observed as filtration advances was probably related with the state of the membrane after some operational time (*i.e.* irreversible fouling do not show important effects at the start of filtration since all membrane area is available, thereby blocked pores not so important since a large amount of them are still operative).

UF – PSE – 1.2 g L^{-1}			
Section 1	Section 2	Section 3	Section 4
First days of operation (day 6 in Fig. 5.3)	Evolution after some weeks of filtration (day 26 in Fig. 5.3)	Last days of operation (day 33 in Fig. 5.3)	Raise on the air sparging and backwashing intensity (hour 16 after finishing the filtration period)

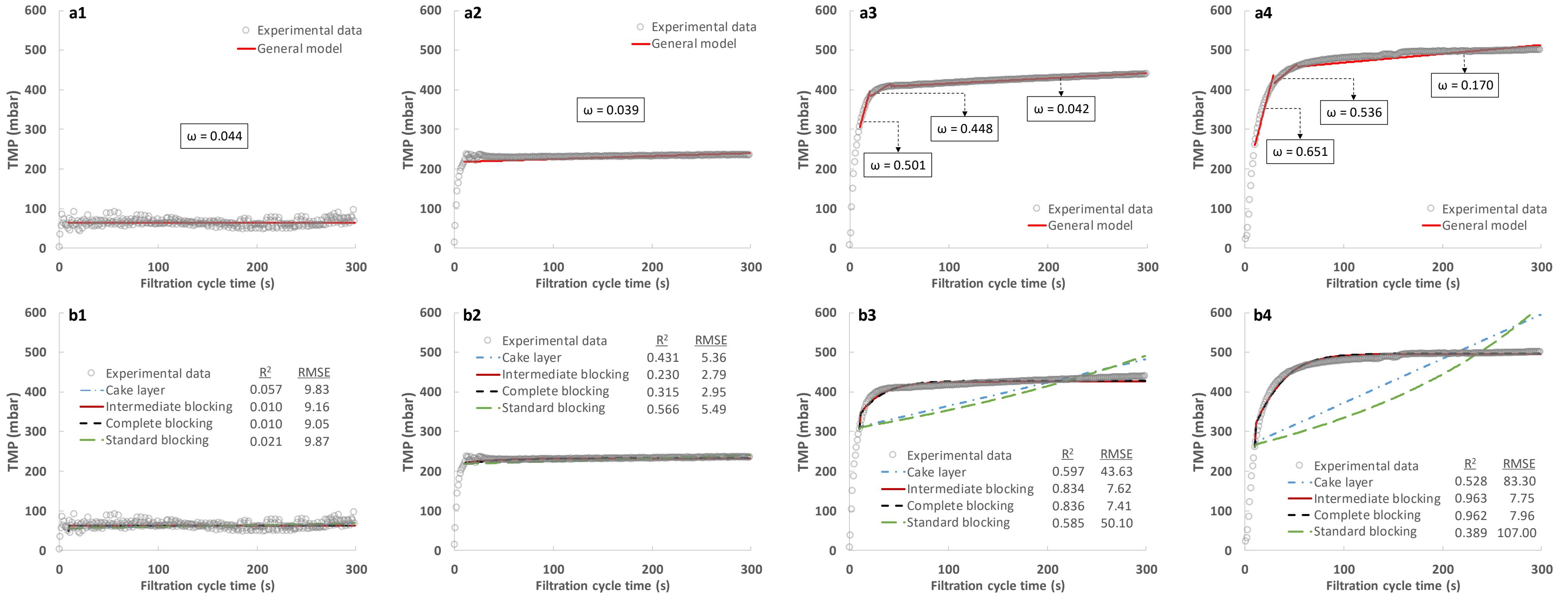

Figure 5.7. Cont.

MF – PSE – 1.1 g L^{-1}			
Section 1	Section 2	Section 3	Section 4
First hours of operation (hour 0.75 in Fig. 5.3)	Evolution after some hours of filtration (hour 4.75 in Fig. 5.3)	Last hours of operation (hour 6 in Fig. 5.3)	Raise on the air sparging and backwashing intensity (day 3 in Fig. 5.5b)

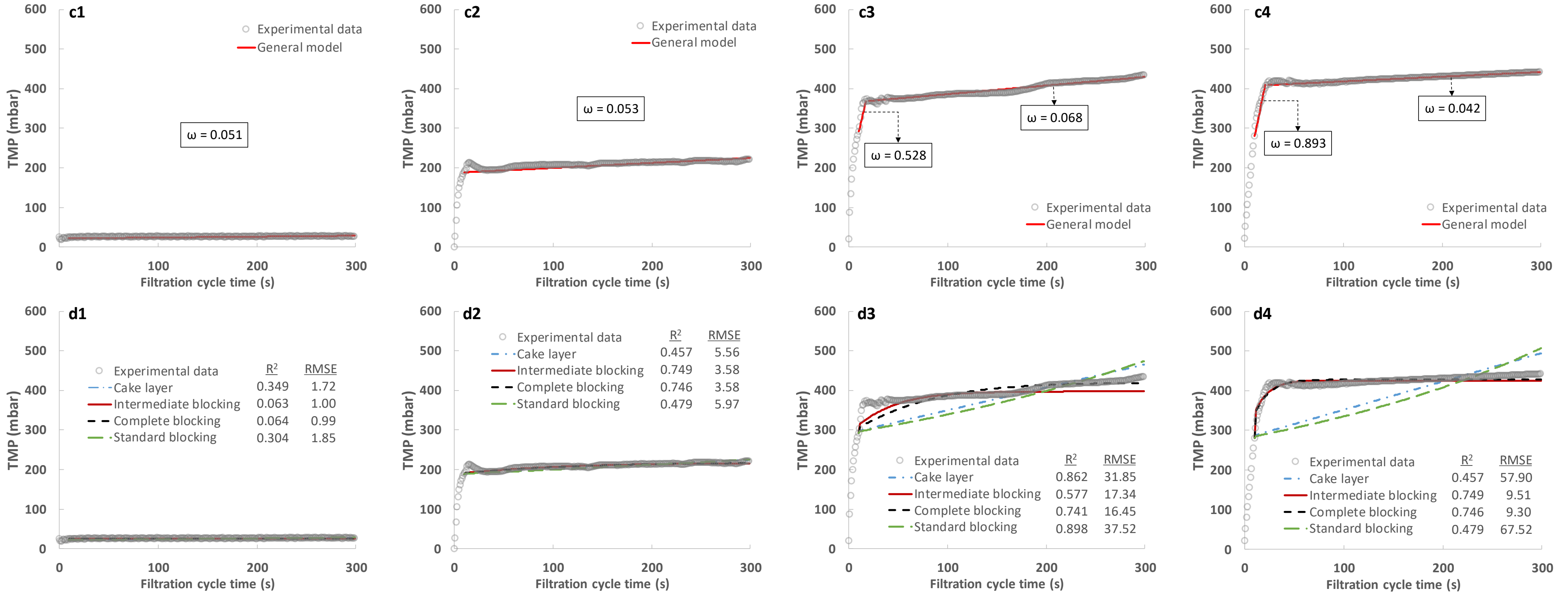

Figure 5.7. Examples of the fouling mechanism study for the TMP evolution during independent filtration cycles: (a) (c) general model; (b) (d) cake layer, intermediate blocking, complete blocking and standard blocking models. R^2 and RMSE represent the square of the Pearson correlation and the root-mean-square error, respectively. PSE: Effluent of the full-scale wastewater treatment plant primary settler.

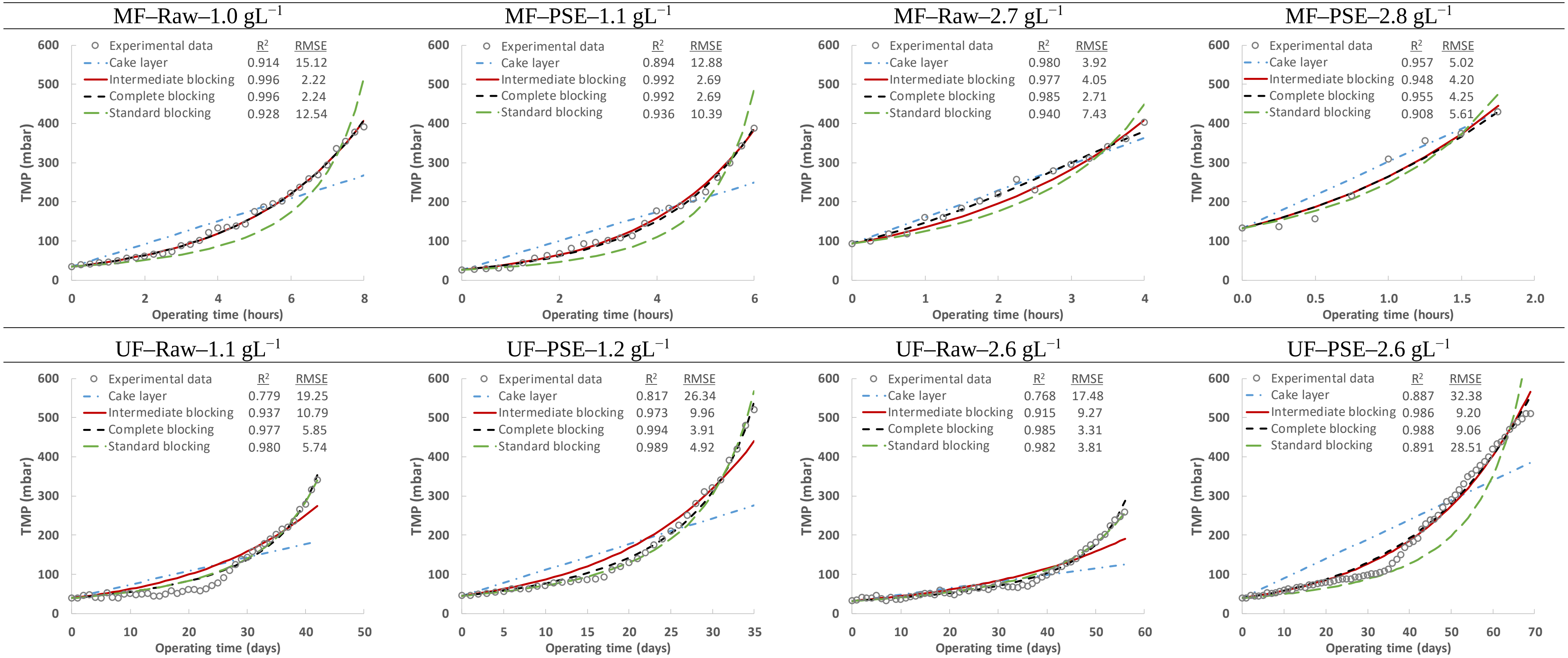

Figure 5.8. Fitting of the specific fouling mechanism models (cake layer, intermediate blocking, complete blocking and standard blocking models) to the hourly (MF membrane) and daily (UF membrane) TMP evolution obtained during the direct filtration of MWW. R^2 and RMSE represent the square of the Pearson correlation and the root-mean-square error, respectively. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

Table 5.5. Results of the general fouling model (Eq. 5.1) for the hourly (MF membrane) and daily (UF membrane) TMP evolution

Exp. nomenclature	***ω* value**
MF-Raw-1.0	0.978
MF-PSE-1.1	1.194
MF-Raw-2.7	0.893
MF-PSE-2.8	1.515
UF-Raw-1.1	0.717
UF-PSE-1.2	1.405
UF-Raw-2.6	0.689
UF-PSE-2.6	0.761

From the results obtained from the fouling study it can be concluded that two different mechanisms control fouling during direct MWW filtration. During the filtration stages, the predominant mechanism would be cake layer formation, which would slightly increase TMP as the filtration cycle advances due to the growth of the cake layer thickness. This is in agreement with the findings of Fujioka and Nghiem [5.20] and Ravazzini [5.28], who also determined that although pore blocking can occur in the early stage of every filtration cycle, the predominant fouling mechanism is cake layer formation. The low filtration-resistance cake layer observed in this study may be due to the relatively low operating TSS concentrations tested and/or due to the effectiveness of the employed fouling control strategies (air sparging and backwashing) to control its growth during each filtration cycle. On the other hand, a steady accumulation of irreversible organic fouling also occurs as the filtration process continues. In this case, the SMP substances or colloidal organic particles could produce intermediate/complete pore blocking of the membrane pores, in time reducing the useful membrane area dramatically. Fortunately, reduce the membrane pore size or promote the formation of a protecting cake layer onto the membrane surface by using raw MWW at higher solids concentrations showed as effective solutions to reduce this irreversible fouling growth.

5.4 Conclusions

The feasibility of applying the DMF concept for treating MWW was studied on a demonstration plant. The effect of MF and UF membrane technologies, treated influent (raw and PSE MWW) and operating TSS concentration (about 1 and 2.6 g L^{-1}) were evaluated. The main findings were as follows:

- A dramatically different performance was found, depending on the membrane used. Filtration periods of 2 – 8 hours were achieved with the MF membrane, while they were notably increased with the UF module (from 34 to 69 days). This extreme difference was due to the severe fouling when operating the MF membrane, which was dramatically reduced by the UF membrane due to the significantly lower pore size of the later compared to the former. The benefits observed when operating with a lower membrane pore size were associated to a reduction of the pore blocking propensity.
- Both the use of raw MWW and increased TSS concentration in the membrane module significantly benefitted the filtration performance with the UF module. This benefit could be associated to the increase in the average particle size, reducing the sludge propensity to block the membrane pores, and/or due to the formation of a more porous cake layer that acted as a fouling protector.
- The physical fouling control strategies used (air sparging and backwashing) proved to be ineffective in controlling fouling of the UF membrane, although they did have a significant impact on MF membrane fouling, extending the operating time from some hours (2 – 8 hours) to some days (5 – 6 days).
- The fouling evaluation showed that cake layer formation seemed to be the predominant fouling mechanism during each filtration cycle, representing the reversible fouling increase during filtration. However, as continuous filtration advanced, irreversible fouling appeared. This irreversible fouling could be related to intermediate/complete pore blocking in the case of the MF membrane, while it could also be produced by standard pore blocking in the case of the UF membrane. Organic matter represented more than 70% of this irreversible fouling in all the conditions evaluated.

5.5 Acknowledgements

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5.7 Supplementary material

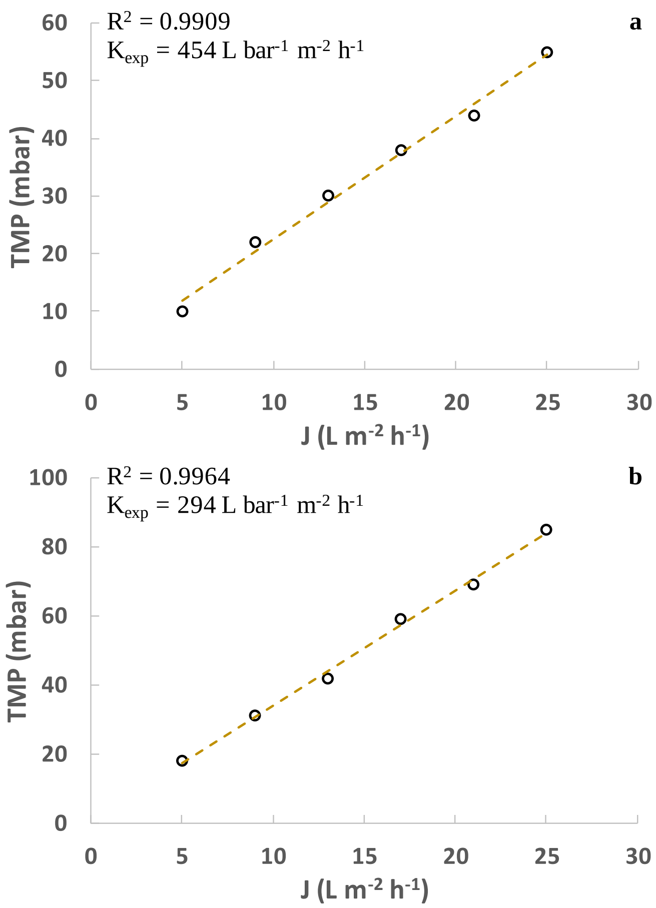

Figure 5.S1. Original permeability of virgin membranes: (a) MF membrane and (b) UF membrane. Dots represent experimentally determined permeability while the lines the linear fits. TMP: transmembrane pressure, J: Permeate flux, R^2: square of Pearson correlation, K_{exp}: membrane permeability average value.

CHAPTER 6. Evaluating Resource Recovery Potential and Process Feasibility of Direct Membrane Ultrafiltration of Municipal Wastewater at Demonstration Scale

Abstract

The aim of this work was to assess the feasibility of applying ultrafiltration membranes for the direct filtration of municipal wastewater. The effect of different influents (raw wastewater and the primary settler effluent of a municipal wastewater treatment plant) and operating solids concentration (about 1, 2.6, 6 and 11 g L^{-1}) was evaluated. Increasing the operating solids concentration sharply reduced membrane fouling, achieving a minimum of about 0.55 mbar per day when operating above 6 g L^{-1}. Filtering raw municipal wastewater also had a beneficial effect on the filtration performance, although only when operating at a low solids concentration (under 2.6 g L^{-1}). High quality permeates were obtained regardless of operating conditions, meeting European discharge standards for no sensible environments. High resource recoveries were achieved thanks to the direct filtration of wastewater, capturing about 80 – 85 and 20 – 40% of the influent COD and nutrients, respectively. The energy and carbon footprint balances showed promising results, achieving energy recoveries and carbon footprints of about 0.46 – 0.40 kWh per m^3 of influent municipal wastewater and -[0.19 – 0.16] kg CO_2-eq per m^3 of influent municipal wastewater when considering the process energy demands and potential energy recovery. Additionally, profits can be obtained from MWW treatment by this alternative, achieving about €0.035 per m^3 of influent MWW when operating at a flux of 10 LMH which entail a payback period for the initial membrane acquisition of 12.3 years.

Keywords

Direct membrane filtration; Ultrafiltration membranes; Resource recovery; Municipal wastewater.

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Prepared for submission

6.1 Introduction

Increasing water scarcity, climate change and the energy and food crisis are some of the most important issues to be addressed by developed counties in the coming years. Global water stress has increased in the last decades, even in regions with consistent rainfall [6.1] finding sources of high-quality water being a prominent policy around the world [6.2; 6.3]. On the other hand, the greenhouse gas (GHG) emissions forecast for 2030 are significantly higher than those necessary for the 2 ºC global temperature stabilization [6.4], not being on track for meeting the Paris Agreement. Other essential resources such as fossil fuels and nutrients are also impossible to sustain and scarcities are forecasted in the not-too-distant future. Due to these threats, a change on our economic model have turned mandatory and in consequence, our current non-renewable resources exploitation system needs to be abandoned in favour of new sustainable circular economic models [6.5]. This focus has gained a special interest in municipal wastewater (MWW) treatment due to all the potential resources that it contains. MWW is identified as a source of reclaimed water, energy and nutrients [6.6] although current treatments fail to recover a significant fraction of all of these essential resources. Aerobic-based MWW treatments (mainly based on conventional activated sludge (CAS)) are energy-intensive processes [6.7], increasing economic expenses. In fact, about 30 – 60% of all the energy consumed by a municipal wastewater treatment plant (WWTP) is associated with CAS treatment [6.8]. Additionally, an important fraction of the influent's energy is lost during the process due to the oxidation of the incoming organic matter to CO_2, hindering even more the energy balance. To overcome these issues, numerous authors have recommended the use of anaerobic systems to: (1) reduce sludge production due to the lower biomass yield of anaerobic biomass regarding aerobic one, (2) reduce the process energy demands by avoiding aeration requirements and (3) improve the energy balance by producing methane from influent organic materials [6.9]. However, due to the relatively low biomass growth rate of anaerobic microorganisms, anaerobic treatments are usually coupled with other processes such as membrane-based systems to reduce their space requirements.

The direct filtration of MWW by porous membranes (or direct membrane filtration (DMF)) has recently appeared as an anaerobic process associated technology to improve the feasibility of MWW treatment [6.10]. This alternative consists of filtering the influent MWW by a porous membrane to capture and concentrate the incoming particulate fraction before feeding the CAS system. Thanks to this previous separation, the energy required by the CAS system can be dramatically reduced since only the soluble organic matter oxidation would be required. Indeed, the subsequent CAS treatment could even be unnecessary depending on the DMF permeate quality [6.11]. On the other hand, the concentrated particulate fraction can be converted into methane via anaerobic digestion (AD), improving the energy balance of MWW treatment. Several studies have shown the potential benefits of applying the DMF for the treatment of MWW [6.10]. However, different issues need to be addressed before considering this alternative as a feasible option. Firstly, the most suitable operating conditions need to be determined to minimize the severe fouling that has been commonly reported when applying this alternative. In this respect, some studies seem to indicate that it is better to use ultrafiltration (UF) membranes instead of microfiltration due to their lower membrane fouling propensity when filtering untreated MWW [6.12] while different studies seem to conclude that using the primary settler effluent (PSE) as influent instead of raw MWW is irrelevant or even slightly harmful for the DMF fouling growth rate [6.12; 6.13]. The operating total suspended solids (TSS) concentration has also been reported to be related to the fouling growth rate [6.12]. Particularly, the fouling growth rate was found to be significantly reduced as the operating TSS was raised, although the benefits obtained would decrease as the TSS concentration is increased above certain concentrations [6.14–6.16]. Further studies are needed to confirm all these reported results and determine the optimum filtration operating conditions. Secondly, the quality of the generated permeate and the energy potential recovery of this technology needs to be studied to properly assess its potential benefits. Finally, the resource requirements for its operation (membrane replacements, membrane cleaning chemicals, etc.) also need to be considered to properly assess this technology’s impact.

The aim of this work was to determine the most suitable operating conditions to minimize membrane fouling and maximize resource recovery during direct MWW ultrafiltration. With that purpose, the influence of two different influents (raw or PSE) at different TSS concentrations (about 1, 2.6, 6 and 11 g L^{-1}) were evaluated. A preliminary energy, economic and carbon foot-print analysis of the process was also performed to assess the feasibility of applying the DMF concept to treat MWW.

6.2 Materials and Methods

6.2.1 Filtration pilot plant

The pilot plant consisted of a membrane tank (567-L working volume) fitted with an industrial ultrafiltration membrane system (PULSION® Koch Membrane Systems, 0.03-µm pore size, filtration area of 43.5 m^2). The membrane tank was equipped with a screw pump (PCM M Series, EcoMoineau™) to perform the vacuum filtration, collecting the produced permeate in a clean-in-place (CIP) tank (400-L working volume) to allow membrane backwashing when required. A blower (G-BH7, Elmo Rietschle) was used to inject air from below the membrane tank to control the cake layer formation via air scouring while a mixing pump (PCM M series, EcoMoineau™) was used to continually recirculate the membrane content to ensure complete mixing during filtration. Further details of the pilot plant can be found in [6.12].

The pilot plant was fed with two different influents during the study: (1) raw MWW (after classic screening and sieving, desanding and degreasing pre-treatment) and (2) PSE from a WWTP. Both influents were provided by the full-scale "Conca del Carraixet" WWTP (Alboraya-Valencia, Spain). Table 6.1 shows the characterization of both influents used. A 0.5 mm screen size roto-filter was used as pre-treatment to protect the membrane module from large abrasive materials. The treated wastewater was stored in an equalization tank (745-L working volume) to feed the membrane tank during filtration. An additional screw pump (PCM M Series, EcoMoineau™) was used to continuously feed the membrane module with fresh MWW according to the filtration requirements.

6.2.2 Operation and experimental plan

The filtration pilot plant was operated at a constant flux of 10 LMH for all the experiments, resulting in an HRT of 1.8 hours. However, after concluding some experimental periods (TSS concentration of 6 g L^{-1}) the flux was increased to 12.5 and 15 LMH for two weeks and one week, respectively, to test its effects on the fouling growth rate. Filtration was performed continuously, carrying out infinity filtration:relaxation cycles. Filtration time was set at 300 s, followed by 60 s of relaxation (operating filtration:relaxation ratio of 5:1). Air sparging and permeate backwashing were used as fouling control strategies during filtration. A low specific air demand per m^2 of membrane area (SAD_m) of 0.1 $m^3\ m^{-2}\ h^{-1}$ was set for all the experiments to minimize the process energy demands. Filtration was operated at a relatively low backwashing rate (120 s backwashing every 10 filtration:relaxation cycles, filtration:relaxation:back-washing ratio of 50:10:2) to maximize the production of recycled water.

Table 6.1. Influent municipal wastewater characteristics

Treated sewage		Raw	PSE
Parameter	**Units**	**Mean ± SD**	**Mean ± SD**
TSS	mg TSS L^{-1}	312 ± 99	132 ± 45
COD	mg COD L^{-1}	498 ± 116	197 ± 62
SCOD	mg COD L^{-1}	63 ± 28	57 ± 21
EPS	mg COD L^{-1}	84 ± 46	69 ± 39
SMP	mg COD L^{-1}	58 ± 11	61 ± 13
TN	mg N L^{-1}	48.1 ± 11.2	45.5 ± 8.5
NH_4^+	mg N L^{-1}	40.6 ± 10.6	37.2 ± 9.2
TP	mg P L^{-1}	6.3 ± 2.1	6.1 ± 2.2
PO_4^{3-}	mg P L^{-1}	3.8 ± 1.9	3.3 ± 1.4
Alk	mg $CaCO_3$ L^{-1}	342 ± 73	335 ± 67
pH	-	7.4 ± 0.7	7.6 ± 0.5

Filtration was performed with two different influents (raw and PSE MWW) and four TSS concentrations (about 1, 2.6, 6 and 11 g L^{-1}) to assess the most suitable operating conditions for direct MWW filtration (Table 6.2 summarizes all the experiments performed). At the beginning of each experimental period, the membrane was operated without waste production until reaching the required TSS concentration (hours or days, depending on the operating TSS concentration), before performing a continuous waste purge. Due to the variability of the influent TSS concentration, the membrane waste flow rate was continuously adjusted to maintain a constant operating solids concentration in the membrane module during filtration.

6.2.3 Analytical methods

The influent, membrane tank sludge and produced permeate were sampled twice a week to study resource recovery. Solids, total and soluble chemical oxygen demand (COD and SCOD), soluble biological oxygen demand (SBOD), total nitrogen (TN), total phosphorus (TP), ammonium (NH_4^+) and phosphate (PO_4^{3-}) concentrations were determined according to standard methods [6.17]. The soluble fraction of the collected samples was obtained by 0.45-µm pore size glass fibre membrane filters (Millipore, Merck). Other glass fibre membrane filter pore sizes (0.45, 0.10 and 0.05 µm, Millipore, Merck) were also used to treat the waste during some experimental periods to study the effect of membrane pore size on the generated permeate soluble pollutants concentration. The SMP concentration of the membrane tank concentrated sludge was also determined twice a week. The SMP content was attributed only to the protein and carbohydrates concentration, determining their content by a commercial total Protein Kit (Micro Lowry Peterson's Modification, Sigma-Aldrich) and the Dubois method [6.18], respectively. The main ions in influent, concentrated sludge and permeate (Cl^-, NO_3^- and SO_4^{2-} anions and Na^+, Ca^{2+}, Mg^{2+} and K^+ cations) were determined to assess their retention percentage by the membrane used and possible contribution to membrane fouling. An ionic chromatograph (919 IC, Metrohm) was employed, using a Metrosep Asupp5-250/4.0 retention column (Metrohm) to determine the anions concentration and a Metrosep C6-150/4.0 retention column (Metrohm) for the cations one. 3.2 mM of Na_2CO_3 and 1.0 mM of $NaHCO_3$ were the solvents for anion determination, using 3.0 mM of HNO_3 and 1.0 mM of $C_2H_2O_4$ as cation solvents. Finally, NaOCl (2000 ppm) and citric acid (2000 ppm) were used for membrane chemical cleaning. Cleanings were performed in two steps, studying membrane permeability recovery with tap water after each reagent cleaning to evaluate the source of the developed fouling (organic or inorganic). Further information on the membrane cleaning protocol used can be found in [6.12].

Table 6.2. Experimental plan

Exp. nomenclature	Sewage treated	Waste concentration (g L^{-1})
Raw-1	Raw	1.1 ± 0.5
Raw-2.6	Raw	2.6 ± 0.4
Raw-6	Raw	5.9 ± 0.4
Raw-11	Raw	11.1 ± 0.3
PSE-1	PSE	1.2 ± 0.4
PSE-2.6	PSE	2.6 ± 0.5
PSE-6	PSE	6.4 ± 0.2
PSE-11	PSE	10.6 ± 0.4

Note that the experimental nomenclature shows the treated MWW followed by the operating total suspended solids concentration used in each experiment.

6.2.4 Calculations

The DMF energy feasibility was assessed considering the energy demands of the equipment used (permeate pump, mixing pump and blower) and the potential energy production of the recovered organic matter. Since other energy requirements would also be necessary when operating a CAS system (feeding pumping, sludge to AD pumping, etc.), only the specific DMF equipment was considered. An average treatment volumetric flow rate of 36625 $m^3\ d^{-1}$ was considered for all calculations based on the *Conca del Carraixet* WWTP treatment flow rate. Equipment energy demands were calculated according to their appropriate theoretical equations (Eq. 6.1–6.3):

$$P_P = \frac{Q_P\ TMP_{ave}}{\eta_P} \tag{6.1}$$

Where P_P is the filtration permeate pump power requirements (W), Q_P is the pump volumetric flow rate ($m^3\ s^{-1}$), TMP_{ave} is the average transmembrane pressure during filtration (Pa) and η_P is the pump efficiency.

$$P_M = Q_M\ \rho\ g\ \frac{\left\{\left[\left(\frac{(L + L_{eq})\ f\ v^2}{D\ 2\ g}\right)_A + \left(\frac{(L + L_{eq})\ f\ v^2}{D\ 2\ g}\right)_I\right] + [z_1 - z_2]\right\}}{\eta_P} \tag{6.2}$$

Where P_M is the mixing pump energy requirements (W), Q_M is the mixing volumetric flow rate ($m^3\ s^{-1}$), ρ is the mixed liquor density ($Kg\ m^{-3}$), g is the acceleration of gravity ($m\ s^{-2}$), L and L_{eq} are the pipe length and equivalent pipe length (m), respectively, v is the liquor velocity ($m\ s^{-1}$), f is the friction factor, D is the pipe diameter and (z_1-z_2) is the height difference (m).

$$B = \frac{M\ R\ T_{gas}}{(\gamma - 1)\ \eta_B}\left[\left(\frac{p_I}{p_A}\right)^{\frac{\gamma-1}{\gamma}} - 1\right] \tag{6.3}$$

Where B is the blower power requirement (W) when considering adiabatic compression, M is the molar air flow rate ($mol\ s^{-1}$), R is the universal constant of gases ($J\ mol^{-1}\ K^{-1}$), T_{gas} is the gas temperature (K), γ is the heat capacity ratio, η_B is the blower efficiency and p_I and p_A are the impulsion and aspiration pressure (Pa), respectively. A pump and blower efficiency (η_B and η_B) of 0.65 was considered.

On the other hand, the following expression was used to estimate the energy production when transforming the recovered organic matter into biogas:

$$E_R = COD_R\, Y^{CH4}\, CV_{CH4}\, \eta_{CHP} \qquad 6.4$$

Where E_R is the energy recovery (kWh m^{-3}), COD_R is the recovered COD concentration in the membrane module (kg m^{-3}), Y^{CH4} is the theoretical anaerobic methane yield of MWW sludge (0.35 m^3 of methane per kg of COD), CV_{CH4} is the methane calorific power (9.13 kWh per m^3 of methane) and η_{CHP} is the CHP system methane electricity generation efficiency. A η_{CHP} of 35% was used considering the different CHP technologies currently available [6.19].

Concerning energy and process costs, €0.20 per kWh was estimated for the electricity cost according to current Spanish high-voltage electricity rates [6.20] while the membrane acquisition cost was estimated at €30 per m^2 of membrane area, according to the data provided by the supplier (Koch Membrane Systems). Membrane lifespan was estimated according to the cumulative tolerance to basic (NaOCl) membrane cleaning. In this case, a maximum NaOCl exposure concentration of 50000 ppm h^{-1} was considered, according to the data provided by the supplier (Koch Membrane Systems). Chemical costs were estimated at €10.04 per L at a volumetric concentration of 14% for NaOCl and €9.52 per kg for citric acid, according to the supplier's data [6.21]. Membrane cleaning requirements and membrane lifespan were calculated based on the fouling growth rate observed in this study. However, a minimum membrane cleaning period of once a year and a maximum membrane lifespan of 20 years were considered to obtain more realistic projections. For the carbon footprint calculations, a global warming potential (GWP) of 0.36 kg CO_2-eq per kWh of consumed energy was considered, in accordance with the GHG emissions ratio expressed in EcoInvent database [6.22].

6.3 Results and Discussion

6.3.1 Filtration performance

Fig. 6.1 and Table 6.3 show the results obtained during continuous direct MWW filtration. As can be seen in the figure, increasing TSS concentration had a marked beneficial effect on the filtration performance and extended the filtration lifespan before requiring chemical cleaning. When the TSS concentration was low (1 and 2.6 g L^{-1}) there were two clear fouling phases: a first stage with no important fouling increases and a second in which fouling consistently increased. However, a more consistent fouling growth rate was obtained as the TSS concentration was raised, as it was impossible to clearly differentiate between these two stages described, at least for the operating periods considered in this study. Regardless of the TSS concentration or fouling growth trend, all the fouling was found to be irreversible, which required removal by chemical cleaning. As explained in Sanchis-Perucho *et al.* [6.12] and reported in other membrane systems [6.23–6.25], increasing the operating TSS concentration could allow the rapid development of a thicker cake layer, which would protect the membrane surface from diverse pollutants with a high fouling potential (*e.g.* colloidal particles and SMP substances). In fact, as Sanchis-Perucho *et al.* [6.12] introduced and Table 6.3 corroborates, the operating SMP:TSS ratio sharply decreases as the TSS were raised in the membrane tank. Then, although a higher SMP concentration was reached at higher operating TSS concentrations, its hindering influence could have been minimized thanks to the formation of a quicker and thicker cake layer. As the results show, this strategy would be effective until achieving an operating TSS concentration at which a consistent cake layer could be obtained during filtration (between 2.6 – 6 g L^{-1} in this study), after which it would become irrelevant, since a further increase would only raise the cake layer thickness (see results for 6 – 11 g L^{-1}). However, although in this study it was necessary to raise the TSS concentration above 2.6 – 6 g L^{-1}, the required solids concentration to create this cake layer could be very different in other cases. Indeed, different authors [6.14–6.16] have reported similar fouling growth rates, regardless of the membrane module TSS concentration when operating MF membranes (from about 4 to 16, 2 to 11 and 1.3 to 7 g L^{-1}, respectively). Thus, this required solids concentration would be highly dependent on the influent characteristics, membrane pore size or filtration operating conditions.

Regarding the influent used, raw MWW showed better filtration performance when operated at low TSS concentrations (1 and 2.6 g L^{-1}), extending the filtration lifespan by several days (see Fig. 6.1). However, the benefits of using raw MWW instead of PSE were less marked as TSS concentration increased, with no appreciable difference when the TSS concentration was raised above 6 g L^{-1}. In this case, using raw MWW seems to show better performance due to the larger average size of the influent particles, reducing their propensity to block the membrane pores and increase the porosity of the cake layer [6.12]. Then, as in the former case, after achieving a TSS concentration at which a consistent cake layer is formed during filtration, the influent used would lose relevance. In this respect, [6.26] also concluded that the influent treated was irrelevant when filtering raw and PSE MWW with UF membranes.

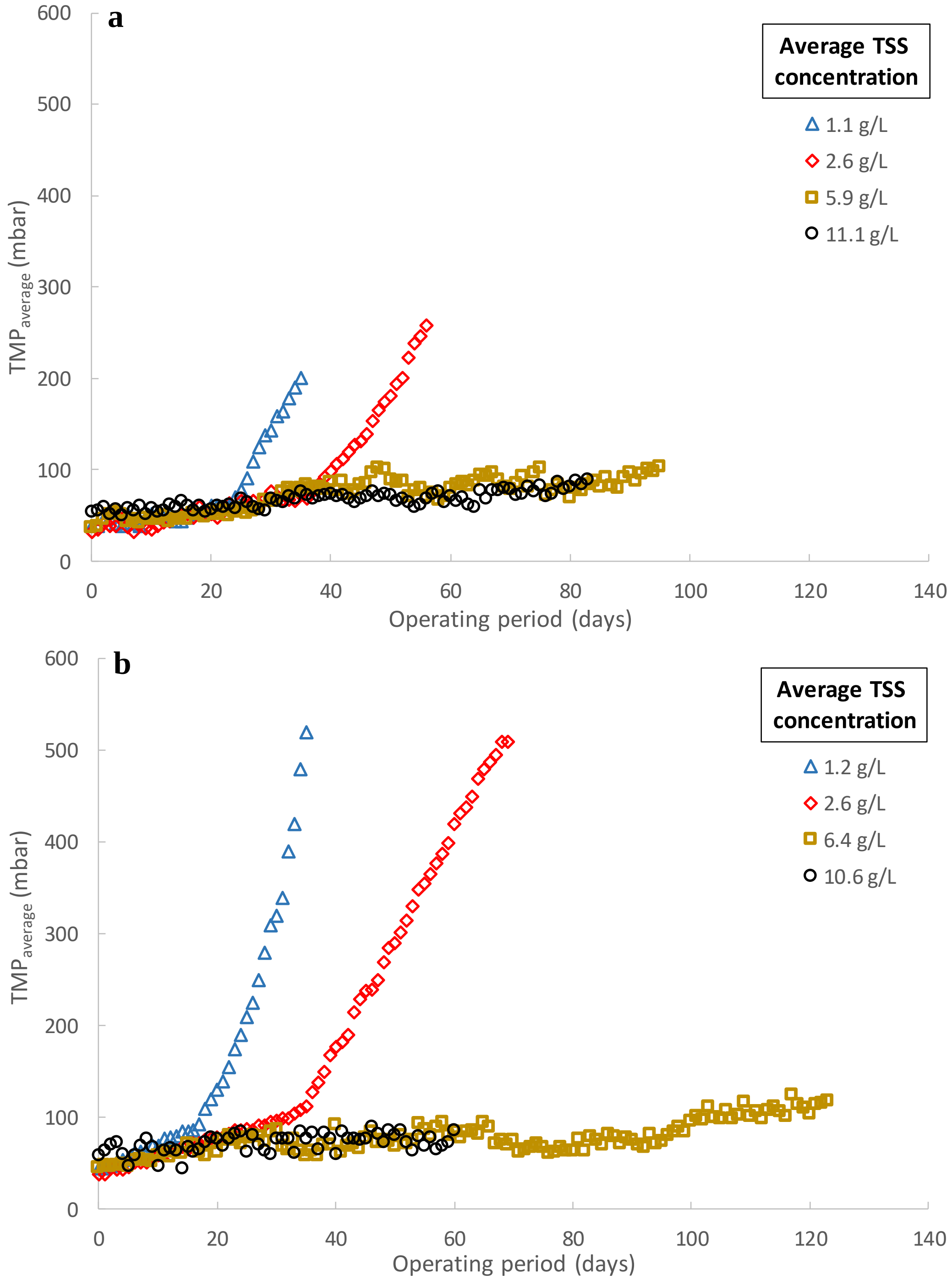

Figure 6.1. Direct municipal wastewater filtration performance: (a) Raw and (b) PSE. Results concerning 1 and 2.6 g L^{-1} were subtracted from Sanchis-Perucho *et al*. [6.12]. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

Table 6.3. Irreversible fouling growth rate during filtration

Sewage treated	Waste concentration (g L^{-1})	SMP concentration (mg L^{-1})	Sludge SMP:TSS ratio	Duration of stable period (days)	Fouling growth rate (mbar day^{-1})	
					1st slope	2nd slope
Raw	1.1	96	86.5	22	1.81	13.22
	2.6	109	42.6	36	1.13	9.31
	5.9	127	21.5		0.54	
	11.1	142	12.8		0.56	
PSE	1.2	78	65.0	16	2.87	21.58
	2.6	101	39.1	34	1.99	12.01
	6.4	116	18.1		0.50	
	10.6	97	9.2		0.62	

Data concerning 1 and 2.6 g L^{-1} sludge concentrations can be found in Sanchis-Perucho *et al*. [6.12]. 1st slope refers to the first stable filtration period when the fouling growth between two consecutive days was inferior to 5 mbar day^{-1}. 2nd slope refers to the rapid-fouling-growth-rate-increase period when the fouling growth between two consecutive days was superior to 5 mbar day^{-1}.

Since the fouling growth rate was sharply reduced when operating above 6 g L^{-1}, an increase in the operating flux was tested to study its effect on fouling propensity (see Table 6.4). Unfortunately, dramatic irreversible fouling growth occurred when increasing the operating flux, achieving fouling growth rates about 11 and 25 times higher than those obtained when operating at 10 LMH for 12.5 and 15 LMH, respectively. This important rise in the fouling growth rate indicates that the cake layer formed could have been compressed to some degree, increasing its density and reducing its permeability. Additionally, since raising the permeate flux also means a higher treated sewage ratio, more pollutants were able to reach the membrane surface and increase fouling. However, when calculating fouling growth per m^3 of water treated (see Table 6.4), important higher irreversible fouling increases were also obtained for the higher tested fluxes (about 9 and 17 times higher than those obtained when operating at 10 LMH for 12.5 and 15 LMH, respectively), so displaying the sharply unfavourable effects of increasing the operating flux on fouling. Despite the results obtained, it should be taken into account that the membrane was operated in the middle-term before these experiments (for about 95 and 123 days for the raw and PSE MWW, respectively), so that the results obtained could have been affected to some degree by the previous fouling and could not fully represent the expected fouling growth rate when operating at the increased fluxes tested. Further experiments on the fouling growth rate during all the filtration process at different fluxes are therefore required to confirm these results.

Table 6.4. Irreversible fouling growth rate during filtration for an operating TSS concentration of around 6 g L^{-1}.

Sewage treated	Flux (LMH)	Fouling growth rate (mbar day^{-1})	Fouling growth rate (mbar m^{-3} of treated water)
Raw	10	0.54	0.052
	12.5	4.43	0.339
	15	11.91	0.761
PSE	10	0.50	0.048
	12.5	5.30	0.406
	15	12.25	0.782

A two-step chemical cleaning was performed on the membrane module after every experimental essay, using NaOCl as basic solution and citric acid. According to the results obtained from after cleaning (see Fig. 6.2), organic matter was found to be the main fouling promotor, representing about 70 – 85% of the irreversible fouling, regardless of the experimental conditions. These results are in agreement with other studies [6.27–6.29] that identified organic fouling as the main culprit in minimizing the filtration lifespan.

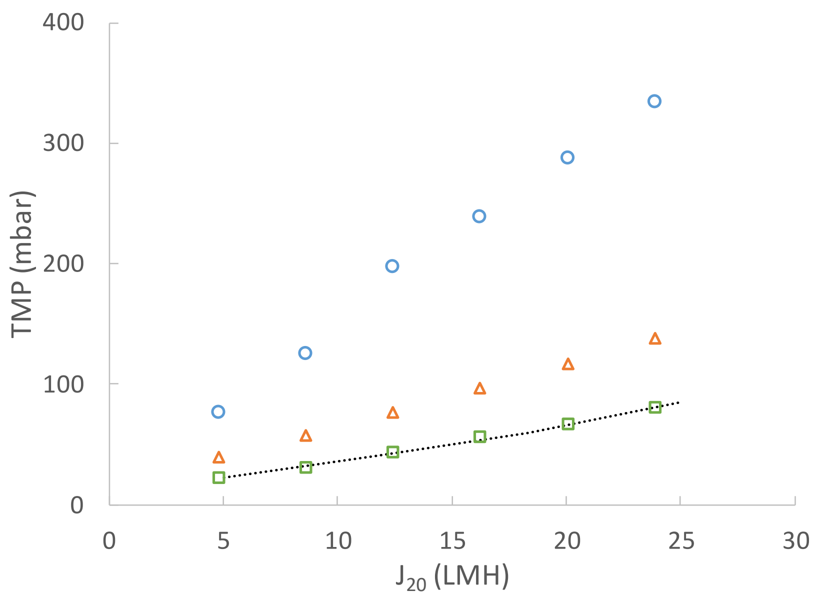

Figure 6.2. Example of membrane permeability recovery after each membrane cleaning. Raw MWW at a TSS concentration of 6.4 g L^{-1} was employed as operating conditions. Tap water was used to determine the membrane resistance. — original membrane resistance; ○ Membrane resistance after the operating period; △ Membrane resistance after the basic cleaning; □ Membrane resistance after the acid cleaning.

6.3.2 Resource recovery and permeate quality

Table 6.5 shows the resource recovery and permeate quality obtained during the filtration periods tested. A similar permeate quality was obtained for all the pollutants analysed, regardless of the test conditions. In fact, the slight differences among filtration periods were more related to the influent MWW's intrinsic variance than with the change of operating conditions. It was therefore concluded that the cake layer formed was not related to the pollutants captured, at least for the influents and TSS concentrations tested, the membrane itself being the main source of resource capture. High organic matter recoveries were reached thanks to membrane filtration, achieving a capture percentage of around 70 – 85% of the influent COD. These high recovery rates were due to the capture of all the particulate fraction of the influent MWW as well as a significant portion of the soluble fraction. Indeed, about 10 – 20% of the influent SCOD was captured by the membrane used, also observing this capture effectivity in the influent SMP concentration (see Table 6.5). Since the capacity to capture soluble substances was independent of the operating TSS concentration, and an accumulation of SCOD and SMP was detected in the soluble fraction of the waste, the possible adsorption of soluble organics by the particulate material was considered negligible. Consequently, the soluble organic matter retention capacity obtained was attributed to the membrane characteristics (*i.e.* pore size and material). Soluble material captured was mainly assumed as colloidal particles larger than the membrane pore size although other strictly soluble compounds could also be captured due to their high viscosity and sticky proprieties or other interactions between them and the membrane material. This phenomenon could be especially important, since these substances (mainly SMP related) could be an important source of membrane fouling, reducing filtration feasibility. However, since no filtration issues were found, even though a large accumulation of SMPs was reached as the TSS concentration was raised, the impact of these substances accumulation in the membrane tank was found to be irrelevant. On the other hand, a significant decrease of organic matter recovery was appreciated when the membrane tank's solids retention time (SRT) was above 3 days, which continued to fall as the SRT was raised. This reduced COD recovery was attributed to the development of aerobic bacteria, which could easily proliferate in the membrane module conditions (continuous input of substrate and oxygen). In this respect, several authors have reported the severe impact of aerobic microorganism on organic matter recovery when applying direct MWW filtration, reporting losses of between 10 and 50%

of influent COD when operating at SRTs of between 1 and 8 days [6.14–6.16; 6.30]. Additionally, a clear mismatch in the COD, SCOD, EPS and SMP mass balances was obtained when the SRT was above 3 days (see Table 6.5), indicating a decline in the output of organic matter mass flow rate (waste + permeate) in relation to the input, which represents the losses of carbon as CO_2 due to aerobic bacterial activity. According to these results, operating under 3 days SRT is strongly recommended to prevent losses of the influent organic matter and maximize process energy recovery. It is important to highlight that thanks to the higher solids content in raw MWW, higher TSS concentrations were reached without the presence of aerobic bacteria. Since increasing the operating TSS concentration as much as possible is an attractive option to reduce the space requirements of this system (whenever it does not negatively affect filtration energy demands or membrane fouling), using raw MWW can be considered a better option for direct MWW filtration. Nonetheless, further studies are required to assess long-term membrane fouling by raw and PSE MWW at different permeate fluxes to confirm this. Reducing membrane tank volume as much as possible would also be an interesting design strategy to reduce the space demands and aerobic bacterial proliferation.

Regarding nutrients recovery, TN and TP captures of about the 20 – 40% were obtained. Recoveries were significatively lower in this case due to the smaller percentage of these nutrients in the influent particulate fraction, which is the main fraction susceptible to recovery by used membranes. A light mismatch in the ammonium balance was generally obtained, which could indicate that a slight amount of the influent ammonium was stripped to the environment together with the injected air as ammonia. On the other hand, small phosphate captures were obtained during filtration (around 5 – 20% of the influent phosphate), achieving a high accumulation of this soluble ion in the waste as the TSS concentration was raised (see Table 6.5). Since the membrane pore size (0.03 µm) should not be able to retain ions, this slight phosphate capture capacity was attributed to the membrane material characteristics, such as its intrinsic superficial charge or hydrophobicity, which could reject this anion in a certain degree. At this respect, a superficial negative charge has been reported when evaluating the zeta potential of pure PVDF membranes due to the preferential adsorption of counter-ions onto hydrophobic surfaces under water [6.31], which could be the cause of this slight phosphate retention in the membrane tank. Moreover, a significant maladjustment of the TP balance was obtained, indicating that a fraction of the influent TP was not properly recovered. Since the phosphate balance did not show this significant mismatch, the TP losses were mainly attributed to a retention of a certain amount of the influent particulate phosphorous in the membrane tank. In this respect, other membrane-based reactor studies have also reported significant TP losses when operating, which could be related to chemical precipitation of the influent phosphate or direct retention of the particulate fraction of the influent TP [6.32]. In any case, this issue should be minimized as far possible to enhance nutrient recovery.

To unveil the effect of membrane pore size on the capture of some of the influent's soluble pollutants (SCOD, NH_4^+ and PO_4^{3-}), the waste was filtered by different membrane filters (pore sizes 0.45, 0.10 and 0.05 µm) once a week in some experimental periods, to determine the remnant soluble pollutants concentration in each permeate and also compare the results with the concentrations obtained during pilot plant filtration (see Table 6.6). The SCOD concentration clearly decreased as membrane pore size was reduced. As mentioned above, this soluble material capture was mainly attributed to the retention of colloidal particles by the membrane, a reduction in the membrane pore size able therefore to increase their capture ratio. On the other hand, no significant differences were found in nutrients concentration in the permeates obtained from each membrane filter. Unlike the organic matter, these results indicate that, as expected, the membrane pore size range used did not have the capacity to capture ions, the phosphate recoveries obtained in this study being more related to the interaction between the membrane material and the present ions than with membrane pore size. The concentration of the main ions in the membrane waste and permeate were also determined every 15 days to study possible phosphate precipitation during filtration or other sources of inorganic membrane fouling. As can be seen in Table 6.7, residual differences were obtained for all the ions studied. Since magnesium is required for the precipitation of phosphate as struvite, these results would confirm the previous conclusions that considered chemical precipitation as a modest source of fouling.

Table 6.5. Resource recovery and permeate quality

Exp.	SRT (days)	Stream	TSS (g L^{-1})	COD (mgCOD L^{-1})	SCOD (mgCOD L^{-1})	TN (mgN L^{-1})	NH_4^+ (mgN L^{-1})	TP (mgP L^{-1})	PO_4^{3-} (mgP L^{-1})	EPS (mgCOD L^{-1})	SMP (mgCOD L^{-1})
Raw-1	0.26	Waste	1.11	1692	99	83	31	11.9	4.6	169	100
		Permeate			56		27		3.2		63
		Recovery* (%)	100	81.6	35.4	42.7	22.4	41.5	24.8	39.8	33.5
		Balance** (%)		9.0	-5.7	8.2	8.6	10.8	-3.5	8.4	-7.0
Raw-2.6	0.58	Waste	2.56	3871	126	171	38	30.3	6.3	294	109
		Permeate			57		34		3.5		59
		Recovery* (%)	100	87.8	23.2	42.4	11.2	54.0	17.5	35.3	18.5
		Balance** (%)		-6.6	-4.6	-8.2	10.5	6.4	2.1	10.4	4.0
Raw-6	1.71	Waste	5.92	8970	172	422	46	38.0	17.3	498	127
		Permeate			58		45		4.7		57
		Recovery* (%)	100	81.2	11.6	31.4	3.7	24.7	17.6	24.0	8.2
		Balance** (%)		6.8	-1.2	-8.9	11.3	16.6	11.0	12.8	7.8
Raw-11	3.28	Waste	11.14	17250	188	523	46	68.3	28.8	703	142
		Permeate			53		44		4.8		54
		Recovery* (%)	100	82.8	6.1	21.1	2.0	23.6	15.6	18.0	5.2
		Balance** (%)		5.9	17.7	1.0	10.5	13.1	15.1	11.7	6.8

Table 6.5. Cont.

Exp.	SRT (days)	Stream	TSS (g L^{-1})	COD (mgCOD L^{-1})	SCOD (mgCOD L^{-1})	TN (mgN L^{-1})	NH_4^+ (mgN L^{-1})	TP (mgP L^{-1})	PO_4^{3-} (mgP L^{-1})	EPS (mgCOD L^{-1})	SMP (mgCOD L^{-1})
PSE-1	0.48	Waste	1.23	1454	108	116	38	15.3	5.0	187	78
		Permeate			57		33		4.1		46
		Recovery* (%)	100	78.4	14.9	28.6	9.6	23.6	11.3	29.4	13
		Balance** (%)		-9.1	6.6	2.6	10.1	13.2	-4.4	5.1	8.2
PSE-2.6	1.28	Waste	2.63	4005	137	234	39	45.4	8.1	233	101
		Permeate			49		35		3.4		52
		Recovery* (%)	100	82.2	10.4	26.2	4.6	29.1	7.5	16.6	7.8
		Balance** (%)		2.6	18.6	-3.5	13.0	19.9	10.3	-2.1	-1.2
PSE-6	3.22	Waste	6.4	9550	186	655	35	86.8	13.9	537	116
		Permeate			47		31		3.5		51
		Recovery* (%)	100	70.0	2.3	19.5	1.2	19.2	4.3	11.5	2.7
		Balance** (%)		3.3	51.2	7.7	14.8	19.1	9.3	12.4	14.0
PSE-11	6.01	Waste	10.6	17950	220	1058	37.4	143	26.5	549	97
		Permeate			64		28.5		3.7		44
		Recovery* (%)	100	54.7	1.8	19.5	0.8	19.3	5.2	7.4	1.4
		Balance** (%)		20.6	32.8	14.1	22.0	16.9	2.3	17.1	16.1

*Recovery was calculated based on mass flow rates.
**Balance term was calculated as the difference between influent and effluent mass flow rates (influent - effluent), using the influent as base for calculating the relative error. Balance was considered close when less than 10% errors were obtained.

Table 6.6. Changes in the permeate SCOD, NH_4^+ and PO_4^{3-} concentrations regarding the filter pore size for samples collected during the filtration of PSE at about 2.6 g L^{-1} of TSS concentration

Filter/membrane pore size (μm)	**Notes**	**SCOD (mgCOD L^{-1})**	**NH_4^+ (mgN L^{-1})**	**PO_4^{3-} (mgP L^{-1})**
0.45	Cellulose commercial filter used during all this study	146 ± 44	39.5 ± 8.5	6.4 ± 1.4
0.10	Cellulose commercial filter	98 ± 29	37.9 ± 8.8	6.2 ± 1.6
0.05	Cellulose commercial filter	76 ± 27	38.2 ± 9.2	6.2 ± 1.2
0.03	Membrane used in the pilot plant (PVDF)	48 ± 8	37.6 ± 7.7	2.2 ± 0.9

Table 6.7. Membrane retention capacity onto main ions during all the experimental periods

	Ion	**Waste**	**Permeate**	**Retention (%)**
Cations	Na^+ (mgNa L^{-1})	108.3 ± 43.4	99.1 ± 37.1	8.5
	Ca^{2+} (mgCa L^{-1})	124.8 ± 44.8	122.6 ± 41.5	1.8
	Mg^{2+} (mgMg L^{-1})	35.2 ± 13.1	33.4 ± 11.8	5.2
	K^+ (mgK L^{-1})	14.8 ± 11.1	12.9 ± 7.4	12.8
Anions	Cl^- (mgCl L^{-1})	192.7 ± 30.0	185.6 ± 28.1	3.7
	NO_3^- (mgN L^{-1})	7.8 ± 9.8	7.7 ± 8.4	0.3
	SO_4^{2-} (mgS L^{-1})	103.0 ± 13.8	97.9 ± 12.0	4.9

From results obtained it can be concluded that the permeate generated during direct MWW filtration is of high quality, almost meeting the European discharge demands. In fact, since solids, COD and BOD restrictions are meet (see Table 6.5 and Table 6.8), this reclaimed water could be used directly when not applied to sensible mediums. Its small soluble nutrients content could also be of special interest for agriculture purposes or could even be commercially valorised by recovering them via ionic exchange or osmosis filtration. On the other hand, the high COD concentration archived with the relatively high operating TSS concentrations tested (about 18 – 9 mgCOD L^{-1}) would turn the membrane waste into an interesting stream for feeding anaerobic bioreactors. A higher fraction of the influent nutrients could be recovered from the waste, which after AD mineralization could also be made use of in agriculture. Despite these potential benefits, further studies are required to determine the best energy, economic and space requirements scenario, considering all the MWW treatment processes together.

Table 6.8. Average BOD achieved in the permeate during all the experimental periods

Sewage treated	BOD_5 (mgBOD L^{-1})	BOD_L (mgBOD L^{-1})
Raw	17 ± 2	21 ± 3
PSE	16 ± 3	18 ± 5

6.3.3 Process feasibility

The feasibility of direct MWW filtration was assessed from three points of view: energy, economy and the carbon footprint of studied alternative. Fig. 6.3 shows the filtration energy demands together with the energy recovery potential. As usual, air scouring was found to be the major energy requirement during filtration, as MBR studies generally report [6.33]. Nevertheless, thanks to the relatively low SAD_m used in this study, little energy would be required in this case. The filtration and mixing energy demands were relatively low for DMF, achieving total energy requirements of about 0.10 kWh per m^3 of influent MWW treated, so that significant energy could be recovered by applying this technology. In this case, energy outputs of about 0.53 and 0.24 kWh per m^3 of influent MWW treated could be obtained when using raw and PSE MWW, respectively. The lower energy recovery obtained when using PSE was due to the lower influent organic matter, which would reduce the energy recovery potential by the same percentage as the influent organic matter recovered in the primary settler (about 55%). However, if the organic matter captured by this pre-treatment is considered, similar energy recovery potentials are obtained (see Fig. 6.3), so that when insignificant aerobic bacterial activity is found, similar energy performance can be expected, regardless of the influent used.

From an economic point of view, important differences were obtained that depended on the operating flux (see Fig. 6.4). As can be seen in Fig. 6.4a, the initial economic investment (CAPEX) can be significantly reduced by using higher operating fluxes, since smaller membrane areas would be required for treat the same MWW influent. However, due to the significant rise in fouling growth rate as the permeate flux is increased, the operating costs (OPEX) sharply increase, dramatically reducing the feasibility of this alternative (see Fig. 6.4b). In fact, not only the amount of chemicals required for membrane cleaning are importantly increased due to higher fouling but membrane replacement costs also rise significantly due to the roused chemical cleaning necessities. Using moderate/low permeate fluxes is thus recommended when directly filtering MWW to reduce fouling and in consequence, operating costs. This recommendation was also reported by Ravazzini *et al.* [6.13] when filtering raw and PSE MWW with UF membranes. Nevertheless, as mentioned above, since short-term experiments were performed for the 12.5 and 15 LMH permeate fluxes, the fouling growth rate obtained in this study may not be representative of the real process, so that further studies are needed to confirm the direct filtration OPEX increases when raising the permeate flux.

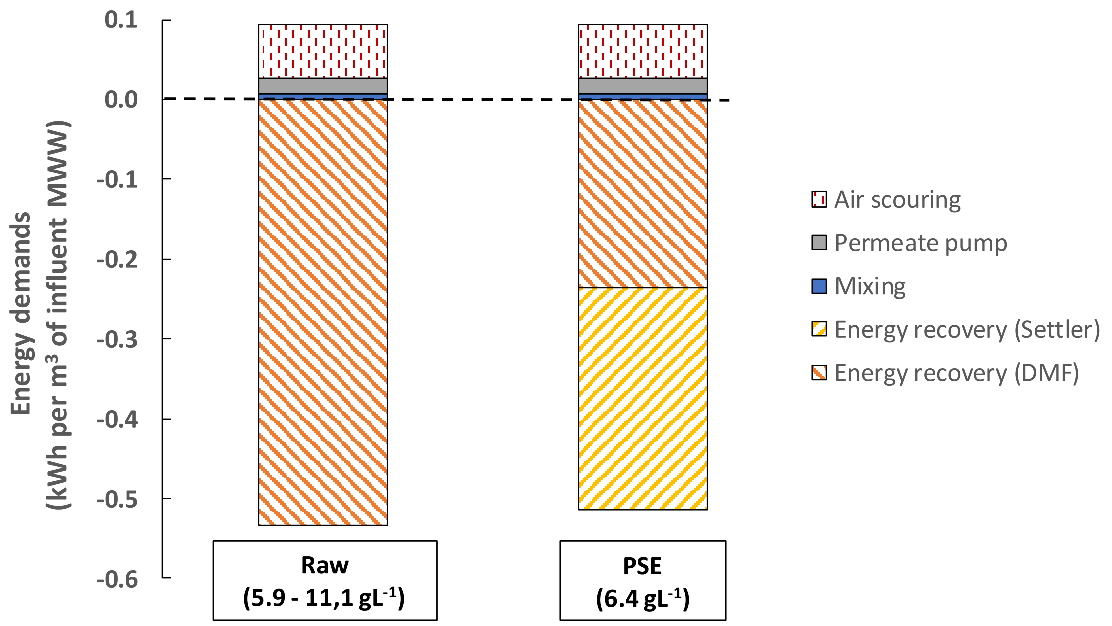

Figure 6.3. DMF average energy demands and potential energy recovery. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

Comparing the results obtained in this study with the CAPEX and OPEX reported by CAS technology promising preliminary results were found. A complete CAS CAPEX (equipment + direct and indirect construction costs) would ascend above M€25.6 (design influent flow rate 30000 $m^3 d^{-1}$), achieving around €0.019 per m^3 of influent MWW when considering the OPEX (operation + maintenance) [6.34]. According to the results obtained, the MWW direct filtration CAPEX would be above M€[3.1 – 4.7] only for membrane acquisition (values calculated under the same calculus base, *i.e.* 30000 $m^3 d^{-1}$). Since the membrane costs alone represent around 15% of the complete CAS CAPEX, a significantly higher DMF CAPEX than those involved in CAS technology can be expected when including all the additional equipment (membrane modules, pumps, blowers, valves, etc.) which would make the DMF a no competitive technology. However, when considering OPEX, promising results were found regarding DMF, achieving economic benefits of around €0.035 per m^3 of influent MWW when operating at 10 LMH in this study, which were reduced to €0.001 per m^3 of influent MWW for a flux of 12.5 LMH and represented a cost of €0.109 per m^3 of influent MWW when flux ascended to 15 LMH. In consequence, since a significant profit could be obtained from moderate flux DMF while treating MWW, the initial membrane investment can be compensated, achieving in this study a payback period of 12.3 years when only considering initial investment and yearly profit. In addition to the above discussed, other considerations should be taken into account to properly determine the economic feasibility of this alternative treatment. DMF OPEX can be significantly increased when including other auxiliary costs (*e.g.* equipment maintenance) thereby reducing the economic benefits of this alternative and increasing the initial investment payback to no competitive values. However, the required chemical prices considered in this study were in accordance with a low/middle-volume commercial supplier (maximum order of about 25 L or kg of chemicals). The cost of these chemicals could therefore be significantly reduced in a real scenario when considering industrial prices. In addition, since the amount of chemicals required is related to the volume of the membrane module (a specific concentration is required during membrane cleaning), a reduction in its free space would also have a beneficial impact on the cost of chemicals. On the other hand, since the important fouling growth rate during direct MWW filtration is the main source of its economic expenses, implementing effective countermeasures would provide enormous benefits for the process's economic feasibility. In this respect, numerous authors have reported promising results regarding possible DMF effective fouling control strategies, such as coagulant dosing, enhanced chemical cleaning and membrane vibration [6.15; 6.27; 6.35]. This means the CAPEX and OPEX reported here could be reduced by implementing these alternatives and thus improve its competiveness.

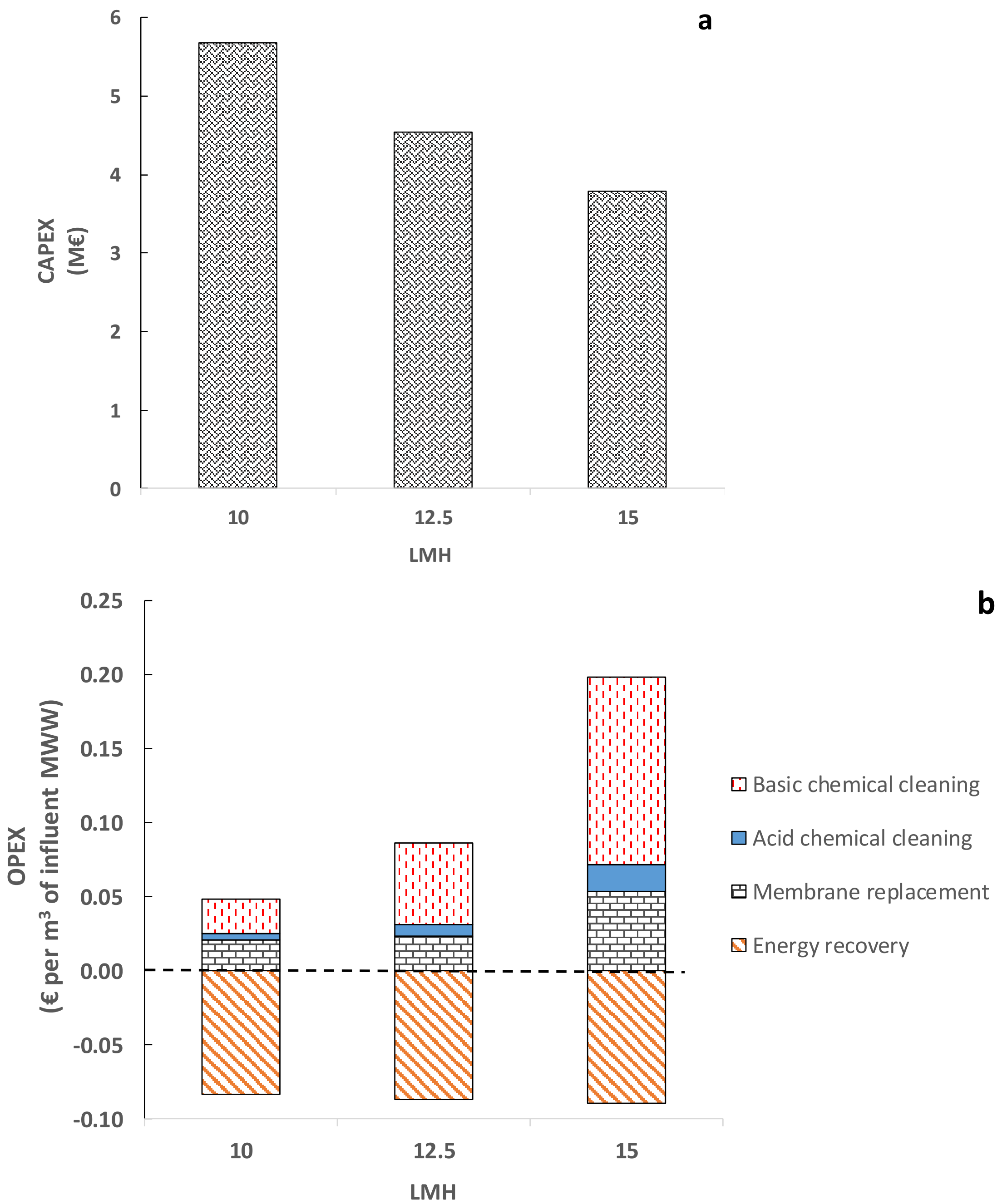

Figure 6.4. DMF average economic impact: (a) initial membrane acquisition investment and (b) operational costs.

When considering the process's carbon footprint, relatively low environmental impacts were achieved. In fact, comparing the results obtained in this work with others currently used MWW treatment technologies, promising benefits can be observed (see Table 6.9). Significantly lower energy demands were found by the DMF regarding aerobic treatment alternatives. Similarly, a lower carbon footprint was achieved by the proposed alternative, since less energy is required for MWW treatment. If the energy recovery potential of these technologies is considered, even better energy demands and carbon footprints could be obtained by the DMF versus aerobic systems, since a relevant fraction of the influent organic matter is lost in the second ones as CO_2 during MWW treatment. Since the DMF could avoid this loss of organic matter, the energy recovery potential would be much notorious also reducing the carbon footprint thanks to the consumption of less fossil fuel energy. Considering the energy that could be produced in a subsequent AD, total energy demands of -[0.46 – 0.40] kWh per m^3 of influent MWW treated and carbon footprints of -[0.19 – 0.16] kg CO_2-eq per m^3 of influent MWW were obtained in this study. These results highlight the potential of DMF for the treatment of MMW, however numerous aspects should be taken into account. The permeate produced could require post-treatment to reduce its soluble nutrients content in some circumstances. In this case, the energy demands of this treatment should be taken into account for the complete MWW management as well as the economic benefits of recovering these nutrients. The treatment of the waste stream should also be considered in the overall process energy demands. In this respect, any necessary change on the WWTP's sludge thickening and anaerobic digester should be considered. Finally, the environmental impact of the membranes used (with replacements) and cleaning chemicals need to be considered, together with all the additional materials that could be required for the process (pumps, blowers, etc.). Further studies considering the complete environmental impact and economic investment (*i.e.* LCA and LCC studies) are required to properly determine the DMF potential for treating MWW.

Table 6.9. Energy demand and carbon footprint of different technologies for MWW treatment

Treatment system	**Energy demands (kWh m^{-3})**	**Carbon footprint (kg CO_2-eq m^{-3})***	**Reference**
Direct membrane filtration	0.09 – 0.10	0.038 – 0.040	This study
Conventional activated sludge	0.30 – 0.60	0.093 – 0.186	[6.36; 6.37]
Extended aeration	0.34 – 0.82	0.150 – 0.254	[6.38]
Aerobic MBR	0.50 – 1.00	0.155 – 0.310	[6.39; 6.40]

*Calculated considering reported energy demands.

6.4 Conclusions

The feasibility of using UF membranes for direct MWW filtering was studied. The effect of using raw and PSE MWW influents and operating TSS concentration (about 1, 2.6, 6 and 11 g L^{-1}) was assessed. The main findings were as follows:

- Increasing the operating TSS concentration was found to sharply reduce membrane fouling. Operating at TSS concentrations of between 6 – 11 g L^{-1} is therefore recommended, according to the results obtained in this study. Filtering raw MWW instead of PSE also showed a slight fouling growth rate reduction. However, since this beneficial effect was only found when low TSS concentrations were used, the influent fed to the membrane module was considered irrelevant when only considering filtration performance. The fouling developed during filtrations was identified as irreversible organic fouling, representing about the 80 – 85% of total fouling, regardless of the operating conditions studied.
- A high quality permeate was produced regardless of the operating conditions tested in this study, meeting European quality standards when considering no sensible environments. High resource recoveries were achieved thanks to the DMF, capturing about 80 – 85 and 20 – 40% of the influent COD and nutrients, respectively. A SRT of 3 days was found as the limit to avoid the proliferation of aerobic microorganisms. Using raw MWW or reducing the design membrane tank volume were thus considered to be interesting alternatives to achieve higher TSS concentrations during filtration.
- The energy and carbon footprint balances revealed promising results for the DMF of MWW, achieving energy recoveries and carbon footprints of about 0.46 – 0.40 kWh and -[0.19 – 0.16] kg CO_2-eq per m^3 of influent MWW when considering the process energy demands and potential energy recovery. Additionally, profits can be obtained from MWW treatment by this alternative, achieving about €0.035 per m^3 of influent MWW when operating at a flux of 10 LMH which entail a payback period for the initial membrane acquisition of 12.3 years. Further studies focused on developing effective fouling control strategies when directly filtering MWW are required to enhance the potential applicability of this alternative.

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CHAPTER 7. Building a Simple and Generic Filtration Model to Predict Membrane Fouling in the Long-Term when Treating Municipal Wastewater

Abstract

This study aimed to propose a simple and generic model to predict membrane fouling in municipal wastewater (MWW) treatment processes. Specifically, this work was conducted using data from a direct membrane filtration demo-system (middle/long-term filtration periods of about 35 – 124 days) to calibrate the model. Two influents were treated by the cited demo-system: the pre-treatment step and the primary settler effluent of a full-scale MWW treatment facility. A resistance-in-series structure was proposed, simplifying fouling as the consequence of two different mechanisms: cake layer formation (from suspended material) and pore blocking (from soluble and colloidal compounds). The model properly represented transmembrane pressure (TMP) dynamics at different operating solid concentrations (about 1, 2.6, 6 and 11 g L^{-1}) by employing 7 model parameters, achieving differences between experimental data and model predictions of around 5 - 25 mbar in all cases (differences calculated as the root mean square error). This model was also able to match results from two different influents (raw municipal wastewater and the effluent of the primary settler) by just modifying 3 of 7 parameters. From the 7 proposed parameters, 4 (δ_C, δ_B, k_I and α') were identified as sensible ones by Morris general sensibility analysis, reducing its number to just 2 (δ_C and α') when operating at solid concentrations above 6 g L^{-1}. However, the uncertainly analysis showed that high errors can be expected for long-term simulations since the estimated membrane fouling strongly depends on former fouling conditions. Consequently, the presented model showed an elevated potential to generate reasonable membrane fouling predictions while maintaining a simplistic and open structure to allow its implementation together with other complementary materials. Further research will be performed to enhance model's accuracy and to validate its potential use for filtration optimization and fouling control purposes.

Keywords

Direct membrane filtration; Filtration modeling; Membrane fouling; Municipal wastewater; Resistance-in-series.

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7.1 Introduction

Membrane technology has achieved an increasing interest within the scientific community in last decades due to fast and important investments, significant acquisition and operating cost decrease, and multiple benefits that they can bring in numerous applications [7.1]. Indeed, their robust and accurate separation capacity, easy scaling-up and low space demands make them an excellent candidate to couple and enhance the performance of numerous processes [7.1]. Such as the case of municipal wastewater (MWW) treatment, where membrane technology represents some of most promising alternatives when aiming to achieve sustainable processes [7.2] (see, for instance, anaerobic membrane bioreactor (AnMBR) technology [7.3]). For moving forward on the successful development and full-scale implementation of (emerging) membrane-based technologies, it is necessary to develop complementary tools such as mathematical models, operational controllers, and optimization techniques, to allow further understanding of the mechanisms involved in membrane filtration and to promote the development of further theoretical studies.

Membrane modeling in MWW treatment is generally based on membrane fouling performance estimations within time, in the form of flux decline or transmembrane pressure (TMP) increase. As membrane fouling develop, filtration energy demands also increase, entailing the use of continuous fouling control strategies (physical and/or chemical). Hence, minimizing/controlling membrane fouling development is usually a key issue to solve in order to achieve feasible results in these systems [7.4; 7.5]. Fouling in membrane filtration systems is commonly categorized according to its most relevant source, being cake layer formation and pore blocking the main mechanisms reported [7.6]. Cake layer formation involves the accumulation of any particulate material onto the membrane surface, which is proportional to filtrate volume production. This kind of fouling is usually identified as reversible fouling that can be controlled by physical fouling control cleaning strategies (*e.g.* gas scouring, crossflow liquid velocity, or backwashing). On the other hand, pore blocking (and pore narrowing) describes the partial/complete obstruction of membrane pores by the superficial/internal deposition of soluble compounds and/or colloidal particles, reducing the effective/available membrane area to filtration. This kind of fouling can be either reversible or irreversible (even irrecoverable) depending on substances involved, differencing its effects according to three sub-categories: (1) standard blocking (membrane pores are narrowed due to the internal accumulation of different substances until their complete obstruction), (2) intermediate blocking (substances block the pores or are accumulated onto former deposited materials to form a pseudo cake layer) and (3) complete blocking (substances in the filtered liquor match membrane pores size, directly blocking them). Fig. 7.1 shows a graphic scheme of the fouling mechanisms described.

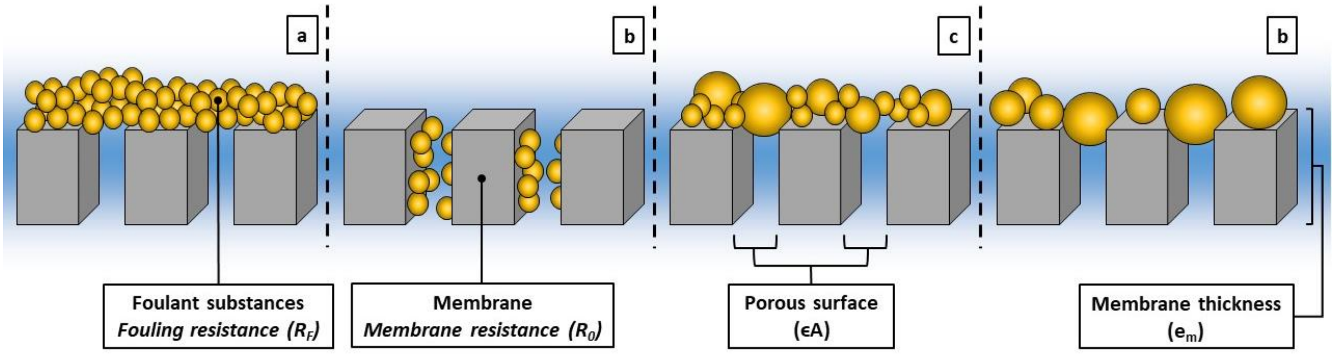

Figure 7.1. Fouling mechanism graphical scheme: (a) cake layer, (b) standard pore blocking, (c) intermediate pore blocking and (d) complete pore blocking.

To model the above-described fouling mechanism, numerous filtration models have been proposed based on a resistance-in-series approach [7.5; 7.7], thus differentiating among fouling mechanisms and linking them to specific substances. Unfortunately, despite the significant amount of model alternatives that can be found in literature for specific filtration processes, they are in general notoriously complicated, increasing in complexity as more general are aimed to be (some examples of complex models could be [7.8; 7.9]). Indeed, the large number of physical/chemical interactions that may occur among liquor substances/structure and membrane surface and internal pores still requires further research to be properly identified, thus demanding the understanding and inclusion of multiple unitary steps that results in increased computational demands. Consequently, the applicability of these models is complex and limited for control and optimization purposes (*i.e.* coupling the model to supervising controllers or optimization algorithms) or when combining with biological models within an integrated framework. The development of simpler and malleable models is accordingly an important milestone to achieve reasonable membrane fouling predictions while allowing its easy combination with the cited complementary materials. Some examples of simple filtration models could be [7.10–7.12], while filtration optimization and control studies by using this kind of models can be found in [7.13; 7.14].

Therefore, the aim of this work is to propose a simple and generic filtration model to allow both membrane fouling prediction regarding influent and operating conditions and membrane performance optimization via supervised controllers. Additionally, a simple and open structure is proposed to allow the easy inclusion of further functions to complement the model according to specific filtration requisites if necessary while facilitating its integration with other models.

7.2 Materials and Methods

7.2.1 Filtration experimental data set

The experimental data set used for developing the model proposed in this study was extracted from two former works based on the direct filtration of MWW using a demo-scale system [7.15; 7.16]. This data set included the experimental results obtained from a filtration plant operated at middle/long-term (above 35 – 124 days each filtration experiment) filtering MWW coming from different steps of a full-scale facility (*'Conca del Carraixet'* wastewater treatment plant (WWTP), Alboraya-Valencia, Spain). A commercial ultrafiltration membrane module (PULSION® Koch Membrane Systems, 0.03-μm pore size, total filtration area of 43.5 m^2) was employed for generating the data set, evaluating the effects on membrane fouling of two influent sources (*i.e.* raw MWW and the primary settler effluent (PSE) of cited municipal WWTP) and four operating suspended solids concentrations (around 1, 2.6, 6 and 11 g L^{-1}). Raw MWW corresponded to the WWTP's influent after a classic pre-treatment of screening and sieving, desanding and degreasing. Further details on the filtration plant can be found elsewhere [7.15]. Table 7.1 and Table 7.2 shows the system constants and main influent characteristics used as input for the model.

Table 7.1. System constants

Constant	Units	Value
Q	$m^3\ s^{-1}$	$1.0875 \cdot 10^{-4}$
A_0	m^2	43.5
e_m	m	$2.5 \cdot 10^{-3}$
ϵ	-	0.7
μ (20 ˚C)*	Pa s^{-1}	$1.003 \cdot 10^{-3}$

*A constant liquid viscosity was employed since experimental data were normalized at 20 ˚C in the source work [7.15; 7.16].

Table 7.2. Model inputs for each set of experiments

MWW treated	X_T (g L^{-1})	SMP (mg L^{-1})	$R_0^{*}\cdot 10^{-12}$ (m^{-1})
PSE	1.2	78	1.7946
	2.6	101	1.5553
	6.4	116	1.8345
	10.6	97	2.3529
Raw	1.1	96	1.5553
	2.6	109	1.2762
	5.9	127	1.4357
	11.1	142	2.1535

X_T: Suspended solids concentration in the membrane tank. SMP: Soluble microbial products concentration in the membrane tank. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler. $^{*}R_0$ calculated from a flux test using the concentrated bulk solution at the beginning of each experimental study.

7.2.2 Model basics

According to Darcy's law, the relationship between the filtration flux (*J*) and transmembrane pressure (TMP) in vacuum filtration can be expressed as follow:

$$J = \frac{Q}{A} = \frac{TMP}{\mu\, R} \tag{7.1}$$

Where Q is the liquid flow rate, A is the total membrane area, μ is the viscosity of the treated solution, and R is the filtration resistance. In general standings, filtration resistance is given by the addition of two different terms: an intrinsically resistance (R_0) that represents the resistance of the membrane itself to filtration, and the developed fouling resistance (R_F) that represents the increasing filtration resistance within time as membrane fouling develops. R_0 is therefore a constant which can be determined by clean water filtration tests (see for instance [7.17]) while R_F is a model's variable which needs to be estimated to achieve proper forecasts.

$$R = R_0 + R_F \tag{7.2}$$

According to the general fouling mechanisms stated before (*i.e.* cake layer formation and pore blocking), this simplified model considers fouling as the result of two independent resistances: (1) the resistance caused by the accumulation of material in the membrane surface (cake layer resistance or R_C), and (2) the resistance linked with the amount of colloidal and soluble compounds attached to membrane pores (pore blocking resistance or R_B). Then, the resulting R_F can be obtained from the addition of these two resistances:

$$R_F = R_C + R_B \tag{7.3}$$

Considering the nature of each defined individual resistance, R_C is built as a function of all the suspended material deposited on the membrane surface (defined as m (kg)), whilst R_B is equivalently built as a function of colloidal and soluble compounds deposited on the membrane pores (defined as n (kg)). m and n terms are therefore the main dynamic variables controlling fouling resistance in this model, which are qualitatively related with the increasing amount of each contaminant accumulated on the membrane during filtration. The following equations are proposed to calculate R_C and R_B:

$$R_C = \alpha' \, m \tag{7.4}$$

$$R_B = \alpha'' \, n \tag{7.5}$$

Where α' and α'' (m^{-1} kg^{-1}) are model parameters representing the contribution of each fouling compound (m and n) to its specific fouling resistance (R_C and R_B). These model parameters can be directly related to the specific cake layer resistance (α_C, m kg^{-1}) or specific membrane pore blocking resistance (α_B, m kg^{-1}) from classic filtration models (see for instance [7.18; 7.19]), respectively, when considering the total filtration area (see Eq. 7.6 and Eq. 7.7).

$$R_C = \alpha' m = \frac{\alpha_C}{A} m \tag{7.6}$$

$$R_B = \alpha'' n = \frac{\alpha_B}{A} n \tag{7.7}$$

Thus, Eq. 7.6 and Eq. 7.7 can be employed when at least one of the cited specific resistances is known, whilst the simplified expressions (Eq. 7.4 and Eq. 7.5) can be used to estimate the qualitative impact of fouling accumulation on filtration resistance when approximations for these physical parameters are not available. In any case, since the total membrane area (A) is a model constant (total filtration area of the membrane module operated), the use of any of proposed expressions will result in same model outputs.

Based on the work performed by Benyahia *et al.* [7.20], the amount of pollutants attached to the membrane (*i.e. m* and *n*), was estimated by considering the mass flux of each related contaminant, proposing the following differential expressions:

$$\dot{m} = \delta_C \frac{Q}{a} X_T \qquad 7.8$$

$$\dot{n} = \delta_B \frac{Q}{\epsilon a\ e_m} S \qquad 7.9$$

Where X_T and S are the concentration of suspended solids and soluble/colloidal material in the bulk, respectively, and 'a' - understand as a(t) - is the dynamic effective filtration membrane area. According to these definitions, δ_C (m^2) is a model parameter involving the impact of the particulate material flux on the cake-layer growth rate (*i.e.* dynamics on particulate material attachment), and δ_B (m^3) is a model parameter involving the impact of soluble and colloidal compounds flux on pore blocking growth rate (*i.e.* dynamics on soluble and colloidal compounds attachment). However, in this later case, since only the internal surface of membrane pores is considered to be affected by these substances in this work, the effective area is recalculated considering the pores percentage of membrane area (ϵ) and the membrane thickness (e_m). Hence, the proposed model calculates fouling resistance (R_F) dynamics as a function of the dynamics on *m* and *n*. For new membranes, the initial values of *m* and *n* will be zero (*i.e.* m(t=0) = 0 and n(t=0) = 0), while these values may be adjusted to adequate initial conditions for the rest of cases.

A membrane permeability impoverishment is expected as filtration advances due to the continuous accumulation of suspended material on the membrane surface and the loss of membrane porosity due to pore blockage by soluble and colloidal substances. Moreover, contrary to several literature studies on membrane fouling modeling, the proposed model considers that the effective membrane area is not constant during the filtration process, as Benyahia *et al.* [7.20] proposed. Consequently, this model traduces this membrane permeability loss not just by an increase in the filtration resistance but also as a reduction of the effective membrane area 'a'. A possible relation between the fouling accumulation (*i.e. m* and *n* increases) and the loss of membrane area could be expressed as follows:

$$a = \frac{A}{1 + \frac{m}{\sigma_C} + \frac{n}{\sigma_B}} \qquad 7.10$$

Where σ_C and σ_B (kg in both cases) are model parameters aimed to consider the effect of each fouling accumulation on membrane area losses. Finally, considering Eq. 7.2, Eq. 7.3 and the fouling dynamics link with the dynamic effective membrane area 'a', Eq. 7.1 can be rewritten as follow:

$$J = \frac{Q}{a} = \frac{TMP}{\mu\,(R_0 + R_C + R_B)} \qquad 7.11$$

According to the presented model, a higher fouling accumulation during the filtration process means that a lower membrane area is available for continuing with the process, increasing consequently the operating filtration flux since an effective shorter membrane area is used for treating the same liquid flow rate. This dynamic creates then a circular interaction between *m* and *n* and the effective membrane area 'a' which is the basic loop used by the model to estimate fouling development.

7.2.3 Further model considerations

Given all different soluble and colloidal substances that may interact with membrane filtration depending on bulk characteristics, the following generalized expression is proposed to considered their overall effect in the proposed model (Eq. 7.9):

$$\dot{n} = \delta_B \frac{Q}{\epsilon a\, e_m} \sum_{i=1}^{N} (f_i\, x_i) \qquad 7.12$$

Where N is the number of soluble and colloidal materials considered in the process, *x* represents their concentration, and *f* represents the relative relevance of each considered soluble and colloidal compound over total relevance (*i.e.* $\sum_{i=1}^{N} f_i = 1$). In the present work, since just data concerning SMP concentration during filtration were available, the membrane pores blocking resistance was completely linked to the total concentration of these substances, arranging Eq. 7.12 as follows:

$$\dot{n} = \delta_B \frac{Q}{\epsilon a\, e_m}\, SMP \qquad 7.13$$

On the other hand, the data set used in this work revealed that an increase in the bulk suspended solids concentration resulted in a sharp beneficial effect on overall filtration performance, achieving less fouling growth rates (see [7.15; 7.16]). This phenomenon was attributed to the formation of thicker cake layers during filtration, which prevented the soluble substances and colloids to reach the membrane surface, as other authors have also theorized [7.21–7.23]. Since increasing the solids concentration (X_T) decreased the fouling propensity related with these substances ($\dot{n}$), an inhibition function for pores blocking fouling was included in the proposed model (Eq. 7.13) to consider this effect. In this case, a classic literature exponential inhibition function [7.24] was used due to the good fits obtained for the studied experimental data set. However, other inhibition functions, or none, could be proposed depending on the process if necessary. Eq. 7.13 can then be rewritten as follows:

$$\dot{n} = \delta_B \frac{Q}{\epsilon a\, e_m}\, SMP\, e^{-k_I\, X_T} \qquad 7.14$$

Where k_I (kg^{-1}) is an inhibitor constant which needs to be determined as a model parameter from experiments.

7.2.4 Model implementation, calibration and validation

The model was implemented in MATLAB®. Function ‘ODE45’ was employed for differential equations operations. To calibrate the model from the data set, the experimental average TMP calculated for each operating day was employed. The experimental data from the operating solids concentration of about 1, 2.6 and 6 g L^{-1} were used for calibration, leaving all experimental data from a solid concentration of about 11 g L^{-1} for validation. Two different sets of parameters were calibrated depending of each MWW evaluated (*i.e.* raw and PSE). The error between experimental TMP (TMP_{exp}) and predictions obtained from Eq. 7.11 (TMP_{teo}) was used as objective value for parameters calibration, minimizing its value by using a nonlinear optimization algorithm function (‘fmincon’, MATLAB/Simulink). The error was calculated as the root mean square error (RMSE):

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N}\left(TMP_{exp} - TMP_{teo}\right)^2}{N}} \qquad 7.15$$

Where N represent the total amount of data.

On the other hand, the theoretical study of the identifiability of the model can be difficult and laborious. To ensure practical identifiability of the model, we adopt a procedure in which parameter optimization is obtained from several sets of initial conditions. By limiting the number of parameters to be identified (chosen following a sensitivity study), we guaranteed to obtain a unique set of parameters for each experimental condition tested [7.25].

7.2.5 Sensibility and uncertainty analysis

Two global sensibility analysis (GSA) methods were applied in this work to determine the most influential parameters of the model after calibration: standardized regression coefficient method (SRC) and Morris screening method [7.26]. In both methods, an input variation factor of ±20% regarding default values (see Table 7.3) was considered. SRC was performed based on Monte Carlo method, applying semi-random Latin Hypercube Sampling [7.27] to generate parameters variation. The number of Monte Carlo simulations was set to 2000. Inputs resulting in standard regression coefficients (β_i) higher than 0.1 were considered as influential factors, establishing a minimum coefficient of determination (R^2) of 0.7 to validate β_i as sensibility measure [7.28]. Morris method was performed by using the scaled elementary effect (SEE_i) proposed by Sin and Gernaey [7.29]. The trajectory-based sampling strategy proposed by Ruano *et al.* [7.30] was used as modification of Morris screening method to improve the calculation of SSE_j finite distribution associated to each input factor (F_j). The absolute mean (μ^*, Eq. 7.16) and standard deviation (σ, Eq. 7.17) were used as statistical parameters to determine the relative importance of parameters variation on model's output [7.28; 7.31].

$$\mu_i{}^* = \frac{\sum_{j=1}^{r} |SEE_j|}{r} \tag{7.16}$$

$$\sigma_i = \sqrt{\frac{1}{r}\sum_{j=1}^{r}\left(SEE_j - \mu_i\right)^2} \tag{7.17}$$

Where *r* is the number of trajectories evaluated (set to 100 in this study) and μ is the mean. A resolution of p=4 was employed in this study [7.32]. Morris total simulations (MTS) were calculated based on Eq. 7.18, ascending in this case to 800.

$$MTS = r(k + 1) \quad 7.18$$

Where k is the number of input factors (*i.e.* analyzed parameters; 7 in this study: α', α'', δ_C, δ_B, σ_C, σ_B and k_I).

The uncertainty analysis (UA) of the model was studied after the identification of model's most sensible parameters. A parameter variation factor of ±20% regarding default values (see Table 7.3) was applied. This analysis was conducted by determining the 5th and 95th percentiles from Monte Carlo simulations [7.33].

7.3 Results and Discussion

7.3.1 Sensibility and identifiability analysis

Table 7.3 shows the model parameters values after calibration. As results obtained from the SRC sensibility study shows (see Fig. 7.2), just 2 out of 7 parameters (δ_C and δ_B) were identified as significant influential model factors (both related to the amount of mass attached to the membrane depending of the fouling source). Unfortunately, low correlation degrees were obtained between variations in parameter values and resulting effects on model output (see Fig. 7.2), which continuously decreased as the simulation time was increased. Since SRC methodology demands correlations above 0.7 to validate the sensitivity results [7.28], it was not possible to validate this methodology in any evaluated scenario, mainly due to the nonlinearity of the presented model.

Table 7.3. Calibrated parameters

Parameter	Units	MWW		DBP (%)
		PSE	Raw	
α'	$m^{-1}\ kg^{-1}$	$5.9501 \cdot 10^{10}$	$5.9501 \cdot 10^{10}$	< 0.01
α''	$m^{-1}\ kg^{-1}$	$5.9501 \cdot 10^{10}$	$5.9501 \cdot 10^{10}$	< 0.01
δ_C	m^2	$4.8108 \cdot 10^{-3}$	$2.8098 \cdot 10^{-3}$	41.59
δ_B	m^3	$1.6578 \cdot 10^{-2}$	$7.2783 \cdot 10^{-3}$	56.10
σ_C	kg	1	1	-
σ_B	kg	1	1	-
k_I	kg^{-1}	0.6078	0.3803	37.43

DBP: Difference between the parameters calibrated for the two MWW studied. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

PSE

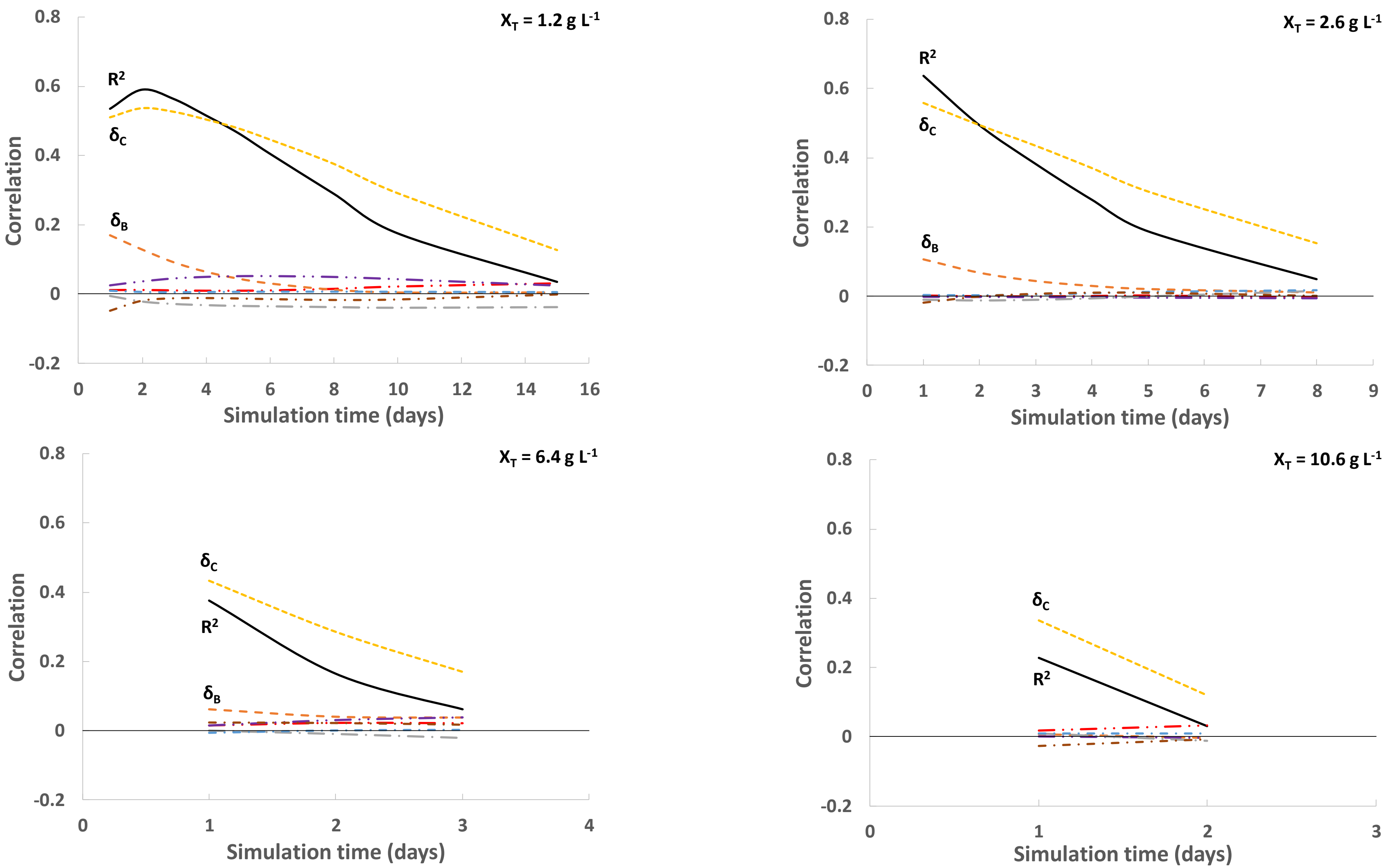

Figure 7.2. Cont.

Raw

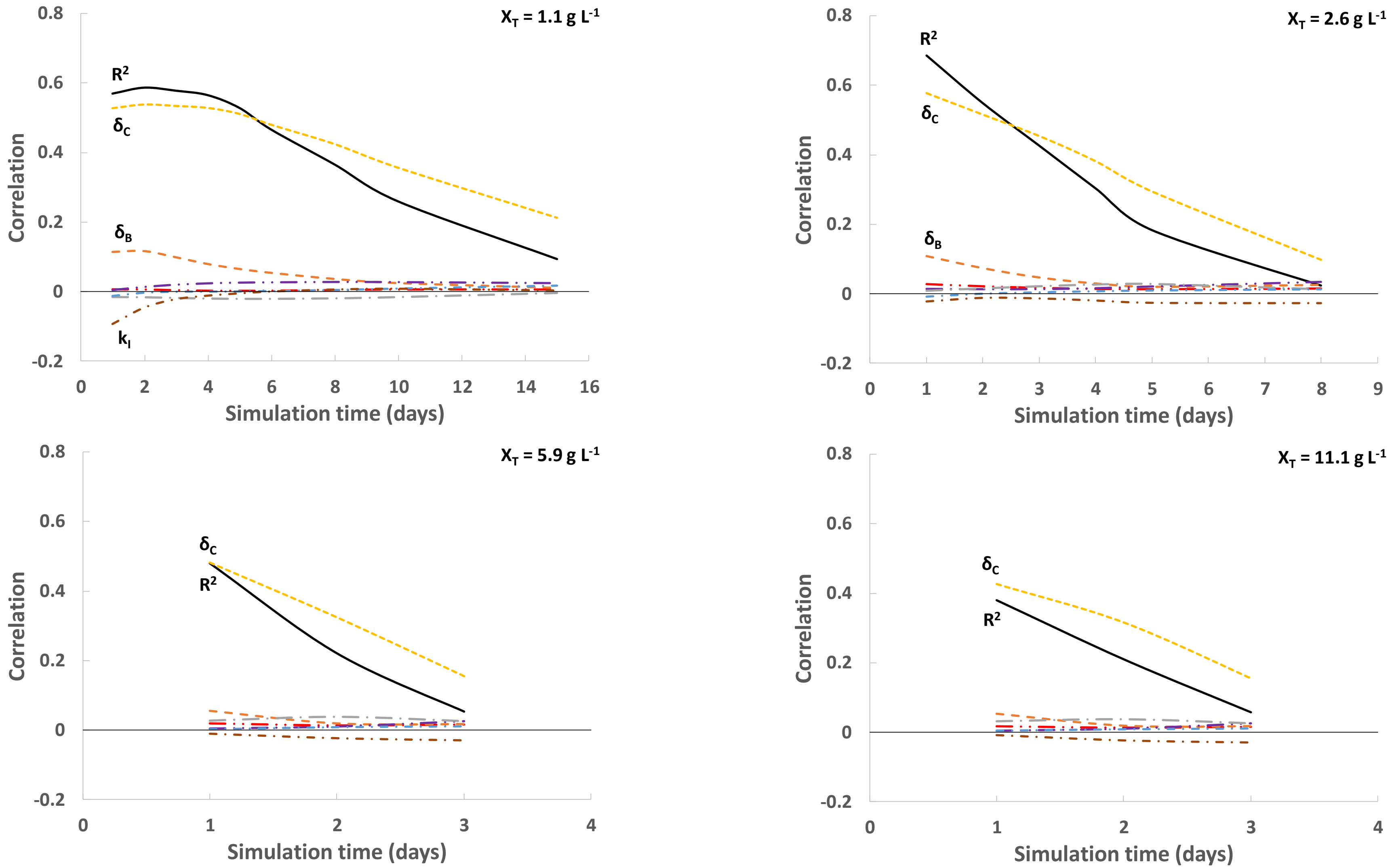

Figure 7.2. SRC method results. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

On the other hand, Morris screening method revealed 4 out of 7 model parameters to be influential on model output (see Fig. 7.3): δ_C, δ_B, k_I and α'. This method suggests that δ_B and k_I are important input factors when operating at low solid concentrations (around 1 and 2.6 g L^{-1} in this work), while their influence decreases in relevance when operating at higher solid concentrations (above 6 g L^{-1}), regardless of the MWW treated. As will be discussed in the next section (see Section 4.3.2), the TMP observed in this work is mainly controlled by the soluble compound fouling (*n*) in the low solids concentration range, reducing its importance as bulk solids concentration raised. Consequently, the higher the operating solids concentration in the membrane tank, the lower the relevance of *n*-related fouling. In any case, δ_C was identified as the most influential model parameter within all experimental conditions, its relevance dramatically increasing as solids concentration raised, obtaining the highest μ^* and σ values for the highest solid concentration tested (about 11 g L^{-1} in this case, see Fig. 7.3). This was due to the direct relationship between δ_C and X_T to calculate *m* (see Eq. 7.8).

Regarding the identifiability analysis, it determined that the proposed set of parameters was 3 degrees of freedom higher than that required in this case. This was due to the lack of information concerning fouling development in the studied experimental sets. Indeed, no information regarding the specific cake layer resistance during the process or the amount of pollutants attached to the membrane was available. Consequently, just a qualitative evolution of pollutants accumulated in the membrane surface or pores (*i.e. m* and *n* evolution) could be expected by the model in this case. To solve this identifiability issue, the value of 2 of the no sensible parameters (σ_C and σ_B) was set to a constant value (1 kg in this case) to reduce the degrees of freedom, while α' and α'' were forced to achieve the same value during calibration thus acting as a unique parameter. The number of parameters to calibrate was consequently reduced to 4 (δ_C, δ_B, k_I and α'), all of them influential parameters in some degree when operating at a low solids concentration range (under 2.6 g L^{-1}), and with just 2 sensible parameters (δ_C and α') when increasing the solid concentration in the treated bulk (above 6 g L^{-1}).

PSE

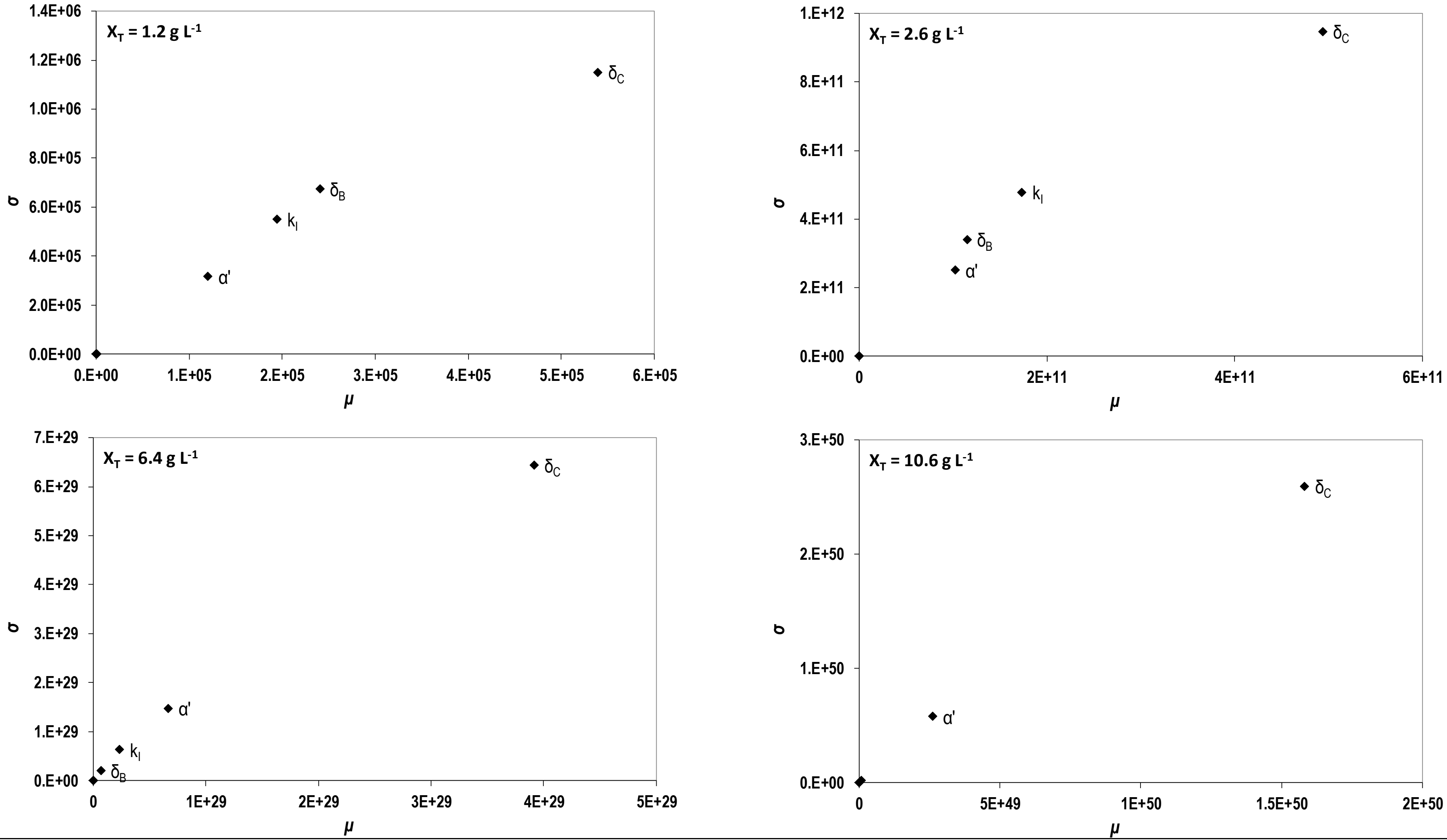

Figure 7.3. Cont.

Raw

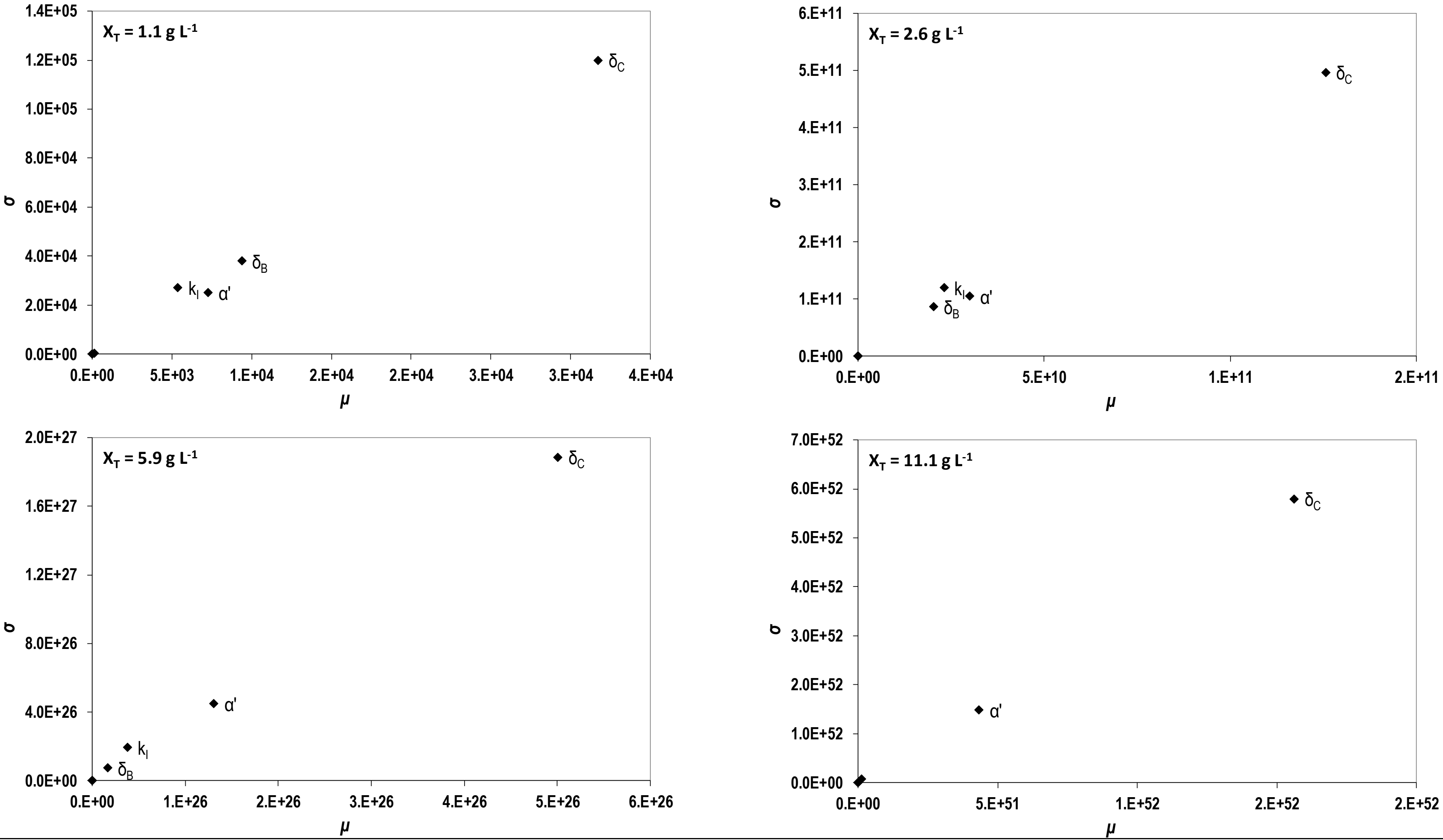

Figure 7.3. Morris screening method results. A filtration simulation time of 20 days was applied in all cases for the parameters sensibility evaluation. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

7.3.2 Model performance and uncertainty analysis

Fig. 7.4 (7.4a and 7.4c) shows the performance of the model for the operating conditions evaluated in this work (calibrated parameters values can be found in Table 7.3). Reasonably good fits were obtained in all cases, achieving low RMSE values between about 5 – 25 mbar (see Fig. 7.4). Indeed, good fits were also obtained during model validation (solid concentration of about 11 g L^{-1}), showing the good predictability capacity of the model. R_C, R_B, *m*, *n* and '*a*' evolution is also displayed in Fig. 7.4 (7.4b and 7.4d) to allow an easier interpretation of model performance. As stated before, this model is based on the interaction between fouling development and membrane permeability loss (*i.e.* *m* and *n* increase and '*a*' decrease), creating a dynamic where the effective operating flux increases virtually due to a decrease in the effective membrane filtration area. This membrane permeability reduction can be achieved by two different paths: cake layer formation (related with *m* term) and pore blocking (related with *n* term), since '*a*' is affected by both through the same pathway (see Eq. 7.10). Cited interaction can be observed when comparing the fouling source from the experimental data with the fits obtained at low solid concentrations (1.1 and 1.2 g L^{-1}, see Fig. 7.4b1 and 7.4d1) and high solid concentrations (10.4 and 11.1 gL^{-1}, see Fig. 7.4b4 and 7.4d4), where the soluble and particulate material are the most relevant fouling sources, respectively, consequently producing different outputs on TMP evolution. Regarding the influent studied (raw and PSE), similar values were reached after model calibration for most of model parameters (see Table 7.3). This was presumably due to the fact that the two studied MWWs shared the same matrix, *i.e.* PSE being obtained from raw MWW after a primary settling step. The lower fouling rate propensity observed when treating raw MWW instead of PSE (see Fig. 7.4) was captured by the model through of a lower value for the δ_C and δ_B parameters. However, this differences could be due to an oversimplification of the fouling promotor pollutants. As Sanchis-Perucho *et al.* [7.15; 7.16] suggests, the particle size distribution of filtered bulk is a key factor concerning membrane fouling propensity when directly filtering untreated (no biological) MWW. Consequently, including information regarding colloidal fraction concentration (identified in other studies as an important fouling promotor [7.34]) and average particle size of bulk suspended solids particles could represent a significant benefit regarding this model fouling prediction capacity. Further studies regarding this later point will be performed to build a more complete model able to predict fouling from the two influents studied with a unique set of parameters.

PSE

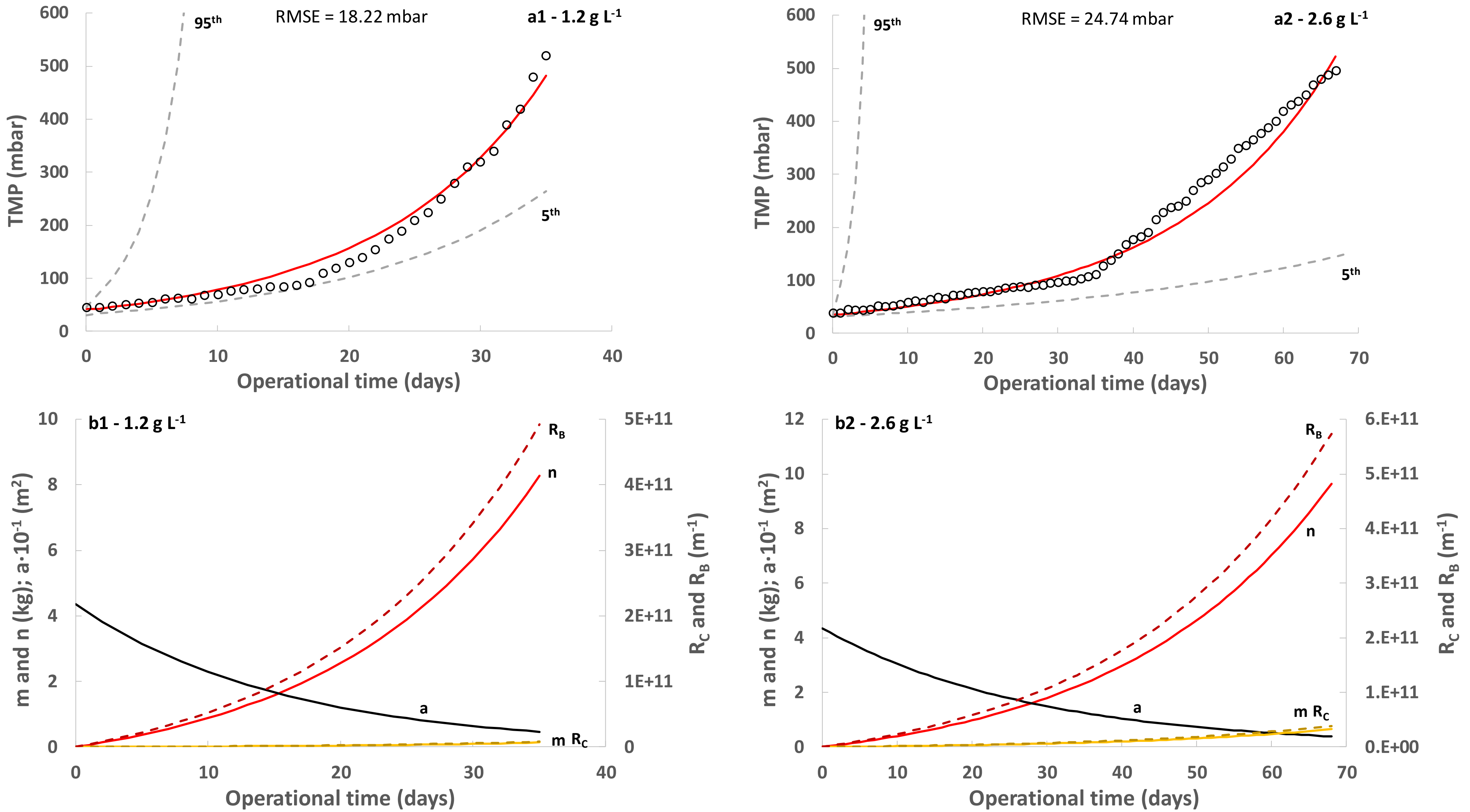

Figure 7.4. Cont.

PSE

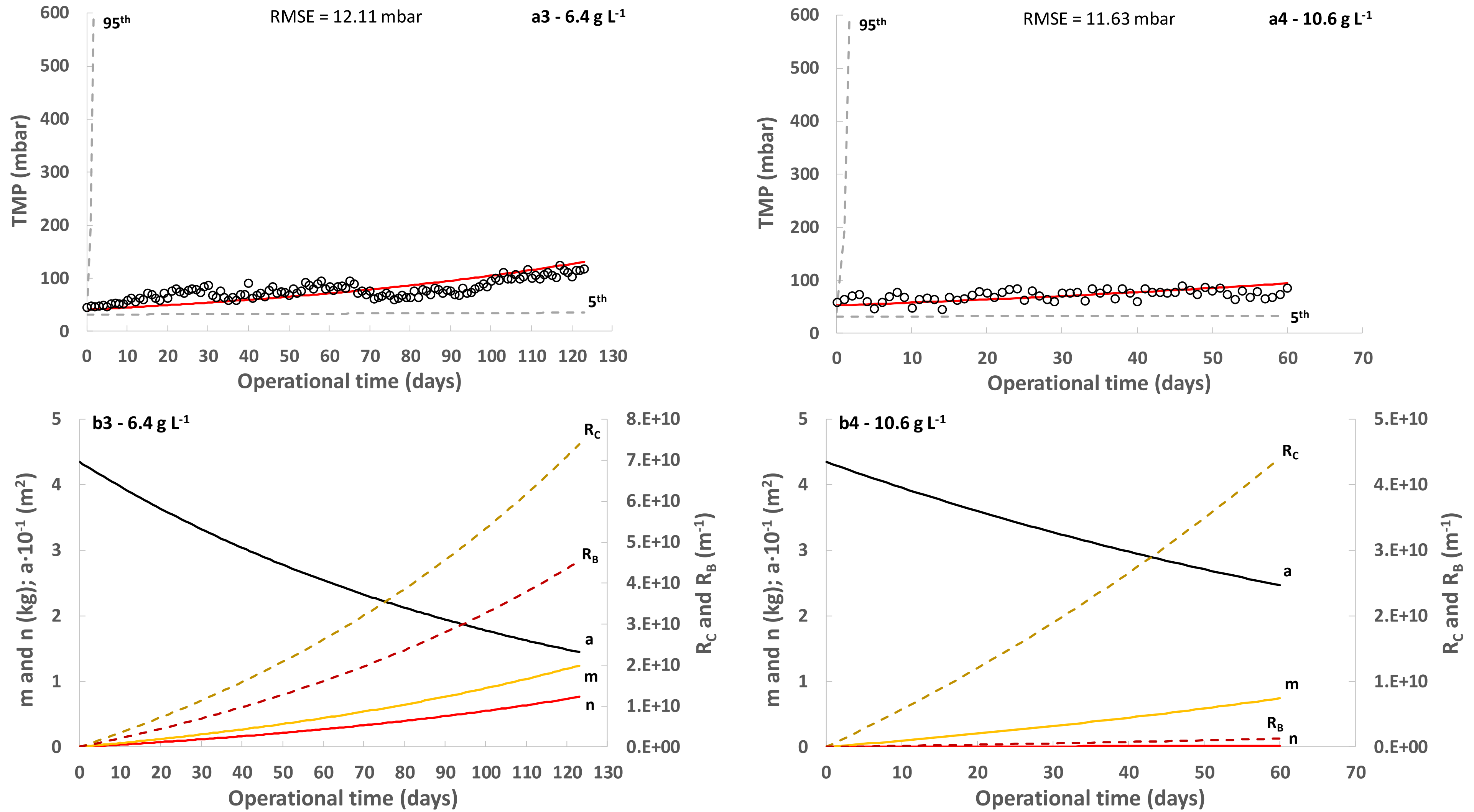

Figure 7.4. Cont.

Raw

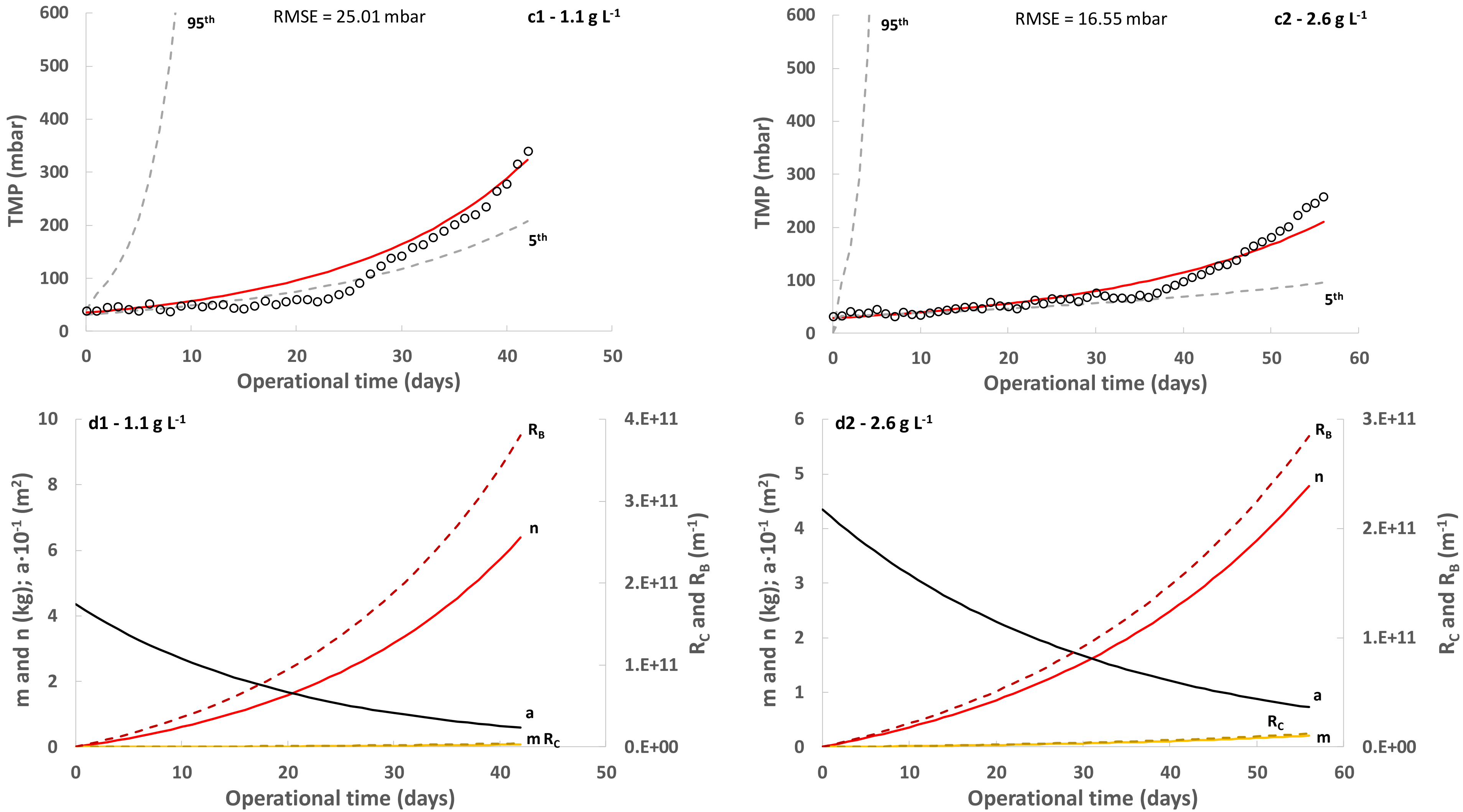

Figure 7.4. Cont.

Raw

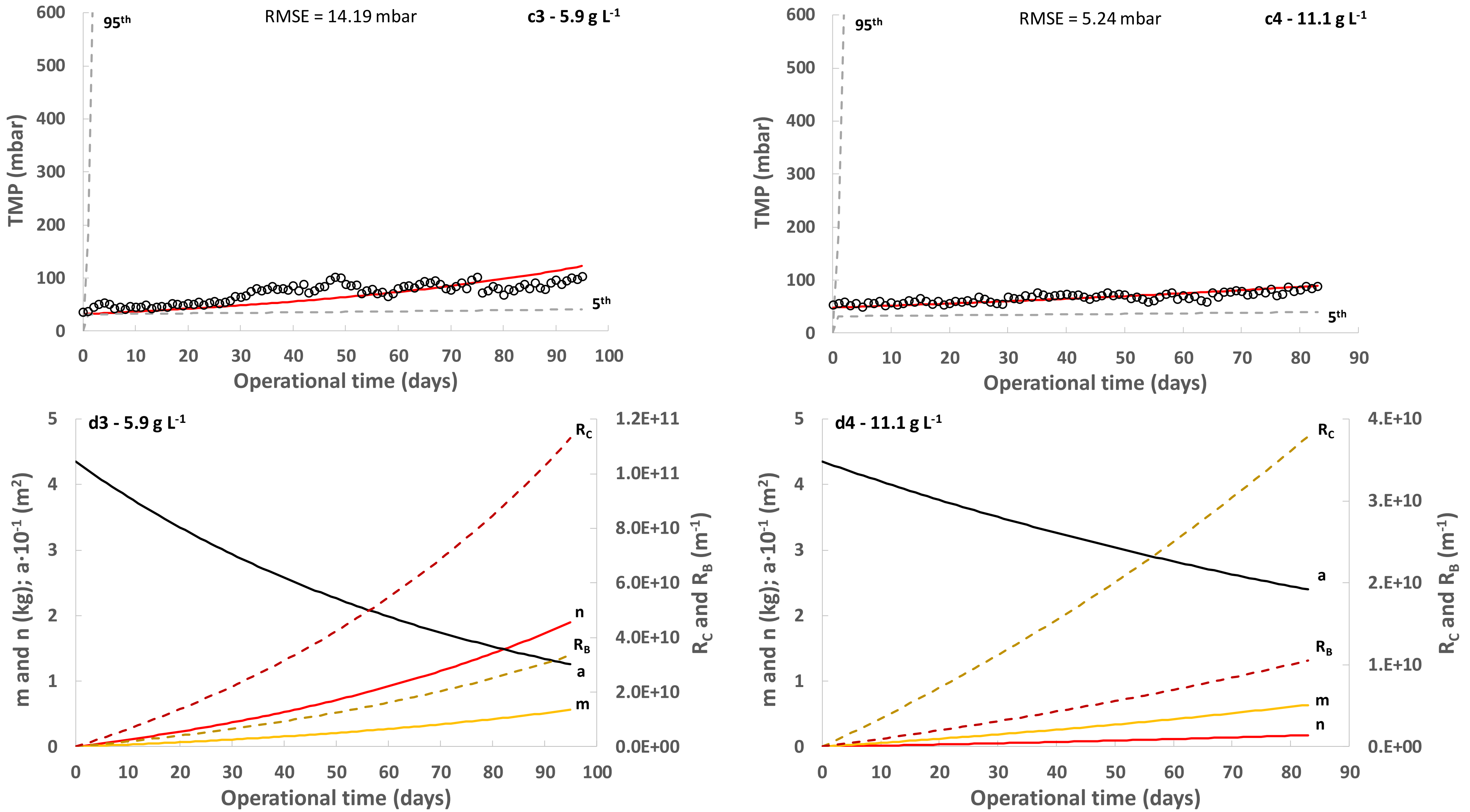

Figure 7.4. Model's TMP predictions in the middle/long-term: (a and c) TMP evolution and (b and d) model variables evolution. Experimental data are displayed by dots while model's predictions are shown by lines. Note that each figure includes the operating solids concentration of the experiment. Two MWW were studied: PSE (a and b) and Raw (c and d). 5th and 95th represent the corresponding uncertainty percentiles from Monte Carlo simulations. Raw: Influent municipal wastewater after a classic pre-treatment (screening and sieving, desanding and degreasing). PSE: Effluent of the full-scale wastewater treatment plant primary settler.

Finally, the UA performed when considering the most influential model parameters (*i.e.* δ_C, δ_B, k_I and α') showed that tolerable uncertainties are obtained in the short-term (some hours, see Fig. 7.5), although model uncertainty dramatically raise when considering middle/long-term filtration periods (weeks and months, see Fig. 7.4). This is related to the fact that that future fouling development in filtration systems depends on former fouling conditions. Consequently, low divergences during parameters calibration could imply substantial discrepancies in the long-term due to accumulative errors. Thus, the accurate calibration of the most influential model parameters is imperative when applying the model.

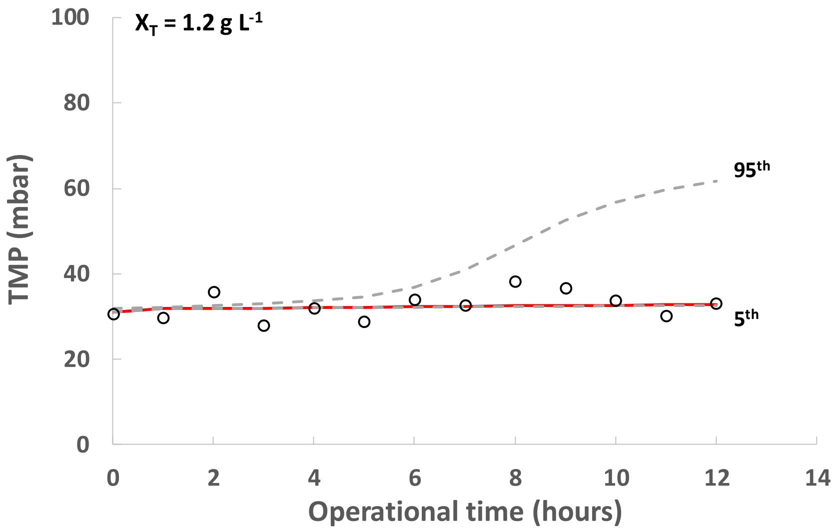

Figure 7.5. Example of the uncertainty assessment in the short-term. Results obtained when treating PSE (primary settler effluent) at a solid concentration of 1.2 g L^{-1}. Experimental data are displayed by dots while model's predictions are shown by lines. 5^{th} and 95^{th} dotted lines represent the corresponding uncertainty percentiles from Monte Carlo simulations.

7.3.3 Model forecasts regarding solids concentration increase in the bulk

As Fig. 7.4 shows, an increase on the operating solids concentration entails a reduction on pore blocking fouling as filtration experimental data indicated. As discussed in Sanchis-Perucho *et al.* [7.15; 7.16], this phenomenon may be due to a reduction on soluble compounds interaction with the membrane surface as increasing the cake layer thickness formed onto the membrane surface, thereby reducing pore-blocking-related fouling propensity. However, numerous membrane systems operate at much higher solid concentrations, where an increase in solids concentration entails higher fouling propensities [7.35; 7.36]. To estimate the model outputs when operating at high solid concentrations and evaluate its performance suitability for other filtration processes, some simulations where performed hypothetical conditions. Parameters calibrated when treating PSE were used in these simulations. Fig. 7.6 shows the results obtained when simulating a consistent increase of the operating solids concentration in the filtered bulk.

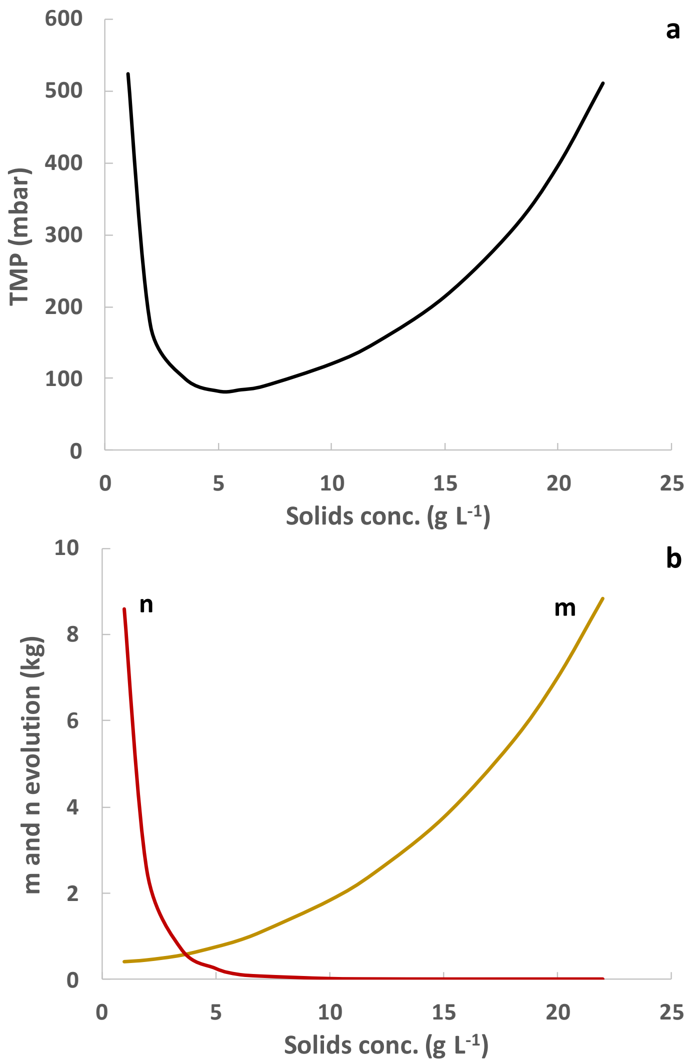

Figure 7.6. Simulation runs regarding solids concentration. Parameters calibrated for PSE treatment were employed (see Table 7.3). All simulations were performed under 120 days of filtration at a constant SMP concentration of 20 mg L^{-1}.

As expected, the main contributor (*m* or *n*) to the resulting TMP varies as the operating solids concentration increases. Furthermore, a changing dynamic on the forecasted TMP was obtained, firstly reducing its value thanks to the reduction of pore-blocking-fouling as increasing the solids concentration but later increasing due to the increment of cake-layer-fouling as solids concentration increased over 5 g L^{-1}. These results show the potential of presented model to match numerous fouling behaviors described in literature. Indeed, the proposed dynamic between *m*, *n* and ‘*a*’ allows this model to consider different fouling sources, overcoming issues from other simple models which work considering fouling as the product of a unique variable (generally solids concentration in the bulk, see for instance [7.10; 7.13]). This fact makes presented model an interesting tool for filtration control and optimization due to its simplicity and easy implementation or combination with other assistant tools (*e.g.* complementary supervising controllers or optimization algorithms). However, further studies aimed to enhance the accuracy of model predictions to different filtration conditions around the evaluated system (*i.e.* direct filtration of MWW at higher solid concentrations and different permeate fluxes) are required. Additionally, future research assessing this model utility in other filtration systems and on filtration optimization is also necessary.

7.4 Conclusions

This work aim was to propose a simple and generic model (7 calibration parameters) to predict membrane fouling in the long-term when directly filtering MWW. Proper predictions were obtained for different operating solid concentrations (about 1, 2.6 and 6 g L^{-1}), achieving RMSE values between experimental data and model predictions around 5 – 25 mbar. Additionally, good fits were also obtained when applying the calibrated model to a higher solid concentration (about 11 g L^{-1}), validating the proposed model for a short solid concentration range. This model was also able to match results from two different influents (raw municipal wastewater and the effluent of the primary settler) by just modifying 3 of 7 parameters. From the 7 proposed parameters, 4 (δ_C, δ_B, k_I and α') were identified as sensible ones by Morris general sensibility analysis, reducing its number to just 2 (δ_C and α') when operating at solid concentrations above 6 g L^{-1}. However, the uncertainly analysis showed that high errors can be expected for long-term simulations since the estimated membrane fouling strongly depends on former fouling conditions. Consequently, the presented model showed an elevated potential to generate reasonable membrane fouling predictions while maintaining a simplistic and open structure to allow its implementation together with other complementary materials. Further research will be performed to enhance model's accuracy and to validate its potential use for filtration optimization and fouling control purposes.

7.5 Acknowledgements

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CHAPTER 8. Dynamic Membranes for Enhancing Resources Recovery from Municipal Wastewater

Abstract

This paper studied the feasibility of using dynamic membranes (DMs) to treat municipal wastewater (MWW). Effluent from the primary settler of a full-scale wastewater treatment plant was treated using a flat 1 µm pore size open monofilament polyamide woven mesh as supporting material. Two supporting material layers were required to self-form a DM in the short-term (17 days of operation). Different strategies (increasing the filtration flux, increasing the concentration of operating solids and coagulant dosing) were used to enhance the required forming time and pollutant capture efficiency. Higher permeate flux and increased solids were shown to be ineffective while coagulant dosing showed improvements in both the required DM forming time and permeate quality. When coagulant was dosed (10 mg L^{-1}) a DM forming time of 7 days and a permeate quality of total suspended solids, chemical oxygen demand, total nitrogen, total phosphorous and turbidity of 24 mg L^{-1}, 58 mg L^{-1}, 38.1 mg L^{-1}, 1.2 mg L^{-1} and 22 NTU, respectively, was achieved. Preliminary energy and economic balances determined that energy recoveries from 0.032 to 0.121 kWh per m^3 of treated water at a cost between -€0.002 to €0.003 per m^3 of treated water can be obtained from the particulate material recovered in the DM.

Keywords

Direct membrane filtration; Dynamic membranes; Resource recovery; Municipal wastewater treatment.

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8.1 Introduction

The municipal wastewater (MWW) management paradigm has changed considerably in recent years. Due to increasing worldwide water demands, the global energy crisis and climate change, the need to find new fresh water sources and develop more energy-efficient technologies with a low environmental impact is becoming imperative to ensure sustainable global economic models. As a result, numerous studies are recommending changing the current development models for new ones based on the circular economy (CE) [8.1]. MWW is thus beginning to be considered a relevant source of essential resources, including reclaimed water, energy and nutrients (mainly nitrogen and phosphate) [8.2]. Unfortunately, the current municipal wastewater treatment plants (WWTP) fail to recover all the potential resources in MWW, focusing on managing sewage as their only goal. In fact, classical aerobic technology, which is the core of the water line treatment, is usually identified as an inefficient system, representing up to 50% of the total energy requirements of full-scale WWTPs [8.3].

Several alternatives have been proposed to transform the current WWTPs to new resource recovery facilities (*e.g.* the direct treatment of MWW in anaerobic membrane bioreactors [8.4]). However, their implementation could represent significant economic investments due to radical changes in the MWW treatment structural scheme and operating conditions. Because of that, the filtration of the MWW before the aerobic treatment, which is commonly known as direct membrane filtration (DMF), have recently been proposed as an interesting option [8.5]. DMF consists of using a membrane filtration system to treat raw influent MWW (after conventional screening, sieving, desanding and degreasing pre-treatment) and capture the particulate fraction of the influent sewage. Thanks to this, the oxygen demands of the biological process can be significantly reduced by lowering the organic loading rate, which in turn reduces the energy demands. Indeed, aerobic treatment could even be unnecessary depending on the generated permeate quality [8.6]. On the other hand, the organic material recovered in the membrane tank can be transformed into methane via anaerobic digestion (AD), enhancing the overall energy balance of the facility. The nutrient content in the particulate fraction of the MWW is also recovered in the concentrated sludge, thus improving the overall resource recovery potential during the MWW treatment without requiring a great deal of structural modifications to the current installations.

Many authors have studied different membrane technologies (*i.e.* microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and forward osmosis (FO) membranes) for the DMF of MWW, usually finding severe membrane fouling during the filtration process [8.7; 8.8]. In fact, the development of effective and energy-efficient fouling control strategies is one of the major issues to be addressed to enhance DMF feasibility [8.9]. To overcome this issue, some authors have proposed using dynamic membranes (DM), showing promising results [8.10–8.12]. DMs consist of the formation of a cake layer on a low filtration-resistance supporting material, making the cake layer formed the main filtering actor [8.13]. Filtration resistance during DM operation can thus be easily controlled by physical cleaning methods and usually achieve significantly lower filtration resistance than those of other membrane technologies [8.10]. Additionally, the required supporting structures are generally low-cost materials such as filter-cloths and woven meshes, representing low investment cost for their acquisition and/or replacement [8.14]. Two different kinds of DMs can be defined depending on how the filtering cake layer is formed namely, self-forming and pre-coated DMs [8.13; 8.15]. Self-forming DMs are formed by developing the cake layer from the direct deposition onto the supporting structure of the suspended material and high molecular weight organics contained in the treated liquor while performing the filtration process. On the other hand, pre-coated DMs are formed by passing an external solution containing one or more particulate materials through the supporting structure, such as powdered carbon or kaolite, for pre-forming a stable structure onto which the filtering cake layer will be formed. In comparison, self-forming DMs are more advantageous since they do not require additional chemical dosing, reducing the operating cost [8.15]. However, pre-coated DMs are essential in some scenarios for reducing the DM forming time and for allowing its formation when not enough particulate material is transported by the treated liquor.

Different strategies have been proposed to improve filtration performance of DM systems when treating MWWs. Among them, coagulant dosing is one of the most recommended for reducing membrane fouling and enhancing permeate quality [8.8; 8.16], helping also in the formation of the DM as a pre-coating material. Several studies agree in recommending inorganic polyaluminum chloride (PACl) coagulants [8.16; 8.17], which can efficiently flocculate the small-size-range particles present in the MWW, and even capture a significant percentage of the colloidal fraction. Phosphate can also be efficiently captured via chemical precipitation, while stronger and more resistant flocs have been reported to be formed when using PACl, achieving better performances than other coagulants when carrying out filtration processes [8.18; 8.19]. Another possible alternative to enhance DM resource capture efficiency is to raise the operating solids concentration, which could promote the formation of thicker and less porous cake layers on the supporting material. This would represent an interesting approach since additional operating chemicals and environmental costs could be avoided. However, increasing operating solids concentration could also raise the sludge filtering resistance, which could dramatically affect the filtration energy demand, achieving counterproductive effects. Using pre-treated influents has also been suggested to reduce the severe fouling reported when treating raw MWWs [8.20]. In this regard, primary settler effluent (PSE) as the DM influent could be an interesting option since a large fraction of the influent particulate material would be recovered and concentrated in a pre-treatment step, presumably reducing the fouling potential of the treated MWW. Additionally, the recovered sludge would be used in the AD in much the same way as conventional MWW treatment schemes, thereby not affecting the energy balance. However, removing the higher size particles when using DMs could also involve important negative effects on the DM self-forming capacity, DM structure and toughness, and permeate quality, which could compromise their applicability.

The aim of this work was therefore to assess the potential benefits of DMs in treating PSE from a WWTP with a preliminary (pilot scale) study to determine the best operating conditions to carry out the filtration process. The effect of using an additional supporting material layer, increasing the filtration flux (15 and 45 LMH), different operating total suspended solids (TSS) concentrations and coagulant dose were evaluated. All the experiments focused on determining: (1) DM forming capacity when treating this influent, (2) the resource recovery capacity of the system and (3) the fouling rate and preliminary economic costs of the process.

8.2 Materials and Methods

8.2.1 Influent and experimental design

The influent MWW used was PSE from the full-scale “Conca del Carraixet” WWTP (Alboraya, Spain) (see the main characteristics of this influent MWW in Table 8.1). Two different systems (a pilot-scale plant and a lab-scale membrane module) were used to evaluate the effect of different operating conditions on the DM performance. The pilot plant was used to assess the effect of the number of supporting layers, operating flux and coagulant dosing and the lab-scale module to evaluate the effect of the operating TSS concentration (performed at lab-scale due to the difficulty of reaching the required TSS concentrations in the pilot plant). Table 8.2 shows the experimental conditions of every experimental period. A flat polyamide open monofilament woven mesh of 1 µm average pore size (NITEX®, SEFAR) was used as supporting material in all the experiments and the membrane surface was cleaned by brushing it with tap water as required.

Table 8.1. Influent characteristics

Parameter	Units	Mean ± SD
TSS	mg TSS L^{-1}	113 ± 22
COD	mg COD L^{-1}	167 ± 42
SCOD	mg COD L^{-1}	57 ± 21
TN	mg N L^{-1}	45.5 ± 8.5
TP	mg P L^{-1}	5.9 ± 1.1
Alk	mg $CaCO_3$ L^{-1}	335 ± 67
pH	-	7.6 ± 0.5
Turbidity	NTU	109 ± 31

Table 8.2. Experimental setup

PILOT-PLANT			
Exp.	**Supporting material**	**Operating flux (LMH)***	**Coagulant concentration (mg Al_2O_3 L^{-1})**
1	1 layer	15.4 ± 0.2	-
2	2 layers	15.1 ± 0.3	-
3	2 layers	45.3 ± 3.1	-
4	2 layers	14.7 ± 0.7	10
LAB-SCALE			
Exp.	**Supporting material**	**Operating flux (LMH)***	**Sludge concentration (g L^{-1})**
1L	1 layer	15.3 ± 0.6	1.9
2L	1 layer	14.8 ± 0.6	4.7
3L	1 layer	14.9 ± 0.9	9.2

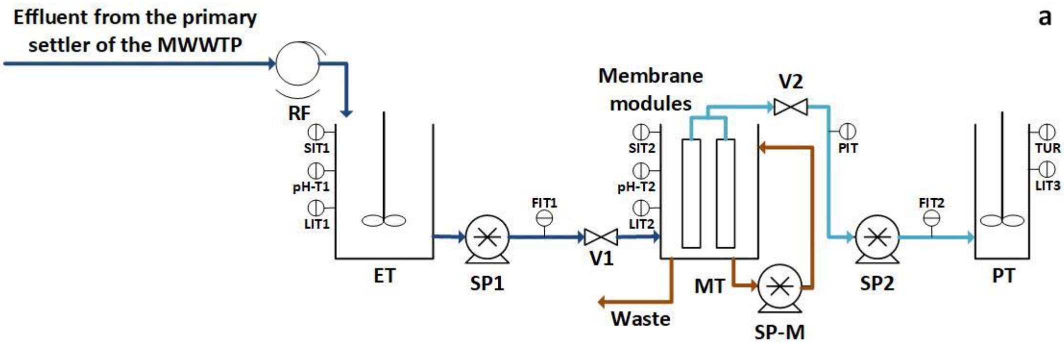
*Operating flux corrected to a temperature of 20 °C.

8.2.2 DM pilot plant

Fig. 8.1a shows a flow diagram of the DM pilot plant, which mainly consisted of a membrane tank (MT) (190-L working volume) equipped with two submerged flat membrane modules. To allow for the proper development of the DM on the supporting material, the employed woven mesh was attached to a rectangular supporting frame (1-m high and 0.5-m wide) to stiffen the supporting material. A large-pore steel woven mesh was added under each textile layer to stiffen the supporting material during filtration. The supporting frames were designed with two external surfaces open to the treatment liquor. One woven mesh was attached to each frame face, recovering the generated permeate in the interstitial space. This design allowed us to increase the membrane areas, providing a total filtration area of 2 m^2 (Fig. 8.1b shows a membrane module schematic draw).

The pilot plant was operated continuously at a given operating flux (see Table 8.2), performing filtration-relaxation cycles with a ratio of 3:1 min. The two membrane modules were connected to a screw pump (PCM, M series, EcoMoineau™) for vacuum filtration. A screw pump continuously mixed the concentrated sludge to ensure homogeneity in the membrane tank. The influent MWW was pre-treated with a 0.5-mm screen size roto-filter (PAM 270/500, Procesos Auto-Mecanizados, Spain) and homogenized in a stirred equalization tank (ET) (745 L). The wasting flow was set to 2.6 L h^{-1} to prevent significant aerobic microorganism development during filtration, operating at a solids retention time (SRT) of about 3 days. When coagulant dosing was required, a peristaltic lab pump continuously injected the coagulant solution into the MT. Fig. 8.1c shows a view of the pilot plant.

The pilot plant was equipped with several on-line sensors and automatic equipment to control and monitor all the involved variables (see Fig. 8.1a). The on-line sensors installed were: two pH-temperature sensors (InPro3100/120/PT100, Endress+Hauser) in the ET and MT; three level sensors (Cerabar PMP11, Endress+Hauser) in each tank (ET, MT and PT); one pressure liquid sensor (IP65, Druck) to monitor the transmembrane pressure (TMP); two solid concentration sensors (LXV424.99.00100, Hach) in the ET and MT; and one sensor to monitor the turbidity level (LXV424.99.00100, Hach) in the PT. For actuators, the pilot plant was equipped with different frequency converters (pumps and blowers) (SINAMICS G120C, Siemens) and control valves. Plant automation was carried out by a programmable logic controller (PLC) which performed the control and data acquisition of all the instrumentation installed in the pilot plant. A SCADA system was also used to gather all the information collected and allow their proper supervision, interaction and control.



RF:	Roto-filter	SP:	Screw pump	SIT:	Solids sensor
ET:	Equalization tank	SP-M:	Mixing pump	LIT:	Level sensor
MT:	Membrane tank	V:	Automatic valve	FIT:	Flow-rate sensor
PT:	Permeate tank			PIT:	Pressure sensor
				TUR:	Turbidity sensor
				pH-T:	pH-temperature sensor

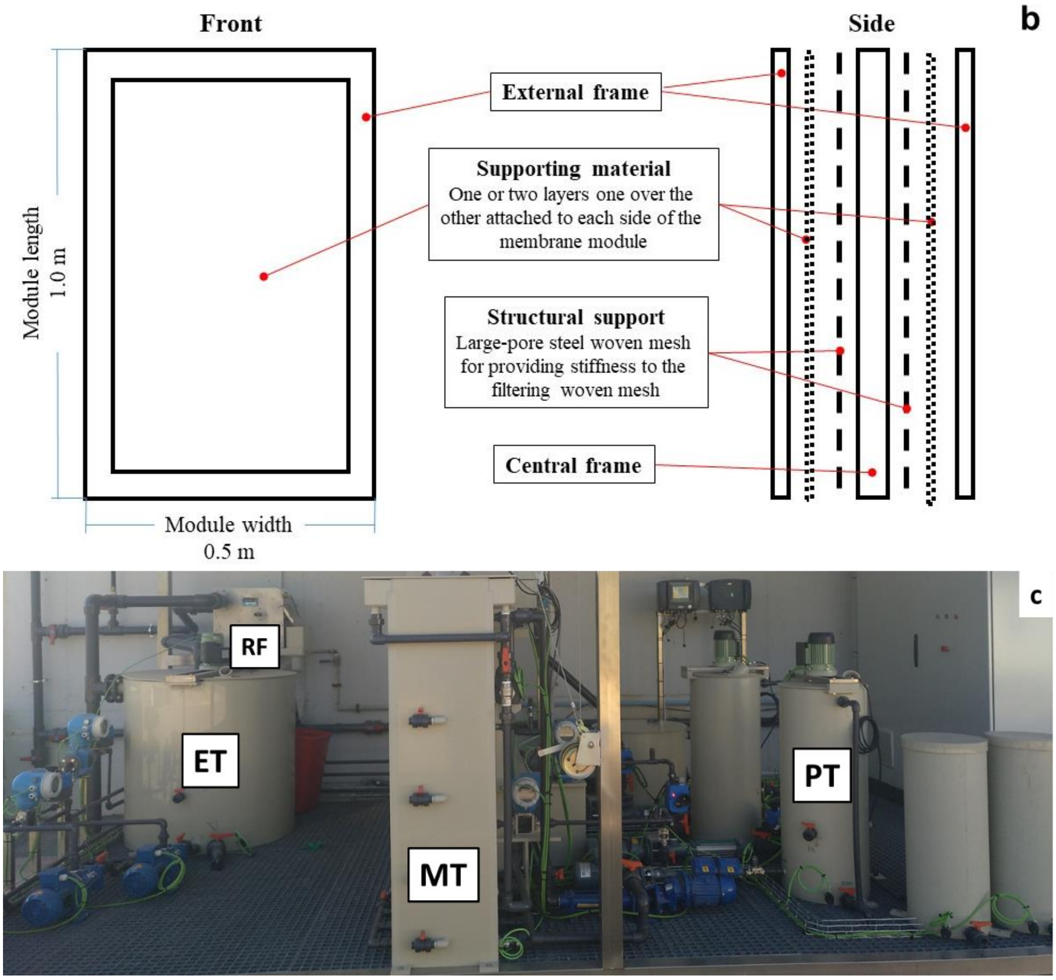


Figure 8.1. DM pilot-plant: (a) diagram scheme, (b) membrane module scheme and (c) picture.

8.2.3 Lab-scale DM

Like the pilot plant, the lab-scale DM mainly consisted of a membrane tank (8-L working volume) with submerged DM module for filtration. To allow for the proper development of the DM on the supporting material, the woven mesh was attached to a rectangular supporting frame (0.18-m height and 0.11-m width; 0.02-m^2 total area) to provide stiffness to the woven mesh. In this case, the supporting frame was designed with only one open surface in contact with the liquor, while the other side was closed to capture the generated permeate. The lab-scale DM was operated continuously, performing the filtration process according to filtration-relaxation cycles with a ratio of 3:1 min at an operating flux of 15 LMH. The permeate was obtained by vacuum filtration using a peristaltic pump and the TMP was recorded by a pressure captor (IP65, Druck) installed in the permeate side. Both TMP recording and peristaltic pump control were performed by a custom-made data acquisition software, processing the input and output signals through a multichannel data acquisition card (PicoLog 1000 series). To avoid the concentration of the MWW in the membrane tank during filtration, the permeate was recycled back to the membrane tank during continuous operation, only extracting the volume required for the sampling analysis. The content of the membrane tank was completely replaced every three days with new MWW to avoid the development of any kind of microorganism and was continuously homogenized by a supplementary peristaltic pump which continuously mixed the membrane tank sludge to avoid stratification or particle sedimentation. A PVDF hollow-fiber UF membrane (0.03 μm pore size, PURON® KMS) pre-concentrated the influent MWW to feed the lab-scale unit with the TSS concentration required in each experiment (see Table 8.2).

8.2.4 Analytical methods and calculations

Influent, membrane concentrated sludge and generated permeate were sampled twice a week to evaluate the DM resource recovery capacity. The soluble fraction of the collected samples was obtained by 0.45-mm pore size membrane filtration with glass fibre filters (Millipore). Solids, total and soluble chemical oxygen demand (COD and SCOD), total and soluble nitrogen (TN and SN) and total and soluble phosphorus (TP and SP) were determined according to standard methods [8.21]. A laser granularity distribution analyser (Malvern Mastersizer 2000; detector range of 0.01 to 1000 μm) was employed to determine the particle size distribution of the evaluated samples. Four different PACl coagulants were tested (Feralco Iberia S.A.) (see Table 8.3) to determine the most suitable and its optimum dosing concentration by means of a conventional jar-test (performed according to ASTM D2035-19).

Table 8.3. Features of the different coagulants used in this study (Feralco Iberia S.A.)

Coagulant	Product code	Formula	% Al_2O_3	% Cl^-	% SO_4^{2-}
1	PHAL 18	$Al(OH)_aCl_b$	17.0 ± 0.5	21.5 ± 1.0	-
2	PHAL 10	$Al(OH)_aCl_b(SO_4)_c$	10.0 ± 0.3	12.0 ± 0.5	2.1 ± 0.2
3	AQUALENC F1	$Al(OH)_aCl_b(SO_4)_c$	9.5 ± 0.5	12.3 ± 1.3	n.a.
4	AQUALENC F2	$Al(OH)_aCl_b(SO_4)_c$	9.0 ± 0.5	10.5 ± 0.5	1.5 ± 0.5

*All compounds are expressed in mass percentages. n.a.: not available.

The membrane performance analysis was studied by means of the recorded TMP, calculating the average TMP in every filtration cycle ($TMP_{average}$), while the 20 °C-standardized operating flux (J_{20}) was calculated according to the following expression:

$$J_{20} = J_T\, e^{-0.0239\,(T-20)} \qquad 8.1$$

Where T is the temperature and J_T is the imposed operating flux. The potential energy recovery from the captured COD (E_R) was calculated as follows:

$$E_R\ (kWh\ per\ m^3) = COD_{Influent}\ \ \%COD_{Captured}\ \ Y^{CH4}\ CV_{CH4}\ \eta_{CHP} \qquad 8.2$$

Where $COD_{Influent}$ represents the COD concentration feed to the DM module (kg m^{-3}), $\%COD_{Captured}$ is the percentage of COD captured by the DM during the filtration process (%), Y^{CH4} is the theoretical anaerobic methane yield of MWW sludge (0.35 m^3 of methane per kg of COD), CV_{CH4} is the calorific power of the methane (9.13 kWh per m^3 of methane), and η_{CHP} is the methane electricity generation efficiency of the employed CHP system. A η_{CHP} of 35% was used in this study considering the different CHP technologies currently available [8.22]. The energy costs were estimated at €0.07 per kWh according to current Spanish high voltage electricity rates [8.23; 8.24] while the coagulant costs were estimated at €200 per ton of coagulant, according to the data provided by the supplier (Feralco Iberia S.A.).

8.3 Results and Discussions

8.3.1 Pilot plant operation: Effect of operating conditions

Fig. 8.2 shows the results obtained during the operation of the DM pilot plant. Exp. 1 focused on determining the possibility of self-forming a DM on the supporting material (1 μm pore size flat open monofilament woven polyamide mesh) when using effluent from the primary settler of a full-scale WWTP. This experiment lasted for 24 days (from day 0 to 24 in Fig. 8.2) and no significant TSS captures were detected during continuous filtration, achieving average values of about 30% (see Fig. 8.2b). These low TSS captures were attributed to the filtering capacity of the supporting material itself, which would be able to retain mainly all the particles above 1 μm in size. Neither were any important TMPs detected during filtration, achieving values of about 20 mbar during the filtration stages (see Fig. 8.2a), suggesting that a negligible cake layer formed on the supporting material. Since an evolution of the DM was not appreciated during the first experimental period, the membrane frame was taken out of the membrane tank to check DM development. As Fig. 8.3b shows, very poor particle deposition was found on the supporting material after Exp. 1, showing that DM had not even started to form. Based on these results, it was concluded that the self-formation of a stable DM did not seem feasible for the supporting material and MWW studied, at least in the short-term. The results obtained during this experiment contrasted with the results reported by other studies treating MWWs by DMs, in which between 2 and 20-h self-forming times were reported using similar or even larger pore size supporting materials (between 1 and 100 μm) [8.10; 8.11]. In these studies, however, raw MWW was used as influent to feed the membrane tanks, which would contain a higher amount of particulate material with a higher average particle size. Additionally, more suspended material (diatomite) was added in one of the cited studies for enhancing the DM formation [8.11]. All this additional particulate material would favour the development of the DM on the supporting material, especially the larger particles, which would boost the formation of a cake layer on the supporting material in the first steps. Therefore, as anticipated, the use of a more pre-treated influent, such as the one used in this study (PSE), could represent a limitation of DM applicability.

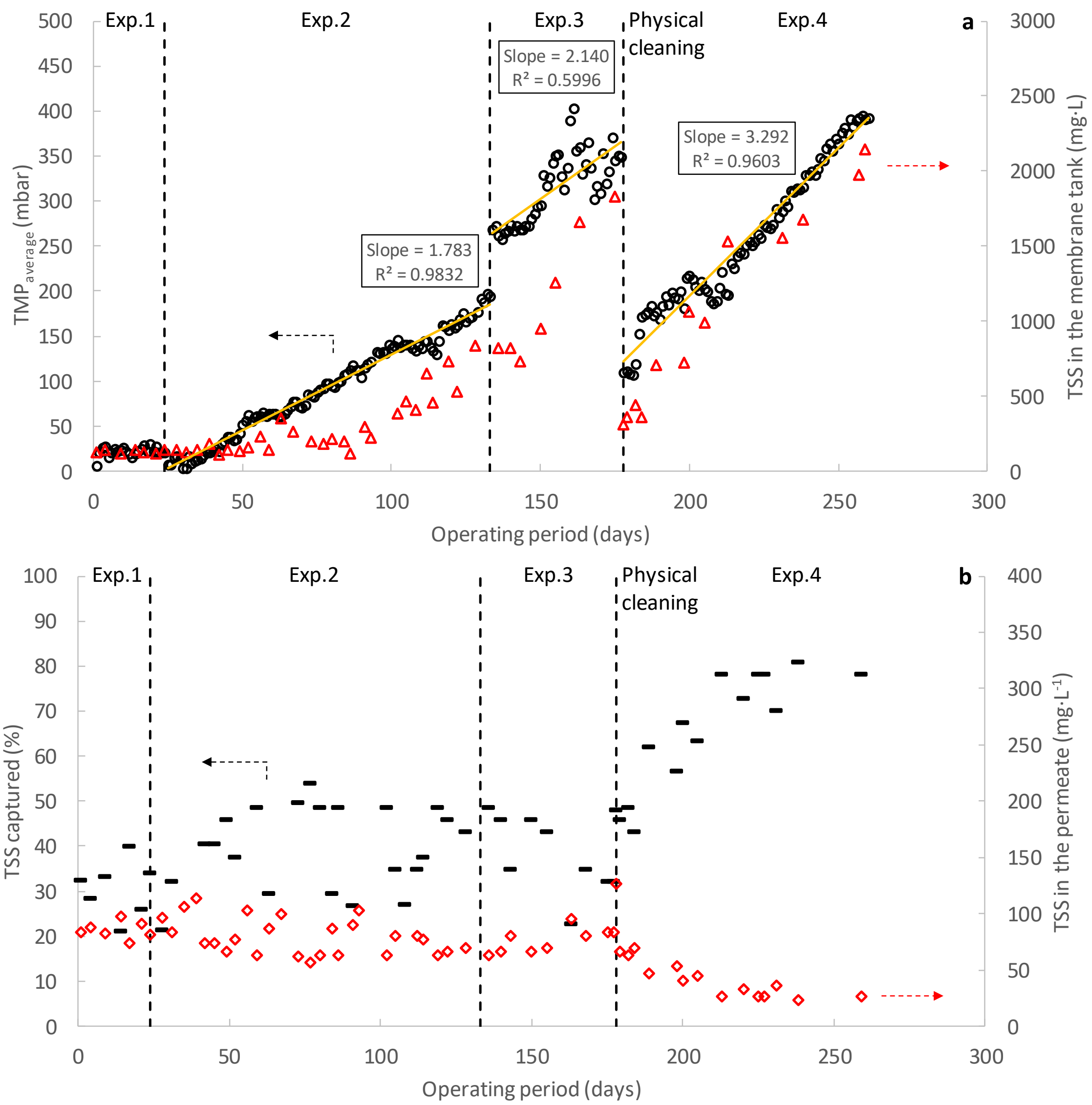


Figure 8.2. Pilot-plant performance. Evolution of (a) transmembrane pressure (TMP) and total suspended solids (TSS) concentration, and (b) TSS capture efficiency and permeate TSS concentration. o $TMP_{average}$; △ TSS in the membrane tank; ‒ TSS capure efficiency; ◇ TSS in the permeate. The continuous lines represent linear fits.

Considering the results obtained in Exp. 1, Exp. 2 was designed to enhance particle deposition on the supporting material to boost DM formation. For this, an additional woven mesh was added to each membrane frame surface, doubling their thickness. Since the new meshes were not aligned with the old, this strategy could improve DM self-forming capacity by both: (1) apparently reducing the average pore size of the supporting material, and (2) increasing the probability of contact between the threads of the woven mesh and the medium suspended particles. In fact, this strategy has been used by other authors to improve solids capture capacity [8.10], reducing turbidity in the generated permeate and increasing filtration resistance. Exp. 2 lasted for 108 days (from day 25 to

133), observing similar TSS captures and TMP values as those achieved in Exp. 1 during the first few days. However, after 17 days of operation (day 42), both TSS capture and TMP gradually increased daily, suggesting the development of a DM on the supporting material. Indeed, TMP steadily increased during the following 91 days (from around 20 to 190 mbar from the day 42 to 133), which could be related to the accumulation of more particles on the supporting material and the consolidation of the pre-formed DM. The TSS in the membrane tank also started a steady increase due to the enhanced DM solids capture efficiency (from around 110 to 840 mg L^{-1}), which contributed to increasing TMP. However, despite the DM consolidation and the consequent rise of solid concentration in the membrane tank, TSS capture efficiency only rose to values of around 45%, reaching a pseudo-steady state after the first 18 days of operation in Exp. 2 (day 43 in Fig. 8.2). From then on, TSS capture efficiency remained static for the rest of the period, regardless of the increase in the operating TMP or the TSS concentration in the membrane tank. Due to this low TSS capture efficiency, a relatively poor permeate quality was obtained in Exp. 2, achieving TSS, COD, TN and TP concentrations and turbidity values on the generated permeate of about 65 mg L^{-1}, 141 mg L^{-1}, 42.3 mg L^{-1}, 4.3 mg L^{-1} and 86 NTU, respectively. Table 8.4 shows the permeate quality achieved during the experimental periods.

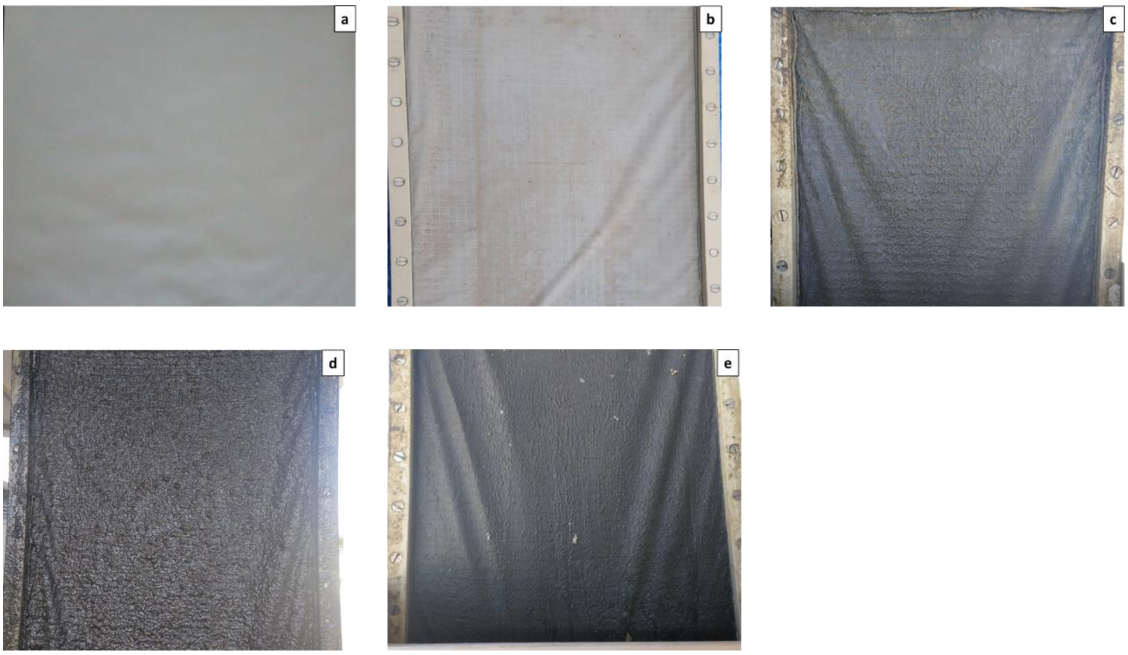


Figure 8.3. Pilot-plant supporting material after every experimental period: (a) New, (b) Exp. 1, (c) Exp. 2, (d) Exp. 3 and (e) Exp. 4.

Table 8.4. Permeate quality

Exp.	TSS		Turbidity		COD		TN		TP	
	(mg L^{-1})	(%)*	(NTU)	(%)*	(mg L^{-1})	(%)*	(mg L^{-1})	(%)*	(mg L^{-1})	(%)*
2	65	58	86	79	141	84	42.3	93	4.3	73
3	59	52	94	86	138	83	42.9	94	4.4	75
4	24	21	22	20	58	35	38.1	84	1.2	20

*Percentage of the influent pollutant remaining in the permeate.

Fewer resources were recovered from Exp. 2 than in other studies treating raw MWW. Indeed, when using similar or even higher pore-sized supporting materials (between 1 and 100 µm), COD and turbidity recoveries between 63–71% and 60%, respectively, are reported in the literature [8.10; 8.11]. Like the DM self-forming capacity, these different results are related to the more treated influent used in this study (*i.e.* PSE), which could affect resource recovery by (1) the development of a less thick DM, and (2) an intrinsic reduction of the resources that can be captured in the DM due to inclusion of the primary settler in the treatment scheme. Initially, the formation of a poor DM when using the PSE was considered, since the lower influent solids concentration could contribute to a weaker and less consolidated DM due to the reduced particle content attached to the supporting material. However, as Fig. 8.3c shows, the DM developed during Exp. 2, although not too thick, seemed homogenous and robust enough to allow proper filtering treatment (day 133 in Fig. 8.2). In addition, the particle size distribution analysis (see Fig. 8.4) showed that together with the increasing TSS concentration, the average particle size of the retained particles consistently increased, which shows the DM's capture capacity. This, together with the increased TMP during the experiment confirms that the DM formed was well developed. It was therefore assumed that the consistency of the DM during Exp. 2 was not directly related to the low resource recovery efficiencies achieved. On the other hand, despite the low resource capture capacity detected, relatively similar permeate qualities to those reported by the cited studies (*i.e.* [8.10; 8.11]) were also obtained. The poor recovery capacity of our study could thus be due to the low suspended material loading influent treated, having captured a considerable fraction of the particulate material from the raw influent MWW in the primary settler. If this was the case, the influent used would not affect the permeate quality and the most suitable treatment scheme for full-scale implementation (*i.e.* direct filtration of raw MWW or the use of a primary settler as pre-treatment step) would be determined by the energy required during the filtration process in each scenario.

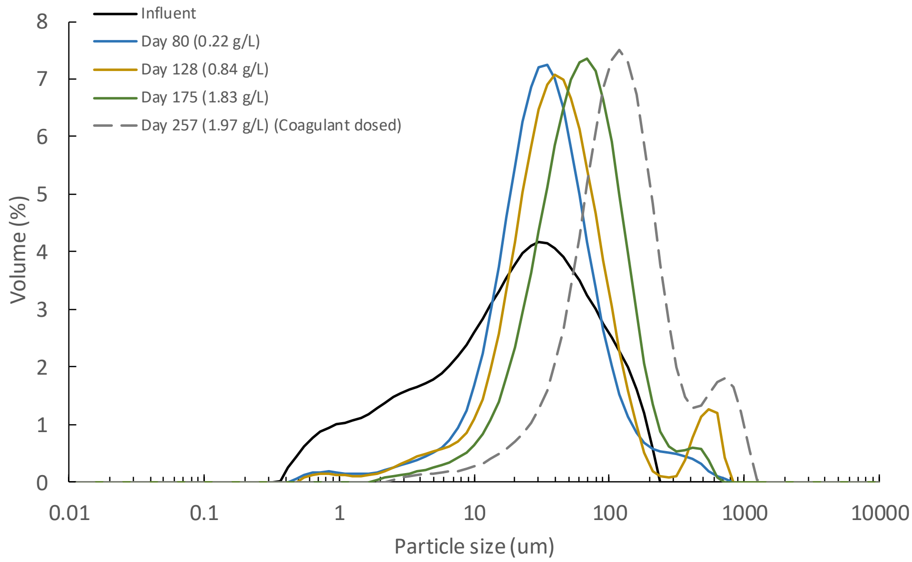


Figure 8.4. Particle size distribution of the concentrated sludge during the pilot-plant operation. Note that legend shows the day, together with the total suspended solids concentration in the pilot-plant membrane tank during sampling.

Considering the results obtained from Exp. 2, Exp. 3 was designed to increase the DM capture capacity. During this experimental period, the operating flux was increased from 15 to 45 LMH to favour particle deposition on the DM and induce cake compression caused by deformation of soft flocs and the structural rearrangement of particles. In fact, other authors have suggested that a more compressive and dense cake layer can be formed when filtering more treated influents due to the large number of small particles present [8.20; 8.25]. Permeate quality could thus be improved by creating a dense DM with a smaller apparent pore size. Due to the significant time required to develop a consistent DM on the supporting material, Exp. 3 used the DM formed during the former experience. Exp. 3 lasted for 44 days (from day 134 to 178 in Fig. 8.2), showing few improvements of the DM resource capture capacity (see Fig. 8.2b and Table 8.4). Although the TMP rose abruptly in the first days of operation due to the larger operating flux, it then behaved like Exp. 2, which indicates similar DM filtering resistance (see Fig. 8.2a). In fact, relatively similar fouling growth rates were achieved during the two experimental periods, obtaining a daily TMP increment of about 1.8 and 2.1 during Exp. 2 and Exp. 3, respectively. The particle size distribution (see Fig. 8.4) and the DM physical observation after this operating period (see Fig. 8.3d) also behaved as in Exp. 2. The results indicate that the increase in the operating flux did not alter DM morphology and constitution in the short-term. Other strategies must therefore be proposed to enhance DM capture capacity.

8.3.2 Pilot plant operation: Coagulant dosing

Coagulant dosing was tested as a second alternative to enhance the pilot plant's resource recovery. Four different PACl coagulants were tested (see Table 8.3). The most suitable and its optimum dosing concentration was first evaluated by a conventional jar-test. As Fig. 8.5 shows, all the employed coagulants achieved considerable pollutant captures at relatively low concentrations, including, as expected, not only a large fraction of the particulate material, but also a significant fraction of the colloidal material (which can be seen by the reduction of SCOD) and the SP. In fact, turbidity, COD, SCOD and SP reductions of up to 86, 78, 42 and 93%, respectively, were achieved during the jar-test at coagulant concentrations between 5–20 mg L^{-1}. However, coagulant concentrations over 40 mg L^{-1} seemed have negative effects on solids capture in some cases, which was attributed to a destabilization of the medium charges when increasing the coagulant concentration [8.26]. The optimum concentration range obtained in this study was similar to that reported by other authors filtering MWW, who usually recommend PACl concentrations of around 15–30 mg L^{-1} [8.8; 8.27]. On the other hand, significant SN captures were not expected or observed during this experience, since there were no relevant chemical interactions between soluble nitrogenous compounds (mainly NH_4^+) and the inorganic coagulants. Finally, no great pH changes were found for the coagulant concentrations tested, although a slight reduction as coagulant concentration was increased can be seen (see Fig. 8.5). Due to the relatively high alkalinity of the MWW studied (see Table 8.1), this perturbation was considered negligible but could be a relevant issue in other situations. The type and optimum concentration of coagulant determined in this study could thus change in different circumstances. In this study, coagulant 2 (PHLA 18) with a concentration of 10 mg L^{-1} was chosen to operate the DM due to its slightly higher COD and SP captures than the rest of the coagulants tested.

A further experiment (Exp. 4) was then carried out focusing on the beneficial effects of continuous coagulant dosing on the DM's performance. To properly determine the improvement in its forming time when dosing coagulant, the supporting material was physically cleaned before the experience. Exp. 4 lasted for 81 days (from day 179 to 260 in Fig. 8.2) and a shorter forming time (of about 7 days) than Exp.2 was obtained. The operating TSS concentration and TMP increased faster during this experiment, especially in the early days. These phenomena were due to the enhanced solids capture efficiency when coagulant was dosed in the membrane tank, capturing more of the smaller particles by forming larger aggregates. Indeed, the results of the particle size distribution analysis showed a significant increase of larger particles than the influent MWW (see Fig. 8.4). A higher amount of particulate material thus ended on the supporting material, boosting DM development. The increased solids capture capacity also accelerated the TSS concentration rate and raised the operating TMP. In this regard, other studies treating MWWs by membrane systems have showed the importance of optimizing coagulant dosing during filtration, achieving severe increases in the operating TMP with high coagulant concentrations due to the sudden accumulation of captured solids on the membrane surface [8.17]. As Fig. 8.2b and Table 8.4 show, a significant improvement of solids capture efficiency was achieved when the coagulant was dosed. As previously mentioned, this increase was due to the capture of the small size particles, reducing significatively the turbidity of the medium. Nevertheless, as the jar-test showed (see Fig. 8.5), coagulant dosed was unable to capture a sensible fraction of the influent colloidal material. Thus, the remaining solids detected in the permeate would be due to this colloidal fraction, together with some formed aggregates smaller than the DM average pore size, being all this particulate material able to cross through the DM and escape with the permeate. Considerable COD and TP recoveries were also achieved during Exp. 4 thanks to the capture of a fraction of the colloidal material and the chemical precipitation of phosphate when dosing the coagulant. Coagulants could thus be used to enhance MWW treatment when using DMs; however, the coagulant dosing protocol plays a critical role in the filtration process and should be carefully chosen to boost resource recovery while minimizing filtration energy demand during long-term operations. Moreover, aluminum-based coagulants, such as PACl, are usually identified as anaerobic digestion inhibitors [8.28], thereby reducing the energy potential of the recovered sludge. Thus, coagulant dosing minimization during filtration should be an imperative matter not only for minimizing chemicals costs, but also for avoiding recovered sludge biodegradability issues. In this regard, Hafuka *et al.* [8.29] studied the effect of PACl coagulants on the biodegradability of sludge recovered from DMF processes, reporting that Al concentrations of about 4.3 mg L^{-1} do not represent problems on the anaerobic digestion methane production. In the performed study, assuming that all the Al was captured by the membrane rejection and considering the operating permeate/waste ratio (30:2.6), Al concentrations of about 10.4 mg L^{-1} could be expected in the recovered sludge, which are not significantly superior to those reported in the cited work. Thus, no important energy recovery issues could be assumed for the recovered sludge in this case.

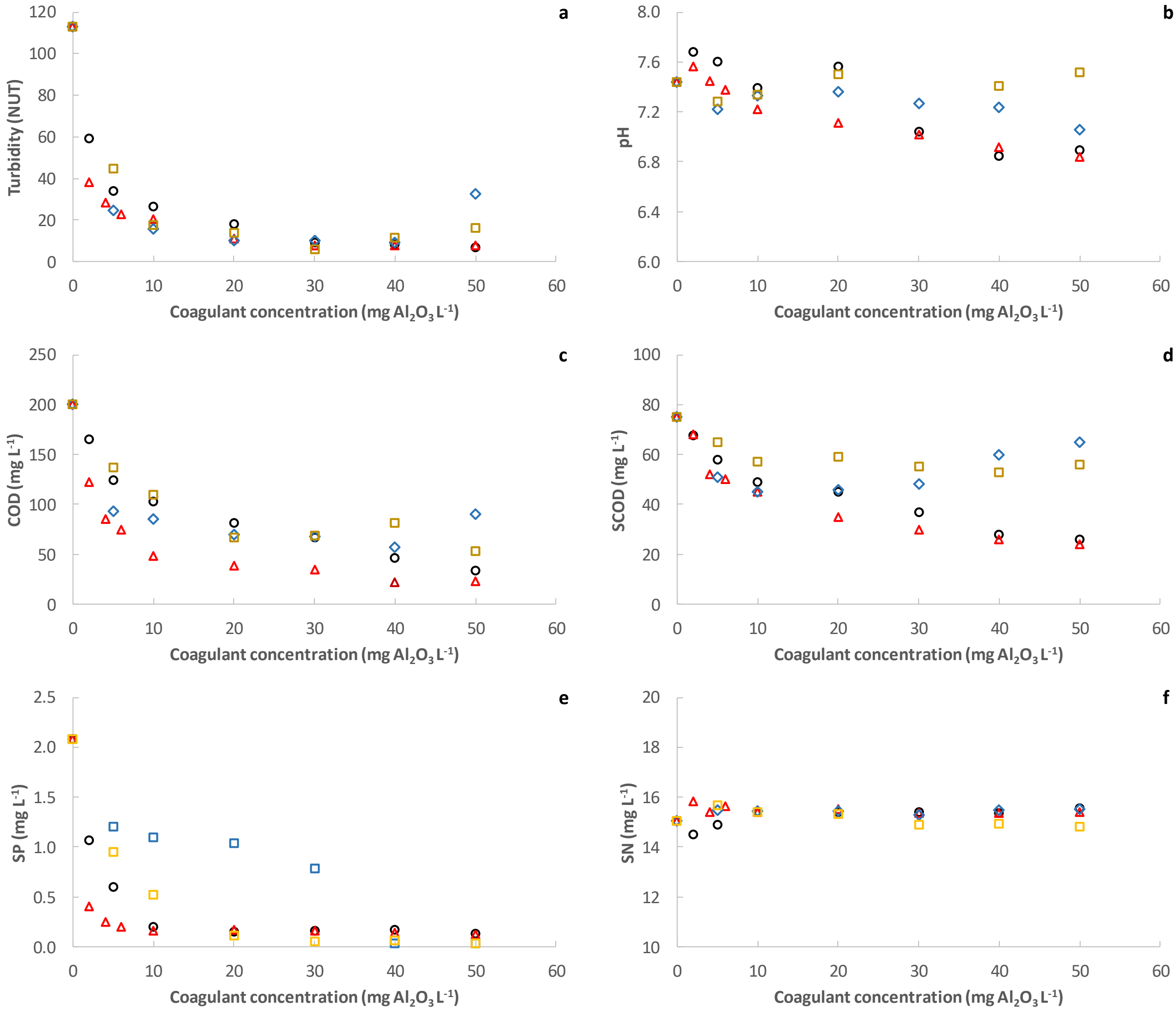


Figure 8.5. Effect of coagulant dosing on pollutant capture during jar-test: (a) turbidity, (b) pH, (c) chemical oxygen demand (COD), (c) soluble chemical oxygen demand (SCOD), (e) soluble phosphorus (SP) and (f) soluble nitrogen (SN). Coagulants used: ○ PHLA10; △ PHLA18; ◇ F1; □ F2.

Finally, after Exp. 4 was concluded, the coagulant dosing was stopped, and the membrane was operated for 5 additional days to study permeate quality (data not shown). Unfortunately, a pretty similar permeate quality to those obtained in Exp. 2 and Exp. 3 was quickly achieved, showing that all the capture improvements in Exp. 4 were only due to coagulant effects and not to a change of DM structure. The visual analysis of the DM formed at the end of Exp. 4 also seemed to indicate that the DM structure remained unaltered, whatever the coagulant dosing (see Fig. 8.3e). Nevertheless, this performance could change in long-term operations, forming a thicker and more compact DM which could itself raise the resource capture efficiency. In fact, the DM maturation period can last for several days, enhancing pollutant capture efficiency on reaching their mature state [8.15]. Further studies focused on dynamically optimizing the coagulant dosing protocol, considering all the important aspects (*i.e.* chemicals cost, filtration energy demand, resource recovery efficiency, sludge biodegradability and permeate quality) therefore need to be performed.

8.3.3 Lab-scale results: Effect of solids concentration

To discover the effect of operating TSS on short-term DM formation, the MWW used during this study was pre-concentrated to different TSS concentrations (see Table 8.2), and then fed to the lab-scale membrane tank before each essay. Each experiment lasted for about 15 days except for the concentration of 9.2 g L^{-1}, when the experiment was stopped on the 6th day due to the severe rise of TMP. Fig. 8.6 shows the results obtained during the lab-scale operation. The DM self-forming time onto the supporting material was significantly reduced by pre-concentrating the treated influent. In fact, self-forming times of between 4–8 days were achieved in this case, although only one supporting material layer was used. This phenomenon was associated with the higher number of particles that can be attached to the supporting material. A significant increase of the particle size distribution to higher particles sizes was also detected when concentrating the influent MWW (see Fig. 8.7). This could be due to the sporadic flocculation of the smaller particles when increasing contact and collisions among particles at higher TSS concentrations. Higher TMPs were also obtained as the TSS concentration was raised in the membrane tank (see Table 8.5), which would be related with a higher accumulation of particulate material onto the formed cake layer during filtration. These results thus confirm that increasing TSS concentration is a feasible alternative to boosting DM development, but at the cost of considerably higher TMP. However, as Fig. 8.6a shows, the permeate quality obtained regarding TSS in all the experiments was pretty similar, practically coinciding with those obtained during the pilot plant operation. Additionally, when calculating the TSS capture efficiency based on the original influent used in this study (see Table 8.1), low values were also obtained, regardless of the TSS concentration in the membrane tank (see Fig. 8.6b). Therefore, these results may indicate that the solid capture efficiency of the short-term formed DM could be related higher with the influent characteristics than with the operating TSS, expecting thereby similar permeate qualities at least concerning solids' concentration. Nevertheless, as commented above, this could significantly change in long-term operations, and further studies are required to determine the most suitable DM solids concentration when treating MWW.

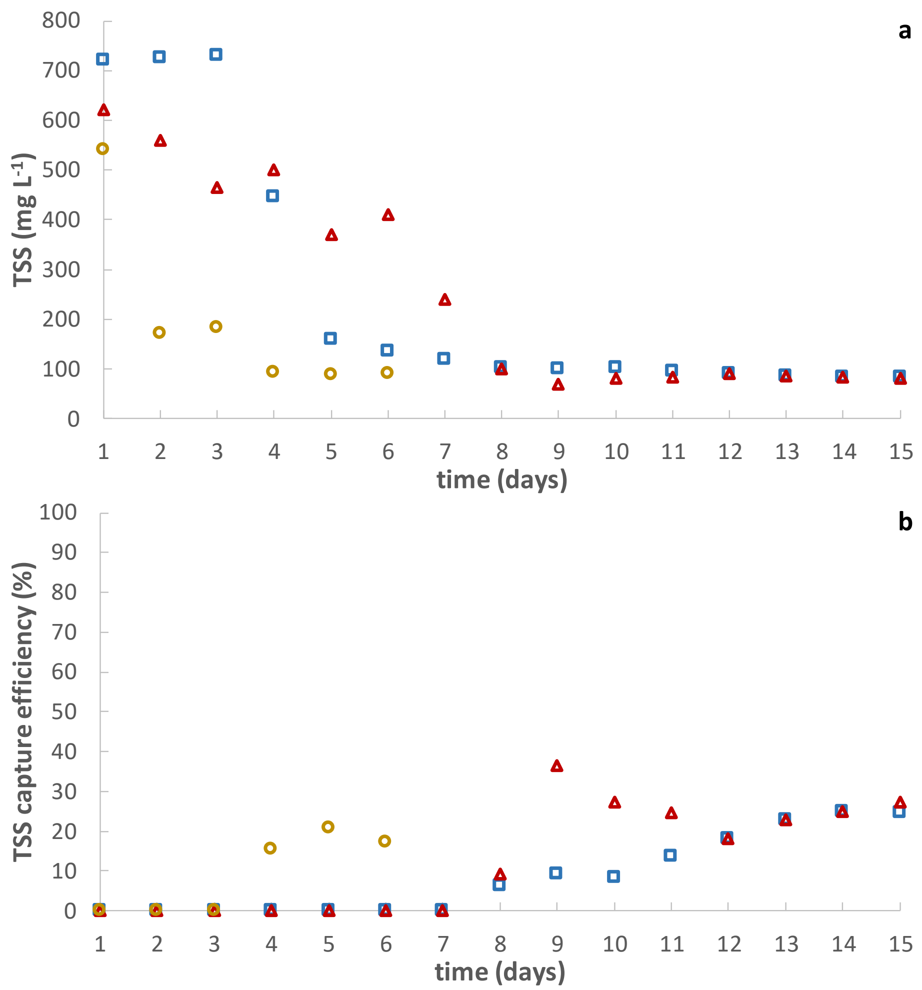


Figure 8.6. Lab-scale DM performance. Effect of influent total suspended solids (TSS) concentration (□ 1.9 g L^{-1}; △ 4.7 g L^{-1}; ○ 9.2 g L^{-1}) on: (a) TSS in the permeate and (b) TSS capture efficiency. Note that the TSS capture efficiency was calculated based on the original influent used (see Table 8.1).

Table 8.5. Average transmembrane pressure (TMP) after the dynamic membrane formation

Exp.	Sludge concentration (g L^{-1})	Operating days	Self-forming period (days)	Average TMP (mbar)
1L	1.9	15	8	56
2L	4.7	15	5	198
3L	9.2	6	4	384

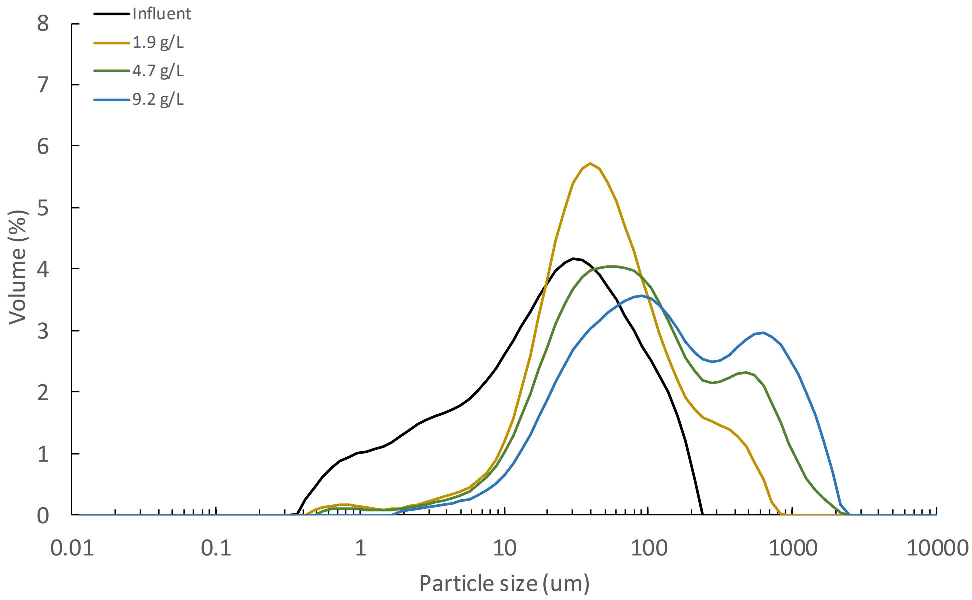


Figure 8.7. Particle size distribution of the concentrated sludge during the lab-scale operation. Note that legend shows the total suspended solids concentration fed to the lab-scale membrane which was obtained by concentrating the influent with an ultrafiltration membrane.

8.3.4 Operating recommendations

Aiming to roughly discern the most suitable operating approach when operating DMs with PSE as feed, a simplified economic balance was performed on every experimental period evaluated in this study, considering only energy recovery and coagulant costs. As can be seen in Table 8.6, there were negligible differences between Exp. 2 and Exp. 3, since the resource capture efficiency was similar in both cases. Since a similar DM formation and fouling was also found in these experimental periods (see Fig. 8.2), increasing the operating flux as much as possible could be recommended to minimize investment and space costs as long as it does not compromise the energy required for filtration or supporting material replace periodicity. On the other hand, the enhanced resource recovery efficiency achieved by the coagulant dosing (Exp. 4) seems not to overcome the expenses of the chemicals involved, requiring slight economic inputs despite the higher energy recovery (see Table 8.6). However, since no great differences were obtained between the economic impact of Exp. 2 and Exp. 4, the use of coagulants can still be recommended as an interesting strategy to boost the DM formation capacity and increase resource recovery. Moreover, other side effects such as higher phosphate recovery or the environmental impact of using these chemicals should also be considered. Thus, further studies are needed to properly assess the suitability of dosing coagulant in this alternative treatment scheme. Regarding the operating TSS concentration, this study showed that although increasing them can favour DM self-forming time, important enhancements of resource capture efficiency cannot be obtained in the short-term. This strategy would thus negatively affect the required filtration energy due to the significant increase in operating TMP. Since coagulant dosing can significatively reduce DM forming time while improving resource capture capacity, relatively low operating TSS could be recommended when operating a DM for treating MWW.

Table 8.6. Energy and operating cost of DM operation

Exp. Num	MWW treated	Energy recovery (kWh m^{-3})	Energy costs (€ m^{-3})	Coagul. costs (€ m^{-3})	Costs output (€ m^{-3})	Reference
2	PSE	0.029	-0.002	-	-0.002	This study
3	PSE	0.032	-0.002	-	-0.002	This study
4	PSE	0.121	-0.009	0.012	0.003	This study
-	Raw	0.101	n.a.	n.a.	n.a.	[8.10]
-	Raw	0.127	n.a.	n.a.	n.a.	[8.12]

n.a.: not available.

Comparing the results obtained in this work with other studies using DMs to treat raw MWW, significantly lower energy recoveries were achieved (see Table 8.6), only reaching similar results when coagulant was dosed. These results were attributed to the different influent used in this study, since raw MWW have a higher number of recoverable resources while in the proposed alternative, the primary settler recovers a significant fraction of these resources. Thus, taking into account that about 50% of the raw influent TSS would be recovered in the primary settler, the overall energy outputs achieved by the proposed alternative would increase to 0.215–0.308 kWh per m^3 of treated MWW, values higher than the energy recovery reported when directly filtering raw MWW. Since a lower fouling rate could be expected when filtering more treated influents due to the reduced fraction of influent pollutants, the proposed alternative could be an interesting approach to boost resource recovery while reducing the required filtration energy.

In addition to the discussion made in this section, other considerations need to be taken into account to properly choose the most suitable DM operating conditions. Fouling development during DM operation should be carefully controlled by employing continuous physical cleaning methodologies (*e.g.* air scouring), determining the optimum conditions to minimize filtration energy requirements without compromising DM integrity or permeate quality. The operating TSS should also be optimized not only considering the energy required for filtration, but also the subsequent use of the concentrated sludge (*i.e.* methane production via anaerobic digestion). Thus, all the extra steps and full energy requirements for using this sludge should also be considered (pumping demands, sludge thickening, etc.) to determine the most feasible operating conditions for the overall process. Similarly, permeate quality should be adjusted according to its foreseen use (direct discharge to water bodies, tertiary wastewater treatments, etc.), which could significantly influence the proper operating flux or coagulant dosing. On the other hand, other improvements could be made concerning the membrane operating parameters. In this study, a high waste/influent operating ratio was used in order to avoid a high sludge retention time in the membrane tank, which would be an undesirable full-scale operating condition due to the high flow rate of the produced waste. Thus, reducing the membrane tank volume as much as possible would be an important design strategy for boosting the energy and economic balances of this technology, as it would reduce the waste stream to treat while increasing its TSS and COD concentration, significantly reduce the membrane tank sludge retention time and also reduce the process space requirements.

It can thus be concluded that treating PSE by DM can be considered an interesting alternative within the DMF approach to improve resource recovery from MWW while reducing process energy requirements. However, further studies, considering all the above exposed and comparing the results obtained with other membrane technologies (*e.g.* other supporting materials, MF and UF membranes, etc.) and influents (*e.g.* raw MWWs) need to be performed to properly determine the best scenarios for full-scale implementation of the proposed alternative.

8.4 Conclusions

DM feasibility for treating MWW was evaluated in this study. The main findings were as follows:

- One layer of the supporting material (a flat open monofilament woven polyamide mesh of 1 µm average pore size) was not enough to self-form a DM in the short-term when treating PSE from a full-scale WWTP, showing the limitations of DMs for treating more depurated influents. Nevertheless, a proper DM was self-formed when using two supporting material layers (17 days of operation) or when increasing the operating TSS concentration (8, 6 and 4 days of operation for a TSS concentration of 1.9, 4.7 and 9.2 g L^{-1}).

- Similar permeate qualities were obtained regardless of filtration flux and TSS tested in this study, achieving TSS, COD, TN, TP and turbidity values of 65 mg L^{-1}, 141 mg L^{-1}, 42.3 mg L^{-1}, 4.3 mg L^{-1} and 86 NTU, respectively.
- Coagulant dosing improved both the required forming time and DM permeate quality. Optimum coagulant (PHLA18) dosing of 10 mg L^{-1} was determined, achieving a DM forming time of 7 days and a permeate quality of TSS, COD, TN, TP and turbidity of 24 mg L^{-1}, 58 mg L^{-1}, 38.1 mg L^{-1}, 1.2 mg L^{-1} and 22 NTU, respectively.
- Preliminary energy and economic balances showed that energy recoveries from 0.032 to 0.121 kWh per m^3 of treated water at an economic cost of from -€0.002 to €0.003 per m^3 of treated water can be obtained from the recovered particulate material.

8.5 Acknowledgments

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CHAPTER 9. Evaluating the Feasibility of Employing Dynamic Membranes for the Direct Filtration of Municipal Wastewater

Abstract

The aim of this study was to assess the feasibility of using dynamic membranes for direct filtration of municipal wastewater. The influence of different alternative supporting materials (one or two layers of flat open monofilament woven polyamide meshes with 1 or 5 μm of pore size) was studied. A stable short-term self-forming DM was achieved (from some hours to 3 days) regardless of the supporting material used, producing relatively similar permeate qualities (total suspended solids, chemical oxygen demand, total nitrogen, total phosphorous and turbidity of 67–88 mg L^{-1}, 155–186 mg L^{-1}, 48.7–50.4 mg L^{-1}, 4.7–4.9 mg L^{-1}, and 167–174 NTU, respectively). A DM permeability loss rate of from 5.21 to 10.03 LMH bar^{-1} day^{-1} was obtained, which depended on the supporting material used. Unfortunately, the preliminary energy, carbon footprint, and economic evaluations performed showed that although DMs obtain higher pollutant captures than conventional treatments (primary settler), the benefits are not enough to justify their use for treating average municipal wastewater. However, this alternative scheme could be suitable for treating higher-loaded MWW with a higher fraction of organic matter in the non-settleable solids.

Keywords

Direct membrane filtration; Dynamic membranes; Resource recovery; Municipal wastewater treatment

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9.1 Introduction

The current economic models based on non-renewable resources are now showing their limitations for long-term sustained development. All the estimates forecast the increasing scarcity of important essential resources, such as fresh water, energy and nutrients [9.1]. Finding alternatives to fossil fuels to produce energy is also essential to minimize climate change. Numerous experts have announced an urgent need to adapt our consumption to circular economy models to achieve sustainable human development [9.2]. Following this approach, municipal wastewater (MWW) treatment is now experiencing an important paradigm shift. In fact, MWW could not only be a source of recycled water, but also a source of energy and basic nutrients (nitrogen and phosphorus) [9.3]. Unfortunately, the current municipal wastewater treatment plants (WWTP) are unable to completely recover all the potential resources contained in MWW. Conventional activated sludge (CAS) systems, which are the core of MWW treatment, fail to recover the potential energy in influent organic matter by removing it via biological oxidation only. The nutrients present are also usually wasted, removing the ammonium concentration by nitrification/denitrification and precipitating the phosphate by chemicals that disable them for reuse. CAS treatments also demand a large amount of energy for removing organics from sewage, representing around 30–60% of the total WWTP energy requirements [9.4]. Alternative treatment schemes thus need to be developed to take advantage of all the potential resources from MWW and change the former conception of WWTPs to new resource recovery facilities.

Several alternatives have been developed to achieve sustainable MWW treatments, anaerobic membrane bioreactor technology being one of most attractive due to the possibility of converting influent organic matter into methane [9.5]. However, the considerable financial investment required to adapt current installations to alternative systems hinders their introduction. In this context, direct sewage filtration, defined in the literature as direct membrane filtration (DMF), has recently emerged as an interesting alternative for upgrading the energy efficiency and resource recovery of current WWTPs [9.6]. This strategy consists of implementing a membrane system before the CAS process to capture the material suspended in the influent. Thanks to this previous filtration, a large amount of influent organics (the complete particulate fraction) would be recovered before CAS treatment, dramatically reducing the aeration demands of the process and MWW energy requirements. The organic matter captured in the membrane tank could be used to produce methane via anaerobic digestion (AD), enhancing the overall process energy balance even further. A significant portion of influent nutrients (suspended fraction) could also be recovered in the membrane tank's concentrated sludge, allowing the DMF to enhance the overall WWTP resource recovery potential without seriously modifying the installations. MF and UF membranes have been extensively tested for this and have obtained promising results [9.7]. Unfortunately, numerous studies have reported severe membrane fouling when operating these membranes with untreated MWW [9.8; 9.9], which compromises their feasibility by requiring energy-consuming membrane fouling control strategies during filtration. Additionally, low/moderate permeate fluxes have been recommended when operating these systems [9.10], sharply increasing the initial investment in membranes for full-scale implementations. This suggests that membrane systems with lower fouling propensities, such as dynamic membranes (DM), could be an interesting alternative to conventional membrane systems to carry out DMF treatment schemes.

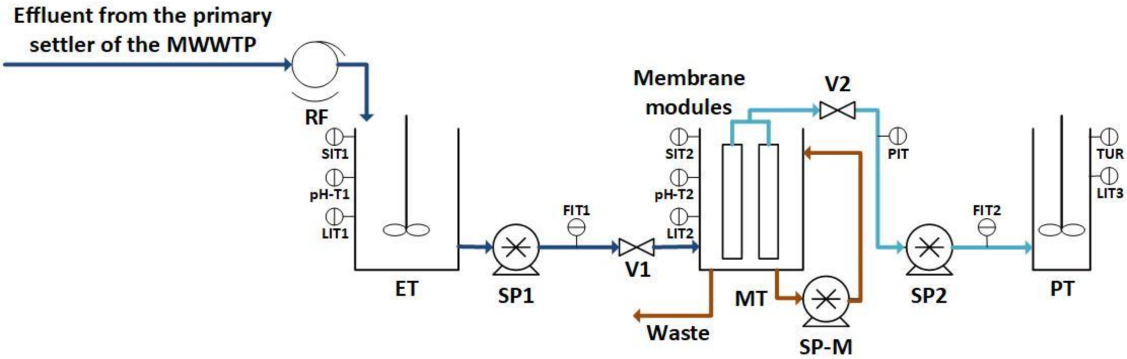
DMs consist of the formation of a stable cake layer on a low filtration-resistance supporting material, which is the main filtration element [9.11]. Thanks to removing the intrinsic filtration resistance of conventional membranes, higher permeabilities can be achieved during filtration, which can be controlled by acting on the thickness and density of the cake layer [9.12]. Membrane fouling thus changes its paradigm, in this case by playing a partially beneficial role that can be easily controlled by physical low energy cleaning [9.12]. In addition, the supporting structures are generally made of low-cost materials, such as woven meshes or filter-cloths, which have a significantly lower acquisition and/or replacement cost than conventional membrane modules [9.13]. However, despite their potential benefits, DMs involve different issues that need to be addressed. Significantly worse permeate qualities can be expected when using DMs instead of MF or UF, since the cake layer formed has a less homogeneous structure with higher porosity than commercial membranes. As the cake layer mainly controls filtration performance, the permeate generated by DMs may be unstable to some degree, changing according to the characteristics of the DM formed during filtration. The formation of a stable DM can also represent difficulty in some cases, since its formation strongly depends on the characteristics and concentration of the material suspended in the influent and its interaction with the supporting structure. In this respect, two different DMs can be distinguished, depending on whether the filtering cake layer developed on the supporting structure is self-forming or pre-coated [9.11; 9.14]. Self-forming DMs are created when the filtering cake layer consists of direct deposits of particulate material on the supporting structure during filtration, while pre-coated DMs consist of a previously stable structure on which the influent particulate material can be deposited to form the filtering cake layer. Pre-coated DMs are in theory less advantageous, since they require auxiliary chemical dosing, which increases their operating costs [9.14]. Selecting a proper supporting material to enable the short-term self-formation of the DM when possible can thus be a key issue for boosting the feasibility of this technology.

Few studies to date have assessed the feasibility of DMs for the DMF strategy and further studies are required to prove their potential. Real MWW instead of synthetic solutions need to be studied to consider more realistic interactions between the influent particulate material and the supporting structure. Larger membrane areas than those used in laboratory-scale studies also need to be tested to consider possible hydrodynamic issues that could hinder DM formation during filtration of difficult sludge recovery in full-scale applications. The aim of this work was thus to evaluate the feasibility of a DM pilot plant (2 m^2 filtration membrane area) for directly filtering the real influent of a full-scale WWTP. The influence of different alternative supporting materials was studied (one or two layers of 1 or 5 µm pore size flat woven polyamide open monofilament meshes) to assess the effect on the self-forming DM capacity, filtration performance, and permeate quality. The proposed alternative potential was evaluated by performing a preliminary energy, economic, and carbon footprint balance.

9.2 Materials and Methods

9.2.1 Pilot plant

The DM pilot plant used in this study consisted of a 190 L working volume membrane tank equipped with two flat submerged membrane modules. Each membrane module consisted of a 1-m high x 0.5-m wide membrane frame that supported the DM supporting materials for filtration (average pore size and number of layers depending on the experimental period). The membrane frames allowed the attachment of two supporting materials (one on each module face), recovering the generated permeate in the interstitial space. The total filtration membrane area of the modules was 2 m^2. A large-pore woven steel mesh was added under each textile layer to stiffen the supporting material during filtration. The pilot plant was operated continuously at a permeate flux of 15 LMH, performing infinite filtration–relaxation cycles. Filtration lasted for 180 s, while 60 s were set for the relaxation stages, achieving a filtration-to-relaxation ratio of 3:1. Filtration was performed by vacuum using a screw pump (PCM, M Series, EcoMoineau™, Milano, Italy). The membrane module content was continuously mixed by a similar screw pump to ensure homogeneity. DM filtration was at a constant total suspended solids (TSS) concentration of about 2.1 g L^{-1} during all experimental periods. Membrane waste was evacuated continuously at a flow rate of 3.3 L h^{-1} to maintain TSS concentration, giving a membrane tank solids retention time (SRT) of 2.4 days. The raw influent MWW was pre-treated with a 0.5 mm screen size roto filter (PAM 270/500, Procesos Auto-Mecanizados, Alicante, Spain) and stored in 745 L working volume equalization tank (1.4 h hydraulic retention time (HRT)) for continuous feeding of the membrane module. A screw pump (PCM, M series, EcoMoineau™, Milano, Italy) was used to feed the DM membrane module according to filtration requirements. Fig. 9.1 shows a schematic diagram of the pilot plant. Further information on this system can be found in Sanchis-Perucho *et al.* [9.15].



RF:	Roto-filter	SP:	Screw pump	SIT:	Solids sensor
ET:	Equalization tank	SP-M:	Mixing pump	LIT:	Level sensor
MT:	Membrane tank	V:	Automatic valve	FIT:	Flow-rate sensor
PT:	Permeate tank			PIT:	Pressure sensor
				TUR:	Turbidity sensor
				pH-T:	pH-temperature sensor

Figure 9.1. Schematic diagram of the DM pilot plant.

9.2.2 Influent and experimental plan

Raw MWW (after classic pre-treatment by screening and sieving followed by desanding and degreasing) from the full-scale *Conca del Carraixet* WWTP (Alboraya, Spain) was used as pilot plant influent. The main characteristics of this MWW can be found in Table 9.1. Four textile-mesh-based alternatives (combinations of two pore sizes in simple or double layers) were evaluated as possible supporting materials for self-forming the DM (see Table 9.2). Flat open monofilament woven polyamide meshes (NITEX®, SEFAR) were used in all cases. Physical cleaning was performed by brushing the membrane surface with tap water as required.

Table 9.1. Influent characteristics

Parameter	Units	Mean ± SD
TSS	mg TSS L^{-1}	321 ± 98
COD	mg COD L^{-1}	512 ± 118
SCOD	mg COD L^{-1}	63 ± 28
TN	mg N L^{-1}	56.7 ± 10.8
TP	mg P L^{-1}	6.4 ± 1.6
Alk	mg $CaCO_3$ L^{-1}	342 ± 73
pH	-	7.4 ± 0.7
Turbidity	NTU	399 ± 124

Table 9.2. Supporting material characteristics and average DM permeability losses obtained during each experimental period

Exp.	Supporting material employed		Fouling growth rate	
	Layers	Average pore size (μm)	Slope (LMH bar^{-1} d^{-1})	R^2
1	2	1	10.03	0.789
2	1	1	9.85	0.888
3	2	5	9.24	0.955
4	1	5	5.21	0.877

9.2.3 Analytical methods and calculations

The pilot plant influent-generated permeate and waste were sampled twice a week. TSS, chemical oxygen demand (COD), total nitrogen (TN), and total phosphorous (TP) were determined according to standard methods [9.16]. A laser granularity distribution analyzer with a detector ranging from 0.01 to 1000 µm (Mastersizer 2000, Malvern, UK) was used to evaluate the particle size distribution of the fresh influent fed to the membrane tank. DM performance was evaluated based on its permeability evolution. 20 °C-standardized permeability (K_{20}) was calculated according to the following expression, which can be deduced from [9.15]:

$$K_{20} = \frac{J_T \, e^{-0.0239\,(T-20)}}{TMP_{ave}} \qquad 9.1$$

Where J_T represents the recorded permeate flux, T is the temperature and TMP_{ave} is the average transmembrane pressure recorded during each filtration cycle.

A preliminary evaluation was made of process energy, carbon footprint, and economic costs. Since this alternative scheme focused on upgrading current WWTPs, general water and sludge treatment equipment was not contemplated. For the energy balance, the energy demands of the permeate pumping and mixing and the potential energy recovery achieved from the organic matter captured by the DM were considered. Accordingly, the carbon footprint analysis only considered the energy required by the process. The economic balance considered both capital expenses (CAPEX) and operating expenses (OPEX). For the CAPEX, only the cost of the supporting material acquisition was considered, while the OPEX included the energy demands of the process and supporting material replacements. An average treatment volumetric flow rate of 36,625 $m^3\ d^{-1}$ was considered for all the calculations. This flow rate coincides with that of the WWTP where the pilot plant operated.

Equipment energy demands were calculated according to their appropriate theoretical equations (Eq. 9.2 and Eq. 9.3) [9.17]:

$$P_P = \frac{Q_P \, TMP_{ave}}{\eta_P} \qquad 9.2$$

Where P_P is the filtration permeate pump power requirements (W), Q_P is the pump volumetric flow rate ($m^3\ s^{-1}$), TMP_{ave} is the average transmembrane pressure during filtration (Pa), and η_P is the pump efficiency, the value of which was set in 0.65 in this study.

$$P_M = Q_M\, \rho\, g\, \frac{\left\{\left[\left(\frac{(L+L_{eq})\, f\, v^2}{D\, 2\, g}\right)_A + \left(\frac{(L+L_{eq})\, f\, v^2}{D\, 2\, g}\right)_I\right] + [z_1 - z_2]\right\}}{\eta_P} \qquad 9.3$$

Where P_M is the mixing pump energy requirements (W), Q_M is the mixing volumetric flow rate ($m^3\ s^{-1}$), ρ is the mixed liquor density ($Kg\ m^{-3}$), g is the acceleration of gravity ($m\ s^{-2}$), L and L_{eq} are the pipe length and equivalent pipe length (m), respectively, v is the liquor velocity ($m\ s^{-1}$), f is the friction factor, D is the pipe diameter, and (z_1-z_2) is the height difference (m). On the other hand, the following expression was used to estimate the energy production when transforming the recovered organic matter into biogas [9.15]:

$$E_R = COD_R\, Y^{CH4}\, CV_{CH4}\, \eta_{CHP} \qquad 9.4$$

Where E_R is the energy recovery ($kWh\ m^{-3}$), COD_R is the recovered COD concentration in the membrane module ($kg\ m^{-3}$), Y^{CH4} is the theoretical anaerobic methane yield of MWW sludge (0.35 m^3 of methane per kg of COD), CV_{CH4} is the methane calorific power (9.13 kWh per m^3 of methane), and η_{CHP} is the CHP system methane electricity generation efficiency. A η_{CHP} of 35% was used considering the different CHP technologies currently available [9.18].

For the carbon footprint calculations, a global warming potential (GWP) of 0.36 kg CO_2-eq per kWh of consumed energy was considered, in accordance with the energy mix GHG emissions ratio expressed in EcoInvent database [9.19]. Concerning energy and process costs, €0.20 per kWh was estimated for the electricity cost according to current Spanish high-voltage electricity rates [9.20], while the supporting material acquisition cost was estimated at €0.7 per m^2 of membrane area, according to Millanar-Marfa *et al.* [9.21]. Although some studies assume that no supporting material replacements will be required [9.21], we estimated a supporting material lifespan of 10 years.

9.3 Results and Discussions

9.3.1 DM self-forming capacity and filtration performance

Fig. 9.2a shows the permeability evolution of the self-formed DM during each experimental period. A slightly shorter self-forming time was achieved as the supporting material pore size was reduced. Similarly, the use of additional supporting material layers also entailed shorter DM self-forming periods. In both cases, the reduced DM self-forming time was due to the higher solids retention capacity achieved in the first days of filtration when reducing the supporting material pore size or adding additional layers (see Fig. 9.2b). Slightly lower permeability of the virgin supporting textile woven mesh was found as pore size was reduced and the layers were raised, indicating that the supporting material presented higher filtration resistance, which helped to retain the influent particulate pollutants (see Fig. 9.2a). Since more particles were retained in the supporting mesh, more material was used to create a preliminary cake layer on the woven textile mesh and promote the formation of a stable DM. Unfortunately, although lower self-forming times were obtained when increasing the supporting material filtration resistance, the enhanced solids retention capacity also entailed a sharp reduction of DM permeability as filtration advanced (see Fig. 9.2a). Permeability was reduced by 90% after 14, 24, 32, and 50 days in Experiments 1 to 4. However, the TSS concentration captured during filtration reached a pseudo-steady state after a stable DM was formed (ranging from several hours to 3 days, depending on the experimental period; see Fig. 9.2b), and did not increase despite the lower DM permeability. According to these results, the drop in DM permeability was related to the quicker increase of DM thickness when using supporting materials with a higher filtration resistance, and relevant short-term alterations of the DM structure are not expected. Since no relevant reduction of the self-forming time was achieved as the supporting material filtration resistance was increased, the use of a 5 µm pore size single layer was considered the most suitable material to extend the filtration lifespan.

The DM formed in Exp. 4 was cleaned by brushing the surface with tap water when the permeability dropped under 50 LMH bar^{-1}. This reduced the DM thickness, recovering a great part of the original supporting material permeability without compromising the DM solids capture capacity (see Fig. 9.2). The filtration lifespan was then extended for about a further 50 days, although at significantly lower permeability (average permeability of about 55 LMH bar^{-1} was obtained after the physical cleaning, in contrast with the 141 LMH bar^{-1} achieved in the first half of Exp. 4). The lower permeability achieved after the physical cleaning may have been due to deficient cleaning not having removed enough DM thickness or, as other studies have found [9.22], it could have been produced by internal blockage of some of the supporting material pores, which cannot be efficiently removed by physical cleaning. In any case, since low-energy physical cleaning methods were relatively effective in controlling DM thickness, applying a proper cleaning schedule would be interesting to enhance process feasibility without compromising energy demands.

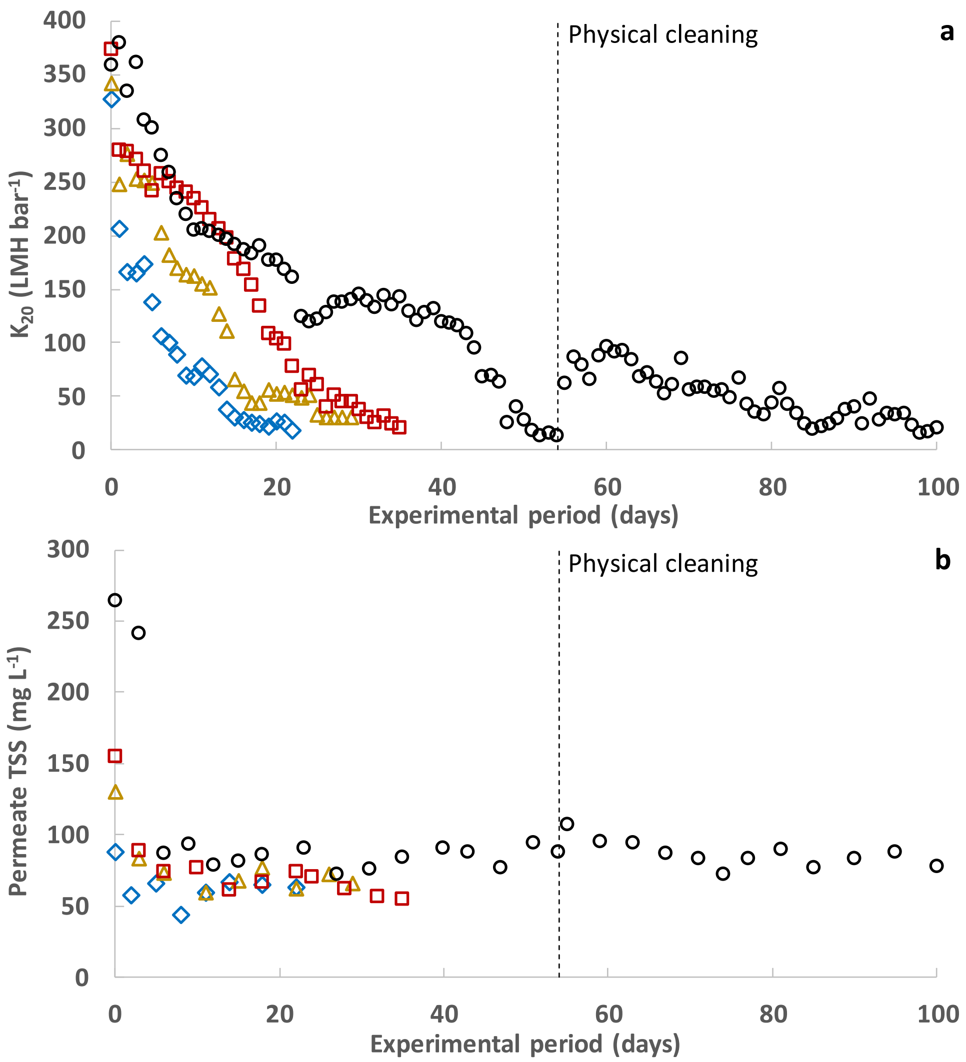


Figure 9.2. DM performance regarding the employed supporting material: (a) permeability evolution and (b) TSS concentration in the permeate. ◇ Two layers with 1 µm of pore size, △ one layer with 1 µm of pore size, □ two layers with 5 µm of pore size, ○ one layer with 5 µm of pore size.

Important differences in performance were achieved between the results obtained in this study using raw MWW as influent and previous studies that used primary settler effluent [9.15]. Much shorter DM self-forming periods were obtained with raw MWW, allowing the larger supporting material pore size. Only a few hours were required to self-form a stable DM with raw MWW in contrast to the 17 days required with primary settler effluent as membrane tank influent (two layers of a woven polyamide mesh with 1 µm pore size in both studies). This significant difference was due to both the higher content of particulate material in the treated influent when using raw MWW and the larger average size of the influent particles. Indeed, the particles between 100 and 1000 µm increased significantly when raw MWW was used (see Fig. 9.3), which favoured the development of a DM by promoting the formation of a preliminary cake layer on the supporting material. However, as the self-forming time was reduced, the filtration fouling growth rate increased, achieving permeability losses in the DM of between 10.03 and 5.21 LMH bar^{-1} day^{-1} (see Table 9.2), depending on the supporting material used in this study with the 2.27 LMH bar^{-1} day^{-1} reached when filtering primary settler effluent [9.15]. Then, although using raw MWW could be considered as a more suitable influent for applying DMs when filtering MWW, more energy may be required to control the DM thickness. Consequently, further studies focused on the overall process energy requirements and resource savings are required to properly determine the best influent to use (filtration energy demands according to DM permeability, applied fouling control strategies energy requirements, percentage of organic matter and nutrients captured from different influents, etc.).

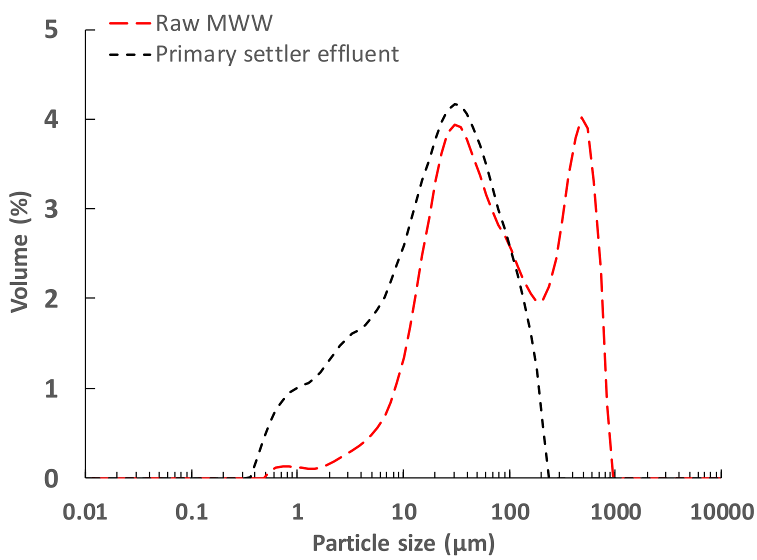


Figure 9.3. Particle size distribution of the raw MWW used in this study and the primary settler effluent filtered in Sanchis-Perucho *et al.* [9.17].

9.3.2 Permeate quality

Table 9.3 shows the average DM permeate quality and resource recovery during each experimental period after the DM was formed. Higher pollutant retention was obtained as the supporting material pore size was reduced and more layers were added, thus increasing the quality of the permeate. However, the benefits in permeate quality after achieving a stable DM were negligible, with only a 6% difference between the most efficient solids-capturing supporting material (two layers with 1 µm pore size) and the less efficient one (one layer with 5 µm pore size). Since there was no great difference in permeate quality, it can be concluded that no significant changes in the surface or internal DM structure can be expected in the supporting textile materials used in this study, the slightly higher retentions being due to the thicker DM formed when increasing the supporting material solids retention capacity. Similar permeate qualities have also been reported by other authors when filtering raw MWW with DMs, despite employing larger supporting material pore sizes (between 1 and 100 µm) or adding an extra suspended material component (diatomite) to boost DM formation [9.12; 9.23]. In fact, the permeate produced after obtaining a stable DM seems to be relatively consistent when filtering untreated MWW, achieving similar qualities when filtering the primary settler effluent of an WWTP [9.15]. Since the supporting material or influent used does not seem to significantly influence the permeate quality, the selection of the most suitable configuration should focus on reducing the filtration energy demands.

Table 9.3. DM permeate quality and resources captured

Exp.	TSS		Turbidity		COD		TN		TP	
	(mg L^{-1})	(%)*	(NTU)	(%)*	(mg L^{-1})	(%)*	(mg L^{-1})	(%)*	(mg L^{-1})	(%)*
1	67	21	167	55	155	30	48.7	86	4.7	73
2	73	23	157	53	159	31	50.1	88	5.0	78
3	70	22	161	55	167	33	49.4	87	4.8	75
4	88	27	174	59	186	36	50.4	89	4.9	77
PS	132	41	-	-	218	43	-	-	-	-

*Percentage of the influent pollutant remaining in the permeate. PS: solids and organic matter captured by the primary settler of the WWTP.

Comparing the DM permeate quality to the primary settling effluent from the *Conca del Carraixet* WWTP, a significantly higher particulate material fraction was captured in the former than in the latter (73% in the DM compared to the 59% captured by primary settling; see Table 9.3), showing its potential as primary treatment. A higher COD content was therefore also recovered in the DM unit (64%) than in the primary settling step (57%). The lower differences achieved for COD were due to the significant concentration of COD in the influent soluble fraction, which none of the compared technologies could capture. If the soluble COD in the influent wastewater were to be negligible, the difference in COD capture between the DM and the primary settler would be higher (and similar to the existing difference in TSS). On the other hand, poor permeate qualities were obtained when comparing the results of this work with other membrane technologies used for DMF (*i.e.* MF and UF membranes). This could be expected, since the lower pore size of MF and UF membranes (from about 1 to 0.01 µm) can capture almost all influent particulate (and colloidal) material, achieving permeates without solids and with COD, TN, and TP concentrations of about 44–88, 46.1–48.2, and 6.44–6.45 mg L^{-1}, respectively [9.24]. In this context, DMs cannot compete in COD recovery, although relatively similar TN and TP captures can be achieved, since the main input of these pollutants comes in the form of soluble compounds (see Table 9.3). In any case, since MF and UF membranes involve significantly higher costs than DMs (about €35 per m^2 of MF and UF membranes compared to the €0.7 per m^2 of DMs) [9.21; 9.25], besides higher operating costs and fouling propensities [9.12], they require substantially, and possibly prohibitive, investment costs when just aiming to upgrade an existing facility. DMs can thus be an interesting alternative when targeting improving the amount of resources recovered during classic MWW treatment at low costs.

According to the results obtained, the permeate quality generated by the DM is far from meeting the European standards regarding direct reuse or discharge into water bodies, especially due to its significant nutrient content. Since a considerable fraction of the influent COD was recovered thanks to the DM treatment, the effluent produced could be treated by a CAS process, which would require less energy thanks to its reduced aeration demands. This CAS process could focus on influent nitrification/denitrification treatment while using the remnant COD, removing the phosphorous concentration by biological capture when possible, or using conventional chemicals for its precipitation. An aerobic bacterial and microalgae consortium to remove the remnant COD while capturing nutrients could be another possible alternative [9.26]. Since the bacterial oxygen demands would be covered by the microalgae activity, this alternative could be proposed as a low-energy treatment. Thus, although significantly worse permeate qualities can be expected when replacing MF or UF membranes by DM, a sizeable fraction of the influent COD would still be recovered via AD, alongside the ability to capture the influent nutrients by secondary alternatives.

9.3.3 Process feasibility

To assess the feasibility of the proposed alternative, three points of view were considered: energy, economy, and the carbon footprint. Fig. 9.4 shows the filtration energy demands together with the energy that could be potentially recovered after transforming the captured organic matter into methane via AD. Since only permeate pumping and mixing would be required when using DMs, significant energy recoveries are achieved by this process, which rise to about 0.33 kWh per m^3 of influent MWW. Indeed, the fouling control strategies required in other membrane systems (membrane bioreactors) are the largest energy consumers [9.27]. Unfortunately, an insignificant enhancement on the energy recovery was achieved compared with a classic WWTP primary settler (see Fig. 9.4), which does not justify the use of DMs. This alternative also showed relevant carbon footprint reductions (about 0.13 kgCO_2-eq per m^3 of influent MWW), thanks to the organic matter captured by the DM, although again with a negligible difference over a conventional primary settler. Positive financial results were obtained for the proposed alternative, thanks to the significant amount of energy recovered, which far exceeded the replacement costs of the supporting material (see Fig. 9.5), while the initial investment was significantly reduced due to the relatively lower cost of supporting materials than those required by other membrane systems. In fact, short payback periods can be expected; we achieved 0.08 years in the present study, with a profit of about €0.065 per m^3 of influent MWW. However, as mentioned above, this profit is not meaningful enough when considering the amount of organic matter captured by a WWTP primary settler, which obtains similar outcomes. Additionally, physical low-energy fouling control strategies need to be studied to improve DM permeability during continuous filtration. The environmental and economic impact of the materials and other auxiliary resources required by this alternative (*e.g.* membrane tanks, maintenance demands, equipment replacements, etc.) should also be considered to properly study its feasibility. The small profit made by the organic matter capture of DMs is therefore not large enough to justify its implementation when treating average MWWs, as this alternative scheme requires a higher resource recovery potential to be competitive. The alternative was found to be unsuitable for treating low/middle-pollutant-load MWWs and further studies considering high-load MWWs or similar influents are required.

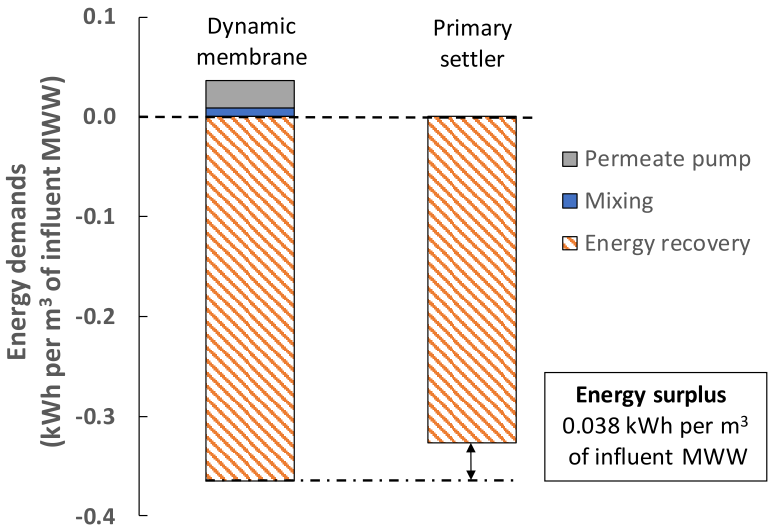


Figure 9.4. Energy recovery potential of the direct raw MWW filtration by DMs.

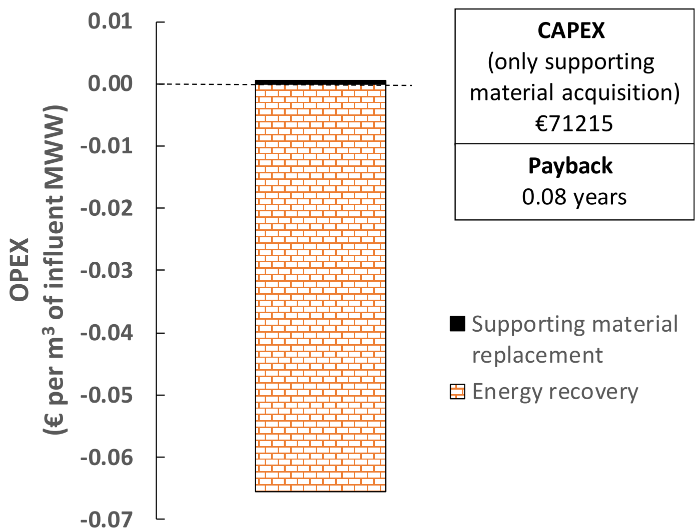


Figure 9.5. Preliminary economic potential of the direct filtration of raw MWW by DMs. CAPEX: capital expenses. OPEX: operating expenses.

To study the potential benefits of DMs over conventional treatment schemes with heavily loaded wastewaters, the energy, carbon footprint, and financial profit of both DM filtration and primary settling were calculated for different increased COD concentrations (from 500 to 1500 mg L^{-1}). The same particulate fraction captures as those obtained in this study were considered for both systems in these simulations. The surplus obtained by DM is shown in Fig. 9.6. Larger profits than those obtained in this study can be achieved when considering a heavily-loaded MWW (COD about 1000 mg L^{-1} [9.28]), with an energy surplus of 0.08 kWh per m^3 of influent MWW with DMs instead of primary settling (see Fig. 9.6). Better carbon footprint reductions can also be obtained in this scenario, achieving a surplus reduction of 0.017 kgCO_2-eq per m^3 of influent MWW. It is important to highlight that these potential advantages only consider the direct benefits of higher DM COD capture. Thanks to the greater particulate COD recovered by DMs, low-loaded effluents could be treated by CAS or other secondary treatments which would require of lower energy inputs thanks to the reduced aeration necessities. Considering 1000 mg L^{-1} of COD in the influent, a direct financial surplus of €0.009 per m^3 of influent MWW would be obtained by substituting the primary settler with a DM system, also reducing the amount of COD to treat in the secondary treatment by 71 mg L^{-1} (see Fig. 9.6). This should be considered in the energy, carbon footprint, and economic balance as indirect profits. This treatment scheme could therefore be attractive for heavily loaded MWWs or industrial wastewaters with a high percentage of non-settable organic particulate material.

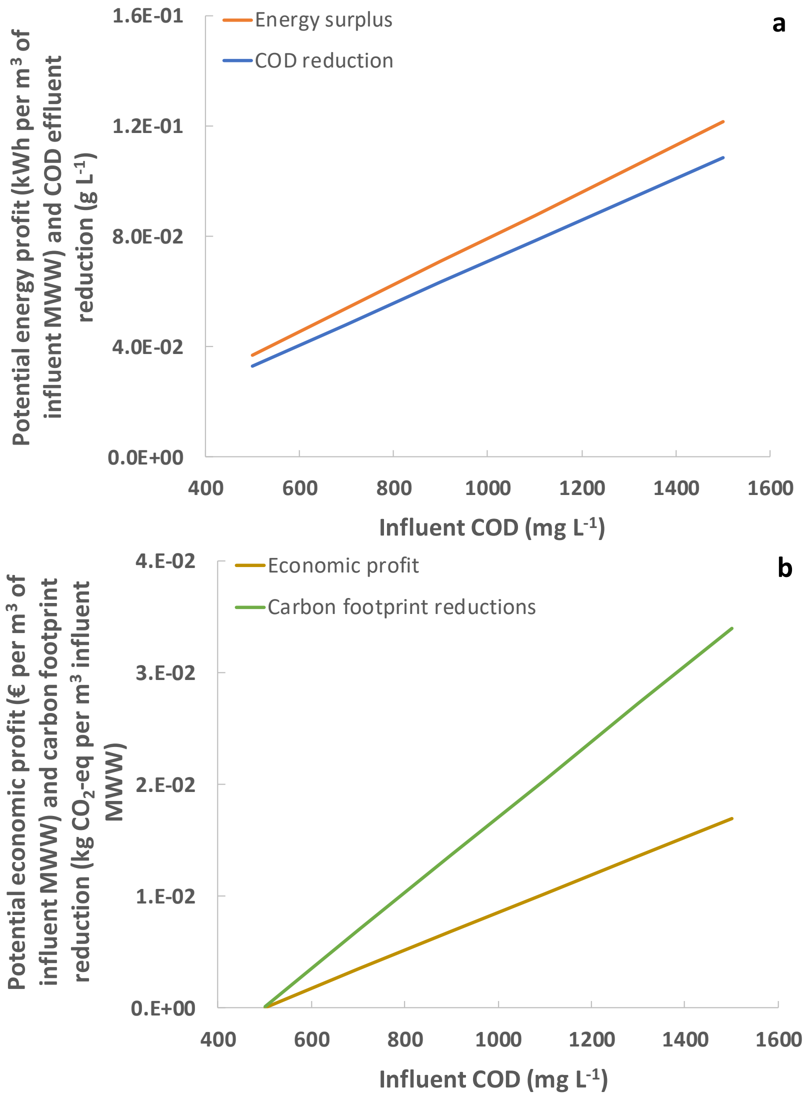


Figure 9.6. DM potential benefits compared to primary settling as influent COD increases: (a) energy surplus and effluent COD reductions, and (b) carbon footprint reductions and economic profit.

Finally, as other authors have proposed [9.12; 9.15], using coagulants to increase the average influent particulate fraction size would be an interesting strategy to increase the potential DM resource recovery even further. In a previous study using the primary settler effluent as the base [9.15], it was determined that relatively low coagulant dosing (10 mg L^{-1}) significantly increased the COD captured by the DM (effluent COD reduction from 141 to 58 mg L^{-1}) and recovered a relevant fraction of the soluble COD fraction along with all the influent phosphate. Further studies are therefore required to evaluate the advantages of dosing coagulants during raw MWW filtration when using DMs.

9.4 Conclusions

This study assessed the feasibility of treating MWW with DMs. A stable short-term self-forming DM was achieved, regardless of the supporting material used (from several hours to 3 days) thanks to the significant concentration of particulate material and the large particle size present in the raw MWW. Relatively similar permeate qualities were obtained for all the supporting materials tested, although higher permeability losses (from 5.21 to 10.03 LMH bar^{-1} day^{-1}) were found as the supporting material filtration resistance increased due to the increasing DM thickness. A single-layer supporting material of 5 µm or larger pore size can be recommended to minimize DM thickness growth rate. Unfortunately, our preliminary energy, carbon footprint, and economic evaluations showed that, although DMs capture more pollutants than conventional treatments (primary settler), the benefits are not enough to justify their use with average municipal wastewater. However, this alternative scheme could be suitable for treating higher-loaded MWW with a higher fraction of organic matter in the non-settable solids.

9.5 Acknowledgments

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CHAPTER 10. Overall Discussion

This PhD work focused on the study of direct membrane filtration (DMF) technology as a potential alternative to boost resource recovery from municipal wastewater (MWW). Three membrane technologies (microfiltration, ultrafiltration and dynamic membranes) were evaluated, studying their feasibility and potential benefits for up-grading current MWW treatment facilities. Different operating conditions (influent characteristics, operating solids concentration, transmembrane flux, and dynamic membrane supporting material pore size depending on the operated membrane) were evaluated aiming to minimize membrane fouling while boosting resource recovery.

Besides conducting different batch experiments at lab-scale, a membrane-based pilot plant equipped with commercial membrane modules treating MWW from a full-scale MWW facility was operated for more than 3 years. Therefore, this work provides of realistic and useful information on the performance of DMF for MWW treatment, as well as delivering guidelines for full-scale implementation of the evaluated technology. The main results obtained during this work are organized in the following sub-points each one representing a chapter of this thesis.

10.1 Assessing the Most Suitable Methodology to Determine Sludge Filterability from Different Municipal Wastewater Treatment systems

The capability of different methodologies to determine sludge filterability and predict the resistance to filtration of different sludge sources in membrane filtration systems was evaluated. Three filterability methods were tested: capillary suction time (CST), time to filter (TTF), and specific resistance to filtration (SRF). These methods were evaluated using three different sludge sources: aerobic activated sludge, supernatant from a primary settler further concentrated by ultrafiltration membranes (PSE), and digestate from the anaerobic co-digestion of microalgae and primary sludge. The capability of CST, TTF and SRF to estimate total suspended solids (TSS) and soluble microbial products (SMP) concentrations was also assessed, while validating the results obtained with the real filtration process of the operated membrane-based systems.

10.1.1 Filterability results

CST method was identified as a suitable alternative for determining sludge filterability regardless the sludge source, within the operating conditions evaluated. However, TTF method was determined as a solid alternative when treating PSE and anaerobic sludge, achieving correlations similar and even superior in some cases than those obtained by CST method. On the other hand, SRF method generally resulted in the poorest correlations within the methods evaluated, especially displaying bad results regarding SMP content. Consequently, the SRF method would be not recommended for monitoring sludge filterability, within the operating conditions evaluated.

10.1.2 Prediction of filtration resistance in different membrane systems

CST and TTF methods were determined as suitable methodologies to predict sludge filtration resistance and estimate TSS and SMP content in two membrane-based systems: DMF treating PSE sludge, and AnMBR treating microalgae and primary sludge. TTF method presented better results when treating PSE sludge, whilst CST method was the only option when treating anaerobic sludge since TTF method resulted in poor correlations. Based on the results obtained, CST was identified as the more convenient method to employ when treating biological sludge. On the other hand, when filtering sludge with low/negligible biological activity (*e.g.* PSE sludge), TTF method was considered as the most attractive alternative since it showed better correlations regarding the medium filterability and sludge filtration resistance.

10.2 Direct membrane filtration of municipal wastewater: studying the most suitable conditions for minimizing fouling rate in commercial porous membranes at demonstration scale

Two membrane technologies were compared in order to determine their suitability for DMF of MWW: ultrafiltration (UF) and microfiltration (MF). The effect of treated influent (raw MWW and PSE) and operating total suspended solids (TSS) concentration (about 1 and 2.6 g L^{-1}) on process performance was evaluated to determine suitable operating conditions resulting in low fouling propensities.

The effectiveness of two physical strategies for fouling control (*i.e.* continuous air sparging and periodic backwashing) was studied during continuous filtration, evaluating the reversibility of fouling under each experimental condition tested. On the other hand, membrane permeability recovery through chemical cleaning was studied to determine the main source of the produced fouling (organic or inorganic matter). Moreover, main fouling mechanisms were studied by correlating experimental fouling data with different theoretical mathematical fouling models.

10.2.1 Effect of membrane pore size

Completely different performances were found regarding the membrane technology employed. It was only possible to operate the MF membrane for extremely short filtration periods (about 2 – 8 hours), while the UF membrane allowed middle-term operation (from 34 to 69 days). The lower fouling propensity when using UF compared to MF membranes was attributed to the reduction of the average membrane pore size which was able to prevent pollutants (especially colloidal particles) to block the membrane pores and/or narrow them by their inside deposition.

10.2.2 Effect of the influent used

Significant lower fouling growth rates were obtained when raw MWW was filtered instead of PSE, regardless the membrane used and the operating TSS concentration. These results suggested that fouling development was indeed strongly affected by the interactions between particle size and membrane pores size. Consequently, higher average particles sizes (as per the case of raw MWW compared to PSE) may result in a reduction in fouling propensity by decreasing the amount of particles able to block or to be deposited inside the membrane pores.

10.2.3 Effect of operating solids concentration

The effect of modifying the operating TSS concentration on filtration performance varied depending on the microporous membrane used (*i.e.* MF or UF). A strong beneficial effect was observed when increasing TSS concentration in UF, reducing the fouling propensity. In this case, since reductions in fouling propensity were recorded regardless of the treated influent, it was concluded that a more robust and thick cake layer is formed when increasing the TSS concentration in the membrane tank, which may protect the membrane to be fouled from diverse pollutants with high fouling potential, such as colloids and SMP substances. Hence, increasing operating TSS concentration could be considered as an interesting operating strategy to improve filtration feasibility when directly filtering MWW. However, contrary results were obtained when operating the MF membrane since even higher fouling degrees were observed when increasing the TSS concentration in the membrane tank. In this case, due to the higher pore size of MF membranes, this strategy may not be so convenient due to the increment in the number of particles able to block or to deposit in the membrane pores. Therefore, particle size distribution was identified as a key aspect to take into account when directly filtering MWW.

10.2.4 Fouling control strategies effectiveness

Air sparging for membrane scouring and backwashing proved to be ineffective to control fouling when operating the UF membrane, although they did have a significant impact on the performance of the MF membrane, extending the operating period from some hours (2 – 8 hours) to some days (5 – 6 days). These results indicated that the main fouling source when operating the UF membrane was related to irreversible fouling. It could be assumed then that the operating conditions stablished during continuous filtration (0.1 m^3 m^{-2} h^{-1} of specific air demand and 2 min of backwashing every 10 filtration:relaxation cycles) were enough to control the thickness of the developed cake layer. Conversely, membrane fouling developed during MF was mainly identified as reversible fouling, which can be minimized by using physical cleaning strategies. Nonetheless, despite the significant enhancement of MF membrane filtration performance when air sparging for membrane scouring and backwashing were employed, MF process efficiency was intensely lower to that achieved in UF, thus UF membranes were identified as the most convenient option when directly filtering MWW, within the operating conditions, and membrane technologies and configurations evaluated.

10.2.5 Fouling characterization

UF membrane chemical cleaning by NaOCl (*i.e.* basic cleaning) resulted in a membrane permeability recovery of about 70 – 85%, while increasing up to 92 – 99% in the case of MF. Therefore, organic matter was identified as the main fouling promotor during filtration for both membranes, showing a special relevance for MF since it was operated during relatively short time periods (around 5 – 7 days). Nonetheless, inorganic fouling (mainly related to inorganic compound precipitation on the membrane) also had a significant effect on UF membrane filtration in the middle-term operating period (around 1 – 2 months). Thus, although fouling related to organic matter should be the main issue be controlled (*e.g.* applying chemical-enhanced backwashes with basic solutions), inorganic fouling must also be taken into account to prolong high membrane permeability in the long-term. Regarding fouling mechanisms, the theoretical models applied to simulated the experimental data indicated that cake layer formation is the predominant fouling mechanism in the short-term (during each filtration cycle). However, as filtration advances, severe irreversible fouling appears, being related to intermediate/complete pore blocking in MF and standard/complete pore blocking in UF.

10.3 Evaluating resource recovery potential and process feasibility of direct membrane ultrafiltration of municipal wastewater at demonstration scale

Since UF technology was found to be much more suitable than MF for direct filtration of municipal wastewater, further research was performed focused on the former. UF performance at higher solids concentrations (about 6 and 11 g L^{-1}) was evaluated for both raw MWW and PSE treatment. Additionally, the resource recovery potential of DMF of MWW using UF membranes was assessed for the experimental conditions evaluated, conducting preliminary energy, economic, and carbon footprint analysis.

10.3.1 Filtration performance

Operating at TSS concentrations above 2.6 g L^{-1} was found to sharply reduce membrane fouling. Minimum fouling growth rates of about 0.55 mbar per day were achieved when operating above 6 g L^{-1}, which allowed to conduct the filtration process for over 120 days. Filtering raw MWW instead of PSE also showed slight fouling growth rate reductions, although this effect was only observed when operating at low TSS concentrations (below 2.6 g L^{-1}). In this case, the beneficial effects of treating raw MWW instead of PSE in terms of fouling control were associated with the reduction in the number of small size particles in the treatment bulk. Thus, increasing the TSS concentration in the membrane tank may result in a quick generation of a thick protecting cake layer, reducing fouling potential due to interactions between the membrane surface and soluble and colloids compounds. These findings validated former results regarding the beneficial effects of TSS concentration on irreversible fouling reduction, reassuring the importance of this operating strategy in the long-term DMF fouling control. Finally, previous results regarding the impact of organic matter on irreversible fouling were also corroborated in the long-term operation (about 120 days), achieving permeability recoveries about 80 – 85% after basic chemical cleaning regardless the operating conditions evaluated or filtration period length (between 35 – 123 days).

10.3.2 Resource recovery and permeate quality

DMF by UF technology was able to properly recover a high percentage of influent resources, capturing about 80 – 85 and 20 – 40% of influent COD and nutrients, respectively. Additionally, biological degradation of influent compounds remained negligible when the membrane tank was operated at sludge retention time (SRT) below 3 days, despite operating at constant levels of air scouring (0.1 m^3 m^{-2} h^{-1}). However, a significant amount of influent COD was biologically degraded when raising the SRT to 6 days. High-quality permeate was produced regardless of the operating conditions, indicating that the employed membrane pore size is the main responsible of resource capture (*i.e.* cake layer plays an irrelevant role regardless of its thickness). Unfortunately, although COD and BOD concentrations met European discharge standards with effluent concentrations between 47 – 64 and 18 – 21 mgCOD L^{-1}, respectively, nitrogen and phosphorus effluent concentrations did not meet the legal limits (ammonium and phosphate concentrations in the permeate ranged between 27 – 45 mgNH_4^+ L^{-1} and 3.2 – 4.8 mgPO_4^{3-} L^{-1}, respectively). Consequently, this permeate could only be safety discharged in non-sensitive environments, otherwise a tertiary treatment to recover soluble nutrients would be necessary.

10.3.3 Process feasibility

Promising results were obtained from energy and carbon footprint estimations of DMF of MWW using UF technology. Indeed, energy recoveries about 0.46 – 0.40 kWh per m^3 of influent MWW can be obtained when considering process energy demands and potential energy recovery from methane production via anaerobic digestion. Similarly, since energy surplus can be obtained, the system acted as a CO_2 sink in terms of operation phase (*i.e.* construction and demolition phases were not evaluated). Specifically, the operating phase resulted in 0.19 – 0.16 kg of CO_2-eq avoided per m^3 of influent MWW

From an economic perspective, it was determined that operating above 10 LMH is not recommended due to the severe fouling development. In this respect, potential savings in capital expenses from reducing membrane area requirements (*i.e.* increasing the design transmembrane flux) would be hindered by a dramatic increase in chemical cleaning requirements, sharply increasing operating costs and reducing membrane lifespan. Consequently, low/moderate operating fluxes (around 10 LMH) are recommended for DMF of MWW by UF technology, achieving profits of about €0.035 per m^3 of treated MWW when operating at a flux of 10 LMH, which entail a membranes payback period of 12.3 years.

10.4 Building a simple and generic filtration model to predict membrane fouling in the long-term when treating municipal wastewater

A simple and generic model was proposed to predict membrane fouling in DFM of MWW using UF membranes. A resistance-in-series structure was proposed, simplifying fouling as the consequence of two different sources: cake layer (from suspended material) and pore blocking (from soluble and colloidal compounds).

10.4.1 Model performance

Proper fouling predictions were achieved at different operating TSS concentrations (about 1, 2.6, 6 and 11 g L^{-1}), resulting in root mean square error (RMSE) values of around 5 – 25 mbar between experimental data and model predictions. The model was also able to match the experimental results from two different influent sources (raw MWW and PSE) by just recalibrating 3 out of 7 model parameters.

10.4.2 Sensibility and uncertainty analysis

A global sensibility analysis based on Morris screening method was performed varying the values of the 7 parameters included in the model. 4 parameters (δ_C, δ_B, k_I and α') were identified as influential factors in model outputs. On the other hand, the uncertainty analysis showed that high errors can be expected for long-term simulations since the estimated membrane fouling strongly depends on former fouling conditions. The presented model showed an elevated potential to generate reasonable membrane fouling predictions while maintaining a simplistic and open structure to allow its implementation together with other complementary materials.

10.5 Dynamic membranes for enhancing resources recovery from municipal wastewater

Since major limiting factors when applying porous membranes for DMF of MWW include the appearance of severe membrane fouling and expensive membrane acquisition and replacement costs, the use of dynamic membranes (DMs) was studied as a potential alternative. Therefore, filtration performance and permeate quality from a DM unit filtering PSE were evaluated. Different strategies were used to enhance the required self-forming time and influent resources capture (increasing the filtration flux, increasing the concentration of operating TSS, and dosing coagulant). Finally, process feasibility was evaluated by performing a preliminary energy and economic study of the proposed alternative.

10.5.1 Filtration performance

One layer of supporting material (a flat open monofilament woven polyamide mesh with 1 µm of average pore size) was found to be insufficient to self-form a DM in the short-term when treating PSE. This was due to both: i) the low solids load entering the system; and ii) the low average size of influent particles which manifested the sharp limitations of DMs for treating some influents despite the relatively low pore size of the employed supporting material. Nevertheless, a proper DM was able to self-form when using two supporting material layers (17 days of operation) or when increasing the operating TSS concentration (8, 6 and 4 days of operation for a TSS concentration of 1.9, 4.7 and 9.2 g L^{-1}), being both effective strategies to allow the treatment of PSE by DMs.

10.5.2 Strategies to enhance resource recovery

Poor influent resource captures (about 10 – 40% depending on the pollutant) were reached by the self-formed DM, achieving TSS, COD, total nitrogen (TN), total phosphorous

(TP), and turbidity values in the permeate of about 65 mg L^{-1}, 141 mg L^{-1}, 42.3 mg L^{-1}, 4.3 mg L^{-1}, and 86 NTU, respectively. This was due to the low percentage of suspended material in the treated influent which hindered the DM capture capacity due to i) the lower amount of resources susceptible to be captured (particulate fraction) and ii) reducing the thickness of the developed cake layer due to the lack of material. Furthermore, no significant improvements were observed neither increasing the permeate flux (from 15 to 45 LMH) nor raising the operating TSS concentration (range tested from 1.9 to 9.2 g L^{-1}). Consequently, it was concluded that non-significant changes on the formed DM occurred by these operational changes, being none of them effective strategies to enhance the DM resource capture capacity. Four commercial coagulant solutions were also tested to enhance DM performance, identifying the PHLA18 at a dosing rate of 10 mg L^{-1} as the optimum reagent dose. Thanks to the coagulant dosing, both the required DM self-forming time and the resource capture capacity were significantly enhanced, achieving in this case a self-forming period of 7 days with a permeate quality regarding TSS, COD, TN, TP, and turbidity of about 24 mg L^{-1}, 58 mg L^{-1}, 38.1 mg L^{-1}, 1.2 mg L^{-1}, and 22 NTU, respectively.

10.5.3 Process feasibility

Since the DM achieved poor resource captures, low energy recoveries of around 0.030 kWh per m^3 of influent MWW were achieved. As stated above, this was due to the low particulate material loading entering the system, reflecting the unsuitability of this technology to treat this type of influents. However, this issue was partially solved thanks to coagulant dosing, achieving increased energy recoveries of about 0.121 kWh per m^3 of influent MWW. Unfortunately, since a continuous reagent dosing would be necessary, the DM operating costs would ascend to €0.003 per m^3 of influent MWW.

Since the permeate produced may need a post-treatment focused on removing the soluble nitrogenous fraction to allow its discharge in sensitive environments, the applicability of the presented alternative would be extremely limited and only competitive against other membrane technologies (*e.g.* UF) when targeting to minimize the investment costs.

10.6 Evaluating the feasibility of employing dynamic membranes for the direct filtration of municipal wastewater

DM technology was further evaluated for DMF of MWW, studying filtration performance, resource recovery, and process feasibility when filtering raw MWW.

10.6.1 Filtration performance and resource recovery

Extremely short self-forming DM periods were required when treating raw MWW in comparison with PSE due to the higher influent solids load entering the former than the later. Indeed, a higher supporting material pore size was able to be used when treating raw MWW, evaluating the effect of four supporting material alternatives (one or two layers of flat open monofilament woven polyamide meshes with 1 or 5 μm of average pore size). The self-forming period ranged from several hours to 3 days, depending on the supporting material, identifying a permeability reduction from 5.2 to 10.0 LMH bar^{-1} day^{-1} as the supporting material capture capacity was increased. The increase observed in fouling propensity as the supporting material layers increased or its average pore size reduced was attributed to the quicker formation of ticker cake layers onto the supporting material due to its particles capture capacity increment. However, relatively similar permeate qualities were obtained regardless of the supporting material alternative employed: the thicker cake layers formed when using more restrictive supporting materials barely enhanced the resource recovery capture. A permeate quality regarding TSS, COD, TN, TP, and turbidity of 67 – 88 mg L^{-1}, 155 – 186 mg L^{-1}, 48.7 – 50.4 mg L^{-1}, 4.7 – 4.9 mg L^{-1}, and 167 – 174 NTU, respectively, was obtained, not finding significant differences regarding the permeate generated when treating PSE. In consequence, since the self-forming period was not an issue and using more restrictive supporting materials just increased the fouling growth rate, the use of a single-layer supporting material with 5 μm or larger average pore size was determined to be the most suitable design strategy for DMF of raw MWW, within the evaluated operating conditions.

10.6.2 Process feasibility

Energy and carbon footprint evaluations showed good results due to the low operating energy demands of DMs. Energy surplus of about 0.33 kWh per m^3 of treated MWW were achieved, resulting in carbon footprint savings of 0.13 kgCO_2-eq per m^3 of treated MWW, related to the operating phase. Unfortunately, these results only showed a slight improvement over currently in use technologies for MWW pre-treatment (primary settler). From an economic point of view, positive results were also obtained thanks to the low cost of the supporting materials and minimal maintenance requirements. A short payback period of 0.08 years was obtained within the experimental conditions tested, with a profit of about €0.065 per m^3 of treated MWW. However, the economic incomes obtained barely exceed that achieved by primary settling pre-treatment. Thus, since the permeate generated by this technology would need a secondary treatment to allow its proper discharge into water bodies, the results obtained do not justify the implementation of this alternative for the treatment of MWW, requiring higher loaded influents to be considered as a competitive option.

10.7 General conclusion and future perspectives

Considering all the results from this PhD work, it can be concluded that UF seems to be the most suitable membrane technology in DMF of MWW. MF technology showed significant membrane fouling propensities when operating under similar conditions. On the other hand, DMs were not able to outcompete UF regarding resource capture from MWW, despite having showed a competitive market niche when aiming to reduce investment and maintenance cost.

Regarding the operating conditions, low/middle permeate fluxes (around 10 LMH) are recommended within the operating conditions evaluated, since a severe fouling is obtained otherwise. In fact, the reduction on the investment costs that could be obtained by increasing the permeate flux do not offset the sharp increment in the operating costs due to chemical cleaning requirement and reductions in membrane lifespan. Operating at relatively high TSS concentrations (between 6 and 11 g L^{-1}) is also recommended to minimize membrane fouling whenever non-significant aerobic bacteria activity is found. SRTs values below 3 days would be recommended to avoid significant organic matter losses due to biological biodegradation.

Regarding influent source, raw MWW and PSE showed similar fouling propensities when operating at relatively high TSS concentrations in the membrane tank. Thus, their selection was concluded not to be a major issue in DMF of MWW, although raw MWW may be considered as a slightly better candidate since lower SRTs would be required in the membrane module when raising the operating TSS concentration. Since organic matter was found to be the major fouling promotor during filtration, the use of fouling control strategies focused on minimizing organics attachment onto the membrane are suggested, such as periodical chemical enhanced backwashing (CEB) with basic reagents.

Hence, the presented work shows promising results concerning the application of DMF technology equipped with UF membranes for MWW treatment. However, although promising energy and carbon footprint results were obtained, economic balance still needs to be further improved to achieve a robust competitor for MWW treatment.

Future perspectives would include:

- Conducting comparative studies of different membrane pore sizes (even nanofiltration and osmosis technologies) to corroborate previous conclusions and properly determine most suitable membrane technology for DMF of MWW.
- Performing long-term filtration trials to properly corroborate chemical cleaning demands. Furthermore, performing long-term filtrations trials at higher fluxes are also required to verify the fouling growth rates obtained in this study.
- Studying fouling mechanisms deeply. Further analysis is suggested to indeed assessing the nature of the developed fouling. These studies would include the determination of the following: the amount of mass attached to the membrane, cake layer porosity and compressibility, and fouling structure by scanning electron microscopy, among others.
- Developing effective fouling control strategies to minimize membrane fouling in the short- and log-term operation. In addition to CEB, continuous/periodical coagulant dosing could be of interest to capture soluble compounds and colloids, while increasing the average particle size in the influent.
- Evaluating the combination of DMF for MWW treatment with (advanced) post-treatments focused on recovering soluble nutrients from the generated permeate, such as complementary membrane systems (reverse osmosis, membrane distillation, etc.), cationic and anionic exchanging materials, or microalgae cultivation, among others, also studying the possible valorization of recovered resources.

- Developing a general filtration model able to capture the dynamics of the system at different operating conditions, configurations, etc.
- Conducting deeper economic and environmental analysis (*i.e.* life cycle assessment and life cycle cost analysis) to properly determine the potential benefits of proposed alternative.

CHAPTER 11. Conclusions

This PhD thesis aimed to evaluate the potential of direct membrane filtration technology to boost resource recovery from municipal wastewater. Membrane fouling remains as a key issue to be attended for achieving feasible full-scale implementation. Hence, this PhD thesis mainly focused on assessing the viability of different membrane fouling control strategies, identifying effective design and operating strategies to minimize fouling propensity. A membrane-based pilot plant fitted with industrial equipment was operated for treating municipal wastewater from a full-scale wastewater treatment facility. From this, the following conclusions can be drawn:

Sludge filterability assessment

1. Sludge filterability was found to strongly depend on the operating solids concentration and soluble microbial products content, which was mainly affected by one of these parameters depending on the characteristics of the sludge source. Biological sludge was more influenced by the microbial products content, while suspended solids concentration was the main responsible of filterability variations when treating sludge form the concentration by UF membranes of the supernatant of a primary settling step.
2. Among the filterability methodologies tested, capillary suction time method resulted in good correlations between sludge filterability and sludge filtration resistance independently of the origin of the sludge treated, within the range of solids and microbial products concentration evaluated. However, when treating sludge with negligible biological activity, the time to filter method was identified as the best alternative to correlate sludge filterability and resulting membrane performance.

Porous membranes (MF and UF) performance assessment

1. A strong relation between membrane pore size and fouling propensity was found, clearly lowering the latter as the former was reduced.
2. Increasing the solids concentration in the membrane tank was found as an effective strategy to reduce fouling propensity when using UF membranes, being advisable to operate at total suspended solids concentrations of above 6 g L^{-1}.

3. The origin of the wastewater entering the system (*i.e.* raw municipal wastewater or primary settler supernatant) had a negligible effect on UF membrane performance, observing similar fouling propensities when operating at solid concentrations over 6 g L^{-1}.
4. Severe fouling intensities were found when raising the operating transmembrane flux in UF. Thus, the use of low/moderate fluxes (around 10 LMH) is recommended since cost savings associated to reduced membrane area are hindered by increased chemical reagents cost for membrane cleaning and increased membrane replacement costs due to associated reductions in membrane lifespan.
5. Fouling was mainly identified as irreversible, being the physical fouling control strategies employed (*i.e.* air scouring and backwashing) inefficient to remove/minimize membrane fouling. Organic matter was found to be the major fouling promotor in the system, representing from 70 to 99% of total membrane fouling.
6. Negligible biodegradation of organics was observed in the UF membrane tank when operating at sludge retention times below 3 days.
7. Generated permeate met European discharge standards for non-sensible environments. They could then be directly discharged in no sensible water bodies, used for fertigation or treated by tertiary processes focused in recover and valorize its nutrients content.
8. Energy and carbon footprint balances showed promising results, achieving positive outputs (net energy producer and carbon footprint sink) in all scenarios. The economic balance identified this direct membrane filtration technology as a potential alternative for resource recovery from municipal wastewater treatment, achieving reasonable payback periods when operating at moderate fluxes.

Modelling

1. A simple and generic filtration model was proposed to estimate UF membrane fouling in the long-term. The proposed model was able to adequately capturing the dynamics on transmembrane pressure, within the experimental conditions evaluated. 4 out of the 7 model parameters were identified as influential on model outputs.

Dynamic membrane (DM) operation

1. Self-forming DMs where achieved when directly filtering municipal wastewater regardless of the influent treated. Nonetheless, using raw municipal wastewater helped in shortening the self-forming period while allowing the use of higher pore size supporting materials.
2. Increasing the transmembrane flux or operating solids concentration when treating primary settling supernatant was found ineffective to enhance permeate quality in the short-term.
3. Reducing the supporting material pore size and adding extra supporting material layers was found as an effective strategy to reduce the DM self-forming period when treating raw municipal wastewater. However, this strategy entailed higher membrane fouling propensities. Since a similar permeate quality was obtained regardless the supporting material employed, the use of just one supporting material layer with an average pose size of 5 µm or higher is recommended.
4. Poor permeate qualities were achieved by using DMs, far from meeting European discharge demands. Coagulant dosing can be used to effectively enhance resources capture (mainly colloidal particles and phosphate), also shortening the DM self-forming period.
5. Positive energy and carbon footprint balances were achieved in DM technology. Similarly, promising economic balances were achieved thanks to the low price and maintenance requirements of DMs. However, the resource recovery surplus of this alternative compared to primary settling was not significant enough to justify its application when treating municipal wastewater. Consequently, its use is recommended for more loaded influents with a low percentage of settable particles.

Appendix. Resumen extendido

La creciente escasez mundial de agua dulce junto con la actual crisis energética y cambio climático están impulsando la necesidad de modificar nuestros modelos de consumo hacia una economía circular. Las aguas residuales municipales (ARM) pueden ser una importante fuente de recursos incluyendo agua regenerada, energía y nutrientes. Lamentablemente, las plantas depuradoras actuales son incapaces de valorizar dichos recursos, basándose estos principalmente en procesos aerobios (fangos activos) que, además de desperdiciar el potencial energético de la materia orgánica afluente, presentan un elevado coste energético. Debido a ello, numerosas alternativas han sido planteadas para mejorar los tratamientos actuales, siendo la filtración directa de agua residual mediante membranas (FDM) una alternativa altamente interesante. La FDM consiste en incluir una etapa de filtración del ARM mediante membranas previamente al tratamiento de fangos activos. De esta forma, la fracción particulada de las ARM podría ser recuperada en el tanque de membranas, requiriendo solamente del tratamiento mediante procesos aerobios de la fracción soluble remanente lo que reduciría de forma muy significativa las necesidades energéticas del posterior proceso de fangos activos. Por otro lado, los lodos retenidos y concentrados en el tanque de membranas podrían ser destinados a digestión anaerobia, mejorando el rendimiento energético global del tratamiento de las ARM. Por tanto, esta alternativa permitiría la valorización de los recursos contenidos en las ARM sin suponer un gran cambio en las instalaciones de tratamiento ya existentes.

El presente estudio tuvo como objetivo evaluar el rendimiento de tres tipos de membranas: ultrafiltración (UF), microfiltración (MF) y membranas dinámicas (MD) con el fin de determinar aquella más adecuada para la implementación de dicho esquema de tratamiento. Asimismo, se estudiaron las condiciones de operación más ventajosas (nivel de pre-tratamiento del afluente alimentado al sistema, concentración de sólidos en el módulo de membranas, flujo transmembrana y tamaño de poro del material de soporte según la membrana operada) con el objetivo de minimizar el ensuciamiento de la membrana y aumentar la recuperación de recursos. Finalmente, se realizó un estudio de viabilidad preliminar centrado en los requisitos energéticos, huella de carbono e impacto económico del esquema alternativo de tratamiento propuesto.

Además de realizar varios experimentos a escala de laboratorio, este estudio se ejecutó principalmente mediante la planta piloto de membranas contemplada en el proyecto con financiación nacional CTM2017-86751-C, desarrollado por la Universitat de Valencia. Dicha planta piloto consta de tres módulos de membranas independientes equipados cada uno con un tipo de membrana: UF (PULSION® Koch Membrane Systems, 0,03 µm de tamaño de poro, área de filtración de 43,5 m^2), MF (TERAPORETM 5000 Mitsubishi, 0,4 µm de tamaño de poro, área de filtración de 18 m^2) y MD (tejido monofilamento de poliamida como material de soporte, NITEX® SEFAR, 1 y 5 µm de tamaño de poro, área de filtración de 2 m^2). La planta piloto fue alimentada con el ARM proveniente de la EDAR a escala industrial "Conca del Carraixet" (Valencia, España), alimentando cada tanque de membranas paralelamente con el mismo afluente para permitir una adecuada comparación entre cada tipo de membrana estudiada. Fricción por burbujeo de aire se utilizó para controlar la capa de torta formada en las membranas de UF y MF. Asimismo, el fango concentrado en cada módulo de membranas se purgó continuamente por rebose para operar a una concentración constante de sólidos según el caso. Las concentraciones de los principales contaminantes (turbidez, DQO y nutrientes) se analizaron en el afluente, lodo y permeado generados para determinar la calidad del efluente y la recuperación de recursos de cada tipo de membrana. Asimismo, la presión transmembrana (PTM) fue monitoreada continuamente para determinar el ensuciamiento de cada membrana según el flujo fijado.

Los principales resultados obtenidos durante este trabajo se organizan en los puntos siguientes, cada uno de ellos tomando como referencia un capítulo completo de esta tesis doctoral.

A.1. Estudio de la metodología más adecuada para determinar la filtrabilidad del fango producido en diferentes sistemas de tratamiento de aguas residuales municipales

Se estudió la capacidad de diferentes metodologías para determinar la filtrabilidad de distintos tipos de fango y estimar su resistencia a la filtración durante procesos reales de filtración. Tres métodos fueron evaluados durante este estudio: tiempo de succión capilar o CST (del inglés *capillary suction time*), tiempo de filtración o TTF (del inglés *time to filter*) y resistencia específica a la filtración o SRF (del inglés *specific resistance to filtration*). Estos métodos se evaluaron utilizando diferentes tipos de fango cada uno proveniente de un tratamiento de agua residual municipal distinto: fangos activos aerobios, sobrenadante de la decantación primaria posteriormente concentrado mediante membranas de ultrafiltración (EDP, de efluente de la decantación primaria) y el digestato producto de la co-digestión anaerobia de microalgas y fango primario. También se evaluó la capacidad de dichos métodos (CST, TTF y SRF) para estimar la concentración de sólidos suspendidos totales (SST) y SMP (sustancias solubles secretadas durante la actividad microbiana, del inglés *soluble microbial products*) en las muestras de fango, al tiempo que se validaron los resultados obtenidos mediante su correlación con la resistencia del fango observada durante un proceso real de filtración por membranas.

A.1.1. Filtrabilidad del fango

El CST se identificó como la metodología más adecuada para determinar la filtrabilidad del fango independientemente de su procedencia para las condiciones de operación evaluadas en este estudio. Sin embargo, el TTF se determinó como una sólida alternativa al tratar PSE y fangos anaerobios, logrando correlaciones similares e incluso superiores en algunos casos a las obtenidas por el método de CST. Por otro lado, en general, el método SRF resultó en las correlaciones más pobres dentro de los métodos evaluados, mostrando especialmente malos resultados con respecto al contenido de SMP en las muestras de fango. En consecuencia, el método de SRF se identificó como una alternativa no recomendable para monitorear la filtrabilidad del fango para las condiciones de operación evaluadas en este trabajo.

A.1.2. Correlación con el proceso real de filtración

El CST y TTF se determinaron como metodologías adecuadas para predecir la resistencia a la filtración del fango durante procesos de filtración mediante membranas o estimar su contenido en SST y SMP. El método de TTF presentó mejores resultados al tratar PSE, mientras que el método de CST fue la única opción disponible para el tratamiento de fango anaerobio, mostrando el TTF en este caso correlaciones insatisfactorias. En base a los resultados obtenidos en este estudio, se identificó al CST como el método más conveniente para el tratamiento de fangos biológicos. Por otro lado, al filtrar fangos sin actividad biológica (como, por ejemplo, PSE), el método de TTF se consideró como la alternativa más atractiva, ya que mostró mejores correlaciones con respecto a la filtrabilidad medida y la resistencia a la filtración de del fango durante su filtración mediante membranas.

A.2. Filtración directa mediante membrana de agua residual municipal: estudio de las condiciones más adecuadas para minimizar la tasa de ensuciamiento en membranas porosas comerciales a escala demostración

Se compararon dos tipos de membranas (ultrafiltración (UF) y microfiltración (MF)) con el fin de determinar la más adecuada para la filtración directa de ARM. Asimismo, se evaluó el efecto del afluente tratado (agua residual bruta tras un pretratamiento de desbaste, desarenado y desengrasado, o el efluente del decantador primario de la EDAR (EDP)) y la concentración de SST en el tanque de membranas (sobre 1 – 2,6 g L^{-1}) con el objetivo de determinar las condiciones más propicias en base a la minimización del ensuciamiento de la membrana.

Se estudió la eficacia de dos estrategias para el control del ensuciamiento (fricción por burbujeo de aire y contralavado periódico de permeado) durante el proceso de filtración, evaluando la reversibilidad del ensuciamiento en cada una de las condiciones experimentales probadas. Por otro lado, se estudió la recuperación de la permeabilidad de la membrana mediante limpiezas químicas, determinando la fuente principal del ensuciamiento (materia orgánica o inorgánica). Además, se estudiaron los principales mecanismos del ensuciamiento correlacionando los valores obtenidos experimentalmente con distintos modelos teóricos de filtración.

A.2.1. Efecto del tamaño de poro de la membrana

El tamaño de poro de la membrana empleada se determinó como una de las variables de mayor relevancia al filtrar ARM. La membrana de MF solo pudo operar por períodos de filtración extremadamente cortos (alrededor de 2 – 8 horas) antes de su completo ensuciamiento, obteniéndose no obstante plazos de filtración mucho más largos (entre los 34 – 69 días) al utilizar la membrana de UF. Está marcada disminución del ensuciamiento al reducir el tamaño de poro de la membrana empleada se atribuyó a una reducción del estrechamiento y/o bloqueo de los poros debido al depósito de sustancias solubles y/o coloidales durante la filtración. La membrana de UF se seleccionó como la opción más conveniente para la filtración directa de ARM.

A.2.2. Efecto del afluente utilizado

Se obtuvieron ensuciamientos de la membrana significativamente más bajos al filtrar ARM bruta en lugar de EDP, independientemente de la membrana utilizada o la concentración de SST fijada en el tanque de membranas. Estos resultados sugirieren que, tal y como se había identificado en el punto anterior, la relación entre el tamaño de partícula del afluente tratado y el tamaño de los poros de la membrana está estrechamente relacionada con la propensión al ensuciamiento al filtrar ARM. En consecuencia, aumentar el tamaño medio de las partículas en el afluente (como en el caso de la ARM bruta con respecto al EDP) puede producir una reducción del ensuciamiento de la membrana al reducir la cantidad de partículas con capacidad para bloquear o depositarse dentro de los poros de la membrana.

A.2.3. Efecto de la concentración de sólidos

La concentración de sólidos alcanzada en el tanque de membranas durante la filtración presentó distintos efectos según el tamaño de poro de la membrana empleada. Un marcado beneficio se observó al incrementar dicha concentración en el tanque de UF, reduciéndose significativamente el ensuciamiento de la membrana. Dado que este fenómeno se observó con independencia del afluente tratado, este se asoció a la capa de torta desarrollada sobre la superficie de la membrana durante la filtración. Dicha capa de torta podría tornarse más gruesa y robusta a medida que aumenta la concentración de sólidos, siendo capaz de proteger la superficie de la membrana de diversos contaminantes con un elevado potencial para producir el bloqueo de los poros de la membrana, como las partículas coloidales o los SMP, a medida que aumenta su espesor. Operar a concentraciones de sólidos relativamente elevadas en el módulo de membranas podría considerarse como una estrategia operacional efectiva para mejorar la viabilidad de la filtración directa de ARM. No obstante, el módulo de MF mostró resultados contrarios a los observados en el módulo de UF, incrementándose el ensuciamiento en este caso al aumentar la concentración de sólidos en el tanque de membrana. En este caso, dado el mayor tamaño de poro de la membrana de MF, esta estrategia podría no ser tan conveniente debido al incremento en el número de partículas capaces de bloquear los poros de la membrana.

A.2.4. Eficacia de las estrategias de control del ensuciamiento

Las estrategias de control del ensuciamiento utilizadas (fricción por burbujeo de aire y contralavado periódico de permeado) demostraron ser ineficaces para controlar el ensuciamiento durante la operación de la membrana de UF. Sin embargo, estas mismas estrategias lograron reducir significativamente el ensuciamiento producido en la membrana de MF, extendiendo el tiempo de funcionamiento de algunas horas (entre 2 – 8 horas) a algunos días (entre 5 – 6 días). Estos resultados indicaron que la principal fuente de ensuciamiento al operar la membrana de UF estaba relacionada con el ensuciamiento irreversible. Podría asumirse pues que las condiciones de operación establecidas durante la filtración (caudal de aire inyectado en el tanque de membrana por área de membrana de 0,1 m^3 m^{-2} h^{-1} y 2 min de contralavado cada 10 ciclos de filtración:relajación) fueron suficientes como para controlar el espesor de la capa de torta desarrollada durante el proceso. Por el contrario, una porción significativa del rápido ensuciamiento observado durante la operación de la membrana de MF parece ser reversible, pudiendo ser reducida incrementando la intensidad y/o periodicidad de las estrategias de control del ensuciamiento empleadas en este estudio, incrementando la viabilidad de las membranas de MF. No obstante, a pesar de las significativas mejoras logradas en el módulo de MF, su rendimiento fue marcadamente inferior al ostentando por el módulo de UF, siendo este segundo pues la opción claramente más conveniente para la FDM de ARM.

A.2.5. Estudio del ensuciamiento de las membranas

Las limpiezas químicas realizadas en cada tanque de membranas determinaron que una recuperación de entre el 70 – 85 % de la permeabilidad inicial de la membrana puede alcanzarse tras una limpieza básica ($NaOCl$ en este caso) al emplear la membrana de UF, aumentando dicha recuperación hasta el 92 – 99 % en el caso de la membrana de MF. La materia orgánica se identificó como el principal promotor del ensuciamiento durante la filtración de ambas membranas, mostrando una especial relevancia en el caso del módulo de MF, el cual solo pudo ser operado por períodos de tiempo relativamente cortos (de 5 a 7 días). No obstante, el ensuciamiento inorgánico (principalmente relacionado con la precipitación de compuestos inorgánicos en la superficie y/o poros de la membrana) también cumplió un papel significativo durante la operación del módulo de UF, cuyo periodo operacional se extendió de 1 a 2 meses. Por tanto, aunque el ensuciamiento de fuentes orgánicas es sin duda el principal factor a controlar (por ejemplo, mediante la aplicación de contralavados mejorados químicamente con soluciones básicas), es importante prestar cierta atención al ensuciamiento inorgánico con el objetivo de prolongar una mayor permeabilidad de la membrana y, por ende, incrementar la viabilidad del proceso. Con respecto a los mecanismos de ensuciamiento, la correlación de los datos experimentales con los modelos teóricos parece indicar que la formación de una capa de torta sobre la superficie de la membrana es el mecanismo predominante a corto plazo (durante cada ciclo de filtración). Sin embargo, a medida que avanza la filtración, el ensuciamiento irreversible se hace más notorio, pudiendo estar esté relacionado según los modelos aplicados a un bloqueo intermedio/completo de los poros de la membrana en el caso del módulo de MF y a una reducción del tamaño de los poros hasta su completo bloqueo en el caso del módulo de UF.

A.3. Estudio del potencial para recuperar recursos y viabilidad de la ultrafiltración directa de aguas residuales municipales a escala de demostración

Dado que la membrana de UF se identificó como la más adecuada para la FDM de ARM en comparación con el módulo de MF, se realizó una investigación más exhaustiva y extensa de dicho modulo. Se incrementó la concentración de sólidos en el tanque de membranas, evaluando dos concentraciones adicionales (6 y 11 g L^{-1} aproximadamente) para cada afluente previamente descrito (ARM bruta y EDP). El potencial de recuperación de recursos de la presente alternativa fue evaluado bajo todas las condiciones estudiadas, concluyendo este estudio con un análisis preliminar de los requisitos energéticos, huella de carbono e impacto económico del sistema de tratamiento propuesto.

A.3.1. Proceso de filtración

Se obtuvo una marcada reducción del ensuciamiento de la membrana al continuar aumentando la concentración de sólidos en el módulo de UF. Una tasa de ensuciamiento en torno a 0,55 mbar por día se identificó como el mínimo en este estudio, alcanzándose al operar por encima de los 6 g L^{-1} y permitiendo extender la filtración durante más de 120 días. El tratamiento de ARM bruta en lugar de EDP también mostró una ligera reducción sobre la tasa de ensuciamiento de la membrana. No obstante, en este caso dicho beneficio solo fue significativo al operar por debajo de los 2,6 g L^{-1}. Dado que los beneficios de filtrar ARM bruta sobre el ensuciamiento se asociaron al menor número de las partículas de menor tamaño en el afluente tratado, la formación de una capa de torta más robusta al operar a concentraciones elevadas de sólidos en el tanque de membranas reduciría la cantidad de material capaz de alcanzar la superficie de la membrana. Estos resultados validarían las conclusiones obtenidas con respecto a los efectos beneficiosos de la concentración de sólidos sobre el control del ensuciamiento irreversible de la membrana, resaltando la importancia de dicha estrategia al filtrar directamente ARM. Asimismo, podría concluirse que, desde el punto de vista de la filtración, el afluente alimentado es irrelevante siempre que se opere a concentraciones adecuadas de sólidos. Las conclusiones previas respecto al impacto de la materia orgánica sobre el ensuciamiento de la membrana también fueron confirmadas en este estudio, obteniéndose similares impactos sobre la permeabilidad (entre el 80 – 85%) debido a estos contaminantes. Dicho porcentaje se mantuvo independientemente de las condiciones operacionales o del periodo experimental (de 35 a 123 días).

A.3.2. Recuperación de recursos y calidad del permeado

El módulo de UF fue capaz de recuperar en torno al 80 – 85% de toda la DQO afluente, reduciéndose este porcentaje en torno al 20 – 40% en el caso de los nutrientes (nitrógeno y fósforo). A pesar de la inyección continuada de aire al sistema, no se observó una actividad microbiana relevante durante la filtración para tiempos de retención de solidos (TRS) por debajo de 3 días. No obstante, una parte significativa de la DQO fue consumida por las bacterias aerobias al incrementar el TRS sobre los 6 días, estableciendo sobre los 3 días el límite operacional. El permeado generado ostentó una elevada calidad gracias a la filtración por membranas, el cual se mantuvo independiente de las condiciones operacionales probadas. Por tanto, el tamaño de poro de la membrana empleada resultó ser el principal actor en la recuperación de recursos, jugando la capa de torta desarrollada sobre la superficie de la membrana un papel irrelevante en este aspecto, independientemente de su espesor. Desafortunadamente, a pesar de que las concentraciones de DQO y DBO cumplen con los requisitos de vertido estipulados por la normativa europea (concentraciones en el permeado alrededor de 47 – 64 y 18 – 21 mgDQO L^{-1}, respectivamente), la concentración de nutrientes en el permeado (nitrógeno y fósforo) sobrepasó las concentraciones máximas permitidas al considerar vertidos en zonas sensibles (concentraciones de amonio y fosfato en el permeado alrededor de 27 – 45 mgNH_4^+ L^{-1} y 3,2 – 4,8 mgPO_4^{3-} L^{-1}, respectivamente). En consecuencia, el permeado generado podría incorporarse a los cuerpos acuáticos de forma segura solamente en zonas no estipuladas como sensibles, requiriendo de un post-tratamiento en caso contrario. Dicho post-tratamiento estaría centrado en recuperar los nutrientes solubles del permeado, los cuales podrían ser posteriormente valorizados.

A.3.3. Viabilidad del proceso

Se obtuvieron resultados prometedores al evaluar el balance energético y huella de carbono de la alternativa propuesta, obteniendo en todos los casos evaluados balances positivos. Una recuperación energética en torno a 0,46 – 0,40 kWh por m^3 de ARM afluente puede alcanzarse gracias a la recuperación de la materia orgánica afluente, cubriéndose pues completamente la energía requerida por el proceso de filtración y generando un exceso energético. Asimismo, dado que producciones netas de energía pueden alcanzarse mediante este sistema de tratamiento, la FDM funcionaría también como un sumidero de CO_2, reduciendo las emisiones globales en 0,19 – 0,16 kg CO_2-eq por m^3 de ARM afluente gracias a la reducción en el consumo de combustibles fósiles. En cuanto al punto de vista económico, se determinó que operar por encima de 10 LMH no es una estrategia recomendable. Esto es debido a que, si bien la inversión inicial en área de membrana puede reducirse debido al mayor caudal de permeado tratado, el severo ensuciamiento obtenido al operar con flujos de filtración más altos obliga a un importante aumento en la periodicidad de limpiezas químicas, incrementándose notablemente los costos operacionales y remplazos de membrana del sistema. Se recomienda operar a flujos de permeado moderados/bajos con el fin de minimizar el ensuciamiento de la membrana, alcanzándose en este estudio un posible beneficio de 0,035 € por m^3 de ARM afluente al operar a un flujo de 10 LMH. Dicho beneficio supondría un periodo de amortización solamente considerando los costos de la membrana de 12,3 años.

A.4. Desarrollo de un modelo de filtración simple y genérico con el que predecir el ensuciamiento de la membrana al filtrar directamente agua residual municipal

Se propuso un modelo simple y genérico para predecir el ensuciamiento de la membrana con respecto a las condiciones operacionales estudiadas en el módulo de UF. Dicho modelo se basó en una estructura de resistencias en serie, simplificando el ensuciamiento como consecuencia de dos fuentes diferentes: la capa de torta formada, relacionada con el material en suspensión, y el bloqueo de poros, relacionado con la concentración de partículas coloidales y compuestos solubles.

A.4.1. Capacidad predictiva del modelo

El modelo propuesto fue capaz de generar tasas de ensuciamiento similares a las obtenidas experimentalmente según la concentración de sólidos en el tanque de membranas. Se obtuvieron reducidas discrepancias entre los valores teóricos y experimentales (entre 5 – 25 mbar), siendo estos calculados como la raíz del error medio cuadrático (REMC). El modelo propuesto fue también capaz de ajustarse al cambio de afluente (ARM bruta y EDP), requiriendo modificar solamente 3 de los 7 parámetros del modelo para obtener una adecuada evolución del ensuciamiento teórico.

A.4.2. Análisis de sensibilidad e incertidumbre

De los 7 parámetros propuestos, 4 (δ_C, δ_B, k_I y α'), fueron identificados como los más sensibles según el método de sensibilidad general de Morris. Por otro lado, el análisis de incertidumbre mostró que modificar el valor de los parámetros del modelo tiene una marcada repercusión sobre las capacidades predictivas del mismo, obteniéndose discrepancias muy significativas al considerar periodos largos de simulación. Esto es debido a que la evolución del ensuciamiento de la membrana predicho por el modelo depende en gran medida de su estado previo, acumulándose pues un error aditivo en el caso de no calibrar adecuadamente uno de los parámetros del modelo. El modelo propuesto presenta no obstante un elevado potencial para generar predicciones razonables a partir de una estructura simple y fácilmente adaptable, permitiendo así pues su uso conjuntamente a otras herramientas complementarias para la optimización y control de procesos de filtración.

A.5. Aplicación de membranas dinámicas para mejorar la recuperación de recursos en las aguas residuales municipales

Dado que el severo ensuciamiento de la membrana durante la filtración directa de ARM, junto con los elevados costes de adquisición y sustitución de la misma, son los principales factores limitantes para la aplicación de la FDM a gran escala, se evaluó la posible sustitución de membranas porosas por membranas dinámicas (MD). En este trabajo se estudió el ensuciamiento y calidad del permeado generado por una MD al filtrar EDP. Se evaluaron distintas estrategias (aumento del flujo transmembrana, aumento de la concentración de sólidos en el tanque de membrana y dosificación de coagulante) para reducir el tiempo de auto-formación de la MD y aumentar su capacidad para la recuperación de recursos. Finalmente, se evaluó la viabilidad de la alternativa propuesta a partir de un estudio preliminar que consideró sus potenciales requisitos energéticos y económicos.

A.5.1. Proceso de filtración

Se determinó que al filtrar EDP, una única capa de material de soporte (malla monofilamento abierta de poliamida con un tamaño medio de poro de 1 µm) es insuficiente para auto-formar una MD a corto plazo. Esto se debió a la baja carga de sólidos en el afluente tratado, así como al bajo tamaño medio de las partículas del mismo, siendo pues el tratamiento de efluentes pre-tratados un claro limitante para el uso de MD a pesar del relativamente bajo tamaño de poro del material de soporte empleado en este estudio. A pesar de estos primeros resultados, el uso de una capa adicional de material de soporte permitió la auto-formación de la MD tras 17 días de operación, presentándose como una posible alternativa para el tratamiento de este tipo de afluentes. Asimismo, un aumento en la concentración de sólidos también consiguió resolver esta limitación, reduciendo el periodo de auto-formación hasta los 8, 6 y 4 días según la concentración de sólidos alcanzada en el tanque de membranas (1,9, 4,7 y 9,2 g L^{-1}, respectivamente).

A.5.2. Estrategias para mejorar la recuperación de recursos

La MD auto-formada durante la filtración de EDP logró recuperaciones relativamente pobres (entre el 10 – 40% según el contaminante), generando un permeado lejos de cumplir con los límites de vertido establecidos por la normativa europea (concentración de SST, DQO, nitrógeno total (NT), fósforo total (PT) y turbidez sobre los 65 mg L^{-1}, 141 mg L^{-1}, 42,3 mg L^{-1}, 4,3 mg L^{-1} y 86 NTU, respectivamente). Esto se debió al bajo porcentaje de material particulado en el afluente tratado, siendo éste el único susceptible de ser retenido por la membrana empleada. Asimismo, la reducida carga de sólidos en el afluente también redujo la capacidad de formar una torta lo suficientemente robusta y espesa como para mejorar la capacidad de retención de la MD. Un aumento del flujo transmembrana (de 15 a 49 LMH) y un aumento de la concentración de sólidos en el módulo de membrana (sobre 1,9, 4,7 y 9,2 g L^{-1}) se evaluaron como posibles alternativas para mejorar la calidad del permeado producido, obteniendo no obstante resultados similares a los anteriormente mostrados. Se concluyó pues que cambios significativos en la estructura de la MD auto-formada no son posibles mediante estas estrategias a corto plazo, siendo la torta formada sobre el material de soporte incapaz de retener el material particulado de pequeño tamaño. Cuatro coagulantes comerciales se probaron con el objetivo de incrementar la calidad del permeado generado, identificándose el PHLA18 a una concentración de 10 mg L^{-1} como la opción más recomendable. Gracias a la dosificación de coagulante se mejoró significativamente tanto el tiempo de auto-formación de la MD como su capacidad de captura de recursos, consiguiendo en este caso un periodo de auto-formación de 7 días junto a una calidad de permeado en cuanto a SST, DQO, NT, PT y turbidez en torno a 24 mg L^{-1}, 58 mg L^{-1}, 38,1 mg L^{-1}, 1,2 mg L^{-1} y 22 NTU, respectivamente.

A.5.3. Viabilidad del proceso

Debido a la limitada capacidad de la MD sobre la recuperación de recursos, se obtuvieron bajas recuperaciones energéticas en este estudio, logrando valores sobre los 0,030 kWh por m^3 de afluente tratado. Tal y como se comentó en el punto anterior, esto fue debido a la baja carga de material particulado en el afluente, identificando las MD como una tecnología inadecuada para el tratamiento de este tipo de afluentes pre-tratados. Sin embargo, esta limitación puede ser resuelta mediante la dosificación de coagulante, consiguiéndose en este segundo caso una recuperación energética mucho más competitiva (sobre los 0,121 kWh por m^3 de afluente tratado). Desgraciadamente, dado que sería necesaria una dosificación continua de reactivos, los costes de explotación de la MD ascenderían a 0,003 € por m^3 de afluente tratado, identificándose un costo operacional en este caso. Teniendo en cuenta que el permeado producido por esta alternativa podría requerir de un post-tratamiento enfocado a recuperar la fracción nitrogenada soluble con el objetivo de una segura incorporación en ambientes sensibles, la aplicabilidad de la alternativa presentada sería extremadamente limitada y apenas competitiva con respecto a otras tecnologías de membrana (por ejemplo, las membranas de UF).

A.6. Estudio de la viabilidad de emplear membranas dinámicas para la filtración directa de agua residual municipal

Dadas las limitaciones de tratar afluentes pre-tratados con MDs, este estudio se centró en evaluar la posible aplicación de esta tecnología de membranas para la filtración directa de ARM bruta. Como en el caso anterior, se evaluó tanto la capacidad de auto-formar una MD a corto plazo como su capacidad para recuperar recursos y la viabilidad energética y económica.

A.6.1. Proceso de filtración y recuperación de recursos

La mayor carga de sólidos del ARM bruta respecto al EDP produjo una marcada reducción en el tiempo requerido para la auto-formación de la MD, siendo posible emplear en este caso materiales de soporte con mayores tamaños de poro. En este estudio se evaluó la mejor alternativa de entre cuatro posibles (una o dos capas de una malla monofilamento abierta de poliamida con un tamaño medio de poro de 1 o 5 µm). Periodos de auto-formación de entre varias horas a 3 días se obtuvieron según la alternativa propuesta, identificándose una reducción de la permeabilidad de 5,21 a 10,03 LMH bar^{-1} $día^{-1}$ a medida que se incrementó la capacidad de captura del material de soporte. La mayor propensión al ensuciamiento a medida que aumentan las capas de material de soporte o se reduce su tamaño medio de poro se atribuyó a la formación más rápida de capas de torta con un elevado espesor sobre el material de soporte. Desafortunadamente, se obtuvieron calidades de permeado relativamente similares independientemente de la alternativa empleada. Por tanto, el espesor de la capa de torta formada fue incapaz de incrementar significativamente la capacidad de la MD para producir permeados de mayor calidad. En este caso, se obtuvo una calidad de permeado en cuanto a SST, DQO, NT, PT y turbidez de 67 – 88 mg L^{-1}, 155 – 186 mg L^{-1}, 48,7 – 50,4 mg L^{-1}, 4,7 – 4,9 mg L^{-1} y 167 – 174 NTU, respectivamente, no encontrando diferencias significativas en cuanto al permeado generado al tratar EDP. En consecuencia, dado que el período de auto-formación no fue un factor limitante en este caso y el uso de materiales de soporte más restrictivos solo aumentó la tasa de ensuciamiento en la membrana, se concluyó que el uso de una única capa de material de soporte con un tamaño de poro medio de 5 µm o superior era la alternativa más adecuada para el tratamiento de ARM bruta.

A.6.2. Viabilidad del proceso

El estudio energético y de huella de carbono preliminar realizado a la alternativa propuesta mostró resultados muy favorables gracias a los bajos requisitos energéticos de las MD. La filtración de ARM bruta mediante MD alcanzó producciones netas de energía y reducciones globales de huella de carbono, lográndose en este estudio un excedente energético del orden de 0,33 kWh por m^3 de afluente tratado con un sumidero de huella de carbono de 0,13 $kgCO_2$-eq por m^3 de afluente tratado. Desafortunadamente, los resultados obtenidos suponen una ligera mejora con respecto a la tecnología actualmente en uso para el pre-tratamiento de ARM (decantación primaria), no siendo el excedente alcanzado suficientemente significativo como para justificar el uso de la presente alternativa frente a instalaciones ya en funcionamiento. La presente alternativa también logró buenos resultados al considerar su impacto económico, alcanzándose en este estudio un beneficio de alrededor de 0,065 € por m^3 de afluente tratado gracias a los bajos requisitos de mantenimiento de la MD. Asimismo, gracias a los bajos costes del material de soporte, periodos de amortización extraordinariamente bajos pudieron lograrse, estimándose en este estudio un periodo de meses (sobre 0,08 años). Desafortunadamente, dado que el permeado generado por esta alternativa necesita de un tratamiento secundario para permitir su segura incorporación a cuerpos acuáticos, su uso en sustitución de la decantación primaria no estaría justificada para el tratamiento de ARM bruta, requiriendo de afluentes con una mayor carga de material particulado no sedimentable para ser considerada como una opción competitiva.

A.7. Conclusión general y perspectivas futuras

Teniendo en cuenta todos los resultados obtenidos durante esta tesis doctoral, puede concluirse que las membranas de UF parecen ser la tecnología más adecuada para la implementación de la FDM de ARM. Las membranas de MF presentaron un ensuciamiento extraordinariamente más elevado al operar bajo condiciones similares, mientras que las MD, a pesar de lograr un nicho competitivo cuando se busca reducir los costos de inversión y mantenimiento de la membrana, no pudieron alcanzar una recuperación de recursos suficientemente elevada como para competir con las membranas porosas.

En cuanto a las condiciones de operación, los resultados obtenidos durante este estudio sugieren el uso de flujos de permeado bajos/moderados (alrededor de 10 LMH) con el fin de reducir tanto como sea posible la propensión al ensuciamiento de la membrana. De hecho, la reducción en los costes de inversión que podrían obtenerse aumentando el flujo de permeado no compensa en ningún caso el marcado incremento de los costes operativos derivados del severo incremento del ensuciamiento de la membrana. Operar a concentraciones de sólidos en el tanque de membrana relativamente altos (entre 6 y 11 g L^{-1}) es también una importante recomendación para minimizar el ensuciamiento de la membrana. No obstante, se recomienda no superar un TRS de más de 3 días con el objetivo de evitar pérdidas significativas de la materia orgánica recuperada debido a la promoción de microorganismos aerobios en el tanque de membrana.

Con respecto al afluente tratado (ARM bruta y EDP), ambos presentaron una propensión similar al ensuciamiento de la membrana al operar bajo concentraciones de sólidos adecuadas. Por tanto, podría recomendarse el uso de ARM bruta dado que su mayor carga de sólidos permitiría alcanzar mayores concentraciones de sólidos en el tanque de membrana a menores TRS. La materia orgánica fue identificada como el principal promotor del ensuciamiento durante la filtración, sugiriéndose por tanto el uso de estrategias de control del ensuciamiento centradas en minimizar su influencia, como la aplicación de contralavados mejorados químicamente con soluciones básicas.

Este estudio reafirma el potencial de la FDM como una prometedora alternativa para transformar las estaciones depuradoras actuales en instalaciones de recuperación de recursos. Sin embargo, aunque los balances de energía y huella de carbono muestran una clara ventaja de la propuesta alternativa en comparación con tratamientos actualmente en uso, aun se requieren de ciertas mejoras en relación a su impacto económico para poder ser considerada como una sólida alternativa dentro del marco de tratamiento de ARM.

Futuros estudios podrían incluir:

- Una comparación más amplia entre las tecnologías de membrana disponibles (incluyendo otros tipos de membrana como la nano-filtración y la ósmosis) con el fin de determinar la más adecuada y corroborar las conclusiones obtenidas en este estudio.
- Procesos de filtración a más largo plazo con el objetivo de alcanzar el ensuciamiento completo de la membrana bajo todas las condiciones operacionales probadas y corroborar adecuadamente la periodicidad de limpieza química de la membrana. Asimismo, también es necesario evaluar la propensión al ensuciamiento al operar a flujos más elevados para verificar los resultados obtenidos en este estudio, requiriendo de filtraciones a medio y largo plazo.
- Un estudio más exhaustivo de los principales mecanismos del ensuciamiento. Análisis específicos a escala de laboratorio tales como determinar la cantidad de masa adherida a la membrana, la porosidad y compresibilidad de la capa de torta formada y la estructura del ensuciamiento de la membrana mediante microscopía electrónica de barrido; podrían sugerirse para corroborar las hipótesis realizadas en este estudio y contribuir con más información sobre la naturaleza del ensuciamiento desarrollado.
- Una vez aclarado el origen y los mecanismos del ensuciamiento, estrategias más efectivas y eficientes para su control deben ser también apropiadamente evaluadas. Además de la aplicación de contralavados mejorados químicamente, la dosificación periódica/continua de coagulante también podría ser una estrategia interesante a evaluar para aumentar la ratio de capturar de compuestos solubles y coloides al tiempo que aumentar el tamaño promedio de las partículas en el fango tratado. Sus efectos sobre el proceso de filtración y su contribución a los costos operativos deben estudiarse adecuadamente al filtrar agua residual real en módulos de membrana comerciales, considerando también su combinación con todas las demás estrategias operativas y de diseño determinadas en este trabajo doctoral.
- Post-tratamientos enfocados a recuperar la fracción soluble de los nutrientes presentes en el permeado, tales como sistemas de membranas complementarios (ósmosis inversa, destilación por membranas, etc.), materiales de intercambio

catiónico y aniónico o cultivos de microalgas, entre otros, necesitan ser evaluados, estudiando también la posible valorización de los recursos recuperados.

- El modelo de filtración propuesto debe someterse a pruebas más exhaustivas, considerando una mayor cantidad de condiciones operativas a largo plazo para validar su utilidad y proponer una estructura más robusta. Su uso para la optimización del proceso de filtración debe también estudiarse adecuadamente para demostrar su utilidad en este campo.
- Análisis económicos y ambientales más profundos (es decir, análisis de ciclo de vida y análisis de costes del ciclo de vida) son necesarios para determinar apropiadamente los beneficios potenciales de la alternativa propuesta.