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Additional Information

# Anaerobic membrane bioreactors for resource recovery from municipal wastewater: a comprehensive review of recent advances

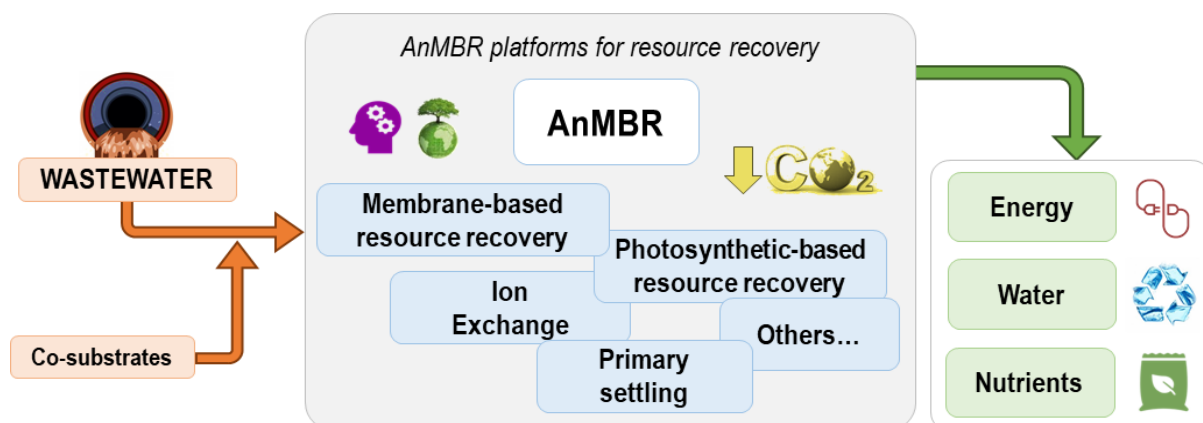
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## Abstract

In a paradigm shift towards a sustainable society based on the Circular Economy, wastewater treatments are rapidly evolving towards simultaneous recovery and reuse of clean water, renewable energy, and nutrients. This review examines recent advances (from 2016 to 2020) in the potential of anaerobic membrane bioreactors (AnMBRs) to serve as the core technology for municipal wastewater (MWW) resource recovery, focusing on the latest technological advances and economic and environmental innovation perspectives. The potentials and limitations of AnMBR for further full-scale application and new platforms to address these challenges are discussed, covering systems based on co-digestion, pre-concentration of particulate organics, removal, concentration and recovery of inorganic nutrients (photosynthesis, membranes and ion exchange) and hybrid systems for enhanced energy recovery. Overall, this review provides important insights into overcoming the challenges that hinder the application of AnMBR to MWW resource recovery from the technical, environmental and economic points of view.



## Keywords

Anaerobic membrane bioreactor (AnMBR), Circular Economy, municipal wastewater, resource recovery,

water resource recovery facility (WRRF)

## Highlights

Recovery systems are needed for recycling key resources from wastewater

AnMBR can be used to recover water, energy and nutrients from municipal wastewater

Challenges that hinder full-scale AnMBR implementation are identified

New AnMBR platforms are gaining attention to improve economic and environmental viability of AnMBRs

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## 1. Introduction

The increased pressure on water supplies, the need to reduce carbon footprints, and the depletion of resources have made it crucial to apply a new development model focused on the Circular Economy (CE)

1. In the CE model, the former wastewater treatment plants (WWTP) are being progressively understood as water resource recovery facilities (WRRF) since wastewater is regarded as a source of resources rather than as a source of contaminants<sup>2-4</sup>. This change of paradigm in wastewater treatment could achieve environmental goals within the water sector whilst producing valuable products from a renewable source, (i.e. nutrients, reclaimed water, energy, etc.) to be re-cycled into other value chains.

Nutrient recycling from wastewater to farmland is of great interest in CE principles. Phosphorus (P) is a fundamental element, which was included in the EU's Critical Raw Material List in 2017<sup>5</sup>, due to its supply risk in the near future and its economic importance. Different authors have estimated that it may be depleted by the end of the 21<sup>st</sup> century, seriously threatening global food security<sup>6</sup>. As the present methods of producing fertilizers from nitrogen (N) need high amounts of energy and significantly impact global warming<sup>7</sup>, N recovery from wastewaters could reduce worldwide energy consumption by reducing fertilizer production.

The increasing demand for water to meet the worldwide population growth, together with the unsustainable management of resources can affect water supply in terms of quality and quantity and therefore increase the existing pressures on water resources<sup>8</sup>. The reutilization of treated wastewater can provide important environmental, economic, and social benefits if regulators and policy makers ensure adequate water reuse implementation around the world<sup>9</sup> and if this includes recycling the nutrients embedded in the effluent (i.e. fertigation) the benefits are even greater.

From the global energy crisis perspective it is necessary to implement more cost-effective and sustainable technologies to replace current aerobic processes that require considerable amounts of energy for organic matter oxidation<sup>10</sup>. In a conventional WWTP, this energy demand accounts for more than 50% of the total energy consumption and a large amount of poorly biodegradable biomass is produced. These drawbacks have increased interest in anaerobic systems since they have the potential to be both net energy producers and allow nutrients to be recovered whilst reducing waste sludge production.

However, anaerobic treatments can present problems in certain operating conditions (e.g. treatment flow rate, temperature, etc.) and when treating particular types of waste (e.g. low-strength wastewaters, such as urban wastewaters) mainly due to the low biomass growth rate at sub-mesophilic temperatures and their sensitivity to process dynamics<sup>11</sup>. Up to now, anaerobic processes have been limited to municipal wastewater (MWW) in warm climates or to highly loaded streams (i.e. industrial wastewaters, sludge, etc.) in which the amount of methane produced can increase the operating temperature of the anaerobic reactor. Anaerobic membrane bioreactors (AnMBRs, see typical treatment scheme in Figure 1) can overcome the drawbacks of applying anaerobic processes to low-loaded wastewater by introducing a membrane filtration system for the complete retention of biomass.

AnMBR technology thus has many advantages that could help WWTPs to become WRRFs: i.e. reduced footprints, effluent quality, organic matter valorization, reduced carbon footprint, low biosolids production and on-site stabilization, and nutrient recovery potential<sup>12</sup>. Nonetheless, different authors have reported several issues that need to be solved in order to make AnMBR a feasible technology for MWW: i) the presence of methane dissolved in the effluent, which needs to be captured both to prevent its stripping to the atmosphere downstream and to enhance energy recovery<sup>13</sup>; ii) membrane fouling, which reduces system productivity and increases the cleaning requirements, thus reducing the membrane lifespan and raising operating expenses<sup>1</sup>; and iii) the competition between sulfate-reducing bacteria (SRB) and methanogens for the available substrate<sup>14</sup>. The presence of nutrients in AnMBR effluent has also been reported as a drawback, since N and P may not comply with the discharge limits imposed in certain

areas<sup>15</sup>, affecting eutrophication. However, nutrient availability in AnMBR effluents should be considered as an opportunity to re-introduce them into a self-sufficient cradle-to-cradle bio-based economy through the development of novel, effective and sustainable platforms (fertigation, microalgae cultivation, ion exchange, etc.). A technological, economic and environmental approach is thus needed for nutrient discharge. Table 1 provides an overview of the effluent quality of different AnMBR systems as well as biogas and sludge composition. This paper aims to review current advances (from 2016 to 2020) in AnMBR-based processes for resource recovery from MWWs.

This review examines advances from 2016 to 2020 in the potential of AnMBR technology to serve as the core technology for resource recovery from MWW. First, key challenges that limit the applicability of AnMBR technology to municipal MWW treatment are critically reviewed (effluent quality, dissolved methane, membrane fouling, etc.), which finally affect overall resource recovery. Then, novel platforms for resource recovery are discussed in depth (co-digestion, pre-concentration of particulate organics, removal, concentration and recovery of inorganic nutrients (photosynthesis, membranes and ion exchange) and hybrid systems for enhanced energy recovery), focusing on the latest technological advances and economic and environmental innovation perspectives. Finally, future perspectives and development outlooks towards full-scale implementation of AnMBR-based systems focused on resource recovery are described in the form of a research roadmap for further development of AnMBR for resource recovery from wastewater.

## **2. Potentials and limitations of AnMBR technology for full scale implementation**

### **2.1. Water recovery**

Due to the extension of water stress issues, reclaimed water can provide important environmental, economic and social benefits if regulators and policy makers ensure adequate water reuse implementation around the world <sup>9</sup>. The main challenges of water reclamation are not only technological, environmental and economic issues, but also social acceptance and the political framework, both crucial to guarantee feasibility and sustainability of water reclamation. The main current concerns are related to

microbial risk to public health and environmental risk<sup>12</sup>. Further advancing on these aspects would demonstrate the capability of AnMBR technology for water resource recovery.

#### 2.1.1. Irrigation, fertigation, and other recycling uses

The use of reclaimed wastewater for irrigation has continuously increased in the last decades due to water scarcity and the depletion of ground water resources. Treated wastewater is considered a reliable source of water with high nutritional value for crops. There are several examples in literature dealing with reclaimed wastewater being used for irrigation of different crops such as olives<sup>16</sup>, nectarines<sup>17</sup>, avocados<sup>18</sup>, etc, although several authors have observed adverse effects related to the higher salinity of reclaimed wastewater in long term experience<sup>16, 18</sup>. These adverse effects are the steady increase of sodium absorption ratio of the soil solution and higher root sodium content. Precipitation should ensure enough seasonal leaching of salts to avoid these issues. In some of these cases, not only was wastewater reused after an appropriate treatment, but the nutrients present in the influent wastewater were not removed in the WWTP or used as fertilizers. Pedrero et al.<sup>17</sup> observed that fruit from trees irrigated with reclaimed wastewater showed higher values in most of the fruit quality parameters than the ones irrigated with fresh water due to the higher nutrient concentration of the reclaimed wastewater. Reclaimed wastewater also has urban uses such as the irrigation of parks, playgrounds, schoolyards, residences, golf courses, cemeteries and highway medians, industrial uses, such as cooling, washing, and boiling, and household uses, such as toilet flushing<sup>19</sup>.

#### 2.1.2. Pathogens and removal of emerging contaminants

The high quality of AnMBR effluent makes this stream interesting for reuse in irrigation and fertigation. Peña et al.<sup>20</sup> characterized the effluent of a pilot-scale AnMBR treating MWW and demonstrated its suitability for agricultural irrigation according to the Spanish law (R.D.1620/207) regarding the reuse of treated wastewater. All the physical-chemical parameters (including heavy metals) were below maximum admissible values, fitting into the most restrictive category. Helminth eggs were below 1 egg per 10L and

*E. coli* was clearly below the limit of 100 cfu per 100mL. Similar results were obtained by Foglia et al.<sup>21</sup>, since they found that the quality of the effluent of an AnMBR pilot plant was significantly better because it was free from total suspended solids and fell into Class A, since ultrafiltration also guarantees disinfection and pathogen removal. These authors also proved that the effluent from an UASB pilot plant did not comply with the EU limits (2020/741) for water reuse.

AnMBR technology is able to produce an effluent complying with the EU Class A recycling water quality standards (EU Directive 2020/741) even when treating other wastewaters other than municipal WW.

Moazzem et al.<sup>22</sup> operated a laboratory scale AnMBR fed with car wash wastewater. The *E. coli* found in the raw car wash wastewater was 4900 orgs per 100 mL. After treating the wastewater in the AnMBR no *E. coli* was found in the permeate.

The membrane pore size usually applied in AnMBRs provides a direct barrier that stops bacteria passing through the membrane by size exclusion, i.e. the membrane pore size is smaller than the bacteria.

However, for viruses this is not often the case, since viruses can vary in size from approximately 20 to 200 nm, so that virus removal in AnMBRs cannot be correlated with fecal coliform present in the effluent as is usual in activated sludge systems. The membrane will exclude coliforms in the effluent. However, the pores are too large to ensure viruses are excluded. The literature on virus removal in AnMBR systems is scarce. Fox and Stuckey<sup>23</sup> observed that 0.4 µm membrane alone showed relatively poor phage rejection for both MS-2 and T4 phages (LRV  $0.7 \pm 0.4$  and  $2.3 \pm 0.2$ , respectively). The AnMBR performance increased virus removal mainly due to membrane fouling. The highest log removal values (LRV) were up to 1.7-2.1 for MS-2 and 5.1-5.3 for T4 and were obtained for the lowest gas sparging rate, which promotes membrane fouling. Further research is needed to gain a deeper understanding of virus removal mechanisms in AnMBR systems.

Antibiotic resistant genes (ARGs) enable bacteria to survive in the presence of antibiotics. They can be classified into different types according to their action mechanism and the class of antibiotics they resist. A general pattern has been observed; each time a new antibiotic is launched on the market ARGs appear not long after. It should be highlighted the capability of ARGs to be transferred between different bacterial



species when they are present in the same environment, such as in biological reactors. As ARGs are capable of persisting and even disseminating into the environment via wastewater discharge, the presence of ARGs in reclaimed wastewater used for irrigation could be a problem for human health in the future since it is commonly acknowledged that ARGs can be transferred widely between microorganisms that are present in the environment through mobile genetic elements. The relative abundance of the genes determines the potential for intercellular ARG proliferation and the antibiotic resistance level of microorganisms. Igwaran et al.<sup>24</sup> examined the antibiotics resistance patterns and the presence of some resistant genes among *E. coli* isolates from effluents from different WWTPs in South Africa. The resistance exhibited by *E. coli* isolates against the antibiotics tested was: clindamycin and erythromycin (100% each), sulphamethoxazole (99%), amoxicilin (94.5%), doxycycline (90%), trimithoprim (83.7%), cefuroxime (64.8%), ofloxacin and ciprofloxacin (60.3% each), colistin-sulfate (58.5.1%), gentamicin (52.2%) and menopenem (48.6%).

The implications of the use of reclaimed water for irrigation in terms of dissemination of antibiotic resistance are still unclear, but different studies have found that some ARGs tend to accumulate in soils after being irrigated with reclaimed wastewater for a long time<sup>25</sup>. These results have encouraged the discussion on including threshold values for ARGs in the effluent from wastewater treatment plants<sup>26–28</sup>. Several obstacles in defining a threshold value have been pointed out in these works, such as: a) the threshold value should be “low enough” to minimize the risk of ARGs spreading, b) it has to be easily reached with affordable wastewater treatments, and c) low cost and ease of use methods for quantification at very low levels are required. Further research is needed to determine the contribution of irrigation with reclaimed wastewater to the spread of antibiotic resistance and to minimize ARG soil contamination.

Regarding AnMBR technology, different studies have demonstrated the ability of anaerobic membrane bioreactors to remove antibiotic-resistant bacteria (ARB) and ARG. Cheng and Hong<sup>29</sup> evaluated the removal of ARB in a pilot-scale AnMBR plant. The results obtained showed that new membranes with a minimal fouling layer could remove more than 5 log units of ARB. However, as the membranes

progressed to subcritical fouling, the ARB log removal values (LRVs) decreased at increasing operating transmembrane pressures but returned to the level achieved by the new membranes with critically fouled membranes. Kappell et al.<sup>30</sup> observed a log reduction of ARG close to 3.5 removal in AnMBR treatment. The reduction in ARG was due to the membrane filtration process since the ARG concentration in the influent wastewater, the effluent of the primary settler and the membrane tank feed were similar. Although the size of free ARGs is clearly lower than the pore size, some membrane pores are partially blocked with fouling matter, which reduces the effective pore size and enhances ARG removal<sup>31</sup>. These authors observed a significant increase in the production of extracellular polymeric substances (EPS) and soluble microbial products (SMP) when a mixture of several antibiotics was added to the synthetic influent wastewater. The removal of several extracellular ARGs were significantly correlated with the protein, polysaccharide, and total contents of the EPS and SMP matrix. The higher the membrane fouling the higher the reduction observed in extracellular ARGs.

In a recent study, Zarei-Baygi et al.<sup>32</sup> found numerous strong positive correlations between certain ARGs and microbial groups (OTUs) in the biomass of a bench scale AnMBR plant. Although a statistically significant positive correlation between a microbial group and a particular ARG cannot be considered as evidence of that group carrying antibiotic resistance, it can indicate that the OTUs are potential host bacteria for ARGs and mobile genetic elements.

Finally, Harb et al.<sup>33</sup> considered AnMBR technology a viable option for reducing the risk of ARG persistence in the environment. They observed a lower concentration of ARGs associated with sulfamethoxazole and trimethoprim resistance in the effluent of an AnMBR system than in the effluent of an MBR system. The average and peak relative abundance of *sul1*, *sul2*, *intl1*, and *dfrA5* genes was over one order of magnitude lower in the effluent of AnMBR than in aerobic systems. On the other hand, co-treatment of MWW with livestock manure has also been shown to improve ARG removal efficiency of AnMBR<sup>34</sup>. These authors operated a lab scale AnMBR fed with municipal wastewater and increasing amounts of cattle manure. For ARGs, the contribution of the wastewater was higher than that of the cattle manure. The more the cattle manure added, the higher the ARG removal efficiency.

## **2.2. Energy recovery and fossil-carbon emissions mitigation**

Numerous studies are available in the literature related to the estimation and minimization of greenhouse gas (GHG) emissions from wastewater treatment by substituting aerobic processes – an energy-intensive activity due to the energy input for organic oxidation – with anaerobic technology. Anaerobic technology transforms biodegradable organics into the gaseous energy carrier CH<sub>4</sub>, suitable for offsetting energy input from fossil sources.

Several studies have demonstrated the potential of AnMBR for energy recovery from MWW. AnMBR allow overcoming the limitations associated to anaerobic processes applied to MWW treatment (e.g. low growth rate at ambient temperature or poor settling properties of sludge) due to the complete physical retention of the biomass, decoupling therefore hydraulic retention time (HRT) and sludge retention time (SRT). Thus, adopting AnMBR as MWW treatment core technology could take full advantage of the organic matter and its associated energy embedded in wastewater. At the same time, reduction of GHG emissions would be expected when grid energy consumption is replaced by energy recovery provided by this methane.

However, as previously commented, these indisputable benefits present three fundamental challenges: i) the competition between SRB and methanogens for the same substrates reduces methane yields when treating high sulfate influents; ii) dissolved methane losses through the effluent reduce energy recovery and, specially, increase GHG emissions; and iii) membrane fouling increases energy demand (i.e. reduces energy recovery and increases GHG emissions) for keeping a given system productivity while increases cleaning requirements and reduces membrane lifespan (i.e. increases raw materials consumption and associated GHG emissions). Further advancing on these aspects would demonstrate the capability of AnMBR technology for energy resource recovery while reducing carbon footprint in MWW treatment.

### **2.2.1. Sulfate-reducing bacteria**

Under anaerobic conditions, sulfate reduction takes place by using sulfate ions as the terminal electron acceptor. This reaction generates a slightly lower redox potential (-220mV) than other reduced conditions such as methanogenesis (-200 mV)<sup>35</sup>. Sulfate reduction is carried out by different bacterial species called SRB, which can be divided into two groups <sup>36</sup>: heterotrophic SRB and autotrophic SRB. Heterotrophic SRB degrade organic compounds, mainly volatile fatty acids, competing with acetoclastic methanogens for the substrate. Autotrophic SRB use dissolved hydrogen as electron donor competing with hydrogenotrophic methanogens for the substrate.

For high influent sulfate concentrations, SRB usually outcompetes methanogens, as has been demonstrated by several authors. Seco et al.<sup>14</sup> reported a methane yield of 0.11 m<sup>3</sup> CH<sub>4</sub>·kg COD<sup>-1</sup> degraded (i.e. 0.084 m<sup>3</sup> CH<sub>4</sub>·kg COD<sup>-1</sup> removed) in a pilot plant operated at 27° C and fed with sulfate rich MWW. The methane yield determined was clearly lower than the theoretical value of 0.35 m<sup>3</sup> CH<sub>4</sub>·kg COD<sup>-1</sup> because only 18% of the influent COD was converted into methane. Thus, 46% of the influent COD was consumed by SRB, affecting the energy balance. Similar results were obtained by Mei et al.<sup>37</sup> in a laboratory-scale AnMBR plant operated at 35°C and fed with low-strength MWW. The methane yield determined was 0.12 m<sup>3</sup> CH<sub>4</sub>·kg COD<sup>-1</sup> removed because only 30% of the influent COD was converted into methane. Robles et al.<sup>38</sup> reported a methane yield of 0.21 m<sup>3</sup> CH<sub>4</sub>·g influent COD<sup>-1</sup> working in an AnMBR pilot plant operated at 27°C and fed with sulfate rich MWW. This value is higher than the previous ones because of the high influent COD values (over 1200 mg COD·L<sup>-1</sup>). The methane yield and the energy balance do not depend on the influent sulfate concentration but on the COD:SO<sub>4</sub>-S ratio.

Tijani et al.<sup>35</sup> analyzed the performance of a laboratory scale AnMBR plant operated at 36°C and fed with different COD:SO<sub>4</sub>-S ratios and found that methane production decreased when COD:SO<sub>4</sub>-S ratio was decreased and dropped below detection limits at a COD:SO<sub>4</sub>-S ratio of 1.

Methane production and energy balance significantly improve when treating wastewater with low sulfate concentrations. The energy production obtained by Lim et al.<sup>39</sup> working a pilot scale (AnMBR volume of 1.3 m<sup>3</sup>) at ambient temperature (between 13 and 31 °C) rose to 0.254 kWh per m<sup>3</sup> due to the low sulfate concentration (less than 10% of the COD removed was consumed by SRB). Energy consumption was

slightly higher (0.357 kWh per m<sup>3</sup>) but energy neutrality could have been achieved by optimizing gas sparging and AnMBR mixing conditions.

It should be highlighted that influent sulfate concentration not only affects biogas production and composition (the H<sub>2</sub>S produced is corrosive and damages CHP devices) but also deteriorates the anaerobic granular sludge. High sulfide concentrations lead to a reduction in granule size, increasing effluent turbidity and membrane fouling<sup>4</sup> and enhancing the production of SMP and EPS<sup>40</sup>.

Different strategies could be applied to mitigate the adverse effects of high influent sulfate concentrations.

One of the advantages of AnMBR technology is the possibility of recovering nutrients from the permeate.

Therefore, adding iron or aluminium salts to precipitate sulfur as it is usually carried out in anaerobic digesters<sup>41</sup> cannot be applied in AnMBR systems. The sulfur formed should be removed from biogas prior to be used for heat and electricity production. Different techniques such as microaeration, scrubbing systems, adsorption with granular activated carbon, etc... have been applied for biogas desulfurization.

Another strategy consists in combining ANMBR with a primary settler and a mesophilic anaerobic digester as it is explained in section 3.1.2. Effluent COD discharge limits are met by means of SRB in the AnMBR while biogas with a very low H<sub>2</sub>S content is produced in the mesophilic anaerobic digester.

### 2.2.2. Dissolved methane

GHG emissions due to methane stripping from AnMBR effluents is still a bottleneck that limits the full-scale application of this technology<sup>11</sup>. Methane loss becomes especially significant for operation in cold/mild regions since the concentration of dissolved methane in the effluent increases as the temperature decreases<sup>42</sup>.

Working in a bench-scale AnMBR, Smith et al.<sup>43</sup> found that dissolved methane in the permeate stream represented a substantial fraction (40–50%) of the total methane generated working at 15°C. In this study, methane oversaturation increased from 2.2±0.41 at 15°C to 4.1±1.2 at 3°C. For UASB systems treating MWW the degree of supersaturation and the dissolved methane losses range from 1.34 to 6.9 and from 11 to 100%, respectively, whereas for AnMBR systems they range from 1.0 to 1.5 and from 19

to 88%, respectively<sup>42</sup>.

Several technologies have been developed for recovering methane dissolved in AnMBR systems including air stripping, aeration, biological oxidation and degassing membranes<sup>44</sup>. The development of effective dissolved CH<sub>4</sub> management strategies for mainstream anaerobic processes will define their respective future roles in reducing wastewater-associated GHG emissions<sup>45</sup>.

Physical stripping based on gas–liquid equilibrium and mixing with gas or a paddle usually showed low energy efficiency and low energy conversion potential<sup>46</sup>. Of the biological oxidation methods, aerobic methane oxidation (AMO) and aerobic methanotrophs are the most prevalent microbial pathways<sup>47</sup>. In AMO, an electron acceptor (i.e. sulfate or nitrite/nitrate) is required for methane oxidation. However, anaerobic processes generally reduce all present sulfate and do not produce nitrite or nitrate, thus AMO may require an extra step to treat these effluents<sup>48</sup>. Regarding aerobic methanotrophs, Matsuura et al.<sup>49</sup> achieved methane removal efficiencies up to 99% using a two stage closed down-flow hanging sponge reactor (DHS). Nevertheless, although this strategy can prevent GHG emissions, it does not maximize energy recovery since methane is consumed and an energy input for oxygen supply is required<sup>47</sup>.

Degassing membrane (DM) technology for removal of dissolved methane from liquid streams is an interesting approach for the treatment of anaerobic effluents due to the following: (i) direct GHG emissions are prevented, (ii) captured dissolved methane can be potentially employed for energy production, and (iii) relatively high recovery yields are achieved<sup>42,50–52</sup>.

Sanchis-Perucho et al.<sup>48</sup> recovered 80% of the dissolved methane in the effluent from an AnMBR prototype-plant treating MWW by using a hollow-fiber degassing membrane. Maximum methane recoveries were achieved when increasing the vacuum pressure and reducing the liquid flow rate up to 0.8 bars and 50 L·h<sup>-1</sup>, respectively. In another study, Lim et al.<sup>39</sup> reported methane recovery efficiencies of 70 ± 5% from the effluent of a pilot-scale AnMBR using a hollow-fiber membrane contactor.

Recovery of methane using membranes also involves different technical issues, such as the efficiency of recovery, the limitation in mass transfer, and the energy required for the recovery process. Several authors have studied the economic impact of recovering methane from the AnMBR effluents using this

technology at psychrophilic temperatures. Crone et al.<sup>42</sup> estimated that an energy-neutral DM operation could be achieved by using the recovered methane, while Henares et al.<sup>52</sup> showed that it is possible to reach net energy productions. Finally, Sanchis-Perucho et al.<sup>48</sup> reported that the combination of AnMBR and degasification membranes resulted in energy demands and GHG emissions of  $-0.04$  kWh and  $0.113$  kg of CO<sub>2</sub>-eq per m<sup>3</sup> of treated water, respectively, which proved the feasibility of DM for scaling-up AnMBR technology for MWW treatment at ambient temperature.

### 2.2.3. Membrane fouling

A key operating challenge in AnMBRs is to optimize membrane performance whilst minimizing membrane fouling. Membrane fouling is a persistent challenge in AnMBR technology due to the energy inputs to mitigate fouling due to the accumulation of inorganic and organic foulants both internally in membrane pores and externally on the membrane surface. The main membrane foulants include suspended biomass, SMP, EPS, attached biofilm, colloidal matter and chemical precipitates. Fouling reduces system productivity and increases chemical cleaning requirements, thus reducing the membrane lifespan whilst increasing operating expenses<sup>11</sup>. Different operating and control strategies can be applied for fouling mitigation and removal, classically entailing the control of gasification, permeate flux and fouling rate by manipulating membrane scouring (gasification flow and cross-flow velocity), filtration cycles (filtration pressure/permeate flux, permeation and relaxation duration, back-flushing duration and initiation), and chemical cleaning (maintenance and recovery chemical cleaning initiation). Nonetheless, other strategies can be found elsewhere<sup>15</sup>.

Regarding membrane scouring, much effort has recently been focused on optimizing gas sparging intensity<sup>53</sup>, while some studies have reported cleaning protocols with crossflow velocity<sup>54</sup>. Other methods include optimizing the frequency and duration of relaxation and back-flushing<sup>55</sup>. Although increasing the duration and frequency of these stages can improve fouling removal, optimizing these is essential for minimizing energy consumption while maximizing membrane flux. On the other hand, hydrodynamic modifications for membrane fouling mitigation have been also evaluated.

Other studies have evaluated the behavior of different types of membranes in terms of irremovable fouling. Liu et al.<sup>56</sup> evaluated the behavior of polymeric flat-sheet membrane and ceramic flat-tubular membrane when treating MWW and found that polymeric membranes increased irremovable fouling and fouling rate, and that membrane recovery by chemical cleaning was less efficient in ceramic membranes. Jeong et al.<sup>57</sup> also compared the performance of polymeric and ceramic membranes. Despite operating at higher MLSS concentrations, the ceramic membranes had less fouling, even when treating higher SMP and EPS concentrations, resulting in a more stable filtration performance. Besides membrane configuration/type, the reactor configuration can also be optimized to mitigate fouling. For instance, Liu et al.<sup>58</sup> studied membrane fouling in an anaerobic baffled membrane bioreactor with intensified turbulence and reported that cake layer resistance accounted for most of the total membrane resistance ( $92.6 \pm 3.4\%$ ).

Physical cleaning effectiveness tends to decrease over time due to the accumulation of irreversible fouling on the membrane surface, making it necessary to apply chemical cleaning. Mei et al.<sup>59</sup> applied alkali-assisted membrane cleaning protocols (chemically-enhanced backflush (CEB) by an NaOH solution), observing that  $10\text{--}20 \text{ mmol}\cdot\text{NaOH}\cdot\text{L}^{-1}$  could significantly reduce fouling rates (4–5.5 times more than deionized water backflush) and slightly improve methanogenic activity. An economic analysis indicated that  $12 \text{ mmol}\cdot\text{L}^{-1}$  of NaOH was the optimal cost-efficient fouling-control dosage. Foglia et al.<sup>21</sup> evaluated the long-term treatment of high salinity low-loaded MWW at ambient conditions, applying citric acid solution to remove scalants. However, continuous application of chemical cleaning protocols is not recommended since they negatively affect membrane lifespan.

Other chemical and biological techniques have been evaluated for fouling mitigation. For instance, Zhang et al.<sup>60</sup> studied the effect of adding versatile zero valent iron (ZVI) for membrane fouling suppression and found that without an electric field it suppressed membrane fouling by 20% via the improvement of mixed liquor filterability. However, with an electric field it accelerated fouling by 17 times due to the formation of a dense Fe-enriched fouling layer with severe inorganic and organic fouling. Quorum quenching (QQ) has also been proposed against biofouling, although long-term operations need to be further evaluated to



identify comprehensive membrane fouling mechanisms and bioprocess performance. Xu et al.<sup>61</sup> deployed a bead entrapped facultative quorum quenching consortium (FQQ), observing that membrane filtration can be prolonged by an average of 75% due to the reduced mixed liquor EPS concentrations leading to reduced cake layer formation and improved sludge dewaterability. Liu et al.<sup>62</sup> embedded *Microbacterium* sp in alginate beads (known as QQ beads, QQB) and prolonged the life of the system by about 8 – 10 times at constant flux operation, with a much lower SMP concentration and 40 – 81% less cake-layer protein-EPS.

In a recent study, Chen et al.<sup>63</sup> evaluated the effect of different SMP and EPS properties (specific production, molecular weight distribution, and adhesion force) on membrane fouling behavior when treating low-strength wastewater at room temperature. Increasing the organic loading rate (OLR) largely affected the SMP/EPS properties of membrane fouling mechanisms. Fouling was mostly due to SMP-induced pore blockages, while increased OLR resulted in EPS-induced fast-growth and a compact cake layer.

In a study of long-term performance with large membrane filtration areas, Dong et al.<sup>64</sup> demonstrated successful operation at a transmembrane flux of 17 LMH for a period of 536 days, without recovery cleaning or membrane replacement for 178 days. Critical fluxes ranged from 21 to 27 LMH depending on the SRT, which affected MLSS and sludge dewaterability. Robles et al.<sup>38</sup> operated a demonstration plant at ambient temperature with gross transmembrane fluxes above 20 LMH, maintaining low membrane fouling propensities for more than 250 days without chemical cleaning requirements.

Membrane operation at sub-critical conditions significantly reduces membrane fouling and therefore chemical cleanings. However, as commented before, membrane fouling enhances removal of ARBs and ARGs. Further research is needed in order to establish the optimum operating conditions considering both issues. Life Cycle Costs (LCC) and Life Cycle Assessment (LCA) would be needed to elucidate if removal of ARGs and ARBs would pay off the increase in energy consumption (higher transmembrane pressures and higher biogas recycling flow rate are required) in chemical reagents (more frequent chemical cleanings) and the decrease in membrane life span.

#### 2.2.4. Novel configurations for membrane fouling mitigation

Different authors have advanced on novel AnMBR configurations for membrane fouling mitigation when treating MWW. Anjum et al.<sup>65</sup> and Vinardell et al.<sup>66</sup> extensively reviewed and summarized these advances (see also the following section). Table 2 summarizes some of the key characteristics of different novel configurations for membrane fouling mitigation. As previously commented, technological advances for membrane fouling mitigation would allow enhancing the capability of AnMBR technology for energy resource recovery while reducing carbon footprint in MWW treatment.

##### 2.2.4.1. Granular sludge and fluidized elements

The use of scouring agents for mechanical cleaning has appeared as an energy-efficient approach to improve flux and reduce fouling<sup>67,68</sup>, such as granular activated carbon (GAC), polyelectrolytes, coagulants and flocculants<sup>69</sup>, although attention must be paid to possible membrane damage by fluidized elements<sup>70</sup>. A better understanding of the links between anaerobic granular sludge characteristics, operating conditions (feed characteristic, hydrodynamics, SRT and HRT) and fouling mitigation efficiencies is also needed<sup>65</sup>. For instance, special attention should be paid to the time required for granular sludge formation and potential destabilization due to process disturbances.

Aslam et al.<sup>68</sup> used an aluminum dioxide ( $\text{Al}_2\text{O}_3$ ) ceramic membrane in a single-stage anaerobic fluidized bed ceramic membrane bioreactor (AFCMBR), which was continuously operated for more than one year at 25 °C at high fluxes (14.5–17 LMH) with only periodic maintenance cleaning (i.e. adding 25 mg·L<sup>-1</sup> of sodium hypochlorite solution). Chen et al.<sup>71</sup> compared the performance of an external granular anaerobic membrane bioreactor (EG-AnMBR) and a submerged granular anaerobic membrane bioreactor (SG-AnMBR). The EG-AnMBR configuration showed less SMP and EPS concentrations, reducing cake layer resistance and fouling propensity and enhancing granule quality. Chen et al.<sup>72</sup> also compared a conventional granular anaerobic membrane bioreactor (CG-AnMBR) with a sponge assisted-granular anaerobic membrane bioreactor (SG-AnMBR). Sponge addition reduced microbial products (e.g. SMP

and EPS) thus alleviating fouling propensity (51% less total filtration resistance).

Aslam et al.<sup>73</sup> applied Al<sub>2</sub>O<sub>3</sub> flat-tubular ceramic membrane in a staged anaerobic fluidized bed ceramic membrane bioreactor (SAF-CMBR) that reduced energy requirements. GAC was fluidized by bulk recirculation without gas sparging, achieving a net permeate flux of 22 LMH when combined with periodic maintenance cleaning using 25 mg·L<sup>-1</sup> of sodium hypochlorite solution under GAC fluidization.

Filtration cycles and membrane scouring have been also optimized in granular AnMBRs. For instance, Wang et al.<sup>74</sup> studied the impact of peak flow on membrane permeability in an AnMBR pilot plant configured as a granular UASB (G-UASB) system. Good permeability recovery was attained through increasing gas sparging during peak flow (critical flux operation), although longer durations of supra-critical operation could be sustained by reactive fouling control strategies. Wang et al.<sup>75</sup> compared conventional and novel gas sparging regimes in a granular AnMBR in order to achieve sustainable fluxes whilst simultaneously conserving energy demand. Filtration was conducted without gas sparging, followed by gas-assisted membrane relaxation. Long-term fouling transient analysis demonstrated low residual fouling resistance. In a later study, Wang et al.<sup>76</sup> also evaluated the mechanisms underpinning solids breakthrough in a UASB-configured AnMBR to mitigate fouling.

On-line monitoring techniques are needed in granular systems to be effective and energy-efficient. Wang et al.<sup>77</sup> evaluated a non-invasive accelerometer as a potential real-time monitoring technique, and diagnosed the de-fluidization of the GAC due to severe clogging issues, which thereby eliminated the fouling mitigation capacity. However, TMP and flux were not adequately reproduced.

#### 2.2.4.2. Vibration and rotating membranes

Membrane vibration or rotation is used to create high shear or turbulence on the membrane surface.

Mertens et al.<sup>78</sup> proposed a magnetically induced vibration (MMV) system that improved the filtration process by a factor of 13. Combining MMV with backwashing proved to be more effective than relaxation, allowing 10 days of continuous operation at a flux of 20 LMH. Concerning rotating membranes, Ruigómez et al.<sup>79</sup> compared the effectiveness of this shear-enhanced filtration system to conventional gas-sparging

configuration at different permeate fluxes, showing increased fouling prevention from 41–44% (gas-sparging) to 93–96% (rotating HF membrane). Critical flux improved by 17–20% and stable operation at moderate supra-critical gross fluxes (16 LMH) were achieved<sup>80</sup>. Ruigómez et al.<sup>81</sup> later applied membrane rotation during backwashing and relaxation phases. Compared to membrane scouring by gas sparging, the contribution of reversible fouling to total filtration resistance was reduced from 60–53% to 56–18%.

#### 2.2.4.3. Forward osmosis

Forward osmosis (FO) can also be used to mitigate membrane fouling. FO is regarded as a potential low-energy solution where the driving force to send water molecules through the membrane is the naturally occurring osmotic pressure between feed and draw solution, i.e. hydraulic pressure is not required for permeation. Compared to other membrane processes, the main FO advantages include low hydraulic pressure operation, low fouling propensity, easy cleaning and high rejection of a broad range of contaminants<sup>82</sup>. Different configurations can be set for resource recovery from MWW while reducing membrane fouling in AnMBR-based treatment platforms, such as an anaerobic osmotic membrane bioreactor (AnOMBR) and a fertilized drawn FO (FDFO) process<sup>83</sup>.

Wang et al.<sup>84</sup> studied the recovery of permeability of strongly fouled membranes by chemical cleaning in long-term AnOMBR operations, indicating that citric acid, disodium ethylenediaminetetraacetate (EDTA-2Na), hydrochloric acid (HCl), sodium dodecyl sulfate (SDS) and sodium hydroxide (NaOH) had cleaning efficiencies below 15%, while hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) effectively removed 100% of foulants. However, attention should be paid to potential H<sub>2</sub>O<sub>2</sub> damage to the membrane structure.

The influence of fertilizer draw solution properties on the process performance of a side-stream fertilizer-drawn FO + AnMBR system with an FDFO process was evaluated by Kim et al.<sup>85</sup>. Flux decline was very severe due to the absence of aeration and the sticky anaerobic sludge, while the use of fertilizers as the drawn solution seriously affected biofouling in FO via reverse salt flux. Reverse salt flux also had a significant impact on nutrient accumulation in the bioreactor, negatively affecting anaerobic activity.

Other configurations for enhanced process performance have also been analyzed. For instance, Chang et al.<sup>86</sup> operated an UASB–forward osmotic membrane bioreactor (UASB-OMBR) for simultaneous wastewater treatment, membrane fouling reduction, and nutrient recovery. OMBR aimed at replacing the traditional three-phase separator of the UASB technology. Low fouling propensities were achieved, while COD, phosphate and ammonium capture efficiencies were all above 95%.

#### 2.2.4.4. Dynamic membranes

Dynamic membranes have recently been combined with AnMBR (AnDMBR) as a sustainable solution to wastewater treatment due to their low operating and capital cost and reasonable treatment efficiency<sup>87</sup>. Li et al.<sup>88</sup> used mesh filter as support material. Mesh fouling was mitigated by in-situ biogas sparging and the fouling rate was comparable to an equivalent aerobic MBR. Alibardi et al.<sup>89</sup> observed that large mesh pore sizes could operate at high membrane fluxes (approximately 15–20 LMH) applying low TMP (usually lower than 50–100 mbar). Yang et al.<sup>90</sup> reported fluxes between 15 and 20 LMH for a period of 30 days applying TMPs below 200 mbar. Hu et al.<sup>91</sup> reported continuous operation without relaxation at 22.5 LMH, although high OLR accelerated the increasing rate of TMP. Yang et al.<sup>92</sup> operated at short HRTs (2, 4, 8 hours) with fluxes varying from 22.5 to 90 LMH. At these three HRTs, significantly high fouling rates were reached of between 380 and 570 mbar per day.

#### 2.2.4.5. Bioelectrochemical systems

Bioelectrochemical systems (BES) and AnMBR have also been also combined to mitigate membrane fouling. Different approaches have been proposed based on the combination of microbial electrolysis cell (MEC) and AnMBR. In most of these systems, the membrane unit performs as an electrode in different ways: Werner et al.<sup>93</sup> evaluated the use of graphene-coated hollow fiber membrane as cathode; Yang et al.<sup>94,95</sup> used natural organic matter by electro-enhanced carbon nanotube hollow fiber membranes and Sapireddy et al.<sup>96</sup> used nickel-based hollow-fibers.

Hou et al.<sup>97</sup> observed fouling mitigation carried by MEC and reduced flux declines, which was related to

the smoother membrane surface and reduced binding properties with the fouling precursors (e.g., divalent ions). An almost full recovery of flux ( $97.3 \pm 0.7\%$ ) was obtained after each cleaning using tap water SEM images, indicating that biofouling/organic fouling contributed more to membrane flux decline than inorganic scaling. Zhang et al.<sup>98</sup> reported lower membrane fouling propensities in a forward osmosis anaerobic membrane bioreactor (AnOMBR) coupled with a microbial electrolysis cell (AnOMEBR), increasing filtration time by about 27%. Ding et al.<sup>99</sup> observed that the membrane fouling rate gradually slowed down when increasing the applied voltage, extending the membrane fouling cycle from 60 to 98 hours. The Protein-EPS to polysaccharide-EPS ratio in the sludge decreased when the applied voltage was increased, which was identified as a key factor affecting membrane fouling.

### **2.3. Biosolids production and recovery**

As commented above, P and N recycling from waste to farmland would alleviate different problems. P is an essential element, irreplaceable for crop production, whose reserves are expected to decrease in the near future and are often obtained from unstable countries. N recovery would reduce worldwide energy consumption by reducing fertilizer production<sup>100</sup>.

The feasible management of the biosolids produced is a major concern in the operation of WWTPs. On a global scale, daily production of sludge varies widely from 35 to 85 g dry matter (dm) per capita per day and can even reach values of 125.5 g dm per capita per day<sup>101</sup>. Due to its generation rate and nature, adequate treatment of biosolids is necessary to avoid undesirable environmental effects as it contains nutrients which can lead to eutrophication and hazardous substances (heavy metals, pathogens and persistent organic pollutants) and has a high degree of biological activity<sup>102</sup>. However, although biosolids have been traditionally considered a residue, they can also be understood as a supply of resources, thus replacing an equivalent amount of materials/energy that would otherwise need to be produced from non-renewable resources with a considerable environmental impact<sup>103</sup>. Reuse of stabilized sludge in agriculture is the most widespread application due to the recoverable sludge nutrient content: according to 2015 Eurostat data. Approximately 40% of the total sludge produced in the EU is used in agriculture

and is currently mostly regulated by limits on the heavy metals (Cd, Cu, Hg, Ni, Pb and Zn) listed in Council Directive 86/278/EEC. This directive is now outdated and does not reflect the current needs for ensuring the safety of sludge application in agriculture<sup>104</sup>. For this reason, some European Member States have set their own limitations on pollutant concentrations (e.g. heavy metals or persistent organic pollutants) and pathogens, while others abandoned sludge disposal in agriculture due to its high environmental risk<sup>103</sup>, which caused great differences among the different EU countries. For example in 2010 the United Kingdom, Denmark, France, Belgium and Spain used more than 50% of sludge in agriculture, while countries like Netherlands, Greece, Romania, Slovenia and Slovakia used none at all<sup>105</sup>. On the other hand, the presence of ARG, ARB<sup>106</sup> and microplastics<sup>107</sup> in biosolids is raising concerns due to their effects on the safe land application perspectives.

Anaerobic treatments usually produce less biosolids to be handled than aerobic-based systems due to the low growth yield of the anaerobic biomass, which has an interesting advantage for reducing operating costs and the carbon footprint. Pretel et al.<sup>108</sup> compared different aerobic-based schemes to an AnMBR coupled with a post-treatment applied to the removal of organic matter, N and P from MWW. They found that AnMBR combined with conventional activated sludge (CAS)-based post-treatment for nutrient removal reduced the environmental impact (global warming potential (GWP), abiotic depletion, acidification, etc.), energy demand (0.04kWh per m<sup>3</sup>), sludge production and sludge handling and disposal costs. In another study, Seco et al.<sup>14</sup> working with an AnMBR pilot plant treating low-loaded MWW at 27 °C and 140 days of SRT the sludge production was 0.218 kg VSS per kg COD removed. Recent AnMBR studies have obtained very low sludge production values of  $0.136 \pm 0.054 \text{ kg VSS} \cdot \text{kg}^{-1} \text{ COD}_{\text{in}}$  when operating at a 70-day SRT and 40 h HRT, which is approx. 36–58% of that produced aerobically<sup>38</sup>.

An AnMBR operated at a high SRT allows onsite stabilization of biosolids and avoids subsequent sidestream treatment of waste sludge<sup>1</sup>. Seco et al.<sup>14</sup> working with an AnMBR pilot plant treating low-loaded MWW at 27 °C and a 140-day SRT produced highly stabilized sludge with only 11% of anaerobic digestibility, indicating that at this high SRT almost all the biodegradable organic matter was degraded

during the anaerobic treatment.

According to its volatile organic contents, which ranges from 21–48%, the energy content of dried sewage sludge varies between 11.10–22.10 MJ·kg<sup>-1</sup>, which indicates similar or even higher calorific values than lignite and various biomass samples<sup>109</sup>. In this respect, novel technologies for energy recovery from biosolids emerge as better options from an environmental viewpoint and now have increased interest within the EU framework<sup>110</sup>. These methods can reduce sludge volume and possibly recover resources (e.g. nutrients). Of these thermal technologies, pyrolysis and gasification are promising alternatives but still involve high costs related to feedstock moisture reduction and additional product upgrading<sup>111</sup>. Hydrothermal carbonization is an emerging technology for sustainable sludge treatment consisting of converting sludge into carbonaceous products (hydrochar) at typical temperatures around 180–250 °C under autogenously saturated pressures (10-50 bar) for several hours together with byproducts, including a large amount of liquid phase (avoiding the need for a previous drying step) and a small amount of gas (mainly CO<sub>2</sub>)<sup>102,111</sup>. The hydrochar obtained has improved dewaterability and better stability and sanitation characteristics for disposal than biosolids.

### **3. AnMBR-based platforms for resource recovery**

Figure 2 shows some of the possible pathways for carbon and nutrient recovery from different AnMBR-based platforms to be configured based on the combination of technologies described in the following sections.

#### **3.1. Enhancing carbon recovery**

Novel strategies for carbon recovery have been evaluated in the last years, including co-digestion of different substrates (i.e. MWW and food waste), particulate organics concentration/recovery in primary treatment units, and forward osmosis as pre-concentration step. These strategies allow enhancing energy recovery from MWW due to both organics concentration/recovery and minimization of dissolved methane losses. Moreover, some of them allow improving nutrient capture associated to particulate



organics recovery. However, special attention should be taken to other aspects that could hinder carbon/energy recovery in these configuration, such as increased membrane foulants concentrations in the mixed liquor due to more complex characteristics of the feeding to the AnMBR.

### 3.1.1. Co-digestion

Anaerobic treatments such as AnMBR have classically been applied to treating high-loaded streams (industrial waste streams or thickened sludge in municipal WWTPs). MWW often has a low organic carbon content and so, is not ideal for anaerobic treatment<sup>112</sup>. To overcome this drawback, several options have been explored and developed to increase the MWW organic load. One of these solutions consists of co-digesting MWW with other high-strength waste streams such as liquid food waste<sup>113,114</sup>. According to Becker Jr. et al.<sup>115</sup>, a positive net energy balance could be achieved for AnMBRs at 40% food waste diversion. The co-digestion of MWW and 100% of food waste would produce around 0.5 kWh per m<sup>3</sup> of treated water. These results agree with those obtained by Moñino et al.<sup>116</sup> in a pilot plant where methane production was significantly increased (over 100%) when 80% of food waste and sulfate-rich MWW were co-digested. The relative abundance of bacterial groups with high hydrolytic and fermentation potentials (*Chloroflexi*, *Firmicutes*, *Proteobacteria* and *Synergistetes*) as well as acetoclastic methanogens increased significantly due to adding food waste to the AnMBR pilot plant<sup>117</sup>. Pretel et al.<sup>108</sup> estimated a positive net energy balance for an AnMBR treating low sulfate wastewater with food waste based on pilot plant results. Also on a pilot scale, Jeong et al.<sup>118</sup> evaluated the performance of an AnMBR treating MWW jointly with food waste recycling wastewater (wastewater obtained after grinding the food waste from restaurants, grocery stores, food processing plants, etc.). The COD removal efficiency was higher than 98%. Net energy balance was improved by adding polyvinyl alcohol-gel beads to alleviate membrane fouling and reduce gas sparging, which is closely associated with the energy demand in AnMBR systems. Amha et al.<sup>119</sup> observed that better performance (higher methane production and biomass diversity) can be achieved with a two-phase than a single-phase AnMBR when treating MWW and food waste mix.

### 3.1.2. Primary settling and chemical enhanced primary treatment

The combination of a primary settling (PS) step and further anaerobic digestion (AD) of the wasted sludge with AnMBR-based mainline treatment may widen the applicability of AnMBR to low temperature climates and high-sulfate MWW treatment. Primary settling reduces the total suspended solids (TSS) and the COD:SO<sub>4</sub>-S ratio in the influent of the AnMBR system, enhancing energy recovery through side-stream AD of the primary sludge. Since methane is mainly produced in the side-stream AD, a high-quality, methane-free effluent is produced in the AnMBR.

Pretel et al.<sup>120</sup> compared different treatment schemes (AnMBR, AnMBR + AD, PS + AnMBR + AD) in different operating scenarios: sulfate-rich and low- sulfate MWW treatment at 15 and 30 °C. The authors showed that the anaerobic treatment of sulfate-rich wastewater at ambient temperature could be enhanced by including a primary settling stage prior to an AnMBR, revealing that the combination PS + AnMBR + AD could result in an energy-neutral solution. In this treatment platform, methane is produced in a sidestream AD that can be operated at an increased temperature (e.g. 35 °C) by using the heat energy recovered from the biogas produced in the system. Since the COD:SO<sub>4</sub>-S ratio entering the sidestream AD is much higher than the one entering the mainstream AnMBR, the growth of methanogens is favored, thus increasing overall energy recovery potential. In addition, since methane is not produced in the mainstream AnMBR (operating at ambient temperature and high flow rates), it is possible to drastically reduce the methane dissolved in the effluent. Seco et al.<sup>121</sup> also evaluated a treatment platform combining primary settling and side-stream AD with mainstream AnMBR for resource recovery from sulfate-rich sewage at ambient temperature. The proposed platform produced a high-quality solid- and coliform-free effluent suitable for nutrient recovery while energy recovery was maximized.

### 3.1.3. Forward osmosis

Several authors have proposed the pre-concentration of sewage by means of FO<sup>122-124</sup> to reduce volumetric flow rates and dissolved methane losses. On the other hand, organics concentration by FO

could widen AnMBR potential for resource recovery, i.e. intermediate products (VFA and H<sub>2</sub>) could also be considered an economically attractive and environmental-friendly option <sup>125</sup>.

The performance of a mesophilic AnMBR at different FO pre-concentration factors has been recently evaluated by Vinardell et al.<sup>126</sup>. The results showed that the methane yield progressively increased from 214 ± 79 to 322 ± 60 mL CH<sub>4</sub>·g<sup>-1</sup> COD as the pre-concentration factor was increased from 1 to 10, highlighting that pre-concentration factors above 10 are an option to make AnMBR economically self-sufficient.

Since one drawback of FO for sewage pre-concentration is its dependence on the availability of draw solution, several authors have focused on selecting the draw solution<sup>127</sup>. In coastal areas seawater has been studied as an open-loop draw solution, but major research is needed to reduce the diffusion of NH<sub>3</sub>-N to prevent the potential environmental impact in coastal areas after seawater discharge. In order to maximize resource recovery, FO should be combined with reverse osmosis (RO) for water recycling. The use of artificial solutions such as draw solutions (NaCl, MgCl<sub>2</sub>, etc.) in FO involves a regeneration step to produce clean water while re-concentrating the diluted draw solution. RO is the most frequently used technology to produce regenerated water from diluted draw solutions<sup>83</sup>. However, the high RO energy requirements (between 1.6 to 2.0 kWh per m<sup>3</sup> of water produced) have been reported <sup>124</sup>. The minimum treatment cost determined by Vinardell et al.<sup>124</sup> for an FO/RO + AnMBR combination was estimated at €0.81 per m<sup>3</sup> by limiting FO recovery to 50% in a closed-loop scheme, although higher costs were estimated when it was set to 80 and 90% (€1.01 and €1.27 per m<sup>3</sup>, respectively). It is therefore necessary to optimize this technology to make this solution economically competitive.

FDFO is another approach to concentrate organics while recovering water for fertigation. For instance, Li et al.<sup>128</sup> evaluated the effect of reverse salt flux on the production of methane when using KCl, KNO<sub>3</sub> and KH<sub>2</sub>PO<sub>4</sub> as draw solutions, KH<sub>2</sub>PO<sub>4</sub> being the lowest reverse salt flux in terms of molar concentration. However, bio-methane production showed an opposite trend with KH<sub>2</sub>PO<sub>4</sub>, followed by KCl and KNO<sub>3</sub>. Li et al.<sup>128</sup> observed that reverse salt flux generally had a negative effect on bio-methane production, probably by influencing the sludge bacterial community via environmental modification.

Operating temperature also plays an important role when maximizing energy recovery from pre-concentrated sewage. Combining FO, AnMBR and sewer mining, Ferrari et al.<sup>123</sup> studied the effect of temperature (34, 23, 17 and 15 °C) for a period of 11 months of treating synthetic wastewater that mimicked the concentrate of an FO process treating sewage with 80% water recovery. COD removal efficiency was 95, 87, 76 and 67% at 34, 23, 17 and 15 °C, respectively, reducing the energy recovery potential accordingly.

Another issue to take into account is the possible inhibition of the process. For instance, Gao et al.<sup>129</sup> showed that COD concentration efficiencies of 80% can be achieved when treating concentrated sewage by FO membrane. However, the process was inhibited as the concentration of  $\text{NH}_4^+\text{-N}$  and NaCl increased.

In a complete review, Ansari et al.<sup>122</sup> summarized and reviewed all the relevant works related to the application of FO-based processes to improve energy and nutrients recovery from several wastewater streams (i.e. synthetic wastewater, secondary effluent, activated sludge, digested sludge, among others), while Vinardell et al.<sup>66</sup> reviewed different open-loop and close-loop configurations for the cost-effective integration of FO and AnMBR. The findings revealed that major research is needed to optimize joint FO and anaerobic processes for biogas production while overcoming salinity accumulation and membrane fouling issues.

### **3.2. Capture, concentration and recovery of inorganic nutrients**

AnMBR mineralizes organic N and P in the form of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  while negligible N and P removals (only microbial assimilation for biomass growth) are expected in these systems. AnMBR also has an intrinsic advantage: the inherent retention of solids due to membrane filtration, which produces excellent permeate qualities that facilitate nutrient concentration and recovery by different post-treatment processes. As commented above, combining AnMBR with water reclamation by fertigation would alleviate not only water scarcity but also the dependence on chemical fertilizers. When fertigation is not possible, AnMBR can be combined with a complementary post-treatment process for nutrient

concentration and/or recovery<sup>130</sup>, e.g. phototrophic-based technology<sup>121,131,132</sup>, membrane-based technology<sup>133</sup> or ion exchange<sup>46,134,135</sup>.

### 3.2.1. Phototrophic-based technology

Photosynthetic-based technologies such as photobioreactors (PBR) or open ponds have emerged as a sustainable and cost-effective alternative<sup>136</sup> for nutrient recovery/removal from AnMBR effluent.

Microalgae are photosynthetic microorganisms which use sunlight as energy to grow, consuming inorganic nutrients and CO<sub>2</sub>. Different studies confirm that microalgal cultures successfully uptake N and P from different wastewater types<sup>137</sup> such as secondary effluent, primary clarifier effluent, AnMBR effluent, and AD supernatants. Indeed, several authors have demonstrated that the combination of AnMBR and microalgae cultivation is a win-win strategy, since it would be feasible to recover both nutrients and other resources such as energy and water from wastewater<sup>138</sup>.

Much research is focused on the study of pure microalgae cultures for wastewater treatment. However, in large-scale outdoor microalgae cultivation systems, microalgae often coexist with other microorganisms. The competition between microalgae and ammonium oxidizing bacteria (AOB) for ammonium uptake especially reduces microalgae N uptake<sup>139</sup>. González-Camejo et al.<sup>131</sup> studied the effect of ambient temperature on an indigenous microalgae-nitrifying bacteria culture dominated by *Chlorella*. They observed that at high temperatures (30–35 °C) microalgae viability was significantly reduced due to the triggered AOB growth in the indigenous microalgae culture, which deteriorated microalgae performance, especially when AOB activity made the system ammonium-limited.

Previous studies have shown that algae PBR could remove 34%–67% of ammonia and >75% of phosphate from the AnMBR permeate, even under harsh conditions (such as in the presence of antibiotics)<sup>140</sup>. In addition, several studies have found that microalgae-based wastewater treatments are strongly affected by many factors<sup>137,141</sup>, some of them related to environmental conditions (outdoor/indoor, light, temperature, etc.), wastewater characteristics (C/N/P ratio, nutrient levels, toxics, etc.) and others associated with operating conditions (batch/continuous, CO<sub>2</sub> addition, hydraulic and

solids retention time, mixing, pH, etc.) and reactor configuration (light path, open systems, PBR, attached-growth, etc.). According to several authors, light intensity is a key parameter in microalgae cultivation<sup>142-144</sup>. Besides this factor, light frequency and photoperiods have also been reported to influence microalgae productivity and nutrient capture efficiency<sup>145</sup>. The optimum light path therefore needs to be defined separately in each microalgae cultivation system, i.e. in each PBR design.

PBRs for microalgae cultivation are classified into open systems, closed PBR and newly designed hybrid PBR<sup>146,147</sup>. The most frequently used system for wastewater treatment is the open pond reactor, which consists of a shallow depth carousel (15–40 cm) in which the water is recirculated by paddlewheels<sup>148</sup>. The final objective of the design is to achieve both cost-effective and high-efficiency wastewater treatment. In this respect, hybrid systems have been proposed such as membrane PBRs that can operate at shorter HRTs and longer SRTs, enabling higher efficiencies<sup>149</sup>. Membrane filtration appears as one of the most competitive separation methods for microalgae harvesting<sup>150</sup>.

The application of photosynthetic bacteria (PSB) to wastewater treatments is a novel technology that can treat both wastewater streams and recycling nutrients<sup>151,152</sup>. Using light as the energy source, PSB can simultaneously assimilate carbon and nutrients at high efficiencies (with biomass yields close to unity, i.e. 1 g COD-biomass·g<sup>-1</sup> COD removed), facilitating the maximum recovery of these resources as different value-added products<sup>153</sup>.

PSB-based treatment processes have been evaluated for the treatment of different types of industrial wastewaters (e.g. vinasses, breweries). All of these are typically highly organic-loaded and easily biodegradable wastewaters, usually generated from food and alcohol production processes. A key future challenge to apply PSB to MWW treatment is the need for more favorable C:N:P ratios such as those found in industrial wastewaters<sup>152</sup>. In this respect, the degree to which external carbon needs to be added in a continuous process to achieve full nutrient capture needs to be identified<sup>154</sup>. Finally, the relatively high capital and operating costs of PSB cultivation make it necessary to recover highly value-added products contained within, excreted by, or synthesized from the PSB biomass<sup>153</sup>. Capson-Tojo et al.<sup>153</sup> showed the contents of the potential value-added products within the PSB biomass, which were in the

following order: crude protein, followed by methane and PHB and finally, CoQ10 and carotenoid.

### 3.2.2. Membrane-based technology

Membrane technology can be used to concentrate and/or recover nutrients from AnMBR effluents. For instance, FO, hollow-fiber membrane contactors (HFMC), electrodialysis (ED), or bioelectrochemical (BES) systems are highly selective on N and/or P and can replenish biofertilizer resources<sup>155</sup>.

#### 3.2.2.1. Bioelectrochemical systems

BES such as microbial fuel cells (MFCs) can also be applied to nutrient concentration and recovery from diluted streams<sup>156</sup>, and MFCs for MWW treatment have garnered increasing interest in terms of attaining energy self-sufficiency due to their theoretical superiority to conventional processes<sup>157</sup>. However, there are several factors that must be further assessed for full-scale implementation combined with AnMBR, such as carbon handling in the liquid stream, power generation, system scalability, or capital cost. Regarding other anaerobic systems for MWW treatment, Nakhate et al.<sup>158</sup> evaluated the continuous operation of a semi-pilot scale BES consisting of a holistic blending of up-flow anaerobic sludge blanket (UASB) and MFC, while hollow fiber ultrafiltration membrane was incorporated in a series. 91.3% COD and 54.4% total dissolved solids removals were achieved during integrated operations, while a theoretical net energy production of around 0.1 kWh per m<sup>3</sup> was reached, which exceeded the system's overall energy requirements.

Ardakani and Badalians Gholikandi<sup>159</sup> recently reviewed the state-of-the-art of MFCs combined with anaerobic processes and membrane bioreactors for simultaneous wastewater treatment and energy recovery. However, despite the potential advantages of MFCs and the encouraging results obtained on a laboratory-scale plant, the low power density and high costs of the initial investment and operation limit their commercial application. Maaz et al.<sup>160</sup> reviewed novel configurations, fouling control and energy considerations in AnMBRs for wastewater treatment, including different studies coupling MFCs with AnMBRs to improve effluent quality. From this review, some authors recommended a novel configuration

of MEC with anaerobic filtration, recovering energy and treated water in single stage, although high membrane fouling propensities were observed. Scouring action due to gas formation on the surface of metallic membranes and using fluidized beds were identified as membrane fouling mitigation strategies. Other configurations were also reviewed, such as anaerobic membrane bioelectrochemical reactors in which membrane worked as cathode and filter media in MFC. In this case, high effluent quality and large bioenergy productions were achieved, while membrane fouling would be minimized due to anode oxidation of substrates that has low organic loading during membrane filtration. Li et al.<sup>161</sup> evaluated the performance of a cathodic fluidized granular activated carbon assisted-membrane BES reactor for wastewater treatment. Although N concentration was not a key focus in this study the BES system was expected to provide a solution for nutrient concentration/recovery.

Besides nutrient recovery, combining BES with AnMBR to produce electricity has recently been shown to reduce the overall energy demand of wastewater treatment, e.g. Chen and Smith<sup>47</sup> combined AnMBR with methane-driven MFCs to recover energy and mitigate dissolved methane emissions. Two replicate air-cathode, single-chamber MFCs and one dual-chamber MFC were operated in continuous mode at 20 °C and 4, 8, and 16 h HRT. Up to 85% of the dissolved methane was removed, generating  $0.55 \pm 0.06$  V.

#### 3.2.2.2. Other hybrid configurations

Other membrane-based processes have shown the potential for nutrient concentration and recovery from wastewater<sup>162</sup> and can also be used for recovering nutrients from AnMBR effluents. For instance, electrodialysis (ED) can selectively concentrate nutrients from wastewater streams into high quality nutrient products by positioning ion-exchange membranes alternately in a direct current field. The combination of different membrane-based technologies can also further enhance the efficiency of hybrid AnMBR systems for nutrient recovery.

The combination of a direct contact membrane distillation (MD) unit with an AnMBR to simultaneously recover energy and produce high quality water for reuse from wastewater was studied by Song et al.<sup>163</sup>. By integrating MD with AnMBR, bulk organic matter and phosphate were almost completely removed, as



well as several trace organic contaminants. Other examples of combining MD and AnMBR can be found elsewhere <sup>160</sup>.

Gao et al.<sup>164</sup> evaluated the performance of inorganic draw solution concentrations in an AnOMBR and obtained high capture efficiencies: more than 96% of COD, 88% of ammonia-N, 89% of total N, and almost 100% of total P. Zhang et al.<sup>98</sup> compared the performance of an AnOMBR to the performance of an AnOMBR coupled with a microbial electrolysis cell (AnOMEBR) for energy recovery and membrane fouling alleviation. The system treated synthetic wastewater while magnesium acetate was used as the draw solution. Methane production in AnOMEBR was around 111% methane production in AnOMBR and it was possible to compensate for the extra AnOMEBR energy input. Hou et al.<sup>97</sup> incorporated a microbial electrochemical unit into an AnOMBR for enhanced recovery of biogas, N, and P, while maintaining high effluent quality with low dissolved methane. This combination provided high P concentration efficiencies (around 99%) and adequate phosphorous and ammonium recovery efficiencies (65 and 45%, respectively).

Giwa et al.<sup>165</sup> reviewed recent developments in membrane bioreactors and electrochemical processes for treating different wastewaters, and highlighted the combination of a low-voltage electric field with AnMBRs and OMBRs could improve the performance of these systems in terms of effluent quality. Yang et al.<sup>166</sup> published a comprehensive review of nutrient-energy-water-solute recovery by hybrid osmotic membrane bioreactors and also included a hybrid AnOMBR combined with BES.

### 3.2.3. Ion exchange

Ion exchange (IEX) processes are a promising cost-effective alternative to concentrate and recover nutrients from MWW by interchanging ions of like valences attached to a solid phase medium. Once the media is depleted, it can be regenerated by a regenerant brine solution, thus recovering the ions in a concentrated stream that can be used to produce nutrient-based commercial products (e.g. struvite and ammonium salts). However, IEX processes are rarely applied in MWW mainly due to media selectivity, bed clogging, and costly regenerations<sup>167</sup>. Nevertheless, nutrient recovery by IEX from the effluent of an

AnMBR process is gaining momentum since AnMBR produces a solids-free high-quality permeate, which can deal with some of the above-mentioned limitations.

Liu et al.<sup>168</sup> evaluated the adsorption and regeneration of P mechanisms from the effluent of an AnMBR by IEX. IEX exhibited stable total P capture efficiency (up to 81.6%) and high long-term exchange/regeneration capacity, although enhanced regeneration protocols are needed for humic acid removal. Zhu et al.<sup>169</sup> investigated the mechanisms of P capture, showing that 88.6% of phosphorous could be captured with a resin dosage of 10.0 g·L<sup>-1</sup>. The experimental regeneration results showed that 0.4 mol·L<sup>-1</sup> NaCl would be needed for effective resin recovery. Smith et al.<sup>170</sup> addressed the performance of an AnMBR including a post-treatment IEX-based N recovery, using a clinoptilolite ion-exchange column for ammonia capture, achieving N capture efficiencies above 99.5% in each run and average effluent concentrations of 0.05 ± 0.05 mg·N·L<sup>-1</sup> (influent N concentration was 37 ± 4 mg·N·L<sup>-1</sup>).

Mullen et al.<sup>171</sup> used IEX media loaded with hydrated ferric oxide (HFO) and copper (Cu<sup>2+</sup>) to selectively capture phosphate, followed by clinoptilolite for ammonium capture and recovery. Regeneration through five IEX cycles using relatively low concentration regenerant solution resulted in the highest phosphate recovery efficiency and the most consistent ammonium recovery. Up to 94% of the removed phosphate was recovered during IEX regeneration. However, although capture and recovery of NH<sub>4</sub><sup>+</sup> was achieved, less than 50% of NH<sub>4</sub><sup>+</sup>-N was recovered during IEX regeneration. Huang et al.<sup>167</sup> performed a modeling evaluation by upscaling a 10,000 population equivalent AnMBR + IEX and compared it with a traditional biological nutrient removal (BNR) plant + iron dosing and a CAS process + IEX. The results showed that all the treatment platforms evaluated could achieve the desired effluent quality of COD < 20 mg·L<sup>-1</sup>, NH<sub>4</sub><sup>+</sup>-N < 1 mg·L<sup>-1</sup> and PO<sub>4</sub><sup>3-</sup>-P < 0.5 mg·L<sup>-1</sup>. Overall, the AnMBR + IEX represented an interesting approach for optimizing biogas sparging for membrane scouring, thus minimizing energy demands and operating costs while keeping high performance efficiencies.

Gu et al.<sup>135</sup> proposed an innovative AnMBR-reverse osmosis-IEX treatment platform for resource recovery from MWW. In this integrated process, nearly 76.8% of influent COD was converted to methane (energy output of 0.41 kWh per m<sup>3</sup> wastewater treated was achieved). Although most of N and P was

concentrated by the reverse osmosis unit, IEX was used as an additional low-energy polishing technology, producing an effluent that met quality requirements for industrial and indirect drinking water uses.

Li et al.<sup>172</sup> compared an anaerobic fluidized-bed membrane bioreactor (AFMBR) with zeolite adsorption and reverse osmosis (RO) with an anoxic-aerobic MBR coupled with RO, both platforms for MWW reclamation. The zeolite column was used to remove ammonia in the AFMBR permeate, which ensured comparable organic and N levels in the feeds to RO units in the two processes.

Finally, other configurations based on IEX technology can be defined for N concentration previous to an AnMBR process. In this respect, Zhou et al.<sup>173</sup> evaluated mainstream N capture by IEX as a previous step to an aerobic membrane bioreactor (MBR). The IEX unit was applied after a coagulation-sedimentation unit for carbon and P recovery. The results showed that around 90% of the influent N to the IEX unit could be removed, reducing the influent load to the MBR.

#### **4. General remarks**

##### **4.1. Energy, cost and environmental considerations**

Table 2 summarizes key aspects of the different AnMBR-based platforms for MW resource recovery. Although different benefits can be achieved by combining different technologies aimed to recover nutrients in a whole-plant platform around AnMBR technology, life cycle analysis (LCA) and life cycle costing (LCC) are needed to fully understand the economic and environmental impacts of each combination/technology towards full-scale implementation. Different aspects related to energy demand, LCC and LCA of different AnMBR configurations have been reviewed and can be found elsewhere<sup>15,160,174,175</sup>.

Harclerode et al.<sup>176</sup> conducted an LCA and LCC to holistically compare multiple AnMBR-based MWW treatment platforms with CAS treatment that included different scouring methods for membrane fouling mitigation (gas sparging and GAC fluidization), upstream treatment considerations (primary settling vs. screening only), downstream treatment (dissolved methane recovery, and sulfide and P removal) and

land application of biosolids. The authors found that chemical P removal significantly increases LCC due to the chemical used and residual generation, and also affected AnMBR-based platform sustainability, showing that nutrient recovery platforms need to be optimized. On the other hand, primary settling and dissolved methane recovery can significantly enhance net energy recovery, while it is necessary to operate at both high transmembrane fluxes and room temperature to reduce the environmental impact. Pretel et al. <sup>177</sup> also compared the economic and environmental sustainability of different AnMBR platforms for MWW treatment with CAS-based systems and showed that AnMBR with CAS-based post-treatment for nutrient removal is a sustainable option for MWW treatment at ambient temperature, resulting in low LCC and reduced environmental impact (GWP, abiotic depletion, acidification, etc.). Cogert et al. <sup>178</sup> assessed the combination of denitrifying Methanotrophs (n-damo), anammox, and AnMBR for reducing costs and environmental impacts of wastewater treatment. The authors compared the cost and environmental impact of traditional nitrification/denitrification relative to AOB/anammox and AOB/anammox/n-damo systems with and without AnMBR. The results highlighted that AnMBR implementation reduced costs and emission rates at moderate to high nutrient loading by lowering aeration and sludge handling demands while increasing the methane available for cogeneration. Indeed, AnMBR/AOB/anammox systems reduced cost and GHG emission by up to  $\$0.303 \cdot d^{-1} \cdot m^{-3}$  and  $1.72 \text{ kg CO}_2 \text{ eq} \cdot d^{-1} \cdot m^{-3}$ , respectively, while AnMBR/AOB/anammox/n-damo resulted in similar savings. However, AOB/anammox does not allow nutrient recovery and thus reduced resource recovery capability, indicating that nutrient recovery alternatives are needed to maximize resource recovery from AnMBRs platforms and improve economic and environmental aspects.

An integrated modeling approach was used by Lin et al. <sup>179</sup> to evaluate the holistic economic and environmental profiles of three alternative nitrogen removal and recovery pathways, i.e. conventional nitrification–denitrification, Anammox, and anaerobic IEX. The authors concluded that IEX is a promising technology showing high MWW N recovery efficiencies while delivering competitive economic and sustainability scores. Nonetheless, the authors observed that there is considerable room for improvement in terms of process design and IEX optimization to achieve economically and environmentally optimal

performance.

Regarding pre-concentration of MWW, Vinardell et al.<sup>126</sup> evaluated the economic viability of combining FO and AnMBR operating at different pre-concentrated factors. The preliminary energy and economic analysis showed that, at a pre-concentration factor of 10, the AnMBR temperature could be increased by 10 °C and achieve a positive net present value of 4 M€ for a newly constructed AnMBR treating 10,000 m<sup>3</sup>·d<sup>-1</sup> of pre-concentrated MWW, highlighting that MWW pre-concentration stands as an option to improve the economic feasibility of AnMBR. Hamid et al.<sup>180</sup> evaluated the integration of FO in AnMBR technology from the economic, energy and carbon footprint perspectives. Three platforms for water reclamation were compared, i.e. FO-MBR, FO-MBR integrated with RO, and FO-AnMBR integrated with partial nitrification/anammox (PN/AMOX) process. Low treatment costs were estimated for FO-AnMBR compared to FO-MBR integrated with RO and FO-MBR due to the good FO rejection performance and its ability to concentrate wastewater. GHG emissions from the third platform were 1.5 and 4.1 times lower than the first and second. However, further research is needed to evaluate if this integration could be a feasible AnMBR-based technology for the next generation of wastewater treatment.

Concerning co-digestion, Pretel et al.<sup>108</sup> and Becker Jr. et al.<sup>115</sup> evaluated the economic and environmental feasibility of co-management of MWW and food waste. Both concluded that co-digestion of MWW and food waste provides synergies that could improve the environmental impact of conventional treatment. Becker Jr. et al.<sup>115</sup> highlighted that AnMBR offers slightly higher energy recovery in its current state of development than other conventional technologies and has the greatest potential to minimize environmental impacts if technological developments are achieved prior to full-scale implementation (e.g. minimizing energy demand for membrane fouling control/mitigation and effectively recovering dissolved methane from the effluent). Pretel et al.<sup>108</sup> concluded that AnMBR technology is likely to be a net energy producer, resulting in considerable cost savings (up to €0.023 per m<sup>3</sup> of treated water) when treating low-sulfate MWW. Regarding LCA results, enhanced environmental performance was determined, reducing environmental impact through abiotic depletion, GWP, and marine aquatic ecotoxicity.

Robles et al.<sup>38</sup> observed in a study of AnMBR implementation at high technology readiness levels (TRL)

that net energy production with negligible GHG emissions (as long as dissolved methane is captured) was possible in demonstration scale AnMBRs operating at ambient temperature. The high quality of the nutrient-loaded permeate would make it possible to use the effluent for fertigation. Fertigation can reduce energy consumption for inorganic ammonia-based fertilizer production and P extraction. The authors estimated that when accounting for the energy savings derived from using the AnMBR effluent for fertigation instead of inorganic fertilizers the net energy demand of the AnMBR would be reduced by 0.95 kWh per m<sup>3</sup> of water reused and, regarding the carbon footprint, the AnMBR GHG emissions would be reduced by 0.30-0.37 kg CO<sub>2</sub> per m<sup>3</sup> of water reused.

Besides methane and nutrient recovery, other aspects to take into account for upscaling AnMBR technology to full-scale are the treatment scale, the population density, and the climate. Cashman et al.<sup>181</sup> calculated the energy and greenhouse gas life cycle and cost profiles of AnMBR and aerobic MBR technology, taking into account the above-mentioned factors. An AnMBR operated at psychrophilic temperature condition provided net energy benefits in all the scenarios evaluated, emphasizing the need to operate at ambient temperature. Indeed, the AnMBR GWP and energy demand were found to be highly sensitive to reactor temperature and climate.

#### **4.2. Perspectives towards full-scale implementation**

There are now several examples of successful full-scale AnMBR plants for treating different industrial wastewaters, mainly in the food industry (see e.g. <https://www.thembrsite.com/search/?q=anmbr>) because wastewater from food processing plants contains high contents of biodegradable organics and low contents of inhibitory compounds for anaerobic microorganisms.

Several pilot-plant applications of MWW can be found in literature, including an interesting review by Shin and Bae<sup>182</sup>. However, despite the great efforts made by numerous research groups around the world and the progresses achieved in membrane fouling, membrane manufacture, and reactor configuration, very few very small municipal WWTPs based on AnMBR technology have been implemented so far, as far as we know. An AnMBR plant went into operation in Tortosa (Spain) in 2019 designed to treat 18 m<sup>3</sup>·d<sup>-1</sup>,

although it is currently treating around half of the design flow rate<sup>183</sup>. The results obtained in the first year showed high organic removal efficiency (93% for COD, 83% for BOD<sub>5</sub>). Methane yield was 0.19 L CH<sub>4</sub>·g<sup>-1</sup> COD removed when considering only biogas methane, and 0.27 L CH<sub>4</sub>·g<sup>-1</sup> COD when considering both biogas methane and methane dissolved in permeate. The main operating problem was the high H<sub>2</sub>S biogas concentration (over 2000 ppm), while there were no Helminth eggs or Legionella in the permeate and 90% of the samples analyzed contained less than 200 cfu per 100mL of E.coli. Another example of an industrial prototype AnMBR (demonstration scale, TRL of 6) was evaluated for MWW treatment at the *Alcázar de San Juan* WWTP (Ciudad Real, Spain) within the LIFE MEMORY project<sup>1,38</sup>. The results obtained showed effluent COD concentrations below the discharge limits, achieving COD removals above 90% and reduction of sludge production of 36–58% compared to theoretical aerobic sludge productions. The performance low power requirements for the filtration process (0.15 kWh per m<sup>3</sup> of treated water) were obtained whilst producing a nutrient-rich, pathogen-free permeate. These results show the viability of this technology for producing high-quality reclaimed water for different purposes. Although moving forward into solving different bottlenecks (dissolved methane recovery, membrane fouling) is a remaining challenge, the success of this experience in a real environment shows that the decentralized treatment strategy could maximize the recovery of water, nutrients, and energy, and that it is possible to implement AnMBR technology for treating MWW, setting TRL at 7.

Key perspectives and challenges for widening the full-scale massive implementation of AnMBR technology are outlined as follows:

- Combining PS, AD and AnMBR can overcome the challenges raised by low BOD to sulfate influent ratios and low temperatures in cold climates in which temperatures go below 15°C. In this respect, it is not economically feasible to raise inflow temperature to mesophilic conditions (around 35° C) by heating<sup>184</sup> since huge amounts of energy would be needed.
- The development of novel reactor configurations, anti-fouling membrane modules, cost-effective fouling mitigation methods, and measures for increasing OLR are key alternatives to further enhance the AnMBR energy balance.

- Optimized post-treatment steps for nutrient recovery are needed since AnMBR permeate contains most of the nutrients present in the influent. AnMBR permeate quality (especially using ultrafiltration membranes) makes fertigation the best option for jointly recycling water and nutrients <sup>184</sup>, although it is not always possible, so that different alternatives must be optimized for nutrient recovery <sup>155</sup>.

- Physical post-treatment recovers the methane dissolved in AnMBR effluents. For instance, degassing membranes has been reported as a cost-effective and reliable method for dissolved methane capture, although the technology needs further development for full-scale implementation.

- Although most of the produced biogas is currently used for heat and electricity generation, a few countries (mainly Sweden and to a lesser extent Denmark, France, Switzerland, and South Korea ) have implemented biogas upgrade to biomethane for transport fuel and natural gas grid injection<sup>185</sup>.

Physical/chemical technologies for biogas upgrading have been developed and are now mature technologies capable of providing biomethane. However, their high energy and chemical requirements limit the environmental and economic sustainability of conventional biogas upgrading technologies <sup>186</sup>. For this reason, emerging biotechnologies (such as H<sub>2</sub>-based chemoautotrophic CO<sub>2</sub> bioconversion to CH<sub>4</sub>, microalgae-based CO<sub>2</sub> fixation, enzymatic CO<sub>2</sub> dissolution or fermentative CO<sub>2</sub> reduction and digestion with in situ CO<sub>2</sub> desorption) can offer a low-cost and environmentally friendly alternative to physical/chemical biogas upgrading.

With the appropriate technological advances, AnMBR could represent a real alternative to conventional aerobic treatments, being able to produce a high-quality effluent, recovered nutrients, and energy, while keeping a low environmental impact, low footprint and overall costs. However, the main challenges are not only technological, environmental and economic: social acceptance and the political framework are also crucial and therefore need to be properly considered in order to guarantee AnMBR feasibility and sustainability. In this respect, the European Commission approved the first pilot Innovation Deal entitled “Sustainable wastewater treatment combining anaerobic membrane technology and water reuse in 2016”. The tasks carried out within the ID included the analysis of several case studies. Jiménez-Benítez



et al.<sup>12</sup> studied two cases in the Mediterranean Region (Italy and Spain) and compared the application of combined AnMBR technology and fertigation with conventional WWTP configurations with aerobic systems, irrigation with freshwater and mineral fertilization. They showed that reclaimed water can be considered a reliable water and nutrient source, return a positive economic balance (up to 376 k€ per year) and provide significant reductions and savings in CO<sub>2</sub> emissions (up to -898.9 tCO<sub>2</sub> eq. per year).

## 5. Conclusions

AnMBR is a promising technology for MWW treatment and resource recovery that has many advantages that could help WWTPs to become WRRFs: reduced footprints, effluent quality suitable for fertigation or nutrients recovery potential, carbon recovery, low biosolids production and on-site stabilization. However, there are still some challenges that need to be addressed to make AnMBR technically and economically more competitive for full-scale application. Energy recovery and GHG emissions mitigation must be optimized by efficiently recovering the dissolved methane from AnMBR effluent. Novel platforms such as the combination of AnMBR with PS and sidestream AD of concentrated particulates should be considered to improve energy balance when the COD/SO<sub>4</sub> ratio is low. Fouling mitigation can be enhanced by novel configurations based on different aspects such as scouring agents for mechanical cleaning, vibration or the combination of BES and AnMBR. Co-digestion of solid wastes such as food waste or the municipal fraction of organic solid waste appears as an interesting alternative to significantly enhance organic carbon use. Different novel platforms for the capture, concentration and recovery of inorganic nutrients are also needed when fertigation is impossible. Some of these alternatives are based on phototrophics (algae PBR, algae-bacteria PBR, or PSB), membranes (e.g. FO, HFMC, ED, or BES) and IEX.

Although AnMBRs have the potential for full-scale resource recovery, there is also room for further improvement of their design, operation and optimization, bringing us one step closer to a sustainable and circular society based on resource recycling. It is important to note that demonstration projects would

need to be carried out to take full advantage of its benefits and favorable and harmonized regulations among the EU States would need to be adopted.

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**Table 1.** Summary of AnMBR performance treating municipal wastewater

Reference	COD (g·m <sup>-3</sup> )	BOD (g·m <sup>-3</sup> )	TN (g N·m <sup>-3</sup> )	TP (g P·m <sup>-3</sup> )	%CH <sub>4</sub>	%VSS <sub>sludge</sub>
14	59	14	47.9	6.7	57.2	67.0
121	81	n.a.	44.9	3.9	62.0	59.3
36	70-100	n.a.	38.0-62.1	5.1-10.2	55.0-70.0	66.5-72.4
1	80-116	n.a.	34.2-54.3	6.1-10.3	74.1-77.5	65.8-73.0
38	91	n.a.	47.9	7.7	76.9	70.3
39	58	25	37.0	4.2	68.0	n.a.
37	50	n.a.	34.2	6.5	50.0-70	63.0-75.0
187	39-54	8-16	n.a.	n.a.	70.0-79.6	82.0-84.0

(n.a.: not available)



**Table 2.** Some characteristics of novel AnMBR configurations for membrane fouling mitigation

Technology	Operating principle	Fouling mitigation principle	Main mitigated foulants	Drawback	Relative energy demand/costs
Granular sludge and fluidized agents	Fluidized elements by cross-flow velocity (biogas and/or sludge)	Physical membrane scouring and contaminants adsorption (colloids, SMP...)	Cake layer fouling, and SMP and EPS (adsorption)	Possible membrane damage by fluidized elements	Low
Vibration	Magnetically induced vibration	Turbulence on the membrane surface	Cake layer fouling	Possible damage of membrane surface by friction	Medium
Rotating membranes	Rotation of membranes	Shear-enhanced filtration	Cake layer fouling	Possible damage of membrane surface by friction	Medium
Dynamic membranes	Self-forming biological filtering layer on a support material	Substitution of ultrafiltration membranes by a mesh filter as support material	n.a.	Time for filtering layer formation and permeate quality	Very low
Bioelectrochemical systems	Membrane unit performs as an electrode	Membrane's smoother surface and less binding properties with the fouling precursors (e.g. divalent ions).	General fouling mitigation	Fouling decreases as the applied voltage is increased	Medium

**Table 3.** Key aspects of new platforms for resource recovery from MWW through AnMBR technology

Technology	Recovered resource	Main applications	Potentials	Limitations	Cost <sup>(1)</sup>	References
Co-digestion	CH <sub>4</sub>	Renewable energy	Positive net energy balance when co-digestion >40% food waste	Severe fouling which leads to high operation and maintenance costs	(2) ≈€0 - €0.051 per m <sup>3</sup> of treated water. -1.24 to 0.79 kWh per m <sup>3</sup>	108 115
Primary settling and chemical enhanced primary treatment (CEPT)	CH <sub>4</sub>	Renewable energy	Increase energy recovery through AD of PS Obtain a high-quality, methane-free effluent CEPT enhances removal of pollutants Easy operation and low costs	The wastewater physicochemical characteristics strongly affect the CEPT efficiency	(1) €0.05 -€0.07 per m <sup>3</sup> and maximum energy surplus of 0.10 kWh per m <sup>3</sup> (PS+AnMBR+AD)	120
Direct membrane filtration	Nutrient-rich water with negligible solids concentration	Agriculture Urban irrigation Nutrient recovery	Simultaneous energy, water and nutrient recovery Enables decentralized anaerobic treatment of MWW	Not full scale applications evaluated yet Sensitive areas require nutrient removal Regulation for nutrient-rich water use in agriculture	<0.4 kWh per m <sup>3</sup> of treated water (<8 kWh·kg <sup>-1</sup> N)	155
Forward osmosis	VFA and H <sub>2</sub> CH <sub>4</sub> Nutrient-rich water	Agriculture Urban irrigation Nutrient recovery	Low fouling propensity, easy cleaning, high rejection of contaminants, reduce volumetric flow rates and dissolved methane losses.	High energy requirements to operate the RO to produce regenerated water from diluted draw solutions	FO/RO + AnMBR €0.81 per m <sup>3</sup> , (FO recovery to 50%)	124
Microalgae	Microalgae-based bio-fertilizer after harvesting and composting or thermochemical processes (biochar)	Biofertilizer	Improves soil properties Reduce nutrient losses through a consistent release of nutrients Small carbon footprint (C sequestration) Competitive yields Up to 90% of nutrient content in WW	Further treatment needed for biofertilizer Low degradability of the cell wall Depends on solar radiation availability Extensive surfaces are required Harvesting phase still challenging	(1) 0.1-2 €·kg <sup>-1</sup>	155 137 132
Photosynthetic Bacteria	PSB-based biofertilizer after harvesting	Biofertilizer	High yield of hydrogen High tolerance over disturbances Assimilates C, N and P in a single stage Content of many high-value substances Reduced sludge production	Further treatment needed for biofertilizer Poor settling properties Depends on infrared light availability Still at lab scale Further safety criteria needed	n.a.	153 136
Membrane contactors	Ammonium sulfate	Fertilizer	High N removal efficiencies (> to 99%) Reduce nitrogen load and aeration energy consumption.	Membrane fouling needs further research prior to develop full scale applications	(1) 3 \$·kg <sup>-1</sup> N recovered	155
Electrodialysis	Solution with high N and/or P concentrations to produce ammonium sulfate and struvite	Fertilizer	Concentrates N and P from dilute streams such as wastewater MWW. P recovery from sewage sludge ashes	Lab scale applications only	(2) 3.5-6.5 kWh·kg <sup>-1</sup> N	155
Bioelectrochemical systems	Free ammonia gas that can be absorbed in an acidic solution	Chemical industry Agriculture	High N removal efficiencies (70-90%) Suitable for nitrogen rich streams: reject water, urine, landfill leachate.	Not full scale applications evaluated yet	(1) Costs < revenues for N removal from urine	155
Ion exchange systems	Nutrient-rich water with negligible solids concentration	Fertilizer	Nutrient recovery by IEX from the effluent of an AnMBR is gaining momentum since AnMBR produces a solids-free high-quality permeate	Rarely applied in MWW due to media selectivity, bed clogging, and costly regenerations	n.a.	167 - 170 135 173

(\*): Costs referred to: (1) Total costs, (2) Operating costs