Using ultrafiltration membrane technology for resource recovery from municipal wastewater: evaluation of direct membrane filtration, anaerobic membrane bioreactor, and tertiary membrane filtration.

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

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Acknowledgements

Writing the acknowledgments section of a PhD Thesis: a case study.

Abstract

This work aims to investigate the most appropriate way to write an acknowledgment section of a PhD Thesis. For this, a methodology is proposed that includes, as far as possible, every person, animal or thing that deserves to be mentioned for having contributed, in one way or another, to not ending up jumping off a bridge, fleeing to a remote island or abandon yourself to despair or laziness. The proposed methodology is applied to a specific case of writing a thesis that, let's face it, has lasted more than a day without bread, without the fact that the author is from the Canary Islands being considered as mitigating. The result of the study shows that the number of elements to be thanked for increases linearly with the time the thesis is being prepared, the most influential factors being: the preparation within a mixed research unit, the overwhelming personality of the author and the hours of sunshine available for breaks.

Keywords

Acknowledgment; gratitude

1. Introduction

Since the beginning of time, it seems that human beings have a need to be grateful for things. It seems somewhat reasonable, since all achievements, both collective and individual, are the result of the support and collaboration of the people around us. PhD Thesis are not an exception to this principle, since in their development a series of situations take place that leads the PhD candidate to go through different stages characterized by despair, euphoria, anger, hope, grief or nihilism.

The objective of this work is to develop a robust methodology to prepare a good acknowledgment section in a doctoral thesis that does not make me look terrible, or at least not very bad with the people around me and from whom I have been able to receive support. Very necessary, not only in aspects strictly related to the investigation.

2. Materials and Methods

The methodology developed, called "my goodness, don't forget anyone, please" (MGDAP) is the result of integrating the most sophisticated review procedures to date, based on thinking over an "n" number of times. Given that "n" has no upper limit and the probability of not-forgetting anybody tend to 1 with "n", making n = infinite, it can be ensured that the acknowledgments section would be perfect, as shown in Figure 1.

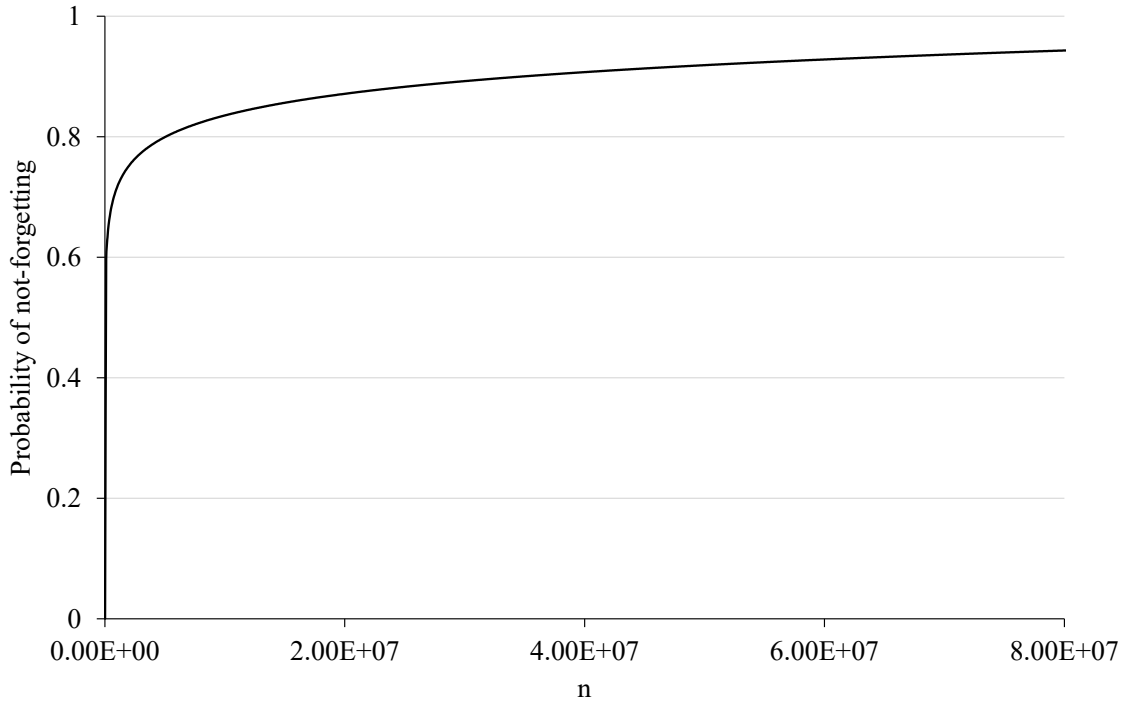

Figure 1. Probability of not-forgetting

However, a compromise must be reached on “n” that reduces uncertainty without causing the writing of the acknowledgments section to be longer than the writing of the thesis. In order to optimize the number of iterations, it is therefore proposed to apply the principle of "man, forget it, you have other things to do" (MFI-YHOTtD). In the event that, finally, an oversight occurs, the "garlic and water" (G&W) theorem is recommended and then apologize.

3. Results

The results obtained from the application of the MGDAP methodology to the case of this PhD Thesis are presented below:

- I want to thank my supervisors, Aurora Seco and Ángel Robles, for their dedication, advice, and support. And for believing that this thesis was possible before I thought about it.
- I also want to thank José Ferrer and Aurora Seco (this time as coordinator of the CALAGUA Joint Unit), for trusting me back in 2008 and continuing to do so throughout these years.

- Many thanks to the UV sector (Luis, Josep, Aurora, Alberto, Ángel, Vicky, Nuria "Martí-sers", Estela, Laura, Juan…), for the laughter in the cafeteria and in the offices. In the same way, also many thanks to the UPV team (José, Ramón, Quique, Joaquín, Dani and María…) for the good times at college and in the agora. How necessary are these breaks is for me.
- To all the PhD candidates at CALAGUA Joint Unit, and a lot currently PhD (there are so many that I am not going to list them), who have operated plants with superhuman suffering and who have not once asked me "what about mine" when I took possession of their data.
- I also want to record my special gratitude to Ruth, for paving the way, and from whom I have learned a lot, without her knowing it.
- To my parents, who have always made it very easy for me. His unconditionality is a great platform to jump off of.
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- Thanks to my sisters María and Begoña, far away, but very close. For laughing at my jokes even when they were horrible.
- Thanks to Óscar and Pablo. Seeing them grow and laugh gives me confidence in the future.
- And I leave Marta for last. Shoulder, arm, ear, heart, lough and good sense. I can't think of another way to synthesize it. I am a lucky man.

4. Conclusions

MGDAP methodology allows reasonable assurance that the acknowledgments section is acceptable, although the doubt always remains whether someone else should not be included.

Acknowledgements

Done.

Abstract

The climate and environmental emergency that society faces represents a challenge that must be addressed from different areas and perspectives. Among the possible options for action, the recovery of resources present in wastewater is considered a promising strategy that respects the foundations of the Circular Economy principles. Under the perspective of this economic model, a conceptual change is required that modifies the perception of urban wastewater, going from waste to being consider as a source of resources, among which stand out reclaimed water, carbon and nutrients (mainly nitrogen and phosphorus). This shift in perception must be accompanied by adequate technological innovation, allowing better use of these resources, guaranteeing the correct treatment of wastewater, and ensuring the health of the population and the conservation of ecosystems.

The incorporation of ultrafiltration (UF) membrane technology to urban wastewater treatment facilities, either as a complementary stage in conventional treatment schemes or as part of more innovative processes (for example, combined with an anaerobic reactor forming an anaerobic membrane bioreactor (AnMBR)) is presented as a promising alternative from an economic and environmental point of view. Its incorporation as a primary treatment unit allows increasing the amount of organic matter that is derived to the anaerobic digester of the sludge line, therefore reducing the amount of organic matter that must be treated in the biological reactor. Being part of an AnMBR system, the production of a high-quality effluent, the valorization of organic matter in the form of biogas, and the recovery of nutrients are favored. As a tertiary treatment, the quality of the treated water is improved, reaching adequate levels of disinfection, which favors its reuse. Additionally, these filtration systems reduce space requirements compared to other conventional treatment elements with a similar function, such as decanters or disinfection systems composed of sand filters and ultraviolet lamps.

However, the application of UF membranes for the treatment of urban wastewater presents some challenges that must be addressed and overcome to promote their adoption in treatment plants, among which the following stand out: the costs of acquisition and maintenance/replacement of the membranes; the fouling of the membranes derived from the filtration process and the consequent reduction in the efficiency of the process; the recovery of dissolved methane from the effluent of AnMBR systems; and the recovery of nutrients present in the effluent of AnMBR systems.

The general objective of this PhD Thesis is to evaluate the technical, economic and environmental feasibility of the application of ultrafiltration membranes in different stages of urban wastewater treatment: primary treatment (filtration of primary settlers effluent), secondary (AnMBR) and tertiary (filtration of secondary settlers effluent). This general objective is specified in the following tasks: 1) evaluate the technical feasibility of the evaluated treatment processes; 2) identify strategies and operating conditions that favor their optimization from an economic and environmental point of view; 3) estimate the investment and operation costs of the systems; and 4) evaluate the environmental impact derived from the implementation of these systems.

To this end, this PhD Thesis uses data obtained from three demonstration-scale plants, which allow them to be scaled up to an industrial level and their subsequent environmental and economic evaluation using the life cycle and cost analysis tools.

The results obtained in this PhD thesis reveal that the membrane processes evaluated can provide economic and environmental advantages over other conventional treatment systems, although the magnitude of the benefits obtained varies depending on whether the membrane acts as primary treatment, as part of an AnMBR or as a tertiary treatment.

With respect to the inclusion of UF membranes after the primary settling stage, the results obtained show that it is possible to increase the production of methane in the sludge line, due to the increase in the energy recovery of the organic matter and reduce the needs of aeration in the biological reactor, which implies a decrease in the electrical energy consumed, improving the environmental behavior of the treatment process. However, the current acquisition cost of membranes prevents the savings in operating costs (OPEX) from offsetting the necessary investment cost (CAPEX). Therefore, the economic viability of this alternative implies optimizing the filtration process and/or reducing the acquisition price of the membranes.

The study carried out on the application of AnMBR technology allows to demonstrate its potential as a core technology for the treatment of urban wastewater, thanks to the following characteristics: 1) great potential for the recovery of resources (energy, reclaimed water and nutrients) from the generation of biogas and its nutrient-rich effluent; 2) favorable energy balances that allow achieving net energy production in certain circumstances; 3) competitive treatment costs compared to conventional treatments; and 4) reduction of the environmental impact of wastewater treatments due to high energy and nutrient recovery.

With respect to the application of UF membranes as tertiary treatment, its economic viability would depend on the use of reclaimed water for fertigation purposes, which allows the use of its nutrient content, and the optimization of operating costs (energy consumption, mainly). On the other hand, the results obtained demonstrate that tertiary treatment by UF can produce regenerated water of the highest quality, in accordance with current regulations. Regarding its environmental impact, energy optimization would make tertiary treatment by UF membranes improve its competitiveness compared to more conventional treatments.

This PhD Thesis has been developed within the framework of the European innovation initiative *Innovation Deal on sustainable wastewater treatment combining anaerobic membrane technology and water reuse* (2016-2018) and three research projects: 1) *Membrane for Energy and Water Recovery-LIFE MEMORY* (LIFE13 ENV/ES/001353) (2014-2018) funded by Life+ Programme; 2) *Application of membrane technology to enhance the transformation of WWTPs in resource recovery facilities-MEM4REC* (CTM2017-86751-C2-2-R-AR) (2018-2021), funded by the State Research Agency-AEI; and 3) *Caracterización de aguas y lodos residuales urbanos e industriales y evaluación de la tecnología adecuada para la recuperación de microplásticos presentes en los mismos-MICROPLAST* (INNEST00/19/058) (2019-2020), funded by the Valencian Innovation Agency -AVI.

Resum

L'emergència climàtica i ambiental que enfronta la societat suposa un repte que ha de ser abordat des de diferents àmbits i perspectives. Entre les possibles opcions d'actuació, la recuperació dels recursos presents en les aigües residuals es considera una estratègia prometedora que respecta els fonaments de l'Economia Circular. Segons aquest model econòmic, es requereix d'un canvi conceptual que modifique la percepció de les aigües residuals urbanes, passant de ser considerades un residu a contemplar-se com una font de recursos, entre els quals destaquen l'aigua regenerada, i el carboni i els nutrients (principalment nitrogen i fòsfor). Aquest canvi de percepció ha d'anar acompanyat per una adequada innovació tecnológica que permeta un millor aprofitament dels recursos, garantint el correcte tractament de les aigües residuals, i assegurant la salut de la població i la conservació dels ecosistemes.

La incorporació de la tecnologia de membranes d'ultrafiltració (UF) a les instal·lacions de depuració de les aigües residuals urbanes, bé siga com a etapa complementària en els esquemes de tractament convencionals, o com a part de processos més nous (per exemple, combinada amb un reactor anaerobi conformant un bioreactor anaerobi de membranes; AnMBR, de l'anglés *anaerobic membrane bioreactor*), es presenta com una alternativa esperançadora des d'un punt de vista econòmic i mediambiental. La seua incorporació com a unitat de tractament primari permet incrementar la quantitat de matèria orgànica que es deriva al digestor anaerobi de la línia de fangs, reduint per tant la quantitat de matèria orgànica que ha de ser tractada en el reactor biològic. Amb el sistema AnMBR, s'afavoreix la producció d'un efluent d'elevada qualitat, la valorització de la matèria orgànica en forma de biogàs, i la recuperació de nutrients. Com a tractament terciari, el sistema de membranes millora la qualitat de l'aigua tractada, aconseguint adequats nivells de desinfecció d'aquesta, la qual cosa afavoreix la seua reutilització. Addicionalment, aquests sistemes de filtració redueixen els requisits d'espai en comparació amb altres elements de tractament convencionals amb funció similar, tal com són els decantadors o els sistemes de desinfecció compostos per filtres d'arena i llums ultraviolats.

No obstant això, l'aplicació de membranes d'ultrafiltració per al tractament d'aigües residuals urbanes presenta alguns reptes que han de ser abordats i superats per a impulsar la seua adopció en les plantes de tractament, entre els quals destaquen: els costos d'adquisició i manteniment/reposició de les membranes; l'embrutiment de les membranes derivat del procés de filtració i la conseqüent reducció de l'eficiència del procés; la recuperació del metà dissolt en

l'efluent de sistemes AnMBR; i la recuperació dels nutrients presents en l'efluent de sistemes AnMBR.

Aquessta tesi doctoral té com a objectiu general avaluar la viabilitat tècnica, econòmica i mediambiental de l'aplicació de membranes d'ultrafiltració en diferents etapes de tractament d'aigües residuals urbanes: tractament primari (filtració de l'efluent de decantació primària), secundari (AnMBR) i terciari (filtració de l'efluent de decantació secundària). Aquest objectiu general es concreta en les següents tasques: 1) avaluar la viabilitat tècnica dels processos de tractament; 2) identificar estratègies i condicions d'operació que afavorisquen l'optimització dels processos des d'un punt de vista econòmic i mediambiental; 3) estimar els costos d'inversió i operació d'aquests sistemes; i 4) avaluar l'impacte ambiental derivat de la implementació d'aquests sistemes.

Per a això, en aquesta tesi doctoral s'utilitzen dades obtingudes en tres plantes a escala demostració, que permeten estudiar el seu escalat a nivell industrial i la seua posterior avaluació ambiental i econòmica mitjançant les eines d'anàlisis de cicle de vida i de costos desenvolupades.

Els resultats obtinguts en aquesta tesi revelen que els processos de membranes avaluats poden suposar avantatges econòmics i ambientals enfront d'altres sistemes de tractament convencional, encara que la magnitud dels beneficis obtinguts varia en funció del tipus d'aplicació, és a dir, de si la membrana actua com a tractament, com a part d'un AnMBR o com a tractament terciari.

Respecte a la inclusió de membranes d'UF després de l'etapa de decantació primària, els resultats obtinguts mostren que és possible incrementar la producció de metà en la línia de fangs, a causa de l'augment de la valorització energètica de la matèria orgànica, i reduir les necessitats de ventilació en el reactor biològic, la qual cosa implica un descens de l'energia elèctrica consumida, millorant el comportament mediambiental del procés de tractament. No obstant això, l'actual cost d'adquisició de les membranes impedeix que els estalvis en els costos d'operació (OPEX) compensen el cost d'inversió (CAPEX) necessari. Per tant, la viabilitat econòmica d'esta alternativa implica optimitzar el procés de filtració i/o la reducció del preu d'adquisició de les membranes.

L'estudi realitzat sobre l'aplicació de la tecnologia AnMBR permet demostrar el seu potencial com a tecnologia central per al tractament de les aigües residuals urbanes, gràcies a les següents

característiques: 1) gran potencial per a la recuperació de recursos (energia, aigua regenerada i nutrients) a partir de la generació de biogàs i del seu efluent ric en nutrients; 2) balanços energètics favorables que permeten aconseguir produccions netes d'energia en determinades circumstàncies; 3) costos de tractament competitius enfront dels tractaments convencionals; i 4) reducció de l'impacte ambiental dels tractaments d'aigües residuals a causa de la recuperació energètica i de nutrients.

Respecte a l'aplicació de membranes d'UF com a tractament terciari, la seua viabilitat econòmica estaria condicionada a l'ús de l'aigua regenerada com a agent fertilitzant, que permet l'aprofitament del seu contingut en nutrients, i a l'optimització dels costos d'operació (consum energètic, fonamentalment). D'altra banda, els resultats obtinguts demostren que el tractament terciari per UF és capaç de produir aigua regenerada de la màxima qualitat, d'acord amb la normativa vigent. Quant al seu impacte ambiental, l'optimització energètica faria que el tractament terciari per membranes d'UF millorara la seua competitivitat enfront dels tractaments més convencionals.

Aquesta tesi doctoral s'ha desenvolupat en el marc de la iniciativa europea sobre innovació *Innovation Deal on sustainable wastewater treatment combining anaerobic membrane technology and water reuse* (2016-2018) i tres projectes d'investigació: 1) *Membrane for Energy and Water Recovery-LIFE MEMORY* (LIFE13 ENV/ES/001353) (2014-2018) finançat per Life+ Programme; 2) *Application of membrane technology to enhance the transformation of WWTPs in resource recovery facilities-MEM4REC* (CTM2017-86751-C2-2-R-AR) (2018-2021), finançat per l'Agència Estatal d'Investigació-AEI; i 3) *Caracterización de aguas y lodos residuales urbanos e industriales y evaluación de la tecnología adecuada para la recuperación de microplásticos presentes en los mismos-MICROPLAST* (INNEST00/19/058) (2019-2020), finançat per l'Agència Valenciana de la Innovació-AVI.

Resumen

La emergencia climática y ambiental que enfrenta la sociedad supone un reto que debe ser abordado desde diferentes ámbitos y perspectivas. Entre las posibles opciones de actuación, la recuperación de los recursos presentes en las aguas residuales se considera una estrategia prometedora que respeta los fundamentos de la Economía Circular. Al amparo de este modelo económico, se requiere de un cambio conceptual que modifique la percepción de las aguas residuales urbanas, pasando de ser consideradas un residuo a contemplarse como una fuente de recursos, entre los que destacan el agua regenerada, y el carbono y los nutrientes (principalmente nitrógeno y fósforo) presentes en las mismas. Este cambio de percepción debe ir acompañado por una adecuada innovación tecnológica, permitiendo un mejor aprovechamiento de estos recursos, garantizando el correcto tratamiento de las aguas residuales, y asegurando la salud de la población y la conservación de los ecosistemas.

La incorporación de la tecnología de membranas de ultrafiltración (UF) a las instalaciones de depuración de las aguas residuales urbanas, bien como etapa complementaria en los esquemas de tratamiento convencionales o como parte de procesos más novedosos (por ejemplo, combinada con un reactor anaerobio conformando un biorreactor anaerobio de membranas; AnMBR, del inglés *anaerobic membrane bioreactor*), se presenta como una alternativa prometedora desde un punto de vista económico y medioambiental. Su incorporación como unidad de tratamiento primario permite incrementar la cantidad de materia orgánica que se deriva al digestor anaerobio de la línea de fangos, reduciendo por tanto la cantidad de materia orgánica que ha de ser tratada en el reactor biológico. Formando parte de un sistema AnMBR, se favorece la producción de un efluente de elevada calidad, la valorización de la materia orgánica en forma de biogás, y la recuperación de nutrientes. Como tratamiento terciario, se mejora la calidad del agua tratada, alcanzando adecuados niveles de desinfección de la misma, lo cual favorece su reutilización. Adicionalmente, estos sistemas de filtración reducen los requisitos de espacio en comparación con otros elementos de tratamiento convencional con función similar, tal y como son los decantadores o los sistemas de desinfección compuestos por filtros de arena y lámparas ultravioletas.

Sin embargo, la aplicación de membranas de ultrafiltración para el tratamiento de aguas residuales urbanas presenta algunos retos que deben ser abordados y superados para impulsar su adopción en las plantas de tratamiento, entre los que destacan: los costes de adquisición y

mantenimiento/reposición de las membranas; el ensuciamiento de las membranas derivado del proceso de filtración y la consecuente reducción de la eficiencia del proceso; la recuperación del metano disuelto en el efluente de sistemas AnMBR; y la recuperación de los nutrientes presentes en el efluente de sistemas AnMBR.

Esta tesis doctoral tiene como objetivo general evaluar la viabilidad técnica, económica y medioambiental de la aplicación de membranas de ultrafiltración en diferentes etapas de tratamiento de aguas residuales urbanas: tratamiento primario (filtración del efluente de decantación primaria), secundario (AnMBR) y terciario (filtración del efluente de decantación secundaria). Este objetivo general se concreta en las siguientes tareas: 1) evaluar la viabilidad técnica de los procesos de tratamiento evaluados; 2) identificar estrategias y condiciones de operación que favorezcan la optimización de los mismos desde un punto de vista económico y medioambiental; 3) estimar los costes de inversión y operación de los dichos sistemas; y 4) evaluar el impacto ambiental derivado de la implementación de estos sistemas.

Para ello, en esta tesis doctoral se utilizan datos obtenidos en tres plantas a escala demostración, los cuales permiten estudiar su escalado a nivel industrial y su posterior evaluación ambiental y económica mediante las herramientas de análisis de ciclo de vida y de costes desarrolladas.

Los resultados obtenidos en esta tesis revelan que los procesos de membranas evaluados pueden suponer ventajas económicas y ambientales frente a otros sistemas de tratamiento convencional, aunque la magnitud de los beneficios obtenidos varía en función de si la membrana actúa como tratamiento, como parte de un AnMBR o como tratamiento terciario.

Con respecto a la inclusión de membranas de UF tras la etapa de decantación primaria, los resultados obtenidos muestran que es posible incrementar la producción de metano en la línea de fangos, debido al aumento de la valorización energética de la materia orgánica, y reducir las necesidades de aireación en el reactor biológico, lo cual implica un descenso de la energía eléctrica consumida, mejorando el comportamiento medioambiental del proceso de tratamiento. Sin embargo, el actual coste de adquisición de las membranas impide que los ahorros en los costes de operación (OPEX) compensen el coste de inversión (CAPEX) necesario. Por lo tanto, la viabilidad económica de esta alternativa implica optimizar el proceso de filtración y/o la reducción del precio de adquisición de las membranas.

El estudio realizado sobre la aplicación de la tecnología AnMBR permite demostrar su potencial como tecnología central para el tratamiento de las aguas residuales urbanas, gracias a las siguientes características: 1) gran potencial para la recuperación de recursos (energía, agua regenerada y nutrientes) a partir de la generación de biogás y de su efluente rico en nutrientes; 2) balances energéticos favorables que permiten alcanzar producciones netas de energía en determinadas circunstancias; 3) costes de tratamiento competitivos frente a los tratamientos convencionales; y 4) reducción del impacto ambiental de los tratamientos de aguas residuales debido a la elevada recuperación energética y de nutrientes.

Con respecto a la aplicación de membranas de UF como tratamiento terciario, su viabilidad económica estaría condicionada al uso del agua regenerada como agente fertilizante, que permite el aprovechamiento de su contenido en nutrientes, y a la optimización de los costes de operación (consumo energético, fundamentalmente). Por otra parte, los resultados obtenidos demuestran que el tratamiento terciario por UF es capaz de producir agua regenerada de la máxima calidad, de acuerdo con la normativa vigente. En cuanto a su impacto ambiental, la optimización energética haría que el tratamiento terciario por membranas de UF mejorase su competitividad frente a los tratamientos más convencionales.

Esta tesis doctoral se ha desarrollado en el marco de la iniciativa europea sobre innovación *Innovation Deal on sustainable wastewater treatment combining anaerobic membrane technology and water reuse* (2016-2018) y tres proyectos de investigación: 1) *Membrane for Energy and Water Recovery-LIFE MEMORY* (LIFE13 ENV/ES/001353) (2014-2018) financiado por Life+ Programme; 2) *Application of membrane technology to enhance the transformation of WWTPs in resource recovery facilities-MEM4REC* (CTM2017-86751-C2-2-R-AR) (2018-2021), financiado por la Agencia Estatal de Investigación-AEI; y 3) *Caracterización de aguas y lodos residuales urbanos e industriales y evaluación de la tecnología adecuada para la recuperación de microplásticos presentes en los mismos-MICROPLAST* (INNEST00/19/058) (2019-2020), financiado por la Agencia Valenciana de la Innovación-AVI.

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1. Introduction

1.1. Introduction

Maintenance of life depends on biological organisms being able to provide themselves with the necessary resources to sustain their metabolism and their ability to get rid of the waste generated. This basic behavior turns into a high-complex network once all interactions between environment and living beings and among living beings acting all together are considered. Furthermore, these geographically located networks constitute what is known as ecosystem, which in turn interacts with other ecosystems on a wider scale.

In this sense, the construction of biological diversity from primitive prokaryotic bacteria to the current situation, including episodes of mass extinction and subsequent recovery, have had as one of their foundations the advantages derived from the ability to take benefit of not only from inorganic and organic resources available in the environment, but also specifically the waste of other organisms, in order to valorize them, both for their use as energy suppliers (catabolism) and as elements for the construction of structures of the organism itself (anabolism). The health of ecosystems and their populations is based, therefore, on a correct and balanced functioning of these interactions. Any imbalance that occurs tends to spread throughout the entire chain and put the survival of the whole at risk. It can be therefore considered that natural ecosystems are based on maximum circular transformation of materials and energy.

Social and economic relationships in human societies can also be understood based on the biological approach. In this sense, societies develop their activities from the resources available and generate a variety of waste that is released into the environment. This human activity was initially restricted to a local geographic environment with low long-distance exchanges, but the access first to the technology and latter to fossil energy sources, and the development of faster and higher-capacity transport systems has made it possible to establish trade networks moving matter and energy that now span the entire world (Naredo Pérez, 1993). This phenomenon of increased connectivity and interdependence has been called “globalization”.

The increase in population, economic development and the rise in living standards have led to an increase in the consumption of materials and energy and an increase in the generation of waste and emissions that already pose a serious threat to the environment and, with it, the human species. By way of example, gross domestic product (GDP) is traditionally defined as the monetary value of all finished goods and services made within a country during a specific period. Since products and services are ultimately made from energy and materials, GDP can also be interpreted as an

indicator of the consumption of these elements. Figure 1.1 represents the evolution of GDP in the last 2 millennia. It is remarkable how for the first 1000 years, GDP remained almost constant, there is a transition period between year 1000 and 1700 with a regular increase, and after 1700, Industrial Revolution started in VXIII century accelerates growth exponentially.

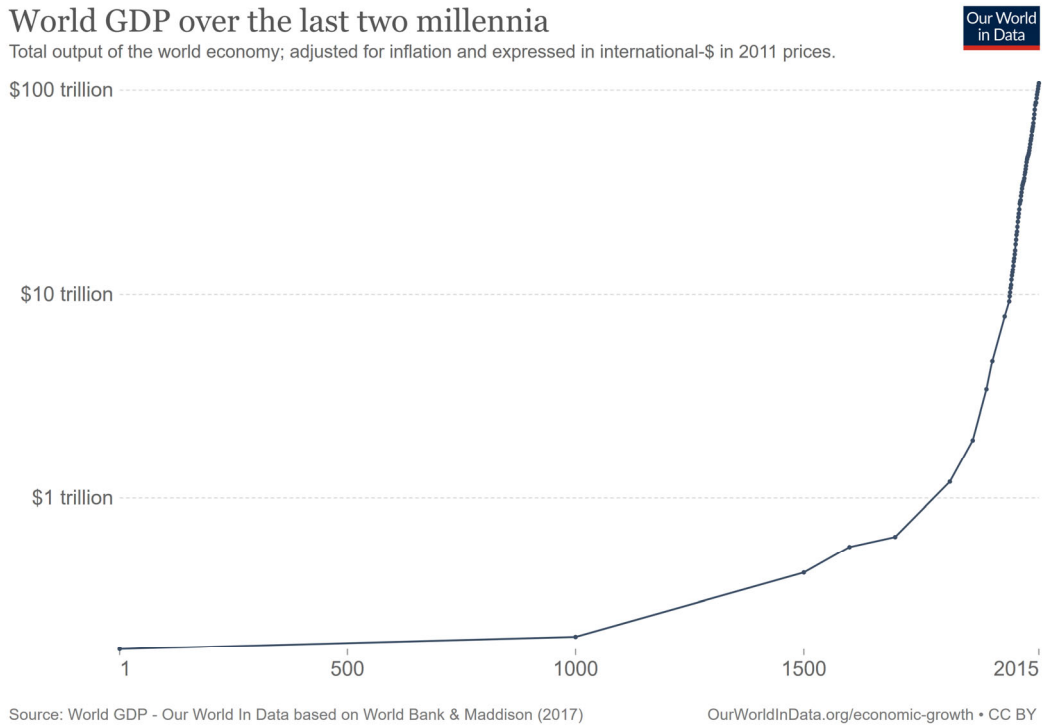


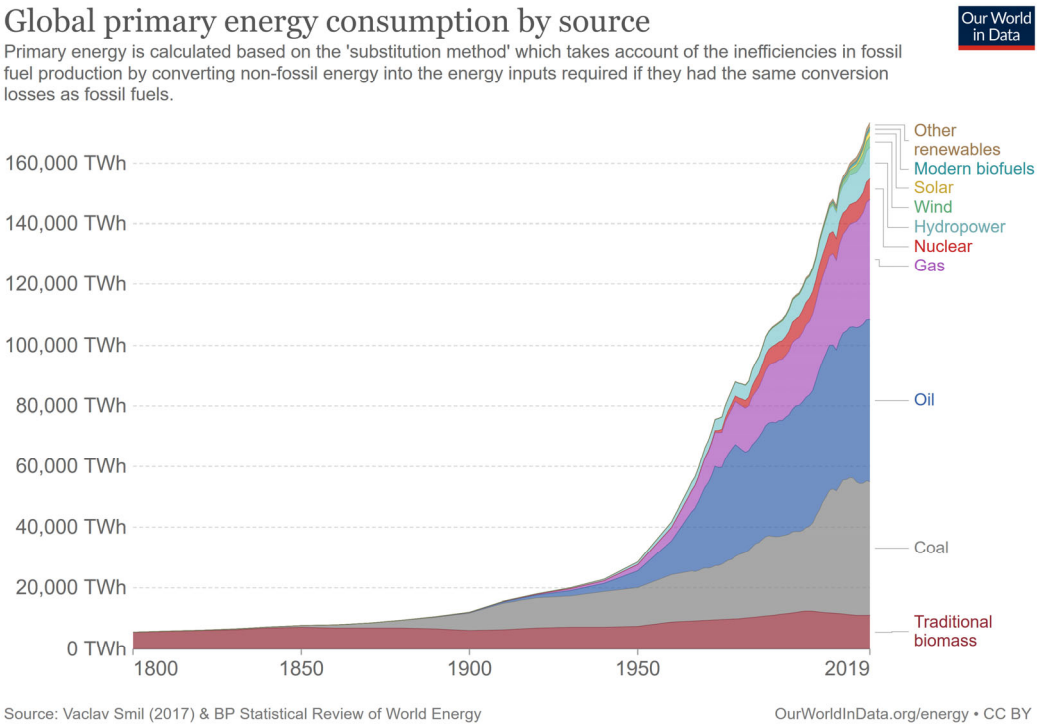
Figure 1.1. Evolution of the world domestic product (GDP) over the last 2000 years (source: https://ourworldindata.org/; accessed 15th March 2023).

Evolution of energy consumption also shows a sharply increase from 1950, driven using non-renewable energy sources, i.e., oil, coal, and gas (Figure 1.2a). Assessment of fossil fuel reserves has received great attention over last decades, since economy and current way of life are critically dependent on energy and its cost (Höök & Tang, 2013). Shafiee & Topal (2009) estimated that reserves of oil and gas will be available up to 2040-2042, while coal reserves will remain until 2112. More recently, Maggio & Cacciola (2012) estimated peak productions for oil, gas and coal to be in the ranges of 2009-2021, 2024-2046 and 2042-2062, respectively. Finally, Yildiz (2018) evaluated reserves-to-production ratios (R/P) for oil, gas and coal obtaining global values of 50.6, 52.5 and 153 years, respectively.

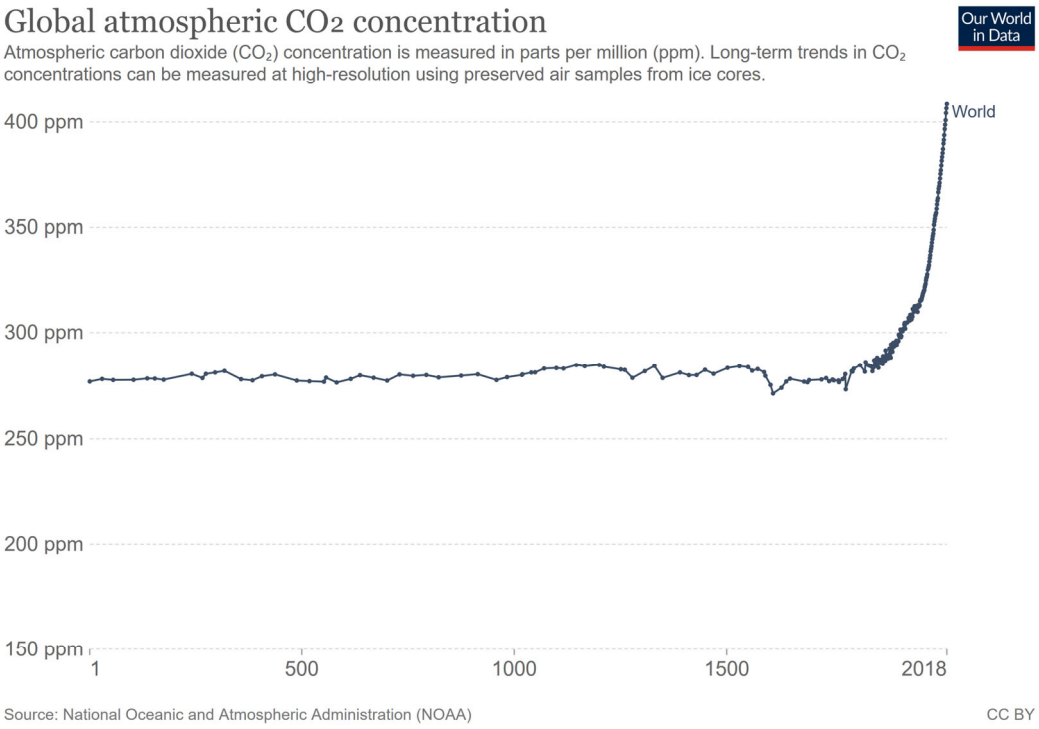
However, global warming and climate change emergency has forced society to substitute fossil fuels to diminish CO_2 emissions (Figure 1.2b). In this sense, Welsby et al. (2021) reported that to

allow for a 50 % probability of keeping global temperature increase below 1.5 °C at the end of the century (according to 2015 Paris Agreement), by 2050 nearly 60 % of oil and fossil methane gas, and 90 % of coal must remain unextracted. Therefore, it is mandatory to look for alternative carbon-neutral energy sources, i.e., solar, wind, biomass, biogas, etc. Biogas can play a role in this transition and the European Union (EU) has adopted the "EU Methane Strategy" (European Commission, 2020a) as a set of policy actions to reduce CH_4 emissions, which will contribute to reach carbon neutrality in 2050. This strategy identifies biogas production from agriculture waste and sewage through anaerobic digestion as a suitable renewable energy source. Furthermore, the European Commission communication "Powering a climate-neutral economy: An EU Strategy for Energy System Integration" (European Commission, 2020b) recognizes a biogas potential production from waste and residues capable of covering between 2.7 and 3.7% of energy consumption in the UE in 2030. It is thus clear that biogas generation had already become a priority in the EU (Scarlat et al., 2018). Furthermore, the invasion of Ukraine has boosted the interest in increasing the weight of this renewable fuel in the European energy mix, as stated in the European Commission (EC) communication "REPowerEU: Joint European Action for more affordable, secure and sustainable energy" (European Commission, 2022a).

Fertilizer availability represents another hotspot for resource sustainability since access to enough and adequate food is based on its application. Figure 1.3 shows how world population raise is, among other reasons, based on food production capacity and, eventually, on fertilization. The Green Revolution that took place around 1950-60 allowed a significant improvement of crops productivity because of introducing new technologies, varieties (especially wheat and rice), mechanization, irrigation, and fertilizers in agriculture. However, this improvement was achieved at the cost of making agriculture more dependent on fossil fuels and their cost. Despite nitrogen is available in the air with no restriction, production of nitrogen-based fertilizers (N-fertilizers) implies a significant energy expenditure and environmental impacts (Ahlgren et al., 2010). According to IRENA & FAO (2021) primary energy consumption for food production worldwide represents around 30% of total energy expenditures, 20 % of them are used in agriculture. Specifically, EU agriculture energy expenditures are 3.7 % of its total energy consumption, with fertilizer production accounting for the 50 % and on-farm diesel around 30 % (Paris et al., 2022).



a)



b)

Figure 1.2. Evolution of (a) world primary energy consumption and (b) atmospheric CO2 concentration (source: https://ourworldindata.org/; accessed 15th March 2023).

Moreover, phosphorus-based fertilizers (P-fertilizers) represents an additional challenge. P is essential for living beings and is intensively used in industry. However, unlike wide nitrogen availability, P source is phosphate rock whose major deposits are located in few countries (China, United States of America, Russia, and West-Sahara) (van Dijk et al., 2016), none of them member of the EU. Therefore, according to phosphorus (P) importance and attending EU vulnerability, P was included in the EU's Critical Raw Materials list in 2017 (European Commission, 2017), which justifies strategies for reducing European dependency on phosphorus importation from a food security perspective as well as from an environmental point of view. It is worth mention that EU imports around 90 % of its total consumption (de Boer et al., 2019).

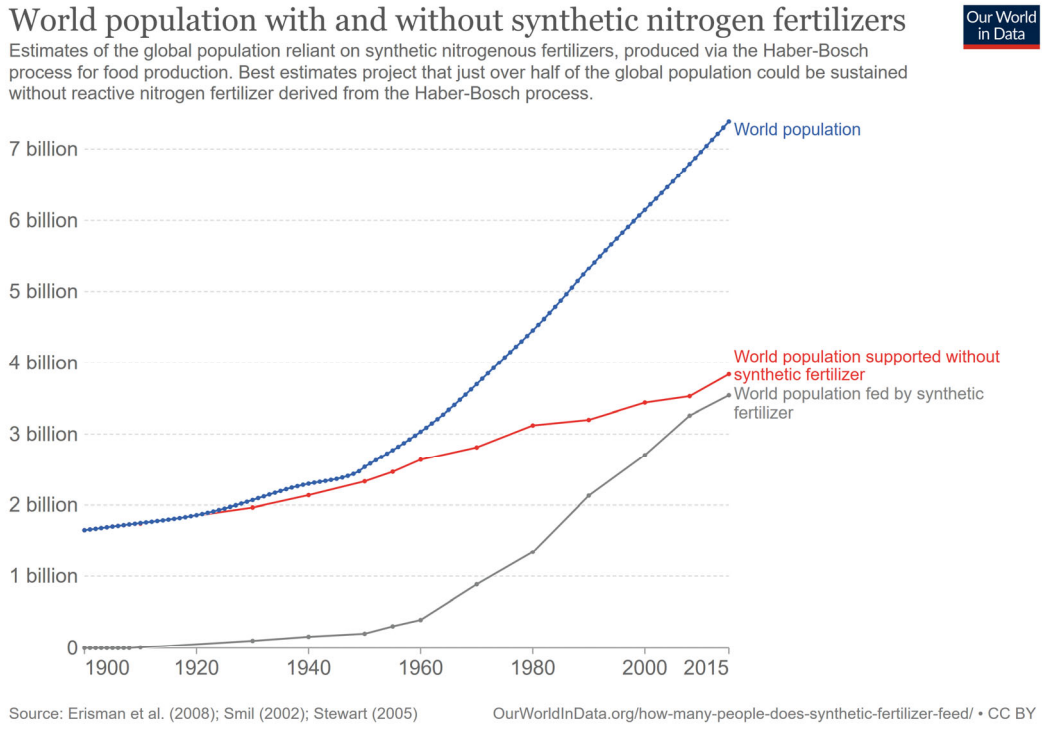
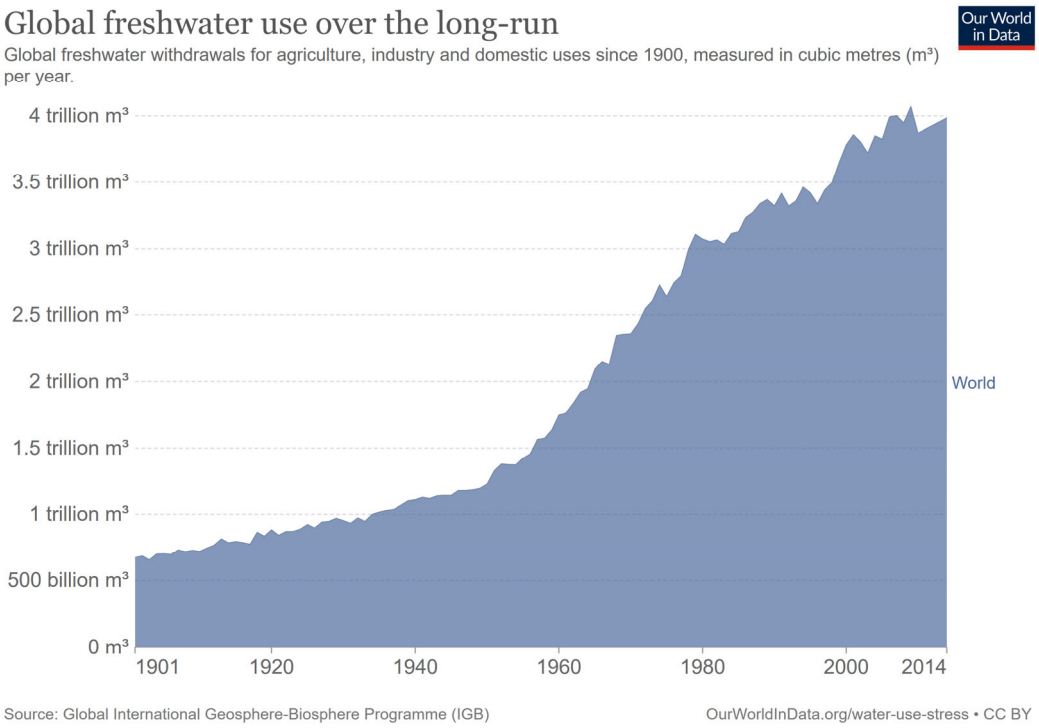
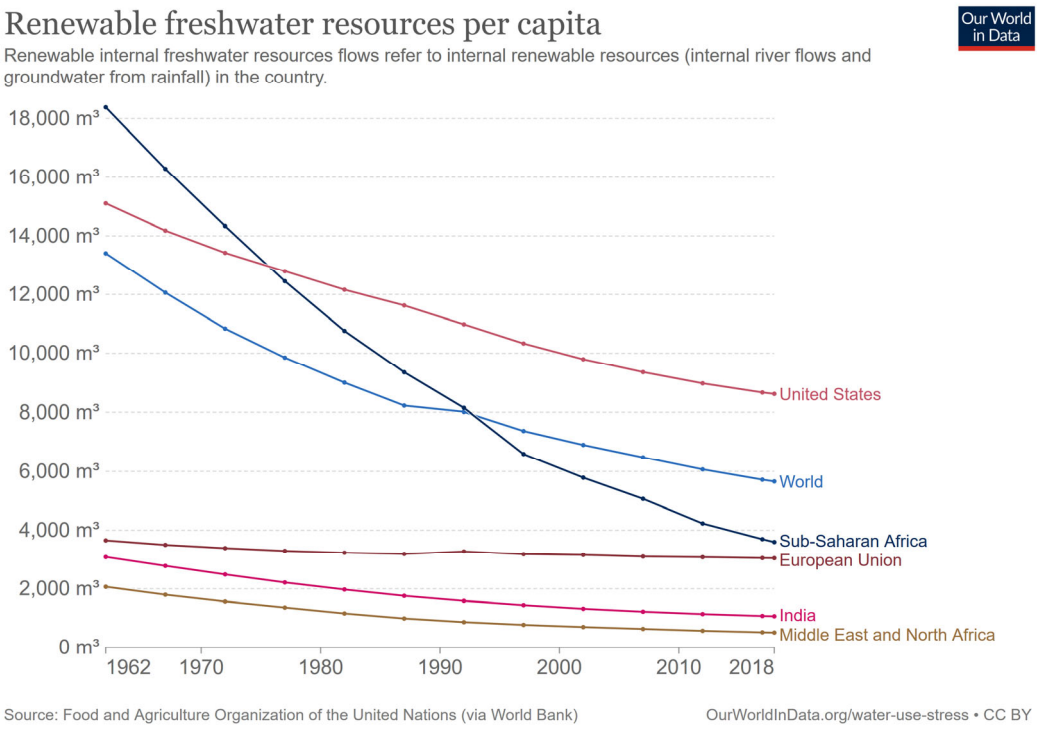


Figure 1.3. Evolution of population fed with fertilizers (source: https://ourworldindata.org/; accessed 15th March 2023).



a)



b)

Figure 1.4. Evolution of (a) freshwater withdrawal and (b) renewable freshwater availability per capita (source: https://ourworldindata.org/; accessed 15th March 2023).

Finally, Figure 1.4 illustrates the increment in water use and freshwater depletion. As depicted in Figure 1.3, Green Revolution (1950-1960) represents a milestone from which water consumption accelerates mainly due to its use in agriculture. In this sense, the world average of crops irrigation is estimated in 80% of freshwater extractions. In Asia, Middle East and North Africa, agriculture use of water rises up to 85 %, while in the EU average is 44 %, with southern member states reaching up to two thirds of total water abstraction. Irrigated areas account for nearly 50% of world's food supply and the area devoted to irrigated crops has almost doubled in the last 50 years, contributing to add pressure on water bodies (Antonelli et al., 2017; Dutta, 2021). Moreover, climatic change is expected to cause widespread spatial and temporal variability in water accessibility with social and governance implications, i.e., transboundary disputes and increased boarder conflicts (Angelakis et al., 2021). In this context, studies have been published anticipating that food demand is expected to increase globally by 30% to 62% between 2010 and 2050 (van Dijk et al., 2021). Therefore, in addition to developing actions to promote savings and efficiency of water usage, alternative sources must be explored to guarantee access to water from a perspective of economic, environmental and social sustainability (Aldaya et al., 2019).

Faced with these challenges, several strategies have been put in place to reduce the consumption of materials and energy with the aim of matching their use to the capacity of the planet. Among them can be mentioned:

- **Cleaner production (CP):** United Nations Environment Programme (UNEP) defined (CP) as *the continuous application of an integrated preventive environmental strategy to processes, products, and services to increase overall efficiency, and reduce risks to humans and the environment*. In the production processes, CP assesses potential actions for saving energy and materials, replacing toxic substances, and preventing toxic waste and emissions. Focusing on product design and development, CP addresses reducing environmental impacts during the entire life cycle. Finally, CP also incorporates the environmental perspective in the services design and delivery.
- **Eco-design:** this strategy aims at designing products and services considering their environmental impact to minimize them during their entire life cycle. Hence, the eco-design partially internalizes the environmental costs and seeks more sustainable means of production, as well as manufacturing materials that are easily reusable or recyclable.
- **Circular Economy (CE):** CE arises in opposition to the traditional linear economy based on the extract-manufacture-acquire-use-dispose sequence. The CE aims at developing an economic system based on the maintenance of energy and material flows in the economy,

so that the waste from one process can become the raw material for others. In this way, the extraction of raw materials from the natural environment is reduced and the emission of waste into the environment is minimized.

- **Degrowth/Post-growth:** Contrary to the previous strategies, degrowth (or post-growth) considers that only a conscious reduction in the consumption of materials and energy, and therefore, a reduction in the economy, would allow balancing the capacities of a planet of finite resources with the needs of living beings. This reduction must be carried out in an orderly and fair manner, with the aim of guaranteeing the right to a dignified life for the entire population, established in the Universal Declaration of Human Rights (Kallis, 2011). For degrowth, technological development or production efficiencies improvement are not solutions for resource depletion since the main problem consist of the existence of an unsustainable way of life.

In any case, moving towards sustainability means reducing inefficiencies in the use of resources and energy and matching the extraction of raw materials with the capacity of ecosystems to renew them, while intensifying efforts to reuse resources already present in the economic systems and reducing waste generation (Marín-Beltrán et al., 2022). In this scenario, concept shift of wastewater from waste to resource source can play a significant role.

1.2. Resource recovery from wastewater

1.2.1. Historical perspective of resource recovery from wastewater

The use of urban wastewater (UWW) as source or resources for agriculture purposes has a long tradition in human story (see Figure 1.5). The first documented infrastructures aimed at channeling UWW to farmland are found in Crete and Indus Valley and dates to 3000 BC, during the Bronze Age. The development of permanent settlements generated the need to establish UWW collection and evacuation systems. Primitive systems evolved during ancient times, in which civilizations such as the Romans set up large hydraulic works. However, the passage from ancient times to the Middle Ages meant a setback in UWW management. In Europe this meant an increase of epidemics of water borne diseases. There are studies that indicate that up to 25% of the population died for these reasons during the Middle Ages.

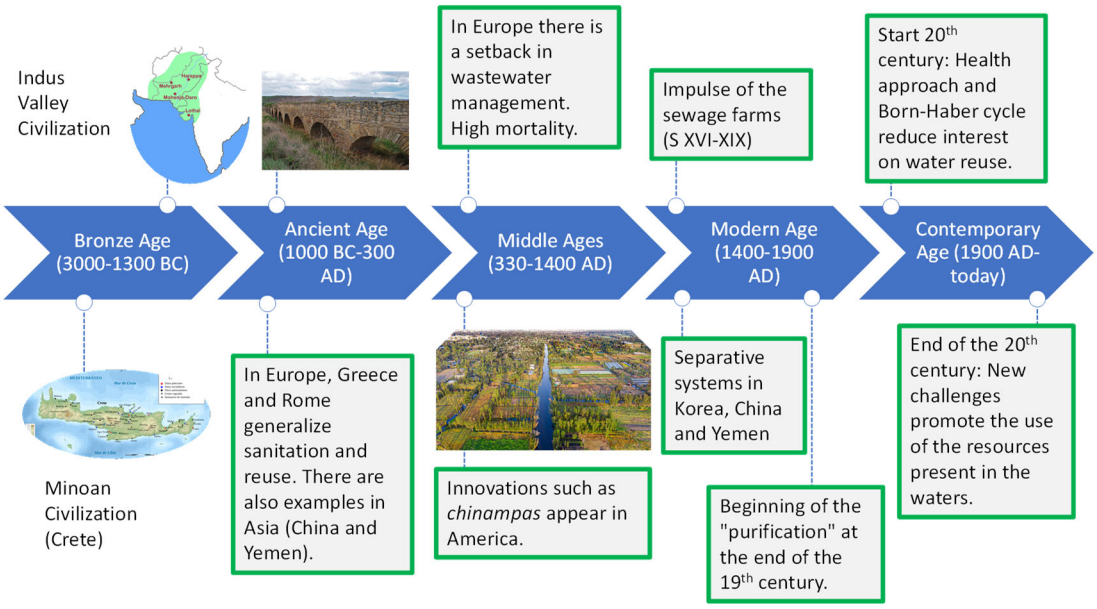


Figure 1.5. Historical evolution of agriculture water reuse (adapted from Angelakis et al. (2018)).

During Modern Age, the increase in population and urban consolidation once again promotes measures to evacuate UWW and use it for agriculture. In this sense, the earliest sewage farms, where UWW was planned for its systematic use in agriculture, were identified at Bunzlau (Poland, 1531) and Edinburgh (Scotland, 1650). Its implementation was imposed and became widespread from the 18th century in Europe and the US and the 19th century in Australia. Finally, the increase in the population of cities and the limitations of space available for sewage farms, prompted the development of mechanical treatment systems.

At the beginning of 20th century, interest in nutrients present in UWW decreased thanks to the production of mineral fertilizer facilitated by the Haber-Bosch process or availability of guano sources. However, at the end of the 20th century, interest in the use of the resources present in sewage was resumed, driven by the increase in the world population, urban growth, the decrease and deterioration in the quality of conventional water resources, the effects of climate change and technological development (Angelakis et al., 2018).

1.2.2. Opportunities for resource recovery from urban wastewater

1.2.2.1. Reclaimed water

The most obvious resource present in UWW is water itself. Reclaimed water and its reuse show some significant advantages to address climate change and drought issues:

- **Flow stability:** where supply of drinking water is the top priority within water resource management systems, e.g., Spain (Gómez-Limón et al., 2021), UWW shows a regular flow towards WWTP. After treatment, the effluent is available throughout the year and contributes to providing a reliable supply of water with less risks that other conventional sources such as aquifers or surface water bodies can present in the event of scarcity or drought.
- **Lower cost than other alternatives:** water reuse is already considered as a lower-cost measure to increase water resources than desalination or water transfer between river basins under certain conditions (European Commission-DG ENV, 2012; Jodar-Abellan et al., 2019).
- **Prevention of regional conflictive:** water reuse, as a strategy for conserving conventional freshwater sources, can also contribute to reduce competition for resources and alleviate water conflicts (Angelakis et al., 2021).
- **Food safety:** increased water availability helps to guarantee crop production and hence food safety. In this sense, peri-urban agriculture, understood as crop lands located where both rural and urban characteristics share space in the surroundings of a consolidated city (Kurian et al., 2013), can greatly benefit from water reclamation, being close to the treatment facilities. In addition, this type of agriculture contributes to improving the environmental indicators related to water-food nexus, as crops are generally produced near its place of consumption (Caputo et al., 2021).
- **Employment and local business:** water represents an essential input for economy activity and therefore jobs creation. Increasing water resources and promoting technological innovation would act as development driver for raising employment and social sustainability indicators. As way of example, Alcalde Sanza & Gawlik, (2014) reported that in Europe 20000 jobs could be created per 1% of growth of the water sector.
- **Environmental benefits:** water reuse shows environmental benefits, among which the following can be mentioned: i) reduction of effluent discharges into natural water bodies; ii) when fertigation is considered, production of mineral fertilizers is reduced and so its

environmental impacts; and iii) lower carbon footprint than other alternatives, e.g., desalination (Kirhensteine et al., 2016).

- **Conventional water resource preservation:** water reuse reduces withdrawals from conventional sources, e.g., groundwater, rivers or lakes. Therefore, reclaimed water contributes to reduce freshwater depletion (Garcia & Pargament, 2015) and restoration of water ecosystems.
- **Quality fit-for-purpose:** reclaimed water fit-for-purpose can be produce by means of physic-chemical and biological processes combined with appropriate technologies (Helmecke et al., 2020). This approach allows production costs to be adjusted to the destination of the water and hence improve the sustainability of the reclamation system.

On the other hand, barriers and bottlenecks have also been reported:

- **Regulation heterogeneity, lack of it or overregulation:** appropriate regulation is a principal requirement for ensuring safe (both in terms of health and environment) water reuse practices. Heterogeneity, absence or overregulation prevent the necessary legal certainty or hinder water reuse expansion (Morris et al., 2021).
- **Environmental and health risk:** unplanned water reuse represents an environmental and health risk due to the presence of chemicals and pathogens. To address this issue, multibarrier approach, appropriate water application and consistent risk management makes it possible to take advantage of the benefits of reclaimed water (Helmecke et al., 2020).
- **Social acceptance:** recognition of the benefits and safety of water reuse in agriculture by end users and citizens still represents a challenge. Reasons for this rejection could include: lack of knowledge about regulation and advantages of this practice; low level of involvement of farmers and policy makers; perception of related costs; and concern about its effects on the soil or crops (Saliba et al., 2018).

Finally, some regions became aware decades ago of the opportunities presented by planned water reuse and have even moved towards potential indirect and direct potable reuse, as is the case of California (Olivieri et al., 2020). On the other hand, reclaimed water remains underexploited in places with great potential and the need to increase its available resources. As way of example, Pistocchi et al. (2018) estimated the effluents of WWTPs in Europe to be 48 km^3 per year and 13 of them could be brought to crop fields, which would represent about 31 % of European irrigation demand (estimated in 42 km^3 per year). Currently, only 1.1 km^3 of treated UWW is reused in the

EU and plans for increasing water reused up to 6 km^3 in 2025 have been set, by improving regulatory and financial framework (Kirhensteine et al., 2016). In the case of Spain, its Mediterranean rim regions (i.e., Cataluña, Comunitat Valenciana, Murcia, Andalucía, and Islas Baleares) show high variability of water reuse implementation and time evolution (see Figure 1.6). As average, Spain reused 10.9 % of the total treated UWW in 2020 and it has remained unvaried since 2013. The region with a highest percentage of reuse is Murcia, which reached 91.4 % and it seems to have made a firm commitment to integrating reclaimed water into its resource management. Comunitat Valenciana and Islas Baleares also reuse a significant percentage of their treated UWW (42.5 % and 45.4 % in 2020, respectively). Finally, in 2020 Cataluña (5.4 %) and Andalucía (5.2 %) show very low reuse ratios. There is, therefore, scope for improvement to take advantage of these alternative resources.

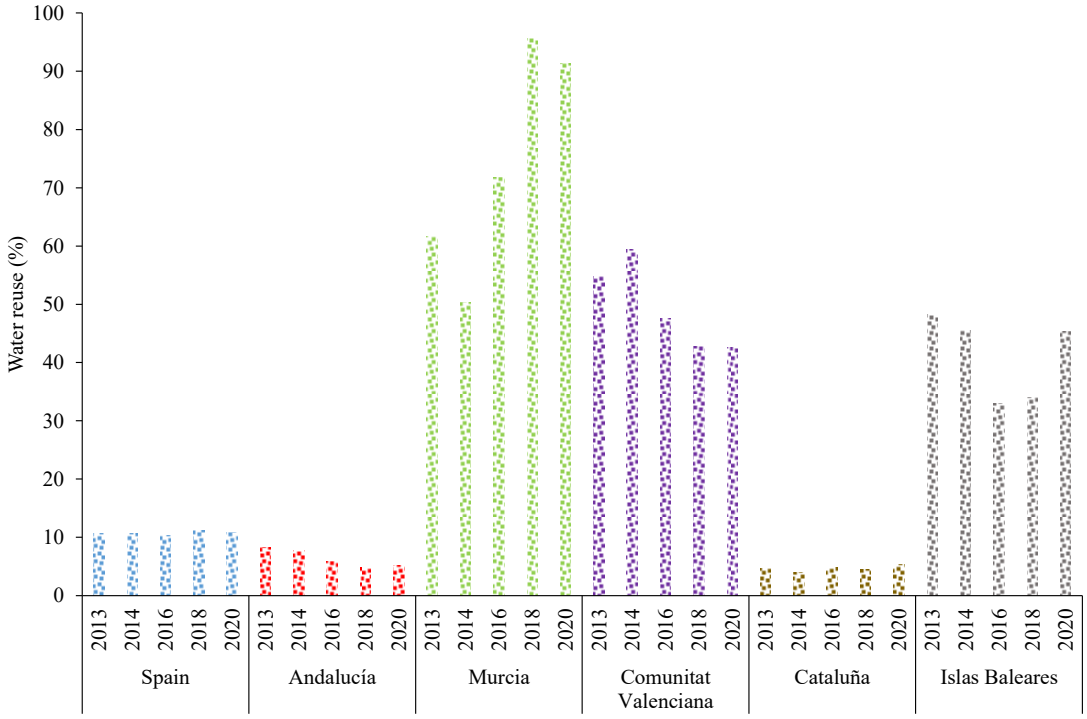


Figure 1.6. Proportion of water reused from treated wastewater (source: Spanish National Statistical Institute)

1.2.2.2. Nutrients

Processes and technologies for the removal of nutrients have been widely applied in water treatment to reduce the environmental impact of effluents and to improve the performance of facilities by reducing problems of incrustation and reduction of effective volumes in the reactors due to precipitation or inhibition of biological processes (Bunce et al., 2018; Mishra et al., 2022). However, UWW can also play a significant role to close nutrient cycles, reducing mineral

fertilizer needs and therefore reducing fertilizing dependency, cost and associated environmental impacts. Major nutrients for crop growth are nitrogen (N), phosphorus (P) and potassium (K) and UWW contains them in higher concentration than conventional freshwater used for irrigation (Shtull-Trauring et al., 2022). In this sense, Qadir et al. (2020) estimated in 16.6 Tg (Tg = million metric ton), 3.0 Tg and 6.3 Tg the amount of N, P and K available in UWW per year worldwide, which would represent 13.4 % of their global demand. Therefore, given the opportunities and benefits of nutrients recovery, great effort has been made to develop technologies to move from their removal to their recovery. A selection of them at different technology readiness level (TRL), focused on N and P, is presented below:

Nitrogen

In the case of nitrogen, the most common process for its recovery is air stripping. Basically, this system consists of a stripping tower which contains a packing media. Ammonia-rich water enters the tower from the top of it, and a current of air flows counter current from the bottom. The ammonia-laden air is then bubbled into an absorber in contact with acid to form the acidic ammonia solution. This process is well-stablished, but it shows some bottlenecks, e.g., scale formation or reduced mass transference at low temperature (van der Hoek et al., 2018).

Bioelectrochemical systems combines electrodes and microorganisms to recover nitrogen through oxidation/reduction processes in electric cells with the participation of an ion exchange (IEx) membrane. Oxidation of organic matter generates an electron flow and a migration of ammonium from the anode to the cathode. Then the ammonium turns into ammonia due to the high pH and recovered in an acid solution via absorption. This technology is still at lab phase, but there is interest in its development. Rodríguez Arredondo et al. (2015) reported removal rates of 10 (standard deviation not available) and 173.3±18.07 g $N \cdot m^{-2} \cdot d^{-1}$ for microbial fuel cells and microbial electrolysis cells, respectively.

Electrodialysis (ED) is based on a sequence of IEx membranes which generate dilute and concentrate chambers with two electrodes at the ends. Once the UWW enters the system the electric field induced by the electrodes causes the migration of ions towards the electrode of opposite sign. The presence of the cation/anion selective membranes generates cation-rich chambers and anion-rich chambers. Nitrogen recovery through ED involves its combination with stripping and absorption. Recoveries in the 85-96 % of nitrogen have been reported (Chen et al., 2021).

IEx has received recently increased attention for nitrogen (and phosphorus) recovery since some limitations (resins selectivity, bed clogging and regeneration cost) are being addresses and improved performance of this technology is being achieved. IEx consists of a substitution of an ion attached to a resin by another ion present in the water, e.g., Na^+ of the resin is substituted by NH_4^+ of water. The valuable ion is then recovered through a regeneration solution for the resin. Huang et al. (2020) carried out an economic evaluation of an IEx system in a 10000-population equivalent WWTP and results were compared to conventional biological nutrient removal. Three treatment schemes were assessed: Activated sludge with nutrient removal and iron dosing as tertiary treatment (S1); activated sludge combined with IEx (S2); and upflow anaerobic sludge bioreactor coupled with IEx (S3). Obtained results for a 40-year whole life cost showed S2 and S3 to be 17 % and 27 % lower than S1, respectively, while estimation of greenhouse gas (GHG) emissions were 936 t CO_{2eq}·$year^{-1}$ (S1), 507 t CO_{2eq}·$year^{-1}$ (S2) and 171 t CO_{2eq}·$year^{-1}$ (S3). Both economic and GHG considerations demonstrate the potential of IEx for urban wastewater treatment (UWWT) and nutrients recovery.

Hollow fiber membrane contactors (HFMC) have also been applied for nitrogen removal with promising results. This technology consists of a gas-permeable membrane separating a nitrogen-rich solution at high pH (> 8.6) and an acid solution. The transfer of ammonia is driven by the concentration gradient between both sides of the membrane. Once the ammonia enters the acid solution, it is converted into ammonium, and it can be used as fertilizer source. As way of example, Noriega-Hevia et al. (2021) evaluated the economic performance of a HFMC for nitrogen recovery in form of ammonia sulphate solution. To this aim, a 40000 m^3·d^{-1} WWTP combined with a membrane contactor was simulated, most favorable operating conditions were defined, and capital and operating cost were estimated. Global warming potential was also considered considering energy consumption and N_2O emissions. Obtained results showed total cost of 0.0095 €·m^{-3} and profit of 0.0090 €·m^{-3} (corresponding to sells of ammonia sulphate and savings due to diminishing nitrification requirements). Furthermore, a reduction of 10.3 t CO_{2eq} per day were estimated, since N_2O emissions associated to nitrification-denitrification process decreases and equivalent mineral fertilizer production is avoided.

Microalgae culture (MC) also can be used for nitrogen recovery. In these processes, microalgae removed nitrogen from UWW by incorporating it to their cellular structure. Then, microalgae can be harvested and treated for different uses (added-value products, biofuels, fertilizers, etc.). In this sense, Acién Fernández et al. (2018) estimated a microalgae recovery potential in the 2-5 t·ha^{-1}·$year^{-1}$ range under low solar radiation conditions and between 5 and 25 t·ha^{-1}·$year^{-1}$ in

tropical locations. However, the authors also identified some bottlenecks: need of long hydraulic retention time (HRT), high surface requirements, optimization gaps or lack of demonstration at industrial scale.

Phosphorus

Biological recovery of phosphorus (BRP) is based on polyphosphate accumulating organisms (PAO). PAO metabolism allows these bacteria to accumulate P (luxury uptake) as polyphosphate under aerobic conditions and release P under anaerobic conditions, but to a lower extend than the previous luxury uptake. As a consequence and with appropriate requirements, a UWWT with an aerobic-anaerobic sequence with presence of PAO shows a net P removal from UWW. Guisasola et al. (2019) evaluated the stability of an enhanced biological phosphorus removal (EBPR) system with a recovery perspective and obtained P recovery efficiencies up to 60 % with an adequate balance between the extraction of supernatant at the end of anaerobic phase for P recovery and PAO requirements in the EBPR system.

P recovery from sludge is also possible through incineration and subsequent acid leaching. Apart from P recovery, this process shows as advantages reduction of sludge volume, destruction of pathogens and energy valorization potential. However, heavy metals can also be transferred to the obtained solution and additional purification processes might be needed. Liang et al. (2021) studied P recovery from ashes of incinerated sewage sludge with and without pre-treatment. Results show efficiencies of P leaching between 83 and 100 % and close relation with Ca content in ashes, which presence hinders P release and recovery.

IEx and ED follow the same process as explained for nitrogen. Finally, as in the case of nitrogen, MC also allows phosphorus to be recovered by another biological route different from PAO bacteria. Acién Fernández et al. (2018) reported microalgae recovery potential of 0.2-1.1 $t \cdot ha^{-1} \cdot year^{-1}$ with low solar radiation and between 0.5 and 2.5 $t \cdot ha^{-1} \cdot year^{-1}$ in tropical areas.

Nitrogen + Phosphorus

Struvite is a magnesium-ammonium-phosphate compound ($MgNH_4PO_4 \cdot 6H_2O$) with high agronomic properties as fertilizer. In WWT context, it represents an opportunity for creating revenues that contribute to the economic sustainability of WWTP, with a jointly valorization of the nutrients presents in UWW. Struvite can be produced in the WWTP in a crystallization reactor by adding Mg under controlled operating conditions. As way of example, Bouzas et al. (2019)

assessed the performance of the implementation of P-recovery system combining and improved sludge line configuration (including elutriation in the gravity thickener) and a struvite crystallization reactor in Calahorra WWTP (Spain). Because of the implementation of the new treatment scheme, P recovery raised from 9 % to 19 % and estimation of struvite production was 15 $t \cdot year^{-1}$ treating supernatant of the thickener. Authors also highlighted that struvite production could reach 38 $t \cdot year^{-1}$ if centrate stream could also be treated in the crystallization reactor.

Direct sludge application in farmlands represents a common practice to valorize the nutrient content (and organic matter) of this waste stream. Dehydrated sewage sludge contains 4 % of nitrogen and 2.5 % of phosphorus, approx., which imply a significant contribution to the soil after its application. However, this practice is being questioned, due to the possible risks that the presence of emerging contaminants in the sludge (drugs, antibiotic resistant genes, etc.) may have for both the environment and human and animal health. Although the real effect of these substances is still controversial, a tightening of the conditions for the application of sludge or even its prohibition is being contemplated in places such as the EU (Buta et al., 2021; Collivignarelli et al., 2019).

Fertigation, understood as planned direct agriculture reuse of the WWTP effluents with their nutrients, is gaining momentum since it would be a simple practice to recover nutrients and save conventional water resources. According to the irrigation method and crop, different qualities for reclaimed water can be obtained to guarantee environment and health safety. Appropriate nutrient management on crop land, water quality preservation along irrigation system, water cost and tariffs, long-term effect on soil properties, and governance are also key elements to address in order to boost fertigation under sustainable criteria. Bolinches et al. (2022) have developed a methodology to quantify investment and operation cost of water reuse projects considering the benefit of the new water source and applied this method to the Upper Guadiana basin (Spain). Capital expenditure (CAPEX) for reclaimed water facility and operation and maintenance costs shows the highest influence on overall cost and net benefits and economic feasibility depends on crops whose irrigation imply a significant additional value.

1.2.2.3. Energy

UWW contains a significant amount of energy in different forms, available for its recovery. These sources are organic matter, nutrients and thermal content (Cornel et al., 2011).

The harnessing the energy present in the organic matter is usually carried out through anaerobic treatments (see section 1.3.2.1), in which this organic matter is partially transformed into biomethane, which, in turn, can be used as fuel to produce electricity and heat, e.g., via combined heat and power (CHP) systems, or injected in the gas grid. Also sludge can be valorized for energy recovery through direct incineration or its conversion into fuel material, e.g., hydrochar, via hydrothermal carbonization (HTC) (Wilk et al., 2021).

In the case of nutrients, in addition to their value as fertilizers, it is also important to consider their energy content, the result of considering the energy involved in producing the same amount that is in UWW in the form of mineral fertilizers. N-based fertilizers are mainly produced after fixation of atmospheric nitrogen via Haber-Bosh process, which is a very energy-intensive method (9-13 $kWh \cdot kg^{-1}$ NH_3-N) (Cornel et al., 2011). P is obtained from mining of phosphorus rocks and, according to Gellings & Parmenter, (2004), this process involves an energy consumption of 2.11 $kWh \cdot kg^{-1}$ P.

Finally, the effluents of WWTP contains internal energy that manifests itself through its temperature. This energy can be used for heating or cooling purposes in the WWTP under certain conditions. By means of taking advantage of this thermal heat, calorific energy obtained from anaerobic digestion can be applied for heating external cycles and contribute to climatize local populations. In this sense, Kretschmer et al. (2021) applied this approach to a case study located in the Kapfenber WWTP (Austria; 6700 $m^3 \cdot d^{-1}$) equipped with anaerobic digestion and a CHP unit to supply internal electricity and heat and also to supply to a residential area of 220 households. Results showed that WWTP internal heat demand could be met and up to 50% of the residential heat demand could be covered with the energy provided by the WWTP.

Some authors have quantified the potential energy content available embedded in UWW. For example, Table 1.1 shows some average values reported by McCarty et al. (2011) for a typical domestic wastewater.

Krzeminski et al. (2017) reported energy consumptions between 0.3 and 0.6 $kWh \cdot m^{-3}$ for CAS and in the 0.6-2.3 $kWh \cdot m^{-3}$ range for aerobic MBR. Therefore, values in Table 1.1 would indicate that energy content in domestic wastewater could potentially cover energy requirements for CAS and significantly reduce energy expenditure in case of aerobic MBR.

Table 1.1. Energy content for a domestic wastewater (adapted from McCarty et al. (2011)).

Parameter	Concentration ($mg \cdot L^{-1}$)	Equivalent energy ($kWh \cdot m^{-3}$)
BOD*	320	1.23
NT	40	0.77
PT	8	0.02
Thermal heat	n.a.	7.00

*Biological oxygen demand

1.2.2.4. Others

Apart from reclaimed water, nutrients and energy, UWW contains other resources whose recovery is the subject of numerous studies. However, these processes are still in many cases at lab scale or pilot scale, so it is still necessary to delve into them to guarantee their sustainability on an industrial scale. Among these resources, the following can be mentioned: hydrogen (H_2), proteins, cellulose, volatile fatty acids (VFA), alginate, fodder, metals, or bio-based biopolymers.

As way of example, Puyol et al. (2017) evaluates emerging technologies for resource recovery during UWWT. Apart from energy and nutrients, the authors identified H_2 production through biophotolysis, photofermentation and dark fermentation as environmentally favorable. Also, biopolymers are mentioned (e.g., polyhydroxyalkanoates (PHA)), since their thermomechanical properties are similar to those of plastics obtained from fossil fuels. PHA production still faces some bottlenecks: use of pure cultures and sterile feedstocks increase costs; low PHA yields from mixed cultures; and extraction and purification. The use of UWW can contribute to lower feedstock expenditures and PHA production can be coupled to other UWWTs, e.g., nutrients removal. Moreover, additional high added value substances, e.g., proteins, can be produced from MC and UWW. Finally, some processes and biological technologies focused on metals recovery (including heavy metals and metallic radionuclides) are also exposed.

Kehrein et al. (2020) identified 11 resources embedded in UWW and technologies for their recovery, again including reclaimed water, energy (methane, biofuels) and nutrients. Among the others, VFA (acetate, propionate, valerate and butyrate) and extracellular polymeric substances (EPS) are mentioned as valuable raw materials for producing high-value products. Cellulose recovery (from toilet paper) is also considered, since it improves WWTP performance and can be

used as soil conditioner, fuel, aggregate for construction sector and feedstock in paper pulp industry. Moreover, membranes are considered as key technologies for water reclamation and reuse. The authors also depicted the main bottlenecks for resource recovery divided into three groups: economic and value chain; environment and health; and society and policy. Within economic and value chain issues are process costs; resource quantity and quality; market value and competition; utilization and application; and distribution and transport. Emissions and health risks constitute the second group. Finally, acceptance and politics make up the third group.

Industrial wastewater shows also great potential for resource recovery and efforts are underway to increase the possibility for materials recovery. The variety of these waters is high, so only a few relevant possibilities will be mentioned here: rare earths; precious metals; metals; nutrients; and inorganics, organics and chemicals (Birungi et al., 2020; Devda et al., 2021; Taghvaie Nakhjiri et al., 2022; Walling et al., 2022).

1.3. Urban wastewater treatment

UWW management, understood as a widespread and comprehensive planification for collecting, treating, and disposing the effluents with the objective of assuring public health and environment preservation is relatively new. Some important urban agglomerations of ancient civilizations developed to some extend drainage systems connected to latrines (e.g., Mesopotamian Empire, Ancient Greeks, etc.) or even sumps to act as settlers before discharge (Indus Civilization) (Angelakis, 2017). The Romans perfected water management by conceiving a system that included supply, collection, cesspools and discharge (Lofrano & Brown, 2010) but the collapse of the Roman Empire and the beginning of the Middle Ages led to the abandonment of the UWW management infrastructure put in place in the previous era. It was not until the 19th century that UWW management, including its treatment, was seriously promoted again (Vuorinen et al., 2007). In this sense, the description of the first biological UWWTs based on activated sludge (AS) (Ardern & Lockett, 1914) represents the milestone that marks the beginning of modern water purification.

1.3.1. Conventional treatment

Current UWT treatment (UWWT) consists of a set of physic-chemical and biological process following a sequence of basic units organized in a facility commonly called wastewater treatment plant (WWTP), which objective is to remove pollutants and produce an effluent without

environmental and health risks. Within UWWT, the biological reactor is considered the core process in the facility. Its combination with a secondary settler and a sludge recycling stream back to the reactor (see Figure 1.7) constitutes what is known as conventional activated sludge (CAS) process.

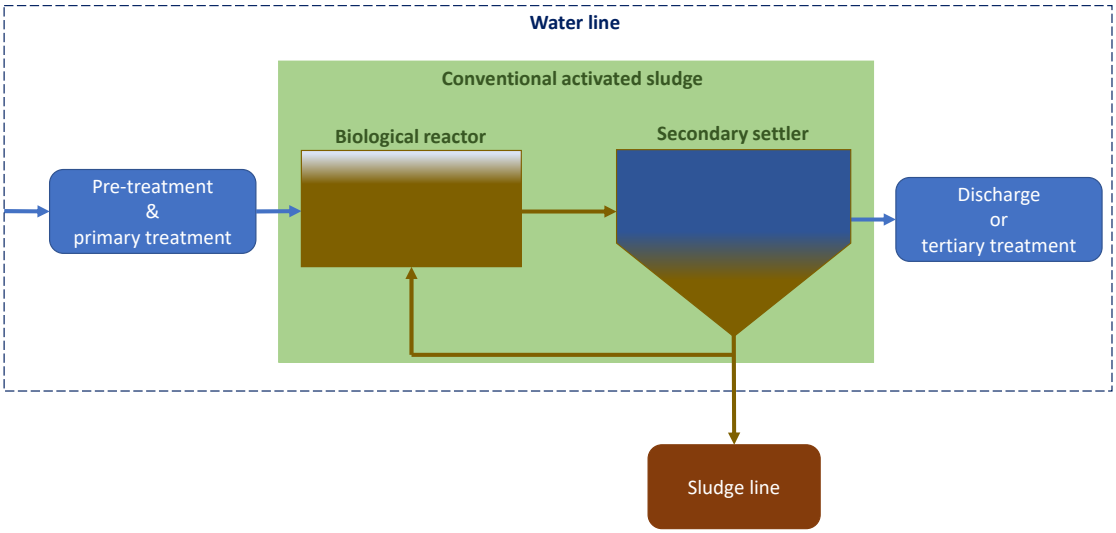


Figure 1.7. Conventional activated sludge (CAS) process.

CAS is the most common biological process applied for UWWT and has been intensively studied since its first description (Orhon, 2013). Different configurations and layouts of CAS have been designed, constructed and operated over time (Gernaey et al., 2004), e.g., modified Ludzack-Ettlinger, Cape Town University configuration, A^2/O, etc. This versatility of CAS allows designers and engineers to address biological removal or organic matter, nitrogen and/or phosphorus. In this sense, CAS contribution to water sanitation has been crucial (Sheik et al., 2014).

However, CAS is based on aerobic metabolism of microorganism, which turns into high oxygen availability requirements to maintain the pollutants removal capacity in the biological reactor. Since the oxygen is supplied from the injection of air driven by blowers, the energy consumption associated with these processes is significant. As way of example, Hernández-Sancho et al. (2011) quantified this expenditure up to 60% of total energy consumption in a WWTP. A similar value, range of 50-70 %, was reported by Capodaglio & Olsson, (2020).

WWTP are also a source of GHG emissions. According to the IPCC2013 application guides, CO_2 emissions from biogenic origin, i.e., emissions from organic matter oxidation, do not contribute to carbon footprint. However, to the extent that the considered energy mix contains contributions from fossil fuels, the energy consumption associated with the UWWTs will be associated with a certain amount of emissions. Furthermore, during biological processes methane (CH_4) and nitrous oxide (N_2O) are also generated (Daelman et al., 2013). The quantity of these gases emitted into the atmosphere is comparatively small, but their influence is high due to their high greenhouse effect potential: 36 kg CO_{2eq} per kg CH_4 (Huijbregts et al., 2017) and 298 kg CO_{2eq} per kg of N_2O (Solomon et al., 2007), respectively.

Moreover, aerobic metabolism shows a high biomass growth yield, which means that WWT produces a high amount of waste sludge with managing needs and costs that affects the economic and environmental performance of sewage treatments. Additionally, the regulation of waste sewage sludge is also expected to be tightened (Collivignarelli et al., 2019), which adds incentives to look for processes that reduce sludge production.

From a resource-recovery perspective, CAS present some weaknesses. First, organic matter removal through aerobic metabolism completely oxidizes organic carbon into CO_2 and water, which means that energy embedded in the organic carbon is lost. Second, nitrogen removal via nitrification-denitrification process also reduces recover potential for this valuable nutrient, which industrial fixation from atmospheric N_2 for fertilizer production implies high energy consumption and includes also the use of natural gas (Epa, 2011).

Therefore, important efforts have been made to improve the economic and environmental performance of conventional UWWT to guarantee its sustainability for the next decades. To this aim, the characteristics of anaerobic metabolism allow organic matter valorization and diminishing waste sludge production, while the use of membranes introduces advantages in WWTP operation and disinfection. Finally, the combination of both anaerobic processes and membranes in the so-called anaerobic membrane bioreactor (AnMBR) add synergies such as higher methane yields than conventional anaerobic digestion, production of methane-enriched biogas and lower footprint.

1.3.2. Alternative treatment

1.3.2.1. Anaerobic processes

Anaerobic treatments comprise a series of stages characterized by different metabolic pathways, each carried out by distinct groups of microorganisms. These stages must be adequately balanced, since the absence of equilibrium between them implies the interruption of the set of processes. If anaerobic treatment proceed adequately, complex organic matter is converted into CH_4 and CO_2 (biogas). Other compounds could also appear if their precursors are present in the influent (e.g., HS^- would be part of the biogas if SO_4^{-2} enters the system). Figure 1.8 summarizes the main metabolic pathways during anaerobic treatment:

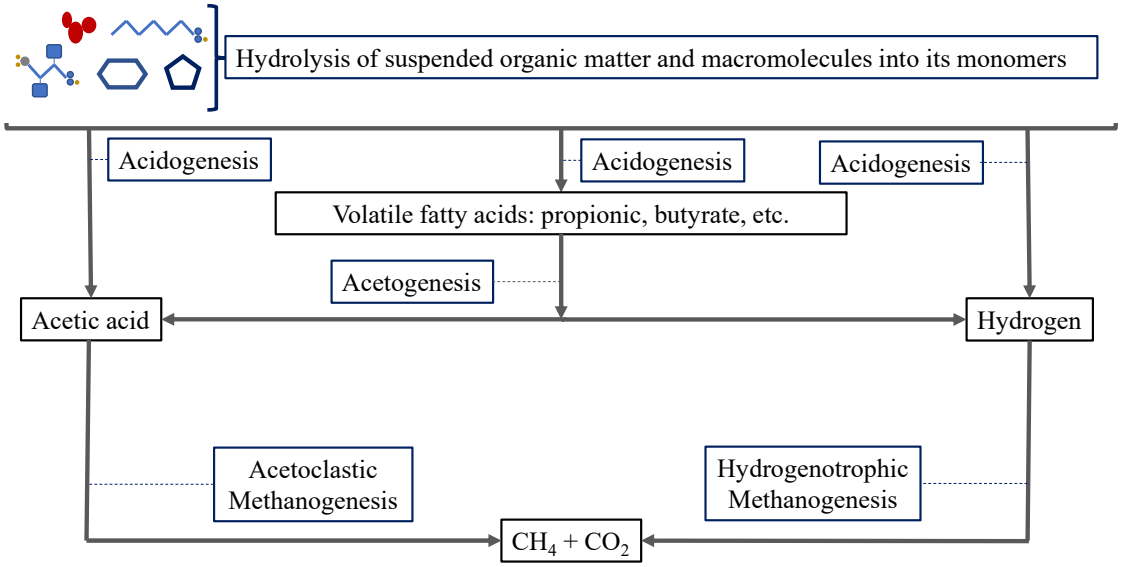


Figure 1.8. Scheme of the anaerobic stages involved in UWWT.

- **Hydrolysis:** during this step, extracellular enzymes fragment complex organic substances, i.e., carbohydrates, proteins, and lipids, into their smaller compounds (single carbohydrates, aminos acids, long-chain fatty acids). This stage is considered the speed-limiting process (Guo et al., 2021).
- **Acidogenesis:** in this step, acidogenic bacteria use compounds from previous step to produce organic acids (propionic, butyric, acetic) and hydrogen.
- **Acetogenesis:** during acetogenesis, acids are converted into hydrogen and acetic acid.
- **Methanogenesis:** in the final step, methanogenic archaea (MA) metabolize the hydrogen and acetic acid and turn them into CO_2 and CH_4.

Finally, if SO_4^{-2} is present, growth of sulphate reducing bacteria (SRB) will occur. These bacteria reduce SO_4^{-2} to HS^- while oxidize propionic acid, acetic acid and H_2. In this situation appears a competition between MA and SRB for the available substrate, where 2 kg of chemical oxygen demand (COD) per kg of sulphate are consumed by SRB. Since SRB have higher growth yields than MA, under low COD:SO_4^{-2} ratios sulphate content critically affects methane production by MA (Durán et al., 2020b).

Anaerobic treatment of UWW has several advantages over conventional aerobic processes, as has been widely recognized over the years. Its main advantages are: low sludge production due to low anaerobic biomass yield (Giménez et al., 2011); potential on-site biosolids stabilization due to high operating sludge retention time (SRT) and temperature; reduced energy consumption because no aeration is required (Lee et al., 2017); organic matter valorization through methane production, which improves the technology's energy balance (Krzeminski et al., 2017b); low GHG emissions when methane is recovered from both biogas and effluent streams; and recovery potential of nutrients content (Batstone & Virdis, 2014).

However, anaerobic process also presents bottlenecks that need to be considered to exploit its benefits:

- **Reduced growth yield:** this characteristic allows reducing sludge production, but also implies operating at high temperatures or at high SRT to achieve a significant concentration of biomass in the reactors or digestors.
- **Sensitivity:** anaerobic metabolism is more sensitive in the presence of inhibitors (e.g., acids, NH_3 or H_2S) or overloads.
- **Settleability:** anaerobic sludge has low settleability properties which hinder water-biomass separation with conventional settlers and forces to use high settling volumes.

The use of AD in UWW has been traditionally applied to sludge treatment, as part of its management before its final disposal (landfill, incineration, agriculture use, etc.). The low growth yield and poor settleability of anaerobic microorganisms hindered its application directly in the water line since high HRT and therefore reactor volumes would be needed. Nevertheless, some technological solutions have dealt with these drawbacks and have overcome them, e.g., by means of immobilization or retention of the biomass in the digestor, decoupling hence HRT and SRT. Examples of these technologies known as high rate anaerobic reactors (HRAR) are anaerobic

contact filters (CF), up-flow anaerobic sludge blanket (UASB), fluidized bed (FB), expanded granular sludge bed (EGSB), internal circulation (IC) or anaerobic baffled (AB) (van Lier et al., 2015). However, the implementation of these technologies for UWWT is scarce, being more frequently applied for industrial wastewater (food, brewery, paper, bagasse, etc.).

1.3.2.2. Membranes

Membranes are barriers capable of separating two phases and allowing the selective passage of substances through them depending on the nature of these substances and the characteristics of the membrane itself (Figure 1.9).

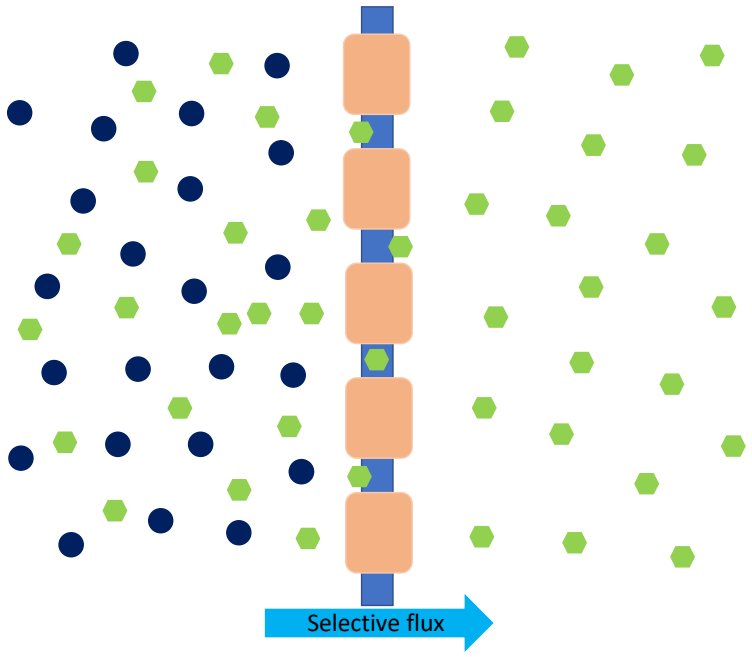


Figure 1.9. Graphical representation of a membrane filtration process.

Membranes can be classified attending to the material they are made from (e.g., polymeric, inorganic), configuration (e.g., tubulars, spiral wound, hollow-fiber, plate and frame, hollow fiber, flat-sheet) or driving force (microfiltration (MF), ultrafiltration (UF), reverse osmosis (RO), dialysis, ED). Among them, the most applied for WWT are polymeric; hollow fiber and flat sheet; and MF and UF.

Membranes have been known since the 18th century (Ezugbe & Rathilal, 2020) and their application to water treatment became widespread in the 1960s boosted by RO for desalination.

Since then, their use has increased and this technology has been integrated into different processes and stages of water treatment, including WWT, due to the advantages it provides. Among them, the following can be mentioned:

- **Improved removal control of pollutants:** membrane production and design include pore size or physic chemical properties selection. Therefore, it is possible to know what type of contaminants will be retained based on their size and/or nature.
- **Decoupling of HRT and SRT:** biomass retention allow**s** achieving high-rate conditions in the reactors and thus increasing biological activity and efficiency per unit of volume.
- **Removal of persistent pollutants:** biomass retention also increases biodiversity, which eventually facilitates persistent pollutants removal by means of microorganism adaptation to these substrates.
- **Smaller footprint:** dens configuration and efficiency make membrane system to be more compact than equivalent settlers or filters.
- **Disinfection without chemicals:** membranes act as a physical barrier that pathogens cannot pass through, as long as these microorganisms are larger than the pore size. Therefore, it is possible to disinfect the water without adding conventional chemicals or radiation. However, the lack of substances in the permeate that prevent the regrowth of microorganisms, may make possible the need for adding a secondary disinfectant to guarantee the quality of the downstream effluent.

However, the use of membranes also presents barriers and challenges that must be addressed to maximize the benefits that their use brings:

- **Membrane lifespan:** during the filtration process, a deposition of substances on membrane surface is produced, as well as incrustations and blockages of its pores. These processes reduce the performance of the membrane, so operating strategies are established to reduce their impact. These strategies include chemical cleanings (acid and basic) that allow the filtering capacity of the membranes to be recovered, but at the cost of reducing their lifespan.
- **Acquisition cost:** although the cost of the membranes has been reduced and is expected to continue this trend, it remains to be a significant item in the investment and replacement costs of these treatments (Judd, 2017).

- **Energy expenditures:** membrane operation include pumps and blowers which significantly increases energy consumption compared to conventional activated sludge processes (Lazarova et al., 2012)**.**
- **Fouling:** as aforementioned, the deposition of substances on and in the membranes causes the so-called *fouling*, which represents the main challenge for membrane-based treatments. It reduces the filtration productivity, affects membrane lifespan, and increases treatment costs. Thus, operation must be optimized to reduce fouling impact.

Membranes as primary treatment (Direct membrane filtration)

Introducing membranes in combination with or as alternative to primary settlers, known as direct membrane filtration (DMF) of raw UWW (Figure 1.10a) or of the supernatant of the primary treatment (Figure 1.10b), would lead to an improvement in the UWWT efficiency by an organic carbon redirection for its valorization through AD. This redistribution of organic matter reduces aeration needs in the water line and increases methane production and therefore enhances the energy balance of the treatment facilities. It would also reduce sludge production and N_2O emissions in the aerobic reactor. In this sense, Kimura et al. (2017) and Lateef et al. (2013) stated that up to 75% of the organic matter present in raw UWW can be redirected towards energy recovery by replacing primary settler with UF modules. Sanchis-Perucho et al. (2023) evaluated DMF of raw municipal wastewater and primary settler supernatant with UF and MF membranes. These authors concluded that MF membranes were affected by a severe pore blocking propensity (filtration periods of 2-8 hours) while UF performance significantly increased filtration performance up to 69 days.

Membranes as substitutes of secondary settler

Replacement of secondary settlers by membranes turns conventional AS into the so-called aerobic membrane bioreactor (AeMBR) technology. AeMBR operates with high biomass concentration by decoupling HRT and SRT and produce a high-quality effluent suitable for many purposes. It also reduces sludge production and improve its stabilization, compared to CAS (Collivignarelli et al., 2019). As main drawback, these treatment systems are characterized by high energy consumption associated to aerobic degradation and membrane scouring to prevent fouling. MBR energy requirements are commonly in the 0.4-1.6 $kWh \cdot m^{-3}$ range (compared to 0.3-0.6 $kWh \cdot m^{-3}$ for CAS), but it can raise up to 6-8 $kWh \cdot m^{-3}$ when treating high-strength UWW (Krzeminski et al., 2017b). Pretel et al. (2016) assessed the economic and environmental performance of AeMBR treating moderate-/high-loaded UWW and compared it to other aerobic-based technologies. Among their results, the authors obtained net energy demands of 0.21-0.23 $kWh \cdot m^{-3}$ for CAS and

0.50-0.56 kWh·m^{-3} for AeMBR. With regards to total annualized cost (TAC), 0.140-141 €·m^{-3} for CAS and 0.192-0.198 €·m^{-3} for MBR was calculated. Finally, AeMBR showed higher environmental impacts for global warming potential, abiotic depletion, and acidification, while CAS showed worse performance for marine aquatic toxicity.

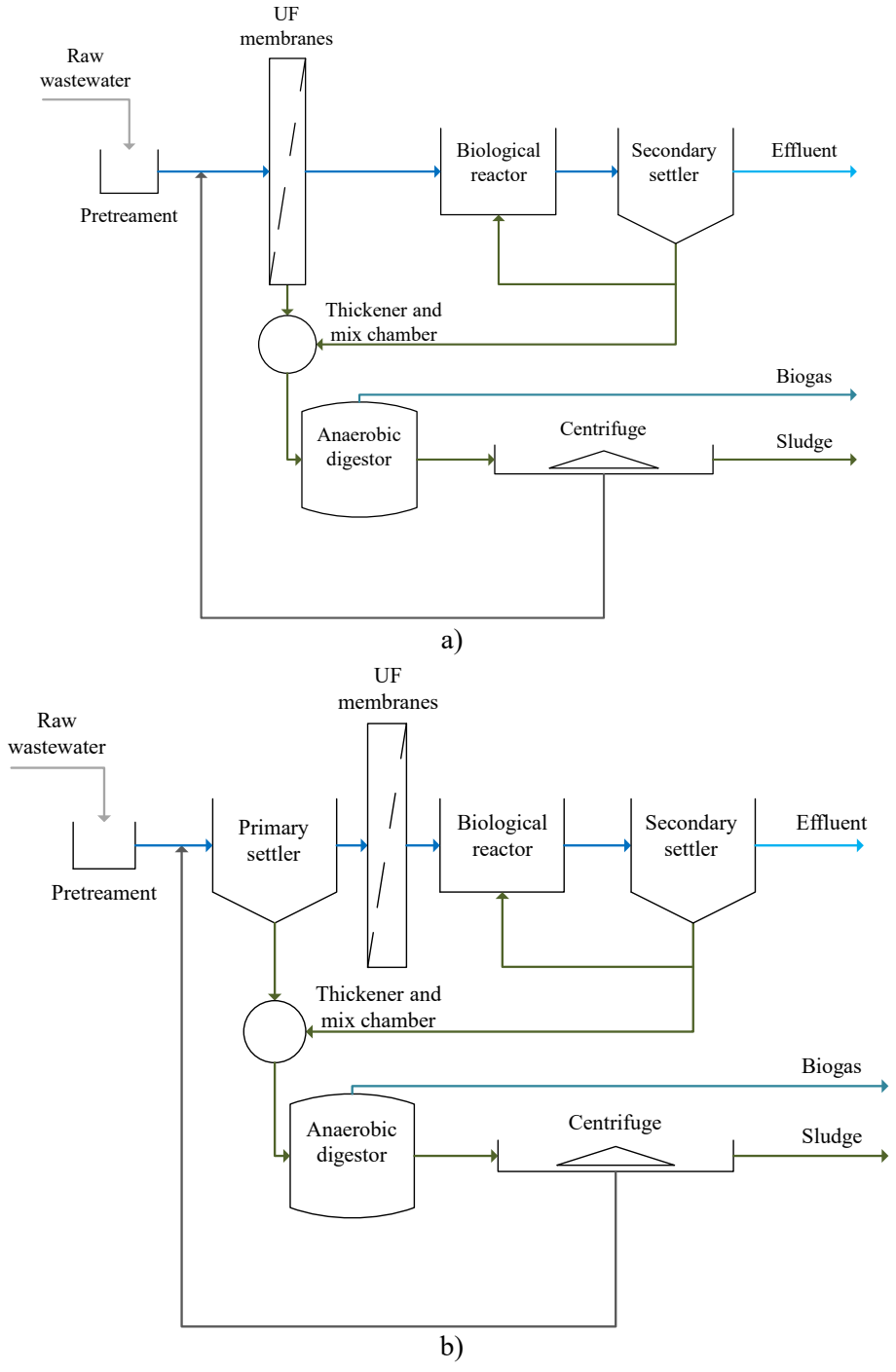


Figure 1.10. Flow diagram of the DMF process of: a) raw UWW and b) supernatant from primary settling.

Membranes as tertiary treatment

The application of membranes as tertiary treatment in water treatment seeks to achieve high quality in water, mainly in what refers to solids and pathogens. Since it is possible to select a membrane with a known pore size, the particle size that will be present in the permeate can be controlled, and therefore, reclaimed water fit-for-purpose can be produced. Literature shows a variety of studies aimed at evaluating tertiary treatments based on membranes, alone or in combination with additional processes. Also, combinations of membranes are common, e.g., UF+RO.

Bray et al. (2021) studied the performance of two different UF membranes of 200 kDa and 400 kDa for tertiary treatment of real treated UWW. Both membranes achieved high water quality in terms of total nitrogen (< 7.12 mg $N \cdot L^{-1}$), total phosphorus (< 0.2 mg $P \cdot L^{-1}$), turbidity (< 0.02 NTU), COD (<7.12 mg $\cdot L^{-1}$) and indicator bacteria E. Coli (5-20 CFU $\cdot L^{-1}$) and fecal enterococci (1-5 CFU $\cdot L^{-1}$). The authors also reported considerable reductions of prokaryotic cells and viruses' particles in the permeate (up to 2.5-3.0 $\log_{10}$ and 4 $\log_{10}$, respectively), but their presence was still significant in terms of quantity. Therefore, permeate quality could be suitable for irrigation purposes (according to Regulation EU 2020/471), but results for viruses and prokaryotic cells were not sufficient for considering a discharge into recreational areas due to health considerations. Ferreiro et al. (2021) combined UF with ozonation for the treatment of a WWTP effluent. Authors reported recovery factor up of 97.92 % of influent flow, concentrations of suspended solids below 1 mg $\cdot L^{-1}$, E. Coli of 0.0 CFU $\cdot$ 100 mL^{-1}, BOD_5 of 2.90 mg $\cdot L^{-1}$ and turbidity of 0.06 NTU. Hence, permeate achieved statements of Regulation EU 2020/741 for A-Quality reclaimed water. Yang et al. (2021) studied the feasibility of a semi-industrial UF pilot plant for UWW reclamation. Among their findings, the authors achieved sustainable long-term operation with transmembrane flux of 80 $L \cdot h^{-1} \cdot m^{-2}$, 40 min of filtration cycle and 1 air backwash followed by 3 classic backwashes. Moreover, reclaimed water accomplished with World Health Organization (WHO) guidelines and EU regulation. Finally, Pérez et al. (2022) evaluated the performance of a prototype (treatment flow of 2.5 $L \cdot m^{-3}$) with a combined UF and RO system for water reclamation to be industrially reused. The authors reported higher CAPEX and operating expenditure (OPEX) for UF than for RO, which is attributed to the pretreatment requirements and fouling. Total treatment cost of 1.096, 0.831 and 0.572 $€ \cdot m^{-3}$ for treatment flows of 2.5, 5 and 20 $m^3 \cdot h^{-1}$ were reported, respectively. These costs are competitive compared to the considered tap water price (0.963 $€ \cdot m^{-3}$).

1.3.2.3. Anaerobic membrane bioreactor

AnMBR technology merges AD and membrane filtration in one compact single unit, which allow taking advantages of both treatment processes benefits already described in Sections 1.3.2.1 and 1.3.2.2. AnMBR consists of an anaerobic digestor combined with membranes. According to the position of membranes in relation with the digestor, different configurations can be obtained: crossflow, submerged, or external submerged (Figure 1.11). The configuration of the AnMBR shows different characteristics, which influence its selection for a particular use.

Crossflow layout (Figure 1.11a) operates with high velocity of mixed liquor in the recirculation cycle containing the membrane. Filtration is also conducted with high transmembrane pressure and membrane scouring by means of biogas flow can be implemented. These elements imply high energy consumption (Gander et al., 2000). On the other hand, crossflow on membrane surface contributes to reduce fouling propensity. Submerged configuration (Figure 1.11b) operates with lower energy expenditures since avoids recirculation of mixed liquor and applies vacuum filtration, but transmembrane fluxes are also lower, which implies higher membrane surfaces, and gas sparging could be needed to reduce fouling development. External submerged (Figure 1.11c) is the most common configuration, since it allows operating at low transmembrane pressures by controlling membrane fouling by membrane scouring by gas sparging. Additionally, having the membranes separated from the reactor also facilitates maintenance tasks.

AnMBR performance has evolved since its first reported implementation in 1978 (Grethlein, 1978). Nowadays AnMBR technology represents a suitable option to be considered among the available UWWT technologies. Innovations in system configuration, operation modes, membrane design and combination with other technologies have allowed it to overcome or reduce some of its initial barriers related to efficiency and cost (Shahid et al., 2020). As way of example, Smith et al. (2015) reported COD removal efficiencies over 95 % at operating temperature as low as 6 °C; competitive energy consumptions have also been reported, even at moderate temperatures (15 °C) (Rong et al., 2022); Robles et al. (2021) reported long-term operation (250 days) without membrane chemical cleaning need; and Sanchis-Perucho et al. (2021) demonstrated the potential of degassing membranes to improve energy balance and reduce GHG emissions by recovering the dissolved methane from the effluent.

Finally, the generalization of AnMBR technologies in UWWT will depend on reaching a level of development that allows economic, social and environmental benefits from all the potential

products that this technology offers in its different streams, i.e., permeate, sludge and biogas (Hu et al., 2020).

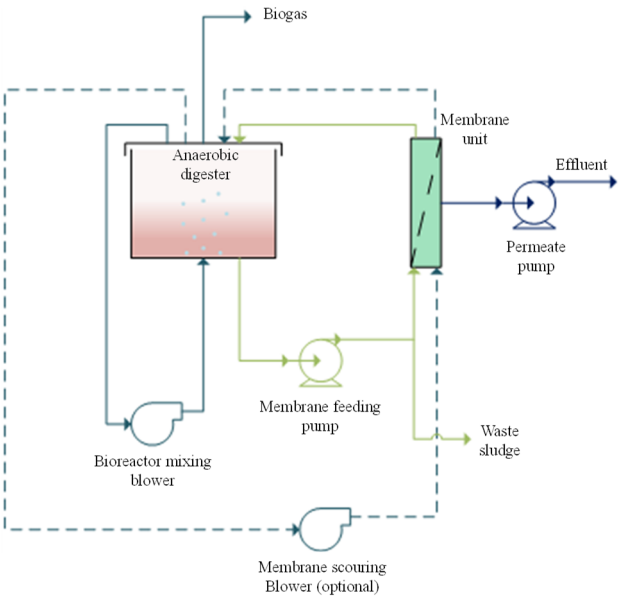
1.4. Modelling of wastewater treatments

1.4.1. Biological models for wastewater treatments

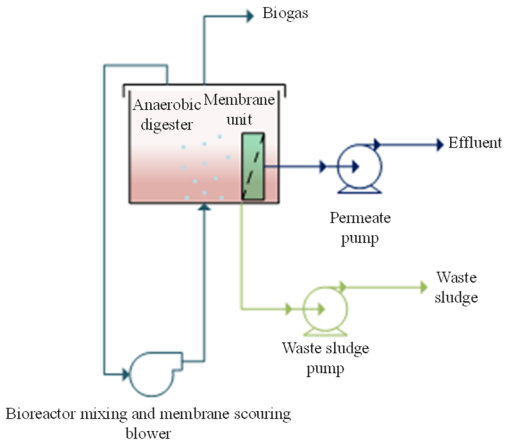
Biological wastewater treatment models are a powerful tool for research, design and optimization of facilities and for personnel training (Petersen et al., 2002). These models aim at representing the biological, physical and chemical processes and their interaction involved in the wastewater treatments, through kinetics and stochiometric expressions.

The first model developed for treatments based on activated sludge was the Activated Sludge Model nº 1 (ASM1) (Henze et al., 1987). This model allowed reproducing and predicting the removal of organic matter and nitrogen. On the contrary, it did not include the biological removal of phosphorus. To cover this gap, further improvements of ASM1 were developed and published. The Activated Sludge Model nº 2 (ASM2) (Gujer et al., 1995) overcame this barrier and it was later expanded in the Activated Sludge Model nº 2d (ASM2d) (Henze et al., 1999) to include the ability to denitrify that polyphosphate-accumulating organisms (PAO) have.

Based on ASMs, a new set of models was developed trying to include all the advantages of those published so far, but also incorporating calculation improvement, additional description of some processes and adding new ones. In this sense, the Biological Nutrient Removal Model nº 1 (BNRM1) (Seco et al., 2004) presents the following characteristics: 1) it uses a unique mathematical model for simulating and modelling the processes and elements in both, the water line and sludge line; 2) organic matter and nutrient removal processes are based on ASM2d with some modifications; 3) anaerobic biological processes are based on the simplified proposal of Siegrist et al. (1993); 4) sedimentation processes are represented by a combination of the proposals of Takács et al. (1991) and Ribes (2004); and 5) the model includes the biochemical processes that occur in primary and secondary settlers and thickeners.



a)



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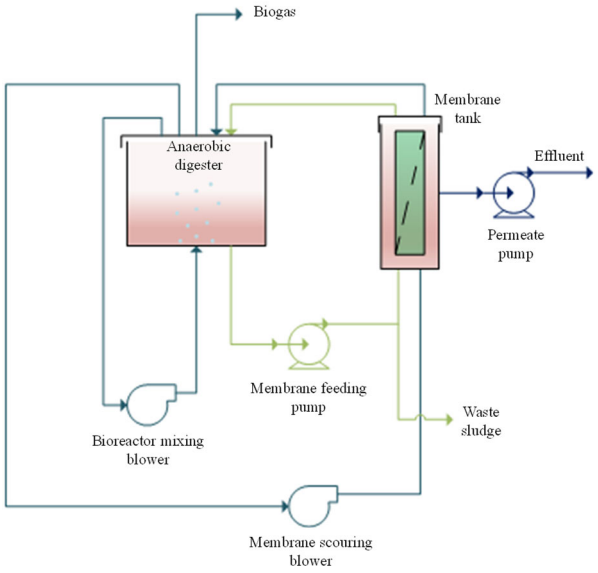


c)

Figure 1.11. AnMBR configurations based on membranes position: a) crossflow; b) submerged; and 3) external submerged.

In 2013 the Biological Nutrient Removal Model nº 2 was published (BNRM2) (Barat et al., 2013), which improved the previous BNRM1 by including all the components and processes required to simulate denitrification via nitrite and precipitation in anaerobic digesters. Finally, Durán et al. (2020) developed the Biological Nutrient Removal Model nº 2d including the competition between methanogenic and sulfate-reducing bacteria (SRB) for the biodegradable COD in the anaerobic processes.

BNRMs set are included in the software Design and Simulation of Activated Sludge Systems-DESASS (Ferrer et al., 2008), which allows the integral assessment of complete treatment layouts including both the water line and the sludge line.

1.4.2. Energy modelling and assessment

Energy modelling and assessment represents a key aspect to address wastewater treatment sustainability. Energy cost affect economic and social feasibility, while GHG emissions associated to fossil fuel consumption influence the environmental and social impact. Detailed breakdown of energy inputs and outputs is crucial for selecting appropriate technologies and equipment focused on diminished undesirable consequences of wastewater treatments.

To this aim, different energy models and approaches have been proposed and their application to treatment schemes and facilities have been published. The research developed in this PhD thesis will be based on the model proposed by Pretel et al. (2016b). This energy model is coupled to the extended BNRM2S plant-wide mathematical model published by Durán et al. (2020). This model can be solved for both steady and dynamic state. More details can be found in chapter 2 (Materials and Methods).

Molinos-Senante et al. (2018) modelled the energy consumption of 305 WWTP divided into 5 categories based on secondary treatment technology: conventional activated sludge; extended aeration; aerate lagoon; trickling filter and biodisk (see Table 1.2). The authors also reported significant economy of scale effect for all evaluated technologies.

Table 1.2. Energy consumption (kWh·m-3) of different WWT technologies (adapted from Molinos-Senante et al. (2018b)).

Conventional activated sludge	Extended aeration	Aerated lagoon	Trickling filter	Biodisk
0.631±0.391	0.562±0.440	0.801±0.663	0.550±0.410	0.399±0.312

Nakkasunchi et al. (2021) reviewed 200 articles focused on water-energy nexus in WWTP. The authors identified assessment tools to explore decarbonization strategies within UWW management and distributed them based on 3 approaches: diminish energy consumption; increase energy production; and integration of renewable energy sources. The article also provides an estimation of the distribution of energy consumption along the treatment phases (see Table 1.3), confirming secondary treatment as the main energy consumer. Benchmarking optimization tools such as Energy Online System (Torregrossa et al., 2016), ENERWATER (Longo et al., 2019) or the methodology proposed by Sabia et al. (2020) are discussed. The need of an integrated system able to link opportunities for energy savings and potential renewable energy production to advance toward a self-sufficient WWTP is one of the main conclusions.

Table 1.3. Allocation of energy demand among a conventional wastewater treatment facility (adapted from Nakkasunchi et al. (2021)).

		Pretreatment	Primary Treatment	Secondary Treatment	Tertiary Treatment	Sludge Line
Energy consumption	%	2-8	2-3	64-74	8-13	6-34

Zaborowska et al. (2021) proposed an extended energy model which would include, in addition to the traditional assessment of facility equipment and energy recovery: i) the associated plants to the overall energy balance and GHG footprint and ii) sub-models of heat and power supply and demand. With this approach, energy balance shifts from a traditional process-oriented perspective to a whole plant-oriented point of view. This change allows improving the energy assessment and clarify the energy profile of the facilities.

Focused on MBR and AnMBR technologies, Pretel et al. (2013) simulated the performance of a AnMBR treating UWW under 3 scenarios: ambient temperatures of 20 and 33 °C and controlled temperature of 33 °C with ambient temperature of 20 °C. Mechanical, heat and recovered energy were considered. Mechanical energy was calculated based on equations proposed by Judd & Judd, (2006). Heat energy comprises the energy required to heat the influent, heat transfer through

walls, heat associated to biochemical reactions and to gas compression and decompression. Finally, energy recovery by means of CHP was also included. All scenarios showed similar mechanical energy consumption (0.2 $kWh \cdot m^{-3}$), while net energy requirements for scenarios operated without temperature control were 0.20 (20 °C) and 0.18 (33 °C) $kWh \cdot m^{-3}$. On the other hand, temperature-controlled scenario obtained an unfeasible result (33.71 $kWh \cdot m^{-3}$), due to the energy demands of maintaining the temperature at 33 ºC in an environment at 20 ºC. The results also revealed the high energy consumption associated with the blowers that drive the biogas for the anaerobic reactors mixing and membrane scouring.

Pretel et al. (2016a) evaluated the energy consumption of an AnMBR coupled with different aerobic based pos-treatment and compared it with aerobic-based technologies for moderate and high-loaded UWWT. A summary of the results is shown in Table 1.4.

Table 1.4. Energy consumption ($kWh \cdot m^{-3}$) of different technologies and their combination for wastewater treatment (adapted from Pretel et al. (2016a)).

CAS	AeMBR	AnMBR+AeMBR	AnMBR+AeMBR:CH_4 $_{DN}$[a]	AnMBR+CAS	AnMBR+CAS:CH_4 $_{DN}$[a]
0.21-0.23	0.50-0.56	0.20	0.20	0.04	0.03

[a] CH_4 $_{DN}$: carbon source of dissolved methane used for denitrification process.

More recently, Cashman et al. (2018) modeled the energy profile of decentralized aerobic MBR and AnMBR for different population densities, treatment flows, climate conditions and considering water reused. When the latter is included in the system boundaries, MBR show net energy production from 1 million of gallons per day (MGD) while mesophilic AnMBRs act as energy producer from 5 MGD. Furthermore, AnMBR performed as net energy producer for al treatment flows at psychrophilic conditions.

Finally, AD and energy recovery from food waste represents an opportunity to improve energy balance in the water management. The water-food-energy nexus is therefore boosting multidisciplinary approaches to take fully advantage of the benefits of integrating food, waste, and facilities management. As way of example, Valenti & Toscano, (2021) applied a GIS-based model to link existing WWTPs and biogas networks in order to optimize the logistic-supplying phase and improve biogas production in the agricultural AD plants. Such approaches would require appropriate tools capable of evaluating energy balances beyond wastewater systems.

1.4.3. Environmental assessment

1.4.3.1. Introduction to life cycle assessment

Environmental concern about the effects of goods production, energy and materials consumption and waste emissions led to the development of tools and approaches to assess impacts to the environment. Among them, life cycle assessment (LCA) has gained prominence and is being applied not only for assessing products and services, but also for evaluating companies functioning or during policy making process. Different definitions of LCA can be found. For instance, the European Environmental Agency (EEA) considers LCA as a "process of evaluating the effects that a product has on the environment over the entire period of its life thereby increasing resource-use efficiency and decreasing liabilities" (EEA Glossary), while ISO 14060:2006 defined it as a "compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle" (ISO/TC 207/SC 5 Life cycle assessment, 2006).

LCA has series of characteristics that make this technique especially suitable for environmental analysis:

- **LCA applies a life cycle perspective:** LCA considers all the processes involved until a certain product or service performs its function and is discarded, which allows for the comprehensive quantification of the impacts at each stage.
- **LCA addresses a broad catalog of environmental issues:** LCA focuses on a diverse group of environmental impacts with the aim of ensuring a proper evaluation of the environmental performance of a product or service.
- **LCA output is quantitative:** results of LCA are obtained by assessing waste emissions and resource consumption and transforming them into environmental impacts through cause/effect models.
- **LCA insight and modelling is based on science:** environmental impacts in LCA are calculated from science knowledge and empirical relationships, which allow obtaining robust results.

With the aim of promoting the use and research related to LCA, the Life Cycle Initiative was launched in 2002 by the United Nations Environment Program (UNEP) and the Society of Environmental Toxicology and Chemistry (SETAC) as a common meeting space for public and private partnership. This initiative aims at widening the application of LCA within governments,

businesses, science, technology, and society to address global challenges and overcome environmental issues faster and more efficiently. To this end, the initiative acts as a platform to facilitate access to environmental, social, and economic life cycle knowledge (LCA data, methods, indicators, etc.) backed by an expert community. An additional milestone to consolidate LCA studies development was the launch of the first academic journal fully dedicated to LCA, the International Journal of Life Cycle Assessment, in 1996.

Finally, the consolidation of the use of the LCA can be contrasted through the evolution of the scientific articles published on this subject. In this sense, He & Yu (2020) carried out a bibliometric analysis for the period 1999-2018 by means of Web of Science database. Results showed a significant increment of published studies, specially from 2009 (see Figure 1.12a). Moreover, Figure 1.12b depicted USA and China as the most productive countries in this field, being Italy and Spain in the third and fourth position. However, if UE member states are considered as a common territory it can be considered the region with most efforts dedicated to this field.

1.4.3.2. Phases of life cycle assessment

Since the first life-cycle-oriented studies emerged in the 1960s, the application of this tool has been evolving, with researcher, practitioners and organizations building consensus until it reached a formal standardization for LCA in the 1990s and 2000, when the International Organization of Standardization (ISO) issued the standard series ISO 14040 (LCA principles and framework), ISO 14041 (Goal and scope definition), ISO 14042 (Life cycle impact assessment) and ISO 14043 (Life cycle interpretation). A revision took place in 2006 which combined the latter three in the ISO 14044.

According to ISO 14040:2006, LCA comprises 4 stages (see Figure 1.13):

- Goal and scope definition (ISO 14040:2006).
- Inventory analysis (ISO 14044:2006)
- Impact assessment (ISO 14044:2006)
- Interpretation (ISO 14044:2006)

As Figure 1.13 shows, the development of a LCA is a non-linear process. During the progress in the development of the study, there are frequent feedback loops with the aim of fine-tuning the previous steps based on the information that is being obtained.

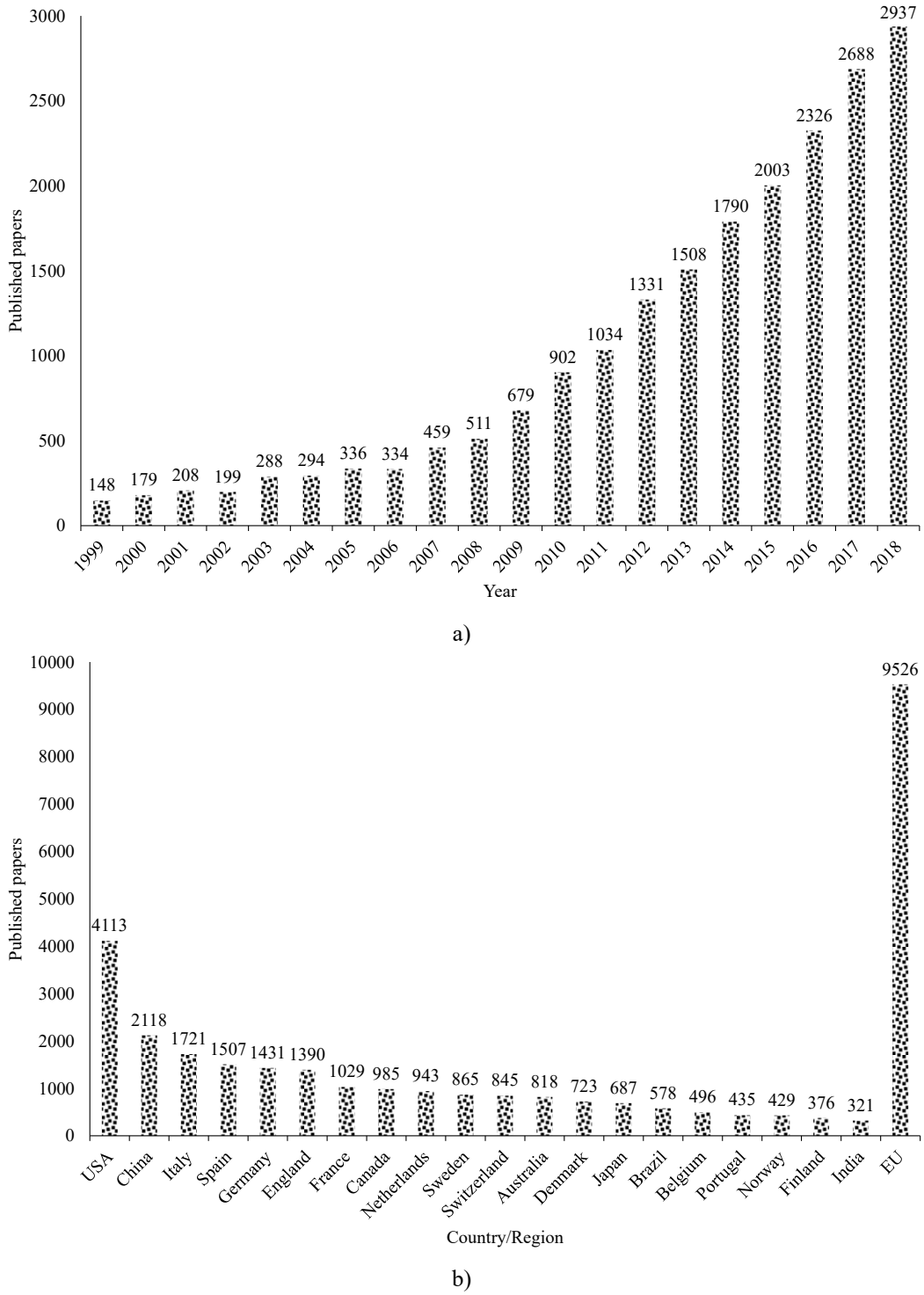


Figure 1.12. Evolution in the period 2018-2020 of a) published papers focused on LCA and b) published papers focused on LCA per country/region (adapted from (He & Yu (2020)).

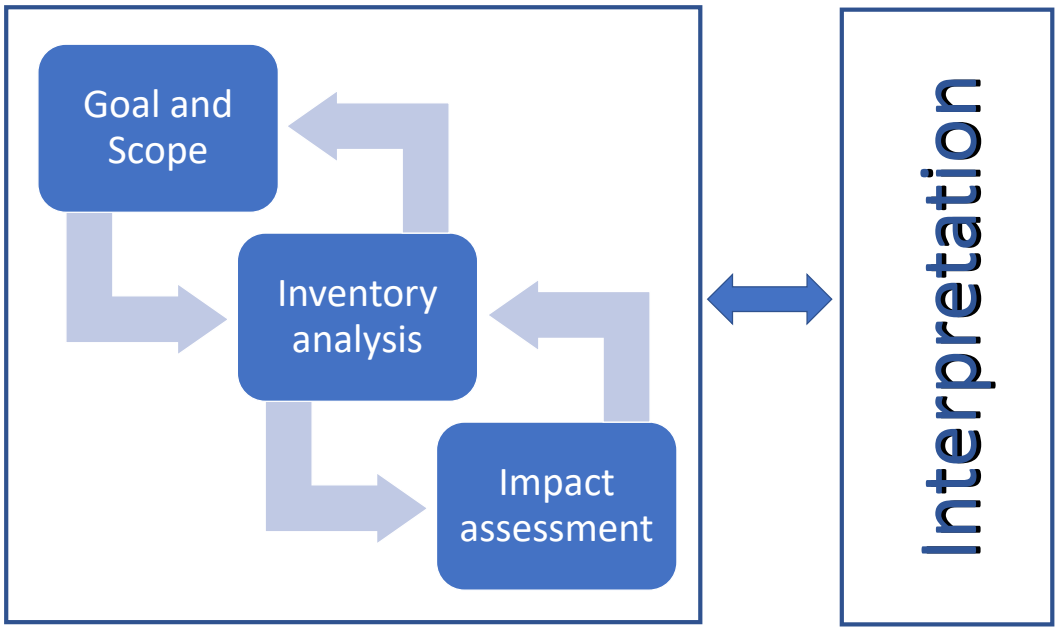


Figure 1.13. LCA phases (adapted from ISO 14040:2006).

1.4.3.2.1. Goal and scope definition

In this phase, the ultimate goal of the study must be clearly defined. The importance of the definition of the goal lies in the fact that it will influence the later phases, since the development of the LCA must be consistent with the definition of it.

ISO standard requirements for an appropriate goal definition comprises:

- The intended application.
- The reasons for conducting the study.
- The intended audience.
- If it is planned to use the results in comparative studies.

On the other hand, the scope definition must address what systems are included in the study and how the assessment will be carried out. ISO stated the following items as part of the scope definition:

- The product system to be studied.
- Product system functions or, in the case of comparative studies, systems: A system can have different functions and the one that is the object of the study must be selected in accordance with the definition of the goal.

- The functional unit (FU): the selection of the functional unit must be made in such a way that it adequately describes the qualitative and quantitative aspects of the function that the system performs (Rebitzer et al., 2004).
- The system boundaries: boundaries delimit the processes that must be taken into account during the evaluation, including the temporal and geographical scope of the study (Tillman et al., 1994).
- Distribution procedures: specification of the impact loads distribution for multiproduct systems (i.e., subdivision of unit process, system expansion or allocation).
- The selected impact categories and impact assessment methodology, and the subsequent interpretation to be used.
- Data requirements.
- Assumptions.
- Limitations.
- Initial data quality requirements.
- The type of critical review, if any.
- The type and format of the report required for the study.

1.4.3.2.2. Inventory analysis phase

Life cycle inventory analysis (LCIA) phase represents the core task of an LCA. During this step, all significant inputs and outputs involved in a process are identified and quantified in a systematic way.

This phase use to be the most time-consuming phase and different type of data are required to achieve a satisfactory description of the product system. Data that describe a particular product system and production system, being specific of it, are known as *foreground data* (e.g., kg of steel to produce one scissor). On the other hand, *background data* accounts for processes that contribute to production system but are not exclusive of it (e.g., energy production or material transport) (see Figure 1.14).

Availability of data is therefore crucial for elaborating a reasonable LCI. According to ISO 14040:2006, it should comprise:

- Energy, feedstock, auxiliary and other physical inputs.
- Products, by-products and residues.

- Emissions to air, water and soil.
- Any other relevant environmental issue.

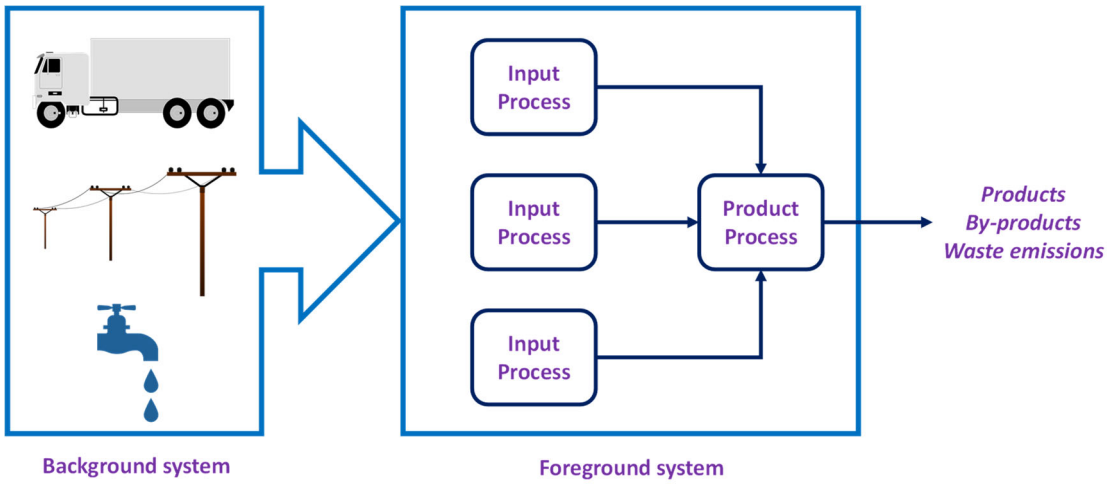


Figure 1.14. Background system vs foreground system in LCIA.

Data can be obtained by different ways and sources: direct measure, modeling, bibliography, data bases or experts. Regarding databases, Table 1.5 show an example of data bases available:

1.4.3.2.3. Impact assessment phase

Once the LCI is compiled, the next phase of a LCA is the life cycle impact assessment (LCIA) phase. The objective of an LCIA is to transform the elementary flows collected in the previous phase (LCI) into environmental impact scores. In this sense, LCIA models the pathway from the emission to its effect and impact by obtaining and applying the so-called *characterization factor* (CF).

During this phase, the elementary flows of LCI, whose interpretation in relation to their consequences on the environment are abstract and difficult to relate, are translated into different impact categories through environmental mechanisms modelling that facilitate their interpretation (Figure 1.15).

Table 1.5. Databases available for inventory analysis.

Database	Characteristics	Link
European Life Cycle Database - ELCD	Database of the JRC of the European Commission, contains more than 300 datasets on energy, material production, disposal and transport.	https://eplca.jrc.ec.europa.eu/LCDN/index.xhtml
Ecoinvent	Swiss database that contains approximately 18000 processes (version 3) under different themes like transport, energy, material production, agriculture, etc. All processes are available as unit- and system-processes and all processes are documented in detail. Updated regularly.	https://ecoinvent.org/the-ecoinvent-database/
Agri-footprint	A comprehensive LCI database of feed, food and biomass, containing around 3500 products and processes.	https://blonksustainability.nl/tag/Databases
GaBi databases	Separate databases mainly based on primary data collection. Cover sectors from agriculture to electronics and automotive industries, textiles and retail, through to services. Contains more than 10,000 Life Cycle Inventory profiles.	https://gabi.sphera.com/databases/gabi-databases/
LCA Food	Data base which provides input/output data on processes in the food sector (and environmental data on food products focused on Denmark	http://www.lcafood.dk/
U.S. Life Cycle Inventory-USLCI	U.S.A data base accounting of the energy and material flows into and out of the environment that are associated with producing a material, component, or assembly.	https://www.nrel.gov/lci/

According to ISO 14040:2006, LCIA contains three obligatory steps:

- **Selection of impact categories:** among all the available impact categories, the relevant ones according to the goal and scope of the study must be selected.
- **Classification:** in this step the elementary flows of the LCI are linked to the impact categories to which they contribute.

- **Characterization:** the classified elementary flows (EF) of LCI are transformed into the impact scores (IS) used to quantify their associated environmental impacts. This transformation is carried out by means of CF, which represents the contribution per quantity of an elementary flow to a specific environmental impact (see Equation 1.1). CF are obtained from different models which represents the transport, fate, evolution, and impact pathways of the evaluated substances.

$$IS = CF \cdot EF \qquad \text{Equation 1.1}$$

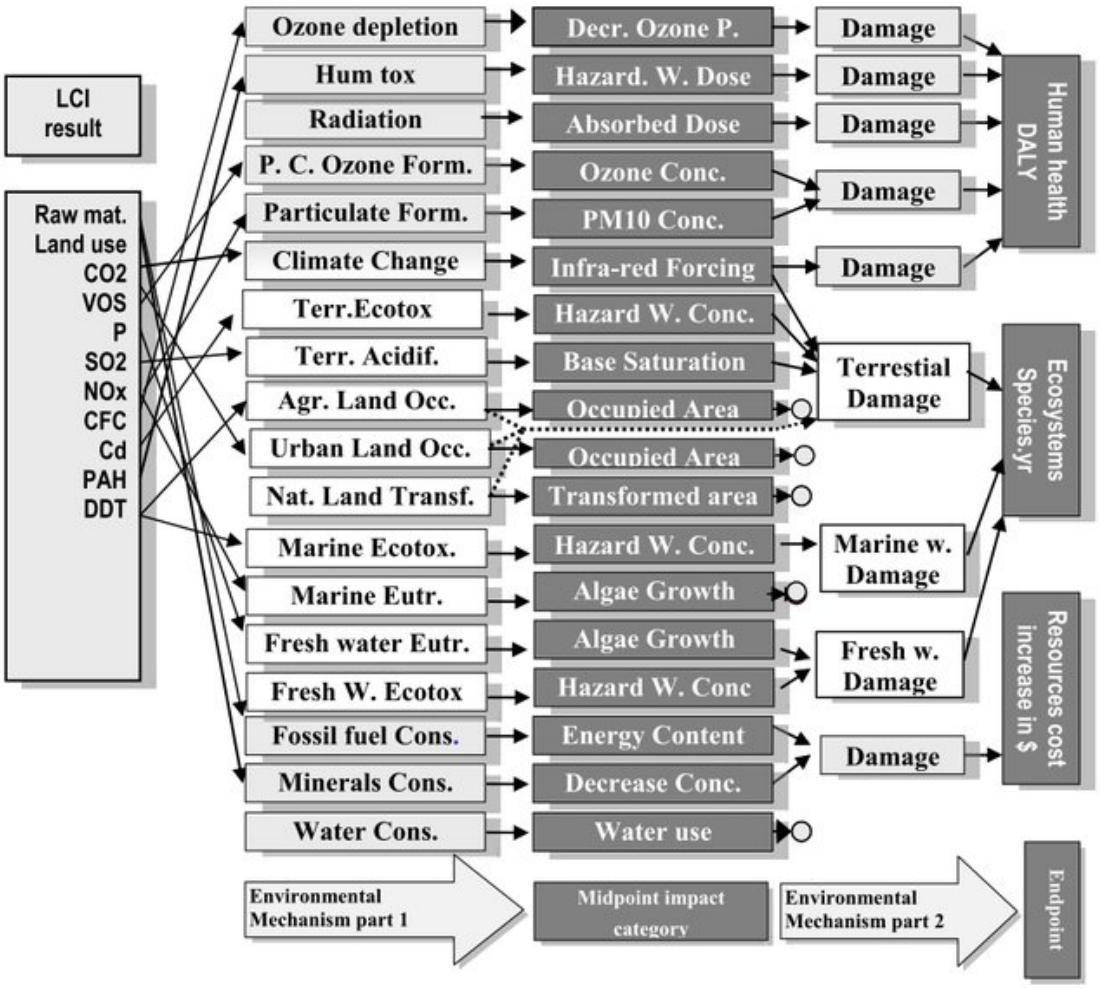


Figure 1.15. Example of scheme of LCIA phase for a given method (Goedkoop et al., 2009).

There are three additional steps, which consideration is optional:

- **Normalization:** since impact categories are expressed in different units, comparison between categories is not possible. Hence normalization consist in transforming the different impacts to a common scale.

- **Weighting:** this step provides importance among categories by assigning weights. This action is based on practitioner criteria, so there is no scientific basis to support the decisions.
- **Grouping:** This step is based on bringing together several impact categories in one or more groups.

Table 1.6. Examples of life cycle impact assessment (LCIA) methods.

Name	Issue	Link/Reference
CM-L IA	Multi	https://www.universiteitleiden.nl/en/research/research-output/science/cml-ia-characterisation-factors
Ecological Scarcity	Multi	Frischknecht et al. (2009)
ILCD 2011	Multi	https://eplca.jrc.ec.europa.eu/uploads/LCIA-characterization-factors-of-the-ILCD.pdf
Impact 2002+	Multi	https://quantis.com/pdf/IMPACT2002_UserGuide_for_vQ2.21.pdf
ReCiPe2016	Multi	Huijbregts et al. (2017)
TRACI	Multi	Bare, (2002)
USETox	Multi	https://usetox.org/
Cumulative energy demand	Single	Hischier et al. (2010)
Ecosystem damage potential	Single	Hischier et al. (2010)
IPCC 2013GWP	Single	https://www.ipcc.ch/report/ar6/wg1/
AWARE	Single	Boulay et al. (2018)

Environmental impact categories and their indicators can be classified into midpoint and endpoint (see Figure 1.15). Midpoint categories focus on single environmental problems (e.g., climate change or freshwater eutrophication) and their indicators can be understood as connections in the environmental mechanism (e.g., infra-red forcing or algae growth, respectively). On the other hand, endpoint indicators illustrate damage levels for an area of protection influenced (e.g., human health) by several midpoint categories.

There are several LCIA methods available midpoint and/or endpoint oriented. LCIA also can addresses multiple environmental issues (multi) or be focused on one (single) (e.g., AWARE assesses water footprint, carbon footprint). A selection is shown in Table 1.6:

Finally, there are also specific software where LCIA are implemented and access to inventory databases are included. A selection of the most common can be found in Table 1.7:

1.4.3.2.4. Interpretation phase

The last phase in an LCA consists in the interpretation of the results obtained during the previous phases, with the aim of drawing useful conclusions, recommendations or establishing necessary actions in accordance with what is defined in the goal and scope step. This phase may result in the need to go back to previous phases if weaknesses or inconsistencies are detected.

Table 1.7. Specific software for LCA.

Software	Characteristics	Link/Reference
SimaPro	Several data bases and methodologies are included. It is product-designer, decision-maker and researcher oriented.	https://simapro.com/
GaBi	It allows expanding LCA with LCC and social assessment. It also facilitates evaluating supply chains.	https://gabi.sphera.com/spain/index/
OpenLCA	Open source and free software for sustainability and LCA. Life Cycle Costing and social assessment tools are also included.	https://www.openlca.org/
Umberto	Software for LCA and resource efficiency evaluation. It includes different methodologies and data bases. It allows graphical modelling to improve transparency and understanding of the processes.	https://www.ifu.com/umberto/
Brightway	Open-source framework for LCA calculations developed in Python with a modular structure. It allows spatial and temporal evaluations and global sensitivity analysis.	https://github.com/brightway-lca

1.4.4. Economic assessment

1.4.4.1. Life cycle costing background

Life cycle costing (LCC) was initially developed during 1960s by the U.S. Department of Defense as an economic tool oriented to evaluate the acquisition of military equipment (Heralova, 2017). Since then, LCC has evolved and different LCCs can be found in literature, depending on their goal and approach: conventional (cLCC), environmental (eLCC) and societal (sLCC) (Ilyas et al., 2021).

cLCC is focused on evaluating capital investment from the perspective of the final user or the manufacturer of a good, product or service. Advantage of cLCC is that standards are available for several industries or economic sectors (e.g., ISO 15663:2021 Petroleum, petrochemical and natural gas industries — Life cycle costing or ISO 15686:2017 Buildings and constructed assets — Service life planning — Part 5: Life-cycle costing), which makes their outcomes more robust. In the case of eLCC, it was developed to support and complement LCA. It does not currently have its own standard, but its life cycle approach, scope and use of the functional unit allows it to be aligned with ISO 14040 and ISO 14044 for LCA. Finally, sLCC aims to include all the environmental and social costs by monetarizing them. In this sense, sLCC is conceived as a decision support tool for authorities and policy maker, but there is no standardization at this time. Given the topic discussed in this thesis, everything that follows refers to the eLCC, although it will be referred to as LCC, unless otherwise specified.

Ilyas et al. (2021) analyzed the published papers focused on LCCs of wastewater treatment since 2020. In the last decades, number of publications on the different LCCs is increasing but it is far from LCA studies (see Figure 1.16a). U.S.A and India are the most productive countries, but the EU shows also high interest in LCC development (Figure 1.16b). The authors concluded that environmental and social LCC publications are gaining momentum versus conventional LCC. Cost breakdown still requires to be improved and oversimplification should be avoided. In this sense, the authors pointed out the need of integrating modern cost estimation and process cost estimation for achieving accurate results.

1.4.4.2. Life cycle costing methodology

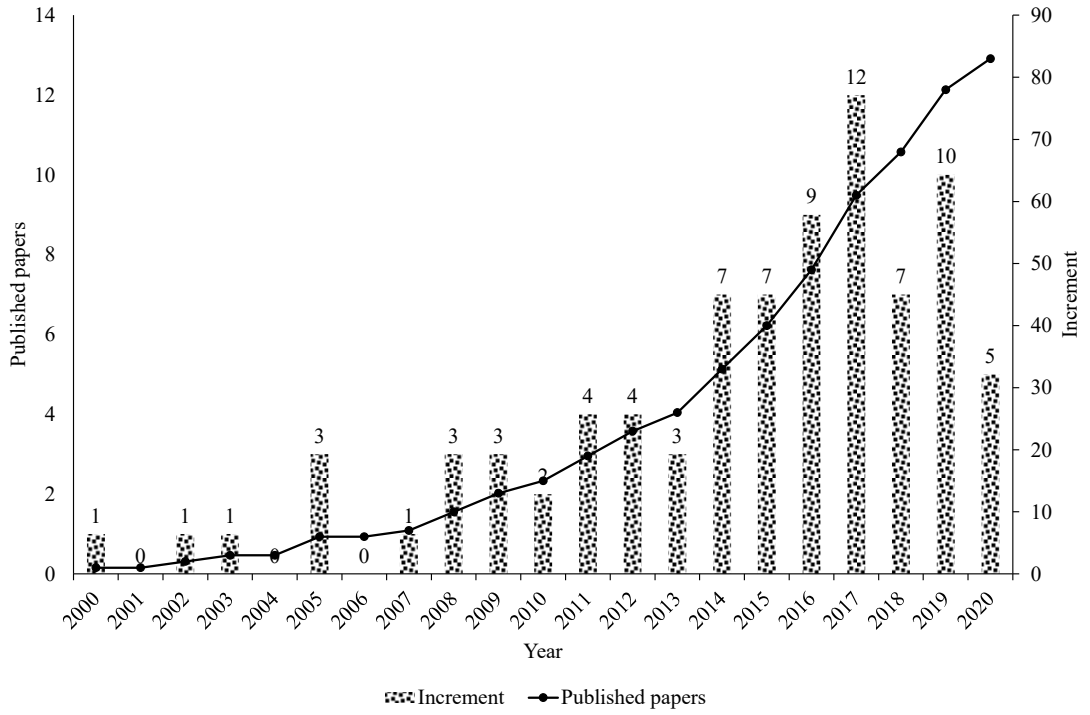
Environmental LCC follows the recommendations and statements stablished in ISO 14040:2006 and ISO 14044:2006 (see section 1.4.3.2). However, some remarks need to be made, since there are some characteristics that differ from LCA:

- **System boundaries:** inclusion of external costs which are expected to be internalized during the life cycle is recommended, if possible (e.g., discharge fee for treated water, costs of sludge management or CO_2 emissions, etc.).
- **Monetarizing:** since eLCC have the vocation to go beyond conventional LCC by internalizing environmental costs, techniques have been described to try to assign a monetary value to these impacts, so that they can be included in the analysis and compared with conventional costs. Among these methods Ecovalue, Stepwise, LIME, Ecotax, MMG or EPS can be mentioned (Arendt et al., 2020)
- **Allocation:** complex systems usually include costs not directly linked to a product or service, but to the general functioning of the company (e.g., overheads or indirect cost). In these cases, a coherent and robust allocation method is required. For example, distributing costs based on the percentage of revenue that each product or service represents for the company is a possible simple and transparent methodology.
- **Inventory:** consistency of currency and year baseline shall be ensured.
- **Discounting:** the discount allows modulating the weight of processes costs to reflect the interest shown by companies in delaying payments and focusing their current resources on investment and/or expansion of their activities. The weights (w) during a period of time (t) are calculated with Equation 2:

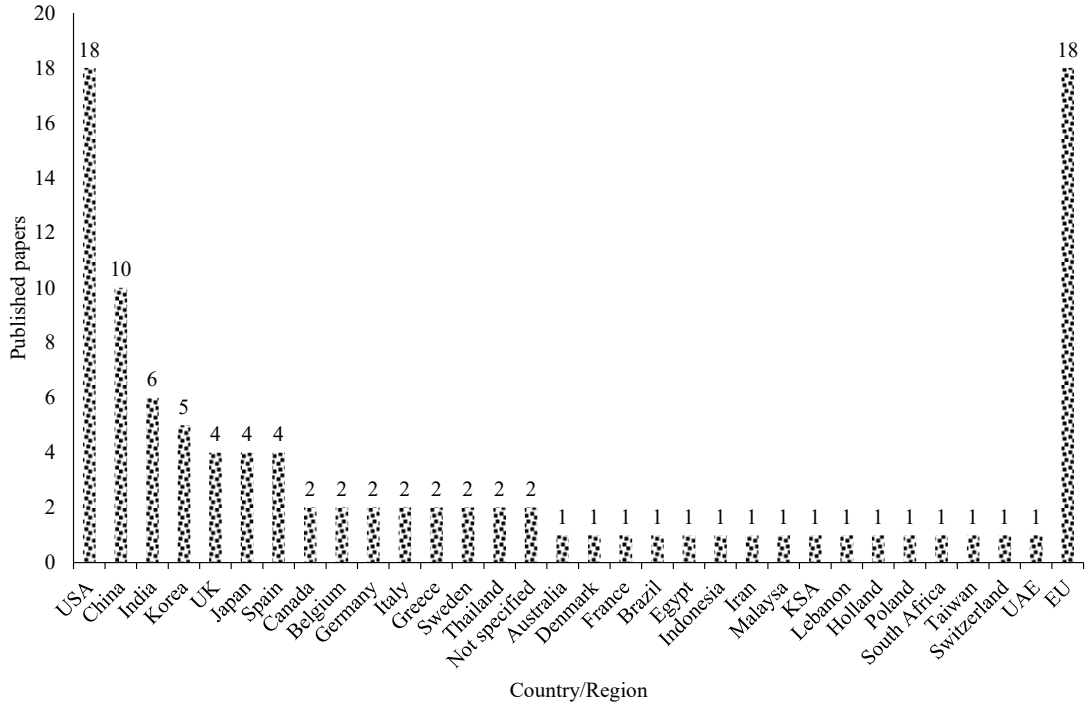
$$w(t) = \frac{1}{(1+r)^t} \qquad \text{Equation 1.2}$$

where *r* is the discount rate.

As in the case of LCA, the stage that requires the most effort and time is the compilation of the inventory. Some data are publicly available (e.g., energy tariffs, taxes) and there are also databases for different sectors (free or under subscription) (Table 1.8). Finally, access to equipment costs might be difficult in some cases since manufacturers usually do not facilitate this information. In these cases, cost estimation (through CEPCI index or specific software, e.g., Aspen Plus) can be applied as alternative to real data.



a)



b)

Figure 1.16. Evolution in the period 2000-2020 of a) published papers focused on LCC of wastewater treatments and b) published papers focused on LCC of wastewater treatments per country/region (adapted from Ilyas et al., 2021)).

Table 1.8. Examples of databases available for life cycle costing inventory (adapted from Hauschild et al. (2018)).

Sector	Database	link
Energy	International Energy agency	https://www.iea.org/data-and-statistics/data-products/?filter=prices
Plastics	The Plastic Exchange	https://www.theplasticsexchange.com/
Chemicals	Independent Commodities Intelligence Services-ICIS	https://www.icis.com/explore/commodities/chemicals/
Metals	Bloomberg	https://www.bloomberg.com/markets/commodities/futures/metals
Commodities	United Nation	https://comtrade.un.org/
Economic indicators (World)	World Bank	https://data.worldbank.org/
Economic indicators (EU)	Eurostat	https://ec.europa.eu/eurostat/web/main/data/database
Salaries	International Labor Organization	https://ilostat.ilo.org/topics/labour-costs/
Construction	Valencian Institute of Edification	https://www.five.es/precios-de-construccion/

Once the inventory is completed, CAPEX and OPEX are combined, and an economic indicator is obtained that allows evaluating the available options based on a certain criterion:

- Net present value (NPV): NPV estimates the present value of all cash flows over a period of time and is calculated by means of Equation 1.3. Positive NPV indicates expected profit.

$$NPV = \sum \frac{P(t)}{(1+r)^t} = \frac{P(0)}{(1+r)^0} + \frac{P(1)}{(1+r)^1} + \frac{P(2)}{(1+r)^2} + \cdots + \frac{P(t_{max})}{(1+r)^{t_{max}}} \qquad \text{Equation 1.3}$$

where P(t) represents the cash flow at time t and r the discount rate.

- TAC: TAC represents de annualized value of the net present cost. It can be calculated with Equation 1.4, by adding the annualized CAPEX to the OPEX:

$$TAC = \frac{r \cdot (1+r)^t}{(1+r)^t - 1} \cdot CAPEX + OPEX$$ Equation 1.4

where r is the discount rate and t the considered time period.

1.4.5. Sensitivity and uncertainty analysis

Modeling are representation of system or processes, most of them with a high inherent complexity. To be useful, models simplify reality trying to combine accuracy and precision in the response with speed of calculation. Also, data quality and coherent assumptions are a crucial element to obtain accurate model outcomes. In order to establish whether the predictions obtained from the application of a certain model can be considered reliable, two tools called sensitivity analysis (SA) and uncertainty analysis (UA) are commonly used.

SA aims to determine, along all parameters involved in a model, which inputs show a higher effect on the model output (Sin et al., 2011). Therefore, input parameters or variables with low or even no effect on the model output can be discarded or set to a default value, thus simplifying the model calibration and validation steps, or even allowing to simplify the model structure. SA analysis can be classified in two main groups:

Local sensitivity analysis (LSA) evaluates the local effect of an input to a model output. With this approach, sensitivities are calculated for a particular set of parameters and most of the existing strategies assumed a linear relation between input and output variables.

Global sensitivity analysis (GSA) assesses the sensitivity of the inputs on the model output in many different points among the parameter space. This strategy allows the evaluation with non-linear relations between inputs and outputs and/or significant uncertainty, but computational demand is also important.

In the UWW fields, two LSA methods have traditionally been used:

- **Fisher information matrix (FIM) method:** this technique is based on the calculation of a series of scalar functions of the FIM matrix, to determine the number of identifiable parameters of the set of parameters that define the model. Then, the covariance matrix is

determined from the FIM, from which information about the correlation of the parameters can be obtained, determining the correlation matrix. This correlation matrix denotes the relationship between parameters and gives insight into the compensating effects of changes in parameter values on the model output.

- **Collinearity index method:** this technique is based on the determination of three sensitivity measures from the calculation of the sensitivity matrix, among which is the collinearity index. This index represents the linear dependence between the parameters of the evaluated set. If the value of this index is very close to 1, there is no dependence between the parameters. Conversely, high values of the index indicate dependence between input and output.

Different GSA tools have also been developed, among which the following can mentioned:

- **Morris screening method:** this method applies modifications of input variable on a one-factor-at-a-time basis and evaluates its effect on the model outputs. This model allows categorizing the effects of inputs on outputs as negligible, linear and additive or non-linear and non-additive.
- **SOBOL method:** this strategy is a variance-based sensitivity analysis that decomposes the model output variance into fractions and, subsequently, it attributes these fractions to the input parameters. Therefore, SOBOL sensitivity analysis aims at determining the contribution of each input parameter and their interactions to the overall model output variance.
- **Fourier amplitude sensitivity analysis (FAST) and extended FAST (eFAST):** FAST methos is also a variance-based method that defines the sensitivity value by means of conditional variances which denote the effect of the inputs on the model output. eFAST method improves the computational efficiency.
- **Standardized Regression Coefficient (SRC) method:** this method applies a multivariate regression between model inputs and outputs and quantify the influence of the considered input on the model output by means of a standardized regression. Hence, the regression coefficient "b_i" denotes the effect of a change in the input on the output of the model.

On the other hand, UA is based on an arbitrary modification of inputs values (built by the user by a selected sampling method) and an assessment of the obtained outputs. UA allows evaluating and quantifying risks and provide evidence on robustness and credibility of the conclusions. In this sense, Monte Carlo is a common method for uncertainty evaluation that can be fed by

different sampling strategies, such as the Latyn Hypercube Sampling (LHS) strategy (Sin et al., 2009). LHS generates a near-random sample of inputs from a multidimensional distribution.

1.5. Legal framework

This section aims to summarize the regulative instruments, guidelines and plans related to resource recovery from UWW. Table 1.9 includes a selection of documents and regulation adapted from Shoushtarian & Negahban-Azar, (2020) and other official sources (e.g., European Commission, standardization agencies, etc.).

Table 1.9. Summary of regulations, guidelines and plans for resource recovery from wastewater (adapted from Shoushtarian & Negahban-Azar (2020) and others).

Resource	Level	Institution	Regulation/guideline/plan
Water	International	WHO	Safe use of wastewater, excreta, and grey water
		FAO	Wastewater quality guidelines for agricultural use
		ISO	ISO 16075:2015 "Guidelines for treated wastewater use for irrigation projects"
		EPA	Guidelines for water reuse
		Australia	National Guidelines for Water Recycling: Managing Health and Environmental Risks
	European Union	European Commission	Communication from the Commission to the European Parliament and the Council - Addressing the challenge of water scarcity and droughts in the European Union {SEC (2007) 993} {SEC (2007) 996}
		European Commission	Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions-A Blueprint to Safeguard Europe's Water Resources (COM (2012) 673 final)
		Council	Council Directive of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources (91/676/EEC)
		European Parliament	Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy
		Council	Council Directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment
		European Commission	Commission Directive 98/15/EC of 27 February 1998 amending Council Directive 91/271/EEC with respect to certain requirements established in Annex I thereof
		Council	Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC)

Table 1.9. Summary of regulations, guidelines and plans for resource recovery from wastewater (adapted from Shoushtarian & Negahban-Azar (2020) and others). (continue)

Resource	Level	Institution	Regulation/guideline/plan
Water	**European Union**	European Parliament and Council	Regulation (EU) 2020/741 of the European Parliament and of the Council of 25 May 2020 on minimum requirements for water reuse
	Spain	European Commission	Guidelines to support the application of Regulation 2020/741 on minimum requirements for water reuse (2022/C 298/01)
		Spanish Parliament	Real Decreto Legislativo 1/2001, de 20 de julio, por el que se aprueba el texto refundido de la Ley de Aguas
		Spanish Parliament	Real Decreto 1620/2007, de 7 de diciembre, por el que se establece el régimen jurídico de la reutilización de las aguas depuradas
		Spanish Parliament	Real Decreto-ley 4/2023, de 11 de mayo, por el que se adoptan medidas urgentes en materia agraria y de aguas en respuesta a la sequía y al agravamiento de las condiciones del sector primario…

Table 1.9. Summary of regulations, guidelines and plans for resource recovery from wastewater (adapted from Shoushtarian & Negahban-Azar (2020) and others). (continue)

Resource	Level	Institution	Regulation/guideline/plan
Nutrients	**European Union**	European Parliament and Council	Regulation (EU) 2019/1009 of the European Parliament and of the Council of 5 June 2019 laying down rules on the making available on the market of EU fertilizing products
		European Commission	COMMISSION DELEGATED REGULATION (EU) 2021/2086 of 5 July 2021 amending Annexes II and IV to Regulation (EU) 2019/1009 of the European Parliament and of the Council for the purpose of adding precipitated phosphate salts and derivates as a component material category in EU fertilizing products.
	Spain	Spanish parliament	Ley 7/2022, de 8 de abril, de residuos y suelos contaminados para una economía circular.

Table 1.9. Summary of regulations, guidelines and plans for resource recovery from wastewater (adapted from Shoushtarian & Negahban-Azar (2020) and others). (continue)

Resource	Level	Institution	Regulation/guideline/plan
Biogas and biomethane	**European Union**	European Parliament and Council	Directive (EU) 2018/2001 of the European Parliament and of the Council on the promotion of the use of energy from renewable sources
		European Commission	RePowerEU Plan
		European Committee for Standardization	EN 16723-1:2016 - Natural gas and biomethane for use in transport and biomethane for injection in the natural gas network - Part 1: Specifications for biomethane for injection in the natural gas network
	Spain	Spanish Association of Normalization	UNE-EN 16723-1:2017 - Gas natural y biometano para uso en transporte y biometano para inyección en la red de gas natural - Parte 1: Especificaciones para la inyección de biometano en la red de gas natural
		Ministry for the Ecological Transition and the Demographic Challenge	Plan Nacional Integrado de Energía y Clima (PNIEC) 2021-2030
		Spanish parliament	Ley 7/2021, de 20 de mayo, de cambio climático y transición energética
		Spanish parliament	Real Decreto 376/2022, de 17 de mayo, por el que se regulan los criterios de sostenibilidad y de reducción de las emisiones de gases de efecto invernadero de los biocarburantes, biolíquidos y combustibles de biomasa, así como el sistema de garantías de origen de los gases renovables

1.5.1. Water reuse

Agriculture is the main usage for water reuse worldwide. Therefore, this section will be focused on regulation and guidelines related to this purpose.

1.5.1.1. World

In this section, guidelines and recommendations of international organizations and relevant national agencies are briefly introduced:

- ✓ **WHO:** the UN agency for health has issued 3 guidelines in 1973, 1989 and 2006. The objective of WHO was contributing to health protection by promoting a safe application of wastewater to crops and aquaculture. 2006 guideline, entitled "Safe use of wastewater, excreta, and grey water" (WHO & UNEP, 2006) includes improved content related to risk assessment and management and provides support for standards establishment to policy-makers. It includes information about presence and risk assessment of pathogens in wastewater, fields and crops and health risk management is quantified based on disability-adjusted life year (DALY) and person per year (PPY) (Jaramillo and Restrepo, 2017).

- ✓ **Food and agriculture organization (FAO):** The UN agency for food and agriculture has published two guidelines in 1987 and 1999. FAO incorporates three categories for water reuse based on its application and exposure risk, according to WHO recommendations: i) Irrigation of crops likely to be eaten uncooked, sports fields, and public parks; ii) irrigation of cereal crops, industrial crops, fodder crops, pasture, and trees; and iii) localized irrigation of crops in category ii) if exposure of workers and the public does not occur (FAO).

- ✓ **International Organization for Standardization (ISO):** ISO released the standard series ISO 16075:2015 "Guidelines for treated wastewater use for irrigation projects". It comprises ISO 16075-1:2015 Part 1: The basis of a reuse project for irrigation and ISO 16075-2:2015 Part 2: Development of the project. The latter was revised by ISO 16075-2:2020 (ISO, 2020). ISO 16075 defines five types of reclaimed water based on the quality: i) very high-quality treated wastewater; ii) high quality treated wastewater; iii) good quality treated wastewater; iv) medium quality treated wastewater; and v)

extensively treated wastewater. The guidelines include, among others, standards for the design of treated wastewater irrigation projects and specifications on correlation between the quality of the treated wastewater, irrigated crops, and applied barriers.

- ✓ **Environmental Protection Agency (EPA, U.S.A.):** EPA has published guidelines for water reuse in 1980, 1992, 2004 and 2012. In its latter version, EPA recognized the need for addressing new reclaimed water usage, new technologies and regulatory developments (EPA, 2012). It also incorporated non-conventional water sources as part of a comprehensive water management perspective. The guidelines include some recommendations for wastewater treatment suitable for the contemplated water reuse categories and recommends following water quality classification for crop irrigation.

- ✓ **Environment Protection and Heritage Council (Australia):** the Australian Environment Protection and Heritage Council, jointly with the Natural Resource Management Ministerial Council and the Australian Health Ministers' Conference issued in 2006 the "National Guidelines for Water Recycling: Managing Health and Environmental Risks" (NRMMC-EPHC-AHMC, 2006). The guideline aims at identifying health and environmental risks and proposed measures to diminish them. It quantified human health risks related to exposure to pathogens based on DALY and used end-points categories for environmental impact assessment.

1.5.1.2. European Union

- ✓ **Communication from the Commission to the European Parliament and the Council - Addressing the challenge of water scarcity and droughts in the European Union {SEC(2007) 993} {SEC(2007) 996}:** This document gathered and presented policy options at European, national and regional to address water scarcity and droughts. Challenges related to Water Framework Directive implementation, water pricing policies and investment tools, land-use planning, water savings, and appropriate data collection were detected, and potential improvement actions proposed.

- ✓ **Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions-A Blueprint to Safeguard Europe's Water Resources (COM(2012) 673 final):** at the time of its publication. This document recognized lower environmental impacts for water reuse than for water transfer or desalination and identified the absence of common EU environmental/health standards as an obstacle to marketing agriculture products irrigated with reclaimed water within the common market.

- ✓ **Council Directive of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources (91/676/EEC):** the objective of the directive is to prevent water pollution by nitrates discharge. To this aim, the directive encouraged the UE Member States to declare vulnerable zones to nitrates pollution and to stablish action programs including good agriculture practices to reduce pollution risk. Manure spreading must ensure that 170 kg $N \cdot ha^{-1} \cdot year^{-1}$ is not exceeded.

- ✓ **Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy:** the so-called Water Directive Framework (WDF) is one of the most relevant legislative actions adopted by the UE. Its ambitious objectives contemplate all water bodies protection (land surface waters, transitional waters, coastal water and groundwater), sustainable water use promotion, pollution reduction and floods and droughts effect mitigation. It considered the river basin district as the geographical unit where its statements shall be applied and established the principle of recovery of the cost of water services in accordance with the polluter pays principle.

- ✓ **Council Directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment and Commission Directive 98/15/EC of 27 February 1998 amending Council Directive 91/271/EEC with respect to certain requirements established in Annex I thereof**: This directive and its amendment constitute the basic legislative action that governs wastewater conveyance and treatment throughout the EU. It contains measures and rules for water treatment and concentration limits for effluent discharge to protect the environment from the harmful effect of pollutants present in these streams. The directive considers as UWW not only the domestic wastewater, but also the combination of domestic wastewater with industrial water and/or rain run-offs. It also mandates that industrial wastewater entering the collecting system does not pose in risk

the health of staff involved in wastewater collection and treatment facilities and does not impede appropriate water treatment and discharge. Finally, it explicitly introduces a mention to reuse water "whenever appropriate" in its article 12.1.

- ✓ **Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC):** This Directive regulates the agriculture use of sewage sludge to avoid harmful effects on soil, animals, vegetation and persons. It stablishes the need of a sludge treatment before its use and concentrations limits for heavy metals above which sewage sludge spreading is prohibited. It also states that nutrient content in sludge and nutrient requirement of crops must be balanced. Furthermore, it must be ensured that qualities of the soil and surface and ground water are not diminished.

- ✓ **Regulation (EU) 2020/741 of the European Parliament and of the Council of 25 May 2020 on minimum requirements for water reuse:** This regulation represents the last legislative action adopted in relation to the reuse of water and aims to make up for some of the bottlenecks detected in this area and published in previous communications. The regulation only affects agriculture reuse of water previously treated according to Council Directive 91/271/EEC. Its objectives are to: mitigate the effects of climate change; guarantee safe scenarios in terms of environment and health; promote the circular economy; contribute to compliance with the Directive 2000/60/EC; and improve the functioning of the common market. It introduces the figure of the "reclamation facility operator" and the necessity of establishing a risk management plan prior to obtain a permit issued by the competent authority. The regulation also clarifies competences and responsibilities and harmonized health standards and water quality requirements for agriculture reuse among UE.

Water reuse has also been addressed by some European states by issuing guidance or adopting normative. The approval of Regulation (EU) 2020/741 at European level mandate their revision to adapt their content (or even adopt new regulations) to the new legislative framework before June 2023. A summary of the current national regulations can be found in Table 1.10.

1.5.1.3. Spain

- ✓ **Real Decreto Legislativo 1/2001, de 20 de julio, por el que se aprueba el texto refundido de la Ley de Aguas*:*** Within the objectives of this law are: i) the regulation of

the hydraulic public domain and the use of water; and ii) the establishment of basic standards for the protection of inland, coastal and transitional waters. Its chapter III estates the legal regime of water reuse and encourage to establish basic conditions for reuse, to define the permitted uses and the associated qualities. It also establishes that the holder of the concession or authorization must bear the necessary costs to adapt the reuse of water to the quality requirements.

Table 1.10. Most important national regulation regarding water reuse.

MS	Regulation title
Cyprus	KDP 379/2015. The Water Pollution Control (Recycled Water Quality). KDP 269/2005. Small-scale WWTPs (< 2000 p.e).
France	JORF num. 0153. Order of 2014, related to the use of water from treated urban wastewater wastewater for irrigation of crops and green areas.
Germany	DIN 19684. Soil structure. Chemical laboratory tests. Part 10: Investigation and assessment; Irrigation. Hygienic aspects of irrigation water. DIN 19650 (1999). Irrigation - Hygienic aspects of irrigation water.
Greece	JMD No. 145116/11. Definition of measures, terms and processes concerning the reuse of treated wastewater effluents.
Italy	DM 185/2003. Regulations laying down technical standards for the reuse of wastewater.
Malta	Subsidiary legislation 545.03. Water supply regulations. Legal Notice 210 of 2017. Water Supply (Amendment) Regulations, 2017.
Portugal	Decree-Law 236/98 – Standards, criteria and water quality objectives. NP 4434:2005. Reuse of reclaimed urban water for irrigation (guidelines).
Spain	RDL 1/2001. Consolidated text of the Water Law. RD 1620/2007. Legal framework for the reuse of treated wastewater.

- ✓ **Real Decreto 1620/2007, de 7 de diciembre, por el que se establece el régimen jurídico de la reutilización de las aguas depuradas:** This law defines the concept of reuse and introduces reclaimed water concept. Urban, agriculture, industrial, recreational and environmental uses are contemplated. The requirements for using reclaimed water, the procedures to obtain the concession required by law, as well as provisions relating quality requirements for each use, are included. Quality requirements per use are listed in Annex I. These criteria are considered as mandatory minimum requirements. Annex II contains the standardized application model that must be submitted to obtain the concession or authorization of reuse of reclaimed water.

- ✓ **Real Decreto-ley 4/2023, de 11 de mayo, por el que se adoptan medidas urgentes en materia agraria y de aguas en respuesta a la sequía y al agravamiento de las condiciones del sector primario derivado del conflicto bélico en Ucrania y de las condiciones climatológicas, así como de promoción del uso del transporte público colectivo terrestre por parte de los jóvenes y prevención de riesgos laborales en episodios de elevadas temperaturas:** This regulation establishes, among other matters, measures to promote the reuse of regenerated water in order to alleviate the effects of the drought that Spain is suffering. The law considers that purified effluents intended for reuse will not be considered "discharge" and, in addition, are exempt from paying the discharge fee.

1.5.2. Nutrients

Nutrients recovery from UWW has faced strong bottlenecks and barriers when they are obtained from a waste stream. However, the situation has improved, e.g., after the recognition as end-of-waste category for struvite.

1.5.2.1. European Union

- ✓ **Regulation (EU) 2019/1009 of the European Parliament and of the Council of 5 June 2019 laying down rules on the making available on the market of EU fertilizing products:** This regulation aims at harmonizing the conditions for making fertilizers, including those obtained from recycled and organic materials. Therefore, fertilizers commercialized in the UE must comply with the provisions of this regulation. This regulation explicitly cites struvite, biochar and ash-based products, but still as recovered wastes. However, the regulation also mandated the Commission to evaluate struvite, biochar and ash-based products for inclusion in the list of fertilizer products.

- ✓ **COMMISSION DELEGATED REGULATION (EU) 2021/2086 of 5 July 2021 amending Annexes II and IV to Regulation (EU) 2019/1009 of the European Parliament and of the Council for the purpose of adding precipitated phosphate salts and derivates as a component material category in EU fertilizing products:** This regulation amended Regulation (EU) 2019/1009 and explicitly allow considering as fertilizer those products containing phosphate salts produced by precipitation processes from, among other sources *"wastewaters and sewage sludge from municipal wastewater treatment plants, other than animal by-products or derived products within the scope of Regulation (EC) No 1069/2009".*

1.5.2.2. Spain

- ✓ **Ley 7/2022, de 8 de abril, de residuos y suelos contaminados para una economía circular:** This law includes a provision regarding the end of the status of waste for waste used in the manufacture of fertilizer products.

1.5.3. Biogas and biomethane

The use of anaerobic processes for energy recovery from organic matter has been driven by the climate crisis, the rise in fossil fuel prices and the vulnerability of dependence on unstable countries or regions. The most important regulation and strategic reports relates at European and Spanish level are presented.

1.5.3.1. European Union

- ✓ **Directive (EU) 2018/2001 of the European Parliament and of the Council on the promotion of the use of energy from renewable sources:** The recast Renewable Energy Directive (RED II) aims at stablishing a common framework for boosting renewable energy across European Countries. Among other considerations, e.g., targets for renewable energy share in the final energy consumption mix, financial aspects or self-consumption, it also proposes criteria on sustainability and GHG emissions for biofuels and biogas. Focus of the RED regarding biogas is mainly on its use in the transport sector.

- ✓ **RePowerEU Plan:** As a result of the war in Ukraine, the European Commission drew up a plan that aims at reducing energy dependence by promoting alternative energy sources and imports and progress toward an energy union within the EU. The plan includes measures for energy savings, supplier's diversification, clean energy transition acceleration and investment and regulatory innovation. It stablishes the goal of achieving a 55 % of net GHG emissions reduction by 2030 compared to 1990, and carbon neutrality by 2050. Regarding biogas and biomethane, it considers an investment need of € 37 billion to increase biomethane production to be able to substitute up to 35 billion of cubic meters (bcm) of natural gas by 2030. For that purpose, RePowerEU also proposes a Biomethane Action Plan to overcome potential barriers and boost biomethane production.

- ✓ **EN 16723-1:2016 - Natural gas and biomethane for use in transport and biomethane for injection in the natural gas network - Part 1: Specifications for biomethane for injection in the natural gas network**: This European Standard specifies the requirements and test methods for biomethane at the point of entry into natural gas networks.3

1.5.3.2. Spain

- ✓ **UNE-EN 16723-1:2017 - Gas natural y biometano para uso en transporte y biometano para inyección en la red de gas natural - Parte 1: Especificaciones para la inyección de biometano en la red de gas natural:** This standard coincides in its content with the European equivalent EN 16723:1:2016.

- ✓ **Plan Nacional Integrado de Energía y Clima (PNIEC) 2021-2030:** The Spanish Integrated National Plan of Energy and Clime 2021-2030 includes in its point 1.8 promoting renewable gases. This measure states the adoption of specific plans for boosting the production and use of renewable gases (e.g., biogas, biomethane and hydrogen, in line with RED II.

- ✓ **Ley 7/2021, de 20 de mayo, de cambio climático y transición energética:** in its article 12, this law establishes that the Government will promote, through the approval of specific plans, the production of renewable gases, including biogas, biomethane and renewable hydrogen, among others from organic waste or by-products of animal or vegetable origin.

- ✓ **Real Decreto 376/2022, de 17 de mayo, por el que se regulan los criterios de sostenibilidad y de reducción de las emisiones de gases de efecto invernadero de los biocarburantes, biolíquidos y combustibles de biomasa, así como el sistema de garantías de origen de los gases renovables:** The Royal Decree transposes part of the RED II. It establishes that each MWh of 100% renewable gas will give rise to the issuance of a guarantee of origin with information on where, when and how the gas was produced. The Royal Decree also creates a Census of Gas Production Facilities from renewable sources and a Committee of Producers.

1.6. Environmental and economic performance of urban wastewater treatment facilities

This section presents a brief summary of the state of the art about the environmental and economic performance of urban UWWT facilities, including those using UF membrane technology (e.g., DMF, AnMBR and tertiary membrane filtration), as well as a compendium of relevant studies in this field.

Regarding LCA, this assessment tool was introduced in WWT studies in the 1990s to get insight on environmental performance of the treatment processes and facilities` impacts (Emmeson et al., 1995). Since then, the number of studies focused on WWT has increased, according to the total number of papers available based on LCA (see section 1.4.3.1). In this sense, several reviews have been published that summarize the main findings and point out the controversies still presents in LCA field. As way of example, Corominas et al. (2013) analyzed 45 studies on WWT and LCA. The authors reported that, despite the issue of ISO standard, a high variability of functional units, system boundaries, methodologies and interpretations can be found, which make it difficult a rigorous comparison. As major challenges, the review considers boosting the role of LCA to tackle with UWW consideration as source of resources, expanding LCA methodologies to include new harmful compounds (e.g., UseTox model), better representing location-specific characterization, reducing data uncertainty and normalizing the use of LCA as policy-making tool. Moreover, Byrne et al. (2017) analyzed 256 papers focused on water infrastructure, 173 of them dealing with wastewater system. In this case, trends for definition of volume of treated water as functional unit; system boundaries limited to operation; and selection of CML, ReCiPe and Eco-Indicator methods for LCIA were detected. Authors also identified the potential benefit of combining LCA with LCC, social assessment and risk assessment and the necessity of developing case studies to

assess these approaches. All in all, big efforts have been made in the last years to collect succeed cases and best practices to improve LCA studies within water treatment. In this respect, Corominas et al. (2020) reviewed 121 papers aiming to developed useful guidance for researcher and practitioner to facilitate LCA application and favor robust conclusions and comparison between studies. The authors gathered outcomes and experiences and elaborated a consistent set of considerations and recommendations to support decisions along LCA phases.

With respect to economic performance, LCC tool has been developed to a lesser extent than LCA and the number of articles and research papers on its application to WWT is therefore lower. The first LCC study of a full-scale WWTP was published in 1997. As far as author knowledge, the most recent review on LCC studies on wastewater treatment is Ilyas et al. (2021), which compiled results of 83 papers from the period 2000 to 2020, 63 % of them correspond to 2015-2020. The authors detected an increment in research interest of LCC and a trend of combining LCA and LCC. However, LCC studies still lack detailed cost inventories and end-of-life considerations are usually left out of the analysis. The authors also pointed out the need of approaches harmonization to establish a coherent methodology able to facilitate comparison and obtaining robust conclusions.

Finally, and regarding economic and environmental performance of membranes and AnMBR technology, a selection of some relevant results of published studies are presented below:

- Lundie et al. (2004): the authors modelled the Sydney Water (main water service provider in Australia) environmental impact until 2021. LCA was conducted for an integrated water and wastewater system, which represented a novelty of the study. Among others, considered impact categories were global warming potential, eutrophication potential, photochemical oxidant formation potential, human toxicity potential, freshwater and marine aquatic ecotoxicity potential and terrestrial ecotoxicity potential. Functional unit was defined as annual water flow throughout the system. Results showed environmental benefits when keeping nutrients in the agriculture and recovering energy from biosolids. Moreover, the impact of chemical consumption cannot be ignored, while the contribution to environmental loads of infrastructure construction is small, below 4% in all categories considered.
- Rodriguez-Garcia et al. (2011): this study assessed the environmental and economic performance of 24 WWTPs of 6 different typologies based on quality requirements for discharge or reuse according to European Directive 91/271/EEC and Spanish Royal

Decree 1620/2007. Two functional units were defined for comparison purposes (1 m^3 of treated water and 1 kg of $PO_{4\,removed}^{3-}$). CML method was selected for LCIA and inventory was elaborated by means of previous studies, WWTP operators contribution and databases accessed via SimaPro (e.g., Ecoinvent). Regarding economic assessment, operation cost was considered and grouped into energy, staff and others category. Results showed selection of functional unit as a key factor for both environmental and economic performance. The study suggests 1 kg of $PO_{4\,removed}^{3-}$ over 1 m^3 of treated water since it reflects better the objective of UWWT. This work also recommends including social aspects when evaluating the sustainability of water treatment.

- Pretel et al. (2013): In this study, the author aimed to evaluate the environmental impact of an AnMBR for UWWT operating at 20 and 33 °C. Real data from a pilot plant were used to carry out an energy balance and a LCA (CML2 baseline 2000 methodology; Ecoinvent database; 1 m^3 of treated water as functional unit), considering energy consumed and recovered, potential nutrient recovery and sludge management. Results pointed out the importance of operating at ambient temperature and with an optimized filtration strategy to ensure AnMBR feasibility. An average energy consumption of 0.19 $kWh \cdot m^{-3}$ was obtained and LCA showed nutrient and methane recovery as a crucial factor to enhance AnMBR environmental performance.

- Lateef et al. (2013): authors investigated the use of hollow-fiber MF unit at lab-scale for DMF of low-strength raw UWW from the inlet of the primary settler of a WWTP (Sapporo, Japan). Up to 75 % of organic matter could be recovered and membrane fouling was mitigated by means of chemically enhanced backwash (CEB) using NaOCl and citric acid. Assuming 90 % of COD degradation and 40 % of electric conversion, energy recovery could be in the 0.14-0.25 $kWh \cdot m^{-3}$ range, which could increase up to 0.5 $kWh \cdot m^{-3}$ in a full-scale facility with higher COD concentration. This energy recovery potential is considered lower than the energy consumption for MBR, estimated in <0.4 $kWh \cdot m^{-3}$. Finally, DMF is considered as an alternative for retrofitting existing WWTP with space constrain, since its implementation and operation can be done within the current facilities.

- Pretel et al. (2014): This study assessed OPEX of an AnMBR treating sulphate-rich UWW and ambient temperature. Energy consumption, methane production and sludge management were considered. Minimum energy consumption of 0.07 $hWh \cdot m^{-3}$ was

achieved with appropriate sparging gas demand and SRT and minimum OPEX of 0.01 €·m^{-3} was obtained. Sulphate content was identified as a key element affecting methane production and, therefore, operating expenditure. Finally, energy production of 0.11 kWh·m^{-3} could be reach when treating low sulphate-loaded UWW in warm climates.

- Smith et al. (2014): in this study a comparison of AnMBR with i) high-rate activated sludge with anaerobic digestion (HRAS+AD); ii) conventional activated sludge with anaerobic digestion (CAS+AD); and AeMBR with AD (AeMBR+AD) was carried out to analyze economic (LCC) and environmental (LCA) performance. UWWT of 18950 $m^3 \cdot d^{-1}$ was considered as functional unit, inventory analysis was compiled by means of U.S. LCI, Ecoinvent and ELCD databases and TRACI method was selected for LCIA. Results determined sludge disposal as a key element for LCC: HRAS+AD showed the lowest life cycle cost in land-application or incineration scenarios and HRAS+AD and AnMBR presented lower LCC than CAS+AD and AeMBR+AD for all sludge fate scenarios, respectively. The highest LCC was obtained for AeMBR+AD for all sludge disposal practices. LCC results also showed CAPEX to be higher than OPEX for all technologies and scenarios. On the other hand, LCA results indicated AnMBR as the technology with higher global warming potential, but up to 75 % of the environmental impact was associated with dissolved methane. Hence, the use of recovery technologies (e.g., degassing membranes) can mitigate this effect. When treating high-strength UWW, environmental performance of AnMBR is improved and show comparable results than HRAS+AD.

- Ferrer et al. (2015): the authors presented guidelines for designing AnMBR for UWWT based on the operation of a AnMBR pilot plant. The objective is to minimize both CAPEX and OPEX considering HRT, SRT, MLTSS concentration, J_{20}, SGD, and sludge recycling flow. The methodology was applied with two operating temperatures (15 and 30 °C) and sulphate-rich (5.7 mg COD·mg^{-1} SO_4-S) and low-sulphate (57 mg COD·mg^{-1} SO_4-S) influent. TAC result with sulphate-rich UWW was between 0.101 and 0.097 €·m^{-3} with no energy recovery and energy recovery, respectively. On the other hand, result with low-sulphate UWW resulted in 0.097 and 0.070 €·m^{-3}.

- Pretel et al. (2015a): In this study, some UWWT layouts based on AnMBR were evaluated combined with primary settling and AD of wasted sludge. Two different UWW qualities were tested (sulphate-rich and low-sulphate) and two operating temperatures (15

and 30 °C). The AnMBR design was conducted based on real data from a pilot plant and simulation. The results showed that, in the case of low sulphate UWW and at high temperature, the AnMBR alone presented the lowest costs (0.05 €·m^{-3}) and energy recovery of 0.1 kWh·m^{-3}. On the contrary, the treatment scheme with primary settler, AnMBR and AD turned out to be the most economical option for UWW rich in sulphate treated at high temperature (0.05 €·m^{-3}). Finally, the total costs of AnMBR treatment are lower when treating low sulphate water (up to 45% lower).

- Pretel et al. (2015b): In this study, the authors assessed the technological, environmental, and economic sustainability of AnMBR technology considering specific design and operating strategy, e.g., SRT, MLTSS concentration, sludge recycling ratio, J and SGD. Methane recovery (both from biogas and dissolved methane), nutrient recovery and sludge fate (land application, landfill, or incineration) were also evaluated. To this end, LCC and LCA methodologies were applied, and a SA and UA were also conducted. Results confirmed that minimizing SRT improved environmental and economic performance and J, SGD, MLTSS and sludge recycling ratio required optimization to contribute to AnMBR feasibility. Additionally, MLTSS and J showed the highest effect on LCA and CAPEX and J also on CAPEX. Finally, operating at high temperature led AnMBR to perform as net energy producer.

- Pretel et al. (2016b): This study aimed at comparing the economic and environmental sustainability of AnMBR technology with aerobic-based treatments for moderate and high-loaded UWW. LCA and LCC were applied two different treatments schemes: AnMBR coupled with an aerobic-based post-treatment; AeMBR; and CAS. Results showed AnMBR with a CAS-based post-treatment for nutrient removal as a sustainable scheme for moderate- and high-loaded UWWT. Energy consumption as low as 0.04 kWh·m^{-3} and low sludge production could be obtained and significant reductions in different environmental loads on global warming potential, marine aquatic ecotoxicity, abiotic depletion and acidification could be reached. Finally, a minimum total cost of 0.135 €·m^{-3} was reported.

- Pretel et al. (2016a): In this study, the authors evaluated the economic and environmental performance of an AnMBR treating UWW jointly with kitchen waste at ambient temperature in mild/hot climates. The assessment included an energy balance, methane recovery, chemical consumption and sludge management. Obtained results showed

AnMBR as potential net energy producer with significant cost savings when treating low-sulphate UWW (up to 0.023 €·m^{-3}). Moreover, LCA outcomes indicated a positive effect of treating UWW jointly with kitchen waste on abiotic depletion, global warming potential and marine aquatic ecotoxicity.

- Morera et al. (2017): this study focused on assessing the environmental impact of the construction phase and equipment to the overall LCA of a conventional WWTP based on activated sludge process. To this aim, a systematic methodology starting from the existing Girona WWTP (Spain) was applied to compile all data of civil works and equipment required for LCI. A total of 45 materials and 1240 devices were included in the study, which was developed with ReCiPe methodology and Ecoinvent as process database. Lifetime for the WWTP was set to 20 years and 15 years for the equipment. Functional unit was defined as 1 m^3 of treated water. Unlike other studies (e.g., Lundie et al. (2004)), the results showed contributions above 5% for most of the impact categories considered (climate change, ozone depletion, freshwater eutrophication, marine eutrophication, human toxicity, metal depletion and fossil depletion). Especially relevant was the contribution of civil works and equipment to metal depletion (63%). The study concluded that an oversimplification of inventories might lead to an underestimation of the construction phase in the LCA.

- Carré et al. (2017): the authors evaluated different tertiary treatment platforms for water reclamation for 2 years, implemented downstream of a conventional secondary treatment in a WWTP. The platforms investigated were: i) sand filtration, storage and ultraviolet (UV) dynamic reactor disinfection (SF-UVD); ii) sand filtration and UV batch reactor disinfection (SF-UVB); iii) UF; iv) UF and UV batch reactor disinfection (UF-UVB); and v) MF, storage and dynamic UV disinfection (MF-UVD). Functional unit was 1 m^3 supplied water with highest quality, according to French regulation. Ecoinvent and PE International databases were used to construct the LCI and ReCiPe method was selected for LCIA. Results show operation phase as the main influencing phase for LCA for almost all environmental impact categories. SF+UVD and SF+UVB show similar impacts than UF, when membranes are operated without additional sterilization module. Otherwise, SF+UVD and SF+UVB present lower environmental loads. Finally, authors obtained similar operation cost for filtration+UV and deep filtration, 0.04-0.07 €·m^{-3}.

- Jodar-Abellan et al. (2019): this article reviews the water management context in Spain considering the water balances at river basin, legislative framework, sewage treatment, investment plans and reuse potential. Authors also addressed UWWT and reclamation costs. In the case of UV disinfection, investment costs were considered to be in the € 7.5-8.6 range per $m^3 \cdot d^{-1}$ of treatment flow and operating cost between 0.01 and 0.02 $€ \cdot m^{-3}$. For membranes (RO, UF and MF), investment costs between 200 and 400 € per $m^3 \cdot d^{-1}$ and operating cost of 0.05-0.09 $€ \cdot m^{-3}$ were reported. Finally, operating average costs of tertiary treatment in Comunitat Valenciana (Spain) were also included (0.06 $€ \cdot m^{-3}$).

- Xu et al. (2020): the objective of this study was to compare the environmental and economic performance of an aerobic vs anaerobic MBR (AeMBR and AnMBR, respectively) treating UWW under two scenarios: discharge and reuse of the effluent. The study included construction and operation phase and was developed by means of TRACI methodology. Functional unit was defined as 0.22 $m^3 \cdot s^{-1}$ of treated UWW achieving minimum USEPA secondary treatment effluent standards. Inventory was made from, among others, Ecoinvent and LCID databases. Results showed higher environmental loads and LCC for AnMBR under discharge scenario, mainly due to the post-treatment requirements for nutrients removal. On the other hand, reuse scenario made AnMBR able to operate with better environmental performance compared to AeMBR, thanks to nutrients recovery. Fertilizer value of the effluent also contributes to improve AnMBR economic competitiveness.

- Mohamad Mazuki et al. (2020): this article assesses the techno-economic performance of potential alternatives for disinfection based on single and multiple strains. Single units were UV, chlorination (Cl), MF and UF, while multiple platforms included UV, Cl, MF/UF, enviromulti-media (EMM) adsorption, or granular activated carbon (GAC) adsorption. Economy of scale was also evaluated considering two treatment capacities: 100 and 5000 $m^3 \cdot d^{-1}$. Obtained results showed the best removal efficiency for UF (single) and combination of UF+Cl (multiple) while economic evaluation established Cl (single) and EMM-UV (multiple) as the best cost-effective alternatives for restricted water quality, Finally, MF (single) presented the lowest cost for unrestricted reuse.
- Foglia et al. (2021): the authors investigated the environmental and economic impacts via LCA and LCC of an existing WWTP and compare different schemes to produce reclaimed water for agriculture. Disinfection methods (enhanced UV, peracetic acid), replacement of CAS with up flow anaerobic sludge blanket (UASB) and use of AnMBR

as the tertiary treatment were investigated. Current WWT (CAS) in the facility was considered as baseline. Ecoinvent and ReCiPe were applied for LCI and LCIA, respectively, and 1 m^3 of treated water was set as functional unit. The three scenarios evaluated (1: enhanced UV disinfection; 2: chemical disinfection using peracetic acid; and 3: biological treatment with UASB followed by AnMBR as tertiary treatment) reached class A effluent quality, according to Regulation (EU) 2020/741. Water reuse improved environmental performance of WWT compared to effluent discharge and the combination UASB+AnMBR showed the best environmental profile except to freshwater eutrophication, but this combination also obtained the highest overall cost, with a high influence of membrane investment and replacement.

- Ferreiro et al. (2021): apart from the operation and technical aspects of combining UF+ozonization (already commented in section 1.3.2.2), an energy assessment and economic evaluation of the proposed tertiary treatment was carried out and compared with the current tertiary treatment implemented in the Galindo WWTP (coagulation-floculation with $Al_2(SO)_4$ and NaClO, lamellar settler, sand filter and UV disinfection). According to the authors, energy consumption was 0.189 $kWh \cdot m^{-3}$ and 0.315 $kWh \cdot m^{-3}$ for conventional and UF+ozonization treatments, respectively. With regards to treatment costs, which also include chemical expenditures, conventional treatment account for 0.0534 $€ \cdot m^{-3}$ and UF+ozonization 0.0325 $€ \cdot m^{-3}$, from which 0.024 $€ \cdot m^{-3}$ corresponds to UF and 0.0042 $€ \cdot m^{-3}$ to chemicals.

- Pérez et al. (2022): this study evaluated the performance of a prototype plant for water reclamation (2.5 $m^3 \cdot h^{-1}$) based on UF and RO for industrial reuse. The authors offer results for each filtration technology for both water quality and costs. In the case of water quality, UF allows achieving a high quality permeate, with average E. Coli, turbidity and TSS of 17 $CFU \cdot 100\ mL^{-1}$, 0.25 NTU and 1.16 $mg \cdot L^{-1}$, respectively. Total costs for UF (CAPEX+OPEX) were calculated to be 0.630, 0.476 and 0.316 $€ \cdot m^{-3}$, for 2.5, 5 and 20 $m^3 \cdot h^{-1}$ of treatment flow, respectively. Results for RO were 0.470, 0.358, 0.256 $€ \cdot m^{-3}$.

- Hu et al. (2022): in this review, the authors analyzed different studies focused on AnMBR technology for urban and industrial wastewater, focusing on treatment configuration, energy balance and life cycle considerations. It is concluded that AnMBR process has great potential to achieve organics removal, bioenergy recovery and environmental protection. LCA studies showed that there is still room to enhance the economic and

environmental performance as well as the flexibility of the AnMBRs. Process integration requirement in terms of pre-treatment (e.g., preconcentration) and post-treatment technologies (e.g., dissolved methane management) is also highlighted.

- Shehata et al. (2023): the authors studied the contribution of membrane in achieving the sustainable development goals (SDGs). To this aim, nanofiltration, UF, MF, and forward osmosis and RO membranes were evaluated. As main conclusions can be mentioned: i) membrane-based water/wastewater treatment technologies have numerous applications in water reuse, water treatment, wastewater remediation, desalination, food industry, biofuel, and value-added materials production; ii) membrane-based technologies in water and wastewater treatment directly impact 15 SDGs out of 17; and iii) high initial cost, high energy requirements, membrane fouling are identified among the major bottlenecks for membrane technology implementation.

- Mateo et al. (2023): this research evaluated the technical feasibility of combining AnMBR, primary settling and AD to treat sulphate-rich UWW at ambient temperature with the aim of reducing CO_2 emissions and enhance energy recovery. This treatment scheme reduces the COD:SO_4^{-2} ratio in the water line while increases it in the sludge line entering the AD. Therefore, SRB removes COD in AnMBR, which produces a methane-free effluent, and particulate organic matter is valorized in the AD, producing high-quality biogas. The authors reported an energy production increase of 0.03 $kWh \cdot m^{-3}$ over conventional AnMBR production, estimated in 0.05-0.60 $kWh \cdot m^{-3}$, and reduction in CO_2 between 0.397 kg $CO_{2eq} \cdot m^{-3}$ (operating at 25 ºC) and 0.478 kg $CO_{2eq} \cdot m^{-3}$ (operating at 15 ºC).

1.7. Scope and outline of this thesis

Based on previous studies on the use of UF membrane technology at different UWWT steps (i.e., primary, secondary and tertiary), it is expected that UF technology would allow, under certain conditions, to contribute to boosting the necessary paradigm shift that definitively considers UWW as a valuable source of resources from which to obtain, among others, reclaimed water, energy and nutrients. At the same time, this resource recovery perspective would promote the economic and environmental sustainability of WWTPs.

Therefore, the main objective of this PhD Thesis is to elucidate the technical, economic and environmental feasibility of applying UF membranes at different treatment steps for UWW treatment, considering a Circular Economy approach. To this aim, data from three different UF-based demonstration plants (DMF, AnMBR and tertiary membrane filtration) was used to obtain robust and representative results:

The general objective of this thesis can be divided into the following specific objectives:

1. To assess the technical feasibility of the studied UF-based technologies for UWWT:

    a) To estimate the potential of DMF to maximize organic carbon valorization in existing WWTPs.
    b) To estimate the potential of AnMBR for resource recovery from UWW at ambient temperature.
    c) To estimate the potential of tertiary membrane filtration to produce reclaimed water in existing WWTPs.
    d) To evaluate the potential of the evaluated treatment solutions to produce reclaimed water with different qualities.

2. To elucidate operating conditions and strategies to optimize the performance of the evaluated technologies from an economic and environmental point of view.

3. To estimate the CAPEX, OPEX and TAC of the evaluated technologies.

4. To assess the environmental performance of the evaluated technologies.

5. To navigate environmental, economic, and technological trade-offs through sensitivity and uncertainty analysis of AnMBR technology for UWWT.

This PhD Thesis has been developed within the framework of the four projects listed below:

- Innovation Deal on sustainable wastewater treatment combining anaerobic membrane technology and water reuse (2016-2018). European Commission. (Chapter 3)
- Application of membrane technology to enhance the transformation of WWTPs in resource recovery facilities (*Modelizacion y control para la implementacion de la*

tecnologia de membranas en las EDAR actuales para su transformacion en estaciones de recuperación-MEM4REC) (CTM2017-86751-C2-2-R-AR). Spanish National Research Agency-AEI. (Chapter 4)

- Membrane for ENERGY and WATER RECOVERY-LIFE MEMORY (LIFE13 ENV/ES/001353) (2014-2018). Life+ Programme. (Chapters 5, 6, 7 and 8)
- *Caracterización de aguas y lodos residuales urbanos e industriales y evaluación de la tecnología adecuada para la recuperación de microplásticos presentes en los mismos* - MICROPLAST (INNEST00/19/058). Valencian Innovation Agency-AVI. (Chapter 9)

Finally, chapters 3 to 9 from this PhD Thesis correspond to articles published or submitted for publication to scientific journals (compendium of articles):

- Chapter 3: A. Jiménez-Benítez, F. J. Ferrer, S. Greses, A. Ruiz-Martínez, F. Fatone, A. L. Eusebi, N. Mondéjar, J. Ferrer, A. Seco (2020) AnMBR, reclaimed water and fertigation: Two case studies in Italy and Spain to assess economic and technological feasibility and CO_2 emissions within the EU Innovation Deal initiative. *Journal of Cleaner Production*, 270; 122398 (IF: 9.297; Q1 89.81 (6/54) in Environmental Engineering category) (source JCR 2022; accessed 18-10-2023).
- Chapter 4: A. Jiménez-Benítez, P. Sanchís-Perucho, J. Godifredo, J. Serralta, R. Barat, A. Robles, A. Seco (2023) Ultrafiltration after primary settler to enhance organic carbon valorization: energy, economic and environmental assessment. Submitted to *Journal of Water Process Engineering,* (IF: 7.0; Q1 85.6 (21/142) in Chemical Engineering category) (source JCR 2022; accessed 18-10-2023). Currently under second peer-review process.
- Chapter 5: A. Robles, A. Jiménez-Benítez, J. B. Giménez, F. Durán, J. Ribes, J. Serralta, J. Ferrer, F. Rogalla, A. Seco (2022) A semi-industrial scale AnMBR for municipal wastewater treatment at ambient temperature: performance of the biological process. *Water Research,* 215; 118249 (IF: 12.8; Q1 90.0 (6/55) in Environmental Engineering category) (source JCR 2022; accessed 18-10-2023).
- Chapter 6: A. Jiménez-Benítez, A. Ruiz-Martínez, A. Robles, J. Serralta, J. Ribes, F. Rogalla, A. Seco, J. Ferrer (2023) A semi-industrial AnMBR plant for urban wastewater treatment at ambient temperature: Analysis of the filtration process, energy balance and quantification of GHG emissions. *Journal of Environmental Chemical Engineering,* 11; 109454 (IF: 7.7; Q1 89.1 (16/142) in Chemical Engineering category) (source JCR 2022; accessed 18-10-2023).

- Chapter 7: A. Jiménez-Benítez, A. Ruiz-Martínez, J. Ferrer, J. Ribes, F. Rogalla, A. Robles (2023) Life cycle costing of AnMBR technology for urban wastewater treatment: A case study based on a demo-scale AnMBR system. *Journal of Environmental Chemical Engineering,* 11; 110267 (IF: 7.7; Q1 89.1 (16/142) in Chemical Engineering category) (source JCR 2022; accessed 18-10-2023).
- Chapter 8: A. Jiménez-Benítez, J.R. Vázquez, A. Seco, J. Serralta, F. Rogalla, A. Robles (2023) Life cycle costing of AnMBR technology for urban wastewater treatment: A case study based on a demo-scale AnMBR system. *Journal of Environmental Chemical Engineering,* 11; 111141 (IF: 7.7; Q1 89.1 (16/142) in Chemical Engineering category) (source JCR 2022; accessed 18-10-2023).
- Chapter 9: A. Jiménez-Benítez, J. González-Camejo, N. Martí, A. Robles, A. Seco (2023) Effect of seasonal dynamics on regenerated water reclamation from ultrafiltration membrane-based tertiary urban wastewater treatment: energy expenditure, cost, environmental and effluent quality assessment. Submitted to *Journal of Water Process Engineering,* (IF: 7.7; Q1 89.1 (16/142) in Chemical Engineering category) (source JCR 2022; accessed 18-10-2023). Currently under first peer-review process.

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2. Materials and methods

2.1. Introduction

For the development of this PhD Thesis, different devices, techniques, procedures and methodologies have been applied This section intends to briefly describe the most representative ones, without prejudice to the fact that a description is also made in the following chapters based on each specific study.

2.2. Demonstration plants

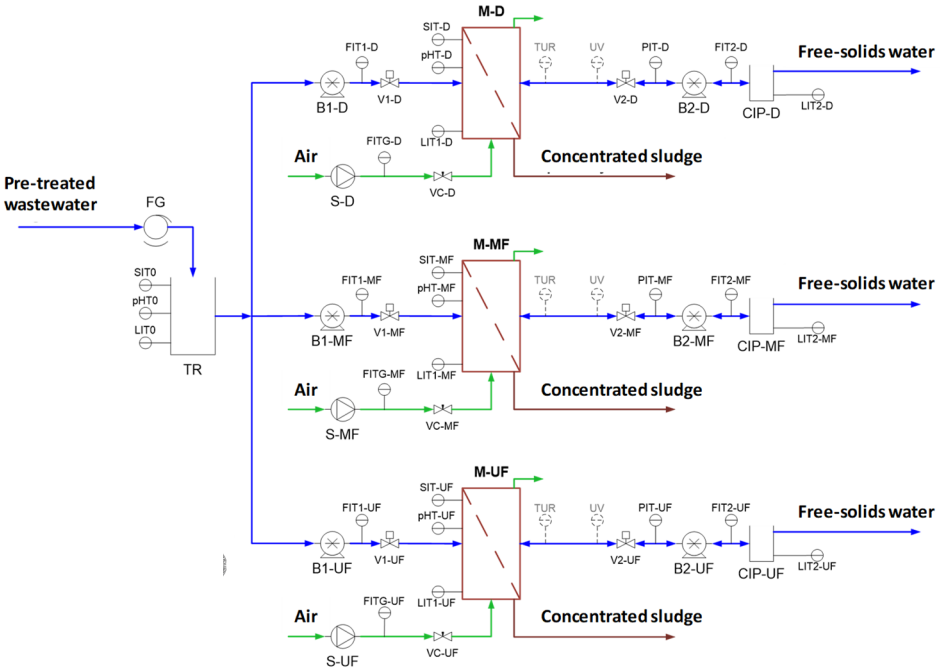
Data from three demonstration plants has been used in this PhD Thesis. Their most relevant components and characteristics are described below. It is important to clarify that the design and operation of these plants are outside the scope of this PhD Thesis, although the foreground data obtained from these systems constitute a crucial part of the data set used in the research carried out here.

2.2.1. MEM4REC plant

MEM4REC project (CTM2017-86751-C2-1-R) studied the combination of conventional wastewater treatments (e.g., primary settler, activated sludge and anaerobic digestion) with ultrafiltration (UF)-based direct membrane filtration (DMF) process to improve the organic matter valorization in the treatment facility. The adoption of membrane technology in existing treatment plants is a possible way to upgrade them, allowing treatment capacity to be increased and improving energy balance. To this aim, a demonstration plant was designed, commissioned and operated. Figure 2.1 shows the flow diagram and a picture of this system.

MEM4REC demonstration plant consisted of three independent treatment lines, similar in all their components but the membrane type. Each membrane tank was fitted with a different industrial membrane module: dynamic (M-D; average pore size depending on the experimental; period filtration area 2 m^2); microfiltration (M-MF; TERAPORETM 5000, 0.4-µm pore size; filtration area 18 m^2); and UF (M-UF; PULSION® Koch Membrane Systems, 0.03-µm pore size; filtration area 43.5 m^2). Vacuum filtration proceeded through a screw pump (PCM M series, EcoMoineau™). The system included a clean-in-place (CIP) tank in each line to store the generated permeate and allow membrane backwashing during continuous operation. Fouling propensities was addressed by means of air scouring using blowers (G-BH7, Elmo Rietschle) and a recirculation stream provided by pumps PCM M series, EcoMoineau™ in the membrane tanks, to guarantee complete mixing and prevent solids stratification. Two different influents were used

to feed the plant: i) pre-treated raw urban wastewater (screening and sieving, desanding and degreasing); and ii) effluent from primary settler from the wastewater treatment plant (WWTP). Further details can be found elsewhere in Sanchis-perucho et al. (2023) and Sanchis-Perucho et al. (2022). For the purpose of this PhD Thesis, data regarding UF treatment line and raw urban wastewater feeding was considered.



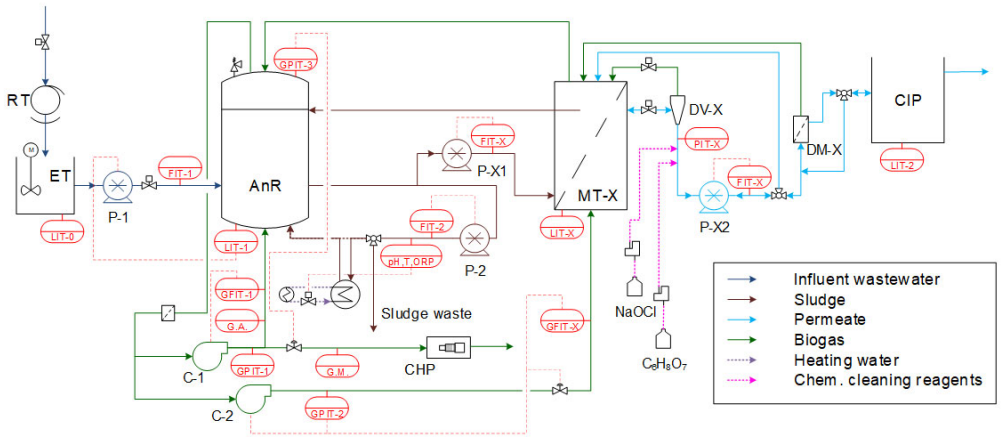
a)

b)

Figure 2.1. MEM4REC demonstration plant: a) process diagram and b) photo.

2.2.2. LIFE MEMORY plant

The LIFE MEMORY project (LIFE13 ENV/ES/001353) aimed at demonstrating the technical, economic and environmental feasibility of anaerobic membrane bioreactor (AnMBR) technology to treat urban wastewater (UWW) as an alternative to conventional treatment schemes based on activated sludge process. To this aim, a demonstration plant was designed, commissioned and operated. Figure 2.2 shows the flow diagram and a picture of this system.



a)

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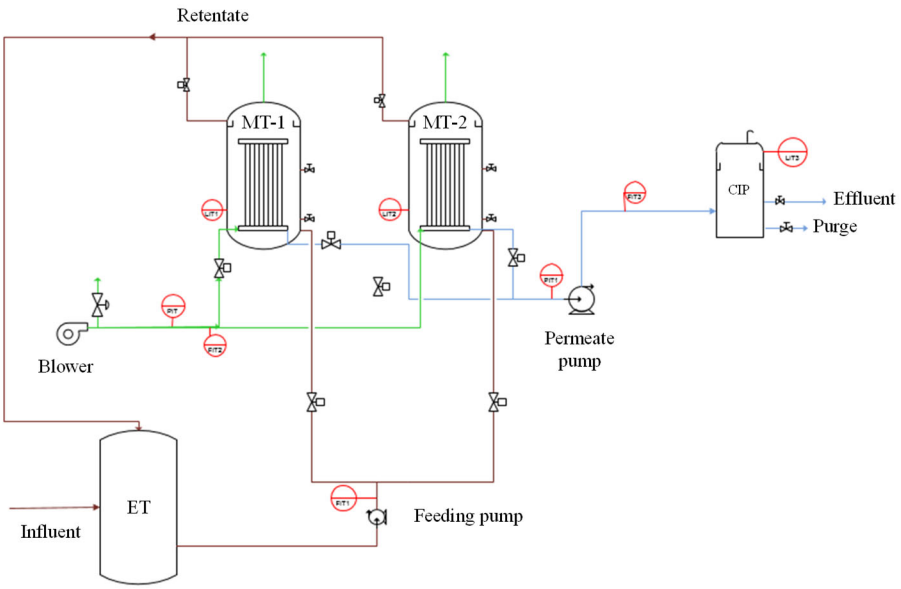
Figure 2.2. LIFE MEMORY demonstration plant: a) process diagram and b) photo.

LIFE MEMORY demonstration plant consists of a 40 m^3 (AnR, 34.4 m^3 working volume + 5.6 m^3 headspace) anaerobic reactor connected to three 0.8 m^3 membrane tanks (MT-X, 0.7 m^3 working volume + 0.1 m^3 headspace). Each membrane tank was fitted with one ultrafiltration membrane system (PURON® PSH41, KMS, 0.03-µm pore size, total filtration area of 41 m^2), giving a total filtration area of 123 m^2. A sieve screw (RT, 1.5-mm screen size), an equalization tank (ET, 1.1 m^3) and a CIP tank (CIP, 0.37 m^3) were the other main elements in the plant. Total sludge volume was recorded considering the working volumes of both the anaerobic reactor and membrane tanks, accounting for 36.32±1.63 m^3 during the operating period. The AnMBR was equipped with a 2.1 m^2 degassing membrane (DM) unit for dissolved methane recovery, consisting of a polydimethylsiloxane (PDMS) hollow-fiber commercial module provided by PermSelect®, MedArray Inc. USA. The plant was fed with effluent from the pre-treatment step (sand and grease removal) of the Alcázar de San Juan WWTP (Alcázar de San Juan, Ciudad Real, Spain). For further details, see Robles et al. (2020).

2.2.3. MICROPLAST plant

The main objective of MICROPLAST project (INNEST00/19/058) was to study the presence of microplastics and their fate along the different WWTP steps. Among the specific objectives of the project, it included the design, commissioning and operation of a UF-based tertiary treatment of UWW. Figure 2.3 shows the flow diagram and a picture of this system.

The MICROPLAST demonstration plant mainly consisted of two 15-L membrane tanks (MT1 and MT2) with a working volume of 30 L, each containing a 2.5-m high hollow-fiber bundle (KMS Puron® Koch Membrane Systems) with a membrane area of 3.44 m^2 each and average pore size of 0.03 µm. One bundle was used for continuous operation and the other was kept in reserve for use during membrane cleaning or system maintenance. The plant also had a 30-L clean-in-place tank (CIP) to store permeate for membrane backwashing as required. Any tank overflow was recycled to a secondary clarifier in the WWTP. To reduce cake layer formation on the membrane surface, a blower provided air bubbles for sparging purposes. The membrane combined the conventional filtration–relaxation (F–R) stages and backwashing. Duration of filtration and relaxation stages were set to 150 and 30 s, respectively, while a backwashing for 60 s was conducted every 5 F-R cycles. Filtration process was controlled by an instrumentation, control and automation (ICA) system based on sensor-transmitters and actuators connected to a programmable logic controller (PLC), an automation system and a Supervisory Control and Data Acquisition (SCADA). The control philosophy used was the one proposed by Robles et al. (2012).



a)

b)

Figure 2.3. MICROPLAST demonstration plant: a) process diagram and b) photo.

2.3. Energy model

As already mentioned in Chapter 1 (Introduction), energy assessment was based on the model proposed by Pretel et al. (2016b), which was coupled to the extended BNRM2S plant-wide mathematical model published by Durán et al., (2020). In this energy model, Equation 2.1 represents the total energy demand:

$$\Delta E_P + \Delta E_K + \Delta h = W + Q \qquad \text{Equation 2.1}$$

Where E_P is the potential energy, E_K is the kinetic energy, h is the internal energy, W the work applied by the system and Q is the transferred heat. Each element of Equation 2.1 is then breakdown into their components. Table 2.1 shows the principal equations for the purpose of this PhD Thesis.

Table 2.1. Equations employed for power assessment (adapted from Judd & Judd (2006)).

Energy element	Equation	Nº
P_B ($J \cdot s^{-1}$): blower power requirement (adiabatic compression)	$P_B = \frac{(M \cdot R \cdot T_{gas})}{(\alpha - 1) \cdot \eta_{blower}} \left[\left(\frac{P_2}{P_1} \right)^{\frac{\alpha - 1}{\alpha}} - 1 \right]$	2.2
P_g ($J \cdot s^{-1}$): power required by a general pump	$P_g = q_{imp} \cdot \rho_{liquor} \cdot g \cdot \frac{\left\{ \left[\left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{asp} + \left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{imp} \right] + [Z_1 - Z_2] \right\}}{\eta_{pump}}$	2.3
P_{stage} ($J \cdot s^{-1}$): power requirement during filtration, degasification or back-flushing	$P_{stage} = \frac{q_{stage} \cdot TMP_{stage}}{\eta_{pump}}$	2.4
TER ($kWh \cdot m^{-3}$): total energy recovery	$TER = -\frac{(Q_{biogas} \cdot y_{biogas}^{CH_4} + Q_{dissolved} \cdot y_{dissolved}^{CH_4}) \cdot CV_{CH_4} \cdot \eta_{CHP}}{Q_{inf}}$	2.5

M: Molar flow rate of gas ($mol \cdot s^{-1}$)
R: Gas constant for gas ($J \cdot mol^{-1} \cdot K^{-1}$)
P_1: Absolute inlet pressure (atm)
P_2: Absolute outlet pressure (atm)
T_{gas}: Inlet air/gas temperature (K)
α: Adiabatic index
η_{blower}: Blower efficiency
ρ_{liqour}: Sludge density ($kg \cdot m^{-3}$)
g: Acceleration of gravity ($m \cdot s^{-2}$)
q_{imp}: Impulsion volumetric flow rate ($m^3 \cdot s^{-1}$)
L: Pipe length (m)
L_{eq}: Equivalent pipe length of accidental pressure drops (m)
V: Velocity ($m \cdot s^{-1}$)
f: Friction factor
D: Pipe diameter (m)
Z_2 -Z_1: Height difference (m)
η_{pump}: Pump efficiency
TMP: Transmembrane pressure (Pa)
q_{stage}: Pump volumetric flow rate ($m^3 \cdot s^{-1}$)
Q_{biogas}: Biogas production in the anaerobic reactor at normal conditions of temperature and pressure (NTP) (m^3 NTP$\cdot d^{-1}$)
$y_{biogas}^{CH_4}$: Methane fraction in the anaerobic reactor biogas
$Q_{dissolved}$: Biogas flow recovered from the effluent at NTP (m^3 NTP$\cdot d^{-1}$)
$y_{dissolved}^{CH_4}$: Methane fraction in the biogas recovered from the effluent
CV_{CH_4}: Calorific value of CH_4 (10.6 kWh$\cdot$NTP m^{-3})
η_{CHP}: CHP efficiency
Q_{inf}: Wastewater influent flow ($m^3 \cdot d^{-1}$)

2.4. Life cycle assessment (LCA)

Environmental impact was evaluated by means of LCA according to ISO 14040:2006 and ISO 14043:2006. Among the different LCA methodologies available to perform LCIA phase, ReCiPe 2016 has been selected since it is a harmonized method which covers a wide range of environmental impacts at midpoint level (18 categories) and also aggregates them into three areas of protection or endpoint levels (see Figure 2.4).

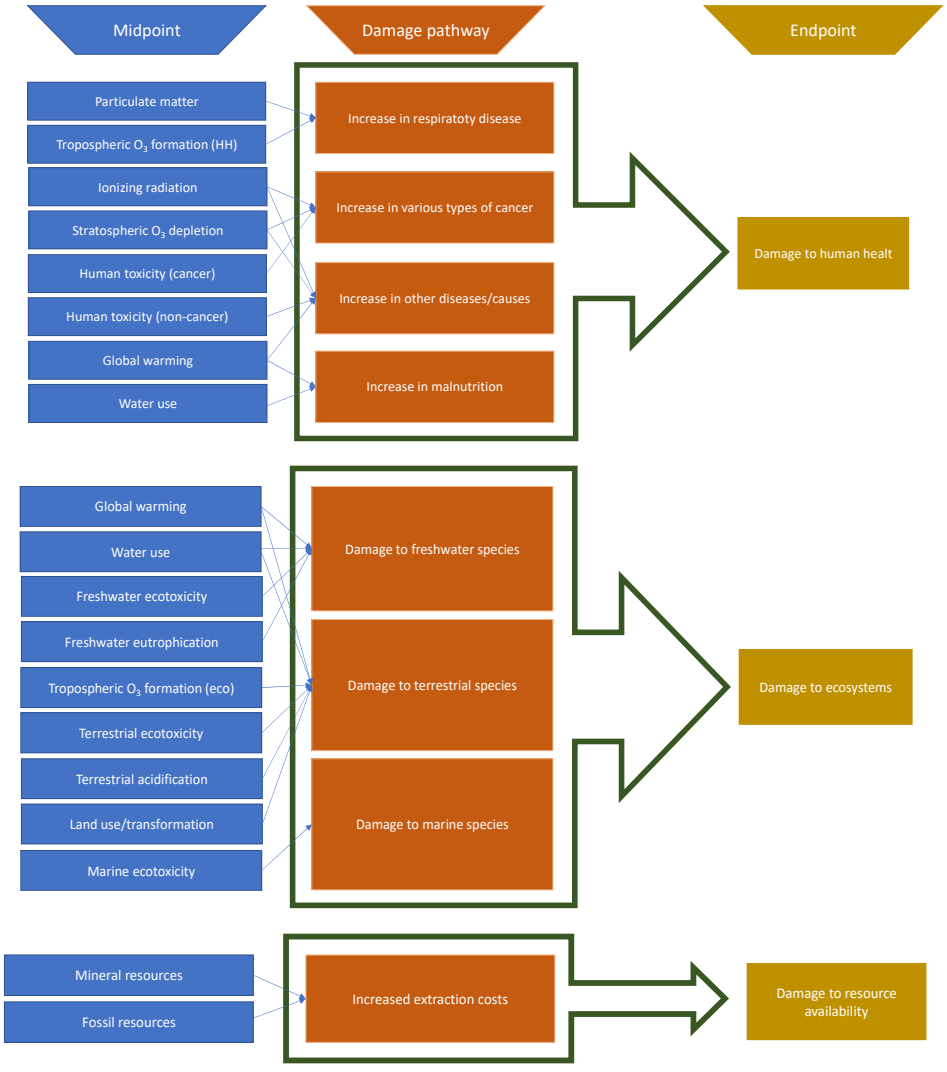


Figure 2.4. Impact categories at midpoint, endpoint and their relation in ReCiPe 2016 method (adapted from Mark A.J. Huijbregts et al. (2017).

ReCiPe 2016 also contemplates three approaches under which it estimates the characterization factors that take into account, among others, different time horizons for the life of contaminants

or the available scientific evidence on the environmental effects associated with some impacts (Huijbregts et al., 2016). These three approximations are the following:

- **Individualist:** This approach is based on optimism in relation to human adaptation capacity thanks to technology, considers the impacts with solid well-stablished evidence and proposes a short time horizon to assess the effects of pollution (20 years).
- **Hierarchist:** This perspective considers the current scientific consensus regarding the time horizon (100 years) and the processes that originate the impacts from contamination.
- **Egalitarian:** is the most conservative approach. It considers the longest time horizons (1000 years or more) and all impact possibilities for which data is available.

Finally, ReCiPe 2016 uses impact mechanisms and processes appropriate for a global perspective. However, it also includes site-specific characterization factors (i.e., at country, region, continent level) for some impact categories which were considered relevant for (e.g., particulate matter formation, photochemical ozone formation or acidification).

Both midpoint and endpoint impact categories were considered in this PhD Thesis, which were applied throughout hierarchist approach.

2.5. Life cycle costing (LCC)

The economic feasibility of the evaluated systems was assessed through LCC by means of the experimental data obtained from the different demonstration plants, background data (e.g., energy cost), cost functions and suppliers information. As per LCA, LCC was carried out following the recommendations and statements stablished in ISO 14040:2006 and ISO 14044:2006. LCC was used to calculate capital and operating expenses (CAPEX and OPEX, respectively) and the total annual costs (TAC), defined in Equation 2.6:

$$TAC = \frac{r\,(1+r)^t}{(1+r)^t - 1} \cdot CAPEX + OPEX$$

Equation 2.6

where r is the annual discount rate and t is the depreciation period in years.

The general considerations and assumptions considered in this PhD Thesis were: i) 20-year depreciation period for building works (annual discount rate 3%) and 10 years for equipment (annual discount rate 5%); ii) energy costs considered both biogas and dissolved methane recovery by DM; iii) UF membrane lifespan was calculated from the permissible maximum total contact with chlorine before membrane replacement, i.e., 500000 ppm-hours cumulative, according to the supplier, and experimental chemical cleaning frequency; iv) lifetime of blowers and pumps was based on the total working hours recommended by the manufacturers (50000 and 75000 h, respectively); v) equipment costs were estimated by correlations between capacities (mainly based on flow rates and power) and the unit costs provided by the manufacturers, when possible; and vi) labor costs were not included.

2.6. Global sensitivity and uncertainty analyses

Sensitivity analysis (SA) was carried out to determine the most significant input parameters to LCA and LCC, while uncertainty analysis (UA) was used to evaluate the related uncertainty in the obtained results. Regarding SA, the Standardized Regression Coefficients (SRC) method was applied to select the most influential parameters of all those that make up the LCA/LCC model. A threshold of 0.1 (absolute value) for the standardized regression slope (b_i) was chosen to select the influential input factors. UA was conducted by means of Monte Carlo simulations to evaluate the propagation of uncertainty in the results, which were assessed by means of i) the 5th and 95th percentiles and ii) the empirical cumulative distribution function (eCDF). The sampling matrix was generated by the Latin hypercube sampling (LHS) method.

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3. AnMBR, reclaimed water and fertigation: two case studies in Italy and Spain to assess economic and technological feasibility and CO2 emissions within the EU Innovation Deal Initiative

Abstract

The use of anaerobic membrane bioreactor (AnMBR) technology on urban wastewater can help to alleviate droughts, by reusing the water and nutrients embedded in the effluent in agriculture (fertigation) in line with Circular Economy principles. The combination of AnMBR and fertigation reduces CO_2 emissions due to the organic matter valorization and the partial avoidance of mineral fertilizer requirements. However, both AnMBR and fertigation still face technological and regulatory barriers that need to be overcome. These bottlenecks were tackled within the first Innovation Deal approved by the European Commission in 2016 and gave rise to several case studies on water reuse systems. The results of the Oliva Wastewater Treatment Plant (Spain) and Peschiera-Borromeo Wastewater Treatment Plant (Italy) showed that reclaimed water can be considered as a reliable water and nutrient source, return a positive economic balance (up to 376 k€·year^{-1}) and provide significant reductions and savings in CO_2 emissions (up to -898.9 tCO_2·year^{-1}). According to the new EU regulation, a new key player known as the *Reclaimed Water Manager*, was also proposed to be in charge of supplying reclaimed water with appropriate quantity and quality to end-users. This new agent would also be responsible for drawing up and implementing a Water Reuse Risk Management Plan in cooperation with the parties involved. Applying AnMBR technology to water reuse thus shows potential for contributing to catchment-scale Circular Economy while preserving natural water bodies, reducing the carbon footprint and creating new business opportunities. However, to take full advantage of its benefits demonstration projects would need to be carried out and favorable and harmonized regulations among the EU States would need to be adopted.

Keywords

Agricultural water reuse; Anaerobic digestion; Membrane technology; Reclaimed water; Nutrient recovery and reuse; Sustainability assessment

Highlights

AnMBR technology appears as a feasible technology for valorizing urban wastewater.
Reclaimed water can be considered a reliable water and nutrient supply source.
Water reuse risk management plan is needed for environmental and health safety.
AnMBR + fertigation can contribute to significant reductions of CO_2 emissions.
Positive economic balance can be obtained in different water reuse scenarios.

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3.1. Introduction

The worldwide population growth has caused and increasing demand for food and thus greater water extraction and higher consumption of fertilizers (Hussain et al., 2019). According to Ledger et al., (2012) as an additional pressure the current climate change conditions are expected to affect rain patterns and cause water scarcity in many regions. This situation calls for better efficiency in the use of water resources.

In this context, Circular Economy (CE) principles are gaining momentum as part of the solution (D. Puyol et al., 2017). Their adoption in the water value chain is shifting the conventional wastewater treatment plant (WWTP) model to the water resource recovery facility (WRRF) concept. This can help to achieve economic and environmental goals within the water sector whilst producing valuable goods from a renewable source, including reclaimed water, nutrients, energy, etc.

To promote this paradigm shift, anaerobic membrane bioreactors (AnMBR) have been proposed as potential substitutes for conventional WWTPs, which are generally based on aerobic processes. AnMBR technology combines the advantages of membrane filtration with those of anaerobic processes: effluent quality, organic matter valorization, reduced carbon footprint, low biosolids production and on-site stabilization and nutrients recovery potential. AnMBR applications for industrial wastewater are already well-established (Dereli et al., 2012) and promising results for urban wastewater treatment have been reported: Giménez et al., (2011) showed almost 90 % COD removal and a negligible fouling rate when operating a submerged pilot-scale anaerobic membrane bioreactor with a 10 LMH transmembrane flux and 22 $g \cdot L^{-1}$ suspended solids in the reactor treating real urban wastewater; Ozgun et al., (2013) reported on the membranes' contribution to maintaining high treatment efficiency despite temperature fluctuations due to the retention of particulate COD and biomass in the reactor; Foglia et al., (2019) demonstrated the ability of a combination of UASB and AnMBR to treat high-salinity low-loaded municipal wastewater in the long term with a reduced methane production but without membrane fouling and producing a high-quality effluent, whereas Pretel et al., (2016b) compared different aerobic-based WWTP to AnMBR coupled to a post-treatment for nutrient removal, observing that AnMBR combined with a conventional activated (CAS) sludge-based post-treatment reduced the environmental impact; finally, Becker et al., (2017) showed AnMBR's energy recovery potential when treating joint wastewater and food waste.

However, some issues still need to be addressed in order to improve AnMBR technical, economic and environmental feasibility: the sensitivity and control of process dynamics have been pointed out by Robles et al., (2018); Velasco et al., (2018) focused on the release of dissolved methane with the effluent; several authors (e.g. Aslam et al., 2018; Judd, 2017; Maaz et al., 2019) coincide in considering membrane fouling as one of the major bottlenecks; and Smith et al., (2012) underlined eutrophication potential due to the nutrients content of its effluent. However, it is precisely the presence of N and P in the AnMBR effluent, together with the high quality of the water, that makes their application for fertigation an attractive approach for resource recovery.

Fertigation (simultaneous reuse of water and nutrients) helps to conserve freshwater sources and reduces the energy consumed for industrial ammonia-based fertilizer production (19.3 kWh·kg N^{-1} by the Haber-Bosh process, according to McCarty et al., 2011) and P extraction (2.11 kWh·kg P^{-1} according to Gellings and Parmenter, 2004). Financial benefits for farmers can also be expected from savings in mineral fertilizers (Scheierling et al., 2011). P was included in the EU's Critical Raw Materials list in 2017 (European Commission, 2017), which justifies strategies for reducing European dependency on nutrient importation from a food security perspective as well as from an environmental point of view.

Several documents, regulation proposals, reports and scientific papers have been published on the benefits of water reclamation and its related issues (Alcalde-Sanz and Gawlik, 2017; General Secretariat of the Council, 2019; WWAP, 2017). As stated by Condom et al., (2012), 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 feasibility and sustainability of water reuse projects and plans The EU has already recognized that the use of reclaimed water has a smaller environmental impact than other alternatives, i.e. water transfers or desalination, while it also has financial and social benefits (European Commission-DG ENV, 2012). The main current concerns are related to microbial risks to public health and the environment risks. These are generally addressed by a multibarrier approach in wastewater treatment/reclamation and the development and implementation of water reuse risk management plans (WRRMP).

Based on this background, and with the aim of investigating whether the AnMBR technology for reclaiming water for fertigation faces regulatory barriers in the EU, the European Commission (EC) approved the first pilot Innovation Deal (ID) entitled *Sustainable wastewater treatment*

combining anaerobic membrane technology and water reuse in 2016. The ID consortium consisted of 14 partners from 5 European member states (Portugal, Spain, Malta, Italy and France) and included a comprehensive group of stakeholders involved in water management: national and regional authorities, universities, research centers, innovators and end-users. Three EC Directorate-Generals (Research and Innovation, Agriculture and Rural Development and Environment) were also represented.

The tasks carried out within the ID included the analysis of several case studies, two of which are presented in this study. These cases are located in the Mediterranean Region (Italy and Spain) and compare the application of combined AnMBR technology and fertigation with conventional configurations of WWTPs with aerobic systems, irrigation with freshwater and mineral fertilization. To the best of authors' knowledge, this has not been reported on before. The work carried out also included the development of the novel concept of the Reclaimed Water Manager (RWM), a new key player in accordance with the EU regulation, who will be in charge of operating water reclamation and reuse systems and responsible for drawing up and applying the WRRMP. Both case studies examined different scenarios and compared them to the current case. An economic study was performed for the RWM and the levels of wastewater resource recovery and CO_2 emissions were assessed for all configurations.

3.2. Material and methods

Each case study encompassed different scenarios in which at least one was AnMBR technology. A four-step assessment methodology was developed to analyze them: (i) water balance, (ii) nutrient balance, (iii) economic analysis and (iv) CO_2 emission estimations. Several reference values were used (see below in this section), based on the literature and on data provided by stakeholders (e.g., efficiency factors, water and nutrients needs, energy needs, energy prices, etc.). Further studies must find the most appropriate variables for each specific case in order to increase the accuracy of the results. Information on the end-users' perception on water reuse for agriculture was also gathered and analyzed.

3.2.1. Water balance

In the first step, the *net crop requirements* or *net provision* ($m^3 \cdot ha^{-1} \cdot year^{-1}$) is calculated, considering the type of crop and the location of the site, since meteorological (effective precipitation) and soil data (available water stored in the soil) exert a considerable influence on water availability. The second step is a water balance applied to the irrigation network in order to calculate the *gross irrigation volume* (V_B in $hm^3 \cdot year^{-1}$) that will be extracted from the water source, since transport, distribution and irrigation networks suffer water losses, known as transport network return flows (r_t), distribution network return flows (r_d) and irrigation return flows (r_a). There is also a water loss associated with evaporation throughout the irrigation network: evaporation in the transport network (p_t), in the distribution network (p_d) and in the irrigation volume applied to the land (p_a) (see Figure 3.1).

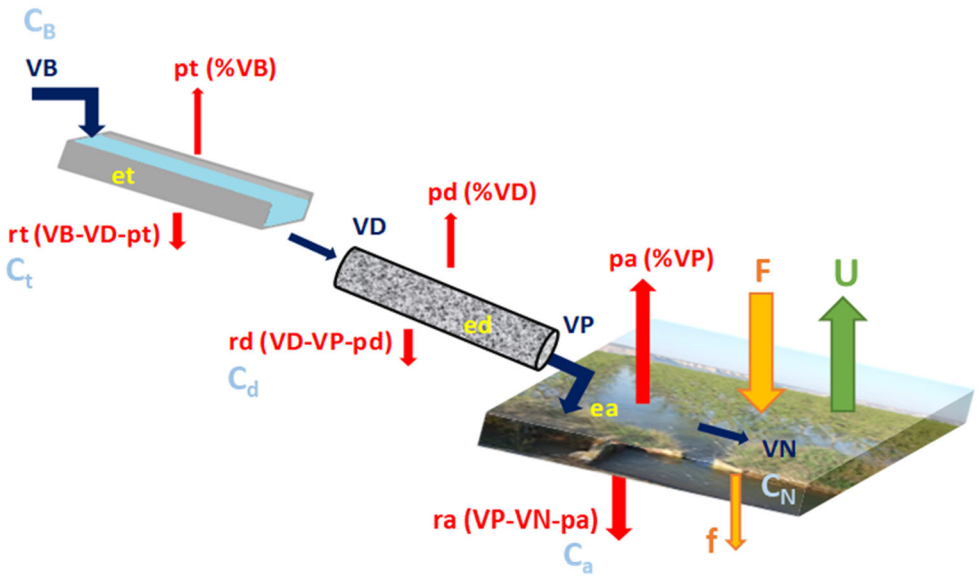


Figure 3.1. Scheme of water and nutrient balances applied to the transport, distribution and irrigation network.

Evaporation water losses are determined experimentally throughout the year (measuring campaigns by the Júcar River Basin Authority) and return flows are calculated as follows:

$$r_t = V_B - V_D - p_t \quad \text{Equation 3.1}$$

$$r_d = V_D - V_P - p_d \quad \text{Equation 3.2}$$

$$r_a = V_P - V_N - p_a \quad \text{Equation 3.3}$$

where V_D ($hm^3 \cdot year^{-1}$) is the irrigation influent water volume to the distribution network, V_P ($hm^3 \cdot year^{-1}$) is the volume applied to cropland and V_N ($hm^3 \cdot year^{-1}$) is the net irrigation volume.

Taking into account the water losses in the water flow sections, an efficiency percentage was obtained for the transport (e_t), for the distribution network (e_d) and for the irrigation method (e_a) (Eqs. 3.4 to 3.6). These efficiency factors were used for all conversions from gross to net irrigation volumes.

$$V_D = V_B \cdot e_t \quad \text{Equation 3.4}$$

$$V_P = V_D \cdot e_d \quad \text{Equation 3.5}$$

$$V_N = V_P \cdot e_a \quad \text{Equation 3.6}$$

3.2.2. Nutrient balance

During the nutrient balance step, the total amount of nutrients in the irrigation water is calculated together with the complementary mineral fertilizers needed to fully provide the crops with the optimal nutrients supply. The balance also evaluates the total amount of nutrients released into water bodies and is applied taking into account that water losses by evaporation give rise to higher nutrient concentrations throughout the water network (Eq. 3.7, Figure 3.1).

$$V_B \cdot C_B + F = r_t \cdot C_t + r_d \cdot C_d + r_a \cdot C_a + f + U \quad \text{Equation 3.7}$$

where C represents nutrient concentration (N or P), F is the fertilizer surplus, r is the return flow. Subscripts B, t, d and a indicate the water flow section in which they are identified, while f considers mineral fertilizer losses and U represents the crop's nutrient uptake.

The following assumptions were made:

- $V_P \cdot C_d + F = U + r_a \cdot C_a + f$: total nutrients provided by net irrigation water ($V_P \cdot C_d$) and mineral fertilizers (F) encompasses the crop's nutrient uptake (U), nutrient losses related to return flow in cropland ($r_a \cdot C_a$) plus mineral fertilizer losses (f).

- $C_N = C_a$: nutrient concentration in the irrigation return flow (r_a) is equal to nutrient concentration in the net irrigation water volume required by the crop (V_N).

Other agronomic considerations, e.g., micronutrients, soil quality, etc., which could change crop nutrient uptakes were outside the scope of this study.

3.2.3. Economic balance

Figure 3.2 shows the different water, nutrient and money flows in a water re-use system to which the economic balance was applied to evaluate the feasibility of AnMBR for fertigation. As proposed by the authors and in line with the new EU regulation proposal (General Secretariat of the Council, 2019), the Reclaimed Water Manager was included. The RWM is the managing body responsible for operating, controlling and supervising the re-use system and would be in charge of drafting and implementing the Water Reuse Risk Management Plan in collaboration with the relevant parties.

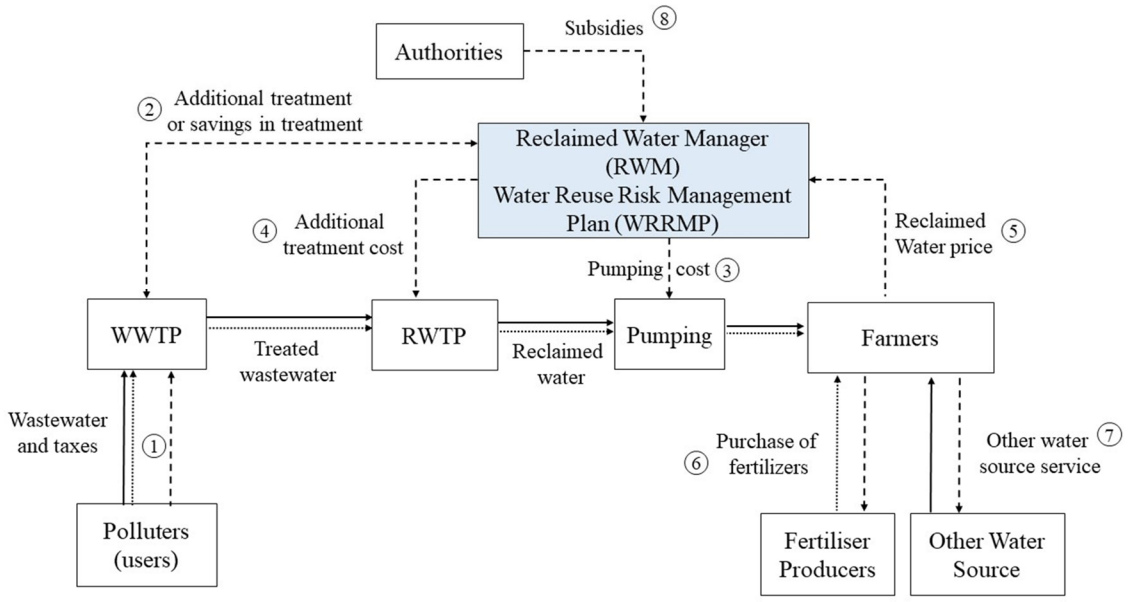


Figure 3.2. Water (full line), nutrients (dotted line) and money flows (dashed line) for an agricultural water reuse system.

The different flows shown in Figure 3.2 are described as follows:

- Nutrient-rich wastewater reaches the WWTP via the sewer system. Based on the "*polluter pays principle*", there is a parallel money (tax/tariff) flow from the users (polluters) to the WWTP.
- Treated water and nutrients enter the Reclaimed Water Treatment Plant (RWTP) for additional treatment (e.g., further filtration, disinfection and/or advanced oxidation processes) to produce good quality reclaimed water.
- The water to be reused is pumped into the irrigation transport/distribution network. Farmers can also use other water sources, such as groundwater and/or surface water, which have to be paid for.
- According to the crop requirements, farmers might need to add mineral industrial fertilizers, which creates a money flow to fertilizer producers. If enough nutrients are present in the reclaimed water, this money flow can be reduced or even disappear.
- The farmers would pay the RWM for controlling the water re-use system and providing enough good-quality water. This opens a new market niche and business opportunity. The RWM would pay the RWTP for the water and nutrients and for using the pumping system. The RWM could also pay the WWTP for any additional treatment needed. In cases where savings can be made in wastewater treatment (WWT) (e.g., avoiding or reducing nutrient removal) or in discharge fees to surface or marine water due to water re-use, the WWTP might pay the RWM.
- The authorities could subsidize the system through the RWM, based on environmental, social and territorial considerations.

The total cost associated with running the WWT and providing reclaimed water for fertigation (C_{Total} , €·year^{-1}) was obtained as follows:

$$C_{Total} = C_{WWT} - C_{Energy\ recovery} + C_{Chemical\ precipitation} + C_{treat_for_reuse} + C_{Pumping} + C_{Fertilizers} + C_{Discharged\ fee}$$

Equation 3.8

where C_{WWT} is the cost of treating wastewater to meet Urban Wastewater Treatment Directive (UWWTD) (91/271/EC) requirements, $C_{Energy\ recovery}$ is the saving associated with the energy recovered as methane during wastewater treatment, $C_{treat_for_reuse}$ is the additional treatment cost of reclaimed water reuse, $C_{Discharge\ fee}$ is the cost of discharging treated wastewater into water bodies, $C_{Pumping}$ is the energy cost for pumping a natural source of water (surface water or groundwater) or reclaimed water and $C_{Fertilizers}$ is the cost of buying fertilizers.

The cost balance of the RWM (B-RWM) was calculated by adding the cost flows in Figure 3.2:

$$\text{B-RWM} = \pm 2\ (€) + 5(€) - 3\ (€) - 4\ (€) + 8\ (€) \qquad \text{Equation 3.9}$$

Where Flow 8 (subsidies) was assumed to be zero. The current price paid by polluters and farmers (current scenario, no water re-use) was kept constant in all the scenarios studied. Polluter prices were calculated according to the individual characteristics of each case, and the farmer's price was calculated as follows:

$$\text{Farmer's price} = C_{Fertilizers} + C_{Pumping} \qquad \text{Equation 3.10}$$

The flow from WWTP to the RWM (Flow 2) was calculated as the difference between the (constant) paid polluter price and the real cost. Savings were made in wastewater treatment or by the absence of chemical precipitation. The flow from farmers to the RWM (Flow 5) was calculated in the same way: as the difference between the farmer's price, which was to remain constant, and the real farming costs. Since the purchase of mineral fertilizer was partially or totally avoided, the RWM could profit from the associated savings.

3.2.3.1. Unit costs and energy consumption

Commercial fertilizer prices and energy consumption in Spain was estimated as shown in Table 3.1, assuming a WWTP energy cost of 0.125 €·kWh^{-1}. Energy consumption of 0.26 kWh·m^{-3} was considered for the CAS system with nutrient removal in the Italian case (value provided by system experts), together with an energy cost of 0.14 €·kWh^{-1}. Costs of 4.35 €·kg$P_{removed}^{-1}$ were considered for the consumption of chemical reagents during P removal (data provided by EPSAR, Spanish public body in charge of WWT in Valencia) and 0.006 €·m^{-3} for chemical membrane cleaning related to AnMBR technology (Pretel et al., 2016b). A pumping energy cost of 0.137 €·kWh^{-1}was considered for a pumping height of 60 m and 9.5 m for groundwater and reclaimed water, respectively.

It should be noted that: (i) the discharge fees applied in Valencia ($C_{Discharged\ fee}$) are on average 0.004 €·m^{-3} and 0.0135 €·m^{-3} for discharging water into coastal and inland water bodies, respectively (data provided by Júcar River Basin Authority and Regional Government of

Valencia). (ii) Italy does not apply discharge fees ($C_{Discharged\ fee}=0$). (iii) C_{WWT} already includes the cost of P chemical precipitation ($C_{Chemical\ precipitation}$) and disinfection ($C_{treat_for_reuse}$) in Peschiera Borromeo, due to regional regulations. (iv) C_{WWT} includes the cost of chemicals for membrane cleaning.

3.2.4. Carbon dioxide emissions

CO_2 assessment included CO_2 emissions related to WWT energy consumption ($CO_{2treatment}$), pumping ($CO_{2pumping}$) and fertilizer production ($CO_{2fertilizers}$) as well as the CO_2 emissions avoided by energy recovered as biogas ($CO_{2biogas}$). The WWT-related CO_2 emissions were based on the energy consumption given in Section 3.2.3.1 considering a European average electricity specific emission factor of 0.2958 kg $CO_2 \cdot kWh^{-1}$ (EEA, 2018). The energy required for fertilizer production was assumed to be 19.3 $kWh \cdot kg\ N^{-1}$ by the Haber-Bosh Process (Perry L. McCarty et al., 2011) and 2.11 $kWh \cdot kg\ P^{-1}$ (Gellings and Parmenter, 2004b)

Total CO_2 emissions ($tCO_2 \cdot year^{-1}$) were calculated as follows:

$$\text{Total } CO_2 \text{ emissions} = CO_{2treatment} + CO_{2pumping} + CO_{2fertilizers} - CO_{2biogas} \quad \text{Equation 3.11}$$

3.2.5. End-users' perception

One of the social bottlenecks in agricultural water reuse identified in the Innovation Deal is the lack of current knowledge on the public perception and social acceptance of the reuse concept. A survey was therefore carried out to collect the end-users' feedback. 15 irrigators from the Mediterranean coast in Spain (Catalonia, Valencia, Alicante and Murcia) and one from Murviel Lès Montpellier in France collaborated in the survey and responded to questions related to crops, water use, water source, water quality, economics of water reuse, water supply problems and their attitude to using reclaimed wastewater for their crops.

Table 3.1. Cost of most usual mineral fertilizers and unitary treatment energy needs and energy reuse of WWT.

Mineral Fertilizer	Cost (€·kg^{-1})
Complex 39N-11P-0K	0.39
Urea 46N-0P-0K	0.31
DURAMON 26N-46P-0K	0.32
Lime superphosphate 0N-20P-0K	0.15
Complex 12N-61P-0K	0.66

	Units	Extended aeration (includes nitrogen removal)	AnMBR (without N.R.)	References
Wastewater Treatment	kWh·m^{-3}	[0.35-0.42]	0.269	EPSAR (based on Valencian Community average) and Pretel et al., 2013
Additional treatment for disinfection	kWh·m^{-3}	0.036	----	EPSAR (based on Valencian Community average)
Energy recovery	kWh·m^{-3}	----	[0.12-0.24]	Pretel et al., 2016a
Pumping from WWTP	kWh·m^{-3}·h^{-1}	0.004	0.004	EPSAR (based on Valencian Community average)

3.2.6. Case studies description

The ID consortium carried out four case studies as part of the tasks involved in the project. This paper presents only the representative cases of Oliva (Spain) and Peschiera-Borromeo (Italy) WWTPs, as they could be replicated in many similar sites in both European countries, where the proposed technology could enhance agriculture sustainability with water reclamation and nutrient recovery and re-use.

3.2.6.1. Oliva WWTP

With a WWT flow (V_T) of 5.5 hm^3·year^{-1} (39,500 population equivalent (p.e.)), Oliva WWTP has an extended aeration (EA) process without P removal by precipitation. The WWTP is in Oliva (Valencia, Spain) and its effluent is discharged into the Mediterranean Sea. It is surrounded by 582 hectares of agricultural land, supplied with 3.4 hm^3·year^{-1} by ground water extraction and flooding irrigation. This land is a non-sensitive area, according to UWWTD, but a vulnerable

zone according to the Nitrates Directive (NiD) (91/676/EC). The polluter price in this case included $C_{Discharged\ fee}$ together with the cost of the treatment.

3.2.6.1.1. Water and nutrients needs

Citrus fruit is the most abundant crop in the study area. Citrus trees need water the whole year round with a net requirement of 3,145 $m^3 \cdot ha^{-1}$, according to the data provided by the Jucar River Basin Authority (CHJ). Citrus N and P needs were considered as follows: 260 kg $N \cdot ha^{-1} \cdot year^{-1}$ and 80 kg $P \cdot ha^{-1} \cdot year^{-1}$ for flood irrigation and 220 kg $N \cdot ha^{-1} \cdot year^{-1}$ and 70 kg $P \cdot ha^{-1} \cdot year^{-1}$ for drip irrigation (López Bellido et al., 2010; Valencian Ministry of Agriculture; Environment; Climate Change and Rural Development, 2018).

3.2.6.1.2. Analyzed scenarios

The scenarios studied for Oliva WWTP are shown in Figure 3.3. SI (a) represents the current situation, in which all the treated wastewater is discharged into the environment, while all the water needed for irrigation is extracted from the ground. SI includes pumping costs as well as the continuous impact on the availability of fresh water.

EA energy consumption is in the range of 0.35-0.42 $kWh \cdot m^{-3}$ (Table 3.1). In the case of the Oliva WWTP, a value of 0.375 $kWh \cdot m^{-3}$ was considered due to the treatment flow and the extent of N removal. Although SI meets the UWWTD requirements, it is still far from complying with the CE objectives, since water, energy and nutrients are not re-used. Fertigation could be used for agricultural water and nutrient demands, as shown in Scenarios II, III and IV (Figure 3.3), in which no groundwater is extracted, with consequent savings. SII includes fertigation after wastewater treatment, with reclaimed water that contains concentrations of N and P of 15 and 6 $mg \cdot L^{-1}$, respectively. In SIII it is assumed that 100 % of WWTP inflow is treated with AnMBR technology instead of EA, providing a nutrient-rich effluent with concentrations similar to the usual values in WWTPs influent (50 mg $N \cdot L^{-1}$ and 8 mg $P \cdot L^{-1}$ according to EPSAR) and biogas generated by anaerobic digestion of organic matter in wastewater. Although the Oliva WWTP discharges into a non-sensitive area, an SIV was evaluated (Figure 3.3) in which wastewater is treated by two parallel lines (AnMBR and EA) to reduce the total nutrients released into the environment.

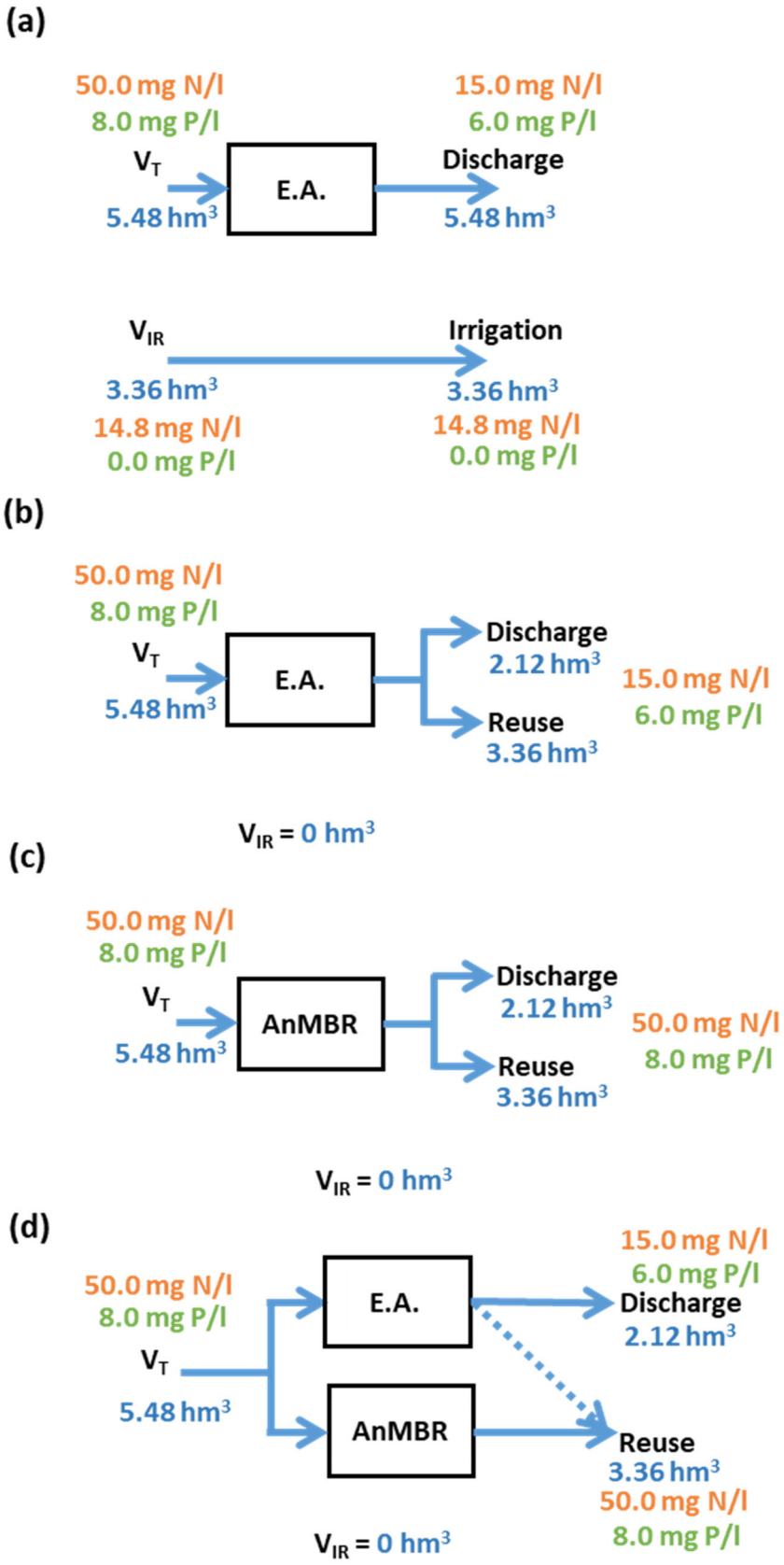


Figure 3.3. Scenarios in Oliva WWTP case study: (a) SI, (b) SII, (c) SIII and (d) SIV.

3.2.6.2. Peschiera Borromeo WWTP

The Peschiera Borromeo WWTP is currently treating an average inflow rate of 78.84 $hm^3 \cdot year^{-1}$ (566000 p.e.) by two water lines that receive wastewater from the Milan metropolitan area. Line 1 consists of a CAS process followed by tertiary treatment (disinfection with peracetic acid) while line 2 consists of an upflow biological filtration for nutrient removal and UV disinfection. This WWTP is situated in the municipality of Peschiera Borromeo (Lombardy, Italy) and discharged into the Lambro River, which was declared a sensitive area by the UWWTD and a vulnerable zone by the NiD. It should be noted that the Italian regulations set the total nitrogen (N_T) and total phosphorus (P_T) standard for water re-use at 15 and 2 $mg \cdot L^{-1}$, respectively, so that the WWTPs must remove the nutrients before water reclamation. This is one of the major economic barriers to possible fertigation and in this case study the polluter price includes the cost of chemical precipitation and disinfection.

3.2.6.2.1. Water and nutrients needs

The surrounding agricultural land which could potentially be irrigated with reclaimed water has an extension of 1,500 ha and its water demand is 12.03 $hm^3 \cdot year^{-1}$. This demand could be covered either by surface water or by a flow rate from the WWTP's water line 2. This plant is in a peri-urban agricultural park (Parco Agricolo Sud Milano) whose main crop is tomatoes. The average water requirements are 7.318 $m^3 \cdot ha^{-1}$ from April to September (183 days). Tomato N and P needs for drip irrigation systems are 160 kg $N \cdot ha^{-1} \cdot year^{-1}$ and 20 kg $P \cdot ha^{-1} \cdot year^{-1}$ (Valencian Ministry of Agriculture; Environment; Climate Change and Rural Development, 2018).

3.2.6.2.2. Analyzed scenarios

The scenarios studied in the Peschiera Borromeo WWTP are depicted in Figure 3.4. SI represents the current situation, in which all the treated wastewater is discharged into the environment, while all the irrigation water is extracted from a fresh water source, affecting both the resource and the surrounding ecosystem. Although a large amount of energy is required for WWT, SI is the common situation in many parts of Italy due to the restrictive legislation, long distances, unfavorable slope between the WWTPs and irrigation areas, and the high monitoring and distribution costs (Ventura et al., 2019).

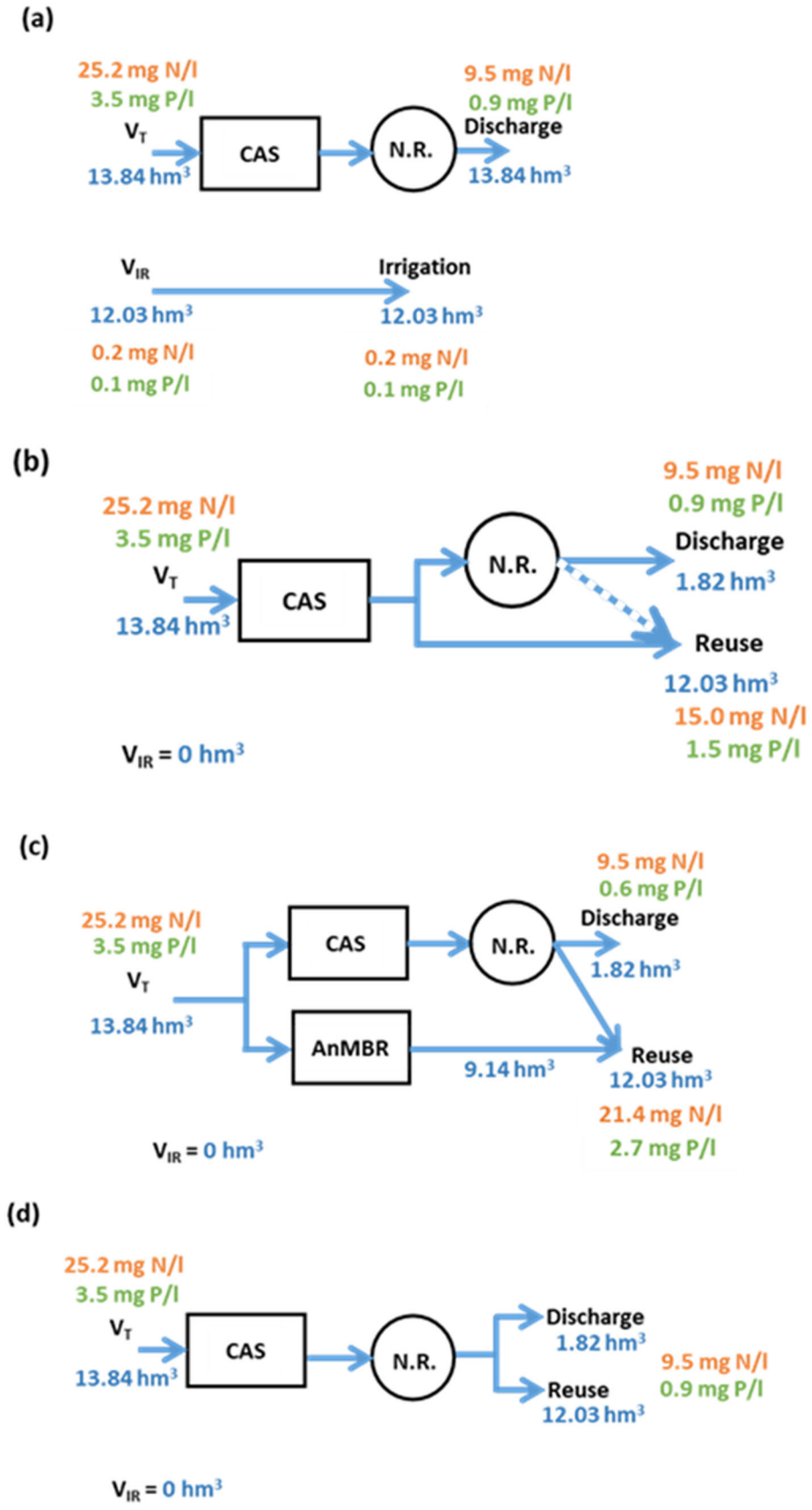


Figure 3.4. Scenarios in Peschiera Borromeo WWTP case study: (a) SI, (b) SII, (c) SIII, (d) SIV.

In SII a fraction of water flow treated in the CAS process bypasses the nutrient removal process and is used for fertigation. This treatment scheme means that the bypass flow can be modified or even cancelled according to the demand for nutrients. The possibility of mixing treated water is also included (dashed line). SIII is proposed with the aim of maximizing energy and nutrient recovery, complying with CE principles. In this scenario, a fraction of the influent is treated by AnMBR, which could reduce the total WWT energy consumption through biogas production, while generating a nutrient-rich effluent that would notably reduce fertilizer costs. In this scenario and in SII, a modification of the Italian regulations is assumed to allow either land application as a wastewater post-treatment, or the use of reclaimed water as a fertilizer product.

In SIV all wastewater is treated through a CAS system with nutrient removal and water is re-used for agriculture. SIV represents the current situation of some Italian WWTPs, such as Milano San Rocco and Milano Nosedo, where farmers supply part of the effluent for irrigation together with their own artificial fertilizers.

3.3. Results and discussion

This section presents and discusses the main results obtained for the two case studies analyzed.

3.3.1. Efficiency factors

Preliminary studies (not shown) allowed for the calculation of efficiency factors for conversion between gross and net irrigation volumes, which were applied in this study and are shown in Table 3.2. Peschiera Borromeo is more efficient since the distribution network consists of pipelines and the irrigation method used is the drip system. In the case of Oliva, the pipeline distribution and drip irrigation systems were included as an optimized version of each scenario, assuming transport, distribution and application efficiencies of 95 %, 97 % and 97 %, respectively.

Table 3.2. Return flows and supplied water efficiency for both Oliva and Peschiera Borromeo case studies.

Irrigation system	(%)	Oliva[(1)]	Peschiera Borromeo[(2)]
Transport network	Water loss by evaporation (p_t)	0.3	0.0
	Water loss through return flow (r_t)	14.7	3.0
	Efficiency (e_t)	85.0	97.0
Distribution network	Water loss by evaporation (p_d)	0.0	0.0
	Water loss through return flow (r_d)	15.0	3.0
	Efficiency (e_d)	85.0	97.0
Irrigation method	Water loss by evaporation (p_a)	8.1	2.0
	Water loss through return flow (r_a)	16.5	1.0
	Efficiency (e_a)	75.4	97.0

(1) Percentages calculated from the data supplied by Júcar River Basin Authority.
(2) Percentages calculated for a pipeline network.

3.3.2. Oliva WWTP

3.3.2.1. Water balance

The current situation is represented by SI. In this scenario, 3.36 $hm^3 \cdot y^{-1}$ of groundwater is used for irrigation (Figure 3.3), which intensifies natural resource depletion. This consumption can be avoided by using reclaimed water for irrigation, which would improve the conservation of water bodies. In Scenarios II and III, 61.3 % of the wastewater treated in the Oliva WWTP is reused.

A total supplied water efficiency of 54.5 % was calculated (Table 3.2), showing that there are high water losses throughout the irrigation network due to the return flows and the irrigation method used (flooding). The application of an optimized irrigation system (pipeline irrigation network and drip feed) would significantly reduce water losses from 1.32 $hm^3 \cdot year^{-1}$ to 0.18 $hm^3 \cdot year^{-1}$ with the consequent increase in network efficiency.

The results show that irrigation system optimization is a key factor in minimizing water losses. As expected, the water balance calculated for the four scenarios in Oliva WWTP with an optimized irrigation network showed a smaller water re-use flow of 2.05 $hm^3 \cdot year^{-1}$.

3.3.2.2. Nutrient balance

Used as the baseline, SI discharges treated wastewater into coastal waters, releasing nutrients into the environment. In this scenario, the total crop nutrient demand is met by mineral fertilization.

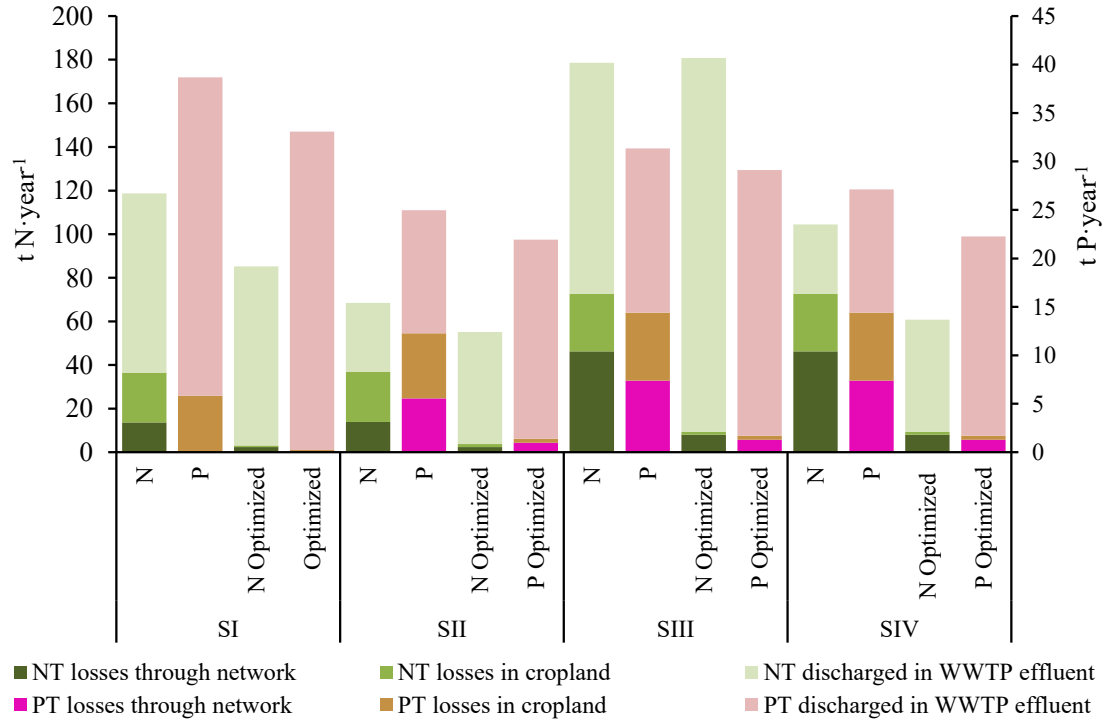
The smaller discharge flow in SII reduces nutrient losses to the environment: 42.2 % less N and 35.4 % less P (Figure 3.5). At the same time, SII recovers the P contained in the reclaimed wastewater: 20.6 kg $P \cdot ha^{-1} \cdot year^{-1}$, reducing mineral P addition by 29.4 % (Figure 3.5a).

The application of AnMBR technology in SIII reduces P losses by 18.9 % (SIII compared to SI, Figure 3.5a) while N discharged into the environment increases from 118.7 $t \cdot year^{-1}$ in SI to 178.6 $t \cdot year^{-1}$ in SIII. The reason behind this result lies in the fact that the N content in the AnMBR effluent is higher (from 15 to 50 $mg \cdot L^{-1}$) than for P concentration (from 6 to 8 $mg \cdot L^{-1}$,Figure 3.3), so that the reduction in discharged flow in SIII compensates for the increase in the effluent's P concentration, whereas in the case of N its higher AnMBR effluent concentration causes higher N losses, even though the discharge flow is smaller. In any case, SIII can reduce the N and P added by mineral fertilizers by up to 71.6 % and 39.2 %, respectively (Figure 3.5a). SIV has the same added mineral needs as SIII and reduces nutrient losses with respect to Scenarios I and III.

It should be noted that the application of an optimized irrigation system under all circumstances not only reduces water losses, but also nutrient losses and therefore the amount of N and P released through the network was lower in the four optimized scenarios (Figure 3.5). This entails an extra environmental benefit for groundwater bodies.

3.3.2.3. Economic balance

The results obtained from the economic analysis are shown in Table 3.3. The operating costs of AnMBR wastewater treatment are 63.4 % lower in SIII and 39 % lower in SIV. The coastal body discharge fee in Scenarios II to IV is reduced in proportion to the amount of water reclaimed. Disinfection costs only apply in SII, since membranes are used in Scenarios III and IV. A pumping cost is required in all the scenarios, but groundwater extraction pumping costs disappear in SII to SIV. Fertilizer costs are also reduced due to fertigation, especially when the reclaimed wastewater is obtained from AnMBR.



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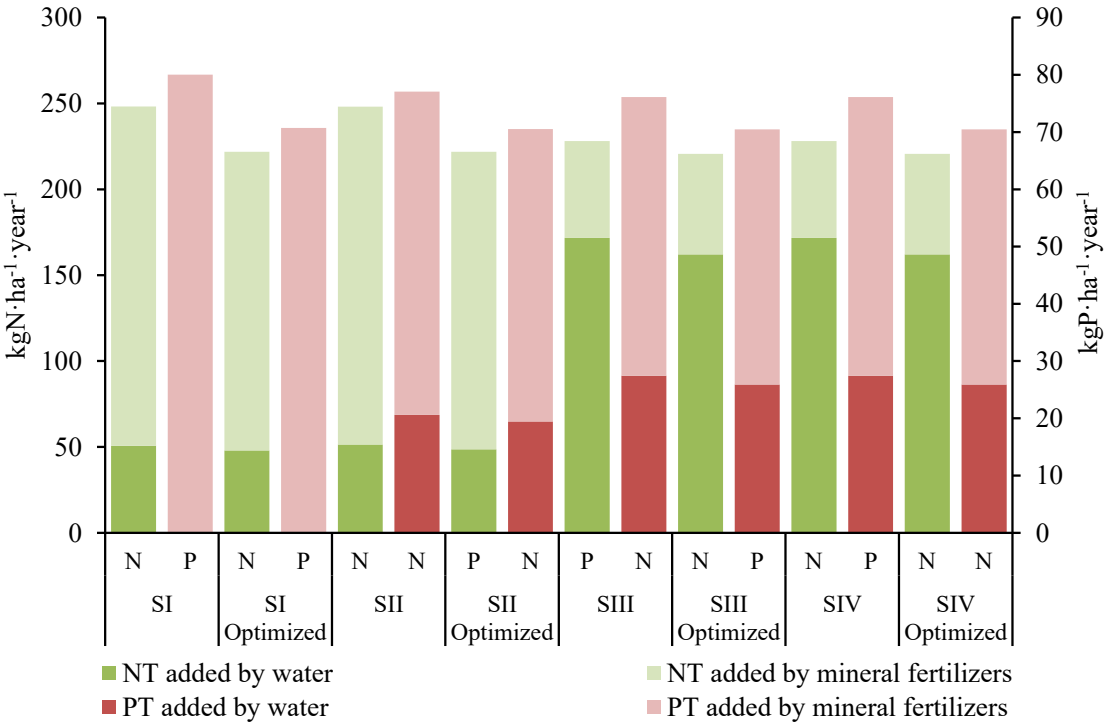

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Figure 3.5. a) Nutrients discharged into the environment in the four scenarios proposed in Oliva WWTP; b) Results of nutrient balance applied to cropland in the four scenarios proposed in Oliva WWTP.

As explained above, the authors propose to maintain the total cost for polluters and farmers in all cases equal to that of SI (278.9 and 202.0 k€·year^{-1}, respectively). A flow from the WWTP to the RWM would be possible in the water re-use scenarios, since WWTP cost would actually be smaller due to lower discharge fees and C_{WWT}. Similarly, due to savings in fertilizers and groundwater pumping, part of the farmers' expenses could be transferred to the RWM, who would have a total income of 0.029 (SII), 0.099 (SIII) and 0.080 (SIV) €·m$^{-3}_{reused}$ (Table 3.3).

The results show the economic viability of AnMBR technology, since the total cost was reduced from 480.9 k€·year^{-1} (SI) to 149.4 k€·year^{-1} (SIII) or 212.3 k€·year^{-1} (SIV). At the same time, Scenarios III and IV do not only meet the UWWTD and NiD requirements, but also reduce the costs associated with fertilizers and pumping. This means that the RWM budget can be devoted to covering the reclaimed water management expenses (including the WRRMP).

Table 3.3. Economic results from the four scenarios studied in Oliva WWTP.

Costs	Units	SI	SI opt	SII	SII opt	SIII	SIII opt	SIV	SIV opt
Wastewater treatment C_{WWT}	k€·year^{-1}	257.0	257.0	257.0	257.0	93.9	93.9	156.8	195.7
Discharge fee $C_{Discharged\ fee}$	k€·year^{-1}	21.9	21.9	8.5	13.7	8.5	13.7	8.5	13.7
WWTP cost	k€·year^{-1}	*278.9*	*278.9*	*265.5*	*270.7*	*102.4*	*107.6*	*165.3*	*209.4*
Extra treatment for reuse $C_{rteat_for_reuse}$ (4)	k€·year^{-1}	0.0	0.0	15.0	9.0	0.0	0.0	0.0	0.0
Pumping for reuse $C_{Pumping-WWT}$ (3)	k€·year^{-1}	0.0	0.0	16.0	9.7	16.0	9.7	16.0	9.7
WWTP + RWTP cost	k€·year^{-1}	*278.9*	*278.9*	*296.5*	*289.4*	*118.4*	*117.3*	*181.3*	*219.1*
Fertilisers $C_{Fertilizers}$	k€·year^{-1}	92.0	81.1	87.0	77.2	31.0	31.1	31.0	31.1
Pumping groundwater $C_{Pumping}$	k€·year^{-1}	110.0	67.3	0.0	0.0	0.0	0.0	0.0	0.0
Farming cost	k€·year^{-1}	*202.0*	*148.4*	*87.0*	*77.2*	*31.0*	*31.1*	*31.0*	*31.1*
Total C_{Total}	k€·year^{-1}	**480.9**	**427.3**	**383.5**	**366.6**	**149.4**	**148.4**	**212.3**	**250.2**
Flow WWTP to manager (2)	k€·year^{-1}	0.0	0.0	13.4	8.2	176.6	171.3	113.7	69.5
Flow farmer to manager (5)	k€·year^{-1}	0.0	0.0	115.0	71.2	171.0	117.4	171.0	117.4
RWM Balance	k€·year^{-1}	0.0	0.0	97.4	60.5	331.6	279.0	268.7	177.1

The economic balance for the four scenarios shows that, while the wastewater treatment costs with and without optimized irrigation networks remain constant for Scenarios I to III, discharge fees increase and the available money flow from the WWTP to the RWM is reduced. The same wastewater flow needs to be treated per year, but a higher flow of treated water is discharged since

a smaller water flow is reclaimed. On the other hand, as the reclaimed water flow is only 61 % of that in the non-optimized scenarios, disinfection and pumping costs and mineral fertilization need to be reduced (Table 3.3). Water re-use costs are therefore reduced due to the smaller water flow needed by a more efficient system.

In SI, where treated water is not reclaimed, groundwater pumping costs also decreased for the same reason. A lower farmer's price was thus obtained for the optimized scenarios (148.4 k€·year^{-1}) and the following total RWM income reduction was calculated: 37.7 % in SII, 15.9 % in SIII and 34.1 % in SIV. The reasons for this are the downscaled re-use system, while wastewater treatment costs remain the same (except for SIV, where two different technologies with two different costs are involved), together with the decrease in the farmer's price due to the lower cost of optimized SI.

An optimized irrigation system saves water ready for re-use and reduces its cost, with a lower RWM budget. However, certain strategies could be applied to increase the RWM economic balance, including subsidies, changes in discharge fees, extension of the irrigated area, etc.

3.3.2.4. Carbon dioxide emissions

Carbon dioxide emissions are associated with the energy cost of mineral fertilizer production, wastewater treatment, disinfection, pumping and energy recovered as biogas. Water re-use by conventional treatment technologies (SII) would reduce emissions by between 6.0 and 15.2 % of CO_2, depending on whether optimized or non-optimized scenarios are considered. AnMBR technology (SIII) would provide the biggest environmental improvement in terms of CO_2 emissions, since it would reduce them by between 74.7 % and 75.3 % CO_2 as a consequence of the high N content in the reclaimed water and the biogas generated. An improvement is also observed when comparing Scenarios II to IV, with reclaimed water, to SI, where no water is reclaimed.

As expected, the optimized Scenarios I to III show an improvement in CO_2 emissions, due to reduced fertilizer use, pumping and disinfection (when applied) (Figure 3.6). The greatest improvement, when optimized, is obtained for SI (11 %). In SIV, given the fact that the amount of reused nutrients is equal to that in SIII, the CO_2 emissions associated with fertilizer production are reduced by the same proportion. The high energy consumption in the EA process and the

absence of energy recovery in this treatment produce higher CO_2 emissions than SIII. This scenario, in which AnMBR is combined with an optimized irrigation system, provided the highest environmental benefit in terms of total CO_2 emissions (reduction of up to 1153 t $CO_2 \cdot year^{-1}$).

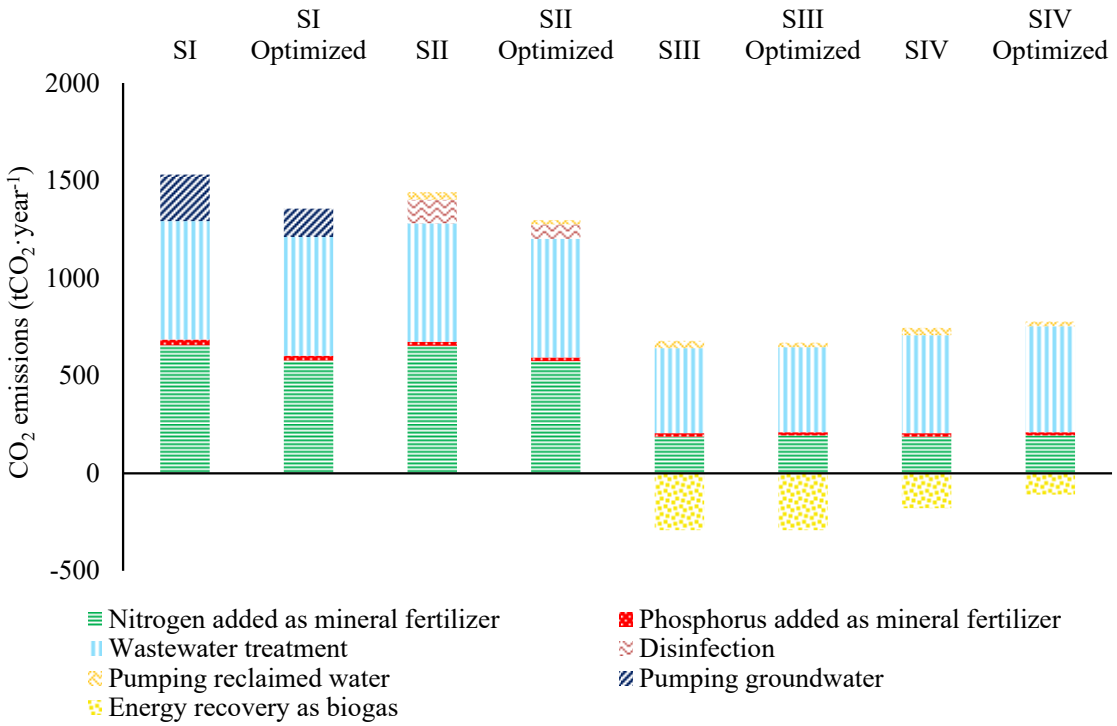


Figure 3.6. CO_2 emissions for the four scenarios studied in Oliva WWTP.

The study carried out in Oliva WWTP (Spain) showed that in this case AnMBR is suitable for the combined purpose of wastewater treatment and fertigation, since it maximizes energy and nutrient recovery and reduces both costs and CO_2 emissions.

The positive impact of AnMBR would be still higher if groundwater conservation could be quantified. Applying this technology in vulnerable zones requires risk assessment plans, including nutrient balances. Methods such as drip irrigation are recommended, since they generate smaller return flows than flood irrigation.

3.3.3. Peschiera Borromeo WWTP

3.3.3.1. Water balance

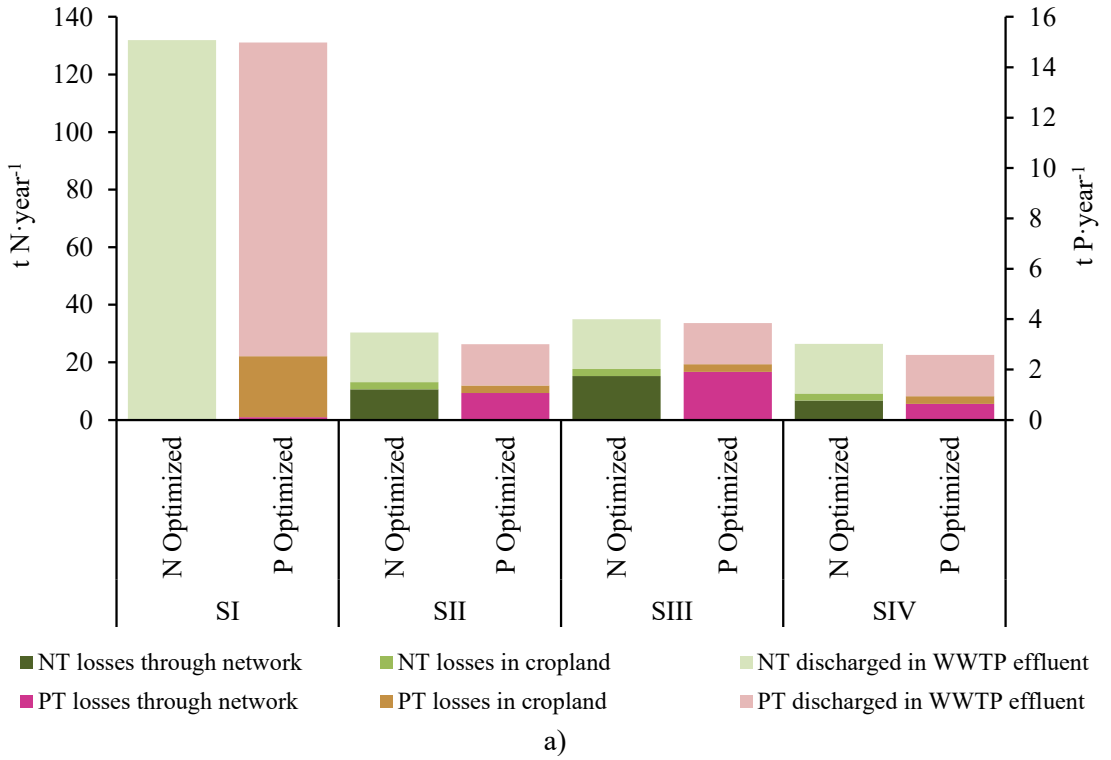
In the present situation (SI) 12.03 $hm^3 \cdot year^{-1}$ of fresh water is used for irrigation (Figure 3.4), depleting natural resources. Fertigation (Scenarios II and III) or irrigation (SIV) would improve the conservation of water bodies. In these scenarios, 87 % of the wastewater treated in the Peschiera Borromeo WWTP would be re-used. The irrigation network in the surrounding area was designed with pipelines and the present irrigation method is by drip, so that the results are those of an optimized irrigation system.

3.3.3.2. Nutrient balance

SI is the situation in which the highest amount of nutrients is discharged into the environment (Figure 3.7a), which was expected in a situation that does not re-use treated wastewater. Scenarios II to IV show the expected lower nutrient discharge into the environment, with a 7.6 times smaller discharged water flow in all cases. Total N losses fall by 77.0 % (SII), 73.5 % (SIII) and 79.9 % (SIV) with respect to SI. P losses are 79.9 %, 74.3 % and 82.7 % less for Scenarios II, III and IV, respectively and with respect to SI. The biggest difference between the scenarios lies in the nutrient losses through the network, which are greater in SIII, with the highest nutrient content of reclaimed water.

In SI almost all the required nutrients are supplied by mineral fertilizers, whereas higher percentages of nutrients are provided by the reclaimed water in Scenarios II, III and IV (70 %, 100 % and 44 %, respectively) (Figure 3.7b). SIII illustrates the flexibility that AnMBR technology gives to the system. In fact, water quality can be modified in terms of nutrient content, by mixing the reclaimed water with the effluent from the nutrient removal process, in such a way that no mineral fertilizers are needed.

Since there is no clear advantage regarding nutrient losses for any of the three water re-use scenarios, the economic and CO_2 emissions analyses become decisive.



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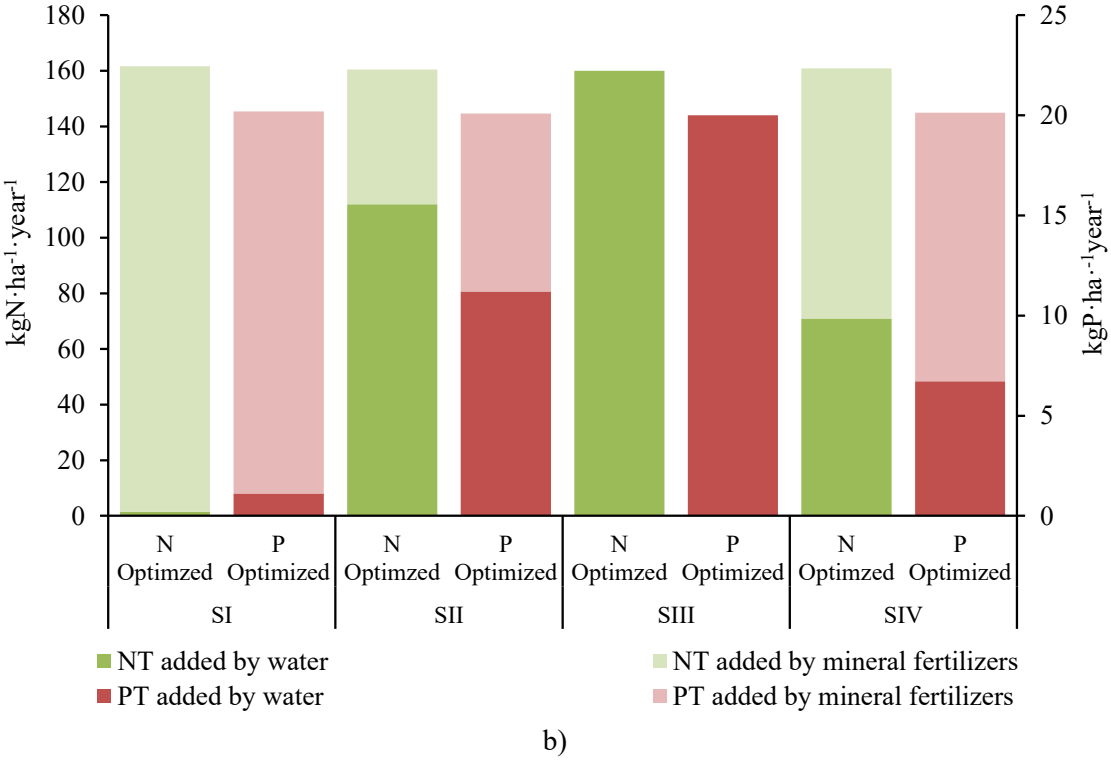

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Figure 3.7. a) Nutrients discharged into the environment in the four scenarios studied in Peschiera Borromeo WWTP; b) Nutrients balance applied to cropland in the four scenarios studied in Peschiera Borromeo WWTP.

3.3.3.3. Economic balance

Table 3.4 shows the results of the economic analysis. Disinfection and nutrient removal are considered part of the wastewater treatment cost. In fact, they are not an additional treatment because, according to the Italian regulations, nutrients must be removed from the final effluent up to the allowed quality standard. This cost in SI (512.9 k€·year^{-1}) was established as the price polluters pay in all scenarios. Treatment costs are lower in Scenarios II and III than in SI (6.1 % and 40.1 %, respectively). Since irrigation water has to be pumped either from surface water bodies or from WWTP effluent, all water scenarios include water pumping costs. Fertilizer costs decrease in the situations with water re-use, being zero when AnMBR technology is applied, which brings considerable savings in farming costs. Water re-use scenarios provide a relevant money flow from farmers to the RWM, especially in Scenarios II and III, due to the higher contribution of the WWTP effluent to fertilization needs.

Table 3.4. Economic results of the four scenarios studied in Peschiera Borromeo WWTP.

Costs	Units	SI opt	SII opt	SIII opt	SIV opt
Wastewater treatment C_{WWT}	k€·year^{-1}	407.0	407.0	261.0	407.0
Disinfection $C_{Disinfection}$	k€·year^{-1}	69.8	69.8	23.7	69.8
Chemical precipitation	k€·year^{-1}	36.1	4.7	22.6	36.1
WWTP cost	k€·year^{-1}	*512.9*	*481.5*	*307.3*	*512.9*
Pumping for reuse $C_{Pumping-WWT}$	k€·year^{-1}	0.0	80.8	80.8	80.8
RWTP cost	k€·year^{-1}	*512.9*	*562.3*	*388.1*	*593.7*
Fertilisers $C_{Fertilizers}$	k€·year^{-1}	170.8	53.2	0.0	97.3
Pumping surface water $C_{Pumping}$	k€·year^{-1}	80.8	0.0	0.0	0.0
Farming cost	k€·year^{-1}	*251.6*	*53.2*	*0.0*	*97.3*
Total C_{Total}	k€·year^{-1}	**764.5**	**615.5**	**388.1**	**691.0**
Flow WWTP to manager	k€·year^{-1}	0.0	31.4	205.6	0.0
Flow farmer to manager	k€·year^{-1}	0.0	198.5	251.7	154.4
RWM Balance	k€·year^{-1}	0.0	149.0	376.4	73.6

Although SIV gives rise to the lowest nutrient losses to the environment (see Figure 3.7a), it results in the highest total cost of all re-use scenarios. This is a direct consequence of removing all the nutrients from the influent, which means a higher quantity of mineral fertilizers is needed. This, in turn, results in fewer economic resources available for the RWM and shows that the

current situation in some Italian WWTPs is not the optimal. AnMBR technology (SIII) could reduce the total cost of the water re-use system to 388.1 k€·year^{-1} due to the following factors: (i) there is no need for mineral fertilizers and (ii) treatment cost is 40.1 % lower than SI, because of the lower energy consumption of the biological process and the reduced disinfection and chemical precipitation costs.

3.3.3.4. Carbon dioxide emissions

The highest CO_2 emissions were those of the current SI, followed by SIV, which represents the most frequently adopted alternative in the area (see Figure 3.8). Water re-use with CAS treatment (SII) reduced CO_2 emissions by 46.4 % with respect to the present situation, while the smallest carbon footprint was obtained for SIII, with AnMBR technology (-898.9 tCO_2·year^{-1}). On the one hand, the possible energy recovery as biogas is substantially higher with this wastewater treatment than with CAS, while on the other the avoidance of mineral fertilizers also prevented a major CO_2 emissions source. These results show that the AnMBR/fertigation combination can considerably reduce the carbon footprint and the financial costs in comparison with conventional treatments such as the CAS system, provided that a high percentage of the influent can be treated by AnMBR (in this case 86.8 %) and application to farmland can be considered as a post-treatment.

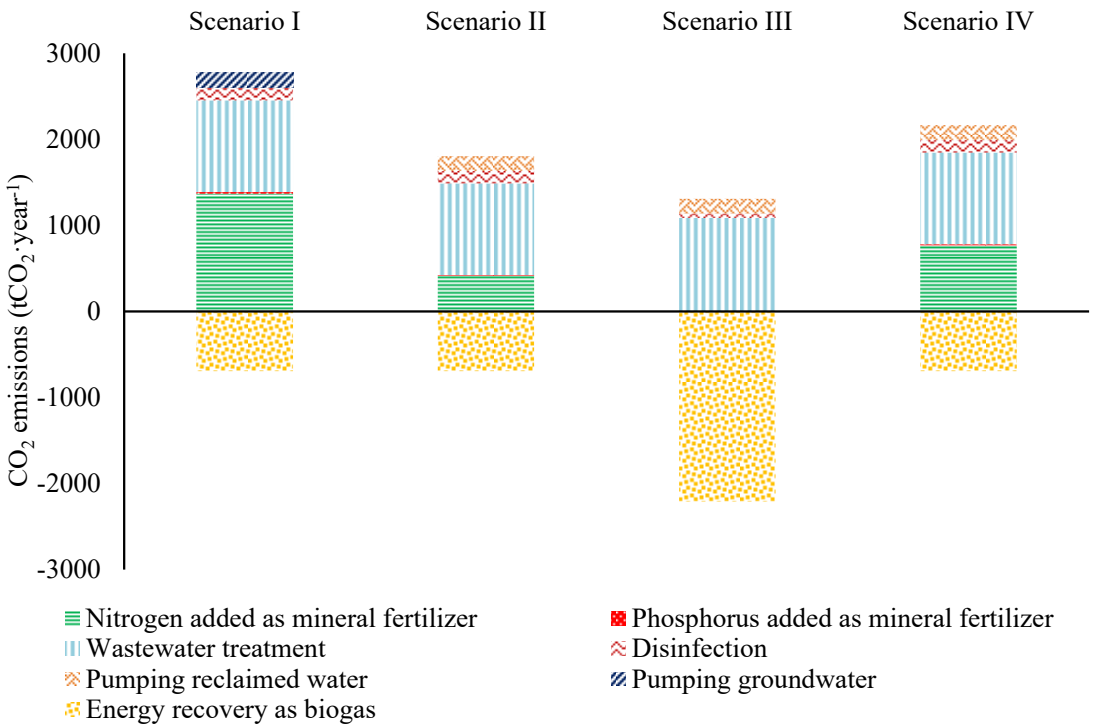

Figure 3.8. CO_2 emissions for the four scenarios studied in Peschiera Borromeo WWTP.

3.3.4. End-user acceptance

The survey was disseminated among irrigators whose average annual water usage is 5000 m^3/ha. The responses revealed that 87.5% of those surveyed have at some point if not currently irrigated their crops with reclaimed water and 64% of these cases declared that they achieved an economic benefit in doing so. The study showed that scarcity problems are more frequent or are more important to the farmers surveyed than water quality problems. Finally, one of the main results of this study was that all (100%) the irrigators surveyed claimed that they would be willing to use water from a wastewater treatment plant if they could be sure it is of high quality, which is consistent with their acknowledgement that reclaimed water guarantees the water supply.

3.4. Conclusions

Reclaimed water is a reliable resource that can be associated with a relevant renewable nutrient supply. It is independent of seasonal droughts and weather variations and thus acts as an alternative resource to alleviate pressure on freshwater sources, while reducing the economic and environmental costs of mineral fertilizers. However, a risk management plan is needed to ensure environmental and health safety when reclaimed water is used for both irrigation and fertigation.

This work has shown the positive economic balance that can be obtained in different water reuse scenarios. The authors propose that savings can be used for drawing up, updating and implementing the Water Re-use Risk Management Plans under the responsibility of the new *Reclaimed Water Manager*.

Since AnMBR technology can also recover energy from the wastewater flow, its application presents an optimal scenario according to Circular Economy principles and environmental sustainability. In this regard, the combination of AnMBR technology and fertigation can significantly reduce CO_2 emissions.

Lastly, in order to comply with the current legislation, future combined AnMBR and fertigation demonstration projects need to be carried out in non-sensitive areas, following the ID conclusions. These trials should include environmental and health risk assessments, agronomic studies and social and economic analyses to obtain comprehensive information of the proposed water management scheme.

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4. Ultrafiltration after primary settler to enhance organic carbon valorization: energy, economic and environmental assessment

Abstract

A membrane-based alternative consisting of introducing an ultrafiltration step after the primary settler of the Carraixet WWTP (Valencia, Spain) is analyzed. This alternative aim at increasing the organic carbon valorization in the anaerobic digestion and reducing the aeration requirements in the biological reactor. A comparison is made between the current WWTP and the membrane-based alternative based on their energy balances and applying the life cycle costing and life cycle assessment methodologies. Assessment of the membrane-based alternative shows a reduction in the energy demand of 0.053 kWh per kg of COD removed due to a decrease of the aeration needs and a higher methane production in the anaerobic digestion. However, a bypass stream of 25 % to the reactor is needed to guarantee enough COD for a proper nitrogen removal to accomplish with discharge limits. The improvement in the energy balance allowed a reduction in OPEX of €0.013 per kg of COD removed, while the implementation of the membrane system represents CAPEX of €0.105 per kg COD removed. Finally, membrane-based alternative shows a better environmental performance in all the categories evaluated with ReCiPe 2016. Specifically, lower N_2O emissions allowed a reduction of 0.3 kg CO_{2eq} per kg of COD removed.

Keywords

UF membranes; organic matter valorization; LCC; LCA

Highlights

The performance of a WWTP with an UF process after primary settler was evaluated.
Energy recovery and aeration reduction allowed savings of 0.053 kWh·kg COD_{rem}^{-1}.
OPEX was reduced by €0.013 kg COD_{rem}^{-1}, but CAPEX of €0.105 kg COD_{rem}^{-1} is needed.
LCA outcome showed enhanced environmental performance in all impact categories.
More favorable financial framework is required to leverage environmental benefits.

A. Jiménez-Benítez, P. Sanchís-Perucho, J. Godifredo, J. Serralta, R. Barat, Á. Robles, A. Seco. Ultrafiltration after primary settler to enhance organic carbon valorization: energy, economic and environmental assessment. *Submitted to Journal of Water Process Engineering on Oct 03, 2023.*

4.1. Introduction

The ever-increasing demand for water also implies an increase in the production of wastewater and greater pressure on the treatment systems. It is thus relevant to explore and assess innovative solutions to enhance the sustainability of the whole system by ensuring adequate treatment capacity and improving the economic and environmental performance of wastewater treatment plants (WWTP) (Hernández-Chover et al., 2023).

Conventional WWTPs based on aerobic processes (e.g., activated sludge) have been shown to be effective in removing traditional pollutants (organic matter and nutrients). However, the high energy consumption derived mainly from aerating the biological reactor is an important drawback (Lee et al., 2017). Improving the efficiency of these facilities necessarily involves an organic carbon redirection to 1) reduce aeration needs and 2) enhance the energy balance.

Boosting anaerobic digestion (AD) in the sludge line over the aerobic biological reactor in the water line appears as a feasible option to meet these objectives. Its main advantages are (Robles et al., 2022): i) lower sludge production due to the low anaerobic growth yield; ii) no aeration is required; iii) organic matter can be valorized through methane production; iv) reduction of net CO_2 emissions by means of energy recovery; and iv) increased nitrogen recovery potential in the sludge line. Reducing the organic load treated in the aerobic reactor would also lower the aeration requirements and would diminish N_2O emissions by reducing the nitrification-denitrification process (Kampschreur et al., 2009). However, as the capacity of primary settlers to capture particulate organic matter is limited, additional processes should be introduced in the water line to drive all the particulates towards AD. For this purpose, membrane technology has shown a promising performance, since it controlled pore size guarantees a selective suspended fraction retention (Foglia et al., 2019). Implementing membranes as the primary treatment before the biological reactor, known as direct membrane filtration (DMF), thus appears as a potential option for retrofitting existing WWTPs to maximize the recovery of organic matter and use it in AD.

Initially, most DMF studies focused on using membranes after primary settling to address the role of membrane fouling on the economics of the process. Indeed, severe fouling propensities have usually been reported (see, for instance, Kimura et al. (2017); Jin et al. (2015); and Gong et al. (2015)), much more so than in other aerobic/anaerobic membrane bioreactors (Jin et al., 2016; Nascimento et al., 2017). However, this problem can be controlled by applying effective anti-fouling strategies. For example, Abdessemed & Nezzal (2003) used ultrafiltration (UF)

membranes and coagulation by adding ferric chloride and adsorption with activated carbon, achieving a chemical oxygen demand (COD) effluent concentration below 15 $mg \cdot L^{-1}$ under the best operating conditions. These authors highlighted the improved membrane performance after coagulation in terms of fouling propensity and treatment capacity. The studies by Sethi & Juby, (2002) are one of the few references to long-term evaluation (> 1 year) of microfiltration (MF) technology in a pilot plant in which the primary clarifier supernatant was treated to achieve disinfected solid-free water. These authors showed it was possible to achieve long-term stable filtration without irreversible fouling. More recently, Hube et al. (2021) assessed fouling and mitigation mechanisms of cross-flow MF and UF membranes for DMF of the supernatant from the primary settler of a municipal WWTP. The authors reported pore blocking and the formation of a cake layer during filtration in both MF and UF membranes. Although short-term physical cleaning was able to remove the cake-layer from UF membrane, its effectiveness was gradually reduced in MF membranes due to the residual particulates remaining inside the pores. The MF performance was improved by increasing the downtime for physical cleaning (i.e., shortening the length of the filtration stage), but it affected the efficiency of the process. Similar results were reported by Sanchis-perucho et al. (2023), who also concluded that MF membranes are not recommended when directly filtering municipal wastewater (MWW). In this latter study, UF and MF membranes were operated in treating both raw MWW and the primary settler supernatant and it was determined that the MF membrane suffer more severe fouling than UF membranes due to the former's more severe pore blockage propensity. These authors also found that the DMF of MWW is possible in the middle/long-term with UF membranes, achieving operating periods longer than 4 months without serious fouling issues when operating a UF membrane pilot-plant at a moderate transmembrane flux (about 10 LMH) and solids concentrations in the membrane tank around 6-11 $g \cdot L^{-1}$ (Sanchis-Perucho et al., 2023a).

With regards to the effect of DMF on AD, some studies have reported an increment in the methane production, but with differences in its extension. By way of example, (Gong et al., 2019) studied the preconcentration of the influent organic matter with polyvinylidene fluoride hollow fiber membrane (0.02mm pore size) and its subsequent AD, obtaining productions of 360-367 ml $CH4 \cdot g^{-1}VS$, which is significantly higher than that obtained from excess sludge (155-250 ml $CH_4 \cdot g^{-1}VS$), but lower than the methane obtained from primary sludge (415 ml $CH_4 \cdot g^{-1}VS$). Net energy productions in the 0.029-0.050 $kWh \cdot m^{-3}$ range were reported. Hafuka et al. (2020) studied the digestibility of the recovered organic matter from DMF (flat sheet ceramic membranes; 0.1 μm pore size) and compared it with mixed sludge and waste activated sludge. Result showed methane yields of 0.54±0.05, 0.43±0.01 and 0.50±0.13 L $CH_4 \cdot g^{-1}VS$, respectively. Kimura et al. (2021) studied a DMF (MF membranes; 0.1 μm pore size) followed

by AD system treating raw urban wastewater. These authors reported 390 ml $CH_4 \cdot g^{-1}VS$, which was higher than the excess sludge produced in the wastewater facility (230 ml $CH_4 \cdot g^{-1}VS$). Also, energy consumption of the DMF system was significantly lower than the potential energy recovery, 0.013 $kWh \cdot m^{-3}$ and 0.15-0.30 $kWh \cdot m^{-3}$, respectively.

Despite the challenge of fouling, this technology is attractive due to its potential to recover resources from MWW, e.g., carbon, nutrients or reclaimed water. Direct filtration systems of raw MWW to concentrate organic matter with UF membranes in mid-term trials (up to one month) have been developed by, among others, Kimura et al. (2017) and Lateef et al. (2013), who found that 75% of the organic matter present in raw MWW can be redirected towards energy recovery. Modise et al. (2006) studied the performance of seven lab-scale MF membranes to mitigate pollution (organic matter, E. Coli, Fecal Coliforms, and Enterococcus) from discharges into water bodies from the primary clarifier, highlighting the importance of pore size for microbial retention, and suggesting a maximum membrane cut-off value of 0.2 μm. Delgado Diaz et al. (2012) studied the effect of a coagulation/sedimentation process prior to filtration of the primary clarifier supernatant with hollow fiber UF membranes (pore size 0.03 μm), demonstrating a high efficiency in the capture of organic matter (81-95% in terms of COD. Tuyet et al. (2015) applied DMF to raw MWW, obtaining a permeate with COD under the discharge limits (< 50 $mg \cdot L^{-1}$). The retentate was co-digested with kitchen waste and produced 64 $kWh \cdot capita^{-1} \cdot d^{-1}$. Nascimento et al. (2017) used DMF for urban wastewater by means of UF and obtained a solid-free permeate, which favored its reuse potential.

This work evaluates UF membranes for treating the supernatant from the primary settler of the Carraixet WWTP (Valencia, Spain) to divert all the particulate and colloidal organic matter to the side-stream AD process, while reducing energy requirements for organic matter removal from the mainstream activated sludge (i.e., avoiding energy for organic matter oxidation). The current WWTP treatment scheme and the membrane-based alternative treatment were mathematically modelled and the economic and environmental outcomes of both treatment schemes were compared. This comparison included an energy balance, a life cycle costing (LCC) assessment and an environmental life cycle assessment (LCA).

4.2. Materials and methods

This study compared the economic and environmental issues in two scenarios: a) Scenario 0 represented the current Carraixet WWTP configuration, and b) Scenario 1 introduced a UF unit

after the primary settler. Only the elements that differed in both scenarios were included in the assessment (membranes; membrane tanks; additional blowers and pumps; methane production; aeration requirements; chemical consumption; effluent quality; and sludge production and quality), since the variation of the common equipment and processes was considered negligible.

Both scenarios were assessed from an energetic, economic and environmental perspective. First, the Biological Nutrient Removal Model nº 2 (BNRM2) (Barat et al., 2013) implemented on DESASS 7.1. software (Ferrer et al., 2008) was calibrated by simulation and used to reproduce the current operation of the Carraixet WWTP (Scenario 0). Once the model was dynamically calibrated (i.e., represented the operation of the WWTP), the Scenario 1 simulations were carried out. The results were used to assess the energy, economic and environmental performance.

4.2.1. Current Carraixet WWTP

Carraixet WWTP is in Alboraya (Valencia, Spain) and serves a population equivalent (p.e.) of 133000. The water line consists of pretreatment (grids, sieves, sand trap and degreaser), primary settler, activated sludge for biological nitrogen removal and phosphorus precipitation, coagulation-flocculation and disinfection (UV). After treatment, a fraction of the effluent is pumped into the surrounding irrigation channels for reuse on farmland. The sludge line includes a gravity thickener for primary sludge, a dissolved air flotation unit for secondary sludge, AD, and a centrifuge for sludge dewatering. Cogeneration for energy recovery completes the facility, as shown in Figure 4.1.

4.2.2. WWTP membrane-based alternative

Figure 4.2 depicts the alternative layout of Scenario 1, based on including UF after the primary settler. By means of this membrane unit, only dissolved COD (COD_S) is treated in the activated sludge reactor, reducing the aeration needs associated with organic matter oxidation. All the particulate COD (COD_P) and a fraction of soluble COD (colloidal; with a particle size greater than the membrane pore size, i.e., 0.03 μm) is therefore sent to AD. It is important to highlight that Scenario 1 considers the possibility of a bypass stream from the outlet of the primary settler to the biological rector (dotted line) to provide the organic matter requirements for denitrification.

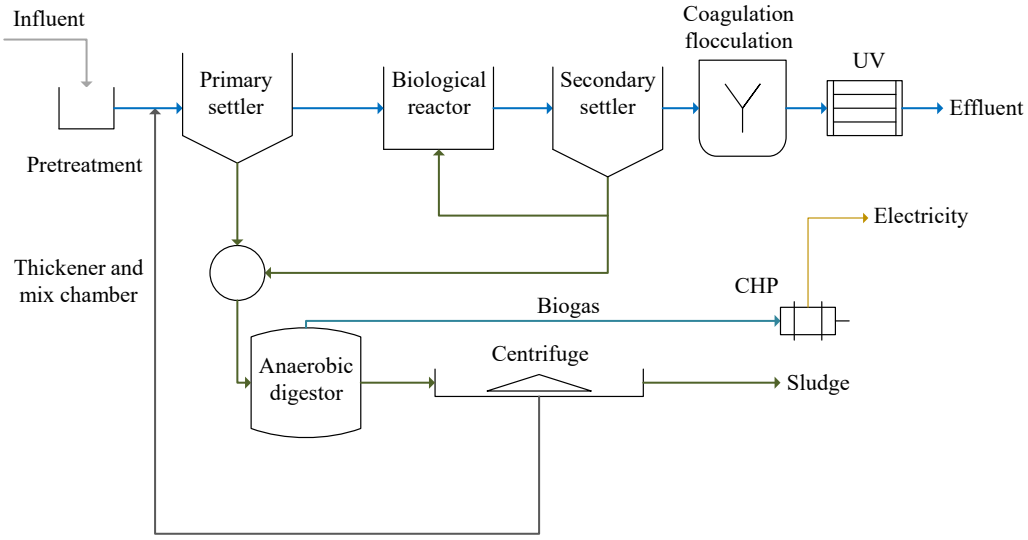


Figure 4.1. Scenario 0: Current Carraixet WWTP.

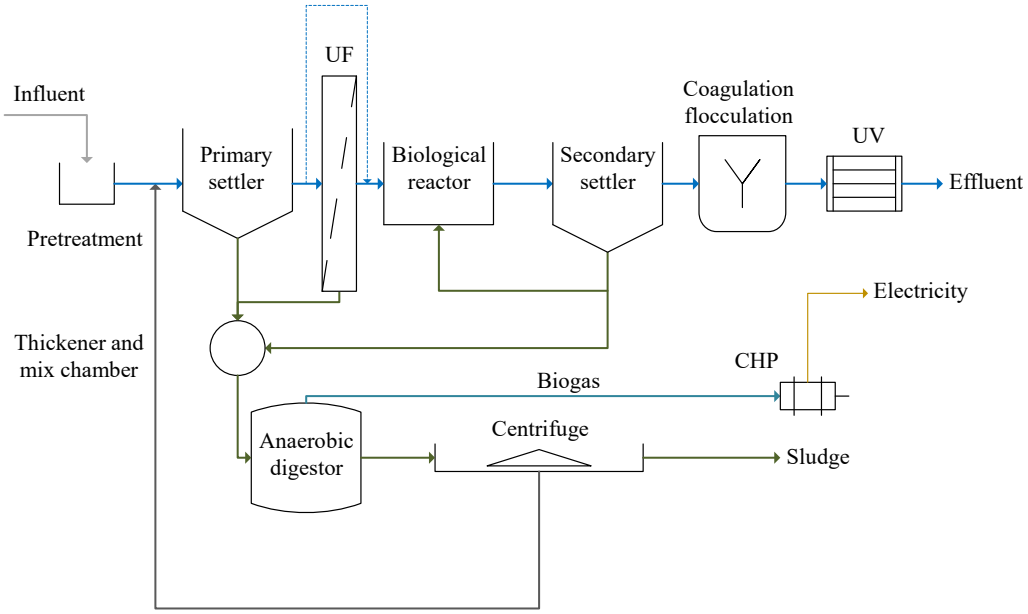


Figure 4.2. Scenario 1: membrane-based alternative proposed in the MEM4REC project (bypass in dotted line).

4.2.3. Calibration and simulation

Scenarios 0 and 1 were simulated on DESASS 7.1 software (Ferrer et al., 2008), which allows the complete wastewater treatment scheme to be evaluated, both in the water and sludge lines, based on the BNRM2 model (Barat et al., 2013). BNRM2 considers a two-step nitrification process based on Jiménez Douglas (2010) (for further details of model parameters, see Supplementary Material). As a previous step, a simulation-based calibration was conducted to adjust the model parameters to the WWTP performance.

The average filtration data from the UF pilot plant operated in the MEM4REC project were used for the assessment in Scenario 1. Based on the project results, the net 20ºC-standardised transmembrane flux (J_{20}) was set to 10 LMH and the specific ambient air demand per m^2 of membrane (SAD_m) to 0.1 $Nm^3 \cdot h^{-1} \cdot m^{-2}$. Average TMP during filtration of 250 mbar was considered. The membranes were chemically cleaned every 10 months, according to the results obtained by Sanchis-Perucho et al. (2023b).

Table 4.1 shows the Carraixet influent quality. The simulations and filtration data provided the information required for the treatment, energy and environmental assessment: flows, composition, chemicals consumption, methane production, aeration requirements, etc.

Table 4.1. Carraixet WWTP influent quality.

Parameter	Units	Concentration
COD_T	$mg \cdot L^{-1}$	512
COD_S	$mg \cdot L^{-1}$	104
P_T	$mg\text{-}P \cdot L^{-1}$	8.8
N_T	$mg\text{-}N \cdot L^{-1}$	57.6
NH_4^+-N	$mg\text{-}N \cdot L^{-1}$	28.0
SO_4^{3-}-S	$mg\ SO_4\text{-}S \cdot L^{-1}$	119.2

4.2.4. Energy balance

The energy balance was calculated according to the energy model proposed by Judd & Judd (2006) and Pretel et al. (2016), included in DESASS 7.1. The energy consumption for aeration of the biological reactor was obtained directly by simulation, while that associated with the membrane scouring blowers, permeate pumps and bypass pumps was calculated by means of equations included in the Supplementary Material.

Energy recovery was estimated from the biogas flow rate, methane percentage in the biogas and methane calorific power. An average power efficiency of 35% was selected, in accordance with the power efficiencies reported by Darrow et al. (2017) for reciprocating engines used as combined heat and power (CHP) systems.

4.2.5. Life cycle costing

The economic evaluation of both scenarios was carried out by life cycle costing (LCC) to calculate the total annual costs (TAC). A functional unit of 1 kg of removed COD (COD_{rem}) was selected for the economic assessment. Therefore, the TAC (€·kg COD_{rem}^{-1}) of the whole process was calculated by adding the annual investment cost of the CAPEX to the annual OPEX, as shown in Eq. 4.1:

$$TAC = \frac{r\,(1+r)^t}{(1+r)^t - 1} \cdot CAPEX + OPEX \qquad \text{Equation 4.1}$$

where r is the annual discount rate and t is the depreciation period in years.

LCC included construction and operation phases. The main considerations when calculating CAPEX and OPEX are summarized as follows:

<u>CAPEX</u>

- CAPEX in Scenario 0: Since Scenario 0 corresponds to the current operation of the treatment plant, its CAPEX is 0 €·kg COD_{rem}^{-1}.
- Depreciation: A depreciation period of 20 years for building works and 10 years for equipment and membranes was used to calculate the TAC. The annual discount rate was set to 3% for the former and 5% for the latter.
- Membrane tank: The membrane tank volume was estimated according to the dimensions of a commercial membrane unit (PULSION® Koch, 0.03-µm pore size).
- Pipelines: The maximum velocity of the liquid in pipes was set to 1.5 $m \cdot s^{-1}$ to calculate the pipe diameter, thus preventing excessive losses of mechanical energy.
- Equipment: Acquisition of blowers, pumps and UF membranes was included. Equipment expenditures were estimated by correlating the capacities (mainly based on flow rates) and unit costs provided by the manufacturers.

OPEX

- Power requirements: Both energy and power tariffs (Spanish) are included in the appraisals according to Somenergia (2022).
- Energy recovery from methane: Energy production from biogas generated in the AD was considered.
- Chemical reagents for membrane cleaning: Sodium hypochlorite and citric acid were considered for membranes chemical cleaning. 2000 ppm was adopted as the concentration of both sodium hypochlorite and citric acid, while the contact time was set to 6 hours. A frequency of 1.2 chemical cleaning events per year was considered.
- Membrane replacement: The lifespan of UF membranes was used to calculate the membrane replacement costs. The lifespan of the UF membrane was calculated from the maximum total contact with chlorine permissible before membrane replacement: 500000 ppm-hours cumulative, according to the supplier. Maximum lifespan is 12 years in any case.
- Equipment replacement and maintenance: The lifetime of blowers and pumps was established based on the total working hours in the manufacturers' recommendations. A maximum lifespan of 10 years was considered.
- Building works and construction maintenance: Maintenance of works and buildings were considered as a percentage of the associated CAPEX, according to the technical specifications for general WWTP constructions (Inyges Consultores S.L., 2015).
- Labor costs: labor costs were not included in this LCC since both scenarios were considered to be operated by the same staff.

Table 4.2 shows the unit costs and correlation functions used to assess the CAPEX and OPEX in the proposed scenarios.

Table 4.2. Unit costs and correlation functions used to evaluate capital and operating expenses (CAPEX/OPEX) in both scenarios.

Unit costs and correlation function of capital and operating expenses	
Pipes, (final cost is function of diameter) €·m^{-1}	8-220
Polypropylene, €·m^{-2}	9.6
Ultrafiltration hollow-fiber membrane, €·m^{-2}	Membrane = -2.985·ln A[a] (m^2) + 68.159
Energy (function of time period), €·kWh^{-1}	0.231 - 0.340
Sodium hypochlorite, €·l^{-1}	0.72
Citric acid, €·kg^{-1}	0.75
Polyelectrolyte, €·kg^{-1}	2.35
Wasted sludge for farming, €·t^{-1}	4.81
Blower, €·unit^{-1}	Blower = 2152.4·ln Q[b] (m^3/h) -7849.6
Pumps, €·unit^{-1}	Pump = -0.6582·Q$^{2\,b}$ (m^3/h) + 263.54·Q[b] (m^3/h) + 1005.4

[a] Membrane surface
[b] Flow

4.2.6. Life cycle assessment

Environmental performance of both scenarios was calculated by means of a life cycle assessment (LCA) according to ISO 14040:2006. The life cycle inventory (LCI) of individual materials and processes (see Table 4.3) were compiled using the Ecoinvent 3.4 and Industry data 2.0 databases accessed via SimaPro v9.1.1 (PRé Consultants; The Netherlands). Life cycle impact assessment (LCIA) was based on the ReCiPe 2016 method and hierarchical approach, also accessed via SimaPro v9.1.1. The construction phase for additional infrastructure in Scenario 1 and operation phase for both scenarios was considered. The dismantling phase was not included as it was considered negligible, according to Foley et al. (2010). In the operating phase, the following aspects were included in the LCA: i) biological and filtration performance, ii) sludge handling and disposal, iii) energy consumption, iv) chemicals consumption, v) energy use. As in LCC (see section 4.2.5), a functional unit of 1 kg of COD_{rem} and a 20-year lifespan were set. The selected environmental categories were Global Warming Potential (GWP), Freshwater Eutrophication (FWE), Marine Eutrophication (ME) and Fossil Resource Scarcity (FRS). The results of other impact categories included in ReCiPe 2016 can be found in the Supplementary Material.

Table 4.3. Materials and processes selected for the Life Cycle Inventory (LCI).

Construction materials, kg·kg COD_{rem}^{-1}	Chromium steel *(1 kg Steel, chromium steel 18/8 {GLO}| market for | APOS, S (Ecoinvent 3))* Cast iron (*1 kg Cast iron {GLO}| market for | APOS, S (Ecoinvent 3))* Polyester resin (*1 kg Polyester resin, unsaturated {GLO}| market for | APOS, S (Ecoinvent 3))* Polypropylene (*1 kg Polypropylene, granulate {GLO}| market for | APOS, S (Ecoinvent 3))* PVC *(1 kg PVC pipe E (Industry data 2.0))* Transport *(1 tkm Transport, freight, lorry 16-32 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3)*	
Energy consumption, kWh·kg COD_{rem}^{-1}, *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Energy avoided (energy recovery from methane), kWh·kg COD_{rem}^{-1} *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Reagent consumption, kg·kg COD_{rem}^{-1}	Polyelectrolyte *(1 kg Polyacrylamide {GLO}| market for | APOS, S (Ecoinvent 3))* NaOCl *(1 kg Sodium hypochlorite, without water, in 15% solution state {GLO}| market for | APOS, S (Ecoinvent 3))* Citric acid *(1 kg Citric acid {GLO}| market for | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
Discharge to water, kg·kg COD_{rem}^{-1}	Ammonium NT *(1 kg emissions NT to water)* Total phosphorous, PT*(1 kg emissions PT to water)*	
Discharge to soil, kg·kg COD_{rem}^{-1}	Disposal to agriculture *(1 kg Solid manure loading and spreading, by hydraulic loader and spreader {GLO}| market for | APOS, S (Ecoinvent 3))*	NH^{+}_4 to soil *(1 kg emissions NH^{+}_4 to soil through sludge spreading)*
		PO^{-3}_4 to soil *(1 kg emissions PO^{-3}_4 to soil through sludge spreading)*
		N-based fertilizer (*1 kg Nitrogen fertiliser, as N {GLO}| market for | APOS, S)*
		P-based fertilizer (*1 kg Phosphate fertilizer, as P2O5 {GLO}| market for | APOS, S (Ecoinvent 3))*
		N_2O to air *(1 kg emissions N_2O to air through sludge spreading)*
	Disposal to landfill *(1 kg Municipal solid waste {RoW}| treatment of, sanitary landfill | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	

4.3. Results and discussion

The following sections give the results obtained from the simulation of Scenarios 0 and 1, as well as the comparison of the energy balance, LCC and LCA between the current wastewater treatment facility and the alternative proposed in the MEM4REC project for the Carraixet WWTP.

4.3.1. WWTP performance simulation

The simulation started with the calibration of the BNRM2 model to minimize the difference of the Carraixet experimental data performance and the model's outcome. Once the model had been calibrated, different operating alternatives were simulated with UF membranes after the primary clarifier. All the simulations were conducted at 20 ºC.

4.3.1.1. Model calibration and simulation of Scenario 0

The BNRM2 model was calibrated by the simulation-based calibration method. The model parameters were adjusted to minimize any differences between the real Carraixet WWTP performance and the simulation outcome. SRT was maintained at 7 days during the calibration, according to the average value maintained in the Carraixet activated sludge reactor.

Table 4.4 shows a comparison of the experimental values for different stream flows and the corresponding simulation after the model calibration. The agreement between the simulated results and the real plant data shows that the calibrated model could adequately reproduce the WWTP performance (further information on the model parameters are available in the Supplementary Material).

Finally, results also show that Carraixet WWTP would accomplish with the discharge criteria established in the DIRECTIVE (91/271/EEC) for facilities with treatment capacities of more than 100000 p.e. discharging to sensitive zone (< 10 mg $N \cdot L^{-1}$ and < 1 mg $P \cdot L^{-1}$).

4.3.1.2. Simulation of Scenario 1

As already mentioned, an alternative configuration of the treatment plant (Scenario 1) was simulated to study the effect of incorporating the membranes in the primary treatment on the WWTP performance, as well as to compare this alternative with the current plant from an energy, economic and environmental point of view. For this, the objective of the simulations was to obtain a similar effluent quality as in Scenario 0 in terms of total nitrogen. This pollutant was the most complex for its removal from the studied system because of the low COD:N. Phosphorus was removed by chemical precipitation.

Table 4.4. Results of model calibration and simulation in Scenario 0 (current Carraixet WWTP).

Effluent	Unit	Carraixet WWTP	Simulation
COD_T	$mg \cdot L^{-1}$	62 ± 5	53
COD_S	mg·L-1	-	44
NO_2^--N	mg $N \cdot L^{-1}$	-	0.0
NO_3^--N	mg $N \cdot L^{-1}$	-	7.3
NH_4^+-N	mg $N \cdot L^{-1}$	1.4 ± 0.2	1.1
PO_4-P	mg $P \cdot L^{-1}$	1.2 ± 0.2	0.8
N_T	mg $N \cdot L^{-1}$	9.7 ± 0.4	9.3
P_T	mg $P \cdot L^{-1}$	0.9 ± 0.2	0.9
Energy			
Aeration	$kWh \cdot d^{-1}$	-	5860
Biogas			
Q_{biogas}	$m^3 \cdot d^{-1}$	-	4620
CH_4	%	-	61.4
Sludge			
M_{sludge}	$kg \cdot d^{-1}$	-	8426
Centrate			
COD_T	$mg \cdot L^{-1}$	842.6 ± 25.3	-
NH_4^+-N	mg $N \cdot L^{-1}$	741.5 ± 35.1	-
PO_4-P	mg $P \cdot L^{-1}$	-	-
N_T	mg $N \cdot L^{-1}$	750.7 ± 37.2	-
P_T	mg $P \cdot L^{-1}$	-	-

Table 4.5 summarizes the simulation outcomes and includes the values of the simulated facility operation and stream flows. The first subset of simulations is shown under the title of *Scenario 1a*, which comprises four sub-scenarios: a1, a2, a3 and a4. This set of simulations varied SRT (7 and 13 d) and dissolved oxygen (DO) (2.0, 1.0 and 0.5 mg $O_2 \cdot L^{-1}$) in the biological reactor. Sub-scenario 1-a1 was simulated with an SRT of 7 d and 2.0 $mg \cdot L^{-1}$ of DO in the reactor (the same operating conditions as in Scenario 0). In these conditions, the combined metabolism of the ammonia oxidizing bacteria (X_{AOB}) and nitrite oxidizing bacteria (X_{NOB}), both with significant populations in the reactor (61 and 35 mg $COD \cdot L^{-1}$, respectively), raised the nitrate concentration in the effluent up to 21.8 mg $N \cdot L^{-1}$. The lack of organic matter prevented Sub-scenario 1-a1 from denitrifying to remove enough nitrogen and, therefore, the total nitrogen in the effluent (22.9 mg $N \cdot L^{-1}$) clearly exceeded the value obtained in Scenario 0 (9.3 mg $N \cdot L^{-1}$).

Since Sub-scenario 1-a1 was unable to remove nitrogen due to the lack of COD, a new simulation was performed by reducing DO to 1.0 $mg \cdot L^{-1}$ (Sub-scenario 1-a2), with the aim of preventing the growth of X_{NOB}, much more dependent on oxygen (Wang et al., 2016), and boosting denitrification via nitrite, which requires less COD than the nitrate pathway. The simulation results showed that the X_{NOB} were indeed washed out, while the X_{AOB} were maintained. This was also reflected in the concentrations of nitrite (15.8 mg $N \cdot L^{-1}$) and nitrate (1.3 $mg \cdot L^{-1}$) in the effluent. It was also verified that this strategy reduced the total nitrogen in the effluent (17.8 mg $N \cdot L^{-1}$), compared to Sub-scenario 1-a1, but by only 5.1 mg $N \cdot L^{-1}$, this concentration still being much higher than in Scenario 0. SRT was then raised from 7 to 13 d to determine whether this increase improved denitrification (Sub-scenario 1-a3). However, the 13-d SRT led to the appearance of X_{NOB} and therefore a higher effluent nitrate concentration (20.0 mg $N \cdot L^{-1}$), which contributed to raising the concentration of total nitrogen in the discharge flow to 21.2 $mg \cdot L^{-1}$. Sub-scenarios 1-a1 to a3 indicated that an increase in SRT allowed for increased denitrification. However, it also favored the growth of $X_{NOB,}$ whose final effect was to increase the concentration of total nitrogen in the effluent due to an organic matter deficit to achieve sufficient denitrification via nitrate. A simulation with high SRT and low OD was thus proposed to check whether the wash out of X_{NOB} occurred, so that denitrification could be promoted via nitrite, with a lower COD requirement than the nitrate route. This final simulation (Sub-scenario 1-a4) was carried out with an SRT of 13 d and reducing the DO concentration to 0.5 $mg \cdot L^{-1}$. These operating conditions effectively removed X_{NOB} and reduced total nitrogen concentration (15.3 mg $N \cdot L^{-1}$) in the discharge flow. However, the low organic load available for denitrification still kept nitrogen well above the Scenario 0 values.

It should be noted that in the sub-scenarios 1-a1 to a4 effluent COD_T (23-24 $mg \cdot L^{-1}$) and COD_S (20 $mg \cdot L^{-1}$) are lower than those obtained in Scenario 0 (53 and 44 $mg \cdot L^{-1}$, respectively; see Table 4.4). This is because, while the inert soluble organic matter passed through the primary clarifier without changes in its concentration, some of it was effectively retained by the membrane, thanks to its pore size. The effluent in Scenario 1a thus presented a lower concentration for this inert organic matter, while its total and soluble quantity were reduced. There was also a reduction in the total suspended solids concentration (X_{TSS}) in the reactor in Scenario 1a (985-1756 $mg \cdot L^{-1}$) compared to Scenario 0 (3734 $mg \cdot L^{-1}$) as a result of avoiding the solids input from the primary treatment and the reduced available organic matter, due to the increase in the COD diverted to the sludge line, showing that under these conditions the reactor would be over dimensioned. In turn, the diverted COD was responsible for the higher biogas production (from 4620 $m^3 \cdot d^{-1}$ in Scenario 0 to 6079-6996 $m^3 \cdot d^{-1}$ in Scenario 1a) and reduced aeration

requirements, as shown by the energy needs in Scenario 0 (5860 kWh·d^{-1}) and Scenario 1a (1725-2821 kWh·d^{-1}).

Table 4.5. Simulation results of Scenario 1.

		Scenario 1a (without bypass)				**Scenario 1b (with bypass)**			
Operation	**Unit**	*a1*	*a2*	*a3*	*a4*	*b1*	*b2*	*b3*	*b4*
SRT	d	7	7	13	13	7	13	13	13
DO	mg·L^{-1}	2.0	1.0	1.0	0.5	2.0	0.7	0.7	0.7
Bypass	%	0	0	0	0	25	15	20	25
Reactor									
X_{SST}	mg·L^{-1}	1023	985	1756	1703	1803	2576	2838	3079
X_{AOB}	mg·L^{-1}	61	61	74	72	60	68	68	68
X_{NOB}	mg·L^{-1}	35	0	46	0	32	0	0	0
Effluent									
COD_T	mg·L^{-1}	24	23	24	23	27	26	27	28
COD_S	mg·L-1	20	20	20	20	20	20	20	20
NO_2^--N	mg N·L^{-1}	0.7	15.8	0.8	12.7	1.7	9.5	8.3	7.1
NO_3^--N	mg N·L^{-1}	21.8	1.3	20.0	1.2	17.1	0.8	0.7	0.6
NH_4^+-N	mg N·L^{-1}	0.1	0.4	0.1	1.1	0.1	0.6	0.6	0.7
N_T	mg N·L^{-1}	22.9	17.8	21.2	15.3	19.4	11.4	10.1	8.9
P_T	mg P·L^{-1}	0.9	0.9	0.9	0.9	0.9	0.9	0.9	0.9
Energy									
Aeration	kWh·d^{-1}	2821	1789	2523	1725	2996	2137	2207	2281
Biogas									
Q_{biogas}	m^3·d^{-1}	6996	6079	6550	6501	6165	6218	6089	6282
CH_4	%	57.9	64.9	61.3	61.7	58.8	60.9	60.9	57.8
Q_{CH4}	m^3·d^{-1}	4051	3945	4015	4011	3625	3787	3708	3630
Sludge									
M_{sludge}	kg·d^{-1}	8496	8445	8355	8508	8643	8322	8295	8274

Since Scenario 1a could not reach a similar quality than Scenario 0 due to lack of organic matter, a new set of simulations was planned to include a bypass stream from the primary treatment effluent to the reactor to increase the available organic matter. Examples of this set are identified in Table 4.5 as *Scenario 1b* and include 4 sub-scenarios: b1, b2, b3 and b4.

Initially, SRT and DO were set to 7 d and 2 mg $O_2 \cdot L^{-1}$, respectively, as the facility operates as in Scenario 0, and the % bypass was adjusted to achieve the effluent quality in its current operation. Sub-scenario 1-b1 shows the results obtained at a 25 % bypass. Compared to sub-scenario 1-a1 with similar SRT and DO, this additional organic load boosted denitrification and reduced total nitrogen in the effluent by up to 17.1 mg $N \cdot L^{-1}$. However, biogas production was reduced to 6165 $m^3 \cdot d^{-1}$, which was one of the objectives of the membrane-based alternative: re-using organic matter and recovering energy.

Based on the previous results, it was decided to simulate with higher SRT and lower DO to favor X_{NOB} wash-out for different bypass flows, specifically 15, 20 and 25 % of the influent flow. Sub-scenario 1-b2 increased SRT to 13 d, reduced DO to 0.7 mg $\cdot L^{-1}$ and lowered the bypass to 15 %. In this case, the nitrite pathway was enhanced compared to Sub-scenario 1-b1, N_T could be reduced to 11.4 mg $N \cdot L^{-1}$ and biogas production was increased up to 6218 $m^3 \cdot d^{-1}$. Finally, to meet N_T concentration in the effluent of Scenario 0, the bypass percentage was increased first to 20 % (Sub-scenario 1-b3) and then to 25 % (Sub-scenario 1-b4). In this latter simulation, as the result obtained for N_T was equal to that in Scenario 0 (8.9 mg $N \cdot L^{-1}$ both), it was considered that the performance of the two treatment schemes were similar and allowed their comparison. The following sections deal with the comparison of the energy balance, LCC and LCA between Scenario 0 and Sub-scenario 1-b4.

4.3.2. Energy balance

The results of the energy balance are shown in Table 4.6. As explained in section 4.2, energy assessment only includes the elements that differ in both scenarios. In Scenario 0 only the energy consumption of biological reactor aeration and energy recovery were calculated. In this case, the aeration requirements reached 0.223 kWh·kg COD_{rem}^{-1}, or 100 % of the total energy consumption considered. In Sub-scenario 1-b4, the new equipment for UF added extra energy expenses associated with: permeate pumps, blowers for membrane scouring and pumps to propel the bypass stream. Of this equipment, the highest energy consumption was associated with the blowers (0.145 kWh·kg COD_{rem}^{-1}), which required 51.6 % of the energy consumption. This result is in line with other studies with UF membranes (Jiménez-Benítez et al., 2023; Pretel et al., 2014) which pointed out the significance of the blower energy expenditure for membrane scouring. Permeate pumps, with an energy consumptions of 0.015 kWh·kg COD_{rem}^{-1}, accounted for 5.2 % of the energy, while that of the bypass pumps was negligible (0.004 kWh·kg COD_{rem}^{-1}; 1.4 %). On the other hand, the oxygen requirements and their associated energy consumption for organic matter oxidation and nitrification in the aerobic reactor was reduced to 47.1 % of Scenario 0 (from

0.223 kWh·kg COD_{rem}^{-1} to 0.118 kWh·COD_{rem}^{-1}). As a result, energy consumptions in Scenario 0 and Sub-scenario 1-b4 account for 0.223 kWh·kg COD_{rem}^{-1} and 0.282 kWh·kg COD_{rem}^{-1}, respectively, with the membrane scouring requirements being responsible for the higher energy consumption.

With regard to energy recovery, Scenario 0 produced 0.550 kWh·kg COD_{rem}^{-1} while Sub-scenario 1-b4 increased energy recovery by up to 0.662 kWh·kg COD_{rem}^{-1}, or 20.4 % more, taking advantage of the additional organic matter available obtained through UF. As the net energy demand in Scenario 0 was -0.328 kWh·kg COD_{rem}^{-1} and -0.381 kWh·kg COD_{rem}^{-1} in Sub-scenario 1-b4, the alternative membrane-based scheme provided energy savings of 0.053 kWh·kg COD_{rem}^{-1}.

Table 4.6. Comparison of energy consumption (EC), energy recovery (ER) and net energy demand (NED) in each scenario (kWh·kg COD_{rem}^{-1}).

Scenario	Permeate pumping	Membrane scouring	Bypass pumping	Reactor aeration	Energy recovery	Energy consumption	Net energy demand
Scenario 0	0.000	0.000	0.000	0.223	-0.550	0.223	-0.328
Sub-scenario 1-b4	0.015	0.145	0.004	0.118	-0.662	0.282	-0.381

4.3.3. Life cycle costing

The main OPEX, CAPEX and TAC results are shown in Figure 4.3. OPEX in Scenario 0 accounted for €-0.137 kg COD_{rem}^{-1}, while Sub-scenario b4 showed similar values for operating expenditure: €-0.150 kg COD_{rem}^{-1}. This represents savings of €0.013 kg COD_{rem}^{-1} compared to Scenario 0. In Scenario 0, the main contributors to OPEX were reactor aeration (€0.116 kg COD_{rem}^{-1}) followed by chemical phosphorus precipitation (€0.024 kg COD_{rem}^{-1}), sludge conditioning (€0.006 kg COD_{rem}^{-1}) and sludge management (€0.002 kg COD_{rem}^{-1}). Power costs (€0.001 kg COD_{rem}^{-1}) contributed to a lesser extent and energy recovery allowed savings of €0.287 kg COD_{rem}^{-1}. On the other hand, membrane filtration in Sub-scenario 1-b4 appeared with two significant associated costs: €0.076 kg COD_{rem}^{-1} for membrane scouring and €0.025 kg COD_{rem}^{-1} for UF membrane replacement during the considered facility lifespan. Sub-scenario b4 also had reactor aeration as a significant expenditure but it was reduced to €0.061 kg COD_{rem}^{-1} as COD and nitrogen removal in this unit was lowered to boost AD. Chemical precipitation was reduced to €0.011 kg COD_{rem}^{-1} (54 % lower than Scenario 0), but this saving was almost counterbalanced by the inclusion of permeate pumping requirements

(€0.008 kg COD_{rem}^{-1}). The costs associated with sludge conditioning and management did not change (€0.006 and €0.002 kg COD_{rem}^{-1}, respectively), since similar sludge production occurred in both scenarios. Scenario 0 produced 8426 $kg \cdot d^{-1}$ while sub-scenario 1-b4 produced 8274 $kg \cdot d^{-1}$, a difference of 1.8 %. Maintenance of blowers and pumps, power cost (€0.001 kg COD_{rem}^{-1} each) and bypass pumping (€0.002 kg COD_{rem}^{-1}) contributed with low expenditure, while maintenance of pipes and membrane tanks showed negligible costs.

Regarding the overall energy cost of equipment operation (blowers and pumps), the result in Scenario 0 was €0.170 kg COD_{rem}^{-1} and higher in Sub-scenario 1-b4, €0.198 kg COD_{rem}^{-1}, which is consistent with the outcome of energy consumption (see Section 4.3.2). Considering energy recovery, revenues of €0.287 kg COD_{rem}^{-1} for Scenario 0 and €0.346 kg COD_{rem}^{-1} for Sub-scenario 1-b4 were obtained, and net energy profits of €0.170 kg COD_{rem}^{-1} and €0.198 kg COD_{rem}^{-1} were calculated, respectively. This means that Sub-scenario 1-b4 obtained a surplus of €0.028 kg COD_{rem}^{-1}. It is important to highlight that in this study it has been considered that the sale price of the energy surplus was the same as the purchase price of the electricity tariff.

Regarding CAPEX, only Sub-scenario 1-b4 showed values (€0.105 kg COD_{rem}^{-1}) since Scenario 0 represents a business-as-usual case with no investment. As Figure 4.3b shows, UF membranes represents the main cost: €0.099 kg COD_{rem}^{-1} and 94.7 % of CAPEX. Pumps (€0.002 kg COD_{rem}^{-1}), pipes (€0.002 kg COD_{rem}^{-1}) and blowers (€0.001 kg COD_{rem}^{-1}) contribution is much lower and membrane tank construction is negligible.

Finally, Figure 4.3c shows the TAC for both scenarios. As can be seen, the TAC in Scenario 0 is €0.092 kg COD_{rem}^{-1} lower than in Sub-scenario 1-b4. Considering that energy balance and OPEX in Sub-scenario 1-b4 are more favorable than Scenario 0, CAPEX shows that membrane cost is the main bottleneck for implementing the MEM4REC membrane-based alternative, since the operating savings do not offset the membrane investment of €0.099 kg COD_{rem}^{-1}, plus a membrane replacement cost of €0.025 kg COD_{rem}^{-1}.

Despite this, the improvement in the performance of the membranes, including their durability (Siagian et al., 2021), together with their expected lower cost in the future in an energy context in which prices will remain high (OECD, 2022), could improve the economic sustainability of the MEM4REC alternative in the medium term over conventional treatment. In this regard, a 25% increase in the membrane lifespan, going from 12 to 15 years, would mean reducing the TAC

from €-0.045 kg COD_{rem}^{-1} to €-0.058 kg COD_{rem}^{-1}. A 20 % cheaper membrane would reduce the TAC to €-0.074 kg COD_{rem}^{-1}. A 10% increase in the price of energy would mean that the TAC of Scenario 0 would be €-0.146 kg COD_{rem}^{-1} and of €-0.055 kg COD_{rem}^{-1} in case of Sub-scenario 1-b4. Increasing net transmembrane flux ($J_{20\,net}$) a 16 % (from 8.2 LMH to 9.5 LMH) by improving filtration productivity would reduce TAC to €-0.072 kg COD_{rem}^{-1}. This last increase would be justified by considering the use of more effective fouling control strategies during the direct filtration of urban wastewater. In fact, the results obtained during the MEM4REC project only included air scouring and backwashing as fouling controllers (Sanchis-Perucho et al., 2023b). However, other studies have reported far better fouling reductions when applying alternative strategies such as chemical enhanced backwashing, coagulant dosing or membrane vibrations (Jin et al., 2017; Kimura et al., 2017; Lateef et al., 2013).

Given that all these changes are not mutually exclusive and could happen to a greater or lesser extent simultaneously, a combination, for example, of an increase in membrane durability of 10% (to 13.2 years), a 20% decrease in the cost of the membrane, an increase in the cost of energy of 10%, and an improvement in $J_{20\,net}$ of 16 % (to 9.5 LMH) would allow the TAC of Sub-scenario 1-b4 to be reduced to €-0.111 kg COD_{rem}^{-1}. Under these conditions, the operating cost would allow the return period associated with the cost of the membranes to be 5.5 years, below their durability (13.2 years). In other words, the investment in membranes could be recovered during their lifespan.

The results obtained in this study were conditioned by the fact that the discharge from the treatment plant must achieve a nitrogen reduction compatible with the legislation on sensitive areas (DIRECTIVE (91/271/EEC)**)**. This implies that part of the organic matter must be consumed in the nitrification-denitrification process, for which this study suggests a bypass flow. However, the results on biogas production and energy consumption obtained in Scenario 1a indicated that the energy consumption associated with biological reactor aeration and biogas production will be much more favorable in scenarios without a nitrogen discharge restriction, or one less stringent than in Scenario 1b. In Sub-scenario 1-a4, which presents effluent nitrogen concentrations close to the limit established in the directive for populations between 10000 and 100000 p.e. (i.e., 15 $mg \cdot L^{-1}$), the calculated energy consumption in the biological reactor was 1725 $kWh \cdot d^{-1}$, which represents a reduction of 24% with respect to Sub-scenario 1-b4. Additionally, Sub-scenario 1-a4 presented a biogas production of 6501 $m \cdot d^{-1}$ compared to 6282 $m^3 \cdot d^{-1}$ in Sub-scenario 1-b4, or 3.4 % more. The combined effect of reducing energy consumption in the reactor

and increasing biogas production will improve the energy balance and in turn will reduce OPEX, thereby improving the competitiveness of the membrane-based alternative.

Finally, the combination of water treatment and water reuse for agricultural purposes also represents an opportunity to improve the feasibility of the proposed membrane-based alternative. On one hand, the cost of the treatment associated with nutrient removal would be reduced, while the nitrogen and phosphorus content of the effluent could be sold as substitutes for commercial fertilizers. In this regard, considering a urea price of €280 per ton and €480 per ton of triple superphosphate, WWTP effluent in Sub-scenario 1-a4 would contain €31273 and €4905 per year of equivalent N-based and P-based fertilizers, respectively. There is therefore still room for improving the economic feasibility of the proposed membrane-based alternatives, especially in non-sensitive areas and locations with water reuse potential.

4.3.4. Life cycle assessment

The LCA results are shown in Figure 4.4. The selected environmental impact of GWP, FWE and ME are commonly included in life cycle impact assessments of wastewater treatments. In this study, FRS was also considered as an impact category since resource depletion has already become a matter of worldwide concern.

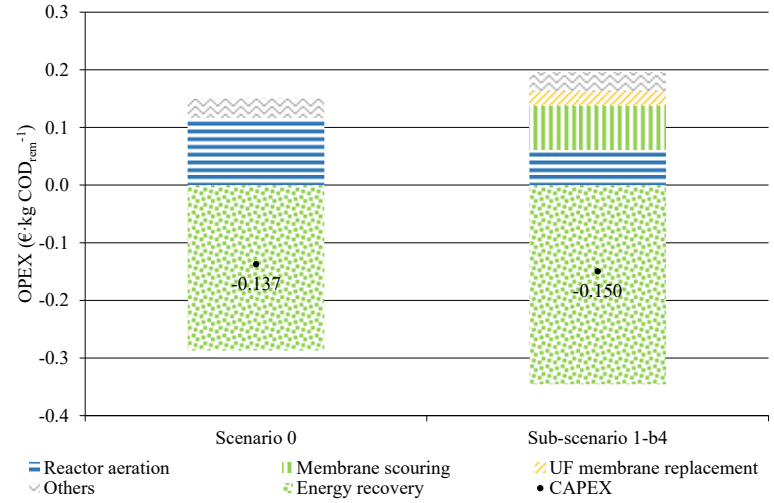
Regarding GWP (Figure 4.4a), estimation of N_2O emissions associated with nitrification-denitrification was the main contributor to GWP, accounting for 91 % and 87 % in Scenario 0 and Sub-scenario 1-b4, respectively, while indirect CO_2 emissions related to energy consumption were comparatively much lower (5 % in Scenario 0 and 7 % in Sub-scenario 1-b4). It should be remembered that we did not consider all the energy consumption in the scenarios, but only those that varied between scenarios. This result is in line with that of Daelman et al. (2013), who reported 78 % of N_2O contribution to the total carbon footprint for a full-scale WWTP, considering all the contributions to GWP in the facility. It is also in line with that of Rodriguez-Caballero et al. (2015), who obtained an average of 61 % for N_2O contribution in the La Roca del Vallès WWTP (Spain). These results highlight the crucial effect of N_2O emissions when assessing the carbon footprint of wastewater treatments, since the characterization factor for nitrous oxide is 298 kg of CO_2 per kg of N_2O. N_2O emissions were calculated as 4 % of the total nitrogen load entering the aerobic reactor, according to Kampschreur et al. (2008). However, the emissions factor reported by Kampschreur et al. (2009) for different processes and facilities present a high variation (between 0 % and 95% at lab-scale and in the range 0-15 % at full-scale) and are

influenced by several conditions: nitrite concentration, dissolved oxygen and aeration rate. Accurate results would thus need specific emission data.

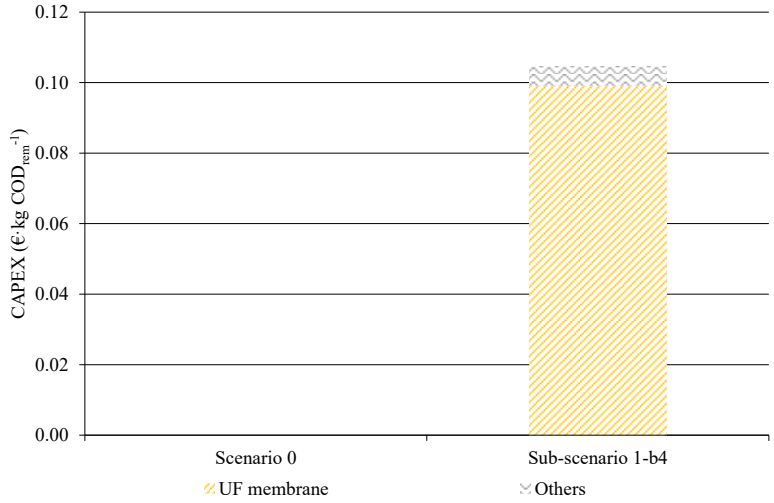
Energy recovery and the avoidance of fertilizer production through nutrient recovery by sludge spreading reduced the GWP by 0.269 kg CO_2·kg COD_{rem}^{-1} (Scenario 0) and 0.305 kg CO_2·kg COD_{rem}^{-1} (Sub-scenario 1-b4). Nutrient recovery showed a similar effect to energy recovery on reducing CO_2 emissions. Therefore, boosting the recovery of nitrogen and phosphorus from wastewater, not only in sludge but also in the effluent, and promoting their reuse as substitutes for mineral fertilizers appears to be an effective way of drastically reducing the water treatment carbon footprint. The GWP related to the construction and implementation of the membrane process showed a negligible contribution compared to the operation phase. Overall, Sub-scenario b4 reduced CO_2 emissions more than Scenario 0, due to the reduction of the total nitrogen load entering the aerobic reactor (and therefore the amount of nitrogen that eventually formed N_2O during nitrification-denitrification).

Figure 4.4b indicates direct emission of phosphorus to water bodies as the major contributor to FWE. The effluent mass flow of phosphorus in Scenario 0 and Sub-scenario 1-b4 involved an environmental load of $6.360 \cdot 10^{-4}$ kg P·kg COD_{rem}^{-1} and $5.668 \cdot 10^{-4}$ kg P·kg COD_{rem}^{-1}, respectively. This process represented approx. 98 % in both scenarios. Moreover, avoiding mineral fertilizer production reduced net FWE by $8.011 \cdot 10^{-5}$ kg P·kg COD_{rem}^{-1} (Scenario 0) and $7.653 \cdot 10^{-5}$ kg P·kg COD_{rem}^{-1} (Sub-scenario 1-b4). Emissions to soil through sludge spreading did not add significantly to the environmental impact, while the construction phase effect was negligible.

Nitrogen emissions to water bodies were the main contributor to ME (Figure 4.4c) with approx. 97-98 % of the total impact in both scenarios. The effluent of Scenario 0 had a slightly higher nitrogen load discharge (366 kg·d^{-1}) than Sub-scenario 1-b4 (355 kg·d^{-1}), so that the environmental impact in ME in Sub-scenario 1-b4 ($5.6 \cdot 10^{-3}$ kg N·kg COD_{rem}^{-1}) was lower than in Scenario 0 ($6.0 \cdot 10^{-3}$ kg N·kg COD_{rem}^{-1}). The contribution of emissions to soil, fertilizer avoidance through sludge spreading and the construction phase had a negligible effect on ME. The results of the ME and FWE impact categories confirmed nutrient recovery and reuse as crucial processes to improve the environmental performance of wastewater treatment. It would be necessary therefore to properly balance the use of sludge and water reuse for agriculture to avoid affecting water bodies due to leakage and runoff of nutrient compounds.



a)



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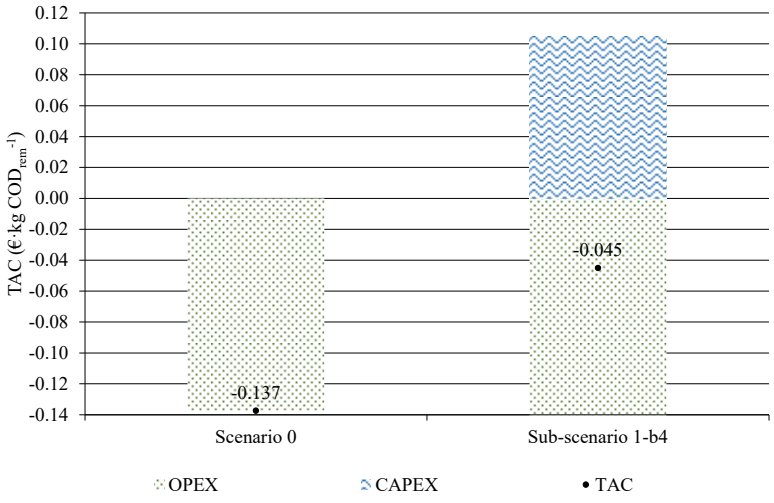

c)

Figure 4.3. Results of LCC for Scenario 0 and Sub-scenario 1-b4: a) OPEX; b) CAPEX; c) TAC.

Figure 4.4d shows the possible contribution of wastewater to reduce global resource depletion. The embedded energy and nutrient recovery can contribute to preserving conventional sources and turn wastewater into a supply stream for social and economic purposes. It is important to highlight that urban wastewater is constantly produced, since the domestic usage of water is legally prioritized. WWTPs could act as resource recovery facilities to regularly reintroduce materials (mainly nutrients and water) and energy into the economy. In this study, only the nutrient content in the sludge was considered for reuse, while water reclamation (with its nutrient content) was outside its scope. The environmental performance of the water treatment could thus be expected to improve by taking advantage of these resources, e.g. by means of fertigation of peri-urban crop fields (Jiménez-Benítez, et al., 2020).

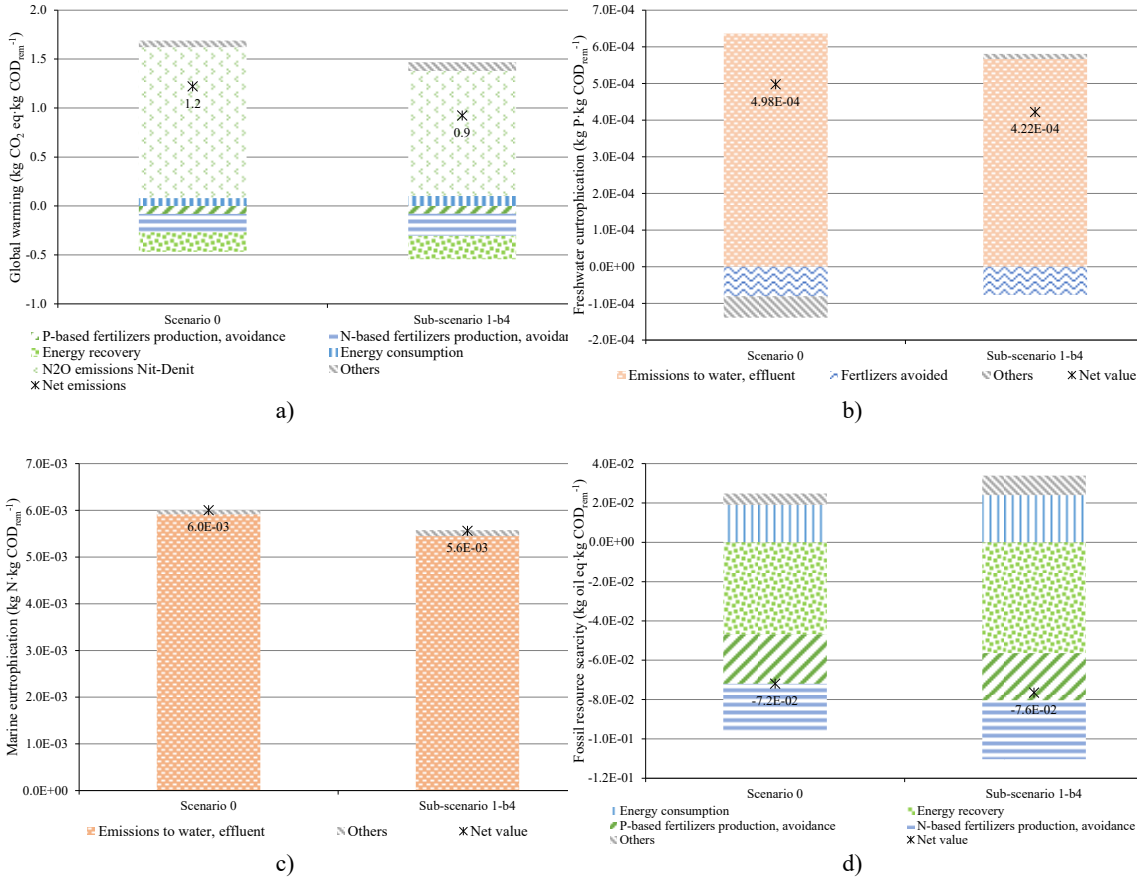

Figure 4.4. Results of environmental assessment in both scenarios for a) global warming potential, b) freshwater eutrophication; c) marine eutrophication; and d) fossil resource scarcity.

The results of this study show that the implementation of UF membrane filtration systems in primary treatment can contribute to progress towards the objectives of sustainability and circular economy within the water treatment sector. The incorporation of this technology allows the updating of existing plants without carrying out significant modifications to the current treatment

plants, which facilitates its adoption. On the other hand, future research and technological development in this area should focus on the optimization of filtration, to improve its competitiveness, and on the treatment of the permeate through processes that reduce or eliminate the bypass stream, given the organic matter required by the nitrification-denitrification process, such as the SHARON and Anammox processes. The objective should be water purification and nutrient recovery. In this sense, technologies such as ion exchange or electrodialysis are presenting promising results to separate and concentrate these resources, and membrane contactors and struvite crystallization also show good performance in order to obtain commercial fertilizer products.

4.4. Conclusion

A comparison between a current WWT facility (Carraixet WWTP, Valencia) and a membrane-based alternative consisting of implementing an UF module after the primary settler was carried out. The comparison was made based on an energy, economic and environmental assessment.

Process performance of the membrane-based layout was simulated for different OD and SRT in the biological reactor and bypass stream to provide adequate COD for nitrification. Equivalent effluent quality to the current facility was obtained with 0.7 $mg \cdot L^{-1}$ of OD, 13 d of SRT and 25 % of bypass. The energy balance shows a reduction of the net energy demand of 0.053 $kWh \cdot kg\ COD_{rem}^{-1}$ boosted by the reduction in the reactor aeration requirements, which decreased from 0.223 $kWh \cdot kg\ COD_{rem}^{-1}$ to 0.188 $kWh \cdot kg\ COD_{rem}^{-1}$, and the increase in energy recovery from 0.550 $kWh \cdot kg\ COD_{rem}^{-1}$ to 0.662 $kWh \cdot kg\ COD_{rem}^{-1}$.

Membrane-based alternative allowed reducing OPEX by €0.013 kg COD_{rem}^{-1}. The main contributor to direct operating cost was membrane scouring (€0.078 kg COD_{rem}^{-1}) followed membrane replacement (€0.025 kg COD_{rem}^{-1}) and reactor aeration (€0.006 kg COD_{rem}^{-1}), while energy recovery allowed operating savings of €0.035 kg COD_{rem}^{-1}. CAPEX was estimated in €0.105 kg COD_{rem}^{-1} and is dominated by membrane acquisition cost (€0.099 kg COD_{rem}^{-1}; 95 %). Finally, membrane-based TAC is €0.086 kg COD_{rem}^{-1} higher than current treatment cost and membrane cost appears as the main bottleneck for implementing the alternative scheme.

Conversely, LCA outcome demonstrate the environmental benefits of introducing UF membranes as primary treatment. Global warming potential can be reduced up to 0.3 kg $CO_{2eq} \cdot$ kg COD_{rem}^{-1}

mainly due to a decrease in N_2O emissions and the improvement of energy recovery. Marine and freshwater eutrophication is also diminished due to the lower N and P discharge with the effluent. And the higher energy recovery contributes to preserve energy resources and hence fossil resource scarcity shows better results.

Therefore, taking advantage of the environmental benefits presented by the implementation of UF membranes as primary treatment necessarily involves improving their economic viability. To this end, some possibilities have been pointed out in this work. The improvement in the performance of the membranes and their cheaper costs, an energy context that favor energy recovery and its sell or considering nutrients and reclaimed water value can contribute to establish a favorable framework to boost this technology for WWTP retrofitting according to sustainable principles.

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4.5. Supplementary material

4.5.1. Energy balance

Theoretical energy consumption of pumps and blowers were calculated as follows, based on the equations proposed by Judd & Judd (2006):

$$P_B = \frac{(M \cdot R \cdot T_{gas})}{(\alpha - 1) \cdot \eta_{blower}} \left[\left(\frac{P_2}{P_1} \right)^{\frac{\alpha - 1}{\alpha}} - 1 \right] \quad \text{Equation 4.1S}$$

where P_B ($J \cdot s^{-1}$) is the blower power requirement (adiabatic compression), M ($mol \cdot s^{-1}$) is the molar flow rate of biogas, R ($J \cdot mol^{-1} \cdot K^{-1}$) is the gas constant for biogas, P_1 (atm) is the absolute inlet pressure, P_2 (atm) is the absolute outlet pressure, T_{gas} (K) is the biogas temperature, α is the adiabatic index and η_{blower} is the blower efficiency.

$$P_g = q_{imp} \cdot \rho_{liquor} \cdot g \cdot \frac{\left\{ \left[\left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{asp} + \left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{imp} \right] + [Z_1 - Z_2] \right\}}{\eta_{pump}} \quad \text{Equation 4.2S}$$

where P_g is the power requirement by a general pump ($J \cdot s^{-1}$), considering both pump aspiration and pump impulsion section, calculated from the impulsion volumetric flow rate ($q_{imp.}$ in $m^3 \cdot s^{-1}$), liquor density (ρ_{liquor} in $kg \cdot m^{-3}$), acceleration of gravity (g in $m \cdot s^{-1}$), pipe length (L in m), equivalent pipe length of accidental pressure drops (L_{eq} in m), velocity (V in $m \cdot s^{-1}$), friction factor (f, dimensionless), diameter (D in m), difference in height (Z_1-Z_2, in m) and pump efficiency (η_{pump}). The friction factor (f) was calculated by means of Swamee-Jaine expression (Equation 4.3S).

$$f = \frac{0.25}{\left[\log_{10} \left(\frac{\varepsilon / D}{3.7} + \frac{5.74}{Re^{0.9}} \right) \right]^2} \quad \text{Equation 4.3S}$$

where Re is the Reynolds number, ε (m) is the pipe roughness, and D (m) is the pipe diameter.

Equation 4.2S was modified to calculate power requirements for permeate pumping (Equation 4.4S):

$$P_{stage} = \frac{q_{stage} \cdot TMP_{stage}}{\eta_{pump}}$$ Equation 4.4S

where P_{stage} is the power requirement during filtration, degasification or back-flushing ($J \cdot s^{-1}$) calculated from average transmembrane pressure (TMP_{stage} in Pa), pump volumetric flow rate (q_{stage} in $m^3 \cdot s^{-1}$), and pump efficiency (η_{pump}). Energy Consumption (EC) was calculated by addition of the power requirements of each individual equipment (P_i) per kg of COD removed (kg $COD_{rem} \cdot h^{-1}$):

$$EC = \frac{\sum P_i}{COD_{rem}}$$ Equation 4.5S

with EC in $kWh \cdot COD_{rem}^{-1}$.

Regarding energy recovery (ER) from biogas, Combined Heat and Power (CHP) technology was considered for methane valorization. ER was estimated based on: i) biogas flow rate V_{biogas} ($L \cdot d^{-1}$);
ii) methane percentage in the biogas $\%_{CH4}$, and methane calorific power CV_{CH4} ($kJ \cdot m^{-3}$). In this case, reciprocating engines were selected as CHP since it is a common technology present in WWTPs. These systems can achieve high power and overall CHP efficiencies (24-36% and 66-71%, respectively) according to Darrow et al., (2017). Equation 4.6S shows the energy from biogas valorization in terms of power (W_{biogas} in kW):

$$W_{biogas} = \frac{V_{biogas} \cdot \%_{CH4} \cdot CV_{CH4} \cdot \%_{power\ efficiency\ CHP}}{1000 \cdot 24 \cdot 3600}$$ Equation 4.6S

a $\%_{power\ efficiency\ CHP}$ of 35% was selected in this study. Based on Equation 4.S6, ER is calculated as follows:

$$ER = \frac{W_{biogas}}{COD_{rem}}$$ Equation 4.7S

Where ER is the energy recovered (kWh·kg COD_{rem}^{-1}), W_{biogas} is in kW and COD_{rem} is the mass flow of COD removed (kg $COD_{rem}·h^{-1}$).

Finally, net energy demand (NED) is calculated by means of Equation 4.8S:

$$NED = EC - ER \qquad \text{Equation 4.8S}$$

where NED, EC an ER are in kWh·COD_{rem}^{-1}.

4.5.2. Model parameters

Nitrification process was modelled based on kinetic and stoichiometric parameters published in Jiménez Douglas (2010). This model considered a two-step nitrification process, with ammonia-oxidizing bacteria (X_{AOB}) responsible for transforming ammonia into nitrite, and nitrite-oxidizing bacteria (X_{NOB}), which oxidize nitrite to nitrate. Table 4.S1shows the parameters for each bacteria.

Table 4.S1. Kinetic and stoichiometric parameters applied to scenarios simulation.

Bacteria	Parameter	Value	Unit
X_{AOB}	Y_{AOB}[a]	0.17	g COD·g NH_4-N^{-1}
	m_{AOB}[b]	0.88	d^{-1}
	b_{AOB}[c]	0.15	d^{-1}
	K_{O2}[d]	0.52	mg·L^{-1}
	K_{NH4}[e]	0.54	mg NH_4-N·L^{-1}
	K_{IHNO2}[f]	0.013	mg HNO_2-N·L^{-1}
	F_{XI}[g]	0.1	
X_{NOB}	Y_{NOB}[h]	0.7	g COD·g NH_4-N^{-1}
	m_{NOB}[i]	0.5	d^{-1}
	b_{NOB}[j]	0.075	d^{-1}
	K_{O2}[d]	0.67	mg·L^{-1}
	K_{NO2}[k]	0.26	mg NO_2-N·L^{-1}
	F_{XI}[g]	0.1	

[a] Cellular performance of ammonia oxidizing bacteria.
[b] Maximum growth rate of ammonia oxidizing bacteria.
[c] Death rate of ammonia oxidizing bacteria.
[d] Half-saturation constant of oxygen.
[e] Half-saturation constants of ammonia of ammonia oxidizing bacteria.
[f] Nitrous acid inhibition constant of ammonia oxidizing bacteria.
[g] Fraction of cellular matter transformed into debris after death.
[h] Cellular performance of nitrite oxidizing bacteria.
[i] Maximum growth rate of nitrite oxidizing bacteria.
[j] Death rate of nitrite oxidizing bacteria.
[k] Half-saturation constant of nitrogen dioxide.

4.5.3. LCA results for additional ReCiPe 2016 environmental impact categories (Midpoint).

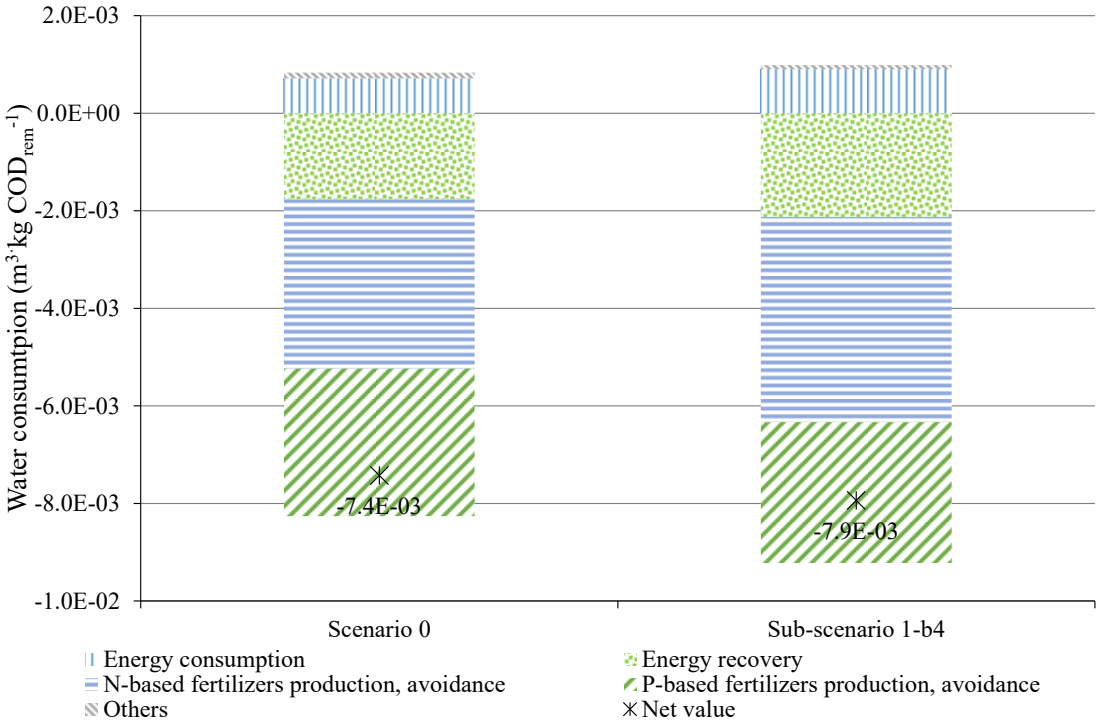

Figure 4.1S. Environmental load allocation for Water Consumption.

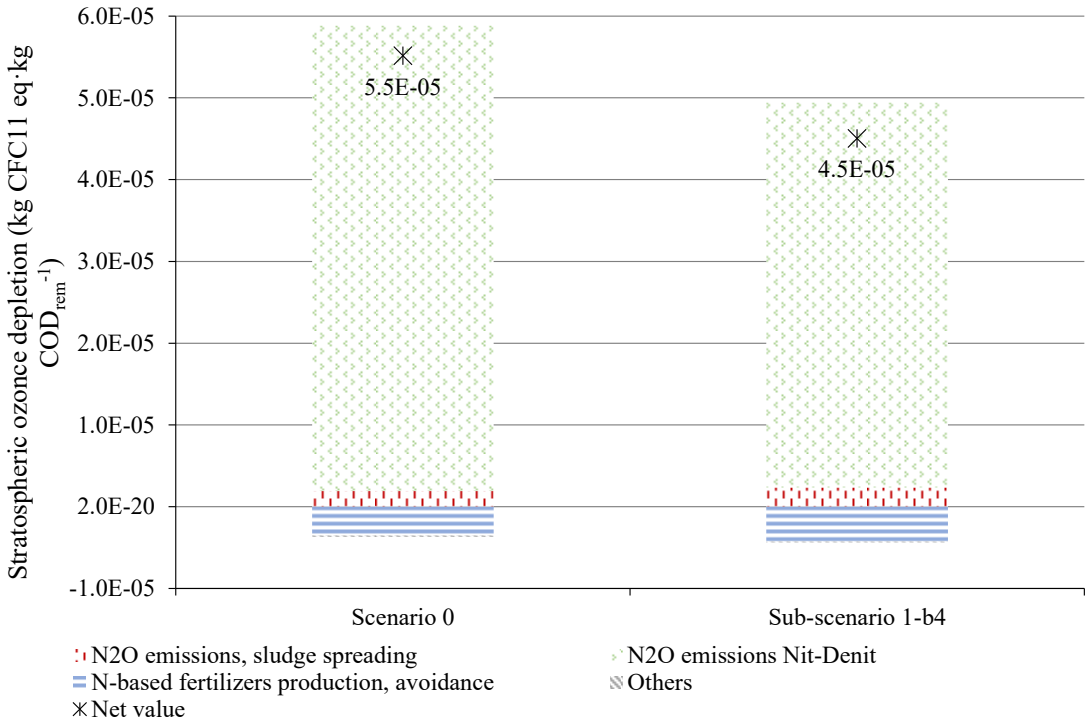

Figure 4.2S. Environmental load allocation for Stratospheric Ozone Depletion.

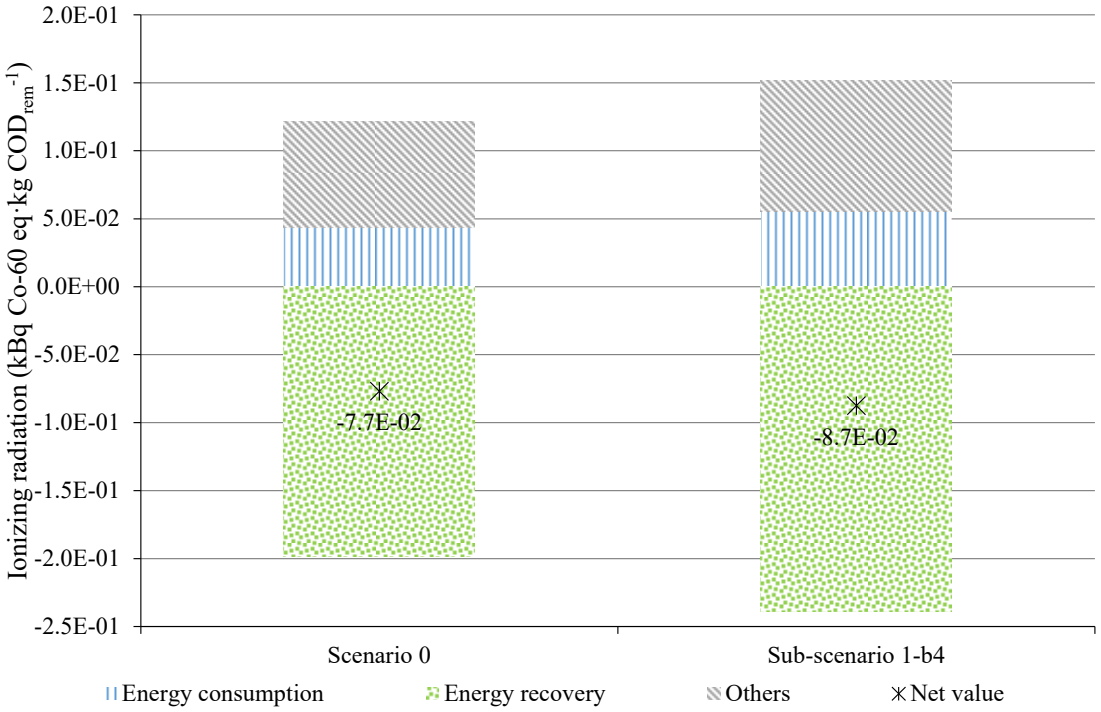

Figure 4.3S. Environmental load allocation for Ionizing Radiation.

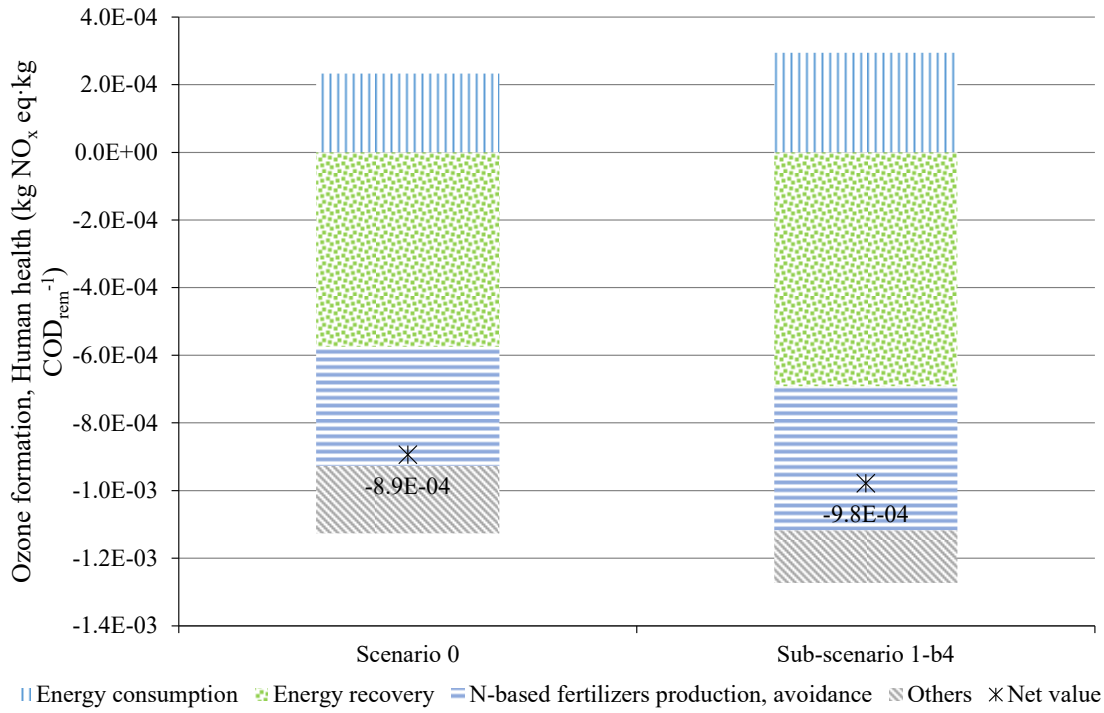

Figure 4.4S. Environmental load allocation for Ozone Formation (Human Heath).

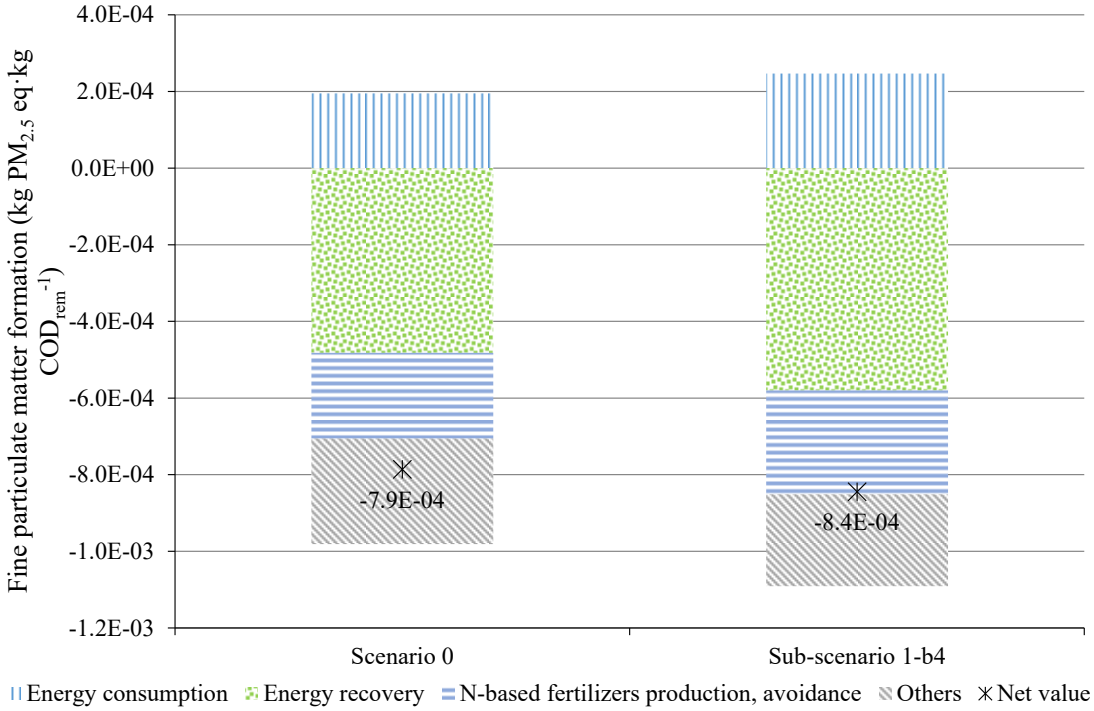

Figure 4.5S. Environmental load allocation for Fine Particulate Matter Formation.

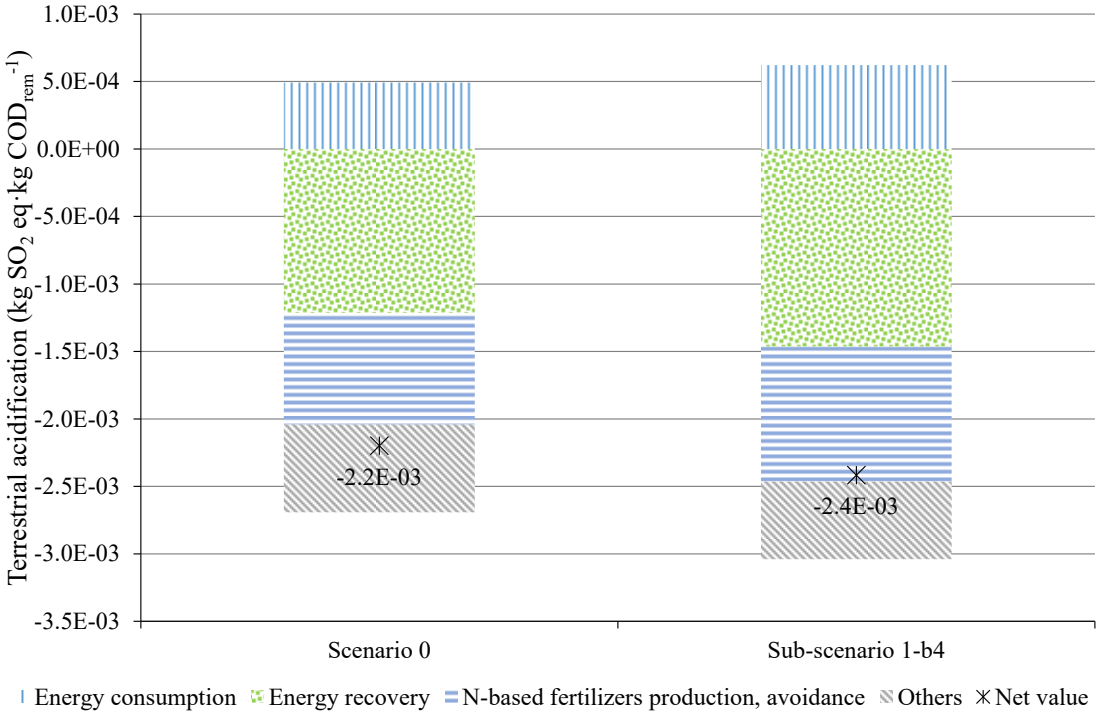

Figure 4.6S. Environmental load allocation for Terrestrial Acidification.

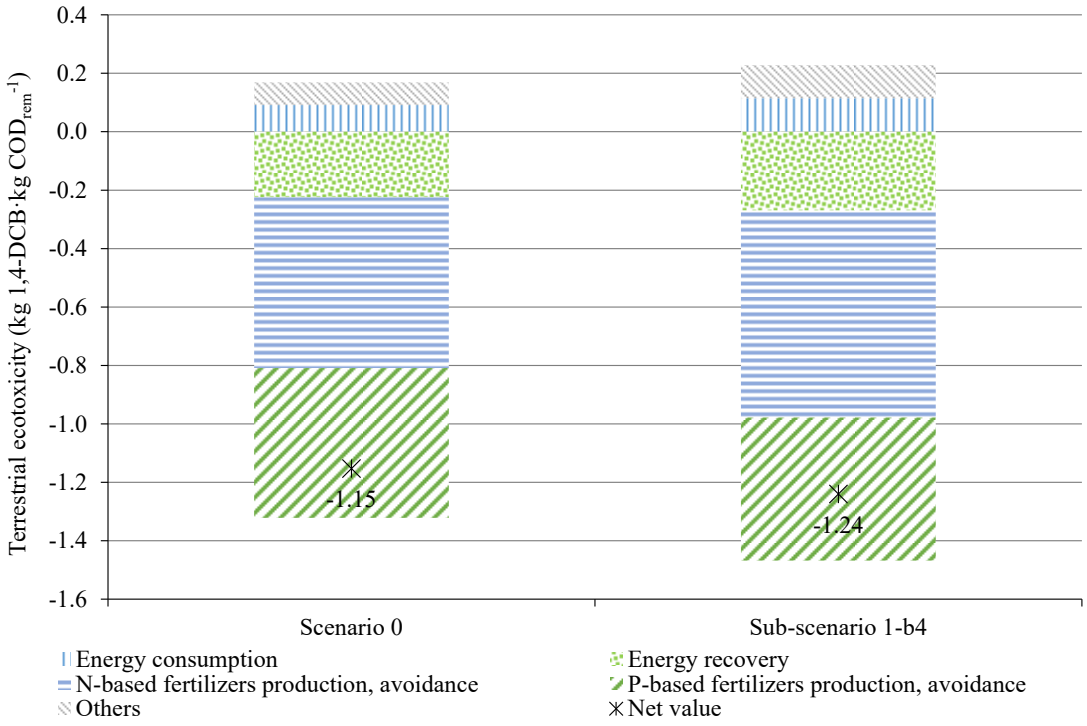

Figure 4.7S. Environmental load allocation for Terrestrial Ecotoxicity.

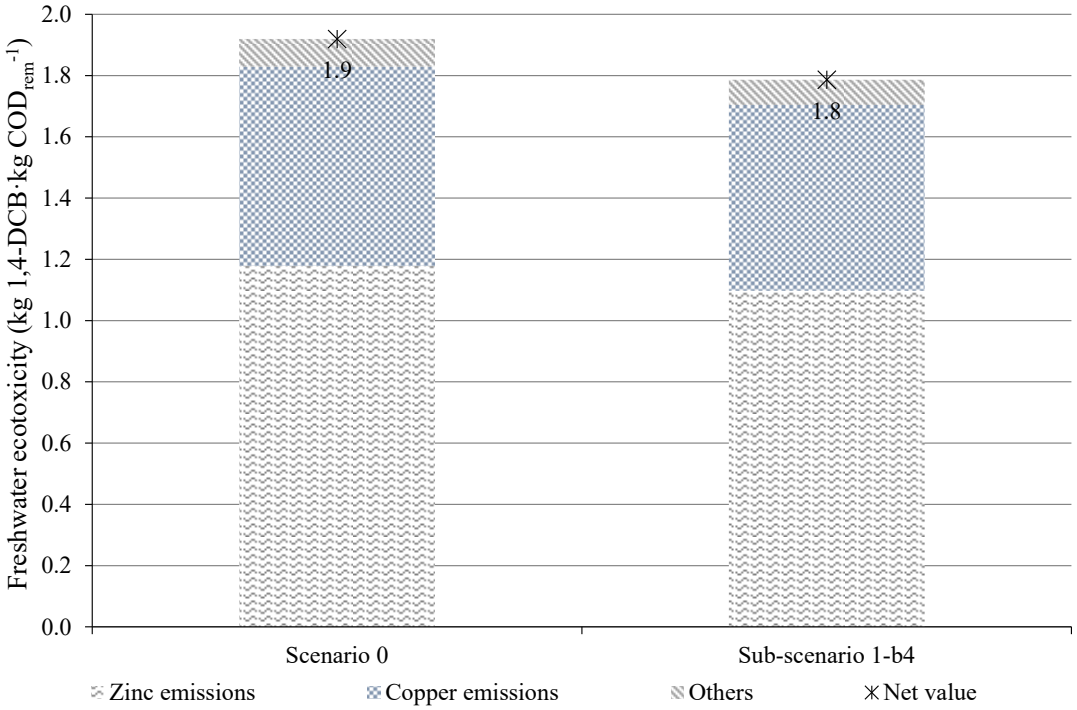

Figure 4.8S. Environmental load allocation for Freshwater Ecotoxicity.

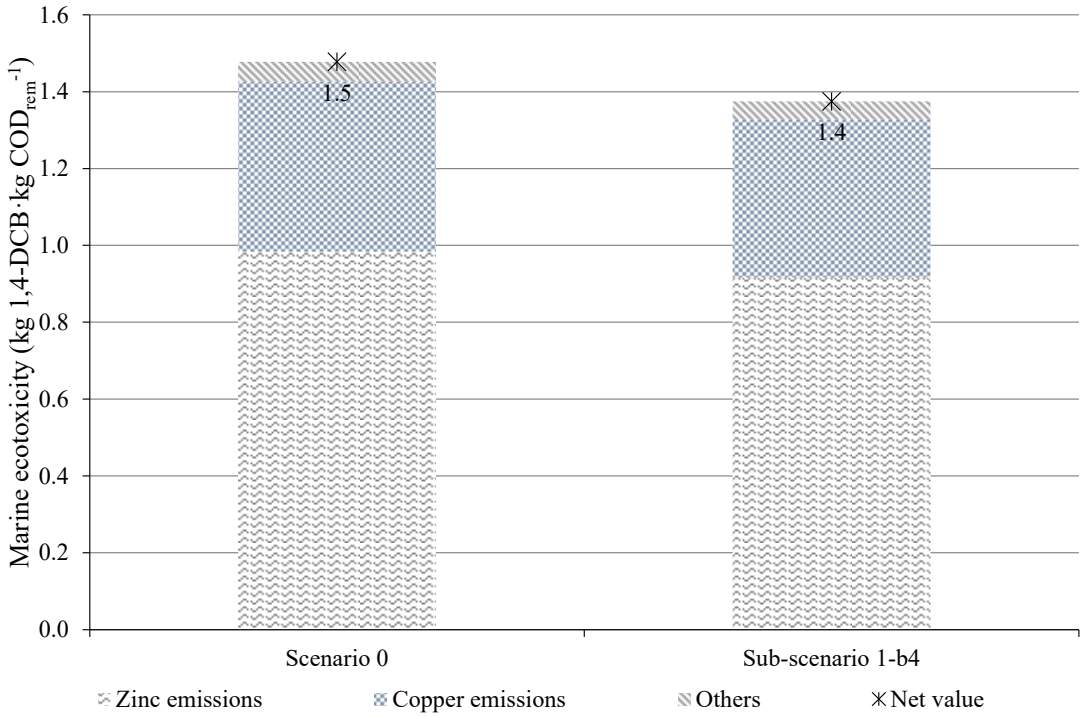

Figure 4.9S. Environmental load allocation for Marine Ecotoxicity.

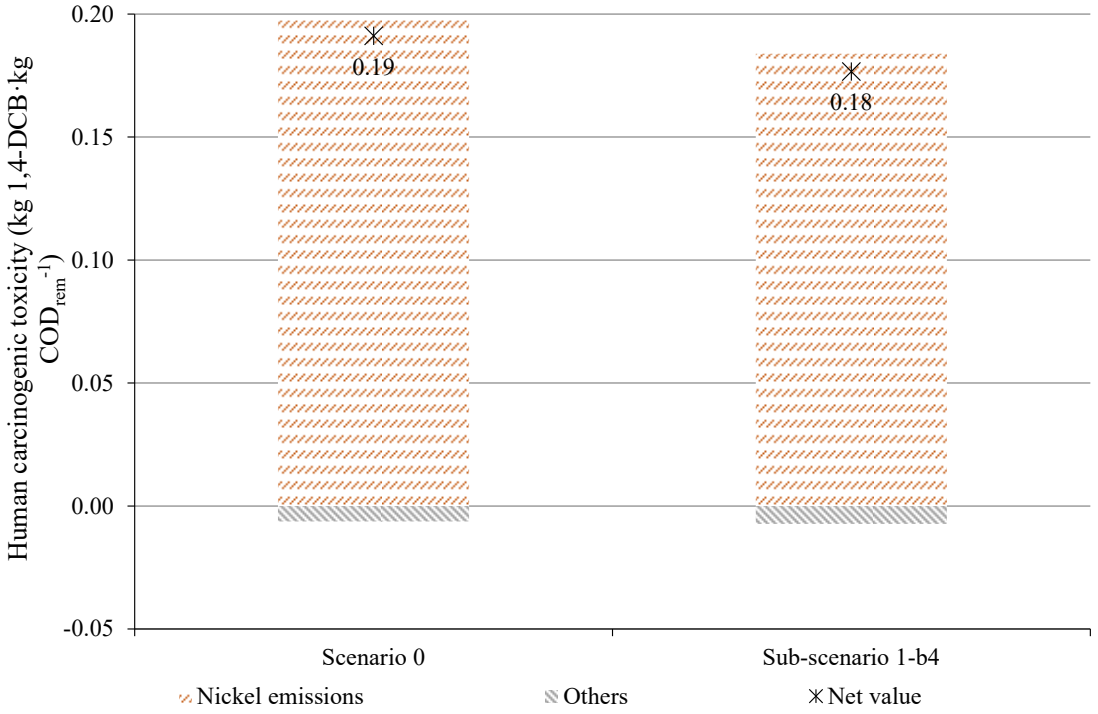

Figure 4.10S. Environmental load allocation for Human Carcinogenic Toxicity.

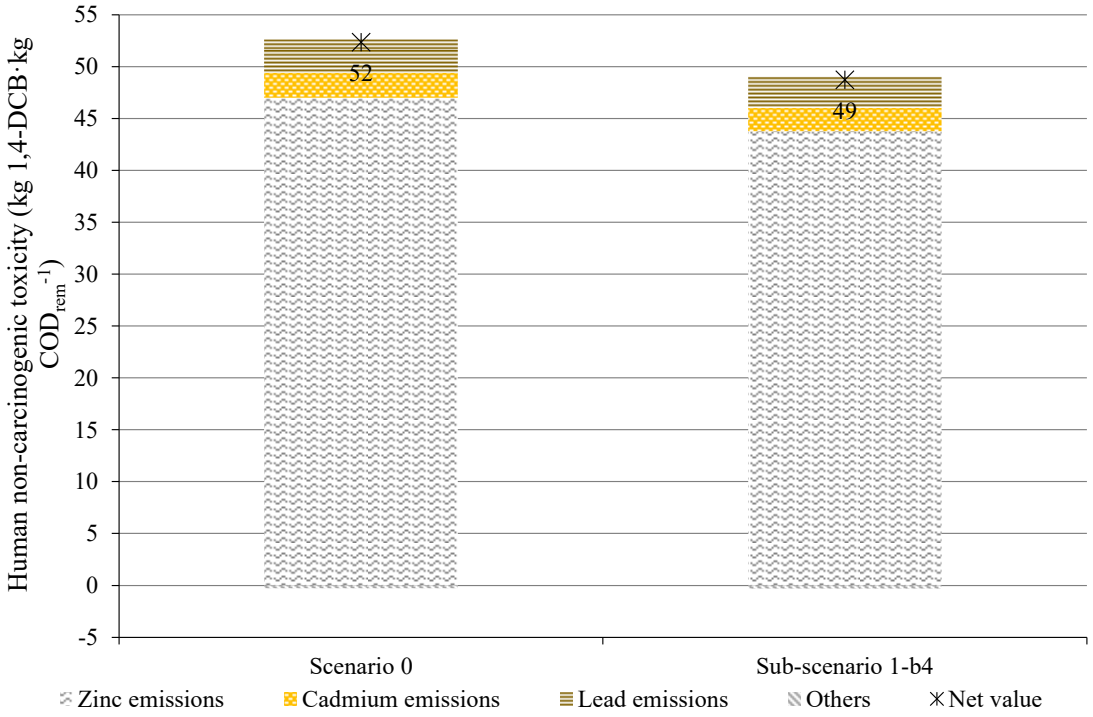

Figure 4.11S. Environmental load allocation for Human Non-carcinogenic Toxicity.

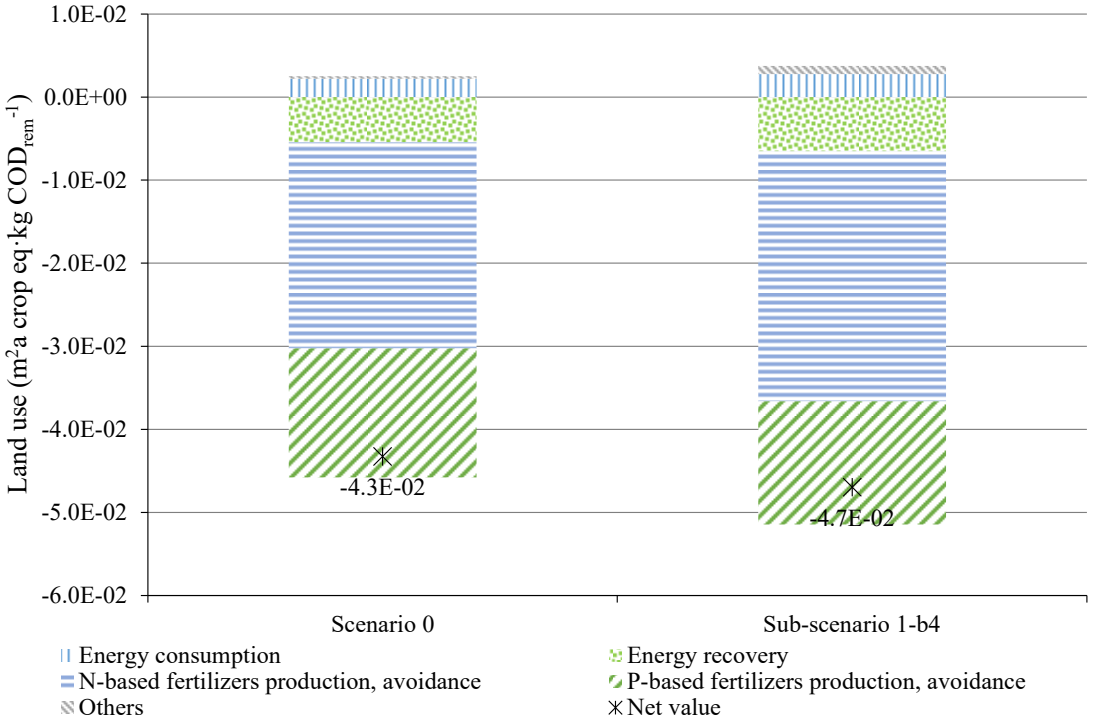

Figure 4.12S. Environmental load allocation for Land Use.

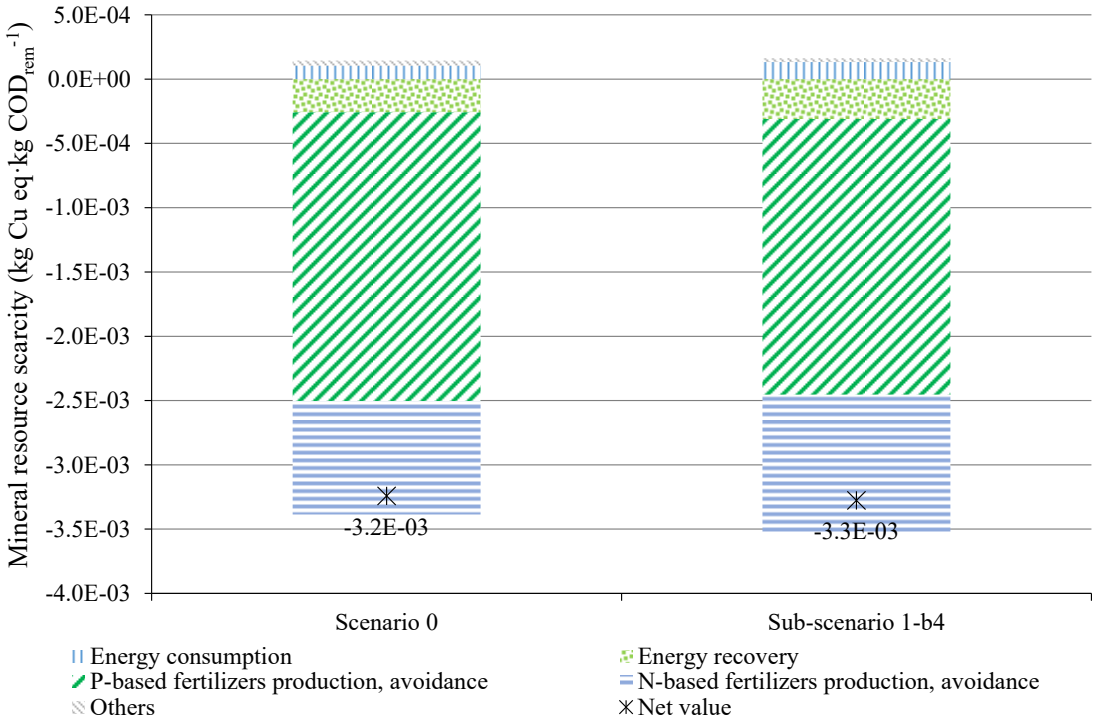

Figure 4.13S. Environmental load allocation for Mineral Resource Scarcity.

5. A semi-industrial scale AnMBR for municipal wastewater treatment at ambient temperature: performance of the biological process

Abstract

A semi-industrial scale AnMBR plant was operated for more than 600 days to evaluate the long-term operation of this technology at ambient temperature (ranging from 10 to 27 °C), variable hydraulic retention times (HRT) (from 25 to 41 h) and influent loads (mostly between 15 and 45 kg COD·d^{-1}). The plant was fed with sulfate-rich high-loaded municipal wastewater from the pre-treatment of a full-scale WWTP. The results showed promising AnMBR performance as the core technology for wastewater treatment, obtaining an average 87.2 ± 6.1 % COD removal during long-term operation, with 40 % of the data over 90 %. Five periods were considered to evaluate the effect of HRT, influent characteristics, COD:SO_4^{2-}-S ratio and temperature on the biological process. In the selected periods, methane yields varied from 70.2±36.0 to 169.0±95.1 STP L CH_4·kg^{-1} COD_{inf}, depending on the influent sulfate concentration, and wasting sludge production was reduced by between 8 % and 42 % compared to conventional activated sludge systems. The effluent exhibited a significant nutrient recovery potential. Temperature, HRT, SRT and influent COD:SO_4^{2-}-S ratio were corroborated as crucial parameters to consider in maximizing AnMBR performance.

Keywords

Anaerobic membrane bioreactor (AnMBR); biological process; demonstration plant; methane production; municipal wastewater (MWW)

Highlights

The AnMBR plant was operated for more than 600 days at ambient temperature.
Effluent COD remained below effluent standards with average COD removal above 87 %.
Waste sludge reduced by between 9 and 43 % less than CAS.
Methane yield ranged from 70 to 169 STP L CH_4·kg^{-1} COD_{inf} during the selected periods.
T, SRT, HRT and influent COD:SO_4^{2-}-S are crucial parameters of AnMBR performance.

Robles, Á., Jiménez-Benítez, A., Giménez, J.B., Durán, F., Ribes, J., Serralta, J., Rogalla, F., Seco, A., 2022. A semi-industrial scale AnMBR for municipal wastewater treatment at ambient temperature : performance of the biological process 215. https://doi.org/10.1016/j.watres.2022.118249

5.1. Introduction

Anaerobic treatment of municipal wastewater (MWW) has several advantages over conventional aerobic processes, as has been widely recognized over the years. Its main advantages are: low sludge production due to low anaerobic biomass yield (Giménez et al., 2011); reduced energy consumption because no aeration is required (Lee et al., 2017); methane is produced from organic matter valorization, which improves the technology's energy balance (Krzeminski et al., 2017b); low greenhouse gas emissions when methane is recovered from both biogas and effluent streams (Jiménez-Benítez et al., 2020b); and potential recovery potential of nutrients content (Batstone and Virdis, 2014). Moreover, the limitations associated with anaerobic processes applied to low-organic strength wastewaters (e.g. low growth rate of anaerobic biomass at ambient temperature or poor sludge settling properties) can be overcome by combining membrane technology and anaerobic reactors in the so-called anaerobic membrane bioreactor (AnMBR) (Orhon, 2013), where the filtration process allows complete physical biomass retention. Hydraulic retention time (HRT) and sludge retention time (SRT) can thus be decoupled, which means high-rate reactors can be designed and high-quality effluent suitable for water reclamation can be achieved with the appropriate membrane pore-size (Jiménez-Benítez et al., 2020a). Eventually, membrane modules are more compact than the equivalent secondary settlers + disinfection units (e.g. UV process), which helps to reduce the layout footprint of wastewater treatment facilities and permits existing space-constrained treatment plants to be retrofitted (Xiao et al., 2019).

A high concentration of nutrients (mainly nitrogen and phosphorus, but also magnesium, potassium, etc.) in the effluent of anaerobic processes has traditionally been considered a significant environmental risk to aquatic ecosystems (eutrophication) and potable water resources when these streams are discharged (Pretel et al., 2016c; Rodriguez-Garcia et al., 2011). This being true, it is also possible to consider it as an opportunity to introduce the fundamentals of the circular economy (CE) to wastewater treatment, since anaerobic treatments can be used to recover nutrients. Recovery plus recycling would contribute to closing both the materials and energy loops, and several processes and technologies are already available or being investigated to take advantage of these essential resources for agriculture and industry (Á. Robles et al., 2020a). Including these resources in a comprehensive nutrient management system becoming more import, as shown, among others, by the interest shown by the European Union in this matter (European Commission, 2015). At the present time, most efforts focus on the P since this element and phosphorite were included in European Critical Raw Materials lists in 2017 and 2014, respectively. However, N recovery is also attracting attention because of the high energy consumption and associated greenhouse gas emissions from its fixation from atmospheric N_2.

Nutrient recovery thus helps to reduce both the dependence on their production and/or imports, which is even more important for those that are exhaustible such as phosphorus and potassium, and greenhouse gases (GHG) associated with mineral fertilizers, whose production results in significant energy expenditure (P.L. McCarty et al., 2011) and has an environmental impact.

Biosolids production has also been identified as a key issue in wastewater treatment; aerobic processes are characterized by high sludge production associated with a high biomass yield of aerobic metabolism, which involves a significant management cost that affects the economic and environmental performance of sewage treatments. Additionally, the regulation of waste sewage sludge, which is a challenge in wastewater treatment processes, is also expected to be tightened (Collivignarelli et al., 2019). AnMBR can help to overcome these problems in two ways: firstly, the anaerobic process mineralizes organic matter with reduced anabolism and low sludge production. By a way of example, Foladori et al. (2015) reported a sludge production in aerobic WWTPs of 0.25-0.35 kg VSS per kg COD removed (unstabilized sludge) and around 0.18 kg VSS per kg COD removed for stabilized sludge, respectively, while Robles et al. (2020) reported a stabilized sludge production in an AnMBR demonstration plant treating MWW of 0.148 kg VSS per kg COD removed. Secondly, biomass retention by membrane modules allows increasing SRT, which contributes to achieve on-site sludge stabilization and reduces or even avoids further sludge treatment.

A final consideration is that anaerobic removal of organic matter generates methane, a storable energy carrier suitable to produce renewable heat and power. Boosting anaerobic processes through AnMBR may thus improve the energy balance of existing wastewater treatment facilities. Using it as the core technology would take full advantage of the organic matter and the 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, despite these indisputable benefits, AnMBR technology presents two fundamental challenges: i) competition between sulfate-reducing bacteria (SRB) and methanogens (MA) for the same substrates reduces methane yields when treating influents with high sulfate contents (Durán et al., 2020a); ii) reduction of sulfate by SRB produces sulfide and hydrogen sulfide, which eventually could entail inhibition in anaerobic metabolism and needs to be considered in energy recovery and water reclamation processes; and iii) dissolved methane losses through the effluent reduce energy recovery and, especially, increase GHG emissions (Jiménez-Benítez et al., 2020b). In this sense, co-digestion (e.g. the organic fraction of municipal waste, food waste, etc.) has been explored to increase influent organic load and thus increase methane production (Karki

et al., 2021; Moñino et al., 2017), while several promising technologies for dissolved methane recovery (e.g. PDMS membranes) are also being studied (Sanchis-Perucho et al., 2020; Crone et al., 2016).

AnMBR technology is thus a clear candidate for renewing the wastewater treatment approach and would contribute to the economic, environmental and social goals of water management, with CE playing an active role. Persevering with a business-as-usual scenario based on conventional technologies, which have been shown to be effective but not efficient, would entail ignoring the current global context: climate change, energy transition, resource depletion, and environment degradation.

This work describes the results of operating of an AnMBR semi-industrial plant (demonstration scale, TRL of 6) for MWW treatment at the Alcázar de San Juan WWTP (Alcázar de San Juan, Ciudad Real, Spain). The size of the prototype, together with being fed with real MWW and operated for more than 2.5 years, means it can be considered a reliable and robust source of performance data. The main objective of the study was to contrast AnMBR with conventional wastewater treatments and demonstrate its potential benefits.

5.2. Materials and methods

5.2.1. AnMBR description and operation

Figure 5.1 shows the process and instrumentation diagram (P&ID) of the semi-industrial AnMBR plant operated in this work. This plant mainly consists of a 40 m^3 (c.a. 34.4 m^3 working volume + 5.6 m^3 headspace) anaerobic reactor connected to three 0.8 m^3 membrane tanks (c.a. 0.7 m^3 working volume + 0.1 m^3 headspace). Each membrane tank was fitted with one ultrafiltration membrane system (PURON® PSH41, KMS, 0.03-µm pore size, total filtration area of 41 m^2), giving a total filtration area of 123 m^2. A sieve screw (RF, 1.5-mm screen size), an equalization tank (1.1 m^3) and a clean-in-place tank (0.37 m^3) were the other main elements in the plant. Total sludge volume was recorded considering the working volumes of both the anaerobic reactor and membrane tanks, accounting for 36.32±1.63 m^3 during the operating period. The AnMBR was equipped with a 2.1 m^2 degassing membrane unit for dissolved methane recovery, consisting of a polydimethylsiloxane (PDMS) hollow-fiber commercial module provided by PermSelect®, MedArray Inc. USA. The plant was fed with effluent from the pre-treatment step (sand and grease

removal) of the Alcázar de San Juan WWTP (Alcázar de San Juan, Ciudad Real, Spain). Further details of this AnMBR system can be found in Robles et al. (2020).

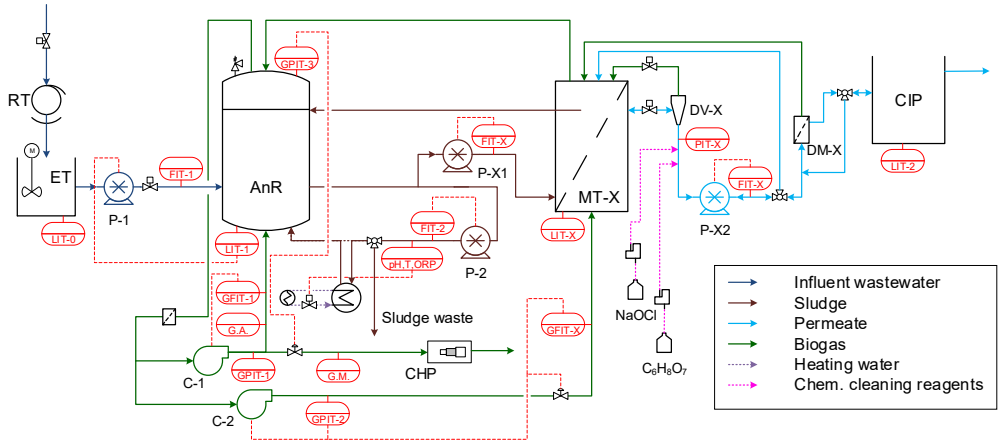

Figure 5.1. Process and Instrumentation Diagram (P&ID).

Regarding filtration process, the system was able to operate with transmembrane flux in the range 14-21 LMH, with moderate sparging gas demand (11-14 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) and significant total suspended solids (TSS) (8-13 $g \cdot L^{-1}$). Transmembrane pressures varied between 87 and 490 mbar and fouling rates between 1.7 and 16.6 $mbar \cdot d^{-1}$. Only two chemical cleanings of membrane were needed during the considered experimental period.

The semi-industrial plant performance was assessed from two points of view: (i) considering the 610 days of operations included in this work; and (ii) selecting 5 periods from the overall operating period. It is important to highlight the variable influent quality and the relatively high COD and BOD concentrations due to the contributions of several nearby dairy and wine industries. During the operation period and based on data obtained in previous AnMBR pilot plant studies (Giménez et al., 2014, 2012; Seco et al., 2018), SRT was maintained at 70 days and the plant was operated at two different HRT (25-26 and 41 hours). Temperature varied freely between 10 and 27 ºC (ambient temperature operation), while OLR was in the range between 0.60±0.20 and 1.28±0.38 kg $COD \cdot m^{-3} \cdot d^{-1}$. Table 5.1 shows the characterization of the influent and effluent streams together with the main operating conditions during the five selected periods.

Table 5.1. Influent and effluent characterization and operation conditions during the studied periods.

INFLUENT	Units	I	II	III	IV	V
$Q_{treatment}$	$L \cdot d^{-1}$	21228±591	33016±2773	33243±3118	34207±1478	20984±7482
TSS	$mg \cdot L^{-1}$	462±188	418±293	332±145	354±116	430±120
VSS	%	64.6±5.9	82.0±11.5	63.1±14.6	75.5±9.6	80.1±13.0
COD	$mg \cdot L^{-1}$	1285±429	1403±532	896±201	755±224	1038±238
OLR	$kg\ COD \cdot m^{-3} \cdot d^{-1}$	0.75±0.25	1.28±0.38	0.82±0.19	0.71±0.23	0.60±0.20
BOD_5	$mg \cdot L^{-1}$	772±277	911±288	569±122	451±154	581±180
N_T	$mg\ N \cdot L^{-1}$	52.0±13.3	54.7±16.3	46.8±6.9	46.0±11.2	35.6±8.0
P_T	$mg\ P \cdot L^{-1}$	10.1±2.8	8.6±2.1	8.5±1.2	6.8±1.3	7.6±1.1
PO_4^{3-}-P	$mg\ P \cdot L^{-1}$	2.5±0.0	5.8±2.1	6.0±0.9	5.3±1.3	3.9±1.7
S^{2-}	$mg\ S \cdot L^{-1}$	37.9±26.7	59.6±19.5	47.0±38.7	30.7±49.2	38.3±14.5
SO_4^{2-}-S	$mg\ S \cdot L^{-1}$	147.7±13.3	172.2±28.5	125.4±47.6	157.3±46.9	149.7±26.9
COD:SO_4^{2-}-S	$g\ COD \cdot g^{-1}\ SO_4^{2-}$-S	8.70±3.69	8.15±4.44	7.14±4.31	4.80±2.86	6.94±2.83
EFFLUENT	**Units**	**I**	**II**	**III**	**IV**	**V**
COD	$mg \cdot L^{-1}$	91±31	116±24	121±15	80±17	79±18
N_T	$mg\ N \cdot L^{-1}$	47.9±6.2	54.0±14.6	47.3±4.3	52.5±10.2	37.0±11.1
P_T	$mg\ P \cdot L^{-1}$	8.8±1.9	9.9±1.9	6.9±2.8	6.3±0.5	7.8±1.2
S^{2-}	$mg\ S \cdot L^{-1}$	174.3±28.7	207.5±73.2	233.1±66.3	183.2±27.0	166.2±27.9
VFA	$mg\ COD \cdot L^{-1}$	8.8±13.7	21.1±45.0	41.3±27.6	0.0±0.0	5.4±10.9
CH_4	$mg \cdot L^{-1}$	16.0±0.5	17.1±0.8	17.9±0.3	16.3±0.5	18.7±0.6
REACTOR	**Units**	**I**	**II**	**III**	**IV**	**V**
SRT	d	70±1	68±2	70±2	71±2	70±0
HRT	h	41±1	25±1	26±2	26±2	41±13
T	ºC	27±1	24±2	19±1	27±1	18±2
TSS	$g \cdot L^{-1}$	8.4±0.5	12.6±0.5	11.3±1.0	10.4±0.8	8.3±1.2
VSS	%	70.3±5.5	68.1±1.8	72.8±3.8	65.7±2.7	68.9±5.9
pH		7.1±0.1	7.0±0.1	7.0±0.1	7.0±0.1	7.0±0.3
Q_{WS}	$L \cdot d^{-1}$	522±5	511±107	521±5	510±18	520±4
COD	$g \cdot L^{-1}$	11.4±0.6	15.8±1.1	14.7±2.0	13.2±0.5	13.0±0.4
Q_{biogas}	$L\ STP \cdot d^{-1}$	5396±4435	5558±3204	1901±597	1359±999	2241±924
$\%CH_4$	%	77±2	77±2	72±2	77±2	76±1

5.2.2. AnMBR monitoring

The following parameters were regularly monitored in the influent: total COD and BOD_5, sulfate (SO_4^{2-}-S), sulfide (HS^--S), total nitrogen (N_T), total phosphorus (P_T), nitrate (NO_3^--N), phosphate (PO_4^{-3}-P), TSS, and volatile suspended solids (VSS). Effluent characterization comprised total COD, N_T, P_T and HS-S, and volatile fatty acids (VFA). Finally, reactor monitoring was based on TSS, VSS, COD, pH, temperature and methane content in the biogas (% CH_4). Dissolved methane in the effluent was calculated by Henry's law and assuming saturation conditions.

pH, temperature and biogas flow rate were registered online. 24-hour-composite samples were taken from the influent twice a week for all measurements except for COD, which was analyzed daily. Grab samples were taken twice a week from the effluent and mixed liquor.

5.2.3. Analytical methods

TSS, VSS, COD, BOD_5, sulfate, sulfide, and nutrients were determined according to Standard Methods (APHA, 2005). VFA concentration was determined by titration according to the method proposed by Moosbrugger et al. (1992).

5.2.4. Data processing and calculations

Different key performance indicators were calculated to evaluate the performance of the system as shown in Robles et al., (2020), i.e., methane yield taking into account the methane dissolved in the permeate, COD removal rates, and biosolids production.

5.3. Results and discussion

The performance of the demonstration plant was evaluated for 610 days. Five periods were further characterized to assess the robustness of the AnMBR-technology against the disturbances in the main operating conditions derived from the natural variations throughout the year.

5.3.1. Organic removal efficiency

Figure 5.2 shows the 15-day moving average for the influent organic load along with its 95% confidence bands, the daily average temperature, and both the removed and biodegraded COD. The influent organic load was highly variable, mainly due to the aforementioned variable industrial discharges. Biodegraded COD accounted for the COD that was biologically removed by SRB or was transformed into methane by MA, whereas the removed COD also considered the physically retained COD (by the ultrafiltration process) besides the biodegraded COD. Conversely, the influent sulfate-concentration variation was moderate, resulting in different COD:SO_4^{2-}-S ratios for each period (see Table 5.1). The temperature followed the natural variations in mid-latitude temperate climates, with strong variations between day to night (data not shown) and summer to winter. The daily-average temperature difference from winter to summer was higher than 17 ºC.

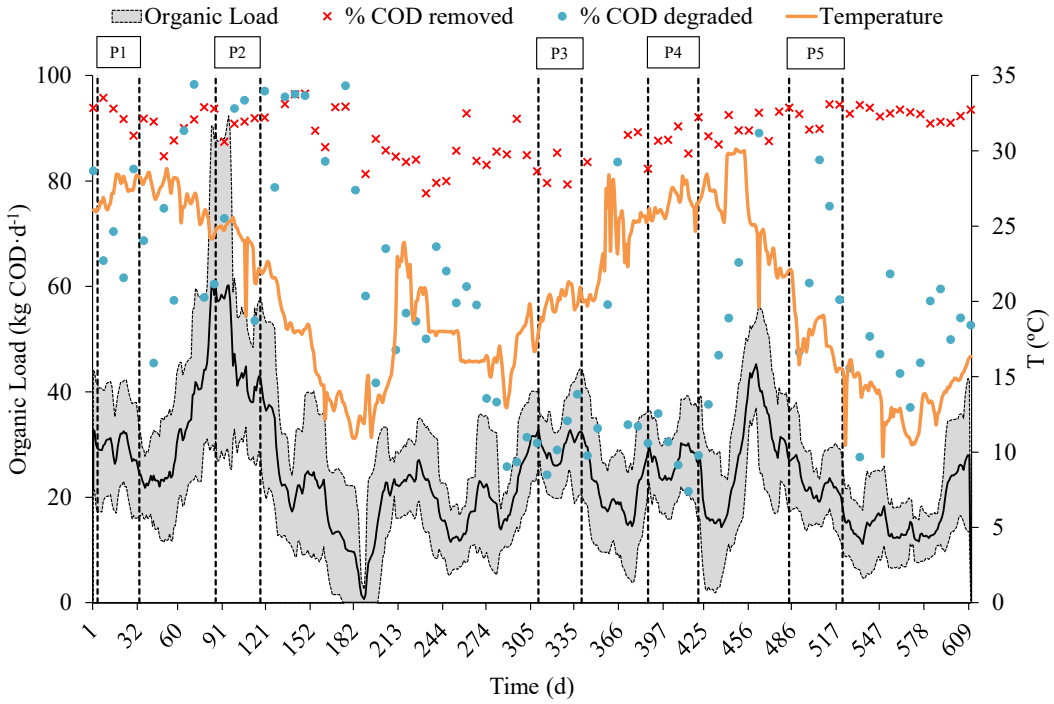

Figure 5.2. Operating conditions during the experimental period.

The HRT was varied to simulate population shifts during summer and winter periods as a result of population density changes during vacation periods. Initially, HRT was set to 41 hours in period P1, in accordance with the low population-density in coastal areas before the summer. HRT was almost halved from P1 to P2 (25±1 h) and P4 (26±2 h) to simulate the intensification of water usage by the massive influx of tourists in coastal areas during vacation periods. Similarly, P3 y P5 were operated at the same low temperature (19±1 and 18±1 °C, respectively) but different HRT (25±1 h in P3 and 41±13 h in P5) to simulate population variation during the winter period. HRT was kept practically constant from period P2 to period P4 (around 25 h), representing densely populated all-year-long areas. However, as shown in Table 5.1, the OLR decreased (1.28±0.38 kg $COD \cdot m^{-3} \cdot d^{-1}$ in P2, 0.82±0.19 kg $COD \cdot m^{-3} \cdot d^{-1}$ in P3 and 0.71±0.23 kg $COD \cdot m^{-3} \cdot d^{-1}$ in P4) as a result of the drop in the influent COD concentrations (1403±532 mg $\cdot L^{-1}$ in P2, 896±201 mg $\cdot L^{-1}$ in P3 and 755±224 mg $\cdot L^{-1}$ in P4), which was attributed to changes in the activity of the industries that discharge into the sewer system. The sulfate concentration also decreased from period P2 (172.2±28.5 mg $S \cdot L^{-1}$) to period P3 (125.4±47.6 mg $S \cdot L^{-1}$), albeit the COD reduction was more significant, resulting in a lower COD:SO_4^{2-}-S ratio. Conversely, the sulfate concentration in period P4 (157.3±46.9 mg $S \cdot L^{-1}$) was higher than in period P3 despite the fact that COD concentration continued to fall, both contributing to a significant reduction in the COD:SO_4^{2-}-S ratio. SO_4^{2-}-S concentration in period P5 (149.7±26.9 mg $S \cdot L^{-1}$) remaining similar to P4. The increased influent COD concentration in

period P5 (1038±238 mg·L^{-1}) thus resulted in a higher COD:SO_4^{2-}-S ratio. Despite the higher COD concentration, OLR was lower in period P5 (0.60±0.20 kg COD·m^{-3}·d^{-1}) than in period P4 as a result of the higher HRT in period P5.

Figure 5.2 shows that COD removal accounted for an average of 87.2 ± 6.1 % of the influent COD. 87 % of the COD removal data was higher than 80 %, whilst almost 40 % of the COD removal data was over 90 %. The membranes were able to virtually retain all particulates. However, the occasional unbalanced performance of the different stages in anaerobic degradation due to low temperatures or organic load peaks accumulated intermediate soluble organics that were able to pass through the membrane, thus increasing the effluent COD and reducing COD removal.

COD was not completely biodegraded by SRB or by MA. Conversely, a fraction of the removed COD was simply retained within the system due to membrane filtration, helping to increase the solids concentration and therefore waste-sludge production. Regarding the biologically removed COD, sulfate concentration was virtually zero in the effluent, indicating that SRB completely reduced influent sulfate (2 grams of COD were consumed per gram of SO_4^{2-}-S reduced). The analysis of the variance ANOVA of the COD consumed by SRB (expressed as OLR-SRB, i.e., the influent OLR which is consumed by SRB) found a significant difference between the mean values in the different periods (P-value = 0.0) (see complementary material). Nevertheless, Fischer's least significant difference (LSD) method for the pairwise comparison of mean values in the different periods were fairly similar in the mean values for the OLR-SRB among periods P1 and P5, and between periods P2 and P4. While P3 had an intermediate mean value.

The remaining COD was theoretically available for MA (i.e., OLR-MA) in the proportion shown in Figure 5.3. As can be seen in this figure, the OLR in period P2 was much higher than the others, as a result of the strengthened influent wastewater organic load. During P2, 24.5 % of the OLR was consumed by SBR (OLR-SBR) while 75.5 % remained available for MA (OLR-MA). On the other hand, P4 showed the lowest OLR-MA (58.3 %). The P-value for the analysis of the variance of the OLR-MA (see complementary material) in each period was lower than 0.05 (P-value = 0.0) again indicating a significant difference between the mean values in the different periods. In this case, Fischer's LSD method found significant similarity between mean values of P1 and P3, and between P4 and P5. P2 had the highest mean value for OLR available for MA due to the high treatment flow and influent COD concentration.

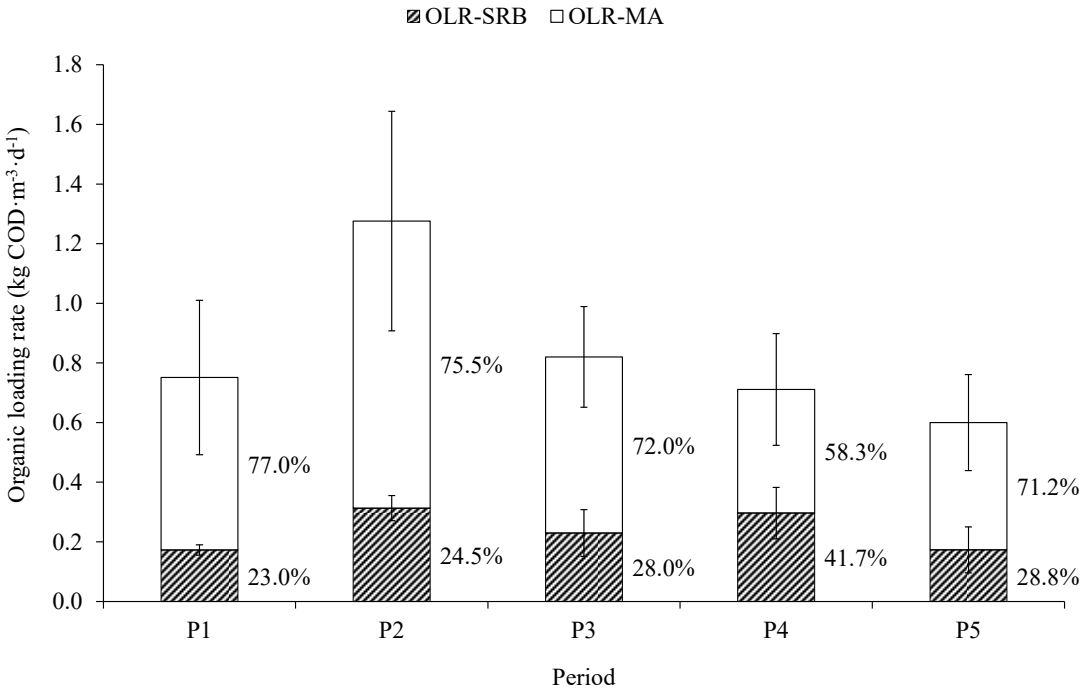

Figure 5.3. Organic loading rate consumed by SRB and available for MA.

The system's response to organic-load peaks entailed an accumulation of VFA, likely due to the faster response of acid-generating than acid-consuming organisms, although the system was able quickly restore the equilibrium. The VFA effluent concentrations were low in the periods with longer HRT (P1 and P5) and moderate organic overloads. Also, higher operating temperatures were associated with lower VFA concentrations (P1, P2 and P4). In contrast, the combination of short HRT and lower temperature of P3 led to more severe organic overloading and hindered accumulated VFA consumption, resulting in the highest VFA effluent concentrations (see Table 5.1). This result is attributed to the more severe impairment of acid consuming organisms, which seemed to be more affected by lower temperatures than acid-producing organisms.

5.3.2. Methane production

Table 5.2 shows the percentages of the COD removed by filtration and biodegradation and biodegraded COD. As aforementioned, the COD removed was higher in periods P1 (93.1±6.3 %) and P5 (92.5±3.5 %), when HRT was higher (41±1 and 41±13 h, respectively), indicating that the extended contact time between soluble organics and anaerobic microorganisms enabled MA to further consume the organic matter and produce CH_4, CO_2. The absence of intermediate products

indicates that the intermediate anaerobic degradation steps were properly balanced, hydrolysis and solubilization being the limiting steps.

The percentage of biodegraded COD in P1 and P2 was higher than the rest of the periods (71.3±27.7 % and 74.4±14.5 %, respectively) due to their high influent COD concentration (1285±429 mg COD·L^{-1} in P1 and 1403±532 mg COD·L^{-1} in P2) and temperature (27±1 °C and 24±2 °C, respectively), which favored bacterial metabolism and hence organic matter consumption. P4, with 63.4±26.2 % of biodegraded COD, also presents a high operating temperature (27±1 °C) but, also the lowest influent COD concentration (755±224 mg COD·L^{-1}). The percentage of biodegraded COD in P4 is similar to those obtained in P3 (61.0±16.2 %) and P5 (61.4±13.0 %), operated at low temperatures (18±2 °C and 19±1 °C, respectively), but with higher influent COD (896±201 mg COD·L^{-1} in P3 and 1038±238 mg COD·L^{-1} in P5).

Figure 5.4 depicts the time evolution of the 15-day moving average biogas production along with its 95 % confidence bands. As can be seen, the biogas production trend was directly related to the fraction of COD load available for MA (OL-MA), although it was also influenced by temperature, which limited the production of substrates for SRB and MA as a result of the reduction in the hydrolysis and solubilization rates.

The capacity of the system to recover energy was evaluated in terms of methane yield (Y^{CH_4}), which stands for the amount of methane produced per unit of influent COD. The biomethane potential (BMP) representing the fraction of the influent COD that ended up as methane (by comparing Y^{CH_4} to the theoretical maximal methane production, i.e., 350 STP L CH_4·kg^{-1} COD) was also calculated. The resulting Y^{CH_4} and BMP in the different periods are shown in Table 5.2. However, only the results regarding Y^{CH_4} will be discussed, since the BMP results followed the same trend.

Table 5.2. Percentage of COD removed and biodegraded, methane yield (Y^{CH_4}), biomethane potential (BMP) per influent COD and waste sludge production (WSP) per kg of influent and removed COD.

Period	COD_{rem} (%)			$COD_{biodegraded}$ (%)			Y^{CH_4} (STP L $CH_4 \cdot kg^{-1}$ COD)			BMP (%)			WSP (kg VSS·kg^{-1} COD_{inf})			WSP (kg VSS·kg^{-1} COD_{rem})		
	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD	Mean	±	SD
P1	93.1	±	6.3	71.3	±	27.7	169.0	±	95.1	48.3	±	28.3	0.112	±	0.045	0.121	±	0.067
P2	91.9	±	2.7	74.4	±	14.5	109.6	±	70.0	31.3	±	22.1	0.095	±	0.032	0.103	±	0.044
P3	86.7	±	4.7	61.0	±	16.2	75.2	±	16.9	21.5	±	4.8	0.144	±	0.050	0.166	±	0.074
P4	89.5	±	4.9	63.4	±	26.2	70.2	±	36.0	20.1	±	10.8	0.135	±	0.055	0.150	±	0.079
P5	92.5	±	3.5	61.4	±	13.0	102.3	±	63.8	29.2	±	18.2	0.137	±	0.061	0.148	±	0.072

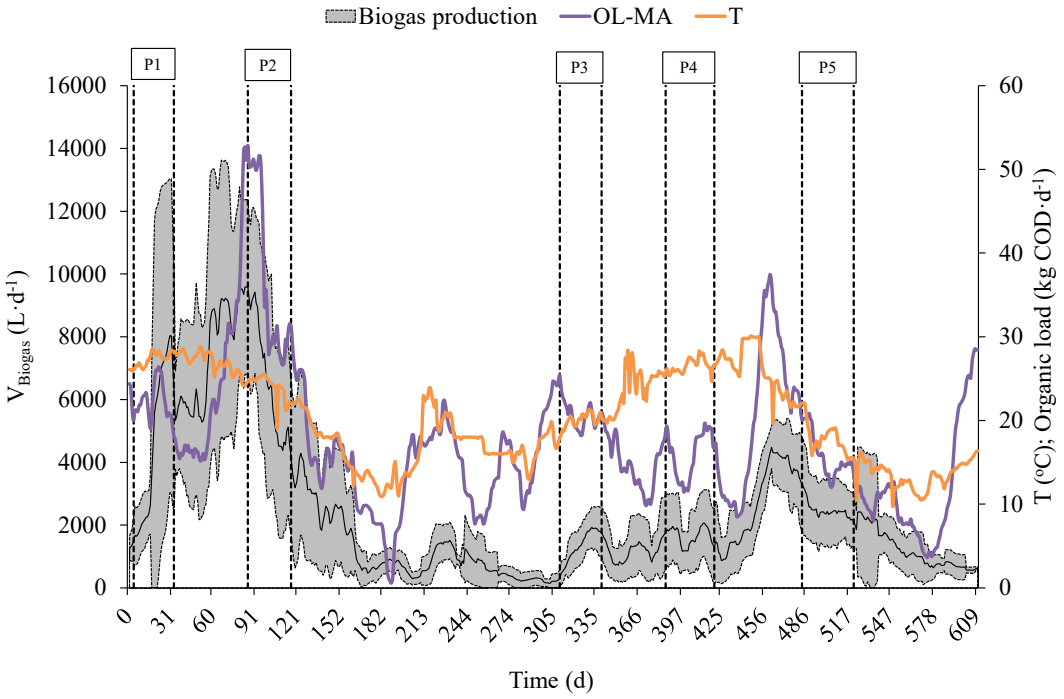

Figure 5.4. Time evolution of the biogas production trend, temperature and organic load available for MA.

The highest Y^{CH_4} value was obtained in P1 (169.0±95.1 STP L CH_4·kg^{-1} COD) with a combination of high T (27±1 ºC) and HRT (41±1 h). Its influent COD:SO_4^{2-}-S ratio was also the most favorable for methane production (8.70±3.69 COD·g^{-1} SO_4^{2-}-S). While a high temperature enhanced the hydrolysis rate, which is considered the limiting stage in anaerobic digestion, methane production increased with the COD available for MA, for which P1 had the highest percentage of influent COD available for MA (77.0 %, see Figure 5.3). In contrast, the slight temperature drop and reduced HRT reduction between periods P1 and P2 (24±2 ºC and 25±1 h, respectively) helped to reduce the Y^{CH_4} to 109.6±70.0 STP L CH_4·kg^{-1} COD, even when Fischer's LSD method showed that the amount of COD for MA in P2 was the highest (35 kg·d^{-1}). It is important to highlight that an error of 18.5 % was obtained when COD mass balance was applied to P2 (see Figure 5.5 and discussed in section 5.3.5). This COD loss could be attributed to underestimated biogas production. As mentioned above, dissolved methane in the effluent was calculated assuming saturation conditions, although there could have been oversaturation episodes (Pauss et al., 1990).

P3 and P4 present the lowest Y^{CH_4} values (75.2±16.9 and 70.2±36.0 STP L CH_4·kg^{-1} COD, respectively). These results can also be explained by the temperature, HRT and COD:SO_4^{2-}-S

ratio. Despite P3 being operated with a moderate influent COD:SO_4^{2-}-S ratio (7.14±4.31 g COD·g^{-1} SO_4^{2-}-S) and available organic matter (72.0 %), influent COD could not offset its low operating temperature (19±1 °C) and HRT (26±2 h), which hindered hydrolysis and possibly also methane production. In P4, the operating temperature was favorable (27±1 °C) and HRT remained at 26±2 h, but the COD:SO_4^{2-}-S ratio was significantly lower than in the other periods (4.80±2.86 g COD·g^{-1} SO_4^{2-}-S), which justifies the lower Y^{CH_4}.

Finally, similar Y^{CH_4} values were obtained for P2 and P5 (109.6±70.0 and 102.3±63.8 STP L CH_4·kg^{-1} COD, respectively). In this case, P2 had high temperature (24±2 ºC) and high COD:SO_4^{2-}-S ratio (8.15±4.44 g COD·g^{-1} SO_4^{2-}-S) but lower HRT (25±1 h). P5 operated at high HRT (41±13 h), a moderate influent COD:SO_4^{2-}-S ratio (6.94±2.83 g COD·g^{-1} SO_4^{2-}-S) but the lowest temperature (18±2 °C). Considering that influent quality cannot be modified, HRT thus becomes the main design parameter for maximizing methane production in low winter temperatures.

It should be remembered that influent sulfate concentration depends on the region in which the wastewater is generated, since it is associated with the concentration of sulfate in water sources as well as the possible contribution of factories that discharge sulfur to the sewage system. Since the COD consumed by SRB is biodegradable and should have undergone at least the first stages of anaerobic degradation (i.e., hydrolysis and solubilization) to be available for SRB, it would also have been available for MA. A sulfate-free influent would thus have yielded a much higher Y^{CH_4} and BMP would have been similar to the percentage of biodegraded COD. As way of example, methane yields without considering the COD consumed by SRB in the studied periods, $Y_{SO_4}^{CH_4}$, would rise to 87.4-191.0 STP L CH_4·kg^{-1} COD. However, in sulphate-rich wastewater, the role of SRB in reducing the effluent-soluble COD when HRT is reduced must be considered, since SRB has higher degradation rates than MA, especially at lower temperatures.

5.3.3. Biosolids

The waste-sludge production (WSP) expressed in kg VSS per kg of influent COD and per kg of removed COD is shown in Table 5.2. The reported values are the outcome of a complex interaction between operational conditions and influent characteristics. The fraction and composition of particulates in the influent and their fate within the system, which depends on the operating conditions (temperature, HRT and especially SRT), influence the accumulation and

retention of biosolids and therefore increased WSP. A greater influx of solids into the system entails greater accumulation and retention unless the operating conditions favor hydrolysis and solubilization. On the other hand hydrolysis is widely accepted as the limiting step in anaerobic degradation and is severely affected by temperature drops (Batstone, 2006). Although temperature also affects the following steps of the anaerobic degradation, results indicated that intermediates accumulation can be mitigated as long as HRT is high enough. In the same way, WSP can also be reduced by increasing system SRT, thus extending the time for further hydrolysis and solubilization of the accumulated solids.

As can be seen in Table 5.2, P3 had the highest WSP (0.166±0.074 kg VSS·kg^{-1} COD_{rem}) mainly due to the low temperature (19±1 °C). The operating conditions hindered biodegradability and therefore raised sludge production. P4 and P5 also had high WSP (0.150±0.079 kg VSS·kg^{-1} COD_{rem} and 0.148±0.072 kg VSS·kg^{-1} COD_{rem}, respectively) although lower than P1. The high production in P5 can be attributed to the lowest operating temperature (18±2 °C). However, despite temperature in P4 being the highest (27±1 °C) the significant WSP in this period was associated with its low COD:SO_4^{2-}-S (4.80±2.86 g COD·g^{-1} SO_4^{2-}-S). In this period, the percentage of influent COD consumed by SRB accounted for 41.7 % while in the other periods was below 29 %. Since the SRB biomass yield was significantly higher than MA, the associated solids production was also higher, as was sludge production in this period (Durán et al., 2020a).

Finally, P1 and P2 had the lowest WSP (0.121±0.067 and 0.103±0.044 kg VSS·kg^{-1} COD_{rem}, respectively). Both were operated at high temperature (27±1 °C in P1 and 24±2 °C in P2) and had the highest COD:SO_4^{2-}-S ratio (8.70±3.69 g COD·g^{-1} SO_4^{2-}-S and 8.15±4.44 g COD·g^{-1} SO_4^{2-}-S, respectively). The combination of both variables enhanced MA organic matter biodegradation and reduced sludge production. P2 was also characterized by the highest percentage of influent VSS, COD and BOD_5, indicating the highest influent wastewater biodegradability and that the operating conditions were appropriate for the maximum biosolids degradation. It is important to highlight that, besides the complex interaction of factors that affect the WSP, it can be as low as 0.103±0.044 kg VSS·kg^{-1} COD_{rem}.

As a way of comparison, Foladori et al., (2015) reported 0.23-0.35 kg VSS·kg^{-1} COD_{rem} (SRT of around 10-20 days) and 0.18 kg VSS·kg^{-1} COD_{rem} for aerobic treatment without and with sludge stabilization, respectively. According to these values, all the selected periods had lower WSP than

the aerobic treatments with percentage reductions between 8 % (P3) and 43 % (P2). Increasing SRT over 70 days could be expected to lead to lower WSP and additional biosolids stabilization, although solids concentration in the reactor would also rise and affect processes such as membrane filtration in the operating stage or increase reactor capacities' requirements in the design stage. In this regard, Ji et al., (2020) obtained a WSP of 0.09 kg VSS·kg^{-1} COD_{rem} operating two different AnMBR with different pore size membranes for 120 days at 25ºC. In this work no excess sludge was wasted during long-term operations except for the sludge samples. This value confirms that it is possible to achieve low WSP with high SRTs. On the other hand, with the same technology Seco et al., (2018) reported a WSP of 0.218 kg VSS·kg^{-1} COD_{rem} with an SRT of 140 days and 27ºC. Design and operating conditions thus need to be carefully selected taking influent characteristics into account and especially the particulate COD and COD:SO_4^{2-}-S ratio.

5.3.4. Nutrients recovery potential

Anaerobic treatments have low nutrients requirements per COD removed, as can be seen from the effluent characterization shown in Table 5.1, which confirms that AnMBR is promising technology for turning wastewater into a source of nutrients. N_T and P_T concentrations in the effluent accounted for 37.0-54.0 mgN·L^{-1} and 6.3-9.9 mgP·L^{-1}, respectively. Considering the treatment capacity of "Alcázar de San Juan" WWTP (16000 m^3·d^{-1}), these concentrations represent 216 – 315 t N·year^{-1} and 37 – 58 t P·year^{-1}. It should be noted that over 90 % of European phosphorus applications are import-dependent, so that wastewater can play an active role as a P supplier by combining AnMBR and CE fundamentals (Rosemarin et al., 2020). Selecting urea ($CO(NH_2)_2$) and P_2O_5 as representative N-based and P-based fertilizers, respectively, nutrients equivalent to 463-676 t Urea·year^{-1} and 84-132 t P_2O_5·year^{-1} could be provided by effluent. If a price of €273.6 per t of urea (www.indexmundi.com) and €783.0 per t of P_2O_5 (www.echemi.com) is assumed, the mineral-fertilizer equivalent-value of the nutrients in the discharge flow would be between k€162 and k€230 per year, and would pay for 10-15% of water treatment costs at €0.269 per m^3 for AnMBR (Jiménez-Benítez et al., 2020a).

To reduce production costs and impact, an alternative embedded nutrient strategy consists of direct reuse (fertigation) instead of producing fertilizers from effluent, which makes AnMBR technology especially suitable for decentralized plants in rural areas where the effluent can be discharged directly to farmland. It is important to highlight that apart from a nutrient-rich stream the combination of an anaerobic process with UF membrane implemented in an AnMBR produces an effluent free of solids and pathogens (Krzeminski et al., 2017b), which makes it especially

appropriate for fertigation. For example, "Alcázar de San Juan" WWTP could cover an annual winter wheat crop (productivity 6.7 t grain·ha^{-1}) of 1080-1575 ha attending to N needs (0.2 t·ha^{-1}) and 1527-2400 ha if P_2O_5 requirements are considered (0.055 t·ha^{-1}) (Roy et al., 2006).

5.3.5. COD mass balance and overall performance considerations

Figure 5.5 shows the COD mass balance in the periods studied. Each sector of the circles depicts the fraction of the influent COD (i.e., COD biodegraded by SRB, COD associated with biogas, COD from dissolved methane, COD wasted with sludge, COD in the effluent and mass balance error).

P1 shows no mass balance error but periods P2 to P5 have positive values in all cases indicating that the influent COD is higher than COD leaving the system through biogas, effluent, wasted sludge and COD biodegraded by SRB. The errors in P4 (1.7 %) and P5 (3.3 %) are negligible, while P2 and P3 show significant errors (18.5 % and 11.6 %, respectively). As all the parameters involved in COD mass balance (e.g., flow rates, COD concentration, biogas production, waste sludge, etc.) are based on analytical and/or on-line measurements except for dissolved methane (calculated as stated in Section 5.2.2), it is hypothesized that in these two periods dissolved methane in the effluent have been underestimated. As mentioned above, dissolved methane is calculated considering saturation conditions based on Henry's law, although oversaturation could take place in these periods due to methane bubbles passing through the membranes or poor reactor mixing.

Figure 5.5 confirms the influence of operating at high T, HRT and COD:SO_4^{2-}-S ratio to enhance methane production, with P1, P2 and P5 showing the highest percentages of biogas associated to COD. P1, operated at the highest temperature, HRT and COD:SO_4^{2-}-S ratio (27±1 °C, 41±1 h and 8.70±3.69 g COD·g^{-1} SO_4^{2-}-S, respectively), shows a percentage of 43.5 % of biogas-associated COD, the highest value. P2 was operated at high temperature (24±2 °C) and a favorable COD:SO_4^{2-}-S ratio (8.15±4.44 g COD·g^{-1} S). Both conditions could offset the lower HRT (25±1 h) and kept biogas production at significant percentages (26.5 %). If the COD mass balance error (18.5 %) is attributed to underestimated methane production, the biogas percentage could rise to 45 %, as in P1. P5 also presented significant biogas production (22.2 %). This period operated at high HRT (41±1 h) and moderate influent COD:SO_4^{2-}-S ratio (6.94±2.83 g COD·g^{-1} SO_4^{2-}-S), but the lowest T (18±2 °C), which hindered methane production.

Since a negligible error was obtained for this period, potential increased biogas production due to underestimated dissolved methane cannot be considered.

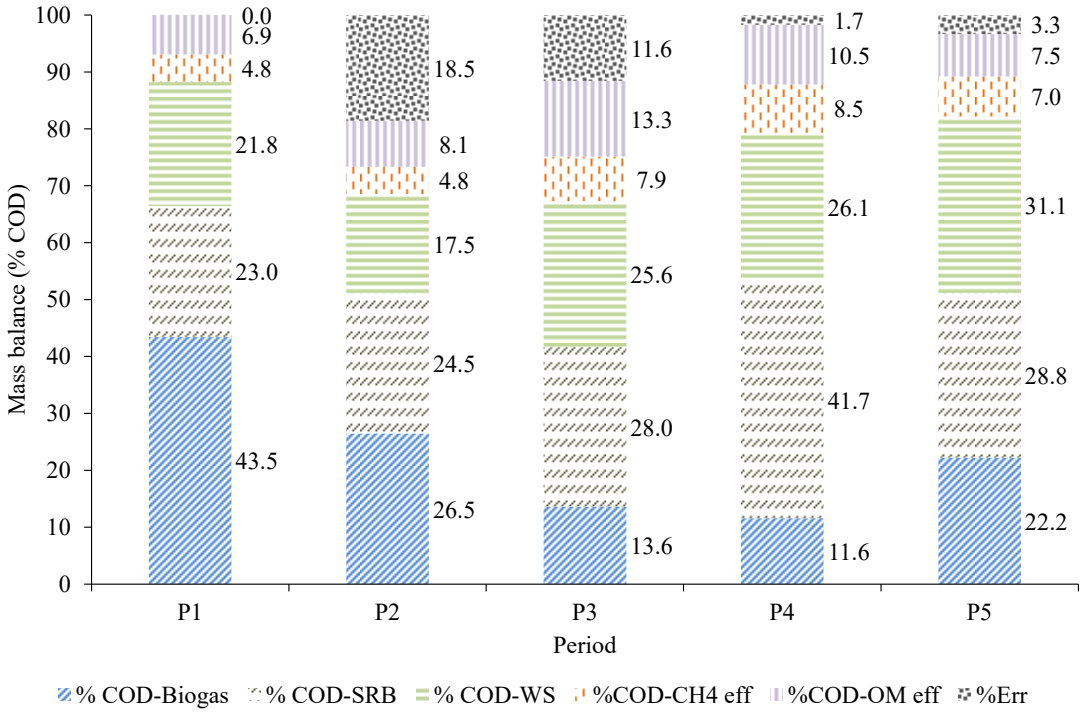

Figure 5.5. COD mass balance.

The lowest percentages of COD associated with biogas were obtained in P3 (13.6 %) and P4 (11.6 %). P3 operated at a low temperature (19±1 °C) and HRT (26±2 h) and therefore with low biogas production despite a favorable influent COD:SO_4^{2-}-S ratio (7.14±4.31 g COD·g^{-1} SO_4^{2-}-S), similar to P5. However, if mass balance error is considered (11.6 %), biogas-associated COD could rise to 25.2 %. P4, with low HRT (26±2 h) and the lowest influent COD:SO_4^{2-}-S ratio (4.80±2.86 g COD·g^{-1} SO_4^{2-}-S) had the lowest COD percentage associated with methane production (11.6 %), despite its favorable operating temperature (27±1 °C).

For the COD associated with waste sludge, Figure 5.5 shows that P5 had the highest percentage (31.1 %). This period operated at the lowest T (18±2 °C), which hindered particulate degradation, and the lowest OLR (0.60±0.20 kg COD·m^{-3}·d^{-1}). Since OLR before P5 was higher than in this period (see Figure 5.2), the sludge wasted during P5 could have been influenced by accumulated VSS. P2 had a percentage of 17.2 %, due to its favorable operating temperature (24±2 °C) and the highest OLR (1.28±0.38 kg COD·m^{-3}·d^{-1}). P3 and P4 show a similar percentage associated

with waste sludge (25.6 % and 26.1 %, respectively), despite operating at very different temperatures (19±1 °C in P3 and 27±1 in P4 °C). This can be attributed to the higher COD percentage consumed by SRB (41.7 %) in P4. As these bacteria have a higher growth yield than MA and therefore increase solids production in the reactor.

The results show an overall promising performance of AnMBR technology for treating MWW at ambient temperature. The COD removal capacity allows compliance with treatment regulation (e.g., MWWTD); methane yields indicate its potential for organic matter valorization and energy recovery; and nutrient content in the effluent indicates its capacity for turning wastewater into a valuable source of nutrients. Low waste-sludge production means AnMBR can be considered a robust candidate as the new core technology to boost the transition of wastewater treatment plants towards the new resource recovery facilities, especially suitable for small and decentralized plants in which anaerobic processes are not currently applied.

Temperature, HRT, SRT and COD:SO_4^{2-}-S ratio are crucial parameters to improve technological, economic and environmental performance of AnMBR technology for wastewater treatment, HRT and SRT being the only ones which do not depend on ambient conditions. Both HRT and SRT thus appear as the main design parameter. It is important to highlight the benefits that the implementation and optimization of dissolved methane recovery technologies (e.g., hollow-fiber membranes) can provide to AnMBR: dissolved methane will improve the wastewater treatment energy balances while the carbon footprint associated with greenhouse gas emissions is minimized.

5.4. Conclusions

An industrial AnMBR prototype was operated for 610 days during which an average of 87.2 ± 6.1 % of influent COD removal was achieved, with 87 % of the data over 80 % and almost 40 % over 90 %. UF membranes contributed to wastewater treatment by retaining almost all the particulates. In addition, in the long-term operations, 5 periods were selected to evaluate the performance of the AnMBR. In these periods, a Y^{CH_4} between 70.2 and 169.0 STP L $CH_4 \cdot kg^{-1}$ COD were achieved and methane percentages in the biogas varied from 75 to 77 %. Sludge production ranged between 0.103±0.044 and 0.166±0.074 kg VSS·kg^{-1} COD_{rem}, which represents a reduction of 9 to 43 % compared to CAS. A significant nutrient recovery potential was also identified.

Finally, temperature, HRT and COD:SO_4^{2-}-S ratio jointly with SRT are crucial parameters to take into account in maximizing AnMBR's technical, economic and environmental performance. However, since temperature and influent COD:SO_4^{2-}-S depend on ambient conditions, HRT could be considered the main design parameter jointly with SRT.

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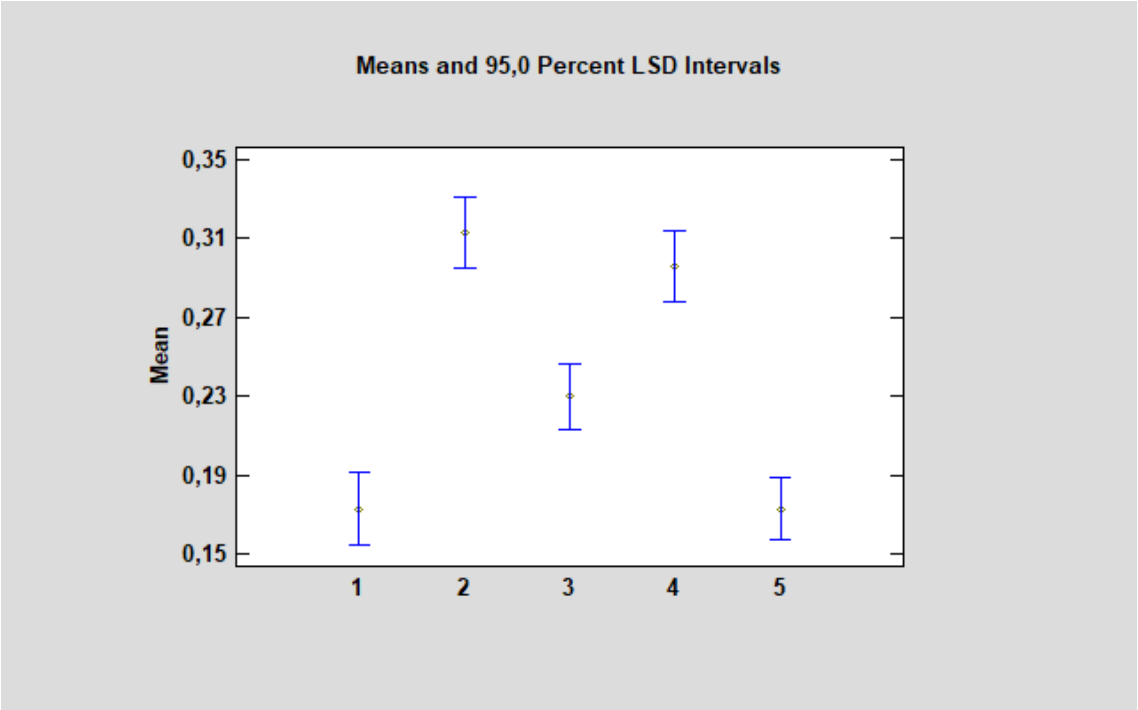
5.5. Supplementary material

5.5.1. Analysis of the variance ANOVA of the COD consumed by SRB

Source	Sum of Squares	Df	Mean Square	F-Ratio	P-Value
Between groups	0,497035	4	0,124259	27,65	0,0000
Within groups	0,633656	141	0,00449401		
Total (Corr.)	1,13069	145			

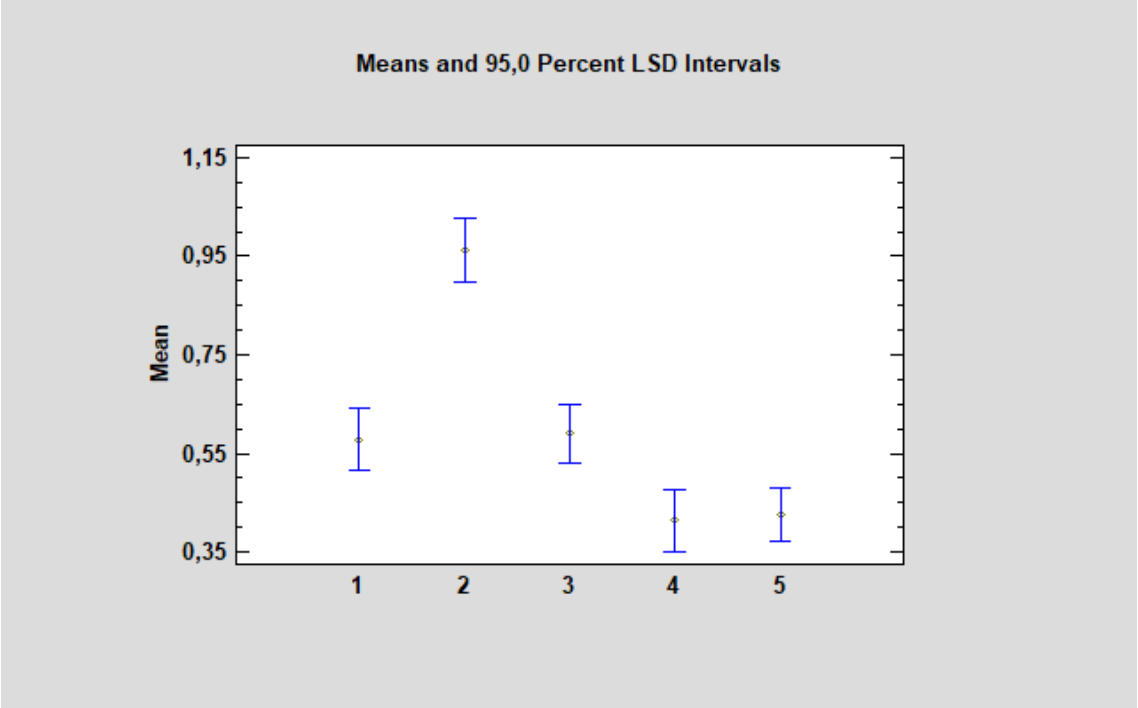
5.5.2. Fischer's least significant difference (LSD) method for the COD consumed by SRB



5.5.3. Analysis of the variance ANOVA of the COD available for MA

Source	Sum of Squares	Df	Mean Square	F-Ratio	P-Value
Between groups	5,39431	4	1,34858	24,55	0,0000
Within groups	7,74563	141	0,0549336		
Total (Corr.)	13,1399	145			

5.5.4. Fischer's least significant difference (LSD) method for the COD available for MA



6. A semi-industrial AnMBR plant for urban wastewater treatment at ambient temperature: analysis of the filtration process, energy balance and quantification of GHG emissions

Abstract

A semi-industrial scale AnMBR urban wastewater treatment plant was operated for 580 days at ambient temperature (ranging from 10-30 °C) to assess its long-term filtration performance, energy balance and GHG emissions. The applied 20ºC-standardized transmembrane flux (J_{20}) was varied between 15 and 25 LMH and the specific gas demand per m^2 of membrane (SGD_m) was modified between 0.10 to 0.40 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to a specific gas demand per permeate volume (SGD_P) between 10 to 20 $Nm^3 \cdot m^{-3}$). The filtration strategy allowed successful long-term operations without any chemical cleaning requirements and little fouling for 233 days. The plant operated as a net energy producer for more than 50 % of the experimental period, with an average net energy demand of -0.169±0.341, -0.190±0.376 and -0.205±0.447 $kWh \cdot m^{-3}$, considering 0 %, 50 % and 70 % of dissolved methane recovery, respectively. Finally, demethanization of AnMBR effluent is needed to achieve an environmentally sustainable operation of the technology. Therefore, the combination of AnMBR with degassing membranes appears as a suitable alternative to conventional wastewater treatment.

Keywords

Anaerobic membrane bioreactor; Ultrafiltration process; demonstration plant; energy balance; Greenhouse gas emissions

Highlights

Urban wastewater can be treated by AnMBR technology at low ambient temperatures.
The process was net energy producer (energy surplus of 0.169 $kWh \cdot m^{-3}$ in average).
Competitive J_{20} were reached (15-20 LMH) with low SGD_m values (0.1-0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$).
Energy consumption in reactor mixing and membrane scouring was < 0.19 $kWh \cdot m^{-3}$.
GHG emissions below 0.2 $kgCO_2 \cdot m^{-3}$ when 70 % of dissolved methane was recovered.

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6.1. Introduction

Although conventional wastewater treatment is usually based on aerobic processes (with conventional activated sludge (CAS) or an aerobic membrane bioreactor (MBR)), anaerobic membrane bioreactors (AnMBRs) are receiving increasing attention due to their advantages (Robles et al., 2021) including: i) organic matter valorization through methane production during anaerobic digestion; ii) a high-quality solid-free effluent production; iii) nutrients are conserved for recovery from the effluent; iv) operations at high sludge retention time (SRT), since membranes decouple hydraulic retention time (HRT) and SRT; v) they can treat low-loaded streams at ambient temperature since the membranes guarantee solids retention and avoid biomass washout; vi) improved stabilization of biosolids and significant reduction in sludge production when long SRTs are applied; and vii) they make reduced layouts possible thanks to the settlers being substituted for compact membrane units.

AnMBR technology has been considered for the treatment of different industrial wastewater streams, such as textile, petrochemical, brewing or pharmaceutical industry effluents with satisfactory results (Chen et al., 2015; Hu et al., 2017). However, despite the potential benefits of AnMBR plants for treating urban wastewater (UWW), they still face bottlenecks that need to be overcome, including: i) membrane fouling; ii) eutrophication potential of the effluent when it is discharged in sensitive zones; iii) dissolved methane in the effluent (Velasco et al., 2018); and iv) competition between sulfate-reducing bacteria (SRB) and methanogens, which can affect methane yield and generate corrosive sulfide (Durán et al., 2020b).

Membrane fouling has been extensively identified as a key issue to be addressed before AnMBRs can be widely used for UWW (Krzeminski et al., 2017b). This phenomenon is governed by different interrelated factors. For instance, operating at high temperatures reduces the viscosity of the mixed liquor, which favors filtration but also involves the release of extracellular polymeric substances (EPS), which are related to fouling (Lin et al., 2009). Although anti-fouling strategies based on gas sparging have been reported as effective, they increase the filtration energy demand and introduce shear forces. Increased SRT can also reduce membrane fouling since it consumes EPS and SMP. However, high SRT increases the solids concentration and hinders the progress of filtration.

The loss of part of the methane production as gas dissolved in the effluent must also be addressed to reduce fugitive emissions, identified as one of the greatest contributions to GHG of this

technology (Smith et al., 2014), and to improve the energy performance of the facilities by recovering part of the methane produced (Vinardell et al., 2021). Among other technologies, degasification membranes (Sanchis-Perucho et al., 2020) and spray aeration towers are thus being studied for this aim.

AnMBR plants have been reported to have a lower energy consumption than CAS and MBR; Lazarova et al. (2012) reported energy demands of 0.25-0.60 $kWh \cdot m^{-3}$ for CAS and 0.50-2.50 $kWh \cdot m^{-3}$ for MBR. Pretel et al. (2013) reported AnMBR net energy production of 0.17-0.19 $kWh \cdot m^{-3}$ without considering dissolved methane recovery, while Jiménez-Benítez et al. (2020b) reported net energy production of up to 0.47 $kWh \cdot m^{-3}$ when treating high-loaded UWW at mild temperatures when dissolved methane was considered and sulfate content in the influent was 112-172 mg $S\text{-}SO_4 \cdot L^{-1}$. The recovery of organic matter through biogas production makes it possible to partially offset the indirect GHG emissions derived from its energy demand. Also, anaerobic processes do not emit nitrous oxide (N_2O), as do aerobic nitrification/denitrification processes (Rodriguez-Caballero et al., 2015). This is an important feature since the global warming potential of this gas has been estimated at 298 $kgCO_{2eq}$ per kg of N_2O (Solomon et al., 2007).

For all the above reasons, AnMBRs appear as a promising technology for the so-called *water resource recovery facility* concept (Guest et al., 2009), as opposed to the conventional urban wastewater treatment approach, which considers wastewater as a source of pollution.

This paper evaluates the performance of a semi-industrial AnMBR plant in terms of filtration process performance, energy balance and GHG emissions. The plant was fed with effluent from the pre-treatment of the "Alcázar de San Juan" wastewater treatment plant (Ciudad Real, Spain). The major novelties of this study include i) long-term operation of 580 days; and ii) technical characteristics and operation (real UWW, reactor volume of volume 40 m^3, commercial hollow-fiber membranes, industrial equipment, ambient conditions, etc.). These characteristics allowed obtaining robust data for scaling-up AnMBR technology to full-scale facilities, thus providing of useful information in terms of membrane performance, energy balance, and GHG emissions for the design and full-scale operation of AnMBR systems. The goal of the research was to determine the potential of AnMBR over conventional treatments by assessing its robustness against variations in the main operating conditions (temperature, organic load, treatment flow, etc.).

6.2. Materials and methods

6.2.1. AnMBR description

The semi-industrial AnMBR plant implemented in the study consisted mainly of the following components: an anaerobic reactor (40-m^3 total volume; 34.4-m^3 working volume); three membrane tanks (MT_A, MT_B and MT_C; 0.8-m^3 total volume each; 0.7-m^3 working volume each) equipped with an ultrafiltration membrane system (PURON® PSH41, KMS, 0.03-µm pore size, 41-m^2 filtration area per tank); a sieve screw (1.5-mm screen size); an equalization tank (1.1 m^3); and a clean-in-place (CIP) tank (0.37 m^3). By including three identical MTs in the system, the effect of applying different J_{20} and gas-assisted membrane scouring intensities on membrane fouling can be tested in parallel treating the same feed at the same ambient conditions. Further details of this system can be found elsewhere in Robles et al. (2020b) and in supplementary material. The plant was also equipped with a degassing membrane unit (2.1-m^2 filtration area) for dissolved methane recovery (PermSelect®, MedArray Inc. USA), using vacuum pressure as the driving force (for further details see Sanchis-Perucho et al. (2021)).

The AnMBR was fed with effluent from the pre-treatment of the "Alcázar de San Juan" wastewater treatment plant (Ciudad Real, Spain), consisting of screening, grit and grease removal. The average influent quality is shown in Table 6.1. Since the dairy and wine industries are important elements in the local economy, the influent wastewater was characterized by high concentrations of COD (1071±467 mg·L^{-1}), BOD_5 (710±274 mg·L^{-1}) and sulfate (140.3±42.5 mg SO_4-S ·L^{-1}).

Table 6.1. Average influent wastewater characteristics.

Parameter	Unit	Mean ± SD
TSS	mg TSS·L^{-1}	405 ± 215
Total COD	mg COD·L^{-1}	1071 ± 467
BOD_5	mg COD·L^{-1}	710 ± 274
Total Nitrogen	mg N·L^{-1}	48.2 ± 11.7
Total Phosphorus	mg P·L^{-1}	8.4 ± 2.4
Sulfate	mg SO_4-S·L^{-1}	140.3 ± 42.5

6.2.2. AnMBR operation

The evolution of HRT, temperature, and organic load available for methanogenesis (OL-MA) during the 580-days experimental operation can be found elsewhere in Robles et al. (2022) and

in supplementary material. Temperature varied freely in the 10-30 °C range according to ambient conditions and season and SRT was set to 70 d. Several periods with different average HRT were operated as follows: 40 h (days 1-32); 25 h (days 33-122); 46 h (days 123-170); 38 h (days 171-291); 26 h (day 292-500) and 70 h (days 501-580). The different HRT simulated population changes during the summer and winter periods. Eventually, HRT was stablished at 70 h in a final experimental stage in order to assess the treatment process performance at high HRT.

The number of membrane tanks in operation depended on the selected HRT, while OL-MA was affected by economic activity, mainly dairy and wine, the latter with a markedly seasonal contribution. The influence of the grape harvest and winemaking can be noted between August and October between days 60 (August) and 100 (October) and between days 430 (September) and 480 (October).

Membrane filtration and scouring took place within the following parameter range: the gross 20ºC-standardized transmembrane flux (J_{20}) was modified between 15 and 25 LMH and the specific gas demand per m^2 of membrane (SGD_m) was between 0.10 and 0.40 $Nm^3 \cdot m^{-2} \cdot h^{-1}$, resulting in a specific gas demand per permeate volume (SGD_P) of between 10 to 20 $Nm^3 \cdot m^{-3}$, (for further details of membrane operation see Robles et al. (2015)).

Membranes were operated according to a specific schedule involving a combination of different individual stages taken from a basic 300 s-filtration : 30 s-relaxation cycle. Besides classical membrane operating stages (filtration, relaxation, and back-flush), degasification and ventilation stages were also considered in the membrane operation. Degasification aims to recover the accumulated biogas in top of the dead-end, hollow-fiber membranes, since this accumulation of biogas reduces the effective filtration area. The degasification stage consists of a period of high flow-rate filtration that is carried out to enhance the filtration process efficiency by removing the accumulated biogas. On the other hand, during ventilation, permeate is pumped from the CIP tank to the membrane tank through the degasification vessel instead of through the membrane. The aim of the ventilation stage is to recover the accumulated biogas in the degasification vessel due to the extraction of gas bubbles during filtration.

The chemical membrane cleaning included a first washing phase with sodium hypochlorite (2000 ppm) for 8 hours followed by a second washing phase with citric acid (2000 ppm) for another 8 hours.

The size of the AnMBR plant and the type and quality of the wastewater (real urban wastewater) involved in this study can contribute to determine the conditions for the technical, economic and environmental feasibility of these technologies as an alternative to conventional wastewater treatment.

6.2.3. Streams characterization and analytical methods

The following parameters were regularly monitored in the influent: total chemical oxygen demand (COD), biological oxygen demand (BOD_5), sulfate, sulfide, total suspended solids (TSS), and volatile suspended solids (VSS). Effluent quality was mainly monitored through COD, sulfide and volatile fatty acids content. Reactor monitoring was based on TSS, VSS, COD, pH and methane content in the generated biogas (% CH_4). TSS, VSS, COD, BOD_5, sulfate and sulfide were determined according to standard methods (APHA, 2005). VFA concentration was determined by titration according to the method proposed by Moosbrugger et al. (1992). Dissolved methane in the effluent was determined according to Giménez et al. (2012).

6.2.4. Data processing and calculations

The instrumentation implemented in the prototype acquired the following data, among others: reactor influent flow, reactor temperature, transmembrane pressures and fluxes, biogas flows for reactor mixing and membrane scouring, mixed liquor feeding flow to the membrane tanks, and biogas production and quality. Different key performance indicators were calculated to assess the performance of the system, *i.e.,* J_{20}, specific energy consumption (EC), total energy recovery (TER), net energy demand (NED) and greenhouse gas (GHG) emissions.

It is important to highlight that a fraction of the influent organic matter is consumed by SRB, which meant it was not available for biogas production by methanogenic organisms. Specifically, 2 grams of influent COD were consumed by SRB for each gram of sulfate (stoichiometrically).

Over-dimensioned equipment in the prototype plant prevented the direct measurement of electrical power consumption from being representative of the energy requirements of AnMBR technology. The power required by the equipment (pumps and blowers) was therefore calculated theoretically using the equations proposed by Judd and Judd (2006) and Pretel et al. (2016). Adiabatic indexes, and blower and pump efficiencies were estimated based on standard values for these components.

Two sources of GHG emissions were considered: i) indirect CO_2 emissions related to the NED of the system ($CO_{2_{NED}}$) and ii) direct equivalent CO_2 emissions from dissolved methane in the effluent ($CO_{2_{D-CH_4}}$). Both sources are considered as the main GHG contributors during the operation of the demo plant. It should be noted that, according to the IPCC2013 application guides, since they are included in a short-term carbon cycle, CO_2 emissions of biogenic origin produced during anaerobic digestion can be excluded from GHG calculations, as well as those associated with the subsequent combustion of biogas in a CHP unit. Three different values were calculated for $CO_{2_{D-CH_4}}$ and therefore for total CO_2 emissions: i) without dissolved methane recovery; ii) with 50 % of dissolved methane recovery and iii) with 70 % of dissolved methane recovery. Percentages of 50 % and 70 % were assumed based on the optimum economic and environmental results from previous studies, respectively (Sanchis-Perucho et al., 2020).

Details on data calculations are provided as supplementary material.

6.3. Results and discussion

6.3.1. Filtration process performance

Figure 6.1 gives the performance of the filtration process during the complete experimental period for the MT_A, MT_B and MT_C membrane tanks (average values can be seen in table 6.1S of the supplementary material). The black dashed bars indicate short maintenance stops (days 94 and 442) and grey dotted-dashed bars indicate membrane chemical cleanings (days 128 and 363). A conservative maximum TMP security value of 500 mbar was selected based on the supplier's information. Finally, the TSS concentration depicted in Figure 6.1 corresponds to the reactor mixed liquor (i.e., MLSS), thus representing the TSS concentration of the mixed liquor entering the three MTs.

As can be seen in Figure 6.1, J_{20} was maintained in similar values for all MTs: 14-20, 15-23 and 15-20 LMH for MT_A, MT_B, and MT_C, respectively. These J_{20} are somewhat higher than previous long-term AnMBR studies (Ji et al., 2020; Peña et al., 2019). The J_{20} applied were also competitive against MBR for urban wastewater treatment. For instance, Xiao et al. (2019) evaluated 19 large-scale Chinese MBRs and reported fluxes in the range of 15-25 LMH (19.6 LMH on average).

Regarding SGD_m, MT_A was operated between 0.1 and 0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$; MT_B mostly at 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$, although higher (0.35-0.40 $Nm^3 \cdot m^{-2} \cdot h^{-1}$; days 130-200) and lower (0.1-0.2 $Nm^3 \cdot m^{-2} \cdot h^{-1}$; days 360-500) values were also evaluated; and MT_C at 0.25 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ at days 294-360 and 510-580 and in the range of 0.1-0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ between days 360 and 510. The modifications in the applied SGD_m aimed to assess gas sparging intensity for fouling control.

In this operation, SGD_P varied between 7 and 23 $Nm^3 \cdot m^{-3}$ for MT_A and between 14 and 16 $Nm^3 \cdot m^{-3}$ for MT_B, with short periods above 20 $Nm^3 \cdot m^{-3}$ (e.g., days 200-216) and below 10 $Nm^3 \cdot m^{-3}$ (days 383-401). Figure 6.1c gives the first period of MT_C, with SGD_P at 13-14 $Nm^3 \cdot m^{-3}$ (days 294-360), a second highly variable period between 5 and 22 $Nm^3 \cdot m^{-3}$ and a third period with SGD_P steady at around 12-13 $Nm^3 \cdot m^{-3}$.

Figure 6.1 allows the comparison of the filtration performance of the three MTs, attending to the differences in their operation. Between days 31 and 91, MT_A and MT_B (see Figure 6.1a and b) operated at similar J_{20} (16-20 LMH) and SGD_P (15-16 $Nm^3 \cdot m^{-3}$) and both tanks started from a close TMP, in the 210-220 mbar range. It can be observed that the behavior of both MTs was very similar, reaching in both cases the limit value of ≈500 mbar at the same time (day 91). It can also be observed a sharp increase in TSS on day 45, reaching a concentration of 15 $g \cdot L^{-1}$ around day 75, which coincides with the sharp TMP increase recorded in both tanks. Therefore, it can be concluded that the membranes were operated at supra-critical filtration conditions due to the combination of the J_{20}, SGD_P and TSS values within the experimental period.

After the maintenance stop conducted between days 91 and 94, both MT_A and MT_B partly reduced their TMP without applying any membrane chemical cleaning, thus the observed recovery was associated with reversible fouling that was removed as a result of prolonged air-assisted membrane scouring without filtration.

After restarting filtration, the behavior of both tanks differed. In the case of MT_A, there was an abrupt rise in TMP after only three days of operation, reaching a stable TMP around 420 mbar. From day 121, the J_{20} was reduced from 20 LMH to 12 LMH, and this caused a drop in the TMP from 420 to 330 mbar, which represents a reduction of 21%. This TMP profile would indicate that reducing J_{20} turned the filtration process from supra-critical/critical (since TMP first raised and then could be maintained at 420 mbar) to sub-critical conditions. In MT_B, the filtration process caused a constant increase in TMP until reaching the maximum threshold value (i.e., 500 mbar)

set for conducting a membrane chemical cleaning. On day 128, the membrane in MT_B was chemically cleaned, which reduced TMP to 161 mbar. A reduction of 43 % in membrane total resistance (R_T) when comparing the R_T achieved after the first maintenance stop on days 91-94 (i.e., 14.1 LMH·mbar^{-1}) and the chemical cleaning on day 128 (i.e., 8.1 LMH·mbar^{-1}) would indicate the amount of irreversible fouling accumulated on the MT_B membrane when the maintenance stop was conducted (between days 91 and 94). Eventually, filtration performance of MT_B between days 128 and 221 confirms that the combination of J_{20} above 20 LMH, SGD_P below 17 Nm^3·m^{-3} and TSS above 10 g·L^{-1} led to significant fouling propensities, increasing TMP from 150-190 to 470 in 35-40 days.

Different profiles between MT_A and MT_B regarding J_{20}, SGD_P and TMP can be observed from day 200 and 295. Specifically, the reduction in J_{20} from 17.5 to 15-16 LMH applied to the MT_A managed to reduce the TMP from the maximum of 450 mbar to 360 mbar (day 245). Moreover, the reduction recorded in the TSS concentration from 11 to 9 g·L^{-1} also favored reducing the SGD_m from 0.37 to 0.29 Nm^3·m^{-2}·h^{-1} (corresponding to a SGD_P of 23 and 19 Nm^3·m^{-3}, respectively), resulting in a slight decrease in TMP, thus confirming that filtration was carried out under sub-critical conditions with this combination of J_{20}, SGD_m and TSS concentration. In this same period, MT_B was operated with a rising J_{20} from 15.5 LMH to 21 LMH and a SGD_m of 0.29 Nm^3·m^{-2}·h^{-1} (SGD_P of 19 Nm^3·m^{-3}) in average. These operating conditions resulted in a sharp TMP increase from 320 mbar to 460 mbar (day 225). Despite this increase, the reduction in TSS concentration to 10 g·L^{-1} allowed to offset the effect of increasing J_{20} on TMP thus managing to remain stable TMP values of around 470 mbar, which indicated that sub-critical filtration conditions were kept. It is also worth to highlight the drop in TMP that occurred between days 290 and 300 due to a decrease in J_{20} from 21 LMH to 16 LMH and the subsequent TMP increase observed when the TSS concentration increased from day 300. Thus, slight changes on J_{20} and/or SGD_m quickly allowed compensating the effect of TSS concentration on TMP, highlighting the potential of gas-assisted membrane scouring as physical fouling control strategy to keep sub-critical/critical filtration conditions thereby controlling membrane fouling propensity.

Between days 300 and 363, when a second membrane chemical cleaning was conducted, MT_B and MT_C were operated in parallel. Figure 6.1b shows how MT_B experimented a TMP reduction due to increasing the SGD_m up to 0.35 Nm^3·m^{-2}·h^{-1} (corresponding to a SGD_P of 20 Nm^3·m^{-3}) between days 344 and 361. MT_C was operated at similar J_{20} values than the ones established in MT_B, although a constant SGD_m of 0.24 Nm^3·m^{-2}·h^{-1} (SGD_P of 13 Nm^3·m^{-3}) was established in MT_C. Thus, it can be concluded that, while it was possible to control TMP in MT_B by increasing

gas sparging intensity, TMP in MT_C continuously increased between days 300 and 363. Therefore, MT_B operated at sub-critical filtration conditions (J_{20} of 15-18 LMH, SGD_P of 15-20 $Nm^3 \cdot m^{-3}$ and TSS of 12 $mg \cdot L^{-1}$) and MT_C at supra-critical filtration conditions (J_{20} of 15-18 LMH, SGD_P of 13-14 $Nm^3 \cdot m^{-3}$ and TSS of 12 $mg \cdot L^{-1}$).

On day 363, a chemical cleaning was conducted in the three MTs. In the case of MT_A, TMP turned out to similar values to the original ones (day 30). Thus, fouling was properly removed through chemical cleaning, revealing negligible levels or irremovable fouling on the membrane within the experimental period. In the case of MT_B, the R_T values achieved after this second chemical cleaning (i.e., 6.7 $LMH \cdot mbar^{-1}$) suggested that the first one (day 128) was not able to completely recovering irreversible fouling, i.e. around 1.4 $LMH \cdot mbar^{-1}$ of R_T was additionally removed compared to the results from the first chemical cleaning applied. In the case of MT_C, R_T was reduced to around 4.9 $LMH \cdot mbar^{-1}$, lower than the initial TMP on day 294 (i.e., 7.9 $LMH \cdot mbar^{-1}$), which would indicate the presence of previous fouling, since this membrane was kept submerged in the mixed liquor.

During days 361-445, the three MTs were operated in parallel. J_{20} between 14 and 15 LMH and low SGD_m (mostly 5-6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$) were established for MT_A and TM_B. MT_C was operated at J_{20} of 15 LMH and higher but fluctuating SGD_P (5-21 $Nm^3 \cdot m^{-3}$). Around day 400, a sharp increase in TMP up to 460-480 mbar was observed in MT_A and MT_B driven by the low agitation, while MT_C could sustain filtration at low TMP for a few days longer (until day 420) since it was operated with higher SGD_P.

Finally, a maintenance stop was conducted on day 443. After this stop, R_T did not reach the value obtained after the previous chemical cleaning, thus some irreversible fouling (around 5 $LMH \cdot mbar^{-1}$ equivalent) may be developed from day 363 to 443 During this period, MT_B and MT_C were operated at similar J_{20} and SGD_m levels. Indeed, both tanks resulted in similar TMP behaviors. From day 505, only the MT_C was operated and its SGD_m was increased to 0.24 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to an SGD_P of 12.5 $Nm^3 \cdot m^{-3}$). This increase resulted in a significant reduction in TMP growth rate and therefore in fouling rate (i.e., from 4.1 $mbar \cdot d^{-1}$ to 1.4 $mbar \cdot d^{-1}$).

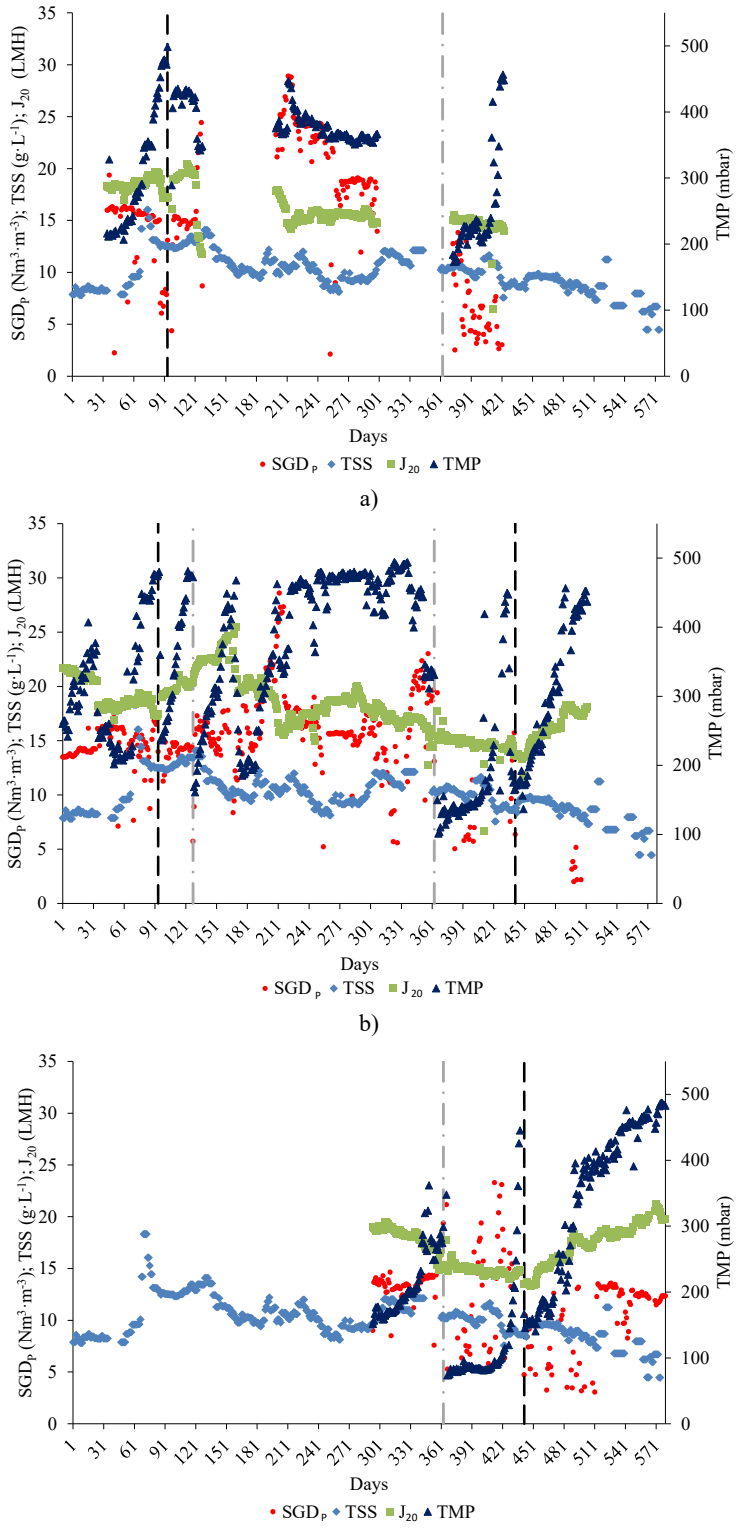

Figure 6.1. Evolution of sparging gas demand per m3 of permeate (SGD_P), gross 20ºC-standardized transmembrane flux (J_{20}), transmembrane pressure during filtration stage (TMP) and total suspended solids (TSS) in a) MT_A; b) MT_B and c) MT_C. (Maintenance stop: black dashed line; Membrane chemical cleanings: grey dotted-dashed lines).

Therefore, Figure 6.1 shows the complex interaction between J_{20}, SGD_m (as shown by SGD_P), TSS in the reactor, TMP and the evolution of fouling. Operation at high J_{20} with low fouling propensities can be achieved with high SGD_m and low mixed liquor TSS concentrations. These results are in line with those obtained elsewhere by Mei et al. (2016).

The results show that the system was able to conduct filtration at a moderate-high J_{20} (15-20 LMH) with competitive SGD_m (at an SGD_P of 10-20 $Nm^3 \cdot m^{-3}$) and moderate TSS (8-13 $g \cdot L^{-1}$), although shifts between supra-critical and subcritical filtration were seen to occur with small changes in the operating conditions. Operating at sub-critical filtration conditions (recommended to avoid quick TMP increase) was possible with TSS concentrations below 10 $g \cdot L^{-1}$, maximum J_{20} of 16 LMH and SGD_m of 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to 15 $Nm^3 \cdot m^{-3}$). Figure 6.1 also shows that fouling control can be achieved even at high TMP (e.g., between days 210 and 300 in MT_A (380-400 mbar) or between days 210 and 335 in MT_B (470-500 mbar). During these periods, the TMP presented a plateau profile, which indicated negligible fouling propensity. In this sense, the rapid response of TMP to changes in J_{20} and/or SGDm contributes to facilitate fouling control. However, operating at high TMP would imply increasing energy requirement for permeate pumping and increasing dissolved methane concentration. Therefore, to avoid these drawbacks operation at high TMP is not advisable.

As filtration performance comparison, Kong et al. 2021 operated a pilot-scale AnMBR to treat municipal wastewater at ambient temperature (25 ºC) and reported significantly lower transmembrane fluxes of approx. 3 and 4.5 LMH for HRT of 48 and 24 h, respectively, despite experiments at HRT of 24 h were conducted with lower mixed liquor suspended solids, in the 6.7-10.2 $g \cdot L^{-1}$ range (mixed liquor suspended solids at HRT of 48 h were not included in the study). Shin & Bae (2018) described the performance of different pilot scale AnMBR treating domestic wastewater. These authors found that the most common transmembrane fluxes were in the 6-14 LMH range, while applied SGD_m varied mostly between 0.15 and 0.31 $Nm^3 \cdot m^{-2} \cdot h^{-1}$. Lee & Liao, (2021) considered 10 LMH as typical design parameter for transmembrane flux.

Finally, it should be noted that the demo plant achieved a successful long-term operation without chemical cleaning requirements, the longest operation period being 233 days of MT_B (days 128-363), although the results indicate that longer operating periods could be achieved. No irrecoverable fouling was detected during the study since chemical cleaning returned the TMP to the initial values or even lower.

6.3.2. Energy assessment

Evolution of biogas production, its methane content expressed in percentage of CH_4, and removed COD (% COD_{rem}) can be found in supplementary material. Moreover, details on the evolution of biogas production process in this demo plant during this operational period can be found in Robles et al. (2022), where it could be concluded that temperature, HRT, SRT and influent COD:SO_2^{-4}-S ratio showed crucial influence in biogas production.

As already mentioned in section 6.2.2, organic load and temperature varied according to economic activities and ambient conditions, respectively. Methane production is therefore highly influenced by these factors. Periods with high temperature and high OL-MA showed higher biogas productions. Conversely, low OL-MA and low temperature led to reduced biogas production. With regards to methane content in the biogas, it remained mostly above 70 % (75±3 % on average). Finally, AnMBR reached a significant COD removal efficiency (89±5 % on average) (further details can be found in Figure 6.3S of supplementary material).

Figure 6.2 shows the evolution of EC, TER and NED. The gaps in the figures are due to maintenance stops and failures in the online equipment required for calculations. Figure 6.2a shows an initial period between days 1-30 with high energy consumption for reactor mixing (around 0.11 $kWh \cdot m^{-1}$). On day 31, mixing was reduced to 0.04-0.08 $kWh \cdot m^{-3}$ until day 500, without affecting reactor performance. EC related to membrane scouring also varied according to the applied SGD_m and number of membrane tanks in operation. Between days 1-30, only MT_B was in operation at an SGD_m of 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$. On day 35, MT_A started operating jointly with MT_B, leading to an increased need for sparging and higher energy consumption (1.2-1.3 $kWh \cdot m^{-3}$). MT_A and MT_B were also operated in parallel between days 194-299, when energy consumption reached the highest value (1.5-2.0 $kWh \cdot m^{-3}$), which was associated with higher applied SGD_m in MT_A (0.36-0.43 $Nm^3 \cdot m^2\text{-}h^1$). From day 300, MT_A was stopped, SGD_m in MT_B was reduced to 0.20 $Nm^3 \cdot m^2\text{-}h^1$ and MT_C started with an applied SGD_m of 0.24 $Nm^3 \cdot m^2\text{-}h^1$. Energy consumption for membrane scouring was thus reduced to 0.5-0.6 $kWh \cdot m^3$. On days 330-355, SGD_m in MT_B was set to 0.35 $Nm^3 \cdot m^2 \cdot h^1$, which led to higher energy consumption for membrane scouring. The period between days 365-500 was characterized by lower applied SGD_m (and thus SGD_P) than previously, which allowed moderate-low energy consumption despite MT_B and MT_C being operated in parallel, and MT_A was jointly operated between days 373-420. The EC associated with membrane tank feeding mostly remained below 0.02 $kWh \cdot m^{-3}$ while the energy requirement for the reactor feeding pump was negligible. After day 500, HRT was set to

70 h, so that the treatment flow was reduced, leading to higher specific energy consumption in $kWh \cdot m^{-3}$.

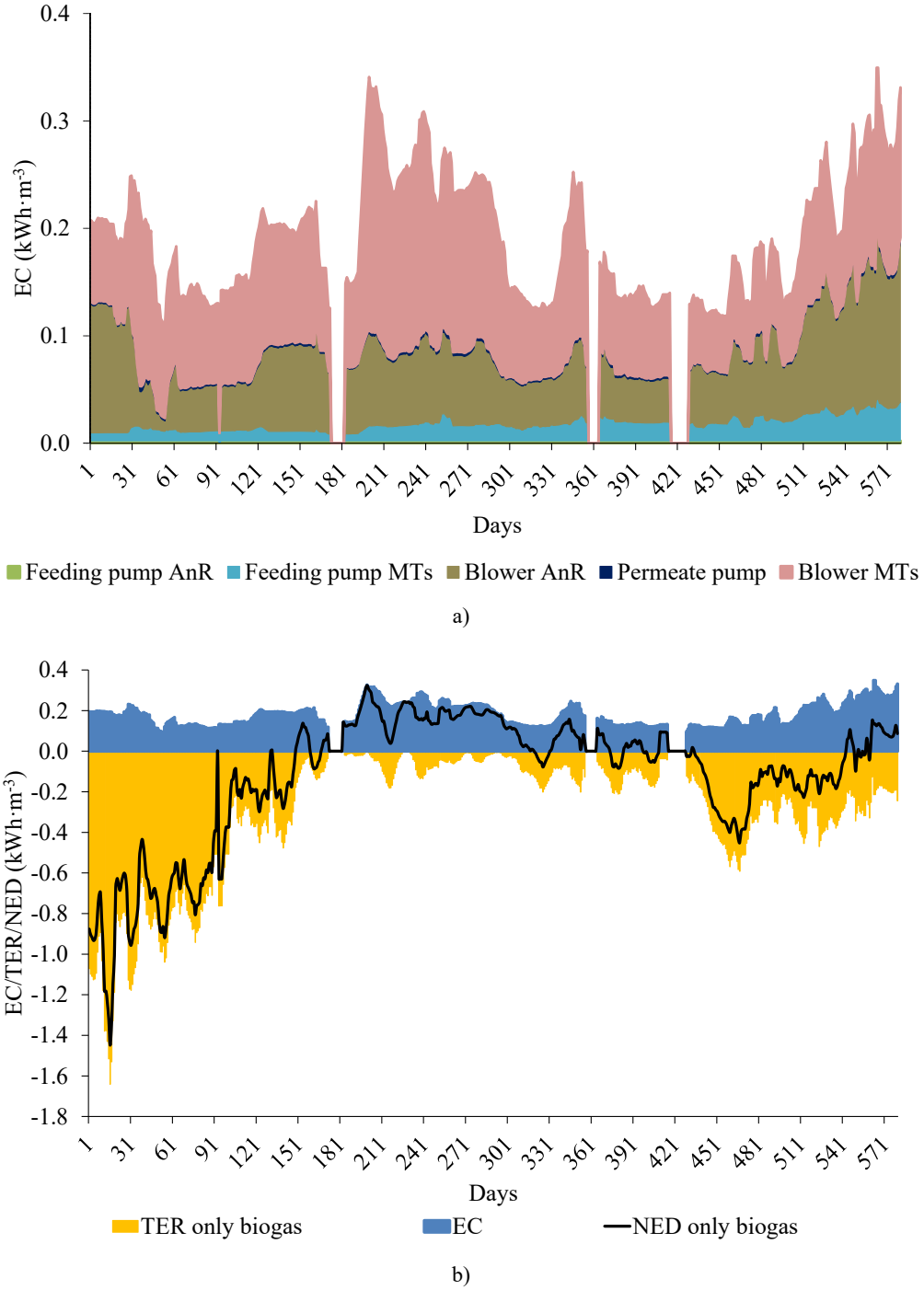

Figure 6.2. Results of (a) allocation of energy consumption (EC) per equipment and (b) total energy consumption (EC), total energy recovery (TER) and net energy demand (NED) considering only the energy recovered from biogas (only biogas).

Figure 6.2b gives the total EC, TER and NED. The TER in Figure 6.2b was based on energy recovered from the biogas stream only (0 % of dissolved methane recovery). This recovered

energy allowed the plant to perform as a net energy producer for over 50 % of the study period (negative values in NED). As can be seen in Figure 6.2b, methane production and therefore energy recovery was boosted by a high OL-MA and temperature (see Figure 6.2S in supplementary material). In this regard, TER followed the same trend as the temperature from day 1 to 160, with a high energy production at 28 °C (days 1-60), which dropped steadily as the temperature dropped to 14 °C (day 160). Days 440-580 also showed high TER values, when energy production increased when the temperature remained above 26 °C (days 400-450) and by the increase in OL-MA (days 450-490). On the other hand, days 160-440 resulted in reduced TER since this period was characterized by a relatively low temperature and OL-MA.

Figure 6.2 gives the average results for the entire study period and shows that the main contributions to EC were associated with the blower requirements for membrane scouring (55.2 %), followed by reactor mixing (34.4 %). Conversely, membrane tanks' feeding pump, filtration pumps, and reactor feeding pump had comparatively very low energy consumption (7.8 %, 1.6 % and 1.0 %, respectively). The contribution of the degassing membrane blowers was negligible (<0.02 %). Overall, average EC accounts for 0.192 ±0.070 $kWh \cdot m^{-3}$ while average TER (0 % methane recovery) was 0.361±0.271 $kWh \cdot m^{-3}$. Dissolved methane produced more energy than that used in its recovery, although its contribution to TER was relatively low (between 0.02 $kWh \cdot m^{-3}$ and 0.04 $kWh \cdot m^{-3}$ for dissolved methane recovery ratios of 50 and 70 %, respectively). However, this recovery definitively reduced GHG emissions, as will be explained in the following section.

These results can be compared with other studies dealing with AnMBR for wastewater treatment. Rong et al. (2022) evaluated the energy balance of an AnMBR at pilot scale and a projected scaled-up facility of 10000 $m^3 \cdot d^{-1}$. Influent COD was 331-414 $mg \cdot L^{-1}$ and temperature ranged between 15 and 25 ºC. These authors obtained a net electricity demand in the 0.09-0.10 $kWh \cdot m^{-3}$ range, with gas sparging accounting for 52.46-74.60 % and reactor mixing for 4.08-6.13 %. More specifically, Hu et al. (2022) reported energy consumptions for gas sparging between 0.25 and 3.4 $kWh \cdot m^{-3}$, accounting for approx. 70 % of total energy consumption, which are between 2 and 32 times the average energy consumption obtained in this study (0.106±0.047 $kWh \cdot m^{-3}$). Concerning degassing membranes, lower energy consumptions were achieved (between $2.3 \cdot 10^{-5} \pm 3.4 \cdot 10^{-6}$ and $4.3 \cdot 10^{-5} \pm 6.3 \cdot 10^{-6}$ $kWh \cdot m^{-3}$, see Table 6.2) compared to the one reported by Velasco et al. (2021), i.e. 0.01 $kWh \cdot m^{-3}$ for 90 % of dissolve methane recovery.

Moreover, Krzeminski et al. (2017) reported 0.3-0.6 kWh·m^{-3} for CAS and 0.6-2.3 kWh·m^{-3} for MBR, including nutrient removal. In this sense, Fraia et al. (2018) reported energy consumption for nitrogen removal up to 6.08 kWh per kg of influent total nitrogen (kWh·TN_{in}) in wastewater treatment facilities with high nitrogen removal efficiency (> 89.2 %), while energy consumption associated to phosphorus removal was considered negligible. Hence, assuming the nitrogen load of the AnMBR influent (see Table 6.1) the energy demand for nitrogen removal in an aerobic/anoxic process would be of approx. 0.29 kWh·m^{-3}, according to this literature value. Hence, extracting this value to the above-mentioned ranges for CAS and MBR energy demand, the energy consumption related to COD removal in CAS and MBR technology would be between 0.01-0.31 and 0.31-1.99 kWh·m^{-3}, respectively. These estimated range of energy requirements would be still higher than the average NED reported in this study when dissolved methane recovery is not conducted (-0.169±0.341 kWh·m^{-3}; see Table 6.2).

These results emphasize the importance of optimizing the parameters governing the reactor mixing requirements and filtration, especially with regard to SGD_m. Increasing sludge recycling would reduce TSS concentration in the MTs and help to reduce SGD_m necessities. Since the MT feeding pump represents only 8 % of EC and membrane scouring accounts for 55 %, this strategy could improve the AnMBR energy performance through a slight increase in the consumption of the sludge pump, which would be compensated by a larger reduction of consumption for membrane scouring. This sludge circulation could also be used to promote reactor mixing and would reduce its energy consumption (34 %).

It should be noted that the significant sulfate content in the influent favors SRB growth, so that improved AnMBR energy performance could be expected when treating low-sulfate wastewater (Robles et al., 2022).

6.3.3. GHG emissions

Figure 6.3 shows the equivalent CO_2 emissions associated with dissolved CH_4 (D-CH_4) when no recovery is considered (grey area) and net GHG emissions (including D-CH_4 and those associated with EC) considering 0 %, 50 % and 70 % of dissolved methane recovery (orange, blue and green lines respectively).

Table 6.2. Energy consumption (EC), total energy recovery (TER) and net energy demand (NED) for 0%, 50 % and 70 % of dissolved methane recovery (D-CH_4).

	D-CH_4 Recovery fraction		
kWh·m^{-3}	**0%**	**50%**	**70%**
EC blower DM[a]	-	$2.3\cdot10^{-5}\pm3.4\cdot10^{-6}$	$4.3\cdot10^{-5}\pm6.3\cdot10^{-6}$
EC blower AnR	0.066±0.030		
EC blower MT	0.106±0.047		
EC AnR feeding pump	0.002±0.000		
EC MT feeding pump	0.015±0.007		
EC permeate pump	0.003±0.001		
Total EC	0.192±0.070		
TER	0.361±0.271	0.382±0.306	0.397±0.377
NED	***-0.169±0.341***	***-0.190±0.376***	***-0.205±0.447***

[a] Degassing Membranes

When no methane recovery was considered, the contribution of D-CH_4 to total emissions accounted for 0.5-0.6 $kgCO_{2eq}\cdot m^{-3}$ during almost the entire period of plant operation. The periods in which the orange line is lower than the corresponding grey bar indicate net energy production (e.g., days 1-130, see Figure 6.2a). On these days, savings in indirect CO_2 emissions related to energy production generated a *GHG credit* which reduced the total carbon footprint. This effect was also found to a lesser extent during days 430-480. Conversely, the orange line over the grey area indicates emissions related to net EC, which is added to those of dissolved methane release (days 210-340).

Net GHG emissions with 0 % of methane recovery (orange line) showed values around 0.4 $kgCO_{2eq}\cdot m^{-3}$, when high volumes of biogas were produced and energy recovery was significantly high (days 1-130), whereas the value oscillates around 0.6 $kgCO_{2eq}\cdot m^{-3}$ when low biogas production was obtained. According to Sanchis-Perucho et al. (2020), the economic optimum for dissolved methane recovery would be 50 % and the environmental optimum 70 % approx. The results obtained with 50 % and 70 % of dissolved methane recovery (blue and green

lines, respectively) show the importance of this process for the improvement of the overall AnMBR environmental performance by reducing CH_4 emissions. For instance, recovering 50 % of the methane would reduce the equivalent CO_2 emissions to 0.1-0.3 $kgCO_{2eq} \cdot m^{-3}$. Operating with 70 % methane recovery kept the emissions below 0.2 $kgCO_{2eq} \cdot m^{-3}$ during the entire study period. By way of comparison, Rodriguez-Garcia et al. (2011) estimated emissions of around 0.3 $kgCO_{2eq} \cdot m^{-3}$ for CAS facilities with organic matter and nutrient removal discharging in sensitive areas, while Chen (2019) estimated small-scale MBR (351 $m^3 \cdot d^{-1}$ treatment capacity) emissions to be around 0.7 $kgCO_2 \cdot m^{-3}$.

The GHG emissions associated with D-CH_4 appear as the main contributor to this environmental impact (between 83.8 % and 95.7 %). Since the other contributors are derived from power consumption, the higher use of renewable energies in the energy mix will raise these percentages even further. The improvement of AnMBR will necessarily involve further research in systems of dissolved methane recovery to offset CO_2 emissions. In this regard, AnMBR combined with degassing membranes represents a promising combination, as it merges greater energy recovery with the possibility of reducing GHG emissions (Henares et al., 2017).

Increasing energy recovery would also help to widen the applicability of AnMBR technology, for example co-digestion of wastewater with other substrates (organic fraction of food waste, food industry wastewater, etc.) has been successfully explored (Pretel, et al., 2016). Biogas upgrading is another strategy that takes advantage of methane; this option has recently received great attention in the UE (e.g. REPowerEU Plan) and promising environmental and economic results have been obtained (Ardolino et al., 2021).

Finally, it is important to highlight the additional benefits that recovering nutrients would entail, since N and P production from traditional sources require high power consumption and GHG emissions. McCarty et al. (2011) reported 19.3 kWh per kg N and 2.11 kWh per kg P. According to these values, considering the average N and P content in the effluent (45.7±10.4 mg $N \cdot L^{-1}$ and 7.8±2.0 mg $P \cdot L^{-1}$, data not shown) and a GHG emission factor for Spanish energy mix generation of 0.14 kg $CO_2 \cdot kWh^{-1}$ (REE), fertigation would allow savings of 0.126 kg $CO_{2eq} \cdot m^{-3}$, which would help the combination of AnMBR+fertigation to achieve carbon neutrality if 70 % of the dissolved methane was recovered.

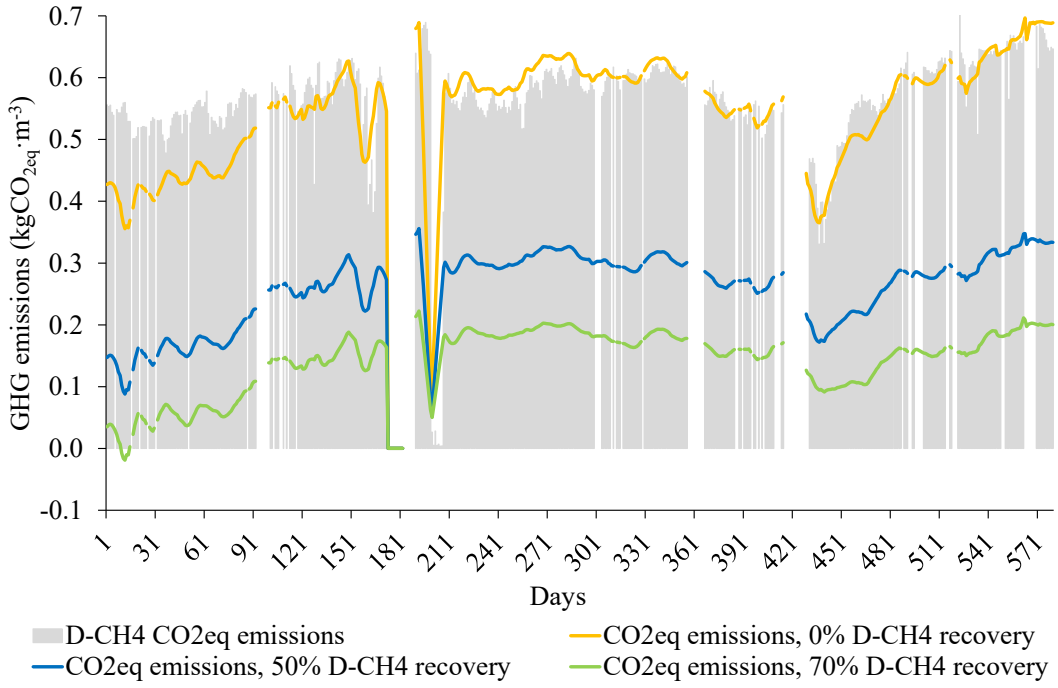

Figure 6.3. Evolution of GHG emissions in the operating phase of the demonstration plant: direct CO2 equivalent emissions of dissolved methane (grey bars), and CO_2 equivalent emissions when considering NED and dissolved methane recovery of 0% (orange), 50% (blue) and 70% (green).

The results of this study suggest that AnMBR shows promise as a potential technology for reducing the energy dependence and overall environmental impacts of wastewater treatment. The combination of anaerobic treatments and membrane technology in AnMBR paves the way for turning wastewater into a source of resources. Energy, nutrients and reclaimed water could be supplied by the continuously generated sewage, while this approach complies with the fundamental principles of the CE and is fully aligned with the Sustainable Development Goals.

6.4. Conclusions

An industrial AnMBR wastewater treatment prototype was successfully operated for 580 days. Fouling propensity remained low and long-term operations were achieved, with a maximum continuous operation of 233 days without applying membrane chemical cleaning. The UF modules were able to operate at a high J_{20} in the range of 15-20 LMH and a competitive SGD_m of between 0.1 and 0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (which corresponded to a SGD_P of 10-20 $Nm^3 \cdot m^{-3}$). TMP showed a quick response to changes in J_{20} and SGD_m, which facilitated fouling control. Moreover, operation with TSS concentrations below 10 $g \cdot L^{-1}$, J_{20} of 16 LMH and SGD_m of 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$

(corresponding to 15 $Nm^3 \cdot m^{-3}$) would further lengthen the filtration periods between chemical cleanings.

With regards to energy assessment, the average EC was estimated at 0.192±0.070 $kWh \cdot m^{-3}$, with most of the power consumed by membrane scouring (55 %) and reactor mixing (34 %). The valorization of organic matter provided an average TER of 0.361±0.271, 0.382±0.306 and 0.397±0.377 $kWh \cdot m^{-3}$ with 0 %, 50 % and 70 % recovery of dissolved methane, respectively, obtaining a NED of -0.169±0.341, -0.190±0.376 and -0.205±0.447 $kWh \cdot m^{-3}$, respectively. The increase in energy production associated with the recovery of dissolved methane is therefore low, but higher than the energy consumption of degassing membranes. Finally, dissolved methane recovery appears as the key challenge to be overcome for achieving low GHG emissions with AnMBR. In this study, recoveries of 50 % and 70 % reduced emissions from 0.5-0.6 to 0.1-0.3 and below 0.2 $kgCO_2 \cdot m^{-3}$, respectively.

Overall, the AnMBR demo plant showed a suitable filtration performance, significant energy productions and low CO_2 emissions when recovering dissolved methane. These results indicate that AnMBR combined with degassing membranes represents a robust alternative to the conventional wastewater treatment.

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6.5. Supplementary material

6.5.1. AnMBR description

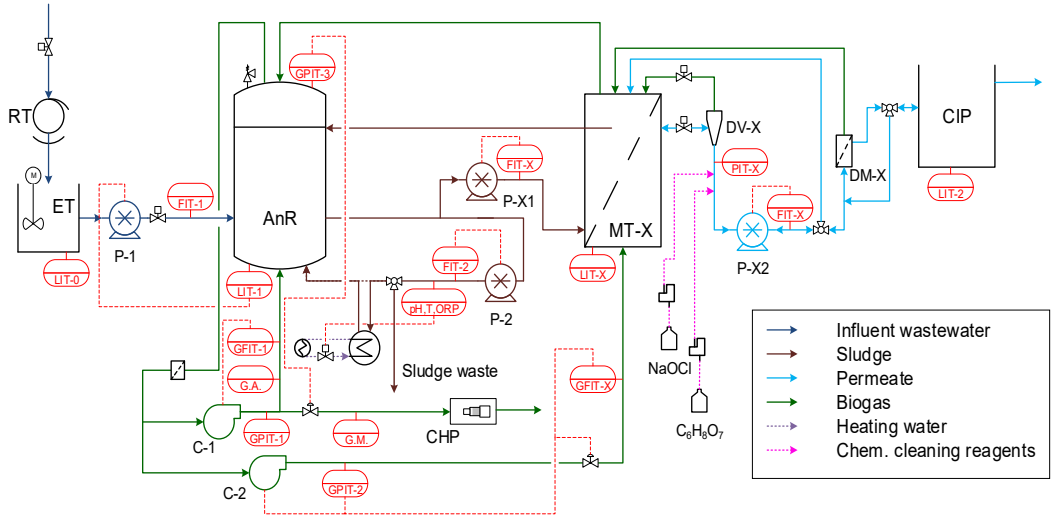

Figure 6.1S. Process and Instrumentation Diagram (P&ID). Note: "X" represents de membrane treatment lane.

6.5.2. AnMBR operation

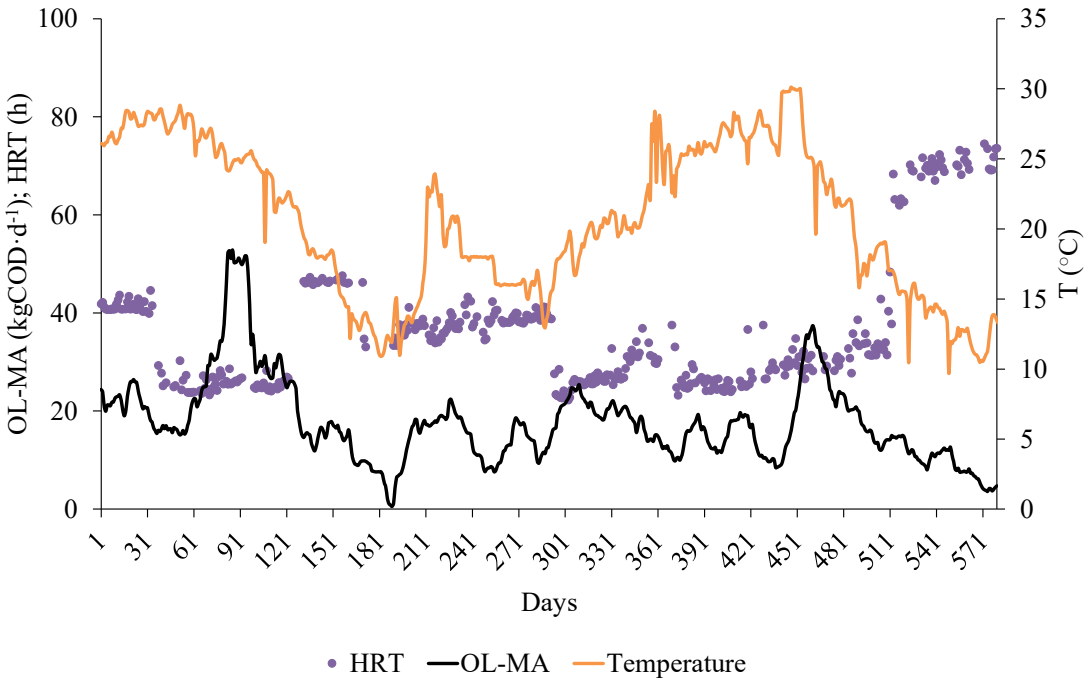

Figure 6.2S. Evolution of organic load available for methanogenesis (OL-MA), HRT and temperature during the experimental period.

6.5.3. AnMBR biological performance

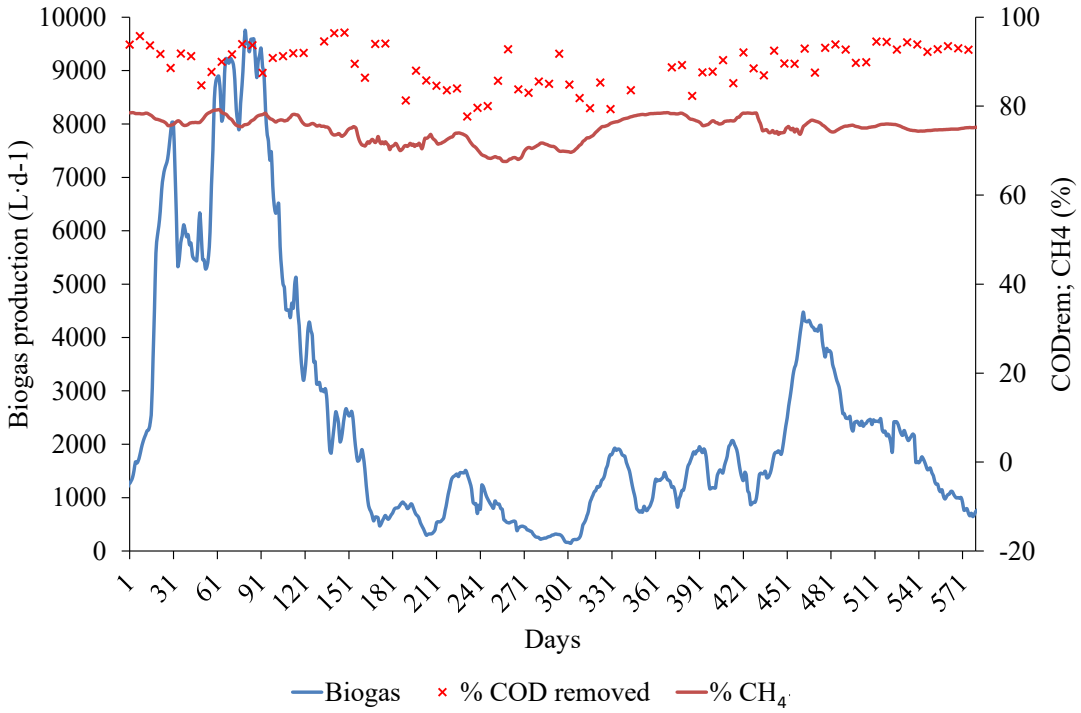

Figure 6.3S. Evolution of biogas production, percentage of methane in biogas (% CH_4) and COD removal (% COD_{rem}).

6.5.4. Energy assessment

Organic load available for methanogenesis was calculated as follows:

$$OL - MA = \frac{Q_{inf} \cdot (COD_T - 2 \cdot SO_4 - S)}{1000} \qquad \text{Equation 6.1S}$$

where OL-MA represents the organic load available for methanogenesis ($kg \cdot d^{-1}$); Q_{inf} is the influent flow to be treated ($m^{-3} \cdot d^{-1}$); COD_T is the total chemical oxygen demand ($mg \cdot L^{-1}$) and SO_4-S is the concentration of sulfate in the influent ($mg\ S \cdot L^{-1}$).

To assess the filtration process, J_{20} was calculated by means of Eq.(6.2S), according to Rosenberger et al., (2006):

$$J_{20} = J \cdot e^{-0.0293 \cdot (T-20)} \qquad \text{Equation 6.2S}$$

where J is the gross flux (LMH) and T is temperature (Celsius).

Moreover, total resistance of the membrane was calculated by means of Eq. (6.3S):

$$R_T = \frac{TMP}{J_{20}}$$ Equation 6.3S

Where TMP is the transmembrane pressure (mbar).

To calculate energy consumption (EC), the online data monitored from the prototype plant was used as input (gas flow, gas temperature, gas pressure, permeate flow, transmembrane pressure, heights). The EC associated with sludge pretreatment, homogenization and dehydration were not considered. Based on these power requirements, it was possible to calculate the EC of the demo-plant as expressed in Eq. (6.4S):

$$EC = \frac{\sum_{i=1}^{n} P_{B,i} + \sum_{j=1}^{m} P_{g,j} + \sum_{k=1}^{o} P_{stage}}{Q_{inf}}$$ Equation 6.4S

where P_B (kW) are the power requirements for each *n* blower, P_g (kW) are the power requirements for each general *m* pumps, P_{stage} (kW) are the power requirements associated with the membrane *o* filtration stages (filtration, degasification and back-flushing) and Q_{inf} is the influent flow to be treated ($m^{-3} \cdot h^{-1}$).

It was not possible to install a Combined Heat and Power (CHP) system in the prototype plant since no proper model was found on the market able to operate with the low biogas flow rate produced. Energy production per treated m^3 was thus calculated by Eq. (6.5S) using a theoretical value for the electrical power efficiency of the CHP (η_{CHP} = 35%).

$$TER = -\frac{(Q_{biogas} \cdot y_{biogas}^{CH_4} + Q_{dissolved} \cdot y_{dissolved}^{CH_4}) \cdot CV_{CH_4} \cdot \eta_{CHP}}{Q_{inf}}$$ Equation 6.5S

where TER is total energy recovery in $kWh \cdot m^{-3}$. Q_{biogas} is the biogas production in normal conditions of temperature and pressure (NTP) (m^3 NTP$\cdot d^{-1}$) in the AnR and $y_{biogas}^{CH_4}$ represents the

methane fraction in this biogas; $Q_{dissolved}$ accounts for the biogas flow recovered from the effluent at NTP (m^3 NTP·d^{-1}) and $y_{dissolved}^{CH_4}$ represents the methane fraction in this biogas; CV_{CH_4} is the calorific value of CH_4 (10.6 kWh·NTP m^{-3}) and Q_{inf} is the influent flow ($m^3 \cdot d^{-1}$).

Using this theoretical approximation and based on the results of Sanchis-Perucho et al., (2021), three values for the total energy recovery (TER) were calculated considering only the methane present in the biogas stream (without methane recovery from the permeate flow), accounting for the recovery of 50 % of the dissolved methane (see Section 6.3.2.3 of the manuscript) and considering the feasible scenario of recovering 70 % of the dissolved methane. Finally, net energy demand (NED) was calculated considering both EC and TER:

$$NED = EC+TER \qquad \text{Equation 6.6S}$$

Theoretical energy consumption of pumps and blowers was calculated as follows, based on the equations proposed in Judd & Judd (2006):

$$P_B = \frac{(M \cdot R \cdot T_{gas})}{(\alpha - 1) \cdot \eta_{blower}} \left[\left(\frac{P_2}{P_1} \right)^{\frac{\alpha - 1}{\alpha}} - 1 \right] \qquad \text{Equation 6.7S}$$

where P_B (J·s^{-1}) is the blower power requirement (adiabatic compression), M (mol·s^{-1}) is the molar flow rate of biogas, R (J·mol^{-1}·K^{-1}) is the gas constant for biogas, P_1 (atm) is the absolute inlet pressure, P_2 (atm) is the absolute outlet pressure, T_{gas} (K) is the biogas temperature, α is the adiabatic index and η_{blower} is the blower efficiency.

$$P_g = q_{imp} \cdot \rho_{liquor} \cdot g \cdot \frac{\left\{ \left[\left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{asp} + \left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{imp} \right] + [Z_1 - Z_2] \right\}}{\eta_{pump}} \qquad \text{Equation 6.8S}$$

where P_g is the power required by a general pump (J·s^{-1}), considering both the pump aspiration and pump impulsion sections, calculated from the impulsion volumetric flow rate ($q_{imp.}$ in

$m^3 \cdot s^{-1}$), liquor density (ρ_{liquor} in $kg \cdot m^{-3}$), acceleration by gravity (g in $m \cdot s^{-1}$), pipe length (L in m), equivalent pipe length of accidental pressure drops (L_{eq} in m), velocity (V in $m \cdot s^{-1}$), friction factor (f, dimensionless), diameter (D in m), difference in height (Z_1-Z_2, in m) and pump efficiency (η_{pump}). The friction factor (f) was calculated by means of Swamee-Jain expression (Eq. (9)).

$$f = \frac{0.25}{\left[\log_{10}\left(\frac{k/D}{3.7} + \frac{5.74}{Re^{0.9}}\right)\right]^2}$$ Equation 6.9S

where Re is the Reynolds number, k (m) is the pipe roughness, and D (m) is the pipe diameter.

Equation 8 was modified to calculate power requirements for permeate pumping (Eq. (6.10S))

$$P_{stage} = \frac{q_{stage} \cdot TMP_{stage}}{\eta_{pump}}$$ Equation 6.10S

where P_{stage} is the power requirement during filtration, degasification or back-flushing ($J \cdot s^{-1}$) calculated from average transmembrane pressure (TMP_{stage} in Pa), pump volumetric flow rate (q_{stage} in $m^3 \cdot s^{-1}$), and pump efficiency (η_{pump}). Total Energy Consumption (TEC) was calculated by the total power requirements of each individual piece of equipment.

Total CO_2 emissions from the AnMBR system were thus calculated as the addition of the two different sources (see Eq (6.11S)) and expressed in $kgCO_2 \cdot m^{-3}$:

$$CO_2 \text{ emissions} = CO_{2_{D-CH_4}} + CO_{2_{NED}}$$ Equation 6.11S

where $CO_{2_{D-CH_4}}$ was calculated following Eq. (6.12S):

$$CO_{2_{D-CH_4}} = \frac{Q_{eff} * CH_{4\,eff} * 34}{Q_{inf}}$$ Equation 6.12S

Where Q_{eff} is the effluent flow rate ($m^3 \cdot d^{-1}$), $CH_{4\,eff}$ represents the dissolved methane concentration in the effluent after the degassing membrane ($kg \cdot m^{-3}$), Q_{inf} is the influent flow rate ($m^3 \cdot d^{-1}$), and 34 $kgCO_{2eq} \cdot kg\ CH_4^{-1}$ is the characterization factor applied to CH_4 emissions from the effluent, according to Huijbregts et al., (2017). Indirect CO_2 emission related to the NED of the system ($CO_{2_{NED}}$) were calculated considering a Spanish electricity-specific emission factor of 0.14 $kgCO_2 \cdot kWh^{-1}$ in 2021 (REE).

6.5.5. Average values for J_{20}, SGD_P and TSS

Table 6.1S. Average values of J_{20}, SGD_P and TSS for the three membrane tanks during the experimental period.

	Time (d)	J_{20} (LMH) Mean	±	SD	SGD_P ($Nm^3 \cdot m^{-3}$) Mean	±	SD	TSS $g \cdot L^{-1}$ Mean	±	SD
MT_A	35-48	17.8	±	1.5	15.1	±	4.4	8.3	±	0.1
	49-90	18.6	±	0.6	14.4	±	3.1	11.9	±	3.2
	94-122	19.2	±	0.9	14.3	±	2.2	12.6	±	0.4
	219-258	15.4	±	0.4	21.3	±	5.6	10.2	±	1.6
	259-300	15.6	±	0.2	18.2	±	1.3	9.0	±	1.4
	373-408	14.9	±	1.3	6.8	±	3.4	10.2	±	0.8
	409-421	13.5	±	2.5	3.0	±	2.7	11.1	±	0.7
MT_B	1-34	21.0	±	0.4	14.1	±	0.8	8.2	±	0.6
	35-48	18.2	±	0.2	14.8	±	3.7	8.3	±	0.1
	49-90	18.5	±	0.5	13.7	±	3.7	11.9	±	3.2
	94-122	19.8	±	0.8	13.9	±	1.0	12.6	±	0.4
	134-164	23.0	±	0.8	15.8	±	1.2	11.6	±	1.4
	171-200	20.0	±	0.5	16.5	±	2.5	10.6	±	2.0
	216-250	16.8	±	0.8	17.4	±	2.1	10.7	±	1.4
	251-305	18.5	±	0.6	14.4	±	3.9	9.2	±	1.2
	306-350	17.2	±	0.5	12.3	±	6.7	11.7	±	0.7
	365-440	14.7	±	1.2	7.5	±	3.9	10.0	±	1.0
	445-512	15.9	±	1.6	2.2	±	1.2	9.0	±	0.9
MT_C	296-363	18.0	±	1.1	13.5	±	1.6	10.8	±	1.3
	364-399	15.2	±	0.6	8.0	±	5.7	10.3	±	0.5
	403-418	14.5	±	0.2	10.5	±	7.5	11.5	±	0.2
	428-441	14.7	±	0.1	5.3	±	5.1	9.0	±	0.3
	443-468	14.3	±	0.8	4.0	±	3.3	9.3	±	0.9
	469-509	16.9	±	0.8	3.9	±	4.1	9.0	±	0.9
	510-575	18.9	±	0.9	11.5	±	3.0	7.7	±	1.9

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7. Life cycle costing of AnMBR technology for urban wastewater treatment: a case study based on a demo-scale AnMBR system

Abstract

This study aims at assessing the economic performance of a projected full-scale anaerobic membrane bioreactor (AnMBR) for urban wastewater (UWW) treatment at ambient temperature. To this aim, data from an AnMBR demonstration plant (industrial prototype, TRL 6) was used, which was operated for 3 years treating real UWW, allowing gathering a robust set of information for scaling-up to full scale. The obtained results revealed that reactor mixing (0.056-0.124 kWh·kgCOD_{rem}^{-1}; 34-57 %), and membrane scouring (0.048-0.120 kWh·kgCOD_{rem}^{-1}; 22-48 %) were the main contributors to the total energy demand; while net energy productions between 0.210 and 0.645 kWh·kgCOD_{rem}^{-1} were achieved. Capital expenditure was highly influenced by UF membranes (€0.029-0.073 kgCOD_{rem}^{-1}; 31-49%), combined heat and power technology for energy recovery (€0.012-0.023 kgCOD_{rem}^{-1}; 8-24%), and reactor construction (€0.07-0.014 kgCOD_{rem}^{-1}; 8-13%); while the main contributors to operating expenditure were energy requirements (€0.042-0.069 kgCOD_{rem}^{-1}; 41-46 %), membrane replacement (€0.011-0.028 kgCOD_{rem}^{-1}; 9-17 %), and discharge fee (€0.010-0.020 kgCOD_{rem}^{-1}; 9-12 %). Total annualized costs showed high variability, between €-0.003 and 0.188 kgCOD_{rem}^{-1}. Results presents AnMBR as a competitive technology for UWW treatment compared to conventional aerobic technologies (e.g., CAS). Membrane fouling control; hydraulic retention time; biogas requirements for reactor mixing and membrane stirring; and energy recovery efficiency were identified as key parameters for improving economic sustainability of AnMBR technology.

Keywords

Municipal wastewater treatment; semi-industrial scale system; prototype AnMBR; economic assessment; life cycle assessment; sewage

Highlights

Economic feasibility of a projected full-scale AnMBR was evaluated.
Net energy productions up to 0.645 kWh·kgCOD_{rem}^{-1} were achieved.
CAPEX varied between €0.080 and 0.147 kgCOD_{rem}^{-1}.
OPEX was in the €-0.099-0.041 kgCOD_{rem}^{-1} range.
TAC between €-0.003 and 0.188 kgCOD_{rem}^{-1} was obtained.

Jiménez-Benítez, A., Ruiz-Martínez, A., Ferrer, J., Ribes, J., Rogalla, F., Robles, A., 2023a. Life cycle costing of AnMBR technology for urban wastewater treatment: A case study based on a demo-scale AnMBR system. J. Environ. Chem. Eng. 11. https://doi.org/10.1016/j.jece.2023.110267

7.1. Introduction

Water management is one of the fields in which new technologies, concepts and approaches are being actively investigated to successfully face the climate change threat, meet Circular Economy (CE) principles and accomplish Sustainable Development Goals (Daniel Puyol et al., 2017). It is therefore crucial to progress towards converting former wastewater treatment plants (WWTP) into full water resource recovery facilities in which urban wastewater (UWW) is regarded as a raw material and no longer as a waste product (Guest et al., 2009). This paradigm shift would take advantage of the value embedded in wastewater and boost the economic framework linked to its management.

Among the possibilities for improving the economic performance of wastewater treatment facilities, the combination of anaerobic digestion (AD) and membrane bioreactors (MBR), resulting in the so-called anaerobic membrane bioreactor (AnMBR), is gaining momentum (A. Jiménez-Benítez et al., 2023; Ozgun et al., 2013; Robles et al., 2022). Reduced growth rates of anaerobic microorganisms can be compensated, even when operating at low reactor temperatures, since an AnMBR can decouple the sludge retention time (SRT) from the hydraulic retention time (HRT) by membrane filtration. The additional benefits of AnMBR also include:

- **Effluent quality:** membrane pore size can ensure microbial quality in the effluent (Skouteris et al., 2012), favoring the production of fit-for-purpose reclaimed water.
- **Organic matter valorization:** anaerobic metabolism converts biodegradable organics into methane, which acts as an energy carrier and enhances the energy balance of wastewater treatment (Smith et al., 2012).
- **Biosolids production and stabilization:** anaerobic metabolism reduces sludge waste management needs due to the low growth yield of anaerobic biomass, while onsite sludge stabilization can be achieved by operating at a high SRT (Seco et al., 2018).
- **Nutrient recovery:** anaerobic systems conserve most of the mineralized nutrients embedded in the effluent, which favors their recovery (Á. Robles et al., 2020a) or direct reuse through fertigation (Jiménez-Benítez et al., 2020a).
- **Reduced footprint:** the MBR configuration eliminates settling units, which reduces the investment costs associated with land for new facilities and favors the retrofitting of existing plants with space constraints (Xiao et al., 2019).

However, AnMBR technology for UWW treatment still faces some operating issues that need to be properly solved. The main bottlenecks for widening the implementation of AnMBR are: sensitivity to process dynamics and development of appropriate control strategies (Robles et al., 2018); membrane fouling (Hu et al., 2020); and loss of dissolved methane with the effluent (Velasco et al., 2018), which worsen the energy balance and increase the carbon footprint.

Despite the aforementioned challenges, AnMBR has continuously improved its performance, e.g., in terms of effluent quality or maintenance of competitive transmembrane fluxes (Á. Robles et al., 2020b). However, from an economic perspective, energy demand needs to be further optimized for enhancing its feasibility. Membrane fouling control through gas scouring has been reported as one of the main contributors to energy costs in AnMBR systems (A. Jiménez-Benítez et al., 2023; Pretel et al., 2016d). Chemical demands for membrane cleaning and membrane replacement also contribute to operating expenses (OPEX) of AnMBR technology (Judd, 2017). Sludge handling and management is also becoming an emerging global problem (Gherghel et al., 2019), so that the reduced sludge production of AnMBRs, jointly with the energy produced through biodegradable organics, can offset the operating costs related to membrane operation and favor the financial framework of AnMBRs against conventional aerobic treatments.

Robust comparisons between different treatment processes and/or operating conditions still represent a complex challenge, especially in the emerging technologies, due to the wide range of assumptions considered, different influent/effluent quality and methodologies applied. In this regard, Ferrer et al. (2015) proposed a method of AnMBR design and assessment and obtained a total annual cost (TAC) including CAPEX and OPEX of between €0.101 and €0.097 per m^3 for sulfate-rich UWW and between €0.097 and €0.070 per m^3 for low-sulfate UWW. Using the same method, Pretel et al. (2016b) calculated a TAC of €0.126-0.169 per m^3 for an AnMBR combined with CAS or MBR-based post-treatment for nutrient removal. These values are in line with those reported by Smith et al. (2014) for an AnMBR treating medium- and high-strength wastewater (around €0.124 per m^3). These authors also studied the influence of sludge fate (landfill, incineration, and land application) and concluded that AnMBR costs are lower than high rate activated sludge+AD and CAS+AD when sludge is landfilled, lower than CAS+AD when it is incinerated and lower than AeMBR+AD in any scenario. Xu et al. (2020) reported significant differences between AeMBR and AnMBR when water and nutrient reuse is considered: in a discharge scenario, the net present value (NPV) of an AeMBR was $11 million lower than AnMBR, but in a reuse scenario fertilizer savings would allow a NPV $41 million lower for AnMBR. Furthermore, tertiary treatments for disinfection are unlikely to be necessary when

ultrafiltration membranes are used, contributing to avoiding or reducing the costs associated with conventional disinfection processes, e.g., UV radiation, chlorination, etc. In this regard, boosting water reuse can also provide revenues for water management systems that could make AnMBR economic performance more attractive.

These results confirm a promising trend, especially when they are compared with the values reported for conventional and aerobic treatments. For example, Iglesias et al. (2017) analyzed the costs of full-scale MBRs and extended aeration (EA) facilities in Spain. These authors calculated an OPEX for the studied MBR in the range of €0.20-0.45 per m^3 and an average of €0.22 per m^3 in case of EA. When EA was combined with reclaimed treatments, operating costs rose to €0.31-€0.40 per m^3. Total costs for general MBR technology at more than 50 % of its treating capacity was estimated to be €0.20-0.50 per m^3. On the other hand, Xiao et al. (2019) reported an OPEX of $0.11-0.20 per m^3 for aerobic MBR in China. EPSAR, the Public Entity for Sewage Treatment in the *Comunitat Valenciana* (Spain), reported €0.33 per m^3 as the average OPEX for all the technologies implemented in this region, based on different aerobic treatments for different treatment capacities (EPSAR, 2021).

This work aimed to assess the economic performance of a full-scale AnMBR for UWW using Life Cycle Costing (LCC) methodology, which has been widely applied to economic appraisals considering all the costs of constructing, operating, maintaining and disposing of products, processes and services (Kloepffer, 2008). The results were obtained using the data from an AnMBR demonstration plant treating UWW under different operating conditions within the LIFE MEMORY Project (A. Jiménez-Benítez et al., 2023; Robles et al., 2022). Given the size of the demonstration plant (equipped with commercial ultra-filtration hollow-fiber membranes, industrial pumping and auxiliary elements and control and monitoring devices) and the long operating period (around 3 years with different ambient conditions), data obtained from the plant can be considered as a robust set of information for scaling-up AnMBR to full-scale facilities and favor the adoption of AnMBR and its advantages over conventional UWW treatments.

7.2. Materials and methods

7.2.1. AnMBR plant

The demonstration plant comprised a 40 m^3 anaerobic reactor (AnR) (34.4 m^3 working volume) connected to three x 0.8 m^3 membrane tanks (MT-X) (0.7 m^3 working volume), equipped with

one ultrafiltration membrane system (PURON® PSH41, 0.03-µm pore size and 41m^2 total filtration area). The total system filtration area is 123 m^2. The plant also includes a rotofilter (RF, 1.5-mm screen size), an equalization tank (ET, 1.1 m^3), a clean-in-place (CIP) tank (0.37 m^3), and degassing membranes (DM) to recover the methane dissolved in the effluent (further details of DM in Sanchis-Perucho et al. (2020)).

For energy calculations, data regarding flow rate, liquid level, pressure (transmembrane, inlet and outlet for blowers, and gas), temperature and methane production was obtained from the available plant instrumentation. Data obtained from the DM unit (transmembrane pressure, flow rates, and methane recovery efficiency) were also used to estimate the dissolved methane recovery potential of the system, according to Sanchis-Perucho et al. (2020). Adiabatic indexes and blower and pump efficiencies were estimated from the standard values for this equipment. Further details of the plant can be found in Robles et al. (2020b). The flow diagram and an overview of the AnMBR are provided in supplementary material.

7.2.2. AnMBR operation

The AnMBR prototype treated effluent from the pre-treatment step (screening and sand removal) of the "Alcázar de San Juan" WWTP. After pre-treatment in the RF and homogenization in the ET, the wastewater was pumped to the AnR. The sludge was continuously recycled through the external membrane tanks (immersed configuration), where the final effluent was obtained by vacuum filtration. A fraction of the biogas produced in the AnR was constantly recirculated from the headspace to the bottom of the reactor through coarse bubble diffusers for stirring purposes. This mixing strategy favored the stripping of dissolved gases from the liquid phase for their collection. Another fraction of the biogas produced was driven to the bottom of the membrane tanks for membrane scouring.

The membrane operating strategy followed a sequence of 300-s basic F-R cycle (280 s filtration and 20 s relaxation), 40 s of back-flush every 5 F-R cycles, 20 s of ventilation every 15 F-R cycles and 40 s of degasification every 50 F-R cycles.

The selected average operating conditions and characterization of the five scenarios evaluated in this study are provided in supplementary material (see Table 7.1S) and further details can also be found in Robles et al. (2022) and Jiménez-Benítez et al. (2023). Influent quality was characterized

by a high COD (755±224-1403±532 $mg \cdot L^{-1}$) and COD:SO_4^{-2}-S ratios between 4.80±2.86 and 8.70±3.69 due to dairy- and wine-industry contributions. SRT of the demo-plant was set to 70 days, based on previous studies on AnMBR technology for urban wastewater treatment at ambient temperature. By achieving high concentrations of biomass in the reactor, an appropriate level of biodegradability can be ensured even at low temperatures. This has been validated in previous studies (e.g., Giménez et al. (2011)). However, to prevent the excessive concentrations of suspended solids in the mixed liquor that could negatively affect the filtration process (A. Robles et al., 2012a), HRT was set at values above 20 h. This allowed maintaining mixed liquor solids concentrations compatible with competitive filtration process performances.

Scenarios I to V were used to scale and estimate the performance of a full-scale AnMBR system that would treat the influent to the "Alcázar de San Juan" WWTP (i.e., 16000 $m^3 \cdot d^{-1}$). From this scaling, both the energy balance and the LCC were evaluated.

Regarding the biological process, an average COD removal efficiency of 87.2±6.1 % was achieved while effluent COD concentration remained below its discharge limits according to European regulations (<125 $mg \cdot L^{-1}$) despite treating medium-high organic load rates (OLR) (0.60±0.20-1.28±0.38 kg $COD \cdot m^{-3} \cdot d^{-1}$). Biogas production varied widely throughout the studied periods (1359±999-5558±3204 STP $L \cdot d^{-1}$), but the methane percentage remained high and stable (75±2-77±2 %).

The filtration process performance demonstrated that it was possible to operate the membranes at competitive 20ºC-standardized gross transmembrane fluxes ($J_{20\ gross}$), 15-25 $L \cdot m^{-2} \cdot h^{-1}$, while maintaining low membrane fouling propensities (0.3-4.9 $mbar \cdot d^{-1}$) by applying specific gas demands per volume of permeate produced (SGD_P) of 10-20 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$ (see Jiménez-Benítez et al. (2023)). These SGD_p were slightly lower than those reported for aerobic MBR by Iglesias et al. (2017) (18.3-24.32 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) and to those applied by Pretel et al. (2014) to an AnMBR (14-32 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$), both also equipped with hollow fiber membranes.

7.2.3. Energy assessment

The energy balance was calculated according to the equations proposed in Judd and Judd (2006) and Pretel et al. (2016a) (see supplementary material). The equipment considered for power energy requirements ($W_{elements}$) included rotofilter, pumps (feeding, recycling and permeating),

blowers and a sludge dewatering system. Heat energy requirements (Q) were not considered in the study since the full-scale AnMBR was assumed to operate at ambient temperature, as per the demo-scale AnMBR.

Combined Heat and Power (CHP) technology was considered for energy recovery from biogas. Since none of the available CHP systems is suitable for low biogas productions, W_{biogas} was estimated from the biogas flow rate Q_{biogas} ($L \cdot d^{-1}$), methane percentage in the biogas $\%_{CH4}$, and methane calorific power CV_{CH4} ($kJ \cdot m^{-3}$). In this case, the widely used reciprocating engines were selected as CHP. These systems can achieve high power and overall CHP efficiencies (24-36 % and 66-71 %, respectively) according to Darrow et al. (2017). Equation 7.1 gives the energy from the biogas recovered in terms of power (W_{biogas} in kW):

$$W_{biogas} = \frac{Q_{biogas} \cdot \%_{CH4} \cdot CV_{CH4} \cdot \%_{power\ efficiency\ CHP}}{1000 \cdot 24 \cdot 3600} \qquad \text{Equation 7.1}$$

An average $\%_{power\ efficiency\ CHP}$ of 35% was selected in this study.

Table 7.1S (in supplementary material) shows the biogas flow rate and the biogas methane percentage for each scenario. The concentration of dissolved methane in the effluent was calculated from Henry's Law. Saturation conditions were considered for all evaluated temperatures (see Giménez et al. (2012)), since coarse bubble diffuser stirring favors methane stripping from the liquid phase thus avoiding oversaturation. The percentage of recovered methane was calculated following previous studies carried out in the demonstration plant (Sanchis-Perucho et al., 2020).

7.2.4. Life cycle costing

The economic feasibility of the projected full-scale AnMBR was assessed by means of the experimental data provided by the prototype through LCC, which was used to calculate the total annual costs (TAC), defined as the sum of the capital and operating expenses (CAPEX and OPEX, respectively) as in Equation 7.2:

$$TAC = \frac{r\,(1+r)^t}{(1+r)^t - 1} \cdot CAPEX + OPEX \qquad \text{Equation 7.2}$$

where *r* is the annual discount rate and *t* is the depreciation period in years.

A functional unit of 1 kg of removed COD ($kgCOD_{rem}$) was selected for the economic assessment, according to the main pollutant target of AnMBR technology. A depreciation period of 20 years for building works (annual discount rate 3%) and 10 years for equipment (annual discount rate 5%) was used to calculate the CAPEX. For OPEX, the energy costs considered both biogas and dissolved methane recovery by DM. Chemicals for membrane cleaning (sodium hypochlorite and citric acid) were included in OPEX appraisals. Since chemical cleaning degrades membranes, the UF membrane lifespan was calculated considering the cleaning frequency requirement obtained during the demo-scale AnMBR operation and the permissible maximum total contact with chlorine before membrane replacement, i.e., 500000 ppm-hours cumulative, or a maximum of 10 years, according to the supplier. The lifetime of blowers and pumps was based on the total working hours recommended by the manufacturers (50000 and 75000 h, respectively) or also set to a maximum of 10 years. When possible, equipment costs were estimated by correlations between capacities (mainly based on flow rates) and the unit costs provided by the manufacturers, while labor costs were not included. LCC was composed of construction and operating costs over 20 years. Dismantling was not included.

Further considerations when calculating CAPEX and OPEX can be found in the supplementary material. Table 7.1 shows the unit costs and correlation functions used to assess CAPEX and OPEX in the proposed scenarios.

7.2.5. Sensitivity and uncertainty analyses

Sensitivity (SA) and uncertainty analyses (UA) were carried out to determine the most significant parameters and the related uncertainty in the obtained results. In this regard, for the purpose of SA, the Standardized Regression Coefficients (SRC) method was applied to select the most influential parameters. A threshold of 0.1 (absolute value) for the standardized regression slope (b_i) was chosen to select the significant factors. The UA was conducted by the Monte Carlo method to evaluate the propagation of uncertainty in the results, which were assessed by means of i) the 5th and 95th percentiles and ii) the empirical cumulative distribution function (eCDF). 2000 model runs were performed for both SA and UA. The sampling matrix was generated by the Latin hypercube sampling (LHS) method with a variability of 5 % for the membrane and equipment cost quoted by manufacturers and 10 % for the rest of parameters considered (further

Table 7.1. Unit costs and correlation functions used to evaluate capital and operating expenses (CAPEX/OPEX) in the scenarios evaluated.

Unit costs and correlation function of capital and operating expenses		
Pipes, (final cost is function of diameter) €·m^{-1} [a]		8-220
Concrete wall/Floor&cover, €·m^{-3}		350/130
Steel cover, €/m^2 [b]		51.11
Ultrafiltration hollow-fiber membrane, €·m^{-2} [c]		€·m^{-2} = -2.985·ln A (m^2) + 68.159
Energy	Energy term €·kWh^{-1} [d] Power term €·kW^{-1}·year^{-1} [d]	0.231-0.340 1.57-24.73
Sodium hypochlorite (NaOCl Cl active 5%), €·L^{-1}		0.72
Citric acid (Citric acid 1-hidrate), €·kg^{-1}		2.96
Polyelectrolyte, €·kg^{-1}		2.35
Wasted sludge for farming, € per t		4.81
Wasted sludge for landfill, € per t		30.05
Wasted sludge for incineration, € per t		250
Blower, €·unit^{-1} [e]		€·unit^{-1} = 970.66·Q (Nm^3·h^{-1})$^{0.6008}$
Water pumps, €·unit^{-1} [f]		€·unit^{-1} = -0.6582·Q^2 (m^3·h^{-1}) + 263.54·Q (m^3/h) + 1,005.4
Sludge pumps, €·unit^{-1} [g]		€·unit^{-1} = 1,071.1·ln Q (m^3·h^{-1}) + 19,643
Rotofilter, €·unit^{-1} [h]		€·unit^{-1} = 13.831·Q (m^3·h^{-1}) + 2,220.8
Thick-bubble diffuser with 4" Membrane Disc Aeration Head (Barmatec S.L.), €·unit^{-1}		16.94
Dewatering system (centrifuge), €·unit^{-1} [i]		€·unit^{-1} = 7,246.1·ln Q (m^3·h^{-1}) + 93,099
CHP, reciprocating engine	€·unit^{-1} [j] O&M €·kWh^{-1} [j]	€·unit^{-1} = -2·10^{-9}·kW^3+5·10^{-5}·kW^2-0.4842·kW· €·kWh^{-1} = 0.0192·e$^{-0.0001·kWh}$
PermSelect® Silicone Membrane Module, €·m^{-2}		37.10

[a] Frans Bonhomme España
[b] BEDEC (iTeC)
[c] Koch Membrane System
[d] Energy and power tariff 6.1 TD. Entidad Pública de Saneamiento de Aguas Residuales de la Comunidad Valenciana (EPSAR). Public Entity for Sewage Treatment of Comunitat Valenciana
[e] Pedro Gil S.L.; Tebyc S.L.; FPZ
[f] Sugein S.L.; Tebyc S.L.
[g] Bombas Ideal S.A.
[h] Drainfilter S.L.
[i] Hutchison Hayes Separation Inc.
[j] U.S. Department of Energy: Combined Heat and Power Technology-Fact Sheet Series

details can be found in supplementary material). OPEX, CAPEX and TAC were used as outputs for SA and UA assessment.

7.3. Results and discussion

7.3.1. Energy assessment

Total energy consumption (TEC), total energy recovery (TER) and net energy demand (NED) of the projected full-scale AnMBR plant were evaluated for each scenario (see Figure 7.1 and Table 7.2). These results are discussed in the following sections.

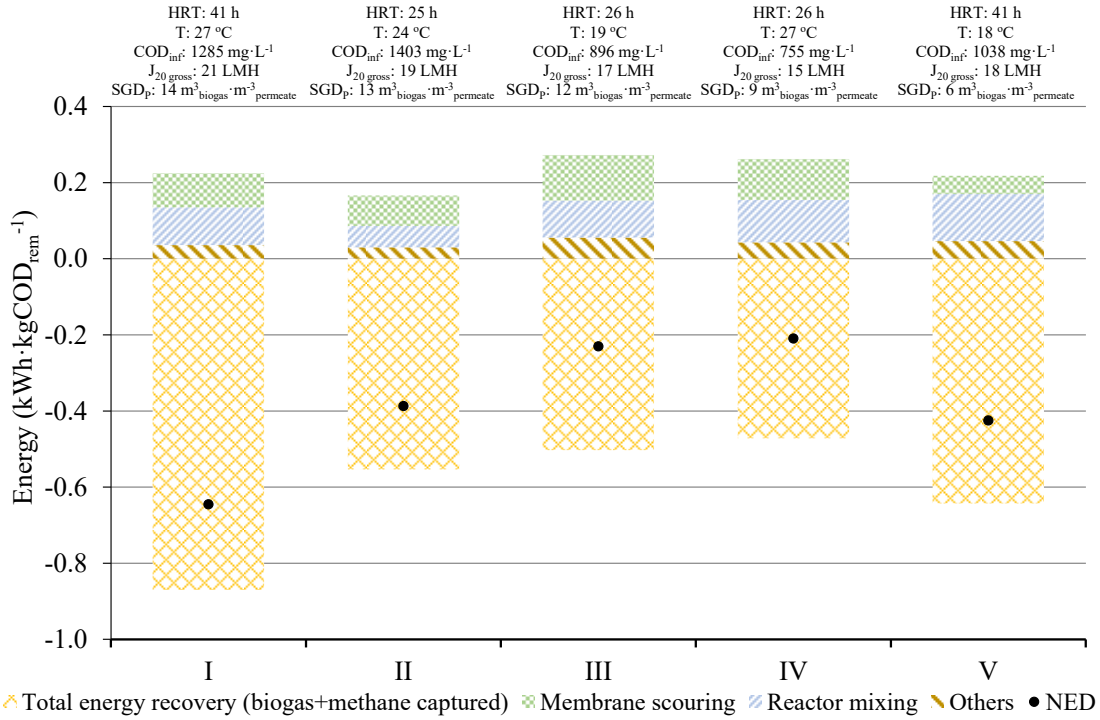


Figure 7.1. Total energy consumption (TEC), total energy recovery (TER) and net energy demand (NED) for the projected full-scale AnMBR plant. "Others" refers to: rotofilter; feeding and permeate pumping; sludge waste, recycling and dewatering and methane recovery blowers.

7.3.1.1. Total energy consumption

Table 7.2 shows that the main contributors to the TEC in all the scenarios were reactor mixing (34-57 %) and membrane scouring (22-48 %). Energy inputs related to methane recovery blowers (0.3-0.5 %), rotofilter (0.5-0.7 %) and waste sludge pumping (0.1 %) were negligible. Moderate energy inputs were required for sludge dewatering (5-9 %), feed pumping (1-2 %), permeate pumping (2-8 %) and sludge recycling (3-4 %).

Table 7.2. Power energy requirements, total energy recovery (TER), total energy consumption (TEC), net energy demand (NED) and % of TEC for each scenario.

Scenario	Rotofilter	Feeding pumping	Permeate pumping	Membrane scouring	Sludge wasting	Sludge recycling	Sludge dewatering	Reactor mixing	Dissolved methane vacuum blower	TER (biogas + dissolved methane capture)	TER (only biogas)	TEC	NED
						$kWh \cdot kgDQO_{rem}^{-1}$							
I	0.001	0.002	0.010	0.089	≈ 0.000	0.006	0.016	0.099	0.001	-0.870	-0.827	0.225	-0.645
II	0.001	0.003	0.010	0.080	≈ 0.000	0.006	0.009	0.056	0.001	-0.553	-0.513	0.166	-0.387
III	0.002	0.005	0.022	0.120	≈ 0.000	0.010	0.016	0.097	0.001	-0.503	-0.429	0.273	-0.230
IV	0.002	0.006	0.005	0.108	≈ 0.000	0.011	0.018	0.112	0.001	-0.472	-0.386	0.262	-0.210
V	0.001	0.003	0.014	0.048	≈ 0.000	0.008	0.019	0.124	0.001	-0.643	-0.581	0.218	-0.425
						TEC (%)							
I	0.5	1.0	4.4	39.8	0.1	2.8	6.9	44.3	0.3				
II	0.6	1.7	6.3	48.2	0.1	3.6	5.4	33.8	0.3				
III	0.6	1.8	8.1	44.1	0.1	3.6	5.7	35.6	0.5				
IV	0.7	2.1	1.9	41.0	0.1	4.3	6.8	42.6	0.4				
V	0.6	1.3	6.3	21.9	0.1	3.6	8.9	56.8	0.5				

Scenario II required the lowest energy consumption (TEC of 0.166 kWh·kgCOD$_{rem}^{-1}$), mainly due to its comparatively low sludge dewatering, reactor mixing, and membrane scouring requirements (0.009 kWh·kgCOD$_{rem}^{-1}$, 0.056 kWh·kgCOD$_{rem}^{-1}$ and 0.080 kWh·kgCOD$_{rem}^{-1}$, respectively). These results were influenced by the highest COD removal efficiency (equivalent to 20631 kgCOD·d^{-1} at industrial scale) and the lowest HRT (25±1 h) within the operating conditions evaluated in this work. On the other hand, Scenario III required the highest energy consumption (TEC of 0.273 kWh·kgCOD$_{rem}^{-1}$) due to its lower COD removal capacity (12428 kgCOD·d^{-1} at industrial scale), which increased the energy requirements per kg of COD removed, especially for membrane scouring (0.120 kWh·kgCOD$_{rem}^{-1}$) and reactor mixing (0.097 kWh·kgCOD$_{rem}^{-1}$), even though its HRT (26±2 h) was almost the same as that of Scenario II.

The permeate pumping results also highlighted the importance of maintaining not only low fouling rates (FRs), but also low TMP. For example, although Scenarios III and IV operated at low FR (0.3 and 0.5 mbar·day^{-1}, respectively), the average TMP in Scenario IV (87 mbar) was significantly lower than in Scenario III (462 mbar), resulting in a reduction of the energy consumption from 0.022 kWh·kgCOD$_{rem}^{-1}$ to 0.005 kWh· kgCOD$_{rem}^{-1}$ (77 % less), associated with permeate pumping. It is important to consider that filtration performance is influenced by several inter-related biological, physical and chemical factors (e.g., temperature, shear rate, SRT, HRT, extracellular polymeric substances and soluble microbial products concentrations, operation mode, etc.) (Orhon, 2013). The higher temperature (27±1 ºC) and lower mixed liquor total suspended solids (MLTSS) concentration (10.4±0.8 g·L^{-1}) in Scenario IV could have led to better sludge filtration properties compared to Scenario III (temperature 19±1ºC and 11.3±1.0 g·L^{-1} MLTSS). Additionally, both Scenarios I and IV operated at similar temperature (27±1ºC) and lower MLTSS in Scenario I than in Scenario IV (8.4±0.5 and 10.4±0.8 g·L^{-1}, respectively), but TMP was significantly higher in Scenario I (318 mbar) than in Scenario IV (87 mbar). Energy consumption for permeate pumping in Scenario I (0.010 kWh· kgCOD$_{rem}^{-1}$) was hence double that of Scenario IV (0.005 kWh· kgCOD$_{rem}^{-1}$) even though COD removal in Scenario IV (10815 kgCOD$_{rem}$·d^{-1}) was significantly lower than in Scenario I (19138 kgCOD$_{rem}$·d^{-1}).

Membrane scouring has been classically identified as a key parameter of AnMBR technology (Shin and Bae, 2018). Scenarios I and II performed with similar high COD removal (19138 and 20631 kgCOD$_{rem}^{-1}$·d^{-1}, respectively) and were operated with similar SGD$_p$ (14 and 13 Nm$^3_{biogas}$·m$^{-3}_{permeate}$, respectively). Both showed therefore low energy consumption associated with membrane scouring: 0.089 kWh·kgCOD$_{rem}^{-1}$ in Scenario I and 0.080 kWh·kgCOD$_{rem}^{-1}$ in

Scenario II. On the other hand, Scenarios II and III were both operated at a similar average SGD_p of 12 and 13 $Nm^3biogas \cdot m^{-3}_{permeate}$ respectively but had different COD removal performance (20631 and 12428 $kgCOD_{rem} \cdot d^{-1}$, respectively). Therefore, Scenario II showed lower energy consumption regarding membrane scouring (0.080 $kWh \cdot kgCOD_{rem}^{-1}$) than Scenario III (0.120 $kWh \cdot kgCOD_{rem}^{-1}$). Finally, Scenario V was operated with the lowest SGD_p (6 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) and performed with an intermediate COD removal rate (15376 $kgCOD_{rem} \cdot d^{-1}$). Scenario V therefore had the lowest energy expenditure for membrane scouring (0.048 $kWh \cdot kgCOD_{rem}^{-1}$).

These results confirm that the challenging issue of optimizing energy consumption in AnMBR performance depends on multiple factors. Biological conditions in the reactor should ensure high COD removal, not only for accomplishing the regulations, but also to better reach high energy efficiencies. Optimizing HRT to reduce reactor mixing expenditures and designing a comprehensive filtration strategy able to maintain low TMP are crucial to reducing TEC. This strategy should also consider appropriate combinations of $J_{20\ gross}$ and SGD_p.

7.3.1.2. Total energy recovery

Table 7.2 shows the TER when energy is recovered from both: only biogas; and biogas and dissolved methane captured from the effluent. The results confirmed the potential of AnMBR for organic matter valorization via methane production, although the energy production values varied widely. Scenarios I and V showed the highest energy recoveries (-0.827 and -0.581 $kWh \cdot kgCOD_{rem}^{-1}$, respectively), while III (-0.429 $kWh \cdot kgCOD_{rem}^{-1}$) and IV (-0.386 $kWh \cdot kgCOD_{rem}^{-1}$) had lower energy productions. Scenario II showed an intermediate result (-0.513 $kWh \cdot kgCOD_{rem}^{-1}$).

These energy outcomes were influenced by both the influent characteristics and the operating conditions (see Table 7.1S in supplementary material). In Scenarios I and II, the high COD:SO_4^{-2}-S ratios (8.7±3.7 and 8.2±4.4) and temperatures (27±1ºC and 24±2ºC) resulted in organic matter conversions with methane yields (Y^{CH4}) of 169.0±95.1 L $STP \cdot kgCOD_{rem}^{-1}$ (Scenario I) and 109.6±70.0 L STP· $kgCOD_{rem}^{-1}$ (Scenario II). Scenario IV showed a reduced methane yield of 70.2±36.0 L STP· $kgCOD_{rem}^{-1}$, even though it operated at a high temperature (27±1 ºC), due to its low COD:SO_4^{-2}-S ratio (4.8±2.9) and OLR (0.71±0.23 $kgCOD_{in} \cdot m^{-3} \cdot d^{-1}$). The comparison between Scenarios IV and V also depicted the importance of the COD:SO_4^{-2}-S ratio: both scenarios operated at similar OLR (0.71±0.23 and 0.60±0.20

$kgCOD_{in} \cdot m^{-3} \cdot d^{-1}$, respectively) but the lower COD:SO_4^{-2}-S ratio in Scenario IV (4.80±2.86) than in Scenario V (6.94±2.83) led the former to reduced methane productions (methane yield of 70.2±36.0 L STP·$kgCOD_{rem}^{-1}$) despite its high operating temperature (27±2ºC).

Hence, increasing the COD:SO_4^{-2}-S ratio or increasing the OLR available for methanogenic organisms appears to be a suitable approach to improve energy recovery. For this, additional co-substrates could be explored, such as adding food waste, as proposed by Moñino et al. (2017) or Galib et al. (2016). Operating at high temperatures also favors the conversion of organic matter into methane while reducing its losses in the effluent. Both effects contribute to improving energy recovery.

Recovering dissolved methane increased TER by 0.040-0.086 kWh·$kgCOD_{rem}^{-1}$, thus improving the energy balance while reducing the carbon footprint related to fugitive CH_4 emissions. The energy demands associated with the methane recovery blowers appeared to be negligible. These results confirm that degassing membranes are an attractive technology for enhancing AnMBR performance.

7.3.1.3. Net energy demand

As Table 7.2 shows, the best NED performance was achieved in Scenario I (-0.645 kWh·$kgCOD_{rem}^{-1}$). In this case, the relatively high reactor mixing energy costs (0.099 kWh· $kgCOD_{rem}^{-1}$) due to the high HRT (41±13 h) could be offset by the highest TER (-0.870 kWh·$kgCOD_{rem}^{-1}$) driven by its operation at higher ambient temperatures (27±1 ºC), OLR (0.75±0.25 $kgCOD \cdot m^{-3} \cdot d^{-1}$) and COD:$SO_4^{-2}$-S ratio (8.70±3.69) than the other scenarios. Scenarios II to V also performed as net energy producers with values between -0.210 kWh·$kgCOD_{rem}^{-1}$ (Scenario IV) and -0.425 kWh·$kgCOD_{rem}^{-1}$ (Scenario V).

By way of comparison, Hernández-Sancho et al., (2011) reported an average NED of 1.684±1.428 kWh·$kgCOD_{rem}^{-1}$ based on data provided by the EPSAR for 177 WWTP in Valencia (Spain), mainly obtained from conventional aerobic-based facilities. More recently, Silva and Rosa (2015) evaluated 17 Portuguese WWTPs based on extended aeration and CAS, and found an average of 1.3 kWh·$kgCOD_{rem}^{-1}$ and a 25-75 percentile range between 0.5 and 1.8 kWh·$kgCOD_{rem}^{-1}$. Gurung et al. (2018) calculated the NED for the CAS-based Finnish Mikkeli WWTP as 0.81 kWh·$kgCOD_{rem}^{-1}$, and identified AnMBR and co-digestion of sludge and

organic waste as suitable technological options for retrofitting the facility. Vaccari et al. (2018) assessed a total of 267 activated sludge WWTPs in Italy and obtained a range between 3.2 kWh·kgCOD$_{rem}^{-1}$ (treatment capacity < 2000 person-equivalent) and 0.85 kWh·kgCOD$_{rem}^{-1}$ (treatment capacity > 100000 person-equivalent). A similar study conducted in Greece (Christoforidou et al., 2020) concluded that WTTPs with above 100000 person-equivalent treatment capacities operated at a NED of 0.35 kWh·kgCOD$_{rem}^{-1}$ and 0.95 kWh·kgCOD$_{rem}^{-1}$ below 10000 person-equivalent. Barillon et al. (2013) reported net energy demands of six full-scale MBR with two membrane configurations (flat sheet and hollow fiber). The results indicated that energy consumption was approximately within the range of 0.7-2.1 kWh·kgCOD$_{rem}^{-1}$. Pretel et al. (2016b) simulated an aerobic MBR treating urban wastewater and obtained net energy demands between 0.99 and 1.02 kWh·kgCOD$_{rem}^{-1}$. Finally, Longo et al. (2016) gathered energy consumption data of 601 WWTP with different treatments schemes and reported average NED of 2.91, 1.30, and 0.57 kWh·kgCOD$_{rem}^{-1}$ for MBR, extended aeration, and CAS, respectively.

It is important to emphasize that the above-mentioned energy consumption from aerobic-based WWTPs also included nitrification, not occurring in an AnMBR treatment. According to Fraia et al. (2018), energy consumption for nitrogen removal would be 6.08 kWh per kg of influent total nitrogen (kWh·TN$_{in}$) in conventional WWTP with high nitrogen removal efficiency (> 89.2 %). Considering the influent nitrogen load in the five scenarios (35.6-54.7 mg N·L^{-1}), the aforementioned removal efficiency for this nutrient would be equivalent to an increase in energy consumption in the 0.202-0.370 kWh·kgCOD$_{rem}^{-1}$ range. NED for AnMBR with nitrogen removal would therefore still be lower than the energy requirements for conventional treatments.

The NED results obtained illustrate the potential of AnMBR systems for recovering resources from organic matter, even when treating sulfate-rich UWW, and for improving the energy efficiency of treatment facilities. The main drivers of the NED results are the OLR and COD removal capacity, HRT, temperature and the COD:SO_4^{-2}-S ratio. In this sense, a previous study focused on the biological performance of the demo-scale AnMBR (Robles et al., 2022) conducted a mass balance and concluded that the influent COD transformed into biogas was between 12 and 44 %, while dissolved methane account for 5-9 %. COD consumed by sulfate-reducing bacteria was calculated as 23-42 % and the organic matter removed via waste sludge 18-31 %. Therefore, there is still room to improve the recovery percentage and, consequently, the energy balance of AnMBR technology.

7.3.2. Life cycle costing

7.3.2.1. Capital expenditure

Figure 7.2 summarizes the CAPEX results obtained for the projected full-scale AnMBR system (detailed results and relative weights can be found in the supplementary material). These results showed the lowest value for Scenarios II (€0.080 $kgCOD_{rem}^{-1}$). Scenarios I (€0.096 $kgCOD_{rem}^{-1}$) and V (€0.109 $kgCOD_{rem}^{-1}$) presented similar investment cost, while Scenario IV obtained the highest CAPEX (€0.147 $kgCOD_{rem}^{-1}$).

UF membranes, anaerobic reactor building, blowers, and reciprocating engines were the main contributors to CAPEX. The reactor construction involved an investment of between €0.007 $kgCOD_{rem}^{-1}$ (Scenario II) and €0.014 $kgCOD_{rem}^{-1}$ (Scenario V), representing around 8-13% of the CAPEX. Since reactor volume is determined by the HRT selected in each scenario, there is clear room for optimization of savings in this constructive element. In this regard, Scenario II required the lowest reactor investment per kg of COD removed due to the combination of the highest COD removal (20631 $kgCOD \cdot d^{-1}$) and the lowest HRT (25±1 h).

In terms of equipment, the results obtained show that UF membranes were among the main contributors to CAPEX. The membrane acquisition cost varied from €0.029 to €0.073 $kgCOD_{rem}^{-1}$ (between 31 and 49 % of CAPEX). Strategies for maximizing filtration process productivity (e.g., optimizing transmembrane fluxes, gas sparging intensities for membrane scouring and downtime for physical membrane cleaning) are required to reduce membrane needs and minimize CAPEX. Enhancing material effectiveness to preventing membrane fouling and therefore maintaining high fluxes; improving membrane scouring procedures; revamping module configurations; and membrane market trends (i.e., reduction of membrane acquisition costs) would also help to reduce this item.

Investment in reciprocating engines also appeared as a relevant cost. Scenario I showed the highest CAPEX allocation for this equipment (€0.023 $kgCOD_{rem}^{-1}$) due to the highest biogas production (5239±4435 $m^3 \cdot d^{-1}$), which entails equipment with greater capacity. Scenario IV required the lowest capital expenditure for this unit (€0.012 $kgCOD_{rem}^{-1}$) because of its low biogas production (1359±999 $m^3 \cdot d^{-1}$). Investment in CHP varied between 8 % (Scenario IV) and 24 % (Scenario I) of CAPEX.

Blowers also showed a significant contribution to CAPEX. In this case, Scenarios III and IV had the highest expenditures, with €0.019 and €0.020 $kgCOD_{rem}^{-1}$, respectively. Despite their low operating HRT (26±2 h in both scenarios) and intermediate SGD_P (12 and 9 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$, respectively), their comparatively low organic matter removal resulted in a high blower cost per kg of COD removed. On the other hand, Scenario II with similar SGD_P (13 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) than Scenario III but the highest COD removal showed the lowest blower cost (€0.012 $kgCOD_{rem}^{-1}$). Investment in blowers was between 13 and 15 % of CAPEX.

Lastly, pumps (8-9 %), DM (4-6 %), centrifuge for sludge dewatering (2-3%), rotofilter (2-3 %), and pipes (1 %) contributed less to CAPEX while membrane tanks (0.2-0.3 %) and diffusers (0.1 %) were negligible in all scenarios.

Reducing CAPEX is thus crucial to improving investment expenditure competitiveness for AnMBR compared to aerobic MBR and CAS. The strategy to reduce CAPEX involves optimizing the HRT to adjust total reaction volume, increasing the filtration productivity to reduce the membrane needs, properly selecting the CHP power to avoid oversizing and optimizing the capacities of the blowers.

7.3.2.2. Operating cost

As can be seen in Figure 7.3, OPEX varied between €-0.099 $kgCOD_{rem}^{-1}$ (Scenario I) and €0.041 $kgCOD_{rem}^{-1}$ (Scenario IV) (detailed results and relative weights can be found in supplementary material). One of the main contributors to OPEX was TEC (€0.042-0.069 $kgCOD_{rem}^{-1}$), representing 41-46 % of the total operating cost, excluding energy recovery.

UF replacement was also a significant OPEX cost. In this regard, Scenario IV, with the lowest $J_{20\ gross}$ (15 LMH) required the largest membrane area and therefore showed the highest replacement cost (€0.028 $kgCOD_{rem}^{-1}$) while Scenario I with the highest $J_{20\ gross}$ (21 LMH) showed the lowest UF replacement cost (€0.011 $kgCOD_{rem}^{-1}$). This expense was between 9 % and 17 % of OPEX, excluding energy recovery. It is important to highlight that FR directly affected UF membrane replacement cost since membrane lifespan and the associated replacement frequency is determined by the maximum total contact with chlorine permissible during membrane cleaning.

However, in this study all the scenarios required one replacement over the considered lifespan of the facility so that there were no differences in this concept.

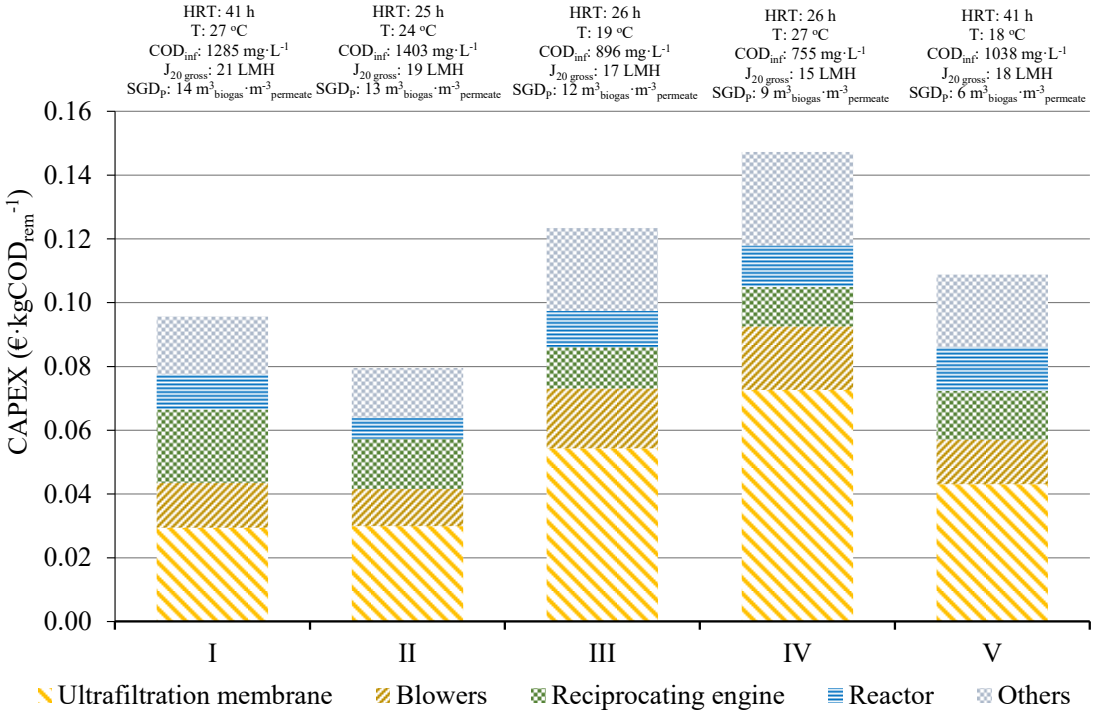


Figure 7.2. Allocation of CAPEX for each scenario. "Others" refers to: pumps; degassing membranes; diffusers; pipes; sludge centrifuge; membrane tanks and rotofilter.

The contribution of reciprocating engines to OPEX was high in Scenarios I (€0.017 $kgCOD_{rem}^{-1}$; 14 %), II (€0.012 $kgCOD_{rem}^{-1}$; 12 %) and V (€0.011 $kgCOD_{rem}^{-1}$; 9 %), in which a significant amount of biogas was produced. However, overall CHP contributed with savings of €0.099-0.181 per $kgCOD_{rem}$ when CAPEX and OPEX are balanced with cost reductions due to energy recovery. Moreover, these revenues do not consider the heat recovered and the environmental benefits of avoiding methane release, which is an additional value to take into account. Further improvement in CHP efficiencies would also improve AnMBR economic and environmental performance. Revenues could also be increased if energy recovery from biogas were to be conducted in the tariff periods with the highest energy costs and not proportionally distributed throughout the day, as considered in this study. However, this optimization strategy would entail an additional infrastructure (e.g., gasometer) and economic appraisals to assess its feasibility.

The fee for effluent discharging to surface water significantly contributed to the operating costs (€0.010-0.020 $kgCOD_{rem}^{-1}$; 9-12 % of OPEX), excluding energy recovery. Reducing or even

removing this charge in case of water (and nutrient) recovery could be a proper governance instrument for promoting this practice, taking advantage of the economic, environmental, and social benefits of water (and nutrient) recovery. By way of example, Jiménez-Benítez et al. (2020) evaluated two case studies of fertigation with AnMBR effluent in Spain and Italy, showing up to 100 % in mineral fertilizers savings, and obtaining reductions of 75 % (Spain) and even a carbon sink effect (Italy) when CO_2 emissions associated with savings on fertilizer production are considered. For this, appropriate reclaimed water management tools should be implemented, e.g., according to the Regulation (EU) 2020/741 on the minimum requirements for water reuse (European Comission, 2020).

Finally, it is important to highlight the reduced OPEX associated with sludge conditioning (€0.002-0.003 $kgCOD_{rem}^{-1}$; 2 %) and sludge disposal (€0.002-0.004 $kgCOD_{rem}^{-1}$; 2 %), which confirm the potential benefits of AnMBR as an alternative to aerobic treatments (e.g., CAS, EA) to address waste sludge management.

These results are in agreement with the trends found by Judd (2017) in which equipment energy needs, UF membrane replacement and discharge fees are the main costs in the OPEX.

7.3.2.3. Total annual cost

Figure 7.4 shows that TAC varied from €-0.003 $kgCOD_{rem}^{-1}$ (Scenario I) to €0.188 $kgCOD_{rem}^{-1}$ (Scenario IV). The results from Scenario I are explained by a low CAPEX (€0.096 $kgCOD_{rem}^{-1}$) mainly due to the lower UF membrane cost (€0.029 $kgCOD_{rem}^{-1}$) resulting from applying high $J_{20\ gross}$ (21 LMH); and the lowest OPEX (€-0.099 $kgCOD_{rem}^{-1}$) thanks to its high energy recovery (€-0.222 $kgCOD_{rem}^{-1}$). On the other hand, Scenario IV had the highest CAPEX (€0.147 $kgCOD_{rem}^{-1}$) influenced by its lower $J_{20\ gross}$ (15 LMH) and therefore high UF membrane cost (€0.073 $kgCOD_{rem}^{-1}$); and OPEX (€0.041 $kgCOD_{rem}^{-1}$) influenced by its high total maintenance and replacement costs (€0.068 $kgCOD_{rem}^{-1}$) and equipment energy costs (€0.067 $kgCOD_{rem}^{-1}$).

Table 7.3 shows TAC results reported in bibliography for different treatment technologies for urban wastewater. It is important to highlight the difficulties involved for achieving robust comparisons due to the variety of functional units, assumptions and hypotheses, or treatment schemes within the reviewed studies. Therefore, the comments presented here should be

considered as indicative. Ferrer et al. (2015) modelled an AnMBR with 50000 $m^3 \cdot d^{-1}$ of treatment capacity. These authors obtained a TAC between €0.14 $kgCOD_{rem}^{-1}$ (low-sulfate influent and considering methane captured from the effluent) and €0.20 $kgCOD_{rem}^{-1}$ (sulfate-rich influent without methane capture). Pretel et al. (2015) simulated an AnMBR treating sulfate-rich and low-sulfate wastewater at 15 and 30 ºC and obtained TAC in the range €0.18-0.43 $kgCOD_{rem}^{-1}$. With regards to MBR technology, Pretel et al., (2016b) obtained a TAC of €0.21 $kgCOD_{rem}^{-1}$, which are significantly lower than those obtained by Bertanza et al. (2017) (€1.74-3.33 $kgCOD_{rem}^{-1}$). In the case of CAS, Pretel et al., (2016b) calculated a TAC of €0.14 $kgCOD_{rem}^{-1}$, while Bertanza et al. (2017) reported much higher TAC for the same technology (€1.18-2.51 $kgCOD_{rem}^{-1}$). Lastly, Jafarinejad (2017) modelled three different treatments schemes: CAS, sequencing batch reactor and extended aeration activated sludge. The author obtained TAC of €0.93-0.95, €1.07-0.13, and €0.95-1.04 $kgCOD_{rem}^{-1}$, respectively. According to these values, the AnMBR system evaluated in the present study would be competitive even in the case of Scenario IV, which showed the highest TAC, i.e., €0.188 $kgCOD_{rem}^{-1}$.

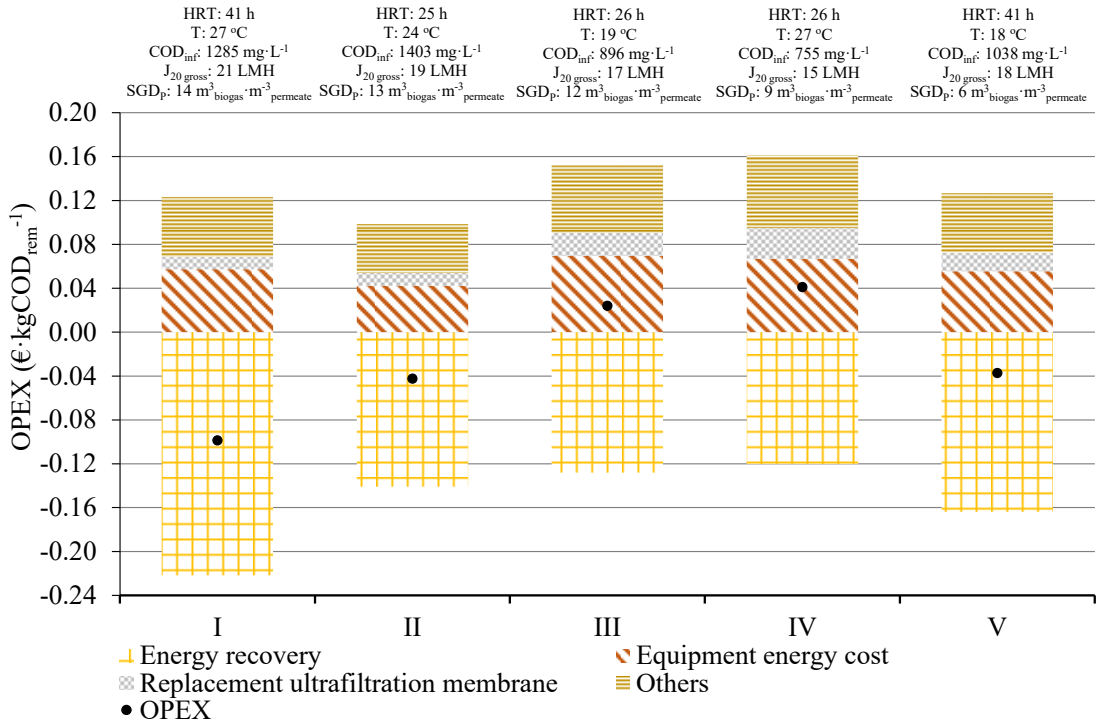


Figure 7.3. Allocation of OPEX for each scenario. "Others" refers to: power cost; membrane cleaning; sludge conditioning; sludge disposal; M&R pumps; M&R CHP; M&R blowers; M&R civil works; M&R pipes and diffusers; M&R rotofilter; discharge fee to surface water; and degassing membrane replacement.

Finally, CAPEX appears as the main contributor to TAC in Scenarios II to V, which is consistent with the results reported by Smith et al. (2014). Scenario I is especially relevant, in which the high energy production offset the other operating costs and CAPEX, bringing the TAC close 0 (€-0.003 $kgCOD_{rem}^{-1}$). Optimization strategies should thus be used to address both CAPEX and OPEX, since both items showed a potential for reducing TAC and contributing to the economic sustainability of AnMBR-based wastewater treatment.

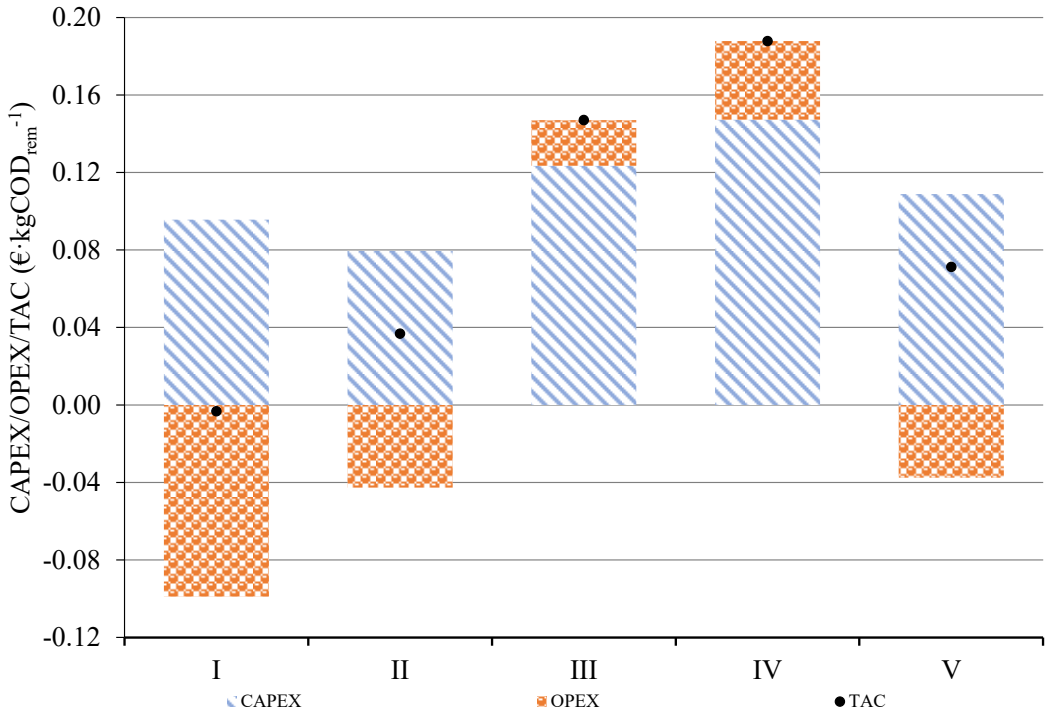


Figure 7.4. Allocation of total annual cost (TAC) for each scenario.

Table 7.3. Total annualized cost (TAC) comparison of different treatment technologies for urban wastewater.

Technology	Scale	Membrane configuration	TAC (€·$kgCOD_{rem}^{-1}$)	Ref
AnMBR	Pilot+simulation	HF	0.14-0.20	Ferrer et al. (2015)
ANMBR	Pilot+simulation	HF	0.18-0.43	Pretel et al. (2015)
MBR	Pilot+simulation	HF	0.21	Pretel et al. (2016c)
MBR	Full	HF	1.74-3.33	Bertanza et al. (2017)
CAS	Full	-	1.18-2.51	Bertanza et al. (2017)
CAS	Pilot+simulation	-	0.14	Pretel et al. (2016c)
CAS	Full+simulation	-	0.93-0.95	Jafarinejad (2017)
SBR	Full+simulation	-	1.07-1.13	Jafarinejad (2017)
EAAS	Full+simulation	-	0.95-1.04	Jafarinejad, (2017)

AnMBR: anaerobic membrane bioreactor; MBR: membrane bioreactor; CAS: conventional activated sludge; SBR: sequential batch reactor; EAAS: extender aeration activated sludge; HF: hollow fiber.

7.3.3. Sensitivity analysis

By way of example, the results from the SA of Scenario I are shown in Figure 7.5, providing the main influencing parameters ($b_i \geq 0.1$).

7.3.3.1. OPEX output

Figure 7.5a shows the SA results for OPEX. The positive main influencing parameters were HRT, blower cost, biogas for reactor mixing, SGD_P applied to membrane tanks and the AnMBR lifespan. Most of these were related to the biogas sparging needs to mix the reactor or stir the membranes. Reducing gas sparging intensities would thus entail significant savings in OPEX. Optimizing HRT would also reduce reactor volume and hence mixing requirements. Finally, extending the AnMBR lifespan by 10 %, from 20 to 22 years, would increase the replacement cost associated with membranes and equipment and thus also the OPEX. In Scenario I, it would be needed to replace both the UF membranes and the equipment (lifespan of 10 years) once throughout the lifespan set for the AnMBR plant (i.e., 20 years). By applying an uncertainty of 10%, AnMBR lifespan would increase from 20 to 22 years in the upper boundary. Thus, new membranes and equipment would be necessary for years 21 and 22. However, the total cost of these new devices would not be distributed in a decade, but only among the two additional years, increasing therefore the total replacement cost for this scenario. In this respect, the second replacement for membranes and equipment would be on service only a 20 % of their potential durability.

In relation to the parameters with negative influence, increasing the membranes' useful life and effective filtration time (i.e., operating period without downtime for physical cleaning of the membrane) would reduce the OPEX related to membrane replacement and filtration productivity. The largest effect would be associated with energy costs and CHP efficiency. Since Scenario I showed high energy production and performed as a net energy producer, favoring energy recovery through improved CHP power efficiency, and increasing the energy price for its sales would produce additional revenues and reduce OPEX. It is important to highlight that the effect of energy price would be the opposite in a net energy consumer scenario.

7.3.3.2. CAPEX output

The parameters with the greatest influence on CAPEX are shown in Figure 7.5b. The SA results indicated that the costs associated with the blowers, membranes, size of the reactors (through the

HRT) and capacity of the CHP (and its consequent cost) had a significant direct influence on CAPEX, as did the discount rates applied to the acquisition of membranes and equipment.

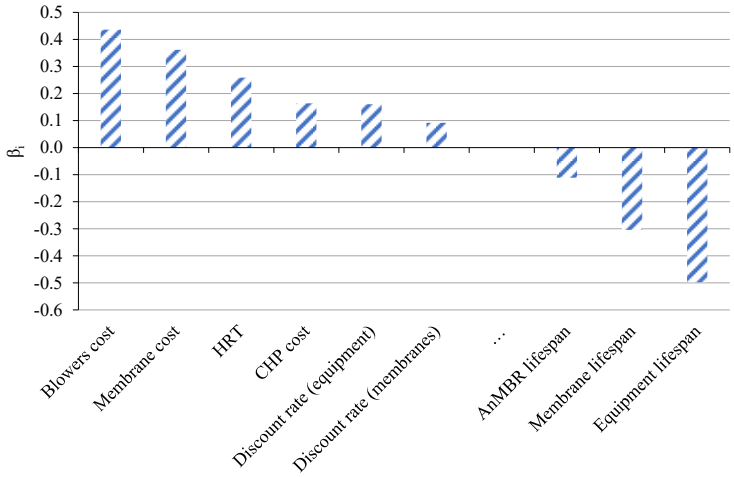
Therefore, reducing the gas sparging requirements, improving the filtration productivity (and therefore the necessary membrane surface), and optimizing the HRT to reduce the volume of the reactors, would be useful strategies to reduce CAPEX. Moreover, an accurate selection of CHP capacity would also be important to avoid over dimension since its influence on CAPEX was also significant.

Finally, CAPEX could also be reduced by increasing the useful lifespan of the plant, membranes and equipment. This necessarily involves adequate maintenance and optimization of the filtration operation, thus reducing the need for chemical cleaning of the membranes, which is mainly responsible for their deterioration.

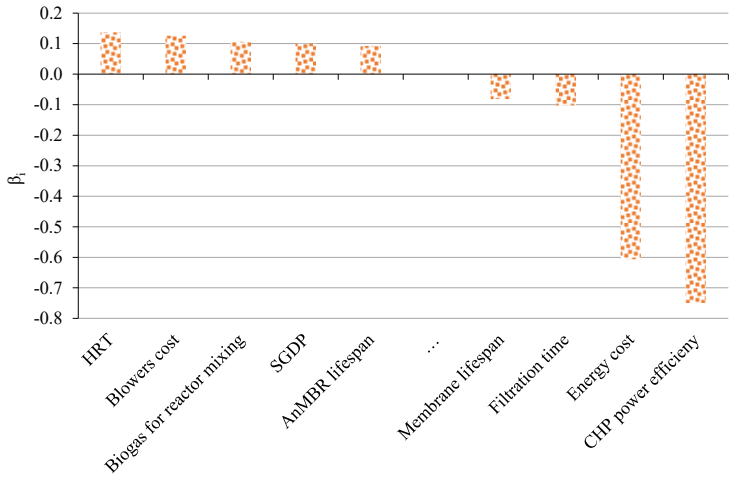
7.3.3.3. *TAC output*

Figure 7.5c shows the input parameters with a strong influence on TAC output, all of which were identified as influential parameters on CAPEX and OPEX outputs. The elements associated with gas sparging (blowers, SGD_P and reactor mixing) jointly with the equipment replacement needs derived from extending the AnMBR lifespan implied an increase in TAC. On the other hand, increasing the membrane and equipment lifespan, improving filtration productivity, and obtaining better conditions for energy recovery (e.g., through higher profits from the sale of electric power, or an improvement in the efficiency of CHP systems) would improve the overall economic assessment represented by the TAC.

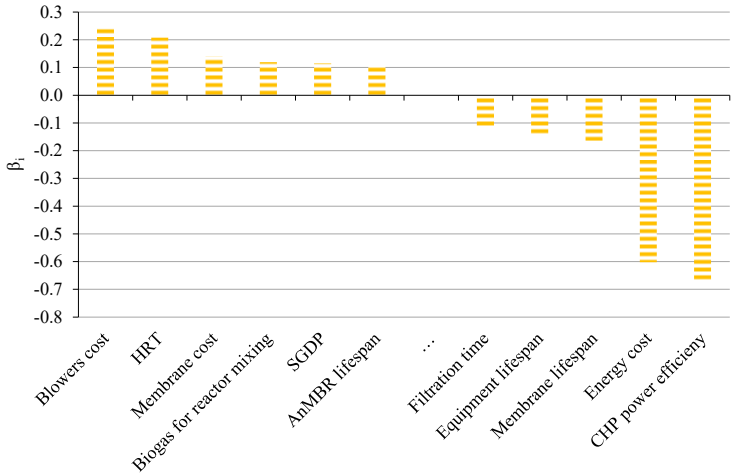
Finally, by way of example, Figure 7.6 shows the Monte Carlo simulation results for TAC with regard to CHP power efficiency (Figure 7.6a) and energy tariff variation (Figure 7.6b) in Scenario I. Figure 7.6a shows that CHP power efficiencies over 38.5 % would lead to a TAC below €0 $kgCOD_{rem}^{-1}$ in all simulations, which represents revenues for treating wastewater. On the contrary, cogeneration energy efficiencies lower than 31.5% obtained a positive TAC in all simulations, which indicates an economic outlay for treating the water. In those situations, with efficiencies between 31.5 and 38.5%, the TAC may be positive or negative depending on the rest of the variables, but the probability of a negative TAC increases as the efficiency of the CHP increases.



a)



b)



c)

Figure 7.5. Sensitivity analysis results for Scenario I. Main influential factors on model outputs: a) CAPEX, b) OPEX, and c) TAC.

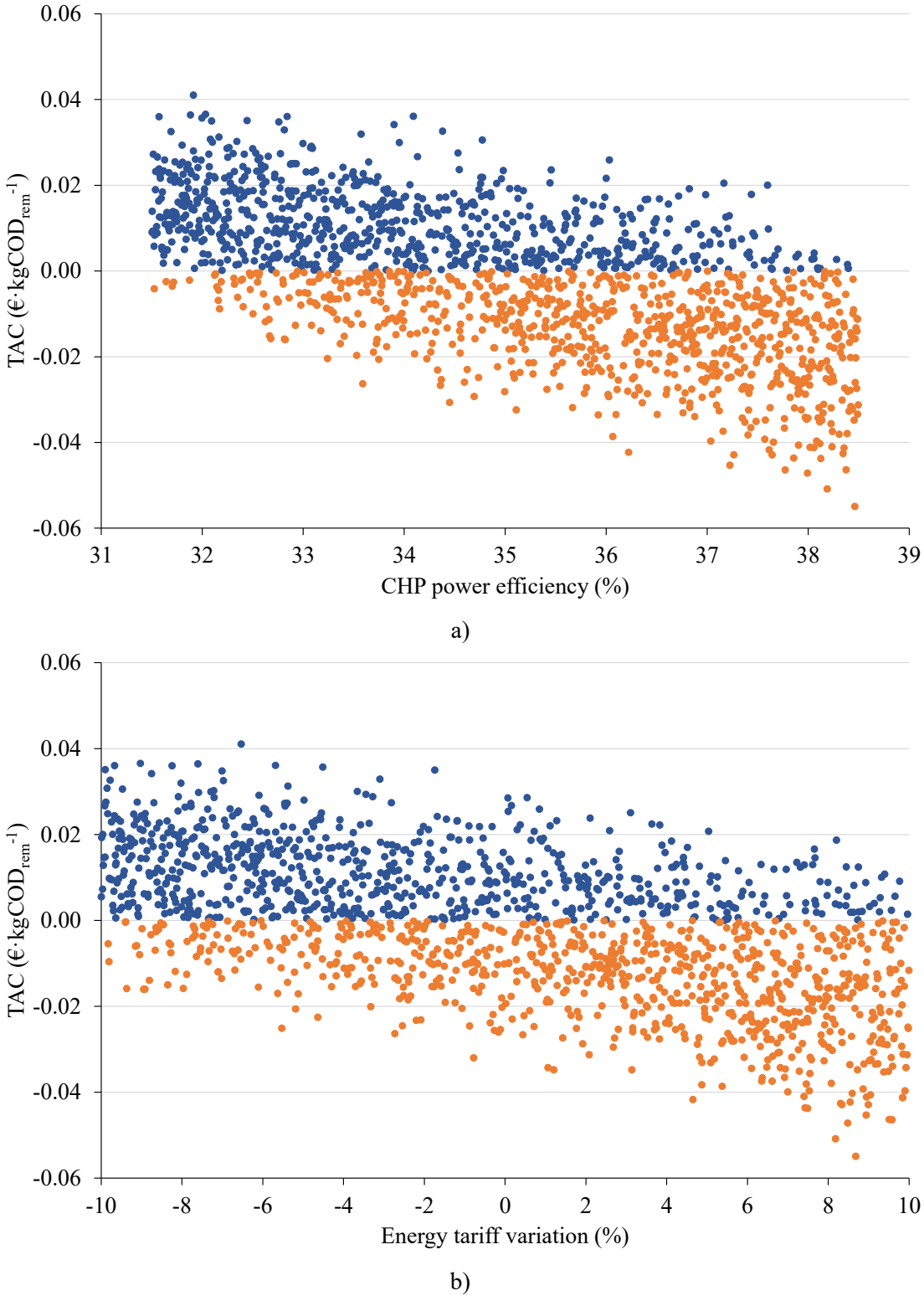


Figure 7.6. Monte Carlo simulation results for TAC regarding a) CHP power efficiency (%) and b) energy tariff variation (%). Red dots indicate simulation outputs with TAC < 0 and blue dots represent simulation outputs with TAC > 0.

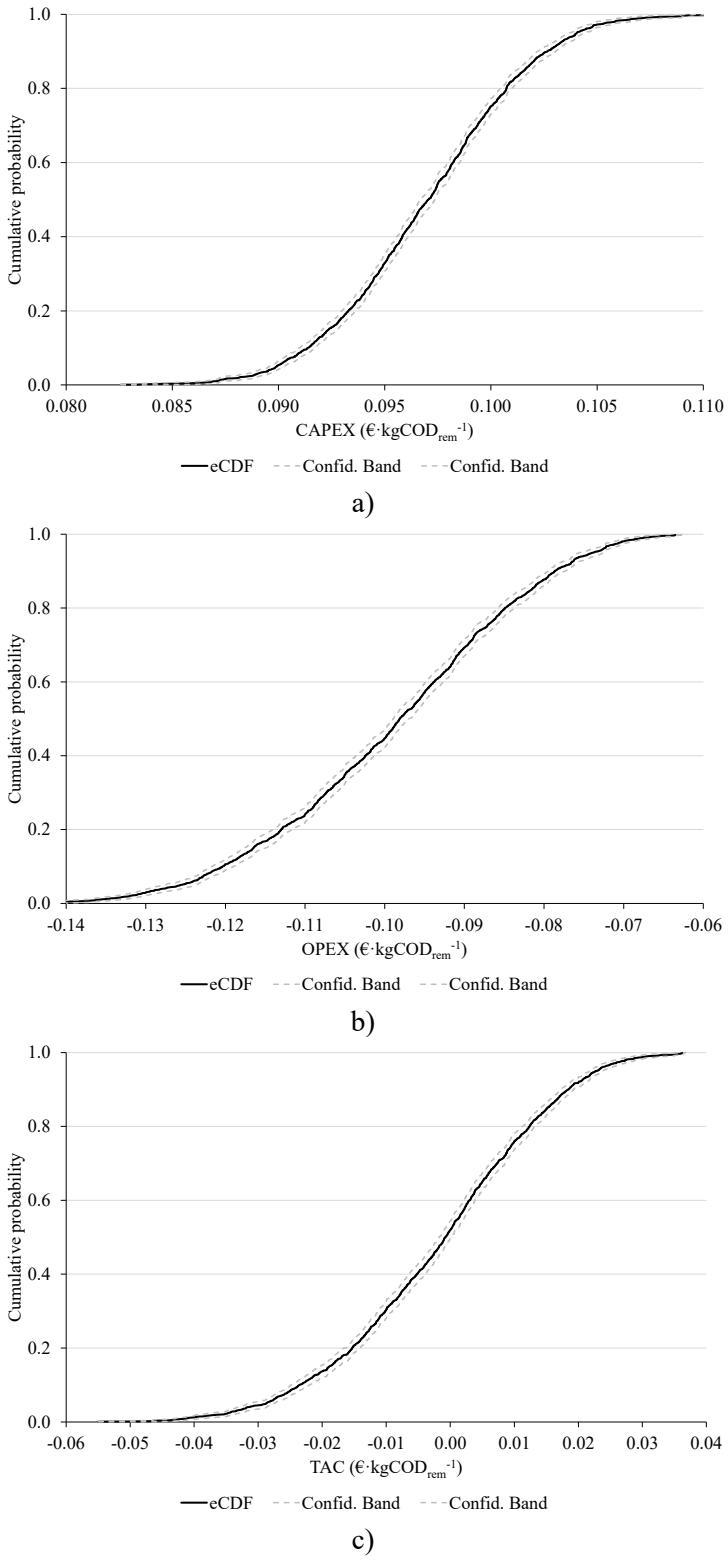


Figure 7.7. Uncertainty analysis results for Scenario I. Empirical cumulative distribution function (eCDF) for: a) CAPEX, b) OPEX, and c) TAC.

Moreover, a 10% increase in the price of the electricity sold would lead to a negative TAC (see Figure 7.6b), while a similar reduction of in the energy price would imply a TAC greater than

€0 $kgCOD_{rem}^{-1}$. As already mentioned in section 7.3.3.1, the influence of the electricity tariff on the results is marked by the fact that the scenario considered performed as a net producer of energy. Otherwise, an increase in the energy tariff would increase costs. These results highlight the importance of considering energy in AnMBR feasibility studies.

Regarding the percentiles, the 5^{th}-95^{th} range for OPEX, CAPEX, and TAC resulted in €0.014, €0.051, and €0.052 per $kgCOD_{rem}$, respectively (see Table 7.4). These results show that the uncertainty in CAPEX is higher than in OPEX, being the former the main contributor to TAC uncertainty. Therefore, in order to reduce sources of uncertainty, efforts should be made to obtain reliable and robust investment data, while a careful design of AnMBR systems should be conducted to ensure an optimal performance. According to section 7.3.3, improving data regarding UF membranes, CHP and blower cost would reduce CAPEX uncertainty. In the case of OPEX, accurate data related to reactor mixing and membrane sparging requirements and revenues through energy recovery (CHP power efficiency and energy tariff) would also improve overall UA.

Table 7.4. Uncertainty results for Scenario I: 5th-95th percentile and 5th-95th range.

	€·$kgCOD_{rem}^{-1}$		
	CAPEX	**OPEX**	**TAC**
5^{th} percentile	0.090	-0.125	-0.029
95^{th} percentile	0.104	-0.074	0.023
5^{th}-95^{th} range	0.014	0.051	0.052

7.4. Conclusions

Favorable energy balances were achieved in all the scenarios evaluated. NED varied between -0.210 kWh·$kgCOD_{rem}^{-1}$ and -0.645 kWh·$kgCOD_{rem}^{-1}$. Reactor mixing (0.056-0.124 kWh·$kgCOD_{rem}^{-1}$) and membrane scouring (0.048-0.120 kWh·$kgCOD_{rem}^{-1}$) showed the highest energy consumption.

The major contributors to CAPEX were UF membranes (€0.029-0.073 $kgCOD_{rem}^{-1}$; 31-49 %), reciprocating engines acquisition (€0.012-0.023 $kgCOD_{rem}^{-1}$; 8-24 %), and reactor building (€0.007-0.014 $kgCOD_{rem}^{-1}$; 8-13 %). OPEX was mainly influenced by equipment energy needs

(€0.042-0.069 $kgCOD_{rem}^{-1}$; 41-46 %), UF membrane replacement (€0.011-0.028 $kgCOD_{rem}^{-1}$; 9-17 %) and discharge fee (€0.010-0.020 $kgCOD_{rem}^{-1}$; 9-12 %). CHP maintenance and replacement was also a significant operating cost (€0.009-0.017 $kgCOD_{rem}^{-1}$; 6-14 %), although it finally provided revenues through energy sale (€0.120-0.222 $kgCOD_{rem}^{-1}$). The TAC varied between -€0.003 and €0.188 $kgCOD_{rem}^{-1}$, with CAPEX as the main cost.

The results obtained from LCC assessment combined with SA and UA showed that AnMBR represents a potential alternative to conventional aerobic treatments. Its definitive impulse will go through, among other aspects, reinforcing its economic sustainability, for which it is necessary to optimize operational aspects (e.g., HRT, transmembrane flux, reactor mixing and membrane sparging) and technological developments that allow increasing membrane lifespan and improve CHP power efficiency. Finally, other aspects that would also result in improving its economic performance have to do with nutrient recovery present in its effluent and favorable pricing for the sale of electricity produced from the methane generated.

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7.5. Supplementary material

7.5.1. AnMBR prototype flow diagram and overview

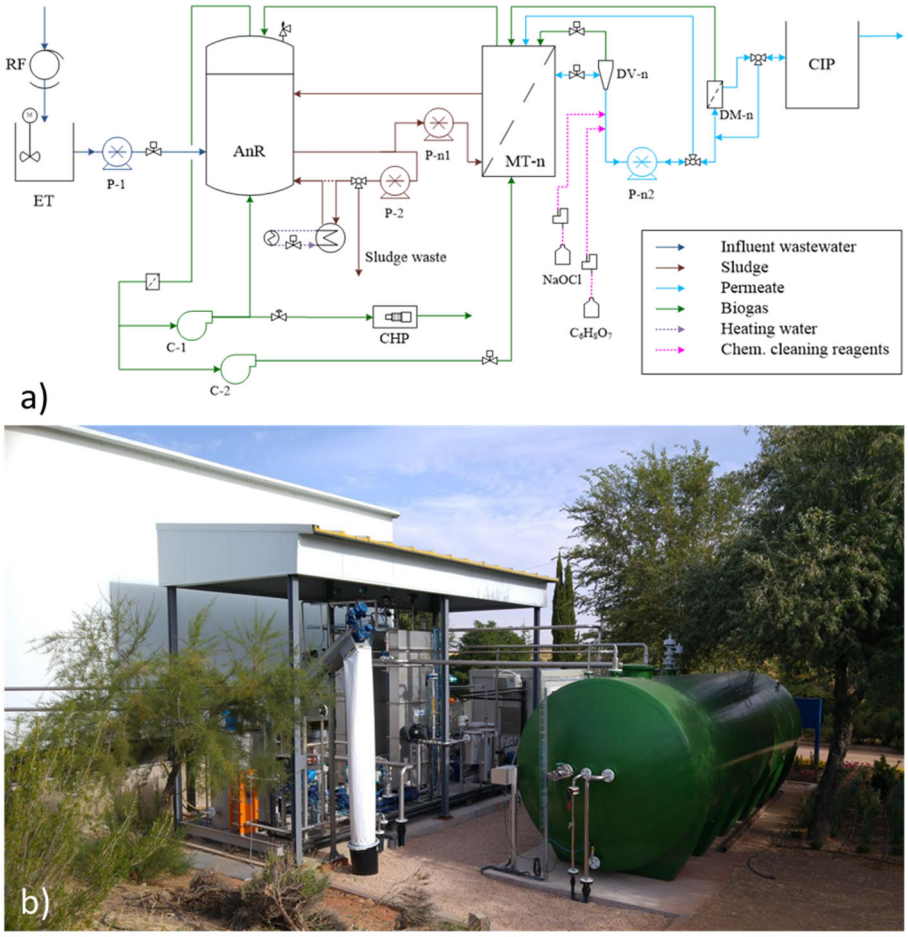

Figure 7.1S. (a) Flow diagram of the AnMBR Plant, and (b) AnMBR plant overview. Note: "n" represents the membrane treatment lane.

7.5.2. Operating conditions and characterization of the selected scenarios

Table 7.1S. Influent and effluent characterization, main operating conditions, and main biological and filtration process performance results from the AnMBR prototype.

INFLUENT	Units	I	II	III	IV	V
Flow rate	$L \cdot d^{-1}$	21228±591	33016±2773	33243±3118	34207±1478	20984±7482
TSS	$mg \cdot L^{-1}$	462±188	418±293	332±145	354±116	430±120
VSS	%	64.6±5.9	82.0±11.5	63.1±14.6	75.5±9.6	80.1±13.0
COD	$mg \cdot L^{-1}$	1285±429	1403±532	896±201	755±224	1038±238
BOD_5	$mg \cdot L^{-1}$	772±277	911±288	569±122	451±154	581±180
OLR	$kg\ COD \cdot m^{-3} \cdot d^{-1}$	0.75±0.25	1.28±0.38	0.82±0.19	0.71±0.23	0.60±0.20
VFA	$mg\ COD \cdot L^{-1}$	149±146	240±132	120±86	33±60	60±35
Alk	$mg\ CaCO_3 \cdot L^{-1}$	625±79	660±126	663±171	734±175	784±129
N_T	$mg\ N \cdot L^{-1}$	52.0±13.3	54.7±16.3	46.8±6.9	46.0±11.2	35.6±8.0
P_T	$mg\ P \cdot L^{-1}$	10.1±2.8	8.6±2.1	8.5±1.2	6.8±1.3	7.6±1.1
PO_4^{3-}-P	$mg\ P \cdot L^{-1}$	2.5±0.0	5.8±2.1	6.0±0.9	5.3±1.3	3.9±1.7
SO_4^{2-}-S	$mg\ S \cdot L^{-1}$	147.7±13.3	172.2±28.5	125.4±47.6	157.3±46.9	149.7±26.9
HS^--S	$mg\ S \cdot L^{-1}$	37.9±26.7	59.6±19.5	47.0±38.7	30.7±49.2	38.3±14.5
COD:SO_4^{2-}-S	$g\ COD \cdot g^{-1}\ SO_4^{2-}$-S	8.70±3.69	8.15±4.44	7.14±4.31	4.80±2.86	6.94±2.83

EFFLUENT	Units	I	II	III	IV	V
COD	$mg \cdot L^{-1}$	91±31	116±24	121±15	80±17	79±18
VFA	$mg\ DQO \cdot L^{-1}$	9±14	21±45	41±28	0±0	5±11
Alk	$mg\ CaCO_3 \cdot L^{-1}$	1110±75	1336±170	996±74	1057±140	1028±198
N_T	$mg\ N \cdot L^{-1}$	47.9±6.2	54.0±14.6	47.3±4.3	52.5±10.2	37.0±11.1
P_T	$mg\ P \cdot L^{-1}$	8.8±1.9	9.9±1.9	6.9±2.8	6.3±0.5	7.8±1.2
PO_4^{3-}-P	$mg\ P \cdot L^{-1}$	7.8±2.5	8.9±1.9	6.9±1.0	5.8±0.6	6.3±2.9
HS^--S	$mg\ S \cdot L^{-1}$	174.3±28.7	207.5±73.2	233.1±66.3	183.2±27.0	166.2±27.9
CH_4	$mg \cdot L^{-1}$	16.0±0.5	17.1±0.8	17.9±0.3	16.3±0.5	18.7±0.6

Table 7.1S. Influent and effluent characterization, main operating conditions, and main biological and filtration process performance results from the AnMBR prototype (continue).

REACTOR	Units	I	II	III	IV	V
SRT	d	70±1	68±2	70±2	71±2	70±0
HRT	h	41±1	25±1	26±2	26±2	41±13
T	ºC	27±1	24±2	19±1	27±1	18±2
MLTSS	$g \cdot L^{-1}$	8.4±0.5	12.6±0.5	11.3±1.0	10.4±0.8	8.3±1.2
MLVSS	%	70.3±5.5	68.1±1.8	72.8±3.8	65.7±2.7	68.9±5.9
N_T	$mg\ N \cdot L^{-1}$	505.8±139.4	579.0±68.7	509.0±133.6	339.9±63.5	269.5±94.4
P_T	$mg\ P \cdot L^{-1}$	96.8±12.3	106.6±10.7	89.4±14.9	93.2±6.2	83.7±6.9
pH		7.1±0.1	7.0±0.1	7.0±0.1	7.0±0.1	7.1±0.3
Waste sludge	$L \cdot d^{-1}$	522±5	511±107	521±5	510±18	520±4
COD	$g \cdot L^{-1}$	11.4±0.6	15.8±1.1	14.7±2.0	13.2±0.5	13.0±0.4
Biogas prod.	$L\ STP \cdot d^{-1}$	5396±4435	5558±3204	1901±597	1359±999	2241±924
$\%CH_4$	%	77±2	77±2	72±2	77±2	76±1
Y^{CH4}	$L\ STP \cdot kgCOD_{rem}^{-1}$	169.0±95.1	109.6±70.0	75.2±16.9	70.2±36.0	102.3±63.8

MEMBRANES	Units	I	II	III	IV	V
SGD_p	$Nm^3_{biogas} \cdot m^{-3}_{permeate}$	14	13	12	9	6
$J_{20\ gross}$	LMH	21	19	17	15	18
$J_{20\ net}$	LMH	18	16	15	13	15
TMP	mbar	318	362	462	87	360
FR	$mbar \cdot day^{-1}$	4.2	4.7	0.3	0.5	4.9
Cleaning frequency	$year^{-1}$	2.8	3.1	0.2	0.3	3.2

7.5.3. Energy calculation

The equipment considered for calculating the power energy requirements term ($W_{element}$) consisted of the following items: rotofilter, pumps, blowers and sludge dewatering system.

The energy consumption of blowers (adiabatic compression), general pumps (feeding and recycling), and permeate pumps was calculated by Eq. 7.1S, 7.2S, and 7.4S, respectively.

$$P_B = \frac{(M \cdot R \cdot T_{gas})}{(\alpha - 1) \cdot \eta_{blower}} \left[\left(\frac{P_2}{P_1} \right)^{\frac{\alpha - 1}{\alpha}} - 1 \right] \qquad \text{Equation 7.1S}$$

where P_B is the power requirement in J/s (adiabatic compression), M (mol/s) is the molar flow rate of biogas, R (J/mol/K) is the gas constant for biogas, P_1 (atm) is the absolute inlet pressure, P_2 (atm) is the absolute outlet pressure, T_{gas} (K) is the biogas temperature, α is the adiabatic index, and η_{blower} is the blower efficiency.

$$P_g = q_{imp} \cdot \rho_{liquor} \cdot g \cdot \frac{\left\{\left[\left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g}\right)_{asp} + \left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g}\right)_{imp}\right] + [Z_1 - Z_2]\right\}}{\eta_{pump}} \quad \text{Equation 7.2S}$$

where P_g is the power requirement by the general pump (J/s), considering both pump aspiration and pump impulsion section, calculated from the impulsion volumetric flow rate ($q_{imp.}$ in m^3/s), liquor density (ρ_{liquor} in kg/m^3), acceleration of gravity (g in m/s), length of straight pipe (L in m), equivalent length of straight pipe for minor pressure losses (L_{eq} in m), velocity (V in m/s), Darcy–Weisbach friction factor (f, dimensionless), pipe diameter (D in m), difference in height (Z_1-Z_2, in m), and pump efficiency (η_{pump}). It has been considered that pipe sections and inlet and outlet pressure remain constant. The Swamee-Jain equation was used to calculate the Darcy–Weisbach friction factor (Eq 7.3S).

$$f = \frac{0.25}{\left[log_{10}\left(\frac{k/D}{3.7} + \frac{5.74}{Re^{0.9}}\right)\right]^2} \quad \text{Equation 7.3S}$$

where f is the Darcy–Weisbach friction factor, Re is the Reynolds number, k (m) is the pipe roughness, and D (m) is the pipe diameter.

Finally, Eq 7.2S was modified for calculating permeate pumping power requirements. In this case, power requirements were determined by Eq. 7.4S:

$$P_{stage} = \frac{q_{stage} \cdot TMP_{stage}}{\eta_{pump}} \quad \text{Equation 7.4S}$$

where P_{stage} is the power requirement during filtration, degasification or back-flushing calculated from transmembrane pressure (TMP_{stage} in Pa), pump volumetric flow rate (q_{stage} in m^3/s), and pump efficiency (η_{pump}).

To calculate the net power required by the permeate pump ($P_{permeate}$), the sum of the power consumed in the following four membrane operating stages was considered: filtration ($P_{filtration}$), back-flushing ($P_{back\text{-}flushing}$), degasification ($P_{degasification}$) and ventilation ($P_{ventilation}$). It should be noted that Eq. 7.2S was used to calculate the power in ventilation stage since the fluid does not pass through the membrane.

7.5.4. Sensitivity and uncertainty analyses

Table 7.2S. Mean value, lower bound (LB) and upper bound (UB) of input factors for sensitivity and uncertainty analyses.

Inputs				
Parameter	**Unit**	**Mean**	**LB[a]**	**UB[b]**
SRT	d	70	63	77
HRT	h	41.0	36.9	45.1
Liquid height in the reactor	m	5.0	4.5	5.5
Steel price	€·m^{-2}	51.11	46.00	56.22
Concrete wall price	€·m^{-2}	350	315	385
Concrete slab price	€·m^{-2}	130	117	143
Liquid height in the membrane tank	m	2.8	2.5	3.1
Discharge fee	€·m^{-3}	0.01346	0.01211	0.01481
Depreciation period for facility	y	20	18	22
Discount rate for facility		0.030	0.027	0.033
Depreciation period for civil work	y	20	18	22
Discount rate for civil work		0.030	0.027	0.033
Depreciation period for equipment	y	10	9	11
Discount rate for equipment		0.050	0.045	0.055
NaOCl price	€·L^{-1}	0.72	0.65	0.79
Citric acid price	€·t^{-1}	2960	2664	3256
Filtration time (i.e., non-downtime period)		0.85	0.77	0.94
Biogas sparging intensity for reactor mixing	$Nm^3 \cdot m_{reactor}^{-3} \cdot h^{-1}$	0.25	0.23	0.28
Dissolved methane recovery efficiency	%	55.0	49.5	60.5
CHP power efficiency	%	35.0	31.5	38.5
Diffuser price	€·$unit^{-1}$	16.94	15.25	18.63
SGD_P	$Nm^3 \cdot m^3$	14.02	12.62	15.42

Inputs (continue)				
Parameter	**Unit**	**Mean**	**LB[a]**	**UB[b]**
Transmembrane flux ($J_{20\,p}$)	LMH	21.0	18.9	23.1
Sludge fate (agriculture)	€·t^{-1}	4.81	4.33	5.29
Sludge fate (landfill)	€·t^{-1}	30.05	27.05	33.06
Sludge fate (incineration)	€·t^{-1}	250	225	275
Polyelectrolyte price	€·kg^{-1}	2.35	2.12	2.59
Energy term 1	€·kWh^{-1}	0.34	0.31	0.37
Energy term 2	€·kWh^{-1}	0.31	0.28	0.34
Energy term 3	€·kWh^{-1}	0.291	0.262	0.320
Energy term 4	€·kWh^{-1}	0.267	0.240	0.294
Energy term 5	€·kWh^{-1}	0.243	0.219	0.267
Energy term 6	€·kWh^{-1}	0.231	0.208	0.254
Power term 1	€·kW^{-1}·y^{-1}	24.73	22.26	27.21
Power term P2	€·kW^{-1}·y^{-1}	21.53	19.38	23.68
Power term P3	€·kW^{-1}·y^{-1}	12.32	11.09	13.55
Power term P4	€·kW^{-1}·y^{-1}	9.90	8.91	10.89
Power term P5	€·kW^{-1}·y^{-1}	2.83	2.55	3.12
Power term P6	€·kW^{-1}·y^{-1}	1.57	1.41	1.73

7.5.5. Power and energy tariff

Spanish power and energy tariff system considers fixed costs, variable costs, and taxes. Tariffs are selected based on the power needed in a facility. For the purpose of this work, tariff 6.1. TD was selected, which corresponds to companies and industries with access tensions between 1 and 30 kV and six periods (www.somenergia.coop). Table 7.3S and Table 7.4S show the details of tariff 6.1. TD applied in this study:

Table 7.3S. Power term for tariff 6.1. TD.

		€·kW^{-1}·year^{-1}					
Description	**Tariff**	**P1**	**P2**	**P3**	**P4**	**P5**	**P6**
1-30 kV	6.1. TD	24.73	21.53	12.32	9.90	2.83	1.57

Table 7.4S. Energy term for tariff 6.1. TD.

		€·kWh^{-1}					
Description	**Tariff**	**P1**	**P2**	**P3**	**P4**	**P5**	**P6**
1-30 kV	6.1. TD	0.340	0.310	0.291	0.267	0.243	0.231

Finally, energy consumption weights for each period according to data provided by Public Entity for Sewage Treatment of Comunitat Valenciana (Spain) (EPSAR) are shown in Table 7.5S.

Table 7.5S. Energy consumption weights (%) for WWTP in Comunitat Valenciana (Spain).

	%					
Tariff	**P1**	**P2**	**P3**	**P4**	**P5**	**P6**
6.1. TD	8	11	5	6	10	60

7.5.6. CAPEX

7.5.6.1. Main considerations for CAPEX

The main considerations when calculating CAPEX are summarized as follows:

- *Depreciation:* A depreciation period of 20 years for civil works and 10 years for equipment was used to calculate TAC. The annual discount rate was set to 3% for civil works and 5 % for equipment.

- *Membrane tank:* The membrane tank volume was estimated according to the dimensions of a commercial membrane unit (PURON®, Koch Membrane Systems, PUR-PSH1500, 0.03-µm pore size, 1500 m^2 total filtering area).

- *Equivalent reactor tanks:* The reactor tanks volume was calculated based on the treatment flow rate and the considered HRT for each scenario. The headspace volume for biogas storage was set to 10 % of the working volume.

- *Pipelines:* The maximum velocity of liquid in pipes was set to 1.5 $m \cdot s^{-1}$ to calculate the pipe diameter, thus preventing excessive losses of mechanical energy.

- *Equipment:* Acquisition of rotofilters, blowers, pumps, diffusers, centrifuges, UF membranes, degassing membranes, and reciprocating engines for methane valorization have been included. To consider scaling aspects, when possible, equipment expenditures have been estimated by correlations between capacities (mainly based on flow rates) and unit costs

provided by manufacturers. Acquisition of additional 20 % of UF membrane for maintenance considerations was considered.

- *Land cost:* land cost is not included in this LCC.

7.5.6.2. Reactor tanks

Table 7.6S shows key technical and economic data from the design of the quadrangular shaped anaerobic reactor tanks. Calculations included the following assumptions:

- ✓ Headspace volume: 10% of the working volume
- ✓ Sludge height: 5.0 m
- ✓ Reactor height: 5.5 m
- ✓ Reactor width: 20.0 m
- ✓ Floor thickness: 0.5 m
- ✓ Wall thickness: 0.6 m
- ✓ Cover thickness: 0.3 m

Table 7.6S. Key technical and economic data from the design of the anaerobic reactor tanks.

Scenario	HRT (hours)	Total Sludge Volume (m^3)	Equivalent Reactor Volume (m^3)	Number of reactors	Reactor Volume ($m^3 \cdot react^{-1}$)	Reactor Length (m)	Concrete WALLS (m^3)	Concrete FLOOR (m^3)	Concrete COVER (m^3)	Concrete TOTAL (m^3)	Concrete FLOOR and COVER (€)	Concrete WALLS (€)	Concrete TOTAL COST (€)	DEPRECIATION ($€ \cdot year^{-1}$)
I	41	27333	30067	4	6834	68.3	1655	2733	1640	6029	568506	579406	1147911	**77158**
II	25	16667	18333	3	5556	55.6	1129	1667	1000	3796	346694	395287	741982	**49873**
III	26	17333	19067	3	5778	57.8	1159	1733	1040	3932	360547	405544	766091	**51493**
IV	26	17333	19067	3	5778	57.8	1159	1733	1040	3932	360547	405544	766091	**51493**
V	41	27333	30067	4	6834	68.3	1655	2733	1640	6029	568506	579406	1147911	**77158**

7.5.6.3. Membrane tanks

Table 7.7S shows the design and economic assessment for AnMBR membrane tanks (MT). Calculations included the following assumptions:

- ✓ MT height: 2.8 m
- ✓ Volume occupied by membranes: 65 %
- ✓ Floor thickness: 0.5 m
- ✓ Wall thickness: 0.6 m
- ✓ Cover thickness: 0.005 m

Table 7.7S. Design and economic assessment of AnMBR membrane tanks.

Scenario	Equivalent MT Volume (m^3)	Concrete WALLS (m^3)	Concrete FLOOR (m^3)	Steel COVER (m^2)	Concrete TOTAL (m^3)	Concrete FLOOR (€)	Concrete WALLS (€)	Steel COVER (€)	MT TOTAL COST (€)	DEPRECIATION (€·$year^{-1}$)
I	287	18.4	53.4	106.9	71.9	6947	6447	5165	18856	**1267**
II	316	19.3	58.8	117.6	78.1	7643	6762	5626	20414	**1372**
III	349	20.3	64.8	129.6	85.1	8424	7102	6086	22150	**1489**
IV	412	22.0	76.2	152.4	98.2	9907	7700	7227	25398	**1707**
V	343	20.1	63.7	127.4	83.8	8278	7039	6086	21826	**1467**

7.5.6.4. Pipes and equipment acquisition

Table 7.8S shows pipes and equipment acquisition costs.

Table 7.8S. Pipes and equipment acquisition costs (€·kgCOD$_{rem}^{-1}$).

	€·kgCOD$_{rem}^{-1}$								
Scenario	**Rotofilter**	**UF Membrane**	**DM**	**Pumps**	**Blowers**	**Diffusers**	**Sludge centrifuge**	**Reciprocating engine**	**Pipes**
I	0.002	0.029	0.004	0.008	0.014	≈0.000	0.002	0.023	0.001
II	0.002	0.030	0.004	0.006	0.012	≈0.000	0.002	0.016	0.001
III	0.003	0.054	0.007	0.011	0.019	≈0.000	0.003	0.013	0.001
IV	0.004	0.073	0.007	0.012	0.020	≈0.000	0.004	0.012	0.001
V	0.003	0.043	0.005	0.010	0.014	≈0.000	0.003	0.015	0.001

7.5.6.5. Allocation of CAPEX weights

Table 7.9S shows the weight of each element as percentage of CAPEX considered for the LCC.

Table 7.9S. Allocation of capital expenditure (%).

	%										
Scenario	**Rotofilter**	**UF Membrane**	**DM**	**Pumps**	**Blowers**	**Diffusers**	**Sludge centrifuge**	**Reciprocating engine**	**Pipes**	**Reactor**	**MT**
I	2.3	30.7	4.3	8.4	14.8	0.1	2.4	24.0	1.1	11.5	0.2
II	2.6	37.6	5.1	8.1	14.6	0.1	2.6	19.9	0.9	8.3	0.2
III	2.8	44.0	5.5	8.6	15.3	0.1	2.8	10.5	1.0	9.2	0.3
IV	2.7	49.3	4.9	8.3	13.6	0.1	2.7	8.4	0.9	8.9	0.3
V	2.5	39.6	4.9	9.2	12.8	0.1	2.6	14.1	1.2	12.6	0.2

7.5.7. OPEX

7.5.7.1. Main considerations for OPEX

The main considerations when calculating OPEX are summarized as follows:

- *Power requirements and total energy consumption (TEC):* the equipment considered for energy input calculations consisted of rotofilter, feeding pumps, permeate pumps, waste sludge pumps, sludge recycling pumps, sludge dewatering system, blowers for reactor mixing, blowers for membrane scouring by gas sparging, and methane recovery blowers. Both energy and power tariffs (Spanish) are included and allocations throughout pricing sections were introduced according to data provided by EPSAR. Energy tariff 6.1 TD have been considered in this study.

- *Energy recovery from methane:* Energy production from the valorization of both biogas and dissolved methane captured from the effluent was calculated as commented in Section 7.2.3 of the manuscript. Reduction in energy consumption and concomitant energy cost follow the same power and energy tariffs provided in supplementary material.

- *Chemical reagents for membrane cleaning:* Sodium hypochlorite and citric acid were considered for membranes chemical cleaning. 2000 ppm was adopted as the dose of both sodium hypochlorite and citric acid while the contact time was set to 5 hours. Chemical cleanings were conducted when TMP of 500 mbar was reached during filtration stage. Cleaning frequency was between 0.2 and 3.2 times·year-1 among the different scenarios (see Table 7.1 in the manuscript).

- *Membrane physical cleaning:* The downtime for membrane physical cleaning through back-flushing and other membrane operating stages was calculated from the membrane operating strategy of the AnMBR prototype-plant.

- *Membrane replacement:* The lifespan of both UF membranes and membrane recovery membranes was used to calculate membrane replacement costs. The lifespan of UF membrane was calculated from the maximum total contact with chlorine permissible before membrane replacement, i.e., 500000 ppm-hours cumulative according to the supplier. The lifespan of methane recovery membranes was set to 10 years.

- *Sludge treatment:* Centrifuges were selected for calculating sludge dewatering costs. To ensure adequate sludge conditioning, the dose of polyelectrolyte considered in this study was 6 kg·tTSS-1 (Sainz-Lastre, 2006).

- *Equipment replacement and maintenance:* The lifetime of blowers and pumps were stablished based on the total working hours recommended by manufacturers (50000 and

75000 h, respectively). The lifetime for the rest of equipment was set to 10 years. Operation and maintenance costs of €0.008-€0.020 per kWh produced was considered (Energy efficiency and renewable energy, 2016).

- *Civil works and construction maintenance:* Maintenance of civil works and buildings were considered as a percentage of the associated CAPEX (0.3 %), according to technical specifications for general constructions for WWTPs (Inyges Consultores S.L., 2015).

- *Sludge handling and disposal:* Three different scenarios were considered for calculating the cost of sludge handling and disposal, i.e., agriculture, incineration, and landfill. Sludge destiny was set to 88% (agriculture), 8% (landfill) and 4% (incineration), according to data published by Spanish Government (Sewage sludge fate, 2020).

- *Discharge fee:* According to Spanish regulation (Ley 22/2013, 2014), a fee of €0.01346 per m^3 of treated water discharged into inland water bodies was considered.

- *Labor costs:* labor costs are not included in this LCC.

7.5.7.2. Operating expenditure

Table 7.10S shows operating expenditures for the five scenarios evaluated.

Table 7.10S. Operating expenditures (€·kgCOD$_{rem}^{-1}$).

	€·kgCOD$_{rem}^{-1}$								
Scenario	**Power cost**	**Net energy cost**	**UF Membrane cleaning**	**Sludge conditioning**	**Sludge disposal**	**M&R Reciprocating engine**	**M&R Rotofilter**	**M&R Pumps**	**M&R Blowers**
I	0.001	-0.164	0.001	0.002	0.002	0.017	≈0.000	0.004	0.014
II	0.001	-0.099	0.001	0.002	0.002	0.012	≈0.000	0.003	0.012
III	0.002	-0.059	≈ 0.000	0.003	0.003	0.010	≈0.000	0.005	0.019
IV	0.002	-0.053	≈ 0.000	0.003	0.004	0.009	≈0.000	0.006	0.020
V	0.001	-0.108	0.001	0.003	0.003	0.011	≈0.000	0.004	0.014

	€·kgCOD$_{rem}^{-1}$						
Scenario	**Replacement UF Membrane**	**M&R Sludge centrifuge**	**M&R Civil work**	**M&R Pipes and diffusers**	**Discharge fee to surface water**	**Replacement DM**	**OPEX**
I	0.011	0.001	≈0.000	≈0.000	0.011	0.002	**-0.099**
II	0.012	0.001	≈0.000	≈0.000	0.010	0.002	**-0.043**
III	0.021	0.001	≈0.000	≈0.000	0.017	0.003	**0.024**
IV	0.028	0.002	≈0.000	≈0.000	0.020	0.003	**0.041**
V	0.017	0.001	≈0.000	≈0.000	0.014	0.002	**-0.038**

M&R: Maintenance and replacement

7.5.7.3. Allocation of operating expenditures weights

Table 7.11S shows the allocation of operating expenditures weights without considering net energy cost.

Table 7.11S. Allocation of operating expenditure (%).

	%														
Scenario	**Power cost**	**Total Energy Consumption**	**Membrane Cleaning**	**Sludge conditioning**	**Sludge disposal**	**M&R Reciprocating Engine**	**M&R Rotofilter**	**M&R Pumps**	**M&R Blowers**	**Replacement UF Membrane**	**M&R Sludge centrifuge**	**M&R Civil work**	**M&R Pipes and diffusers**	**Discharge fee to surface water**	**Replacement DM**
I	0.9	46.2	0.6	1.7	2.0	14.0	0.2	2.8	11.5	9.2	0.7	≈ 0.0	≈ 0.0	8.9	1.3
II	1.1	42.6	0.9	1.9	2.2	12.0	0.2	2.9	11.8	11.6	0.8	≈ 0.0	≈ 0.0	10.4	1.6
III	1.1	45.3	0.1	1.9	2.2	6.3	0.3	3.1	12.4	13.7	0.9	≈ 0.0	≈ 0.0	11.1	1.7
IV	1.2	41.0	0.1	1.9	2.2	5.6	0.3	3.4	12.5	17.2	1.0	≈ 0.0	≈ 0.0	12.1	1.7
V	1.1	43.5	1.0	2.1	2.4	9.0	0.3	3.4	11.0	13.0	0.9	≈ 0.0	≈ 0.0	10.7	1.6

M&R: Maintenance and replacement

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8. Life cycle assessment of AnMBR technology for urban wastewater treatment: a case study based on a demo-scale AnMBR system

Abstract

This study aims at assessing the environmental performance of a projected full-scale anaerobic membrane bioreactor (AnMBR) treating urban wastewater (UWW) at ambient temperature. To this aim, data from an AnMBR demonstration plant equipped with commercially available equipment, including industrial hollow fiber and degassing membranes, was used for projecting a full-scale facility. The use of real operation data allows to obtain robust results that contribute to improve the knowledge of the environmental performance of this technology, pointing out its strengths and the challenges that still need to be addressed. Life cycle assessment (LCA) was applied by means of Ecoinvent data base and ReCiPe2016 methodology considering 1 kg of removed COD as functional unit. Additionally, sensitivity and uncertainty analysis were conducted. Energy balance showed AnMBR performing as energy producer (net energy surplus up to -0.688 $kWh \cdot kgCOD_{rem}^{-1}$) and carbon sink (emissions credit up to 0.223 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$). Results also showed energy recovery, heavy metals in sludge, dissolved methane in the effluent, and effluent nutrient content as the most important aspects affecting LCA outcome. Construction phase affected some impact categories significantly (e.g., 51-71 % in mineral resource scarcity, 18-27 % in fossil resource scarcity, 21-28 % in water consumption), therefore its exclusion should be carefully evaluated. CHP efficiency, dissolved methane recovery, filtration productivity, membrane scouring, reactor mixing, HRT and SRT appeared most influencing parameters. Finally, actions leading to increase the recovery and valorization of dissolved methane and/or of nutrients through, for instance, fertigation, improve the environmental performance of AnMBR for UWW treatment.

Keywords

Municipal wastewater treatment; semi-industrial scale system; prototype AnMBR; environmental assessment; life cycle assessment; sewage

Highlights

Environmental feasibility of a projected full-scale AnMBR was evaluated.
Results of GHG emissions in the -0.223:0.020 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ range.
Environmental benefits in most of the impact categories were reached.
Energy and nutrients recovery identified as key factors for AnMBR LCA.

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8.1. Introduction

The study of the environmental performance of water management has been consolidated as a key element to promote innovation and sustainability within this sector (Corominas et al., 2013). In this context, Life Cycle Assessment (LCA) has been established as a suitable instrument for evaluating the environmental impacts of the water infrastructure (catchment, purification, distribution, sewer systems, wastewater treatment, reclamation, discharge, etc.), providing valuable information on systems design, operation of facilities and policy development (Byrne et al., 2017).

Despite the unquestionable benefits of wastewater treatment (WWT), the conventional processes applied to obtain adequate effluent quality and guarantee the protection of the environment and public health are based on energy-intensive and chemical-dependent technologies (P.L. McCarty et al., 2011) with a considerable associated impact. Furthermore, increased pressure on the environment is foreseen since wastewater production and the associated treatment are expected to increase (Qadir et al., 2010) while the authorities are tightening the regulations on waste (Collivignarelli et al., 2019). Therefore, sewage management strategies should focus on saving energy and chemicals. The progress towards sustainability in the wastewater management sector should also include shifting from the conventional pollutant-removal concept to the resource-recovery paradigm, which offers advantages in reclaimed water, energy and the valuable materials embedded in wastewater, in line with the principles of the Circular Economy (Robles et al., 2020).

Anaerobic membrane bioreactors (AnMBR) have been proposed as an alternative technology to conventional treatments based on aerobic processes (Jiménez-Benítez et al., 2023; Robles et al., 2022) and could overcome some of the drawbacks mentioned above and contribute to enhance sewage treatment sustainability. While anaerobic microorganisms produce methane from organic matter, which acts as an energy carrier and improves the energy balance (Krzeminski et al., 2017b), they also provide a nutrient-rich effluent that can be fed to nutrient recovery processes (struvite crystallization, membrane contactors, photobioreactors…) or directly reused through fertigation, i.e., direct agriculture reuse of the effluent (Jiménez-Benítez et al., 2020a). On the other hand, membrane technology ensures reduced downstream solids and pathogen concentrations making reclaimed water attractive for other uses (irrigation, aquifer recharge, etc.), while the compactness of membrane modules reduces a facility's footprint and thus favors the retrofitting of existing treatment plants operating in a limited space (Xiao et al., 2019). Membrane units also decouple sludge retention time (SRT) and hydraulic retention time (HRT), enabling

AnMBRs to operate at low/middle temperatures since the membranes avoid microorganisms wash-out from the reactor (Smith et al., 2014).

To the best of author's knowledge, few LCAs have been carried out on AnMBRs for urban wastewater treatment, while the comparison of published studies is somewhat challenging due to the diversity of methods (ReCiPe, CML-IA, TRACI, etc.), functional units (m^3 of treated water, kg PO_4^{-3} removed, etc.) and the initial assumptions used. For example, the facility construction phase has traditionally been excluded from the LCA since its impact was considered negligible compared to those related to the operating phase (Renou et al., 2008). However, Morera et al. (2017) more recently concluded that the environmental loads of the construction phase have been underestimated due to a lack of detail when drawing up the inventories. These authors found a significant impact of the construction phase (over 5 %) on most of the categories evaluated, and especially in the case of metal depletion (63 %) using the ReCiPe method. Efforts are now being made to enhance LCA studies on wastewater treatment aimed at clarifying these uncertainties and improving their quality and comparability (Corominas et al., 2020).

A wide implementation of AnMBR for UWW is still hampered by the phenomenon of membrane fouling, whose management may imply an increase in membrane surface requirements and energy and chemicals consumption, thus affecting the economic and environmental viability of this technology. Fouling is governed by different interrelated factors, among which the concentration and nature of the mixed liquor solids, membrane scouring intensity, SRT or operating temperature can be mentioned (Krzeminski et al., 2017a). In this sense, high temperatures diminish mixed liquor viscosity, which boosts filtration but also could make microorganism to release extracellular polymeric substances (EPS) and/or Soluble microbial products (SMP), which increments fouling (Lin et al., 2009; Robles et al., 2013). Fouling-prevention strategies based on gas sparging are effective, but they increase the filtration energy demand. On the other hand, increased SRT might enhance EPS and SMP consumption, although it implies increasing the solids concentration, which hinders the filtration process.

Energy demand, chemical consumption, nutrient release and the methane dissolved in the effluent have been identified as key factors in driving AnMBR environmental performance and determine its comparison with other technologies. Pretel et al. (2016b) estimated the environmental impact of different aerobic-based wastewater treatment plants (WWTP) (conventional activated sludge-CAS; aerobic membrane bioreactors-AeMBR) and AnMBR both alone and combined with post-

treatment for nutrient removal. The results showed that AnMBR alone presented the lowest environmental impact within all the categories evaluated except eutrophication, although eutrophication loads could also be reduced if AnMBR is combined with a CAS-based post-treatment or nutrient recovery processes. Smith et al. (2014) also compared AnMBR to various aerobic-based WWTPs (high rate activated sludge with anaerobic digestion-HRAS+AD; CAS and anaerobic digestion-CAS+AD; AeMBR and anaerobic digestion-AeMBR+AD). The findings showed that the methane dissolved in AnMBR effluent accounted for 75 % of the global warming impact, which indicates that dissolved methane recovery is a key point to be addressed. The results also demonstrated improved AnMBR environmental performance when treating high-loaded wastewater, being comparable to HRAS+AD and lower than CAS+AD in some impact categories. Cashman et al. (2018) compared decentralized AeMBR and AnMBR for different population densities and treatment flows and found that, for all the scales evaluated, AnMBR at 20 ºC presented the highest net energy benefits and concluded that AnMBRs operated at lower temperatures in warm climates were a promising technology. Xu et al. (2020) found that AnMBR had a lower environmental impact than AeMBR when the system boundary was expanded to include effluent reuse and concomitant nutrient recovery through crop uptake. Without this boundary expansion and in disagreement with Pretel et al. (2016b), AnMBR showed worse performance but, as commented in regard to Smith et al. (2014), this contradiction may be due to differences in influent quality: Pretel et al. (2016b) evaluated medium/high-loaded wastewater, while Xu et al. (2020) considered a low-loaded influent. Finally, Pretel et al. (2016a) assessed the economic and environmental performance of an AnMBR treating UWW and food waste. The results showed that codigestion allowed a net energy production, improved the economic sustainability (savings up to €0.023 per m^3 of treated water) and reduced overall environmental impacts of AnMBR technology.

The main goal of this study was thus to apply the LCA method to evaluate the environmental performance of a projected full-scale AnMBR system (treatment capacity of 16000 $m^3 \cdot d^{-1}$) for urban wastewater (UWW) treatment. Unlike other studies, mainly based on pilot plants and simulation data, the results were obtained using real data from an AnMBR demonstration plant treating UWW under different operating conditions (see Jiménez-Benítez et al. (2023); Robles et al. (2022), (2020b)). Due to the dimensions and equipment of this facility, the results can be regarded as robust information for scaling up AnMBR technology for industrial-scale urban WWTPs.

8.2. Materials and methods

8.2.1. AnMBR demo-plant description

The demonstration plant mainly consisted of a 40 m^3 anaerobic reactor (AnR) (34.4 m^3 working volume) connected to three 0.8 m^3 membrane tanks (MT-X) of 0.7 m^3 working volume. Each MT was equipped with an ultrafiltration (UF) hollow-fiber membrane module (PURON® PSH41, 0.03-µm pore size, filtration area of 41 m^2). The plant also included a degassing membranes (DM) system to recover the methane dissolved in the effluent (PermSelect® Silicone Hollow Fiber Membrane; filtration area of 2.1 m^2). The demonstration plant also included different on-line sensors and measurement equipment to monitor and register data on flow rates, liquid levels, pressure (transmembrane, inlet and outlet for blowers, and gas), temperature and methane production and recovery. For further details, see Robles et al. (2020b) and supplementary material.

8.2.2. AnMBR demo-plant operation

Effluent from the pre-treatment unit of the "Alcázar de San Juan" WWTP (screening and sand removal) was pumped into the anaerobic reactor after further pre-treatment in the rotofilter (RF) and homogenization in the equalization tank (ET). The mixed liquor was continuously recycled through the MTs, where the final effluent was obtained by vacuum filtration. In order to maintain appropriate mixing conditions in the anaerobic reactor, part of the biogas produced was constantly recirculated from the headspace to the bottom of the reactor through coarse bubble diffusers, while another fraction of the biogas produced was pumped to the bottom of the MTs for membrane scouring. For further details, see Robles et al., (2020b).

The demo-plant was operated during 580 days under different operating conditions from which five operating periods have been selected to design and simulate the performance of a projected full-scale AnMBR system to treat the Alcázar de San Juan WWTP flow rate (16000 $m^3 \cdot d^{-1}$). The operating periods have been named by combining the letter "H" to indicate the HRT followed by its average value (subindex) and the letter "T" to indicate the temperature followed by its average value (subindex): i) $H_{41}T_{27}$; ii) $H_{25}T_{24}$; iii) $H_{26}T_{19}$; iv) $H_{26}T_{27}$; and v) $H_{41}T_{18}$. Complete characterization of the operating periods evaluated in this study are provided in supplementary material (see Table 8.1S).

The influent wastewater was characterized by a high COD (755±224-1403±532 $mg \cdot L^{-1}$) and sulfate (125.4±47.6-172.2±28.5 mg SO_4^{-2}-$S \cdot L^{-1}$) concentrations due to dairy- and wine-industry contributions. Despite the high influent COD concentration, COD removal efficiencies of 90-92% were achieved with concentrations below the European discharge limits (125 $mg \cdot L^{-1}$). Further details of the biological performance of the demo-plant can be found in Robles et al. (2022).

Regarding the filtration process, membrane modules were operated at 20 ºC-standardized gross transmembrane fluxes ($J_{20\ gross}$) between 15-21 $L \cdot m^{-2} \cdot h^{-1}$ and specific gas demands per volume of permeate (SGD_P) between 6-14 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$. These conditions resulted in fouling rates of between 0.3 and 4.7 $mbar \cdot d^{-1}$. Details on filtration process of the demo-plant can be found in Jiménez-Benítez et al. (2023).

8.2.3. System boundary and functional unit

According to ISO 14040:2006, the LCA framework applied in this study was subdivided into four stages: (1) goal and scope definition phase, (2) inventory analysis phase, (3) impact assessment phase, and (4) interpretation phase. The life cycle inventories (LCI) of individual materials and processes were compiled using the *Ecoinvent 3.0* and *Industry data 2.0* databases accessed via SimaPro (PRé Consultants; The Netherlands) (see Table 8.1). A functional unit of 1 kg of removed COD (kg COD_{rem}) was selected. A total facility life span of 20 years was set.

The following system boundaries were considered:

- The flow diagram considered in this LCA begins with the rotofilter treatment step. Previous screening and sand removal systems are not included since they are common to most treatment systems like CAS.
- Construction (anaerobic reactors, membrane tanks, pipes and equipment), facility operation, and demolition phases (materials disposal and recycling) were considered. Transport of materials, chemicals and sludge (assuming a distance of 20 km for construction and operation and 30 km for demolition) were also incorporated.
- The building work and infrastructure life cycles were set to 20 years (Corominas et al., 2020). Blower and pump lifespans were estimated from the total working hours provided by the manufacturers (50000 and 75000 h, respectively). A 10-year lifetime was set on other equipment (diffuser, reciprocating engines). UF membrane lifetime

was estimated according to the total chlorine contact specified by the manufacturer (500000 h). A 10-year lifespan period was set to DM.

- Neither reuse of nutrients in the effluent for fertigation purposes nor nutrient recovery or removal by other technologies were considered.
- The waste sludge fate was set as follows: 90 % to agricultural application, 10 % to landfill (in line with data published by the Spanish Ministry of Ecological Transition and Demographic Challenge). Nitrogen and phosphorus sludge content was considered available for crops, therefore equivalent avoidance of mineral fertilizer production was contemplated.
- N_2O emissions due to sludge agricultural application were included as 1.18% of total nitrogen content, in accordance with Doka (2009).
- Disinfection treatment step was considered implicit in UF performance as per Ferreiro et al. (2021), Foglia et al. (2020) and Yang et al. (2021), among others.
- According to the IPCC Guidelines for National Greenhouse Gas Inventories (Eggleston et al., 2006), biogenic CO_2 emissions from wastewater treatment were not taken into account to calculate greenhouse gases (GHG).
- According to Kidd et al. (2007), sludge heavy metal content from municipal wastewater treatment was considered, i.e., the industrial contribution is considered to be low (see supplementary material). Displacement of heavy metals in the avoided mineral fertilizers was not taken into account, due to a lack of information about their content in commercial fertilizer.
- The energy balance in each operating period was calculated as shown in Jiménez-Benítez et al. (2023). The material impact of DM and reciprocating engines for energy recovery were thus included. Moreover, it is considered that the energy recovered is used in its entirety, i.e., if the energy recovered exceeds the plant requirement, it is considered that the energy surplus is discharged into the electrical grid and is consumed outside the facility.
- Dissolved methane recovery by means of DM was included. Recovery percentages in the 80-84 % range was considered based on the environmental optimum performance obtained by Sanchis-Perucho et al. (2021).
- Fugitive methane emissions due to the stripping of non-captured dissolved methane in the effluent to the atmosphere were considered. These emissions have been estimated as the difference between the methane content in the effluent and that recovered in accordance with the previous point.

- Potential biogas treatments to adapt its quality to CHP requirements (e.g., removal of hydrogen sulfide or siloxanes) have been excluded from the scope of the analysis due to the lack of information on the concentration of these compounds.
- Emissions to air of given compounds (e.g., CO, SO_2, NO_2, non-methane volatile organic compounds) resulting from biogas combustion (through CHP) were excluded due to a lack of information.

LCA was based on the ReCiPe 2016 method and a hierarchist approach. A general discussion based on the 18 midpoint impact categories provided by ReCiPe 2016 method is offered. Moreover, global warming, mineral and fossil resource scarcity, freshwater eutrophication, marine eutrophication and water consumption were selected in this work as key process indicators and a deeper analysis of them was conducted. The endpoint areas (damage to human health; damage to ecosystems; and damage to resource availability) proposed in ReCiPe 2016 methodology were also considered in this study. The results of all mid-point and end-point impact categories can be found in supplementary material.

8.2.4. Global sensitivity and uncertainty analysis

Sensitivity analysis (SA) and uncertainty analysis (UA) were conducted to identify influential parameters on model outputs and related uncertainty. 2000 model runs (Monte Carlo method) were performed for both SA and UA. In both cases, the Latin hypercube sampling (LHS) method was used to construct the sampling matrix, applying a variability of 5 % for the membrane and equipment weight quoted by manufacturers and 10 % for the rest of parameters considered (further details can be found in supplementary material). The SA was carried out by means of the Standardized Regression Coefficients (SRC) method applied to all variables and constants considered for modelling the projected full-scale AnMBR system. An absolute value of 0.1 was selected as the threshold value for the standardized regression slope (b_i) for identifying influential input model factors. The UA was conducted to evaluate the propagation of uncertainty in the results, which were assessed by means of i) the 5th and 95th percentiles and ii) the empirical cumulative distribution function (eCDF). The above-mentioned midpoint impact categories (global warming, mineral and fossil resource scarcity, freshwater eutrophication, marine eutrophication and water consumption) were used as model outputs for both SA and UA.

Table 8.1. Materials and processes selected for the Life Cycle Inventory.

Construction materials, kg·kg COD_{rem}^{-1}	Concrete *(1 m3 Concrete, normal {RoW}| market for | APOS, S (Ecoinvent 3))* Chromium steel *(1 kg Steel, chromium steel 18/8 {GLO}| market for | APOS, S (Ecoinvent 3))* Cast iron (*1 kg Cast iron {GLO}| market for | APOS, S (Ecoinvent 3))* Polyester resin (*1 kg Polyester resin, unsaturated {GLO}| market for | APOS, S (Ecoinvent 3))* Polypropylene (*1 kg Polypropylene, granulate {GLO}| market for | APOS, S (Ecoinvent 3))* PVC *(1 kg PVC pipe E (Industry data 2.0))* Silicone *(1 kg Silicone product {GLO}| market for | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 16-32 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3)*	
Energy consumption, kWh·kg COD_{rem}^{-1}, *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Energy avoided (energy recovery from methane), kWh·kg COD_{rem}^{-1} *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Reagent consumption, kg·kg COD_{rem}^{-1}	Polyelectrolyte *(1 kg Polyacrylamide {GLO}| market for | APOS, S (Ecoinvent 3))* NaOCl *(1 kg Sodium hypochlorite, without water, in 15% solution state {GLO}| market for | APOS, S (Ecoinvent 3))* Citric acid *(1 kg Citric acid {GLO}| market for | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
Discharge to air, kg·kg COD_{rem}^{-1}	Methane CH_4 *(AnMBR, 1 kg emissions of CH4 to air)*	
Discharge to water, kg·kg COD_{rem}^{-1}	Ammonium NH^{+}_4 *(AnMBR, 1 kg emissions NH^{+}_4 to water)* Total phosphorous, PO^{-3}_4 *(AnMBR, 1 kg emissions PO^{-3}_4 to water)*	
Discharge to soil, kg·kg COD_{rem}^{-1}	Disposal to agriculture *(1 kg Solid manure loading and spreading, by hydraulic loader and spreader {GLO}| market for | APOS, S (Ecoinvent 3))*	N to soil *(AnMBR, 1 kg emissions N to soil through fertigation)*
		PO^{-3}_4 to soil *(AnMBR, 1 kg emissions PO^{-3}_4 to soil through fertigation)*
		N-based fertilizer (*1 kg Nitrogen fertiliser, as N {GLO}| market for | APOS, S)*
		P-based fertilizer (*1 kg Phosphate fertilizer, as P2O5 {GLO}| market for | APOS, S (Ecoinvent 3))*
		Cd to soil *(AnMBR, 1 kg emissions Cd to soil through sludge spreading)*
		Co to soil *(AnMBR, 1 kg emissions Co to soil through sludge spreading)*
		Cr III to soil *(AnMBR, 1 kg emissions Cr^{+3} to soil through sludge spreading)*
		Cu to soil *(AnMBR, 1 kg emissions Cu to soil through sludge spreading)*
		Zn to soil *(AnMBR, 1 kg emissions Zn to soil through sludge spreading)*
		Ni to soil *(AnMBR, 1 kg emissions Ni to soil through sludge spreading)*
		Pb to soil *(AnMBR, 1 kg emissions Pb to soil through sludge spreading)*
		N_2O to air *(AnMBR, 1 kg emissions N_2O to air through fertigation)*
	Disposal to landfill *(1 kg Municipal solid waste {RoW}| treatment of, sanitary landfill | APOS, S (Ecoinvent 3))* Disposal to incineration *(1 kg Digester sludge {GLO}| treatment of digester sludge, municipal incineration | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
Demolition and materials recycling, kg·kg COD_{rem}^{-1}	Steel and Iron (*1 kg Steel and iron (waste treatment) {GLO}| recycling of steel and iron | APOS, S (Ecoinvent 3))* Concrete demolition *(1 kg Waste reinforced concrete {Europe without Switzerland} | treatment of waste reinforced concrete, recycling | APOS, S (Ecoinvent 3))* Polyethylene treatment *(1 kg Waste polyethylene/polypropylene product {Europe without Switzerland} | treatment of waste polyethylene/polypropylene product, collection for final disposal | APOS, S (Ecoinvent 3)* PVC recycling *(1 kg PVC (waste treatment) {GLO}| recycling of PVC | APOS, S (Ecoinvent 3))*	

8.3. Results and discussion

8.3.1. Energy balance

First of all, it should be noted that in a previous life cycle costing (LCC) study carried out for the same full-scale AnMBR plant (Jiménez-Benítez et al., 2023), the energy balance was conducted considering the economic optimum for the operation of the DM unit. This optimum, calculated based on Sanchis-Perucho et al. (2021), was estimated in the 47-55 % range of dissolved methane recovery. Sanchis-Perucho et al. (2021) also included the estimation of the environmental optimum, which raises the % of dissolved methane recovered to the 80-84 % range. The present study, focused on the environmental assessment of the AnMBR technology, has been carried out based on this last recovery percentage range. However, differences in energy balance results are low and it is possible to consult both cases in the supplementary material (Table 8.2S and Table 8.3S).

Since the energy balance has a crucial influence on many of the impact categories evaluated, main results are summarized in this section. All the evaluated operating periods performed as energy producers, especially $H_{41}T_{27}$, $H_{25}T_{24}$ and $H_{41}T_{18}$ (net energy demands (NED) of -0.668, -0.409 and -0.468 kWh·kg COD_{rem}^{-1}, respectively). $H_{26}T_{27}$ showed the less favorable NED (-0.254 kWh·kg COD_{rem}^{-1}), while $H_{26}T_{19}$ obtained an intermediate value (-0.277 kWh·kg COD_{rem}^{-1}). These results were mainly associated with HRT, which affects reactor mixing requirements; membrane scouring; filtration strategy to minimize membrane fouling; reactor temperature; organic loading rate (OLR) and the COD:SO_4^{-2} ratio, which influences methane production and thus recovered energy. AnMBR, thus, appears as a potential alternative to conventional treatments at moderate temperatures for medium-high strength urban wastewater. For further details, see Table 8.2S in supplementary material.

8.3.2. Life cycle assessment

8.3.2.1. Midpoint categories

Table 8.2 shows the LCA results for all the midpoint categories included in ReCiPe 2016 for the five evaluated operating periods. Overall, $H_{41}T_{27}$ resulted in the lowest overall environmental impact of the evaluated operating periods in almost all categories (i.e., global warming, stratospheric ozone depletion, ionizing radiation, ozone formation-human health, fine particulate matter formation, ozone formation-terrestrial ecosystem, terrestrial acidification, freshwater eutrophication, terrestrial ecotoxicity, land use, mineral resource scarcity, fossil resource scarcity

and water consumption). However, $H_{41}T_{27}$ also was found to have a significant impact among the studied operating periods for freshwater and marine ecotoxicity, and human carcinogenic and non-carcinogenic toxicity. Table 8.2 also shows that, in general, the worst environmental results were reached in $H_{26}T_{27}$.

Figure 8.1 shows the effect of the most influential processes on each impact category for $H_{41}T_{27}$ (similar information related to the rest of the operating periods is provided in supplementary material). The main results given in Figure 8.1 generalized to all the operating periods can be summarized as follows:

- **Energy recovery:** high organic loading rate jointly with methane production allowed a higher energy recovery in $H_{41}T_{27}$, $H_{25}T_{24}$ and $H_{41}T_{18}$. Improved total energy recovery offset energy consumption, thus reducing the environmental impact of the following categories: global warming, ionizing radiation, ozone formation (human health and terrestrial ecosystem), fine particulate matter formation, terrestrial acidification and ecotoxicity, mineral and fossil resources scarcities, land use and water consumption.
- **Nutrient recovery:** direct reuse of phosphorus and especially the nitrogen embedded in sludge for fertilizing purposes contributed to reducing the environmental loads associated with the avoidance of mineral fertilizer production. This influence was particularly relevant for stratospheric ozone depletion, terrestrial ecotoxicity, land use, mineral resource scarcity and water consumption. Other categories (e.g., global warming, ozone formation (human health and terrestrial ecosystem), fine particulate matter formation, terrestrial acidification, and fossil resource scarcity also reduced their impact due to nutrient recovery. It is important to highlight that Figure 8.1 foresees that the environmental benefits of nitrogen recovery in agriculture through sludge spreading would compensate for the drawbacks related to the N_2O emissions derived to air (for the categories of global warming and stratospheric ozone depletion), resulting in positive net balances due to this agricultural practice.
- **Release to the environment of the effluent nutrient content:** the concentration of nutrients in the effluent is the factor that determines the eutrophication potential of the technology. However, since functional unit is defined as 1 kg of removed COD, the environmental load for each operating period was also affected by the influent COD:N and COD:P ratios and the organic matter removal efficiency. As a way of example, $H_{41}T_{27}$ and $H_{26}T_{19}$ obtained a very similar total nitrogen concentrations in the effluent (47.9±6.2 and 47.3±4.3 mg $N \cdot L^{-1}$, respectively) but COD removal in $H_{41}T_{27}$ (equivalent to

19138 kg·d^{-1} for 16000 m^3·d^{-1} of treatment flow) was significantly higher than in $H_{26}T_{19}$ (equivalent to 12428 kg·d^{-1}). Therefore, the latter showed higher marine eutrophication. The eutrophication results demonstrated that nutrient recovery in a downstream post-treatment step (e.g., microalgae cultivation, ion exchange, membrane contactors, etc.) or by fertigation (i.e. nutrient-loaded water reclamation for agricultural purposes) should be considered in order to further enhance the environmental feasibility of AnMBR for UWW treatment (Jiménez-Benítez et al., 2020a) and fully leverage resource recovery from the AnMBR effluent. Indeed, reduced environmental loads in other categories influenced by avoiding mineral fertilizer production (e.g., global warming, stratospheric ozone depletion, land use, etc.) could be expected if these nutrients are recovered.

- **Direct GHG emissions due to dissolved methane:** fugitive methane released from the effluent had a significant impact on global warming potential, so that the capture of this gas is a key challenge to be addressed for AnMBR environmental improvement. As way of example, Figure 8.1 shows that 40 % of the global warming load in $H_{41}T_{27}$ was associated with dissolved methane emissions. Increasing its recovery would reduce this impact while improving the NED.
- **Heavy metals:** the heavy metal content considered in the sludge (see supplementary material) mainly affected freshwater ecotoxicity, marine ecotoxicity, and human toxicity, both carcinogenic and non-carcinogenic. Zinc showed a significant impact on human non-carcinogenic toxicity and freshwater and marine ecotoxicity. The two latter categories were also affected by the copper concentration. Nickel appeared as the main metal influencing human carcinogenic toxicity. Figure 8.1 reveals that the benefits of recovering energy and nutrients did not offset the impact of metals associated with human carcinogenic toxicity and the freshwater and marine ecotoxicity impact categories. $H_{26}T_{27}$ obtained the worst performance within these categories due to the combined effect of its moderate dry sludge production (847 t·year^{-1}) and its lower COD removal performance (10815 kg·d^{-1}), while $H_{25}T_{24}$ produced the lowest environmental loads for these categories due to its lower dry sludge production (632 t·year^{-1}) and higher COD removal efficiency (20631 kg·d^{-1}).

As mentioned above, to better analyze the LCA results from the full-scale AnMBR, the following categories were selected as key indicators of the process: global warming potential, mineral and fossil resource scarcity, freshwater and marine eutrophication, and water consumption.

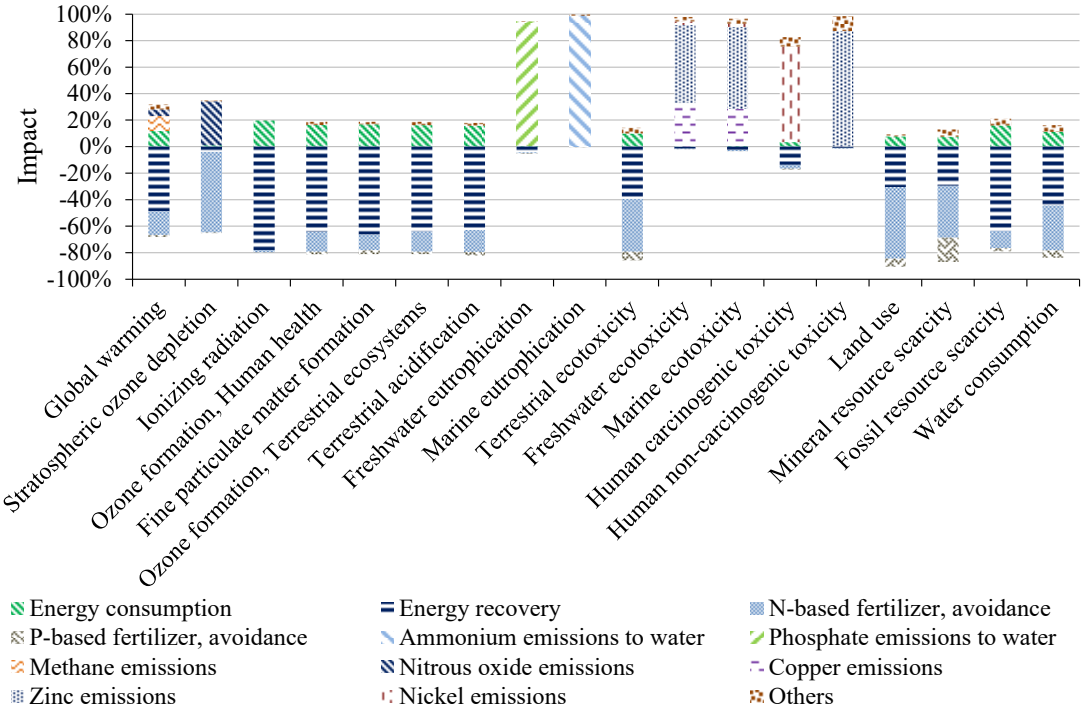


Figure 8.1. Example of allocation of loads per impact category for $H_{41}T_{27}$.

Table 8.2. Relative LCA results for midpoint impact categories in % (the largest impacts are indicated by a blue background and the minor ones by a green background. Negative values indicate environmental benefits).

Impact category	$H_{41}T_{27}$	$H_{25}T_{24}$	$H_{26}T_{19}$	$H_{26}T_{27}$	$H_{41}T_{18}$
Global warming	**-100**	-45	1	**9**	-22
Stratospheric ozone depletion	**-100**	-64	-92	-65	**-63**
Ionizing radiation	**-100**	-61	-43	**-39**	-70
Ozone formation, human health	**-100**	-61	-53	**-42**	-65
Fine particulate matter formation	**-100**	-62	-52	**-43**	-66
Ozone formation, terrestrial ecosystems	**-100**	-61	-53	**-42**	-65
Terrestrial acidification	**-100**	-62	-55	**-45**	-66
Freshwater eutrophication	**76**	81	96	**100**	86
Marine eutrophication	51	54	78	**100**	**49**
Terrestrial ecotoxicity	**-100**	-65	-74	**-53**	-56
Freshwater ecotoxicity	69	**62**	96	**100**	87
Marine ecotoxicity (kg 1,4-DCB)	69	**62**	96	**100**	86
Human carcinogenic toxicity	65	**58**	94	**100**	87
Human non-carcinogenic toxicity	69	**62**	96	**100**	87
Land use	**-100**	-63	-81	**-61**	-66
Mineral resource scarcity	**-100**	-77	-90	-67	**-45**
Fossil resource scarcity	-100	-61	-50	-39	-64
Water consumption	-100	-61	-72	-60	-70

8.3.2.1.1. Global warming potential

Figure 8.2a shows the global warming potential obtained for the evaluated operating periods, considering construction, operating, and demolition phases. The operating phase was the main contributor to this impact category, representing 87-92 % of the direct environmental impact (without considering emission avoidance due to energy recovery and others, e.g., avoidance of mineral fertilizer production) for all the evaluated operating periods. The construction phase weight represented between 8 % and 13 % of the total direct impact, values slightly higher than those reported by Morera et al., (2017) (5-10 %) for a large CAS WWTP using 1 m^3 of treated water as the functional unit. The main contributors to construction phase are concrete applications (44-47 %) followed by PVC of pipes (32-34 %).

Most of the direct environmental load in this category was related to fugitive methane emissions due to downstream stripping of dissolved methane in the effluent (40-55 %). This percentage is lower to that obtained by Smith et al., (2014): 75 % using as functional unit the treatment of 18,950 $m^3 \cdot d^{-1}$). Pretel et al., (2013) evaluated the environmental performance of an AnMBR for urban wastewater treatment and also identified methane emissions as a key point. Reduction of AnMBR carbon footprint therefore necessarily involves improving dissolved methane recovery. In turn, recovering the dissolved methane to produce energy would improve the energy balance. $H_{41}T_{27}$, $H_{25}T_{24}$ and $H_{41}T_{18}$ acted as a carbon sinks (-0.223, -0.101 and -0.050 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$, respectively) when considering the dissolved methane capture efficiencies applied in this study (80-84 %), while $H_{26}T_{19}$ and $H_{26}T_{27}$ showed almost carbon neutral performance: 0.003 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ and 0.020 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$. In this regard, Rodriguez-Garcia et al., (2011) evaluated the operation of different WWTPs, among others, based on conventional activated sludge obtaining values between 0.206 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ and 0.551 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ for facilities without nutrient removal processes and discharging to non-sensitive areas. In the case of the more restrictive situation of conventional activated sludge with nutrient removal and discharging to sensitive areas, the same authors obtained emission values between 0.413 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ and 0.688 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$. It can thus be concluded that AnMBR can be regarded as an alternative treatment to reduce GHG emissions in sewage management when dissolved methane in the effluent is captured.

Enhanced dissolved methane capture efficiency is possible from a technical perspective by increasing the DM filtration area, although this would entail an increase in investment costs, which would affect AnMBR's economic feasibility (Sanchis-Perucho et al., 2020). Moreover, increasing DM filtration area would increment the environmental impacts associated with the use

of DM materials. However, results showed that environmental loads of construction phase were significantly lower than operation phase. Therefore, it is expected that the contribution to the environmental impact associated with an increase in the DM membrane surface would be low. Finally, DM can be combined with additional technologies to prevent dissolved methane release. In this sense, Sanchis-Perucho et al., (2019) reached up to 100 % of methane capture by adding a degassing tower after DM with low investment cost.

On the other hand, it is important to highlight that global warming potential results strongly depended on the energy mix considered. The larger the fossil fuel burden, the higher the CO_{2eq} emissions associated with energy consumption. For example, according to the International Energy Agency (IEA, 2020), in 2018 the main energy sources in Norway, France and Poland were hydro (≈96 %), nuclear (≈72 %) and coal (≈78 %), respectively. Consequently, the global warming potential impact factor in the Ecoinvent database reflects this diversity of technologies and establishes 0.0289 kg CO_{2eq}·kWh^{-1} for the Norwegian mix, 0.0533 kg CO_{2eq}·kWh^{-1} for the French mix and 1.01 kg CO_{2eq}·kWh^{-1} for the Polish one. The appropriate global warming potential comparisons between the WWT technologies should thus take location into consideration (Corominas et al., 2020).

8.3.2.1.2. Mineral and fossil resource scarcity

Mineral resource scarcity was mainly influenced by NED and nutrient recovery through sludge spreading due to avoiding production of P-based and especially N-based fertilizers (see Figure 8.2b). Hence, there is still a great potential for reducing the environmental loads in this category by reducing resource depletion if effluent nutrient content is also recovered and/or reused for agriculture. $H_{41}T_{27}$ showed the lowest environmental impact in this category among studied operating periods due to the high energy recovery and the combination of high total nitrogen and phosphorus concentration in the mixed liquor (505.8±139.4 mg N·L^{-1} and 96.8±12.3 mg P·L^{-1}), the highest dry sludge production (1049 t·$year^{-1}$) and its good COD removal performance (19138 kg·d^{-1}). It should also be mentioned that the construction phase composed between 51 % and 71 % of the direct mineral resource scarcity life cycle impact, when depletion offsets due to energy, nutrient recovery and demolition and recycling are not considered. These results are in line with those obtained by Morera et al. (2017), who evaluated the construction phase contribution to mineral resource scarcity as over 60 %, but considering 1 m^3 of treated water as functional unit. The major contributors to construction phase load are associated to steel chromium used (77-87 %) and concrete applications (12-20 %). Environmental WWT assessment

of scenarios with reduced energy and material recovery may therefore present a non-negligible construction phase impact, at least for the mineral resource scarcity category.

The environmental impact on fossil resource scarcity followed a similar pattern to the global warming potential (see Figure 8.2c), since energy production in Spain still depends strongly on the consumption of fossil fuels (Díaz de la Cruz, 2019). NED is thus the most important factor in the fossil resource scarcity impact category and, accordingly, $H_{41}T_{27}$ presented the lowest environmental load, while $H_{26}T_{27}$ showed the lowest fossil resource preservation. However, this analogous pattern between global warming and fossil resource scarcity may change according to the energy mix selected, as already mentioned. It is also remarkable that both net mineral and fossil resource depletion obtained negative loads in all the operating periods, which indicates that AnMBR contributes to preserving these resources by recycling and valorizing the resources embedded in wastewater and hence contributing to avoid non-renewable resources extraction from conventional sources. These results are in line with Pretel et al. (2013), who also obtained negative loads for abiotic depletion category for wastewater treatments based on AnMBR. Conversely, Niero et al. (2014) and Canaj et al. (2021) obtained positive loads for fossil depletion and fossil and mineral resource scarcity, respectively, assessing the environmental performance of WWTP based on CAS and AD technologies, which confirms the environmental benefits of AnMBR-based treatment over conventional ones.

Even though the operation phase was the major contributor to the fossil resource scarcity input, the construction phase burden forms 18-27 % of the direct load, with PVC of pipes (47-51 %) and concrete applications (27-28 %) as main contributors. Again, this contribution is similar to the results reported by Morera et al. (2017), i.e., ≈20 %, and confirmed that the construction phase could be important in some cases.

8.3.2.1.3. Freshwater and marine eutrophication

Effluent characterization was the main factor affecting both freshwater and marine eutrophication due to its nitrogen and phosphorus content (see Figure 8.2d and e) in line with results obtained by Niero et al. (2014), Pretel et al. (2013) and Rebello et al. (2021), among others. Since freshwater eutrophication is characterized in kg P_{eq}, the trends in this impact category are marked by the effluent phosphorus content and the organic matter removal efficiency in each OP. Results for freshwater eutrophication are in the $2.30 \cdot 10^{-3}$-$2.97 \cdot 10^{-3}$ kg $P_{eq} \cdot kgCOD_{rem}^{-1}$ range, which are higher

than those reported by Canaj et al. (2021) for aerobic-based WWTP ($9.55 \cdot 10^{-4}$ kg $P_{eq} \cdot m^{-3}$), since aerobic-based treatments allows reducing the nutrient content in the effluent.

Despite $H_{26}T_{19}$ and $H_{26}T_{27}$ showed the lowest effluent phosphorus content (6.9±2.8 and 6.3±0.5 mg $P \cdot L^{-1}$, respectively), they had the highest impact within this category due to their comparatively low COD removal (12428 kg·d^{-1} in $H_{26}T_{19}$ and 10815 kg·d^{-1} in $H_{26}T_{27}$). On the other hand, $H_{41}T_{27}$ and $H_{25}T_{24}$ showed the highest phosphorus discharge concentration (8.8±1.9 and 9.9±1.9 mg $P \cdot L^{-1}$, respectively), but their high COD removal allowed them to reduce their relative impact.

Similarly, although marine eutrophication is characterized in kg N_{eq}, the relative impact of each operating period also depended on the COD removal. Results in this category were in the 1.12·10-2-2.27·10-2 kg N_{eq} $kgCOD_{rem}^{-1}$ range, also higher than the one obtained by Canaj et al., (2021), 2.73·10-4 kg N_{eq} m^{-3}. This is coherent with de nitrogen content in its effluent (23.5 mg $N \cdot L^{-1}$), which is half that in this study (37-54 mg $N \cdot L^{-1}$). $H_{41}T_{18}$, which combined the lowest effluent N_T concentration (37.0±11.1 mg $N \cdot L^{-1}$) with a moderate COD removal performance (15376 kg·d^{-1}) showed the lowest marine eutrophication burden. On the other hand, despite presenting similar effluent N_T concentrations, $H_{25}T_{24}$ showed a significantly lower marine eutrophication than $H_{26}T_{27}$, since the COD removal performance of the former (20631 kg·d^{-1}) was double that the obtained in the later (10815 kg·d^{-1}).

The influence of the construction phase on marine eutrophication was found to be negligible, in agreement with Morera et al. (2017). These authors reported a ≈5 % construction phase freshwater eutrophication burden, unlike the present study, in which its contribution was found to be below 1 %. The importance of the N and P sludge concentration on the impact of the agriculture use of biosolids was found to be insignificant.

Finally, it is important to highlight that, although the nutrient concentrations of the effluents from anaerobic treatments present a potential for eutrophication to be considered, it is also true that they represent an opportunity to recover these resources in considerable quantities, which entails obvious environmental and economic benefits. As an example, Jiménez-Benítez et al. (2020) carried out an analysis of the combination of AnMBR and fertigation applied to two case studies in Spain and Italy. The results showed that proper irrigation planning allows reducing the discharges of both N and P into the environment by up to 74 %, which also means a potential of

reducing the need for chemical fertilizers by up to 100 %. In addition, significant economic and CO_2 savings were recorded in the AnMBR+fertigation systems compared to the base scenarios made up of a conventional WWTP and a separate irrigation system.

8.3.2.1.4. Water consumption

Figure 8.2f shows the results of the water consumption impact category. The results indicated that reducing NED through biogas and dissolved methane recovery had a positive effect on the environmental loads. On the basis of the results, it could be also stated that taking advantage of nutrient content and thus avoiding P-based and especially N-based fertilizer production also contributed to a relevant reduction of the total impact in this category.

All the evaluated operating periods resulted in negative environmental loads (between $-4.81 \cdot 10^{-3}$ and $-2.87 \cdot 10^{-3}$ $m^3 \cdot kgCOD_{rem}^{-1}$), which indicates a global water consumption saving. Conversely, Canaj et al. (2021) obtained a water consumption of 0.38 m^3 per m^3 of treated water, but considering groundwater withdrawal for irrigation.

According to the results depicted in Figure 8.1, energy recovery and avoidance of N-based fertilizer production showed a high influence in water consumption impact. Therefore, $H_{26}T_{27}$ comparatively had the worst performance in this impact category since it showed the lowest energy recovery potential and relatively low avoidance of mineral fertilizer production. The opposite results were achieved in $H_{41}T_{27}$, which had the highest methane production and fertilizing potential through sludge spreading. It is important to highlight that water reuse was not considered in this study. Including this practice in the LCA boundaries would prevent freshwater extraction, thus improving this impact category, in line with the results obtained by Cashman et al. (2018) and Canaj et al., (2021).

The construction phase weight in the direct water consumption category made up 21-28 % of its total life-cycle impact (e.g., associated with minerals extraction and processing), without considering the load credits provided by energy recovery, avoidance of fertilizer production and material recycling after dismantling. Demolition also had a significant effect and contributed to reducing the water consumption burden in all the operating periods. Excluding the construction and demolition phases from the LCA boundary for this impact category should be therefore carefully evaluated so as not to affect the results.

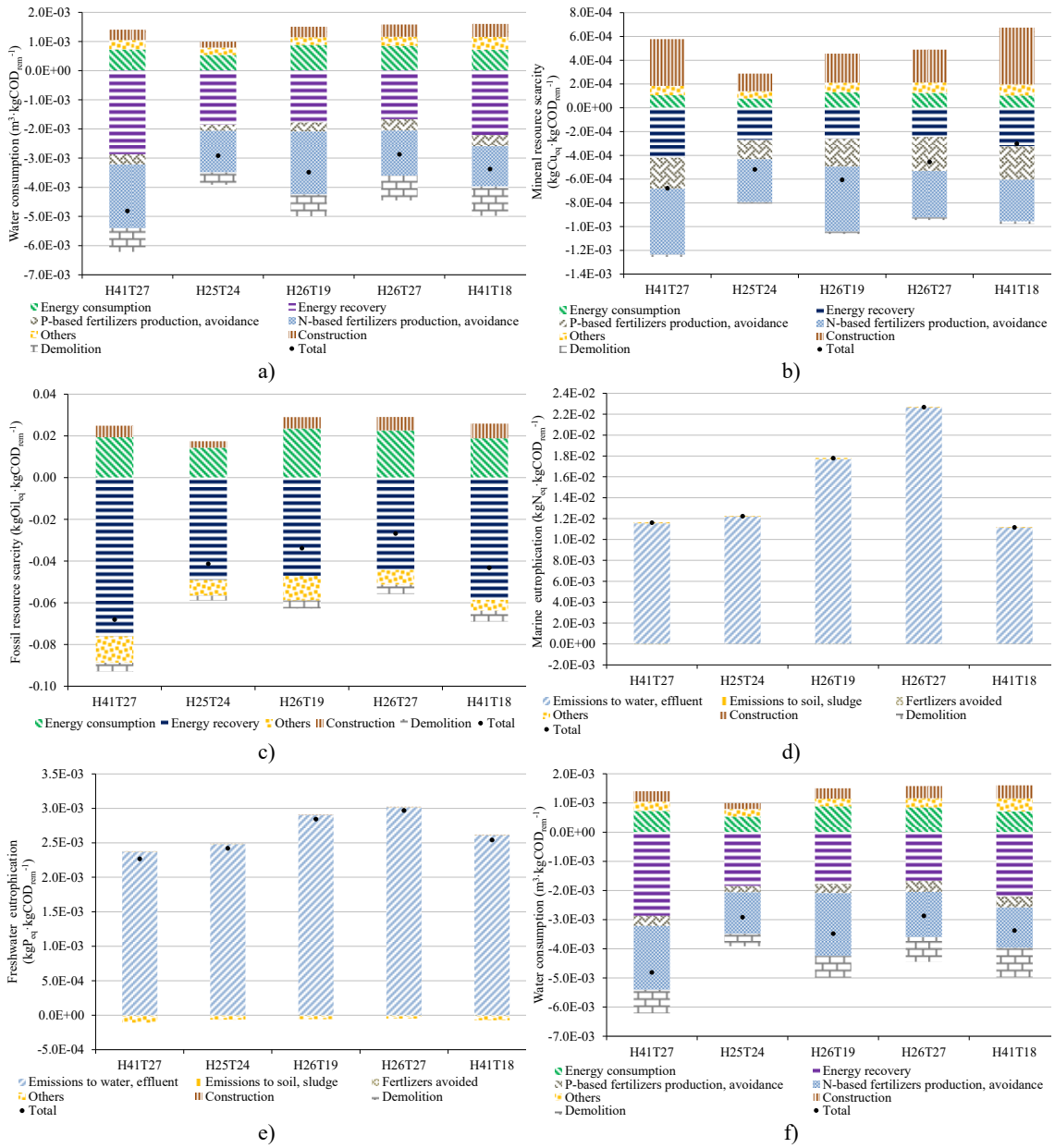


Figure 8.2. Environmental impact through: (a) global warming potential, (b) mineral resource scarcity, (c) fossil resource scarcity, (d) marine eutrophication, (e) freshwater eutrophication, and (f) water consumption.

8.3.2.2. Endpoint categories

Figure 8.3a shows the results for the three endpoint impact categories included in ReCiPe 2016: human health, ecosystems, and resources.

8.3.2.2.1. Human health

The highest impact on human health was reached in $H_{26}T_{27}$, which is consistent with the midpoint results. In this respect, $H_{26}T_{27}$ resulted in the highest impact in terms of human carcinogenic and non-carcinogenic toxicity. As can be seen in Figure 8.3b, the main factor influencing human health was the zinc emissions associated with sludge spreading. Reducing sludge production and its agricultural use or reducing its heavy metal content would address this issue. For example, a reduction of 10% in dry sludge production in $H_{26}T_{27}$ (from 847 $t \cdot year^{-1}$ to 762 $t \cdot year^{-1}$) would reduce its human health impact by 10 % in the operation phase, from $5.070 \cdot 10^{-6}$ to $4.538 \cdot 10^{-6}$ DALY (Disability-Adjusted Life Year). On the other hand, A 10 % reduction in zinc concentration in sludge (from 500 $mg \cdot kg^{-1}$ to 450 $mg \cdot kg^{-1}$) would reduce human health impact to $4.629 \cdot 10^{-6}$ DALY, which represents a reduction of 9 %. In section 8.3.2.2.3 it will be seen that, unlike in the human health, the reduction in sludge production has a detrimental effect on the resources category due to a lower potential use in agriculture of the nutrients present in the sludge. Finally, AnMBR also contributes to human health by the physical disinfection provided by the UF membranes. Despite this effect is not captured by ReCiPe2016, it is clear that preventing pathogens discharge into water bodies enhances human health protection.

8.3.2.2.2. Ecosystems

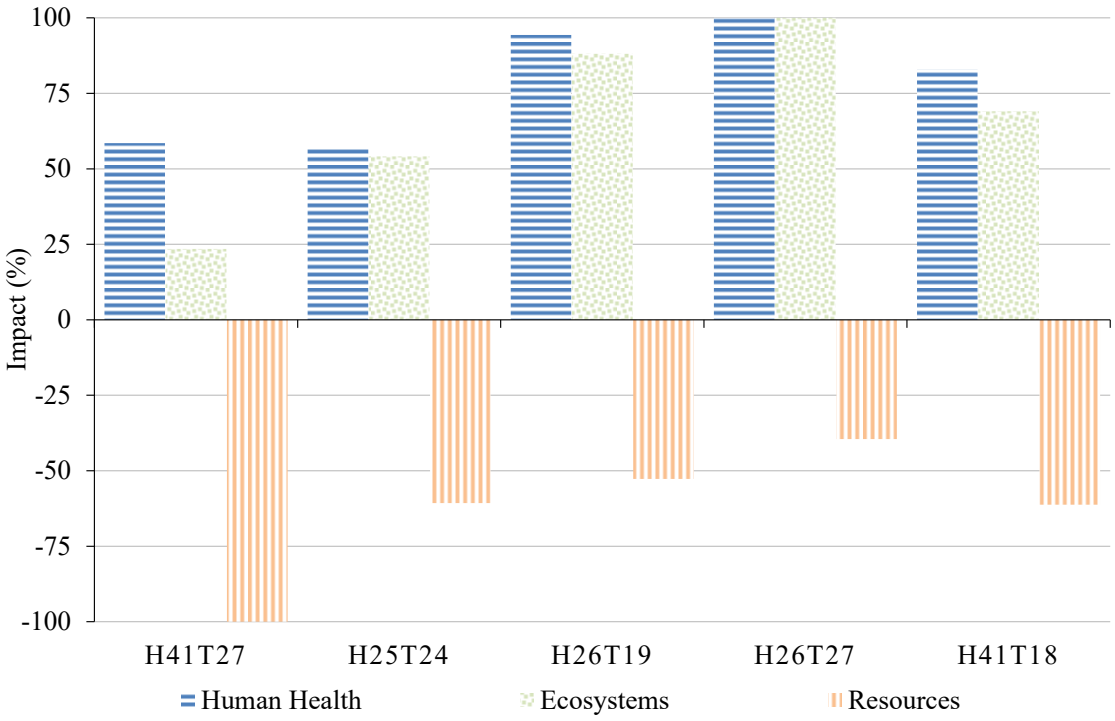
As regards ecosystems, $H_{26}T_{27}$ presented the highest relative impact, while $H_{41}T_{27}$ had the lowest environmental burden. The biggest influence on ecosystems came from phosphorus release, methane emissions associated with effluent discharge, NED, and from zinc released to the soil in a lesser extent. In $H_{26}T_{27}$, the lowest effluent P_T concentration (6.3±0.5 mg $P \cdot L^{-1}$), the relative moderate dry sludge production (847 $t \cdot year^{-1}$), and the low dissolved methane concentration in the effluent (2.6±0.5 $mg \cdot L^{-1}$, mainly due to operating at high temperature: 27±1 ºC) were not able to offset the combination of the lowest resulting NED (-0.254 kWh·kg COD_{rem}^{-1}) and the lowest COD removal performance (10815 $kg \cdot d^{-1}$) of this OP. On the other hand, $H_{41}T_{27}$ presented the minimum impact due to a more favorable NED (-0.668 kWh·kg COD_{rem}^{-1}) and low dissolved methane release (2.6±0.5 $mg \cdot L^{-1}$, operating temperature of 27 ºC), despite its highest dry sludge production (1049 $t \cdot year^{-1}$) and high effluent P_T concentration (8.8±1.9 mg $P \cdot L^{-1}$).

8.3.2.2.3. Resources

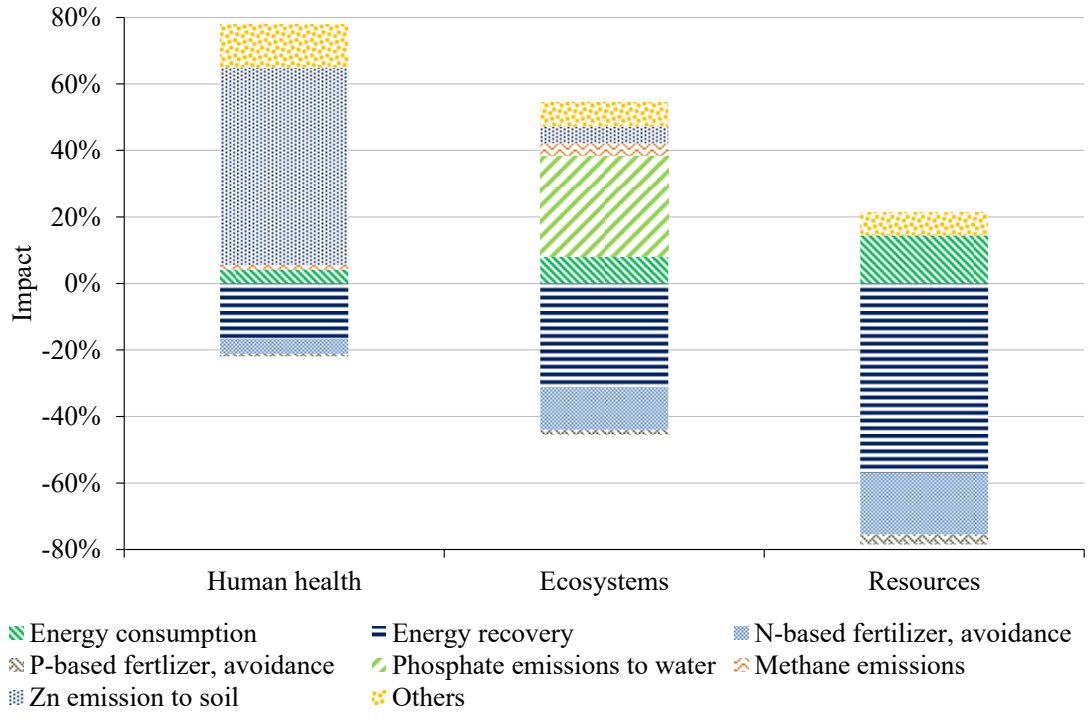
The results obtained for the impact on resources were mainly affected by NED and nutrient recovery for agriculture, especially nitrogen and its associated avoidance of mineral fertilizer production (see Figure 8.3b). Both items gave to all the operating periods evaluated a net impact reduction in this impact category, as previously mentioned for the former midpoint impact categories (mineral and fossil resource scarcities). Again, $H_{41}T_{27}$ showed the best results due to the resulting favorable NED (-0.668 kWh·kg COD_{rem}^{-1}) and high dry sludge production (1049 t·$year^{-1}$), suitable for nutrient recovery through sludge spreading. Conversely, $H_{26}T_{27}$ resulted in a less favorable NED (-0.254 kWh·kg COD_{rem}^{-1}), produced less sludge (847 t·$year^{-1}$) and removed less COD (10815 kg·d^{-1}), with the worst results for ecosystems. It is important to highlight here that sludge production and spreading has opposite effects when analyzing the impact on human health and ecosystems. This is due to the effect of different substances on different areas of protection is considered. In the case of the human health area, the impact is given by the presence of heavy metals in the sludge, therefore, an increase in the amounts destined for agricultural use means increasing the amount of metals deposited in the soil and, consequently, an increased potential for health damage. On the contrary, when the area of resources is considered, the substances of influence are nutrients, so that a greater use of sludge in agriculture reduces the needs of industrial fertilizers and, consequently, a lower consumption of resources.

8.3.2.3. Sensitivity analysis

By way of example, the results from the SA of $H_{41}T_{27}$ are shown in Figure 8.4, providing the main influencing parameters ($b_i \geq 0.1$) for the key midpoint impact categories selected in this work. These factors are CHP electric efficiency; dissolved methane recovery efficiency; total filtration time; SGD_P; rector mixing; HRT; SRT; and emissions of nitrogen to soil.



a)



b)

Figure 8.3. (a) LCA results for endpoint impact categories. (b) Example of allocation of loads per impact category for $H_{41}T_{27}$.

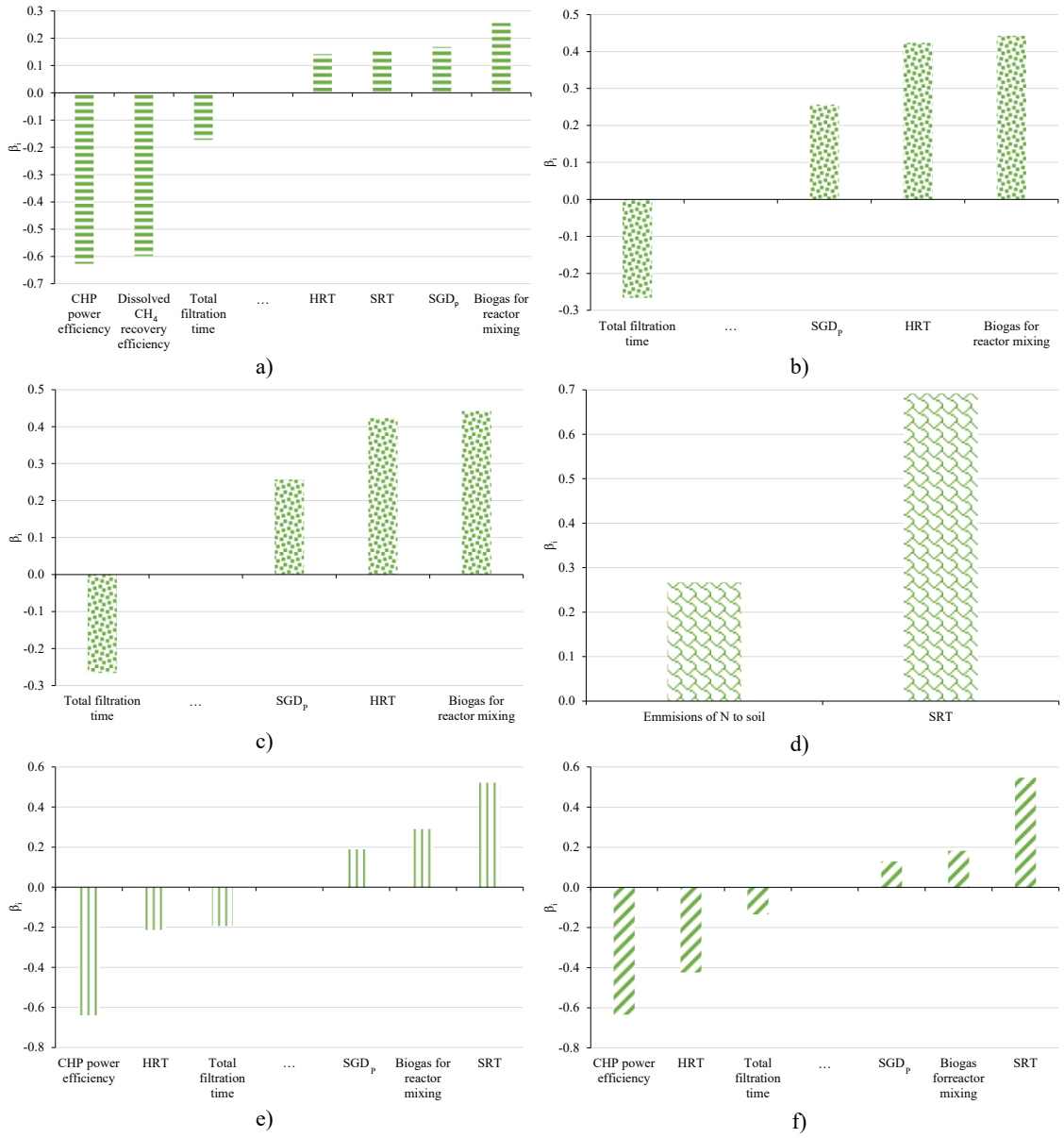


Figure 8.4. Sensitivity analysis of $H_{41}T_{27}$ for (a) global warming potential, (b) mineral resource scarcity, (c) fossil resource scarcity, (d) marine eutrophication, (e) freshwater eutrophication, and (f) water consumption. "…" separates positive from negative results.

8.3.2.3.1. CHP electric and dissolved CH_4 recovery efficiencies

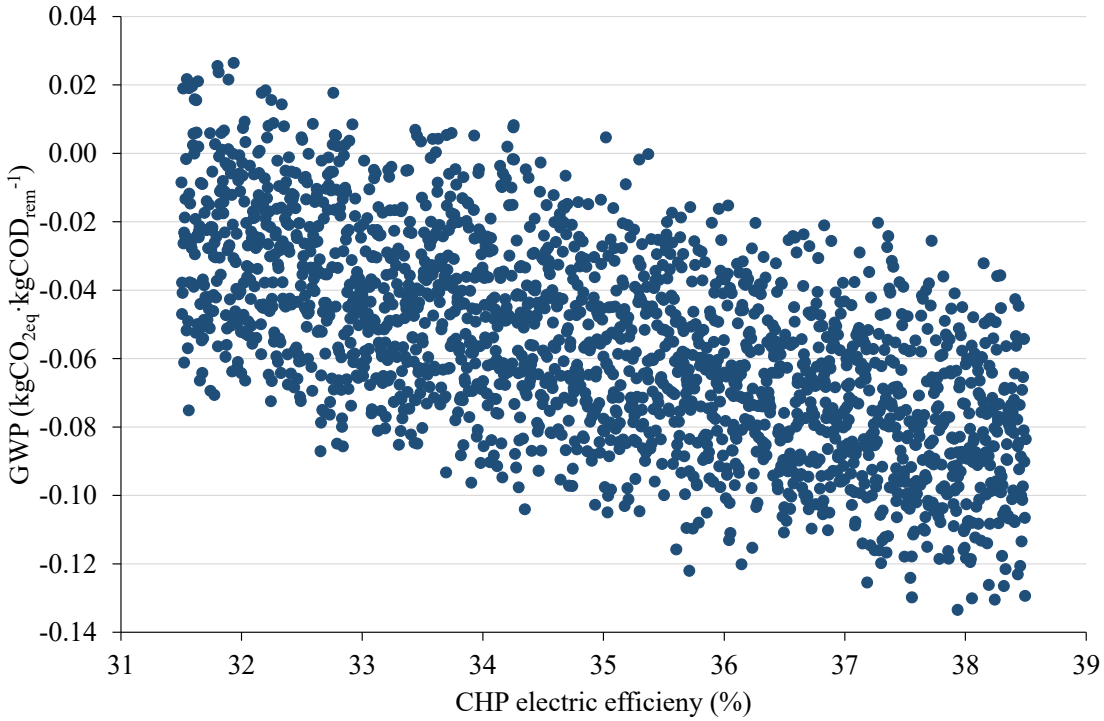
CHP efficiency had a significant influence on global warming potential, fossil resource scarcity, freshwater eutrophication and water consumption, which is consistent with the results given in Figure 8.1. A high energy recovery potential reduces CO_2 emissions associated with fossil fuel

use and prevents fossil resource depletion. Same effect would produce an increase in dissolved CH_4 recovery and the consequent increase in energy production, jointly with a direct reduction in the global warming potential associated with diminishing CH_4 emissions from the effluent. Moreover, reducing the consumption of electricity produced from fossil fuels also makes it possible to improve the water consumption results, since the energy produced from conventional sources (i.e., coal, gas, nuclear, etc.) requires significant water consumption. The relationship between freshwater eutrophication and energy is not so obvious and could be associated with phosphorus emissions during fossil fuel extraction (Gaete-Morales et al., 2019). Therefore, the improvements associated with energy recovery go firstly through the development of CHP technology that allows increasing its efficiency and, secondly, through the optimization of the operation of methane recovery technologies.

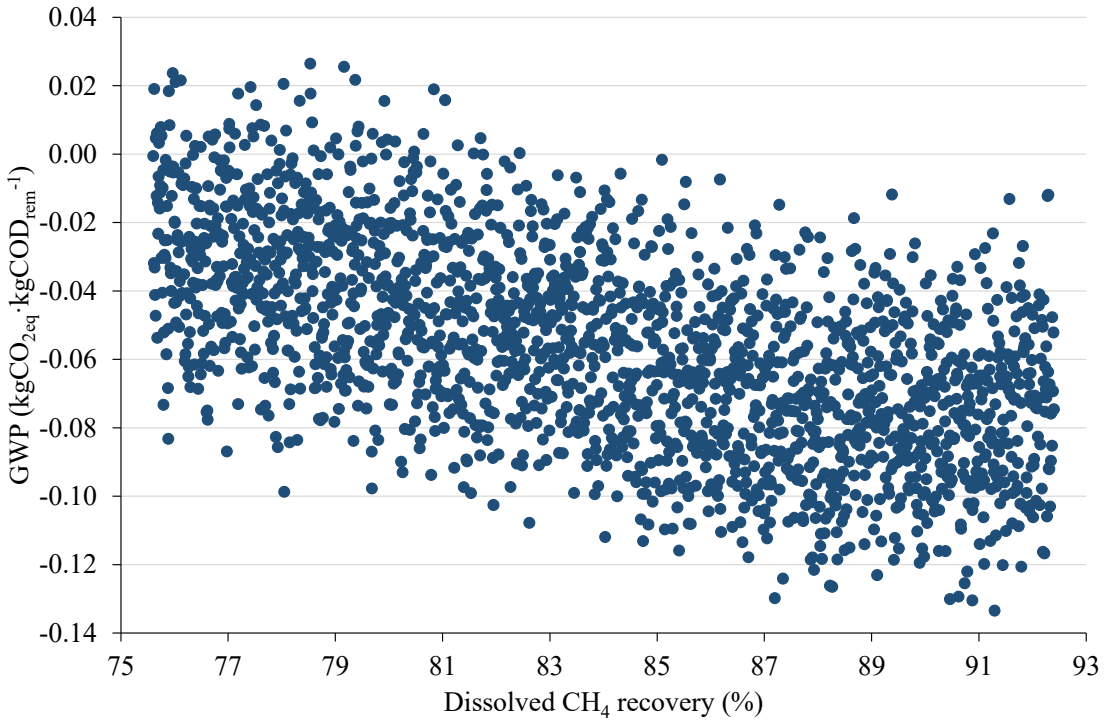
Figure 8.5 shows the Monte Carlo simulation results for global warming potential with regard to CHP power efficiency (Figure 8.5a) and dissolved methane recovery efficiency (Figure 8.5b) in $H_{41}T_{27}$. As already mentioned, higher CHP power efficiencies leads to reduced CO_2 emissions, since the external energy requirements from the grid are lower. The trend of the results shows emissions below -0.4 kg $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ with CHP efficiencies of 38.5 %. Additionally, dissolved methane recovery over 83 % (see Figure 8.5b) would also guarantee negative net GHG emissions. Therefore, the combination of AnMBR and CHP shows a promising performance to contribute to reduce the carbon footprint of wastewater treatment.

8.3.2.3.2. Downtime for physical cleaning

Reducing the downtime for physical cleaning would increase filtration productivity (i.e., total filtration time), while decreasing SGD_P if keeping the same gas sparging intensity for membrane scouring. Under this premise, increasing filtration productivity would reduce energy needs and improve global warming potential, fossil resource scarcity, freshwater eutrophication and water consumption. Therefore, downtime for physical cleaning should be optimized to improve filtration productivity without increasing TMP. Moreover, an enhanced filtration productivity would lead to a reduction in the membrane surface which would also prevent mineral resource scarcity since less material is required in their manufacture.



a)



b)

Figure 8.5. Monte Carlo simulation results. Effect on global warming potential (GWP) output of: a) CHP electric efficiency and b) dissolved methane recovery efficiency for $H_{41}T_{27}$.

8.3.2.3.3. Biogas sparging for membrane scouring and reactor mixing

As commented in section 8.3.1, as membrane scouring and reactor mixing are included among the main energy consumption drivers, increasing them also raises the environmental loads associated with the impact categories significantly influenced by the generation and use of energy: global warming potential, fossil and mineral resource scarcity, freshwater eutrophication and water consumption. Reducing energy consumption for membrane scouring requires going through the optimization of the filtration process, e.g., minimizing gas sparging at constant flux or vice versa while guaranteeing low membrane fouling propensities. On the other hand, an optimization of the reactor mixing would also contribute to minimize the environmental impact associated with this process. This optimization would be based on maintaining the minimum gas flow to avoid sedimentation of the solids in the reactor, considering the rheological and hydrodynamic conditions of the system, e.g., solids concentration, viscosity, gas rising, etc.

8.3.2.3.4. HRT

HRT was identified as an influential input factor on three impact categories. HRT was positively correlated with global warming potential, mineral and fossil resource scarcity. Increasing HRT means increasing the reactor volume for a given treatment capacity. Thus, the emissions associated with material consumption (e.g., concrete) and energy consumption for reactor mixing would increase as the HRT is increased, resulting in increased environmental loads within mineral and fossil resource depletion impact categories. Hence, reducing HRT is a suitable option for improving the environmental performance in these three categories. However, reducing HRT for a given SRT would entail an increase in the mixed liquor total suspended solids (MLTSS) concentration in the reactor, which eventually could increment membrane fouling propensity for a given filtration operating mode (e.g., downtime for physical cleaning, gas sparging intensity for membrane scouring, etc.). Although fouling propensity could be controlled by modifying the filtration operating mode when raising MLTSS, it may impact other dimensions (e.g., energy consumption), thus both filtration process configuration and operation and HRT might be considered jointly for process optimization purposes.

8.3.2.3.5. SRT

SRT was identified to mainly affect methane production, reactor volume, wasting flow rate and nutrients concentration in the effluent. Regarding impact categories, it is positively correlated

with global warming potential, fossil resource scarcity, marine and freshwater eutrophication and water consumption. Increasing SRT generally implies an increase in methane production. However, the operation of the AnMBR demo-plant was already carried out at long SRT (70 d), so the availability of additional methanizable organic matter was not significant and the potential increase in energy recovery was low. On the other hand, maintaining the MLTSS concentration by increasing the SRT implies an increase in the reactor volume. As commented above, increasing the reactor volume would result in increased consumption of construction materials and energy requirements for reactor mixing. In addition, the increase in the SRT implies a slight decrease in the waste sludge and, therefore, in the potential for nutrient recovery through biosolids agricultural use, which limits the environmental benefits associated with the displacement of the manufacture of industrial fertilizers. These combined effects explain why, in this case, an increase in SRT leads to an increase in global warming potential and fossil resource scarcity. However, it is important to highlight that in the case of facilities operating at short SRTs, increasing the SRT could imply a significant increase in methane production, even enough for the effect of increasing SRT to be the opposite of that obtained in this study, thus decreasing global warming potential as increasing SRT. Finally, an increase in the SRT means increasing the mineralization of organic nutrients, increasing the concentration of inorganic nitrogen and phosphorus that is discharged into the environment along with the effluent, although it had a negligible effect in this work due to the long SRT applied. Additional releases of nutrients to the environment would increase the environmental load within eutrophication impact categories.

8.3.2.3.6. Emissions of nitrogen to soil

Emissions of nitrogen to soil from sludge spreading contributes to marine eutrophication (quantified in kg of N_{eq}). On the one hand, A fraction of the nitrogen can percolate through the soil and reach groundwater, through which it is finally transported to the marine environment. On the other hand, runoff can also carry a fraction of nitrogen that will flow towards the sea through surface water. To limit this detrimental effect, the application of agricultural sludge must be carried out following good agricultural practices and adequately balancing the nutrient needs of crops and the amount of sludge to be applied.

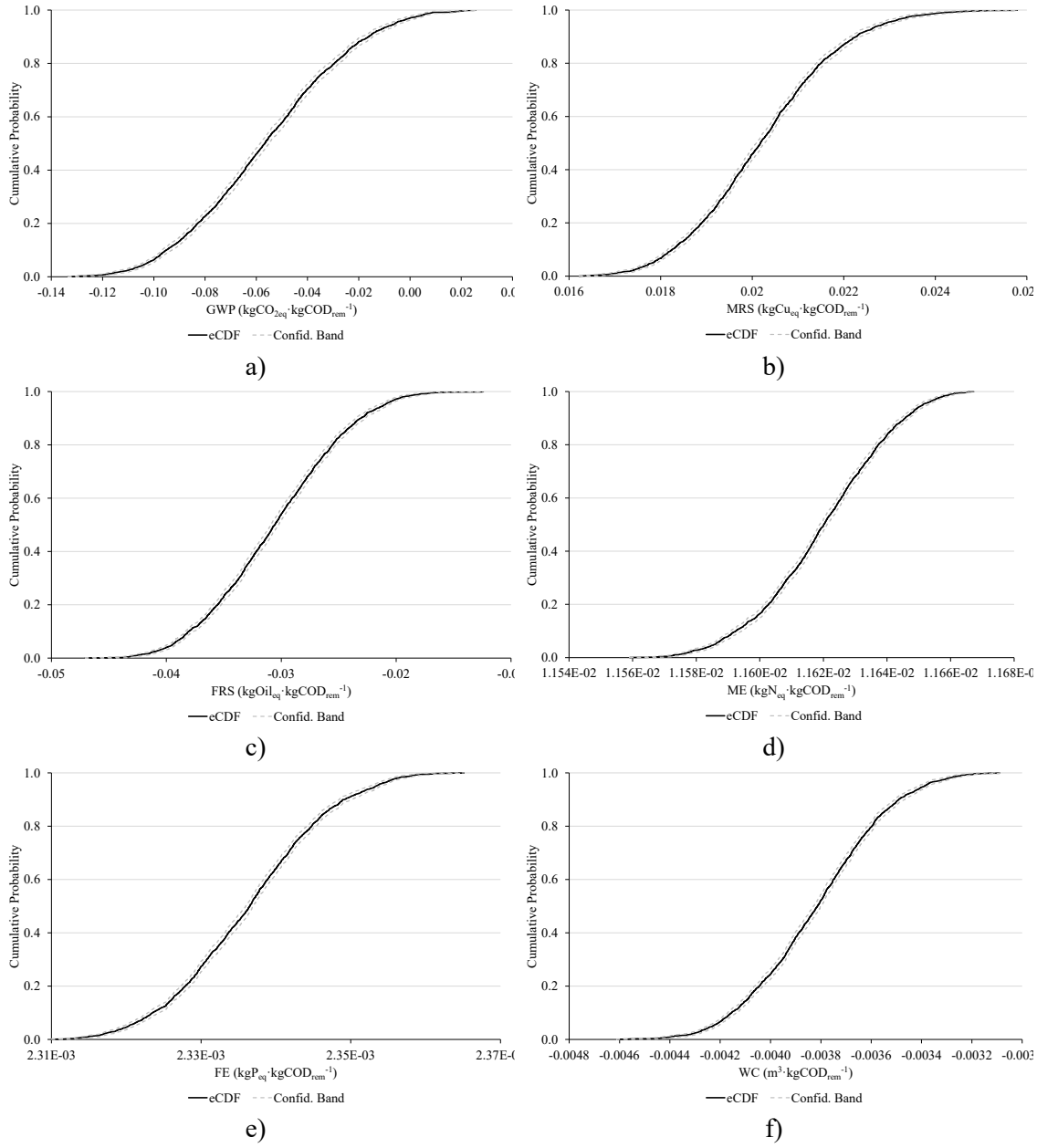


Figure 8.6. Uncertainty analysis results for $H_{41}T_{27}$. Empirical cumulative probability for: (a) global warming potential (GWP), (b) mineral resource scarcity (MRS), (c) fossil resource scarcity (FRS), (d) marine eutrophication (ME), (e) freshwater eutrophication (FE), and (f) water consumption (WC).

8.3.2.4. Uncertainty analysis

Figure 8.6 shows the empirical cumulative function (eCDF) and confidence interval for the six selected midpoint impact categories in $H_{41}T_{27}$ as representative of the study. It can be seen that eCDF follows a standard normal distribution. Table 8.3 shows that the lowest 5th-95th range was obtained for marine and freshwater eutrophication (both almost 0.0 kg N_{eq}·kg COD_{rem}^{-1} and kg P_{eq}·kg COD_{rem}^{-1}, respectively) followed by water consumption, with $1.0 \cdot 10^{-3}$ m^3·kg COD_{rem}^{-1}.

The highest 5th-95th range was for global warming potential (0.144 kg CO_{2eq}·kg COD_{rem}^{-1}) and fossil resource scarcity (0.031 kg Oil_{eq}·kg COD_{rem}^{-1}), while the mineral resource scarcity showed intermediate values (0.009 kg Cu_{eq}·kg COD_{rem}^{-1}). These results therefore indicate that the global warming potential is subject to greater uncertainty and that it is necessary to strengthen the mechanisms to guarantee that the data used in constructing the LCI that influence this impact category are sufficiently robust.

Table 8.3. Uncertainty results for $H_{41}T_{27}$: 5th-95th percentile and 5th-95th range.

	GWP[a]	**MRS[b]**	**FRS[c]**	**ME[d]**	**FE[e]**	**WC[f]**
	kg CO_{2eq}·kg^{-1} COD_{rem}	**kg Cu_{eq}·kg COD_{rem}^{-1}**	**kg Oil_{eq}·kg COD_{rem}^{-1}**	**kg N_{eq}·kg COD_{rem}^{-1}**	**kg P_{eq}·kg COD_{rem}^{-1}**	**m^3·kg COD_{rem}^{-1}**
5th percentile	-0.126	0.017	-0.045	0.012	0.002	-0.004
95th percentile	0.019	0.026	-0.014	0.012	0.002	-0.003
5th-95th range	0.144	0.009	0.031	≈0.000	≈0.000	0.001

[a] global warming potential; [b] mineral resource scarcity; [c] fossil resource scarcity; [d] marine eutrophication; [e] freshwater eutrophication; [f] water consumption

8.4. Conclusions

AnMBR performed as energy producer (NED between -0.277 and -0.688 kWh·kg COD_{rem}^{-1}), being HRT, membrane scouring, filtration strategy, reactor temperature, OLR, and the COD:SO_4^{-2} ratio the main influential parameters on NED.

Energy and nutrients recovery were identified as key factors for improving the environmental performance of a full-scale AnMBR for UWW designed using real data from a demo-scale AnMBR plant. Methane and N_2O release to air, and nitrogen and phosphorus release to soil and water bodies are major drivers for environmental impacts, jointly with heavy metal content in sludge. Degassing membranes for dissolved methane capture appear as a key technology to enhance AnMBR environmental performance, while post-treatment technologies for nutrient recovery/removal (e.g., microalgae cultivation, anammox, ion exchange, membrane contactors, etc.) may be necessary for reducing marine and freshwater eutrophication.

Recovering the dissolved methane with efficiencies of 80-84 % led AnMBR to act as carbon sink (emissions avoidance up to 0.223 $kgCO_{2eq}$·$kgCOD_{rem}^{-1}$). Construction phase contribution to overall LCA was relevant in some operating periods and/or impact categories: 8-13 % in global warming potential, 51-71 % in mineral resource scarcity, 18-27 % in fossil resource scarcity, 21-28 % in water consumption, but negligible in marine and freshwater eutrophication: < 1 %. Therefore, its exclusion from the system boundary should be carefully evaluated.

Finally, the SA confirms that there is room to improve the environmental performance of the AnMBR technology by increasing the potential for energy recovery (improvement in the efficiency of CHP technologies and optimization of dissolved methane recovery technologies), optimization of filtration, membrane scouring and mixing processes, adjustment of HRT and SRT and use of appropriate agricultural practices.

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8.5. Supplementary material

8.5.1. AnMBR demonstration plant

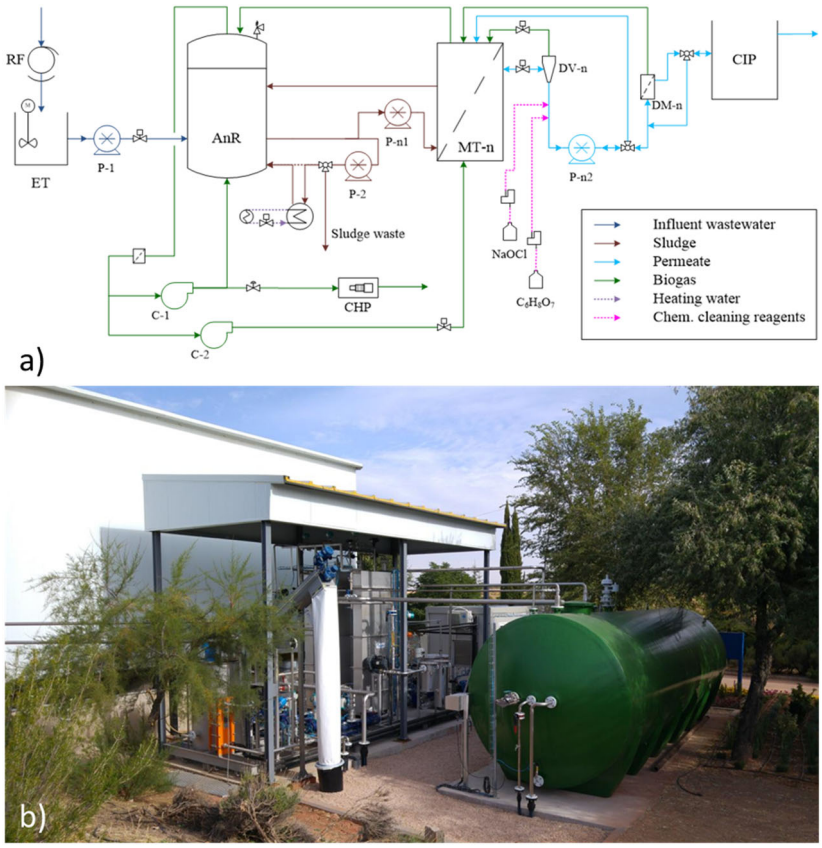


Figure 8.1S. (a) Flow diagram of the AnMBR Plant, and (b) AnMBR plant overview. Note: "n" represents the membrane treatment lane.

8.5.2. Operating conditions and characterization of the selected OPs

Table 8.1S. Influent and effluent characterization, main operating conditions, and main biological and filtration process performance results from the AnMBR prototype.

INFLUENT	Units	$H_{41}T_{27}$	$H_{25}T_{24}$	$H_{26}T_{19}$	$H_{26}T_{27}$	$H_{41}T_{18}$
Flow rate	$L \cdot d^{-1}$	21228±591	33016±2773	33243±3118	34207±1478	20984±7482
TSS	$mg \cdot L^{-1}$	462±188	418±293	332±145	354±116	430±120
VSS	%	64.6±5.9	82.0±11.5	63.1±14.6	75.5±9.6	80.1±13.0
COD	$mg \cdot L^{-1}$	1285±429	1403±532	896±201	755±224	1038±238
BOD_5	$mg \cdot L^{-1}$	772±277	911±288	569±122	451±154	581±180
OLR	$kg\ COD \cdot m^{-3} \cdot d^{-1}$	0.75±0.25	1.28±0.38	0.82±0.19	0.71±0.23	0.60±0.20
VFA	$mg\ COD \cdot L^{-1}$	149±146	240±132	120±86	33±60	60±35
Alk	$mg\ CaCO_3 \cdot L^{-1}$	625±79	660±126	663±171	734±175	784±129
N_T	$mg\ N \cdot L^{-1}$	52.0±13.3	54.7±16.3	46.8±6.9	46.0±11.2	35.6±8.0
P_T	$mg\ P \cdot L^{-1}$	10.1±2.8	8.6±2.1	8.5±1.2	6.8±1.3	7.6±1.1
PO_4^{3-}-P	$mg\ P \cdot L^{-1}$	2.5±0.0	5.8±2.1	6.0±0.9	5.3±1.3	3.9±1.7
SO_4^{2-}-S	$mg\ S \cdot L^{-1}$	147.7±13.3	172.2±28.5	125.4±47.6	157.3±46.9	149.7±26.9
HS^--S	$mg\ S \cdot L^{-1}$	37.9±26.7	59.6±19.5	47.0±38.7	30.7±49.2	38.3±14.5
COD/ SO_4^{2-}-S	$g\ COD \cdot g^{-1}\ SO_4^{2-}\text{-}S$	8.70±3.69	8.15±4.44	7.14±4.31	4.80±2.86	6.94±2.83

EFFLUENT	Units	$H_{41}T_{27}$	$H_{25}T_{24}$	$H_{26}T_{19}$	$H_{26}T_{27}$	$H_{41}T_{18}$
COD	$mg \cdot L^{-1}$	91±31	116±24	121±15	80±17	79±18
VFA	$mg\ DQO \cdot L^{-1}$	9±14	21±45	41±28	0±0	5±11
Alk	$mg\ CaCO_3 \cdot L^{-1}$	1110±75	1336±170	996±74	1057±140	1028±198
N_T	$mg\ N \cdot L^{-1}$	47.9±6.2	54.0±14.6	47.3±4.3	52.5±10.2	37.0±11.1
P_T	$mg\ P \cdot L^{-1}$	8.8±1.9	9.9±1.9	6.9±2.8	6.3±0.5	7.8±1.2
PO_4^{3-}-P	$mg\ P \cdot L^{-1}$	7.8±2.5	8.9±1.9	6.9±1.0	5.8±0.6	6.3±2.9
HS^--S	$mg\ S \cdot L^{-1}$	174.3±28.7	207.5±73.2	233.1±66.3	183.2±27.0	166.2±27.9
CH_4	$mg \cdot L^{-1}$	2.6±0.5	2.9±0.8	3.4±0.3	2.6±0.5	3.7±0.6

Table 8.1S. Influent and effluent characterization, main operating conditions, and main biological and filtration process performance results from the AnMBR prototype. (continue)

REACTOR	Units	$H_{41}T_{27}$	$H_{25}T_{24}$	$H_{26}T_{19}$	$H_{26}T_{27}$	$H_{41}T_{18}$
SRT	d	70±1	68±2	70±2	71±2	70±0
HRT	h	41±1	25±1	26±2	26±2	41±13
T	ºC	27±1	24±2	19±1	27±1	18±2
MLTSS	$g \cdot L^{-1}$	8.4±0.5	12.6±0.5	11.3±1.0	10.4±0.8	8.3±1.2
MLVSS	%	70.3±5.5	68.1±1.8	72.8±3.8	65.7±2.7	68.9±5.9
N_T	mg $N \cdot L^{-1}$	505.8±139.4	579.0±68.7	509.0±133.6	339.9±63.5	269.5±94.4
P_T	mg $P \cdot L^{-1}$	96.8±12.3	106.6±10.7	89.4±14.9	93.2±6.2	83.7±6.9
pH		7.1±0.1	7.0±0.1	7.0±0.1	7.0±0.1	7.1±0.3
Waste sludge	$L \cdot d^{-1}$	522±5	511±107	521±5	510±18	520±4
COD	$g \cdot L^{-1}$	11.4±0.6	15.8±1.1	14.7±2.0	13.2±0.5	13.0±0.4
Biogas prod.	L STP $\cdot d^{-1}$	5396±4435	5558±3204	1901±597	1359±999	2241±924
$\%CH_4$	%	77±2	77±2	72±2	77±2	76±1
Y^{CH4}	L STP $\cdot kgCOD_{rem}^{-1}$	169.0±95.1	109.6±70.0	75.2±16.9	70.2±36.0	102.3±63.8

MEMBRANES	Units	$H_{41}T_{27}$	$H_{25}T_{24}$	$H_{26}T_{19}$	$H_{26}T_{27}$	$H_{41}T_{18}$
SGD_p	$Nm^3_{biogas} \cdot m^{-3}_{permeate}$	14	13	12	9	6
$J_{20\ gross}$	LMH	21	19	17	15	18
$J_{20\ net}$	LMH	18	16	15	13	15
TMP	mbar	318	362	462	87	360
FR	mbar $\cdot day^{-1}$	4.2	4.7	0.3	0.5	4.9
Cleaning frequency	$year^{-1}$	2.8	3.1	0.2	0.3	3.2

8.5.3. Energy balance

Table 8.2S. Power energy requirements for specific equipment, total energy recovery (TER), total energy consumption (TEC), and net energy demand (NED) for each OP considering dissolved methane recovery of 47-55 % (economic optimum).

	Energy (kWh·kgCOD$_{rem}^{-1}$)												
OP	**Rotofilter**	**Feeding pumping**	**Permeate pumping**	**Membrane scouring**	**Sludge wasting**	**Sludge recycling**	**Sludge dewatering**	**Reactor mixing**	**Methane recovery blower**	**TER (biogas+methane captured)**	**TER (only biogas)**	**TEC**	**NED**
$H_{41}T_{27}$	0.001	0.002	0.010	0.089	≈0.000	0.006	0.016	0.099	0.001	-0.870	-0.827	0.224	-0.646
$H_{25}T_{24}$	0.001	0.003	0.010	0.080	≈0.000	0.006	0.009	0.056	0.001	-0.553	-0.513	0.166	-0.387
$H_{26}T_{19}$	0.002	0.005	0.022	0.120	≈0.000	0.010	0.016	0.097	0.001	-0.503	-0.429	0.272	-0.231
$H_{26}T_{27}$	0.002	0.006	0.005	0.107	≈0.000	0.011	0.018	0.112	0.001	-0.472	-0.386	0.262	-0.210
$H_{41}T_{18}$	0.001	0.003	0.014	0.048	≈0.000	0.008	0.019	0.124	0.001	-0.643	-0.587	0.218	-0.425

Table 8.3S. Power energy requirements for specific equipment, total energy recovery (TER), total energy consumption (TEC), and net energy demand (NED) for each OP considering dissolved methane recovery of 80-84 % (environmental optimum).

	Energy (kWh·kg COD_{rem}^{-1})												
OP	**Rotofilter**	**Feeding pumping**	**Permeate pumping**	**Membrane scouring**	**Sludge wasting**	**Sludge recycling**	**Sludge dewatering**	**Reactor mixing**	**Methane recovery blower**	**TER (biogas+methane captured)**	**TER (only biogas)**	**TEC**	**NED**
$H_{41}T_{27}$	0.001	0.002	0.010	0.089	≈0.000	0.006	0.016	0.099	0.003	-0.894	-0.827	0.227	-0.668
$H_{25}T_{24}$	0.001	0.003	0.010	0.080	≈0.000	0.006	0.009	0.056	0.002	-0.577	-0.513	0.168	-0.409
$H_{26}T_{19}$	0.002	0.005	0.022	0.120	≈0.000	0.010	0.016	0.097	0.005	-0.554	-0.429	0.277	-0.277
$H_{26}T_{27}$	0.002	0.006	0.005	0.107	≈0.000	0.011	0.018	0.112	0.005	-0.520	-0.386	0.266	-0.254
$H_{41}T_{18}$	0.001	0.003	0.014	0.048	≈0.000	0.008	0.019	0.124	0.004	-0.689	-0.587	0.221	-0.468

8.5.4. Sensitivity and uncertainty analyses

Table 8.4S. Mean value, lower bound (LB) and upper bound (UB) of input factors for sensitivity and uncertainty analyses.

Inputs				
Parameter	**Unit**	**Mean**	**LB**	**UB**
SRT	d	70	63	77
HRT	h	41	36.9	45.1
Reactor height	m	5	4.5	5.5
Membrane tank height	m	2.80	2.52	3.08
Steel cover MT	m	0.01	0.009	0.011
Concrete wall MT	m	0.150	0.135	0.165
Concrete cover reactor	m	0.30	0.27	0.33
Concrete wall reactor	m	0.30	0.27	0.33
Concrete slab reactor	m	0.50	0.45	0.55
Facility lifespan	y	20	18	22
Membrane lifespan	y	10	9	11
Equipment lifespan	y	10	9	11
Civil work lifespan	y	20	18	22
Diffuser lifespan	y	10	9	11
Disolved methane membranes lifespan	y	10	9	11
NaOCl	ppm	2000	1800	2200
Citric acid	ppm	2000	1800	2200
Filtration time (i.e., non-downtime period)		0.850	0.765	0.935
Biogas for reactor mixing	$Nm^3 \cdot m_{reactor}^{-3} \cdot h^{-1}$	0.250	0.225	0.275
Dissolve methane recovery eficiency	%	84.0	75.6	92.4
CHP Power eficiency	%	35.0	31.5	38.5
SGD_p	$Nm^3 \cdot m^{-3}$	14.02	12.62	15.42
Transmembrane flux ($J_{20\,p}$)	LMH	21.0	18.9	23.1
Emissions of Ammonium to soil	kg $N\text{-}NH_4 \cdot kg^{-1}$ N in sludge	0.10	0.09	0.11
Emissions of N_2O to air	kg $N\text{-}N_2O \cdot kg^{-1}$ N in sludge	0.0118	0.0106	0.0130
Emissions of Phosphate to soil	kg $P\text{-}PO_4 \cdot kg_{-1}$ P in sludge	0.0258	0.0232	0.0283
Transport (O&M)	$tkm \cdot m^{-3}$	20	18	22
Transport (Construction)	$tkm \cdot m^{-3}$	20	18	22
Transport (Dismantling)	$tkm \cdot m^{-3}$	30	27	33

8.5.5. Heavy metals content in sludge

Table 8.5S. Heavy metal content in sewage sludge (P. S. Kidd et al., 2007)

	Total metal concentration in sewage sludge ($mg \cdot kg^{-1}$)
Mn	330
Cd	5
Co	14
Cr	115
Cu	230
Ni	35
Pb	69
Zn	500

8.5.6. Allocation of environmental impacts at mid-point level for each impact category.

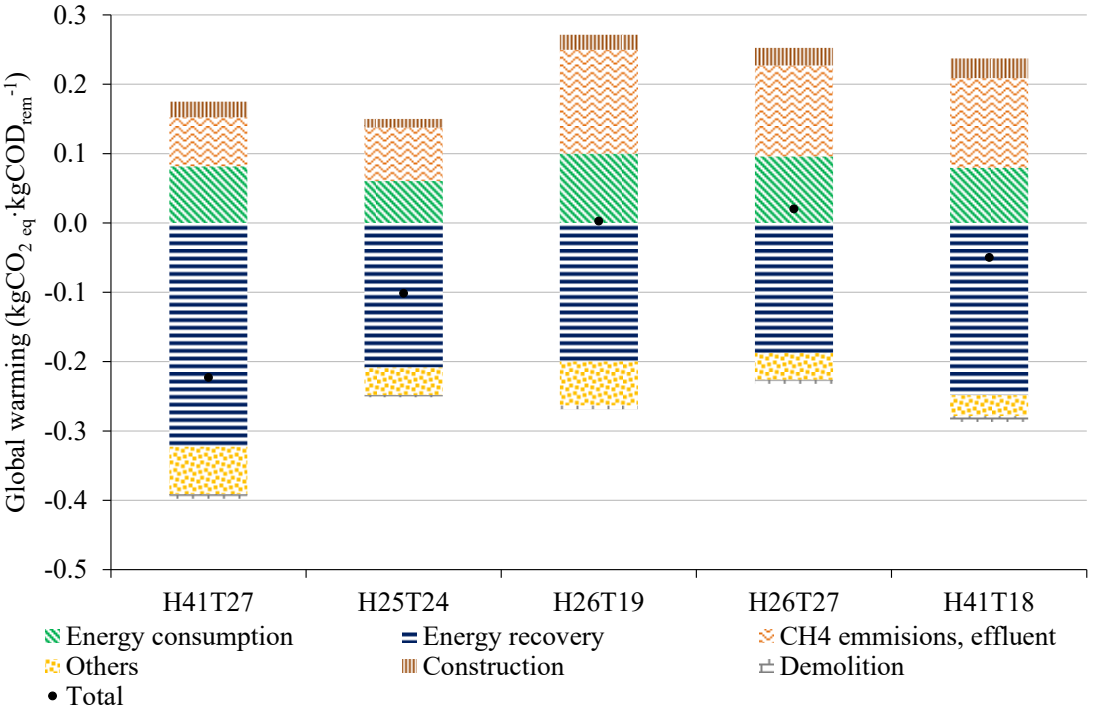


Figure 8.2S. Global warming potential

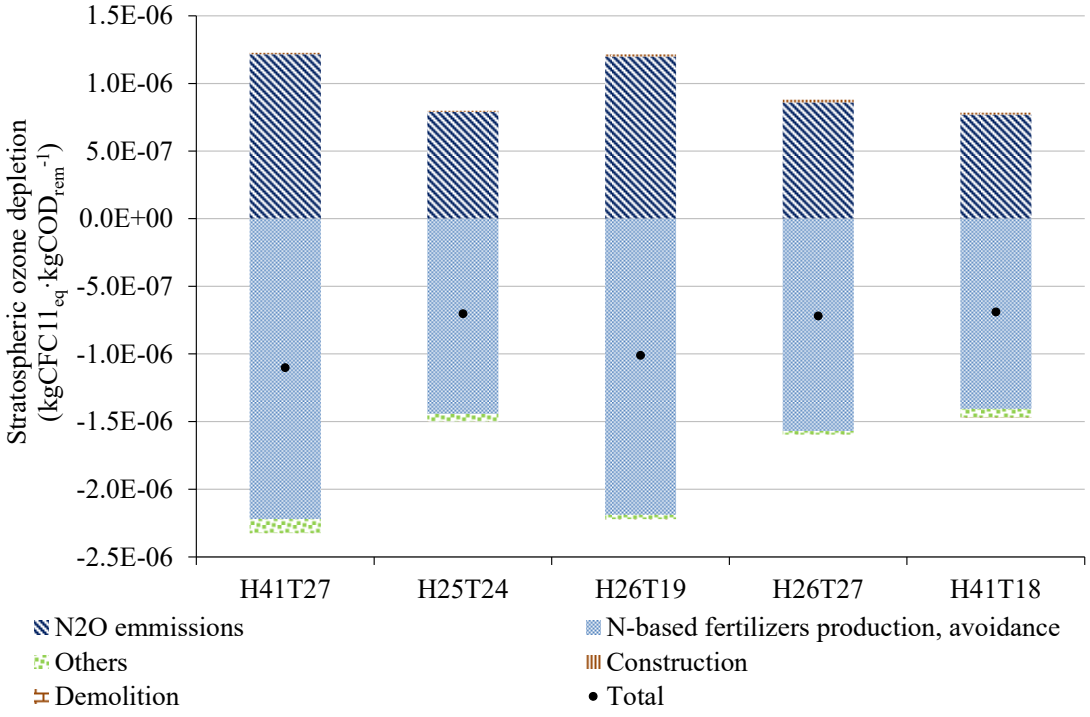


Figure 8.3S. Stratospheric ozone depletion.

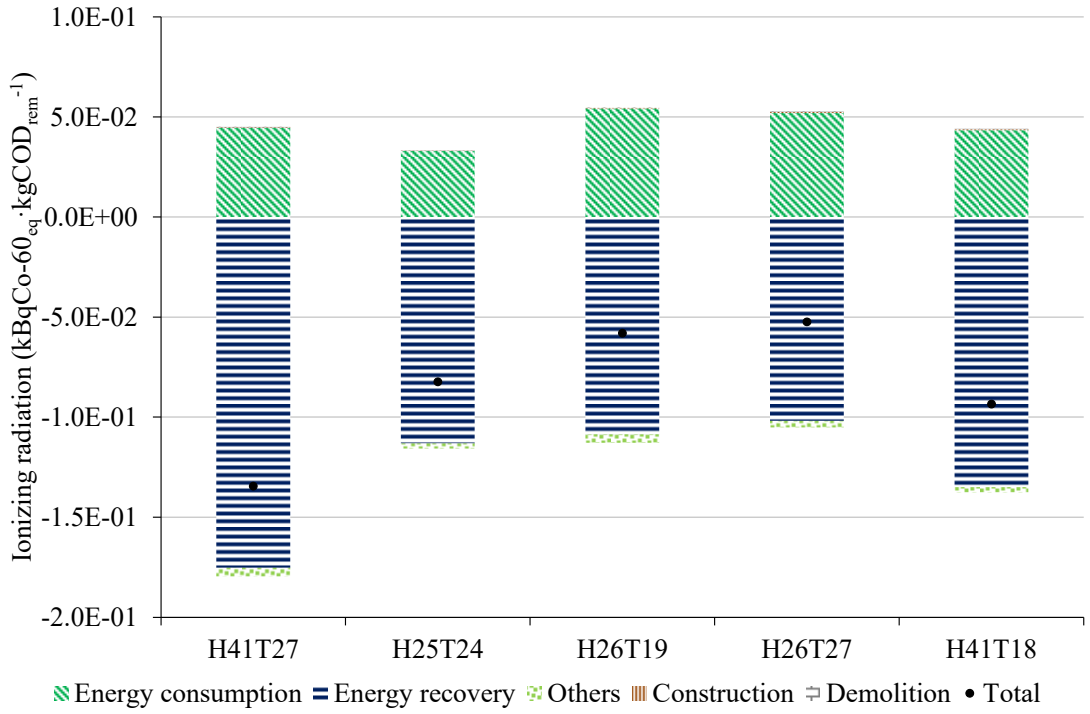


Figure 8.4S. Ionizing radiation.

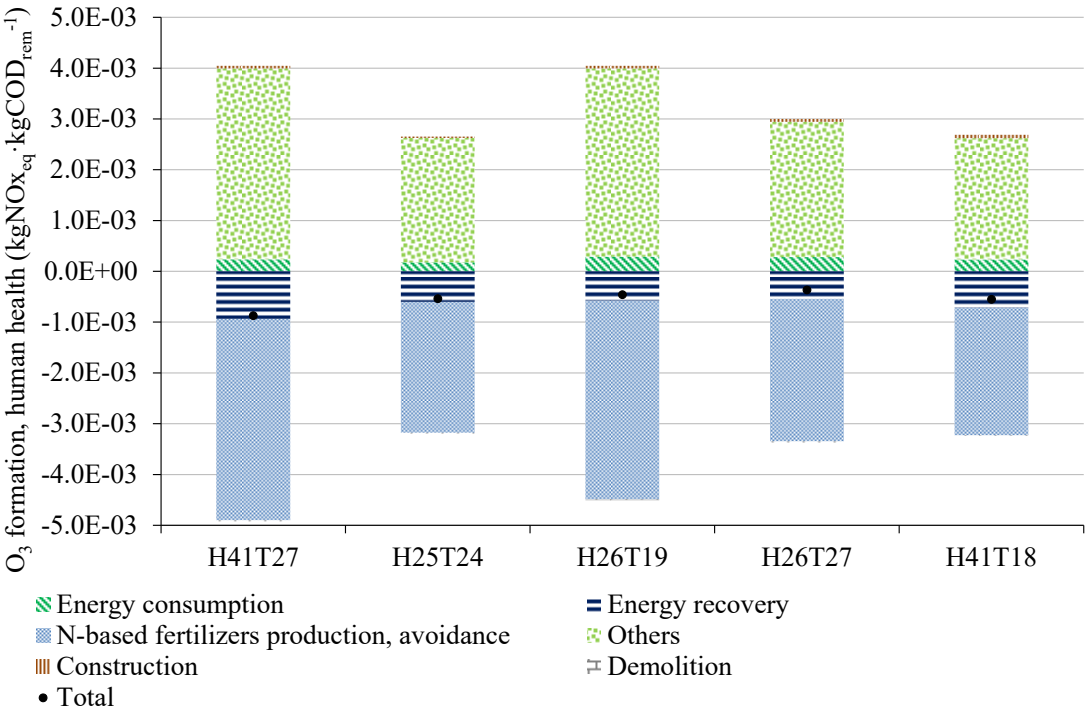


Figure 8.5S. Ozone formation, human health.

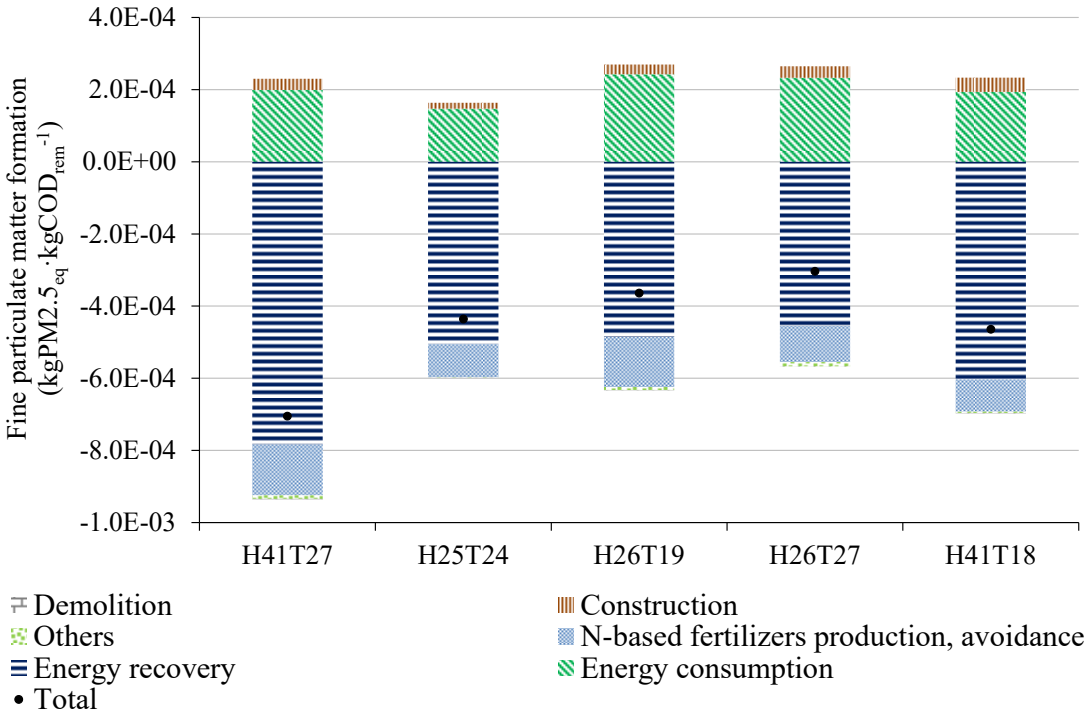


Figure 8.6S. Fine particulate matter formation.

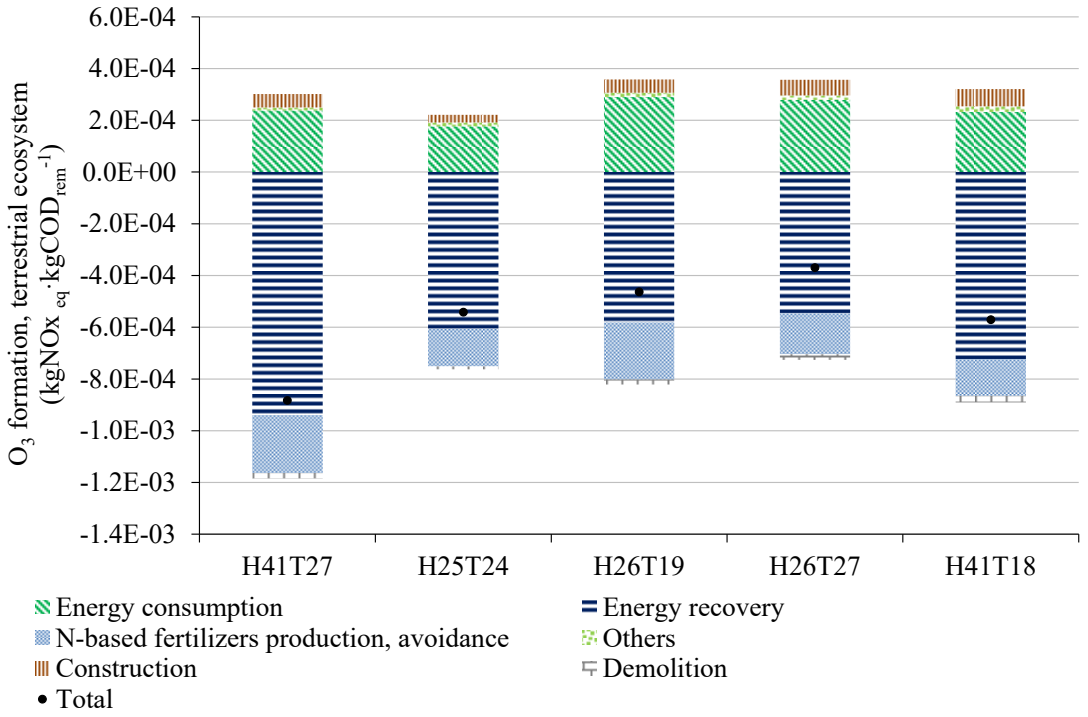


Figure 8.7S. Ozone formation, terrestrial ecosystem.

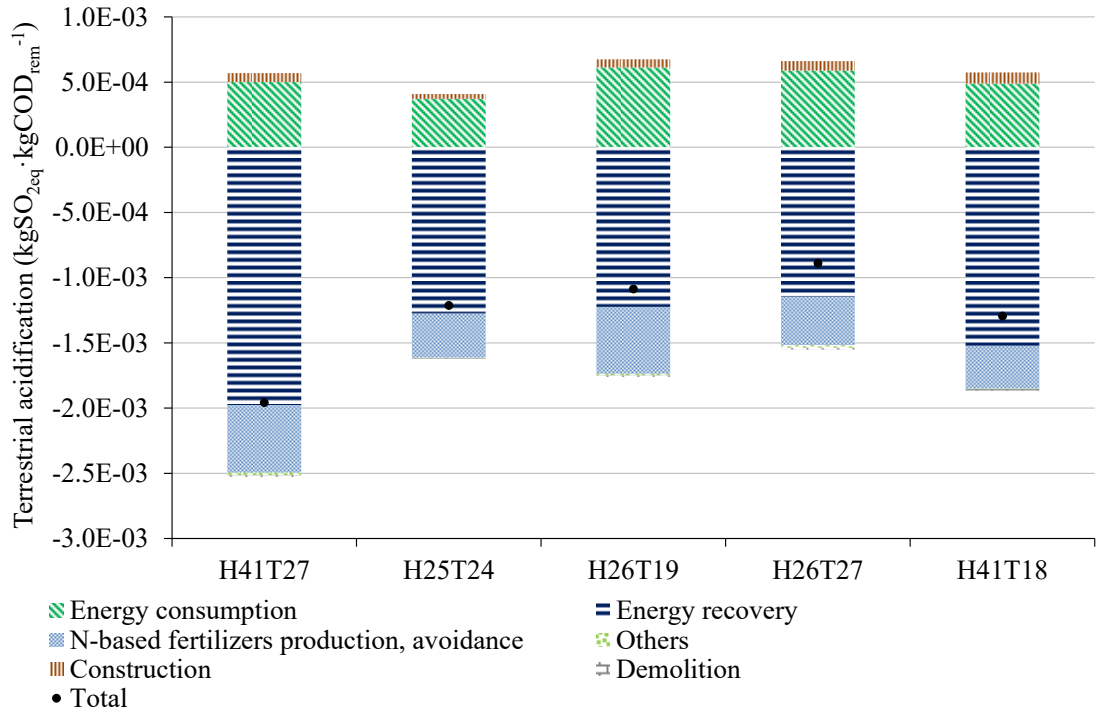


Figure 8.8S. Terrestrial acidification.

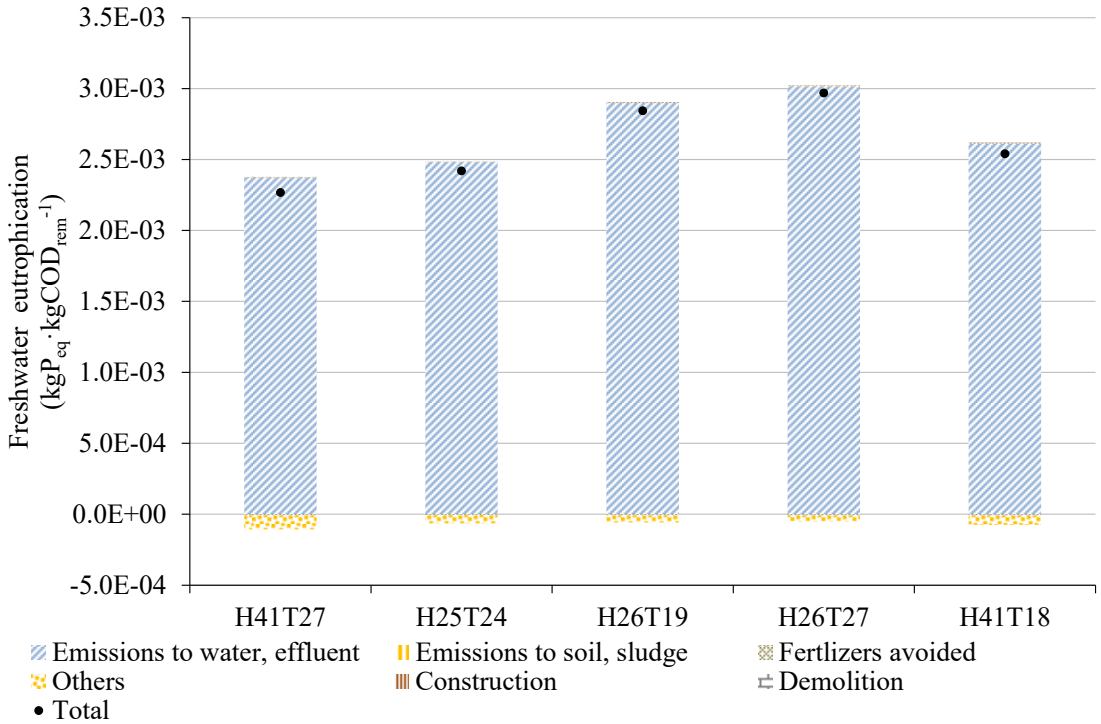


Figure 8.9S. Freshwater eutrophication.

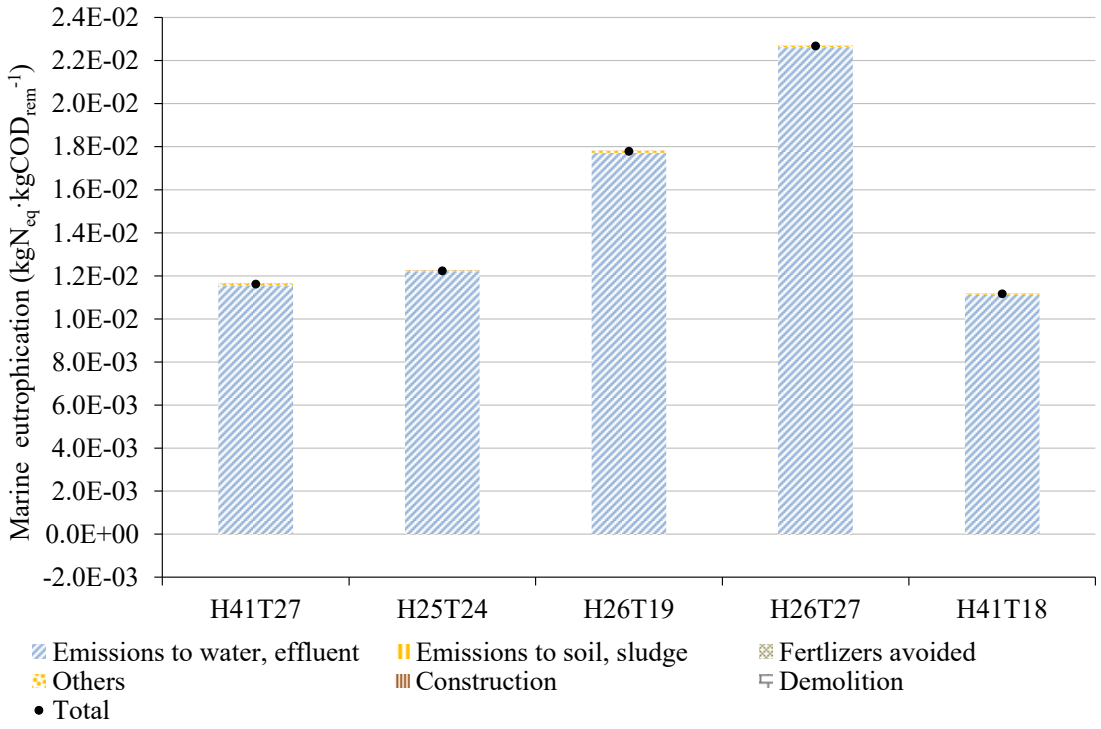


Figure 8.10S. Marine eutrophication.

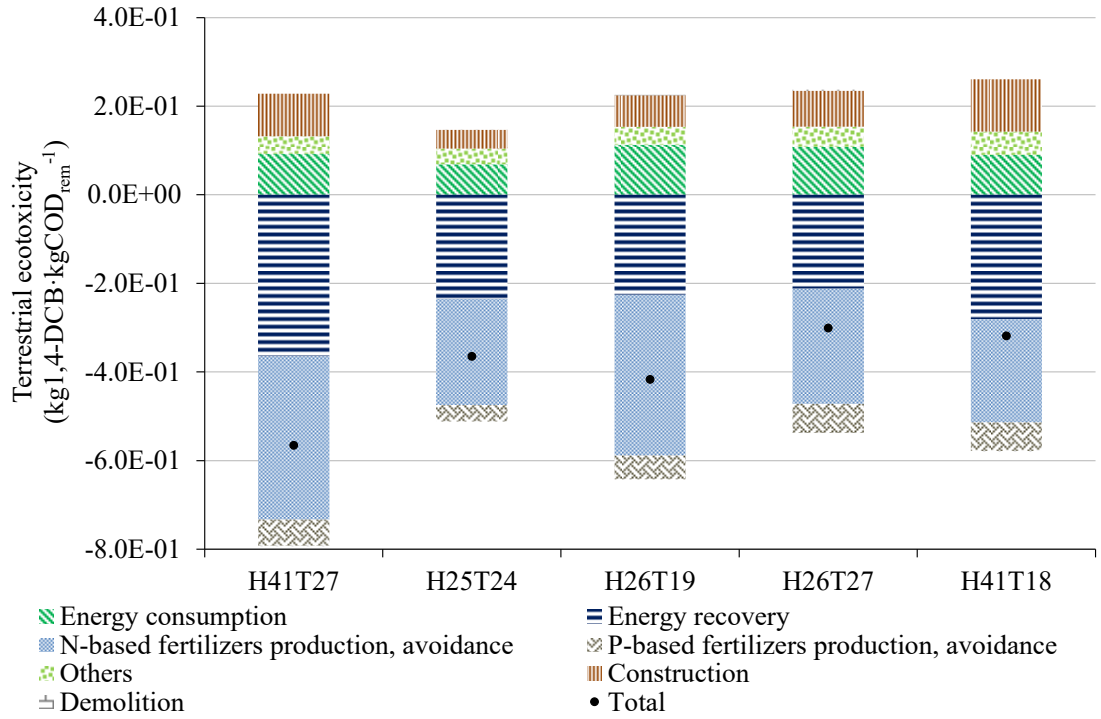


Figure 8.11S. Terrestrial ecotoxicity.

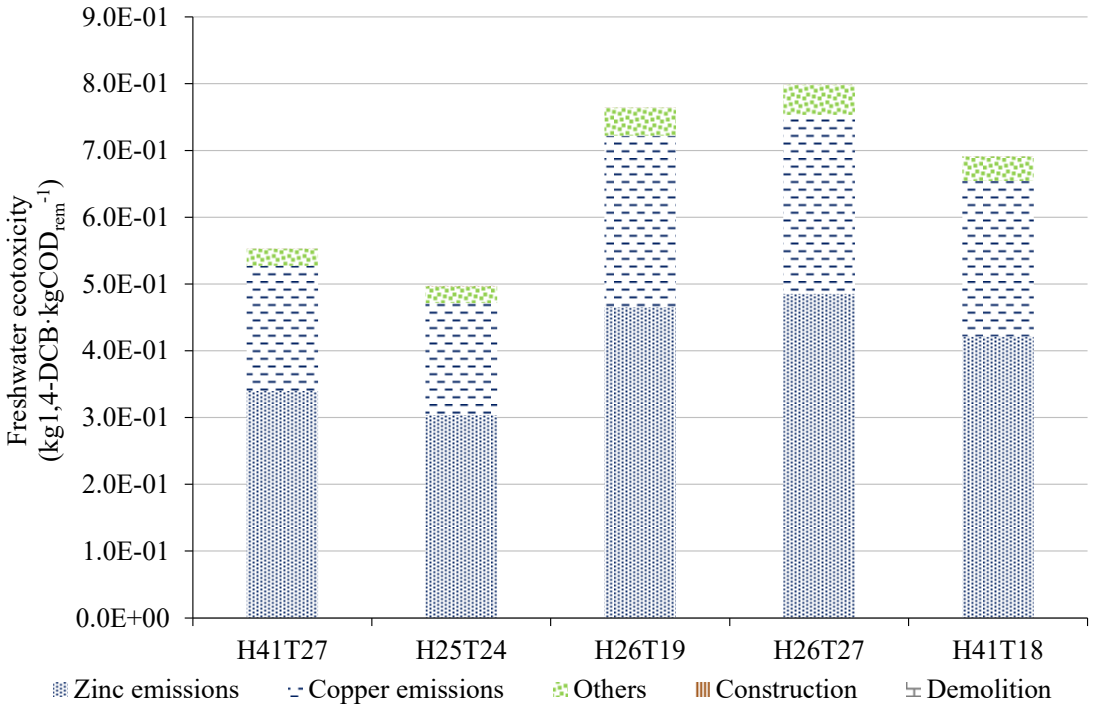


Figure 8.12S. Freshwater ecotoxicity.

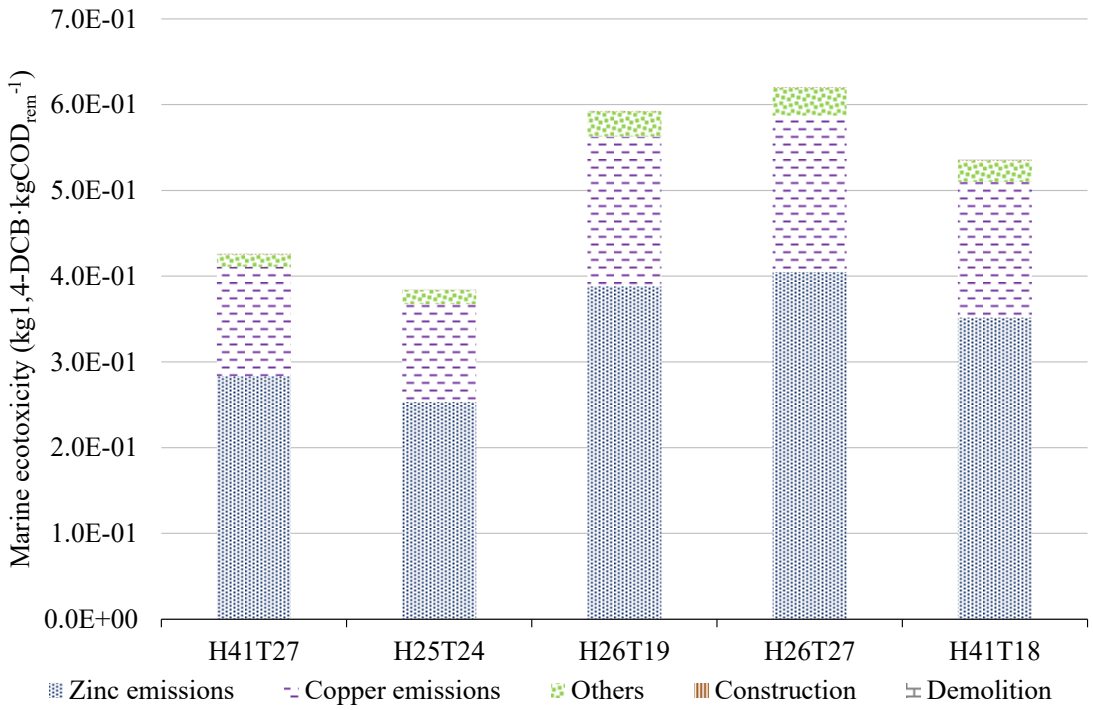


Figure 8.13S. Marine ecotoxicity.

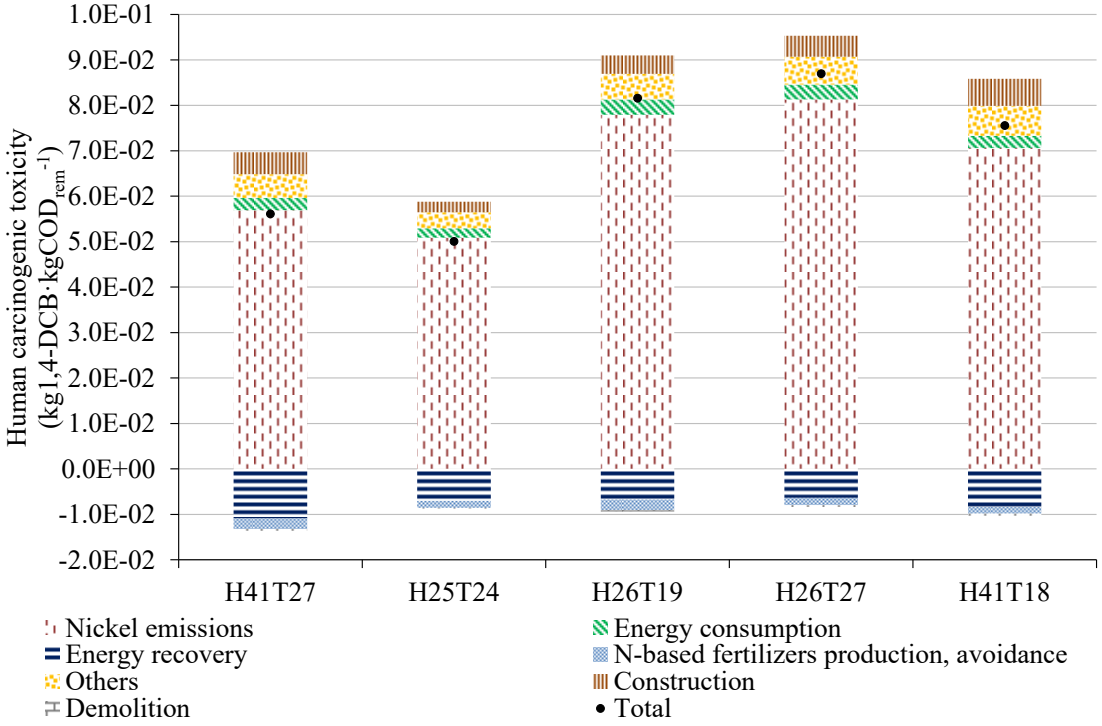


Figure 8.14S. Human carcinogenic toxicity.

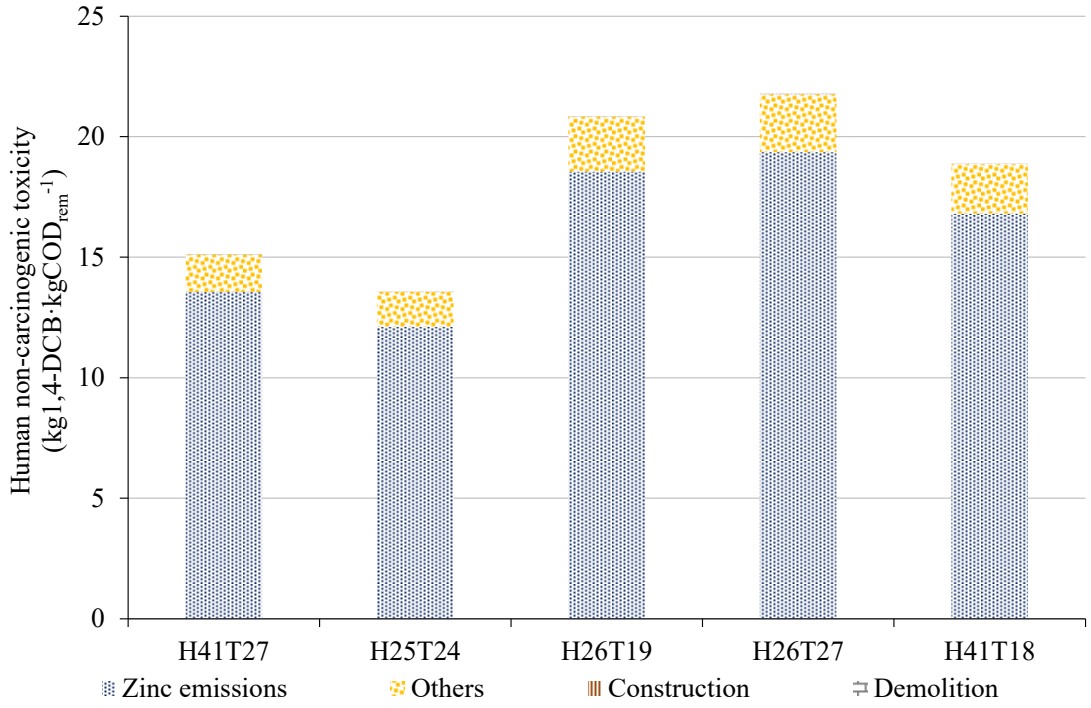


Figure 8.15S. Human non-carcinogenic toxicity.

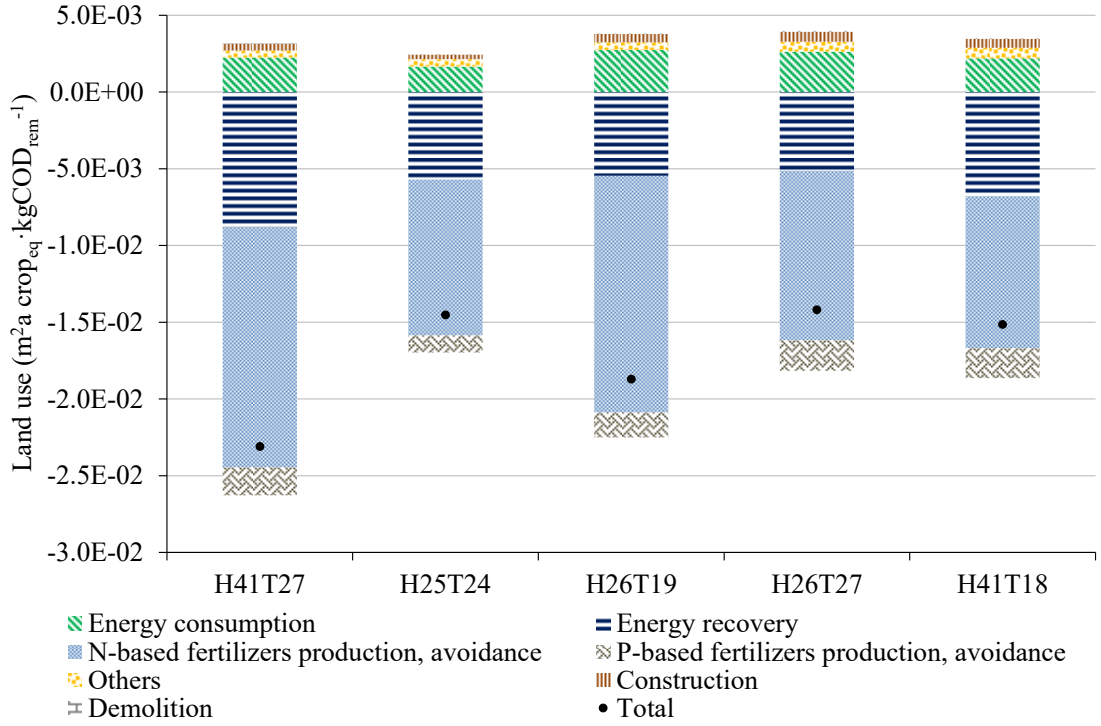


Figure 8.16S. Land use.

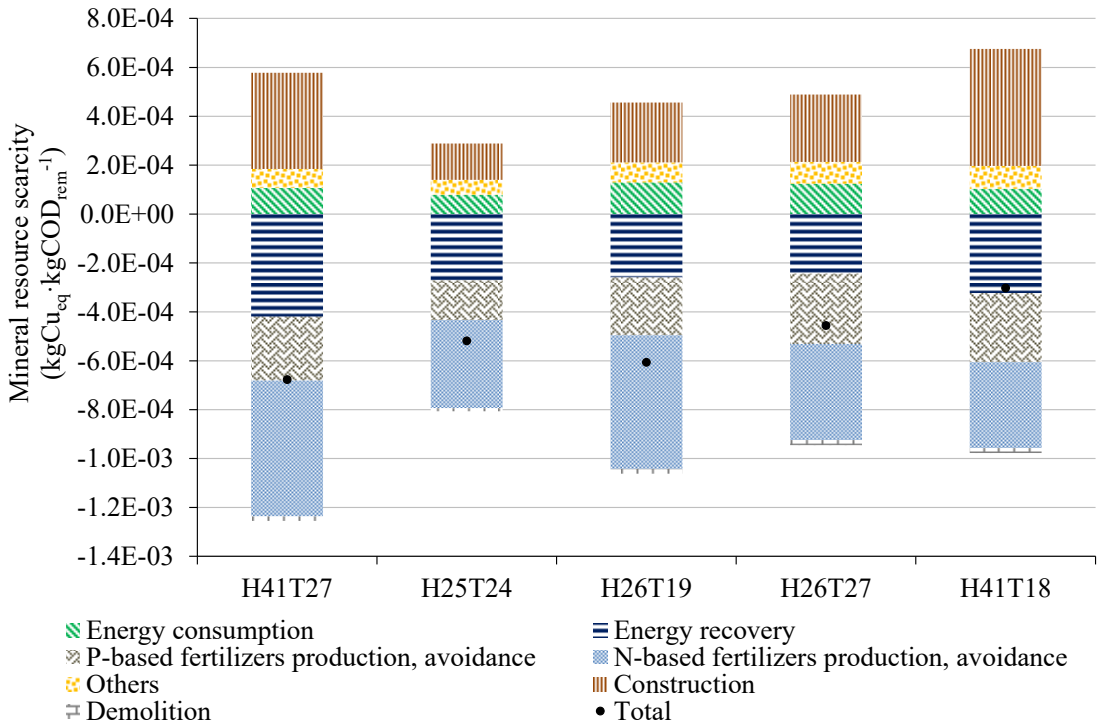


Figure 8.17S. Mineral resource scarcity.

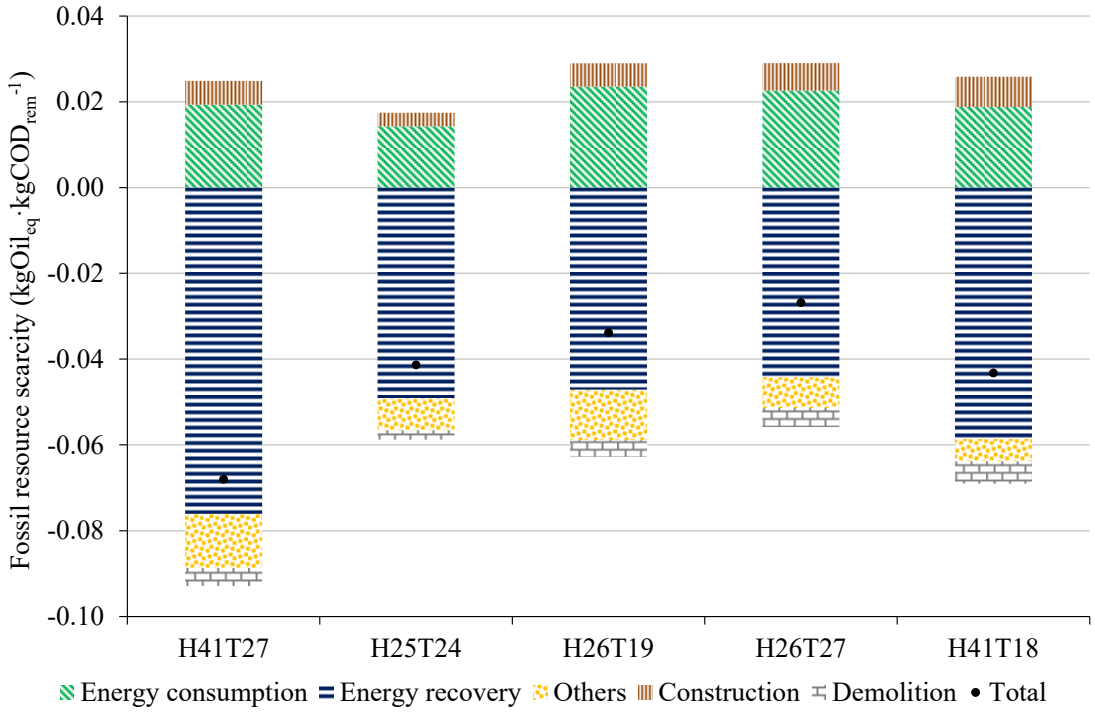


Figure 8.18S. Fossil resource scarcity.

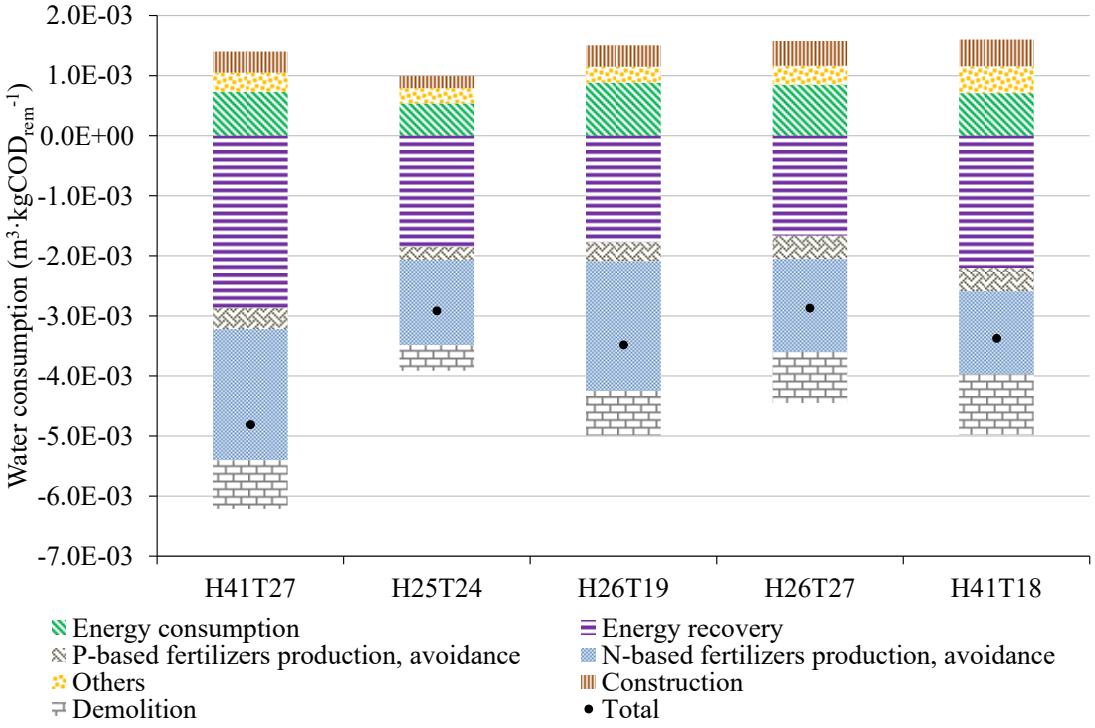


Figure 8.19S. Water consumption.

8.5.7. LCA at mid-point level for the evaluated OPs.

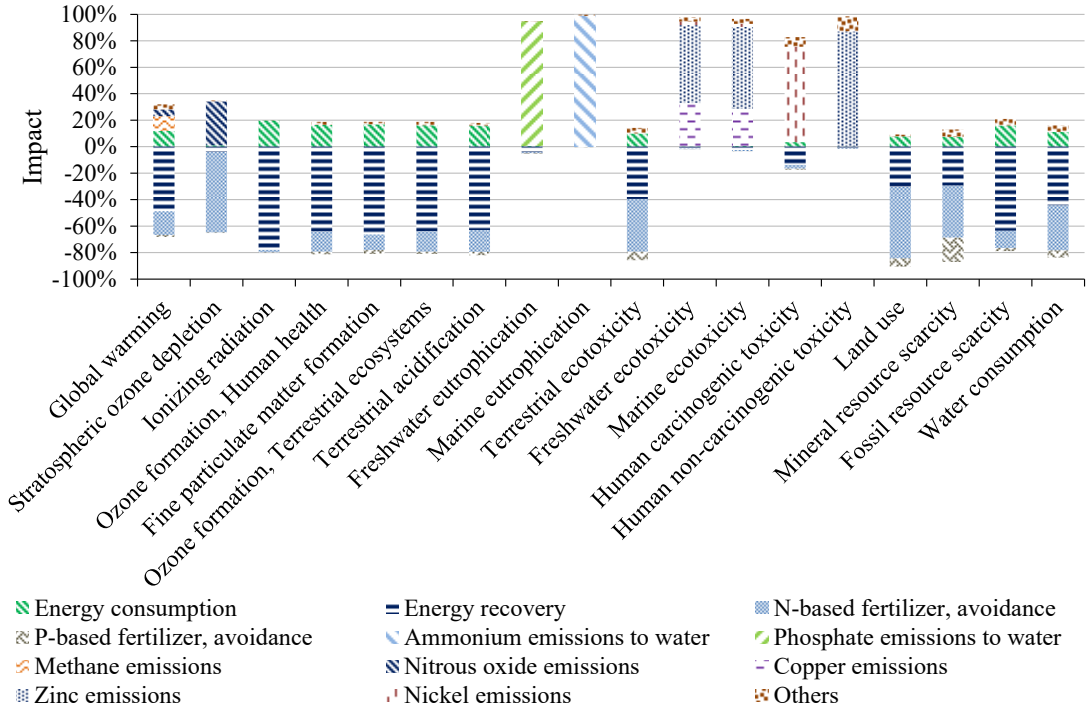


Figure 8.20S. Global LCA at mid-point level for $H_{41}T_{27}$.

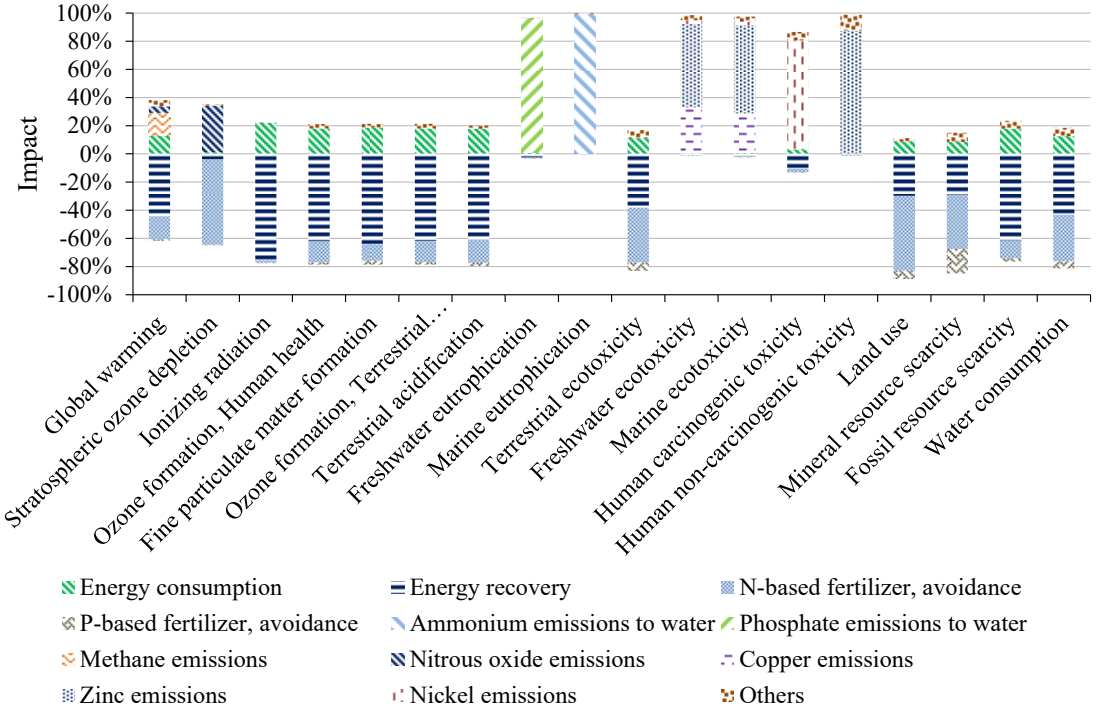


Figure 8.21S. Global LCA at mid-point level for $H_{25}T_{24}$.

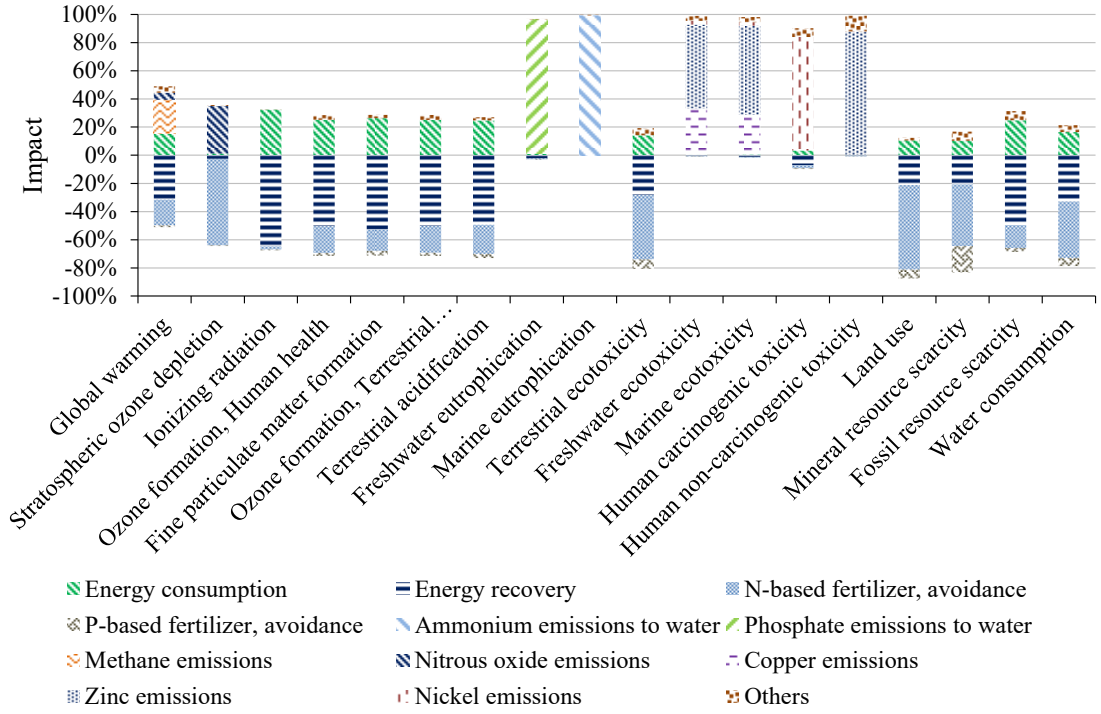


Figure 8.22S. Global LCA at mid-point level for $H_{26}T_{19}$.

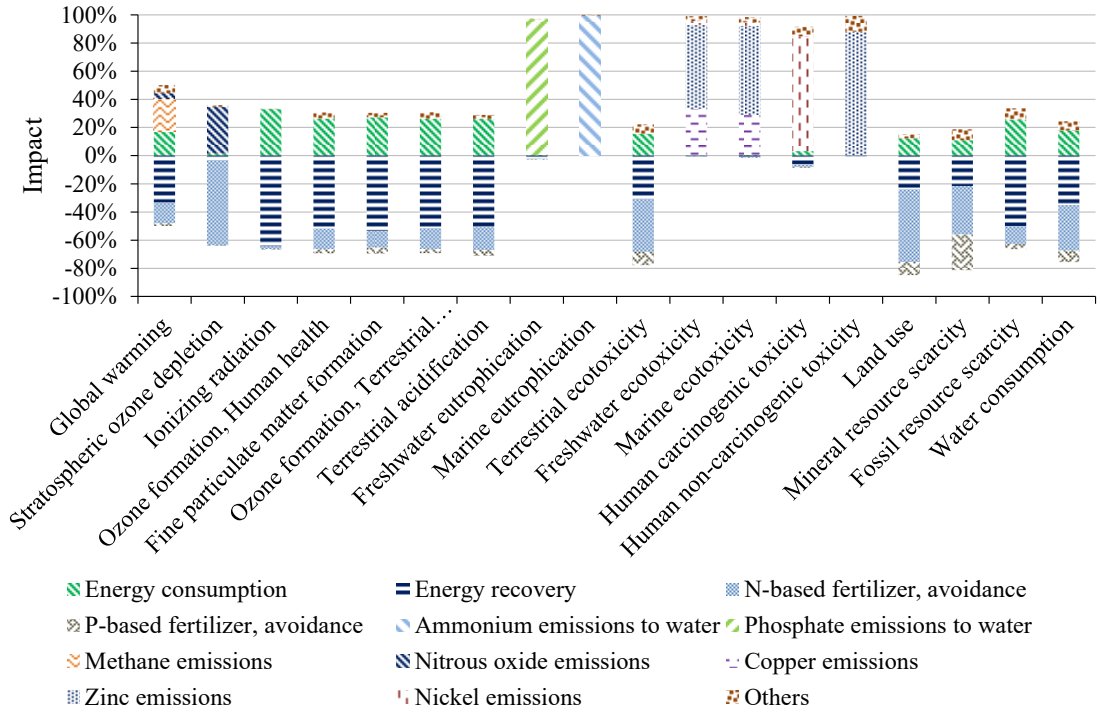


Figure 8.23S. Global LCA at mid-point level for $H_{26}T_{27}$.

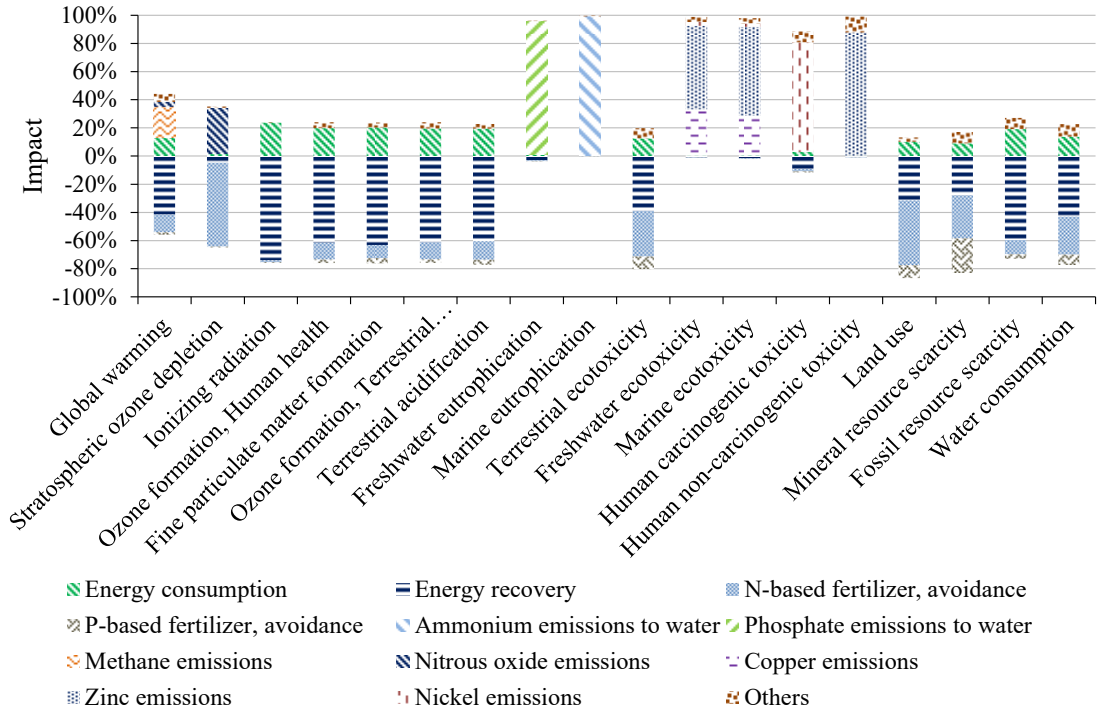


Figure 8.24S. Global LCA at mid-point level for $H_{41}T_{18}$.

8.5.8. LCA at end-point level for the evaluated OPs.

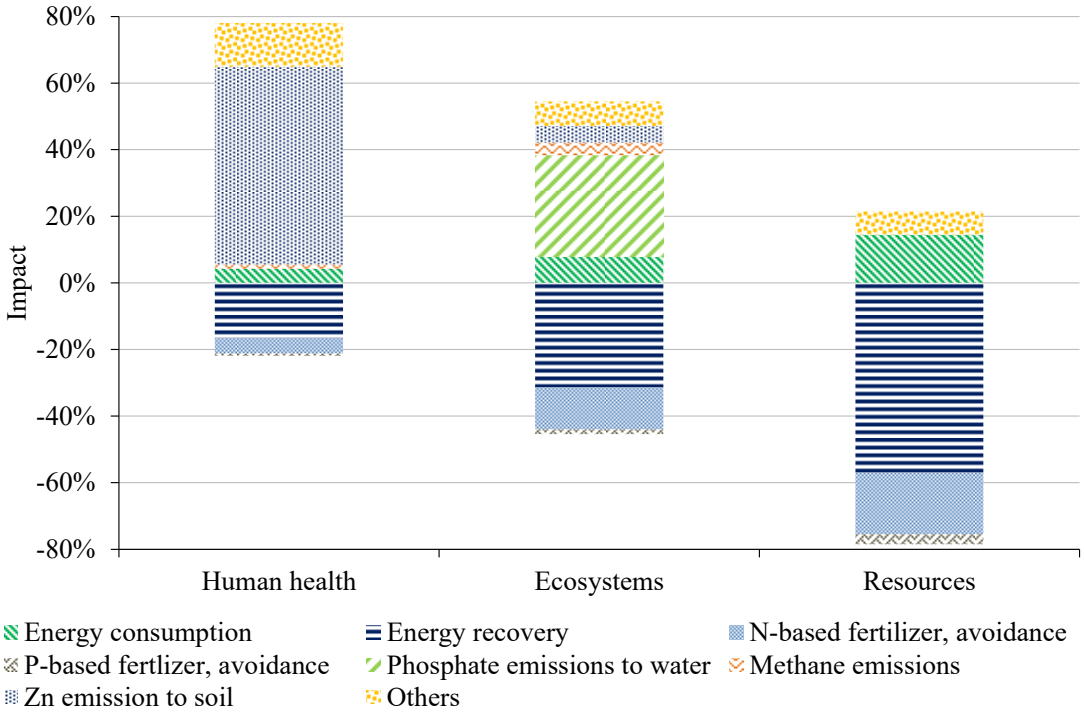


Figure 8.25S. Global LCA at end-point level for $H_{41}T_{27}$.

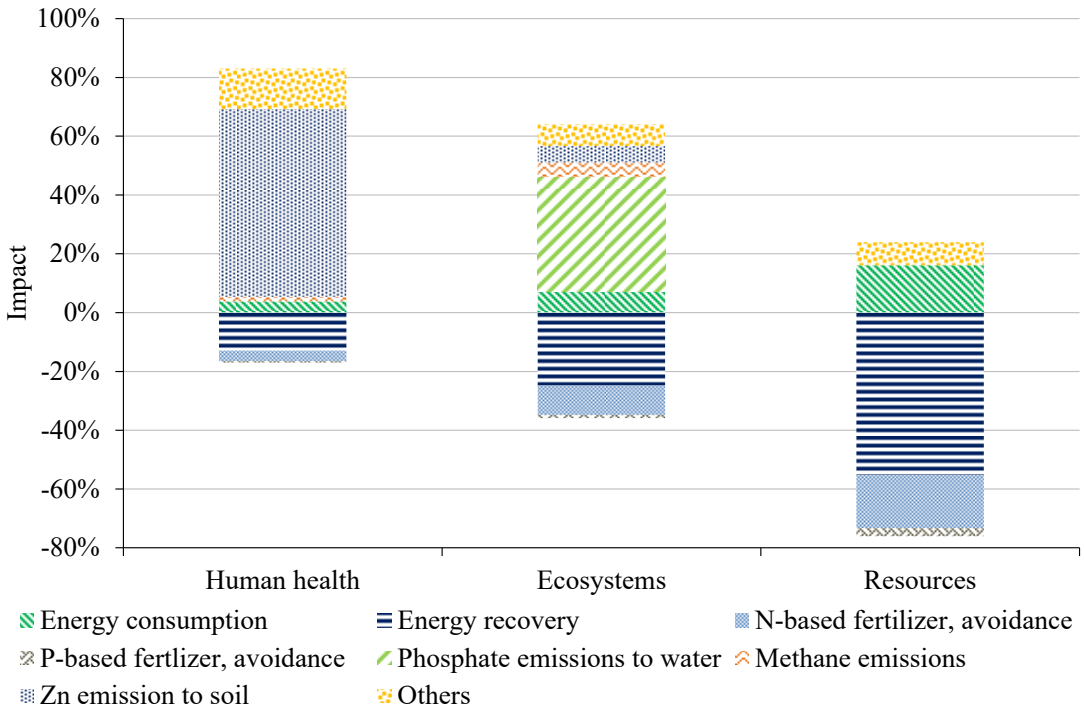


Figure 8.26S. Global LCA at end-point level for $H_{25}T_{24}$.

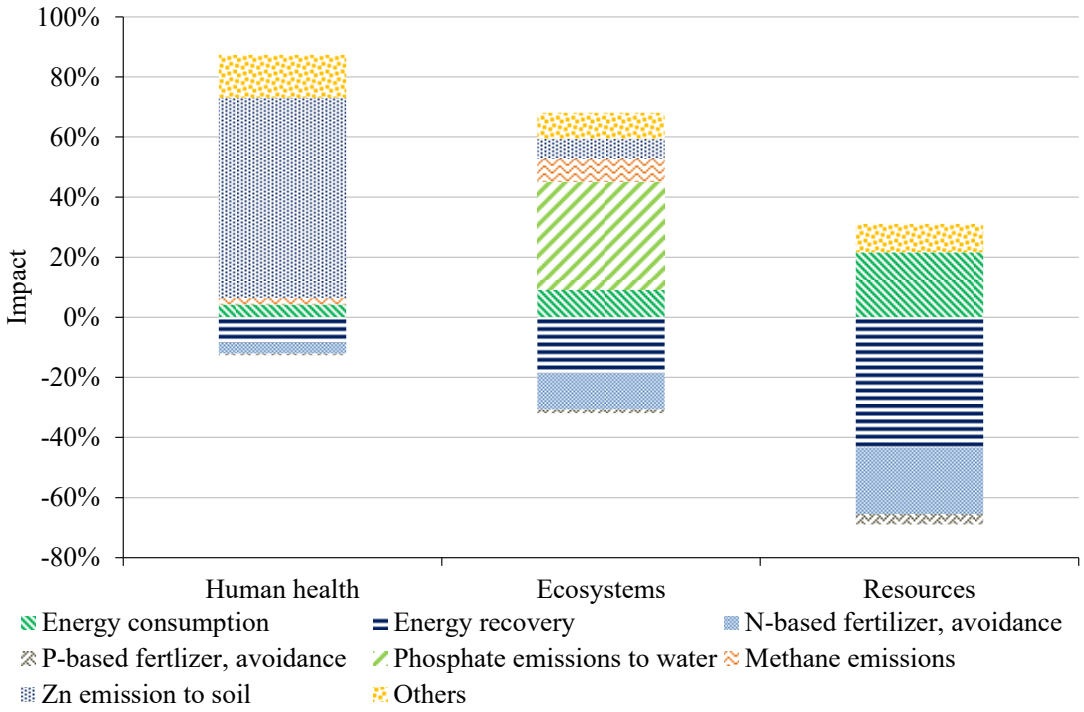


Figure 8.27S. Global LCA at end-point level for $H_{26}T_{19}$.

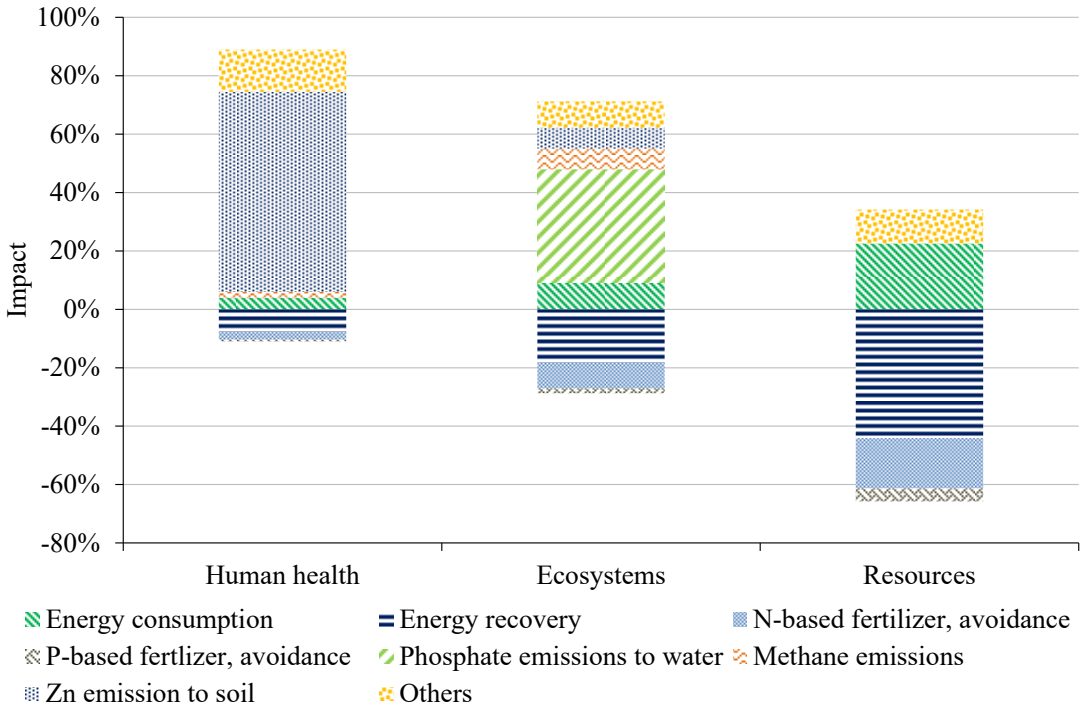


Figure 8.28S. Global LCA at end-point level for $H_{26}T_{27}$.

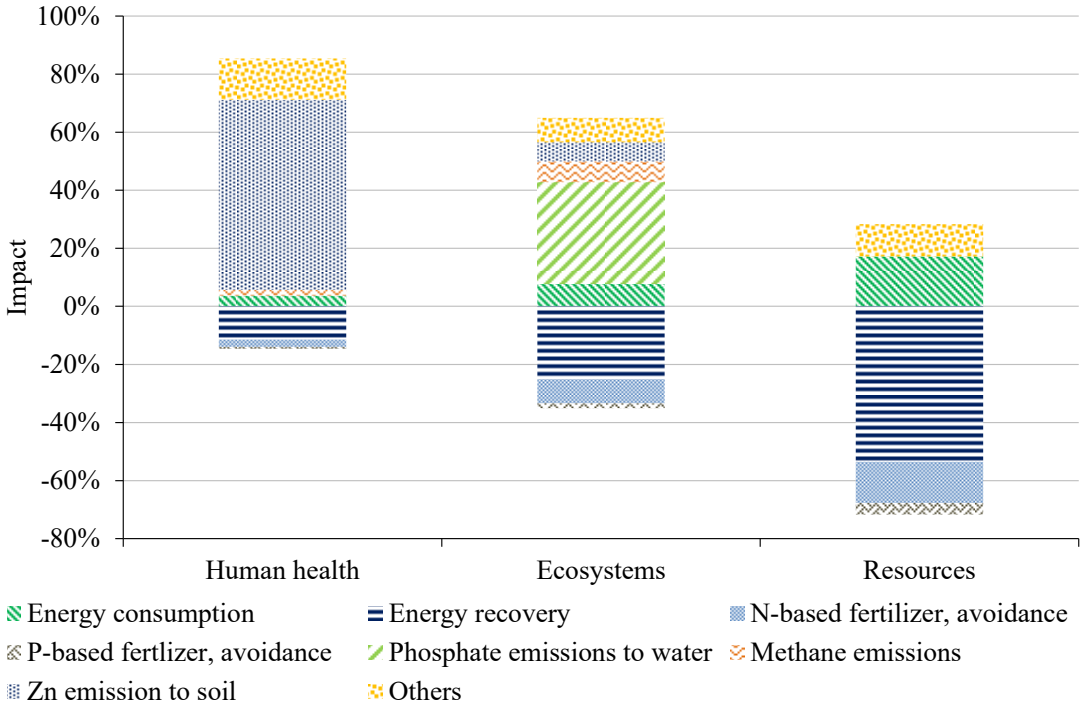


Figure 8.29S. Global LCA at end-point level for $H_{41}T_{18}$.

References

Kidd, P.S., Domínguez-Rodríguez, M.J., Díez, J., Monterroso, C., 2007. Bioavailability and plant accumulation of heavy metals and phosphorus in agricultural soils amended by long-term application of sewage sludge. Chemosphere. https://doi.org/10.1016/j.chemosphere.2006.09.007

9. Effect of seasonal dynamics on water reclamation from ultrafiltration membrane-based tertiary urban wastewater treatment: energy expenditure, cost, environmental and effluent quality assessment

Abstract

This study aims at assessing the performance of an ultrafiltration (UF) tertiary treatment for the effluent of an urban wastewater treatment facility. The assessment is based on the energy balance, the economic and environmental performance and the permeate quality. To this aim, data from an UF demonstration plant made up of commercially equipment, including industrial hollow fiber membranes, was used to project a full-scale facility. Results of the demonstration plant recommended different transmembrane fluxes and sparging air demands under summer and winter conditions, to prevent an excessive fouling propensity. The energy balance of the full-scale facility would be 0.239±0.083 kWh·m^{-3} in summer and 0.151±0.044 kWh·m^{-3} in winter, with blowers' consumption as the main contributor (87-92 %). No difference in CAPEX was obtained between summer and winter scenarios (€0.025±0.001·m^{-3} and €0.026·m^{-3}, respectively) and membrane acquisition accounted for 62-67 % of the investment cost. Energy expenditure represents the major cost of OPEX (67-76 %), with the total operating cost being €0.062±0.017·m^{-3} and €0.044±0.009·m^{-3} in summer and winter, respectively. Environmental assessment confirmed energy consumption as the main factor influencing the environmental sustainability and the need for filtration optimization. Finally, the permeate quality accomplished with the more stringent parameters for water reuse in agriculture according to Spanish and EU regulation.

Keywords

Ultrafiltration; tertiary treatment; energy balance; life cycle costing; life cycle assessment; water reuse

Highlights

Full-scale UF plant shows energy consumption in the 0.151-0.239 kWh·m^{-3} range.
CAPEX was estimated at €0.025-0.026·m^{-3} with membranes as major contributor (62-67%).
OPEX account for €0.044-0.062·m^{-3} being energy cost the main expenditure (67-76%).
LCA showed energy balance as most influential factor on the environmental impact.
UF process allowed accomplishing regulation statements for agriculture reuse.

A. Jiménez-Benítez, J. González-Camejo, N. Martí1, A. Robles, A. Seco (2023) Effect of seasonal dynamics on water reclamation from ultrafiltration membrane-based tertiary urban wastewater treatment: energy expenditure, cost, environmental and effluent quality assessment. *Submitted to Journal of Environmental Chemical Engineering on Nov 06, 2023.*

9.1. Introduction

Providing access to an adequate supply of clean water is a fundamental objective that involves serious challenges. To the growing pressure on the conventional water resources (Châtel, 2018), driven by an ever-increasing world population (Mekonnen and Hoekstra, 2016), we must add the effects of global warming, which will affect precipitation patterns, cause more droughts and modify the reserves of this basic resource (Samantaray et al., 2022). The consequences of these effects are already being seen worldwide (Rodell et al., 2018), e.g., according to the Water Exploitation Index (calculated as total water abstraction per year compared to long-term renewable resources) published by the European Environment Agency, Belgium, Bulgaria, Cyprus, Germany, Italy, Malta, Spain already exceed this indicator by 20%, implying that they are in danger of facing water shortages (European Environment Agency, 2023).

In addition to saving and promoting efficient use of water, e.g. actions included in the European Circular Economy Action Plan (European Commission, 2020c), alternative water sources must be explored to guarantee access to water within the context of economic, environmental and social sustainability (Aldaya et al., 2019). Increasing use is being made of reclaimed water, especially in arid and semi-arid regions, and has become a vector of change and innovation in the water sector. Moreover, wastewater treatment plants (WWTP) are being converted to water resource recovery facilities (WRRF), as wastewater can be considered a usable reserve of resources: e.g., for energy, nutrients and reclaimed water (Qadir et al., 2020).

Water and its nutrient content have been reused since ancient times; the earliest infrastructures for distributing wastewater for agriculture date back to the Bronze Age in the Minoan and Indus Valley civilization 5000 years ago (Angelakis et al., 2018). While this practice has continued throughout history, the increasing population and migration to the cities made it necessary to prioritize sanitary protection and safe discharge into water bodies as the objective of wastewater management. The discovery of the Haber-Bosh process to fix atmospheric nitrogen and produce industrial fertilizers also reduced interest in using the nutrients present in wastewater. However, the situation has now changed and interest in reclaimed water has grown while the population increases, resources are depleted and global warming advances. For example, in the EU more than 40000 hm^3 of wastewater is produced every year, while less than 1000 million m^3 is reused, so that there is clearly a wide margin for improvement and using this resource would also contribute to preserving conventional sources.

Despite the advantages of using the resources embedded in wastewater (e.g., energy, nutrients and water), there are legal, economic, environmental and social conditions that hinder the spread of this practice (Radini et al., 2023). In the legal field for instance, European Directive 91/271/EEC on the treatment of urban wastewater recommends the reuse of water when appropriate. However, its definition of "discharge" and the permitted maximum nutrient concentrations in environmentally sensitive areas prohibits, direct reuse for fertigation without nutrient removal. The different regulations, recommendations and guides for the reuse of water in agriculture in the EU countries was also a legal barrier. This situation has been clearly improved by Regulation (EU) 741/2020 on the minimum requirements for water reuse, which will better harmonize this practice within the EU and is expected to promote reuse (Radini et al., 2023).

The cost of additional treatments to achieve the standards of Regulation (EU) 741/2020 is another hindrance, as well as the preparation, implementation and compliance to mandatory risk management plans for any reuse project that requires administrative authorization. Faced with the low cost of irrigation water from conventional sources, without adequate financial mechanisms there is no economic incentive for the farmer to change his water source, except for scarcity or legal obligations (Foglia et al., 2023).

In the environmental field, the challenges generally have to do with the proper control of the quality of effluents, including emerging contaminants such as microplastics, antibiotics-resistant bacteria (ARB) and genes (ARG), the adequate use of the nutrients present in the waters that prevent adding excessive fertilizers to the soil and soil salinization (Rahman et al., 2022). Last but not least, much remains to be done in terms of raising public awareness of the benefits of water reuse and the use of nutrients in agriculture, as well as the acceptance of the agricultural products obtained under these practices (Radini et al., 2023).

From a technological perspective, membrane-based filtration systems for tertiary treatment present robustness and efficiency in terms of removing microbiological elements and solids (Yang et al., 2021b). Fit-for-purpose high-quality reclaimed water can also be produced by selecting the appropriate membrane pore size, which confers versatility on this technology. However, membrane fouling is still a challenge that reduces filtration productivity and therefore also the environmental and economic performance of ultrafiltration (UF) (Gong et al., 2019; Lei et al., 2021; Qasim et al., 2018). Optimization of permeate productivity (e.g., transmembrane fluxes) and fouling mitigation (e.g., backwash, membrane scouring and chemical cleanings) are

imperative enhance the competitiveness of UF processes. Previous studies evaluated tertiary treatments under certain specific conditions, regardless of the possible seasonal variations in the WWTP, especially in tourist areas such as Valencia. To the best of the authors' knowledge, no study has yet been published on a seasonal analysis of not only technical issues but also wastewater characteristics.

In this context, the present study aimed to evaluate the technical and economic feasibility of UF membranes as the tertiary treatment for water reclamation. For this, a UF prototype that treated part of the effluent from a full-scale WWTP in Valencia, Spain, was operated for a year. The data obtained was then used to evaluate the energy balance and economic feasibility of implementing this UF technology in the same WWTP on an industrial scale (treatment flow rate of 14000 $m^3 \cdot d^{-1}$). The effluent quality was also characterized to confirm that it conformed to the requirements of Regulation (EU) 2020/741 and the Spanish Royal Decree 1620/2007.

9.2. Materials and methods

9.2.1. UF prototype

A prototype UF plant was designed and installed for post-treatment of effluent from a medium-size (50800-p.e.; 14000 $m^3 \cdot d^{-1}$) urban WWTP in Valencia (Spain). The WWTP process is based on primary sedimentation and an activated sludge system for nutrient removal composed of biological reactors and secondary clarifiers.

The UF mainly consisted of two 15-L membrane tanks (MT1 and MT2), each containing a 2.5-m high hollow-fiber bundle (KMS Puron® Koch Membrane Systems) with a membrane area of 3.44 m^2 each and average pore size of 0.03 µm. One bundle was used for continuous operation and the other was kept in reserve for use during membrane cleaning or system maintenance. The plant also had a 30-L clean-in-place tank (CIP) to store permeate for membrane backwashing as required. A flow diagram of the prototype UF plant is shown in Figure 9.1.

A feed pump supplied the system with a flow rate of between 110 and 210 $L \cdot h^{-1}$ and the permeate pump filtered at a flow rate in the range of 95-135 $L \cdot h^{-1}$, corresponding to a transmembrane flux (J) of 27.6-39.2 LMH. Any tank overflow was recycled to a secondary clarifier in the WWTP. To reduce cake layer formation, air was sparged by a blower at a flow rate of between

0.75-4.50 $Nm^3 \cdot h^{-1}$, corresponding to a sparging air demand per m^2 of membrane (SAD_m) of 0.2-1.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$.

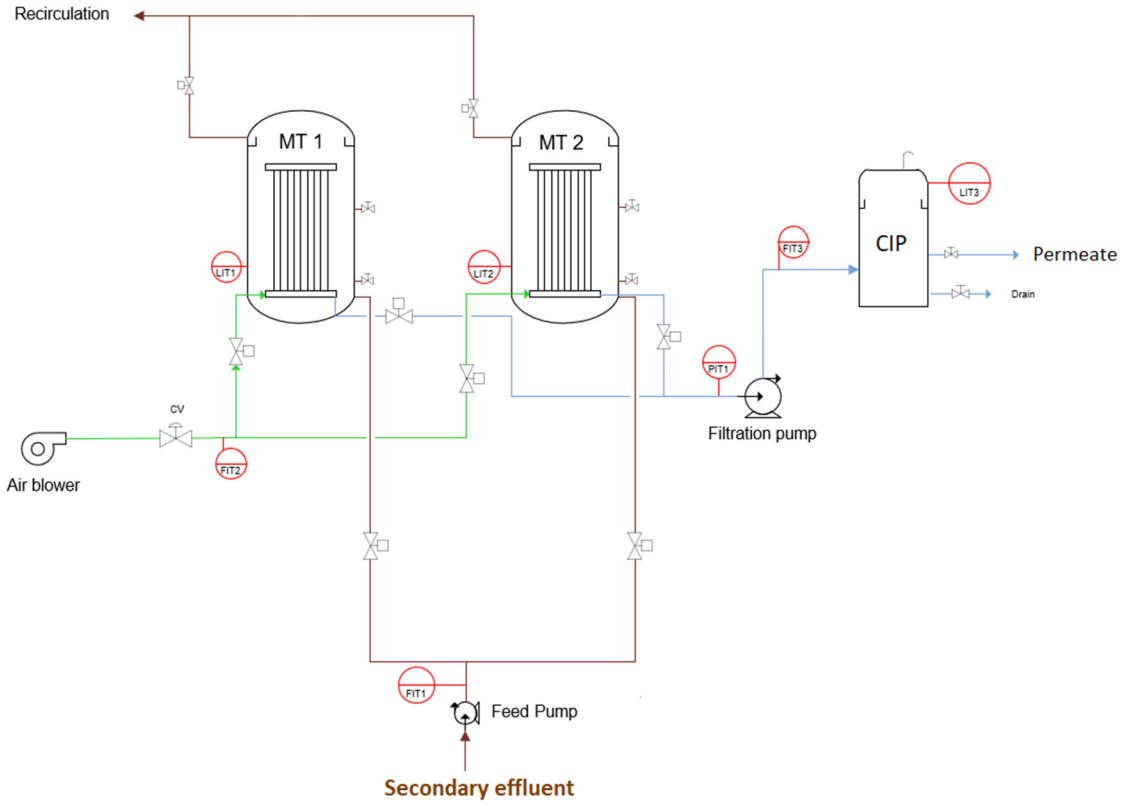


Figure 9.1. Flow diagram of the UF prototype for tertiary treatment.

Filtration process was controlled by an instrumentation, control and automation (ICA) system based on sensor-transmitters and actuators connected to a programmable logic controller (PLC), an automation system and a Supervisory Control and Data Acquisition (SCADA). The control philosophy used was the one proposed by Robles et al. (2012).

9.2.2. Operational periods and conditions

The goal of the UF prototype was to achieve maximum pollutant retention and produce a high-quality effluent suitable for reuse in agriculture at an assumable energy and monetary cost. Thus, different operating conditions were evaluated, such as varying J and SAD_m according to the following aims:

- To operate the membrane filtration at high permeate fluxes (in the range 28-39 LMH) to reduce membrane requirements and their acquisition cost.
- To obtain optimum membrane sparging by operating at the SAD_m with the minimum fouling propensity and energy consumption. A wide SAD_m range (0.2 – 1.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$) was tested to determine its influence on the UF performance with secondary effluent.
- To minimize the waste stream, thus avoiding unnecessary recirculation flows and improve filtration productivity. Following this principle, the feed pump flow was set to 1.05 times the permeate pump flow.
- To control membrane fouling and avoid over-consumption of chemicals for membrane cleaning.

The operating conditions are shown in Table 9.1. Different operating ranges were selected according to the season evaluated: summer (April – October, water temperature of 22-30ºC), winter (November– March, water temperature of 16-19ºC). Filtration was operated in cycles with alternate filtration, relaxation, and backwashing stages. Specifically, the filtration operating mode was set as follows: a combination of filtration–relaxation (F–R) cycles of 180 s (150 s filtration followed by 30 s relaxation) and 60 s of backwashing every 5 F–R cycles.

Table 9.1. Operating conditions of the UF prototype for tertiary treatment.

		$J_{20\ gross}$ LMH	SAD_m $Nm^3 \cdot m^{-2} \cdot h^{-1}$	T °C	Duration d
Summer	*S1*	27.6	0.73	29.6	20.3
	S2	27.6	0.44	28.0	16.0
	S3	39.2	1.31	28.3	17.4
	S4	33.4	1.31	24.9	31.4
	S5	33.4	0.28	24.7	32.8
Winter	*W1*	39.2	0.73	21.9	55.3
	W2	33.4	0.44	18.6	52.6
	W3	39.2	0.44	16.6	31.9
	W4	39.2	0.73	19.2	51.8
	W5	39.2	0.73	18.9	51.2
	W6	39.2	0.44	18.3	42.9
	W7	39.2	0.22	18.6	8.4
	W8	27.6	0.22	18.5	8.8

To obtain comparable results between different experimental periods, each experiment was carried out following the same procedure:

1. Previous chemical membrane recovery (2000-ppm NaOCl and 2000-ppm citric acid; 6 hours each). The initial transmembrane pressure (TMP) was thus similar in all the experiments, i.e., 0.05-0.08 bar.
2. Continuous operation of the UF plant with the pre-selected operating conditions (J and SAD_m).
3. TMP monitoring. When the TMP reached a value of 0.6 bar (maximum security value based on supplier's information), the operation was stopped and the experiment concluded. The experiment durations allowed an estimation of the membrane chemical cleaning frequency and membrane lifespan (see Section 9.2.5).

To evaluate filtration performance, the gross 20 ºC-standardized transmembrane flux ($J_{20,g}$; $L \cdot h^{-1} \cdot m^{-2}$; LMH), the fouling rate (FR; $mbar \cdot min^{-1}$), and the SAD_m ($Nm^3_{air} \cdot h^{-1} \cdot m^2$) were calculated by means on Equation 9.1, 9.2, and 9.3, respectively, an based on on-line monitored data:

$$J_{20} = J \cdot e^{-0.0239 \cdot (T-20)} \qquad \text{Equation 9.1}$$

$$FR = \frac{n \cdot \sum_1^n (TMP_i \cdot t_i) + \sum_1^n TMP_i \cdot \sum_1^n t_i}{n \cdot \sum_1^n TMP_i^2 - \left(\sum_i^n TMP_i\right)^2} \qquad \text{Equation 9.2}$$

$$SAD_m = \frac{Q_{air}}{A_m} \qquad \text{Equation 9.3}$$

Where T is the water temperature (°C), n is the total number of data monitored, TMP_i is the transmembrane pressures (mbar) at time t_i (min); Q_{air} represents the air flow for membrane scouring ($m^3 \cdot h^{-1}$) and A_m is the membrane surface area (m^2).

9.2.3. Analytical methods

9.2.3.1. Water quality analysis

Total and soluble chemical oxygen demand (tCOD and sCOD) were determined according to Standard Methods (American Public Health Association, American Water Works Association,

2012) 5220-COD-C and 522-COD-D, respectively. Ammonium (NH_4-N), nitrite (NO_2-N), nitrate (NO_3-N) and phosphate (PO_4-P) were analyzed according to Standard Methods (APHA, 2012): 4500-NH3-G, 4500-NO2-B, 4500-NO3-H and 4500-P-F, respectively, using an automatic analyzer (Smartchem 200, Westco Scientific Instruments, Westco). Volatile suspended solids (VSS) concentration was also measured in duplicate, according to method 2540 E of the Standard Methods (American Public Health Association, American Water Works Association, 2012).

9.2.3.2. Microbiology

The presence of *E. coli* was quantitatively determined by the standard method for enumeration of *E. coli* β-glucuronidase positive, following the UNE-EN ISO 9308-1:2014: 100 mL of the sample were filtered through a 0.45 μm nitrocellulose membrane (47 mm diameter Microfil©). The membrane was then placed on a Chromocult® coliform agar plate and incubated at 36±2 °C for 24 h, after which the number of positive (blue-green) colonies on the plate was counted. The result was the number of *E. coli* in the samples capable of form a colony during the incubation period, expressed in colony forming units per 100 mL (UFC/100 ml).

9.2.3.3. Heavy metals

The Spanish regulation on agricultural reuse of water is included in Royal Decree 1620/2007, which establishes, among other parameters, limit values for the concentration of boron (B), arsenic (As), beryllium (Be), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), manganese (Mn), molybdenum (Mo), nickel (Ni), selenium (Se) and vanadium (V). These analyses were carried out on an Agilent 7900 inductively coupled plasma mass spectrometer (ICP-MS).

9.2.4. Energy assessment

The operating data obtained from the UF prototype were used to project an industrial system for a WWTP with a treatment flow of 14000 $m^3 \cdot d^{-1}$. The instrumentation used in the UF prototype system acquired, among others, the following data: membrane tank influent flow, temperature, TMP and J, turbidity and air flows for membrane scouring. Based on the data collected, different key performance indicators were calculated to assess the system's performance, i.e., $J_{20,g}$ and SAD_m.

As the over-dimensioned equipment in the prototype plant prevented directly measuring the energy requirements of the UF tertiary treatment, the theoretical energy consumption of pumps and blowers was calculated from the equations proposed in Judd & Judd (2006) and the energy model described by Pretel et al. (2016). Adiabatic indexes and blower and pump efficiencies were estimated from the standard values for these components. Further details can be found in the supplementary material.

9.2.5. Life Cycle Costing

The life cycle costing (LCC) procedure was applied to assess the economic viability of the projected UF tertiary treatment based on the UF prototype performance. The LCC was used to calculate the total annual cost (TAC), which included capital and operating expenses (CAPEX and OPEX, respectively), according to the required inputs (civil works, energy and chemical consumption, equipment acquisition and replacement, etc.) with 1 m^3 of treated water as the functional unit. The main considerations in calculating the CAPEX and OPEX are summarized as follows:

CAPEX

- Depreciation: A depreciation period of 20 years for civil works and 10 years for equipment was used to calculate the TAC. The annual discount rate was set to 3% for civil works and 5% for equipment.

- Membrane tank: The membrane tank volume was estimated according to the dimensions of a commercial membrane unit (PULSION® Koch, 0.03-µm pore size, filtration area of 43.5 m^2).

- Pipelines: The maximum velocity of the liquid in pipes was set to 1.5 $m \cdot s^{-1}$ to calculate the pipe diameter, thus preventing excessive losses of mechanical energy.

- Equipment: The acquisition of blowers, pumps and UF membranes was included. The cost of equipment was estimated from the capacity based on flow rates and the unit costs provided by the manufacturers.

OPEX

- Power requirements: both the energy and power tariffs were included in the appraisals, as proposed in Somenergia (2022).

- Chemical reagents for membrane cleaning: Sodium hypochlorite and citric acid were considered for membranes chemical cleaning at a dose of 2000 ppm of both sodium hypochlorite and citric acid, while the contact time was 6 hours and a frequency of 8.1 chemical cleanings per year.

- Membrane replacement: the lifespan of the UF membrane was calculated from the maximum total contact with chlorine permissible before replacement: 500,000 ppm-hours according to the supplier or a maximum lifespan of 10 years.

- Equipment replacement and maintenance: The lifetime of the blowers and pumps was based on the total working hours in the manufacturers' recommendations, with a maximum of 10 years.

- Civil works and building maintenance: maintenance of civil works and buildings was a percentage of the associated CAPEX, according to the technical specifications for general WWTP structures (Inyges Consultores S.L., 2015)

- Labor costs: labor costs were not included in this LCC as both scenarios (wastewater treatment with and without UF process) were considered to be operated by the same staff.

Details of unit costs and correlation functions used to assess the CAPEX and OPEX in the proposed scenarios can be found in Table 9.1S (supplementary material).

The TAC ($€·m^{-3}$) of the whole process was thus calculated by means of CAPEX and OPEX, as shown in Equation 9.4:

$$TAC = \frac{r\,(1+r)^t}{(1+r)^t - 1} \cdot CAPEX + OPEX \qquad \text{Equation 9.4}$$

where *r* is the annual discount rate and *t* is the depreciation period in years.

9.2.6. Life cycle assessment

Environmental performance was conducted by means of a life cycle assessment (LCA) according to ISO 14040:2006. The life cycle inventory (LCI) of individual materials and processes (based on Ecoinvent 3.4 and Industry data 2.0 databases accessed via SimaPro v9.1.1) can be found in Table 9.2S of the supplementary material. ReCiPe 2016 and hierarchical approach was selected

as life cycle impact assessment (LCIA) method (also accessed via SimaPro v9.1.1). The construction and operation phases were considered, while dismantling phase was not included as it was considered negligible, according to Foley et al. (2010). In the operating phase, the following aspects were included in the LCA: i) filtration performance, ii) energy consumption, iv) chemicals consumption, v) effluent quality. As in LCC (see section 9.2.5), a functional unit of 1 m^3 of treated water, a 20-year lifespan were set to civil works and a maximum of 10 years for the equipment. The selected environmental categories were global warming potential, water consumption, fossil resource scarcity and mineral resource scarcity.

9.3. Results and discussion

The main characteristics of the influent to the UF prototype are summarized in Table 9.2.

The biological performance of the WWTP in terms of organic matter removal (tCOD) showed no significant differences between the seasons (p-value > 0.05). However, the TSS concentration and influent turbidity in winter was significantly higher than in summer as is common in urban WWTPs given that the different seasonal temperatures and WWTP influent characteristics influence the biological process of the treatment facility (Al Ali et al., 2020; Peces et al., 2022). In fact, it was found during the operation of the UF prototype that the urban WWTP presented some operating issues in winter related to bulking and foaming due to increased filamentous bacteria, though no bulking or foaming issues were detected in summer. As a result, the total suspended solids in winter had higher amounts of flocs than in summer.

9.3.1. UF prototype performance

In all the experiments showed in Table 9.1, a similar behavior was observed within the F-R and backwashing cycles (see Figure 9.1S in supplementary material). TMP increased as the deposition of particles on the membrane surface generated a resistance to J. Air sparging efficiently allowed removing the cake layer from the membrane surface. Nonetheless, a little amount of cake remained on the membrane surface throughout F-R cycles, being necessary to conduct backwashing cycles to fully remove reversible fouling from the membrane surface. However, over time, irreversible fouling tended to accumulate resulting in an increase in TMP in the mid/long-term.

Table 9.2. Characteristics of the influent to UF prototype for tertiary treatment.

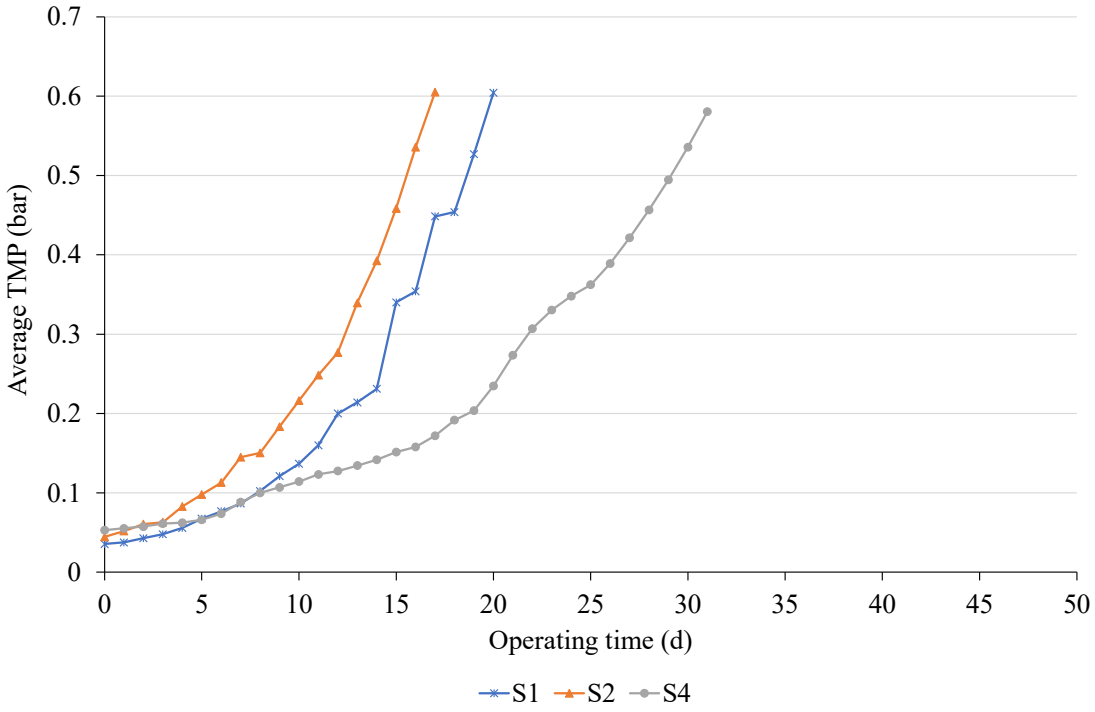
	TSS mg·L^{-1}				tCOD mg·L^{-1}				tCOD:TSS mg tCOD·mg^{-1} TSS				Turbidity NTU				pH			
	Mean	**±**	**SD**	**n**	**Mean**	**±**	**SD**	**n**	**Mean**	**±**	**SD**	**n**	**Mean**	**±**	**SD**	**n**	**Mean**	**±**	**SD**	**n**
Summer	12*	±	5	15	77	±	25	22	5.6*	±	1.7	18	5*	±	2	30	7.7	±	0.2	30
Winter	28*	±	21	30	70	±	44	52	2.6*	±	1.2	48	18*	±	17	60	7.8	±	0.3	60

*Showed statistically significant differences
n: number of samples

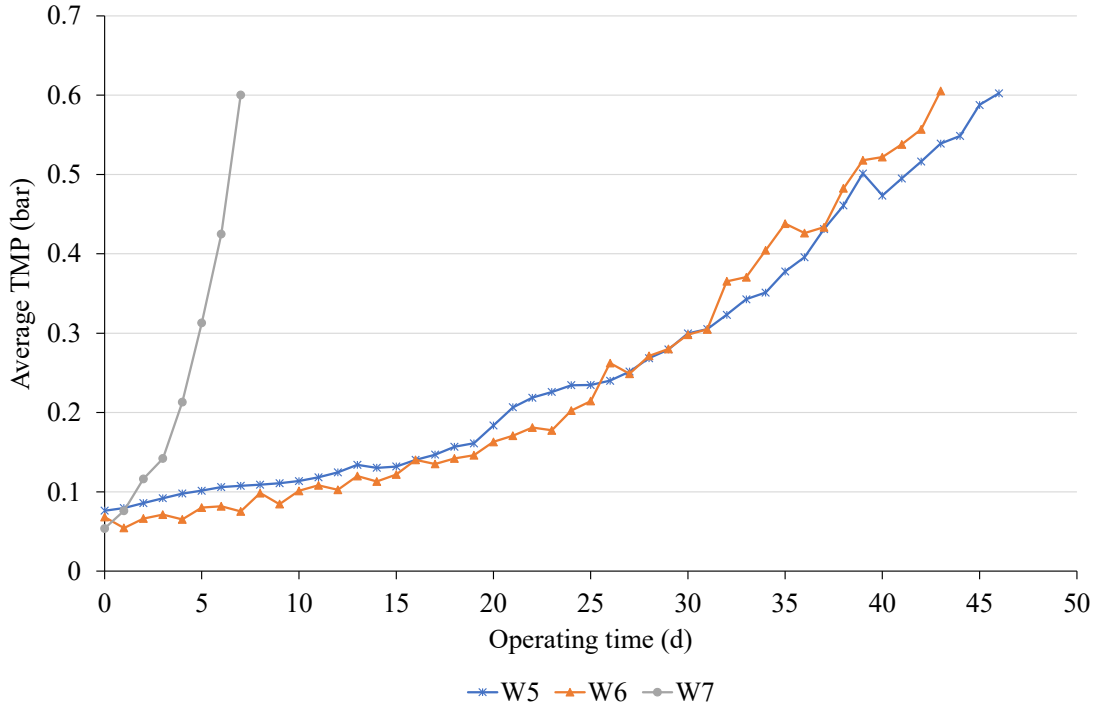
Irreversible fouling depends on many factors, including the water characteristics and the membrane operating conditions. A common strategy used to operate membranes consists of controlling the operating conditions to reduce the irreversible fouling rate and thus the need for chemical cleaning (Shahid et al., 2020).

Figure 9.2 shows an example of the results obtained during the continuous operation of the UF tertiary plant. Figure 9.2a shows the TMP evolution in experiments S1 ($J_{20,g}$: 27.6 LMH; SAD_m: 0.7 $Nm^3 \cdot m^{-2} \cdot d^{-1}$), S2 ($J_{20,g}$: 27.6 LMH; SAD_m: 0.4 $Nm^3 \cdot m^{-2} \cdot d^{-1}$) and S4 ($J_{20,g}$: 33.4 LMH; SAD_m: 1.3 $Nm^3 \cdot m^{-2} \cdot d^{-1}$). As this figure shows, the higher SAD_m, the longer the duration of the experiment, showing membrane scouring by gas sparging as a suitable fouling control strategy. However, likewise other UF systems applied within the wastewater treatment field, a boundary level for SAD_m was identified, above which increasing SAD_m does not enhance fouling mitigation, *i.e.*, reversible fouling is about being completely removed at a given SAD_m level. For instance, results from experiments W5 ($J_{20,g}$: 39.2 LMH; SAD_m: 0.7 $Nm^3 \cdot m^{-2} \cdot d^{-1}$), W6 ($J_{20,g}$: 39.2 LMH; SAD_m: 0.4 $Nm^3 \cdot m^{-2} \cdot d^{-1}$) and W7 ($J_{20,g}$: 39.2 LMH; SAD_m: 0.2 $Nm^3 \cdot m^{-2} \cdot d^{-1}$) in Figure 9.2b indicated then SAD_m values above 0.4 $Nm^3 \cdot m^{-2} \cdot d^{-1}$, for given operating conditions, did not enhanced fouling mitigation in the long term, but would mean unnecessary energy consumptions with no advantage for filtration operations.

Boundary levels were also identified for low SAD_m values. In this respect, Figure 9.3 shows the average fouling rate for the experimental periods summarized in Table 9.1. As this figure shows, W7 ($J_{20,g}$: 39.2 LMH; SAD_m: 0.22 $Nm^3 \cdot m^{-2} \cdot d^{-1}$) and W8 ($J_{20,g}$: 27.6 LMH; SAD_m: 0.22 $Nm^3 \cdot m^{-2} \cdot d^{-1}$) experimented the fasters fouling rate, even doubling the fouling rate observed in W1 ($J_{20,g}$: 39.2 LMH; SAD_m: 0.73 $Nm^3 \cdot m^{-2} \cdot d^{-1}$). These results suggest that an air sparging intensity of 0.2 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ is not enough to reduce membrane fouling propensity under the evaluated operating conditions.



a)



b)

Figure 9.2. Evolution of TMP in the long term. Influence of SAD_m on filtration performance in a) summertime and b) wintertime.

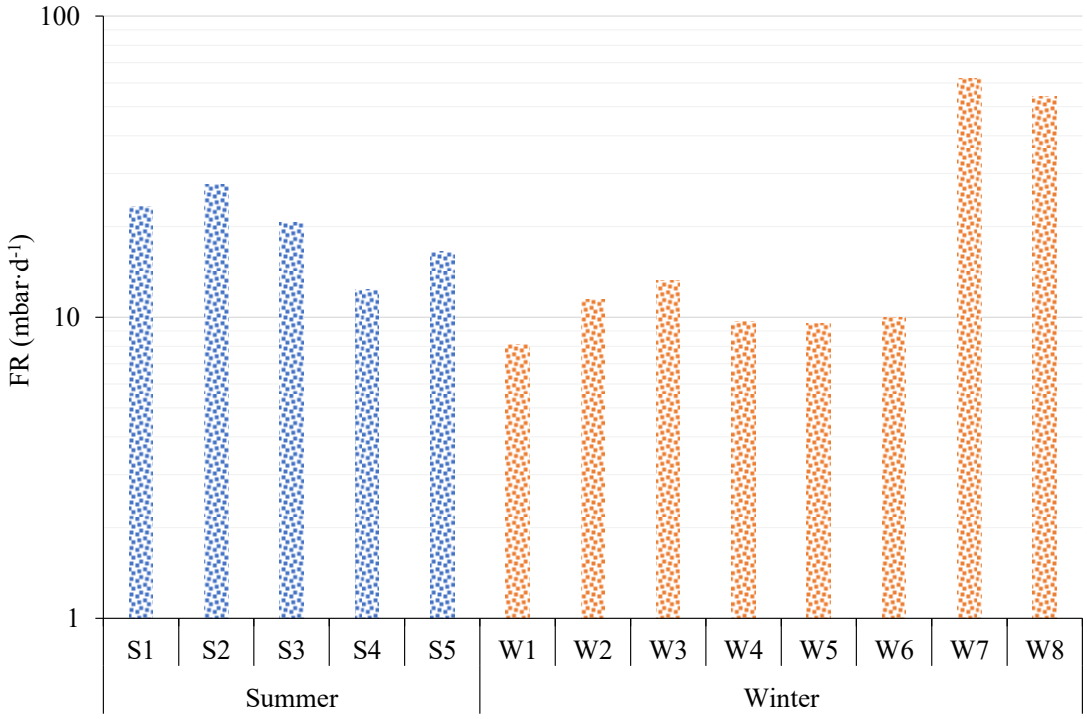


Figure 9.3. Average fouling rates (FR) for each filtration experiment.

On the other hand, Figure 9.3 shows similar fouling rates for S1 to S6 (p-value > 0.05). However, although the operating conditions in S1-S5 were in theory more favorable than in W1-W6 (i.e., high SAD_m with similar $J_{20,g}$, fouling rate was higher in the former than in the later). This behavior was not attributed to cake layer formation, since TSS contentment in the influent was lower in summer than in winter (see Table 9.2). Nevertheless, high temperatures tend to ease filtration as they reduce the resistance to the water flux (Peña et al., 2019), although it affects microbial activity and affects the amount of extracellular polymeric substances (EPS) and soluble microbial products (SMP) in the media. Indeed, the higher fouling rates recorded in S1-S5 compared to W1-W6 may be due to EPS and SMP as the tCOD:TSS ratio was significantly higher in summer than in winter (see Table 9.2). In this respect, although the TSS concentration in the influent was lower in summer than in winter, tCOD was higher in the former than in the later, indicating that a higher fraction of colloids was present in the former thus affecting pore blocking fouling mechanisms. Similarly to other UF systems treating non-biological liquors, pore blocking was expected to be relevant (e.g., Sanchis-Perucho et al., 2023b, 2023a).

Comparing different periods, minor differences were found for similar operating temperatures according to the selected conditions, i.e., periods with higher SAD_m and lower J showed lower fouling propensities. Overall, the results suggests that the UF system should be operated under

different conditions within the year to avoid excessive fouling, e.g., at a $J_{20,g}$ of around 28-33 LMH and SAD_m of 0.4-0.6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ in summertime and around $J_{20,g}$ of 33-39 LMH and SAD_m of 0.3-0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ in wintertime.

Finally, ion precipitation could contribute to some extend to an inorganic fouling development, but ion concentration was not analyzed. However, mayor contribution to membrane fouling is expected to have an organic origin. In this sense, Sanchis-Perucho et al. (2023b) reported contributions of organic fouling in the 70-85% range for UF membranes conducting direct membrane filtration of municipal wastewater with pH of 7.4-7.6, while Robles et al. (2012) obtained low precipitates formation with pH below 7, but an increasing precipitation propensity as pH raises. Therefore, ion precipitation cannot be discarded, but its influence was assumed to be low compared to organic fouling.

9.3.2. Full-scale UF treatment

The information obtained during the operation of the UF prototype was used to project a full-scale 14000 $m^3 \cdot d^{-1}$ UF unit for tertiary treatment and assess its energy performance and its economic feasibility. The size of the UF prototype (equipped with commercial UF hollow-fiber membranes, industrial pumping, auxiliary elements and control and monitoring devices) provided a robust set of data for scaling-up the prototype. The main ambient and operating conditions considered in the ups-scaling are summarized in Table 9.3.

Table 9.3. Selected operating conditions of the projected full-scale UF unit.

		$J_{20,g}$	SAD_m	T
		LMH	**$Nm^3 \cdot m^{-2} \cdot h^{-1}$**	**°C**
Summer	*Sa*	32.3	0.40	26.0
	Sb	32.3	0.60	26.0
	Sc	38.1	0.40	26.0
	Sd	38.1	0.60	26.0
Winter	*Wa*	31.5	0.30	18.0
	Wb	31.5	0.40	18.0
	Wc	37.2	0.30	18.0
	Wd	37.2	0.40	18.0

9.3.2.1. Energy assessment

The total energy consumption (TEC) of the projected full-scale UF unit was calculated considering: i) blower consumption for membrane scouring; ii) feeding pump; iii) permeate pump; and iv) backwash for membrane cleaning. The energy was assessed for summer (S) and winter (W) ambient conditions. The different $J_{20,g}$ and SAD_m applied (according to J and SAD_m suggested in Section 9.3.1) led to four sub-scenarios for each season, i.e., Sa to Sd and Wa to Wb.

The results are shown in Figure 9.4 and Table 9.3S of the supplementary material. The summer TEC was between 0.153 (Sc) and 0.345 $kWh \cdot m^{-3}$ (Sb) (0.239±0.083 $kWh \cdot m^{-3}$ on average); while in winter the maximum TEC was for Wb (0.207 $kWh \cdot m^{-3}$) and the lowest TEC was obtained for Wc (0.105 $kWh \cdot m^{-3}$). The W scenarios required an average energy consumption of 0.151±0.044 $kWh \cdot m^{-3}$. Regarding the energy contributions to TEC, the membrane blowers, which accounted for between 82 % (Wc) and 95 % (Sb), were the biggest energy expenditure. The feeding (3-9 %) and permeate (2-9 %) consumed much less, while backwashing was negligible (< 1 %). Any efforts to reduce energy expenditure should thus focus on reducing the membrane scouring needs. An appropriate filtration strategy should be adopted for this to guarantee significant transmembrane fluxes, which eventually reduce the membrane surface requirements and lead to smaller SAD_m and reduced energy consumption (see Figure 9.4). The summer SAD_m was the same in Sa and Sc (0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$) and in Sb and Sd (0.6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$). Moreover, for a given SAD_m the higher $J_{20,g}$, the lower blower energy consumption: the Sc blower energy requirement was lower (0.134 $kWh \cdot m^{-3}$) than Sa (0.181 $kWh \cdot m^{-3}$) and Sd was lower (0.239 $kWh \cdot m^{-3}$) than Sb (0.327 $kWh \cdot m^{-3}$). The same effect was seen in winter. Maintaining SAD_m kept reversible fouling propensity low, helping to prevent irreversible fouling, thus improving process productivity.

The energy consumption results are similar to those of other studies on tertiary treatments including UF. Melgarejo et al. (2016) evaluated the energy costs of water reclamation in the Rincón de León WWTP (Treatment flow: 52000 $m^3 \cdot d^{-1}$; Valencia, Spain), which combined different tertiary treatment processes: coagulation+flocculation+filtration (CFF), UF, ultraviolet disinfection (UV) and reverse osmosis (RO). The results on energy consumptions were 0.047 $kWh \cdot m^{-3}$ for CFF; 0.236 $kWh \cdot m^{-3}$ for UF ;0.056 $kWh \cdot m^{-3}$ for UV and 0.869 $kWh \cdot m^{-3}$ for RO. The energy consumption was therefore similar to the S scenarios (0.239±0.083 $kWh \cdot m^{-3}$) but significantly higher than to W ones (0.151±0.044 $kWh \cdot m^{-3}$). Results for energy consumption were also obtained by Akhoundi and Nazif (2018) who applied a multi-criteria analysis to assess the

sustainability of wastewater reuse alternatives in a WWTP in the south of Tehran (Iran) which treated 450000 $m^{-3} \cdot d^{-1}$. This study reported energy expenditures of 0.219 $kWh \cdot m^{-3}$, 0.2 $kWh \cdot m^{-3}$, 0.066 $kWh \cdot m^{-3}$ and 1.5 $kWh \cdot m^{-3}$ for coagulation+flocculation, UF, UV and RO, respectively. In this case the UF results were 16 % lower than the average energy consumption obtained in summer but 32 % higher than in winter.

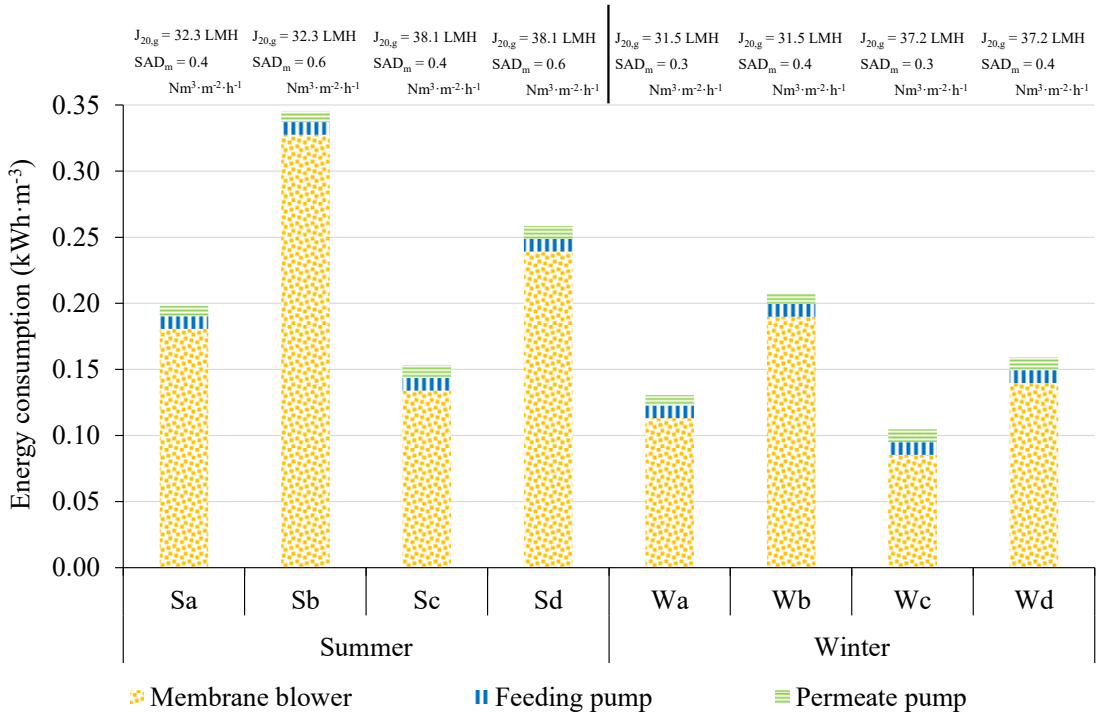


Figure 9.4. Total energy consumption of the projected full-scale UF unit.

9.3.2.2. Life cycle costing

Based on the results obtained during the long-term operation of the UF prototype unit, an economic evaluation was conducted according to the LCC method proposed in Section 9.2.5. Since different operating conditions for J and SAD_m were considered for winter and summer, a seasonal LCC was developed with four sub-scenarios in each (Sa to Sd and Wa to Wd, respectively). The LCC thus represents a cost range for a full-scale UF unit operated for a year.

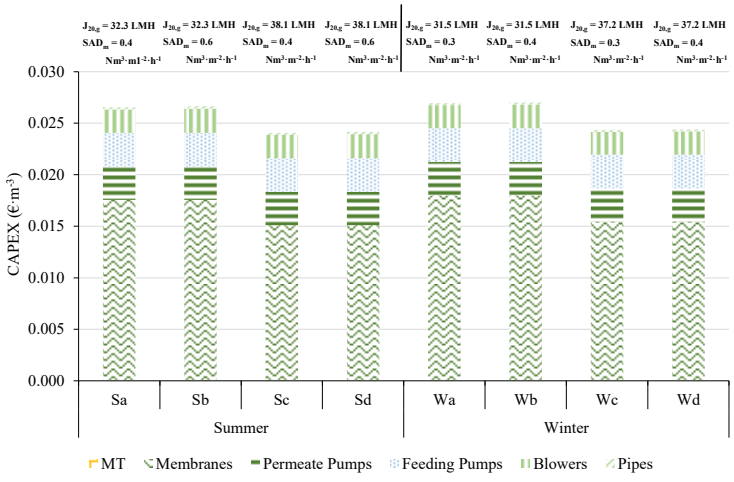
9.3.2.2.1. Capital expenditure

Figure 9.5a and Table 9.4S of the supplementary material show the CAPEX results obtained for the projected full-scale UF unit. Membrane acquisition appeared as the main contributor to

CAPEX, accounting for 62-66 % in summer and 63-67 % in winter. Sub-scenarios with low $J_{20,g}$ led to higher CAPEX, since the membrane requirements depend on filtration productivity. For example, CAPEX in Sa ($J_{20,g}$ of 32.3 LMH) was €0.027·m^{-3} while Sc ($J_{20,g}$ of 38.1 LMH) obtained €0.024·m^{-3}. On average, the summer scenarios produced a CAPEX of €0.025±0.001·m^{-3} and winter scenarios €0.026±0.001·m^{-3}, i.e., non-significant differences. The largest investment cost was for membranes, €0.016±0.001·m^{-3} and €0.017±0.001·m^{-3} in summer and winter, respectively. Permeate pumps and feeding pumps accounted for €0.003±0.000·m^{-3} both in S and W scenarios (13±1 % each), while the blowers' contribution was calculated as €0.002±0.000·m^{-3} (9 %). The SAD_m and $J_{20,g}$ applied in this study did not therefore lead to significant differences in the CAPEX results for pumps and blowers. The cost of pipes and membrane tanks was negligible (≈ 1 %).

These CAPEX results show that the membranes determine the investment cost, so that improving the results implies: i) working with filtration strategies with high transmembrane flows in the long term; and/or ii) a reduction in membrane costs due to market trends. An increase of 10 % in the applied $J_{20,g}$ (to 35.5-41.9 LMH range), while considering no effect on membrane fouling, would reduce membrane acquisition costs by 6 % in both S and W scenarios. On the other hand, a reduction of 10 % in the cost per m^2 of membrane would reduce its contribution to €0.015 and €0.016·m^{-3} under S and W conditions, respectively, which represents savings of 6 % in both scenarios.

Finally, although in this study an economic evaluation of CAPEX is carried out for different operating conditions in winter and summer, the effective implementation of a UF system at an industrial level should be evaluated based on the most unfavorable CAPEX. That is, the economic viability must be evaluated based on the most conservative scenario to guarantee that the investment cost of the implemented system corresponds to a process capable of achieving the treatment requirements.



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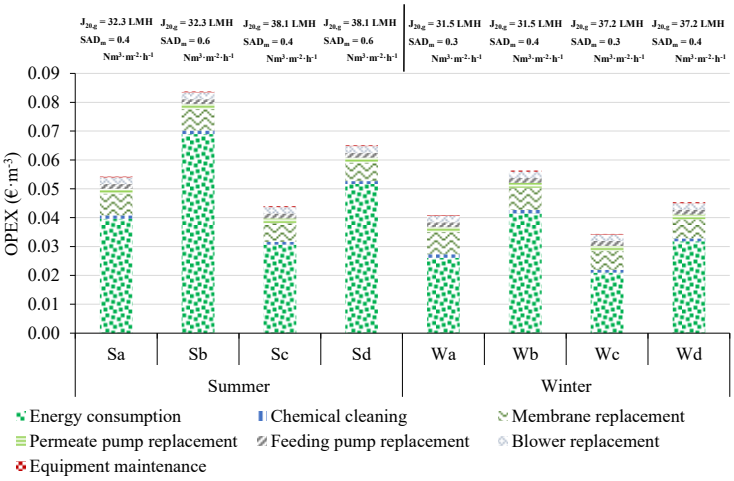

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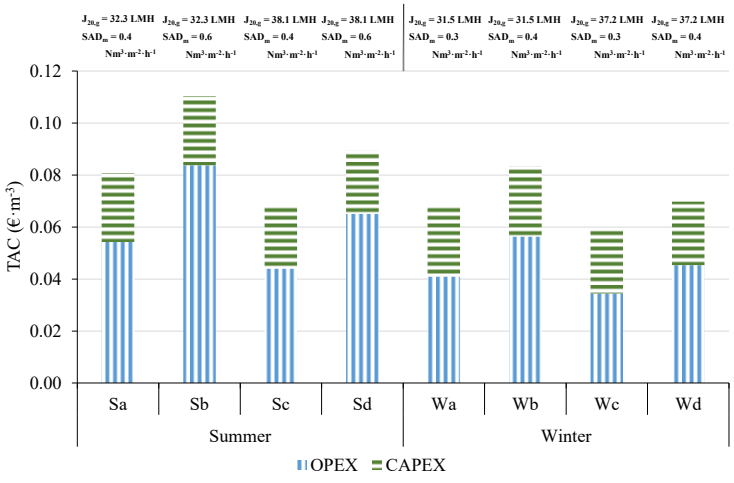


c)

Figure 9.5. Economic assessment of the projected full-scale UF unit: a) CAPEX; b) OPEX; and c) TAC.

9.3.2.2.2. Operating expenditure

As can be seen in Figure 9.5b and Table 9.5S of the supplementary material, OPEX varied in summer between €0.044 (Sc) and €0.084·m^{-3} (Sb), while operating costs were between €0.034 and €0.056·m^{-3} in winter (Wc and Wb, respectively). TEC was the main contributor to OPEX, with an average cost of €0.048±0.017·m^{-3} in summer and €0.030±0.009·m^{-3} in winter, or 76±6 % and 67±6%, respectively. UF membrane replacement was also a significant OPEX cost and accounted for €0.007±0.001·m^{-3} in summer and winter (12±3 % and 17±3 %, respectively). Chemical cleaning of membranes (2-3 %), permeate pump replacement (3-4 %), feeding pump replacement (3-4 %) and blower replacement (4-5 %) were minor contributions to OPEX, while equipment maintenance was negligible (≈1 %).

Reducing OPEX thus involves reducing the aeration requirements for membrane scouring, e.g., reducing SAD_m between Sb and Sa (from 0.6 to 0.4 $Nm^3 \cdot m^2 \cdot h^{-1}$) reduced OPEX from €0.084·m^{-3} to €0.054·m^{-3}, or 36 %. However, reducing SAD_m can increase fouling, which eventually reduces filtration productivity and requires a larger membrane surface, so that aeration optimization should include the complete filtration process and the interrelationships of the different factors.

Operating expenditure of the different tertiary treatments (CFF, UF, UV and RO) can be found in Melgarejo et al., (2016). The results for OPEX were: CFF of €0.014·m^{-3}; UF of €0.034·m^{-3}; UV of €0.007·m^{-3}; and RO of €0.210·m^{-3}. The UF results are similar to those obtained at the lower end of the OPEX range in this study, €0.034·m^{-3} in sub-scenario Wc. OPEX was also calculated by Akhoundi and Nazif (2018) (WWTP which treats 450000 $m^{-3} \cdot d^{-1}$), who obtained the following operating costs: Coagulation-flocculation of €0.062-0.090·m^{-3}, UF of €0.146-0.570·m^{-3}, UV of €0.006·m^{-3} and RO of €0.185-0.292·m^{-3}. This range for UF is significantly higher than the highest OPEX obtained in the present study, €0.084·m^{-3} in sub-scenario Sb. Jodar-Abellan et al., (2019) reported an operating costs range of €0.05-0.09·m^{-3} for tertiary treatment processes based on microfiltration, UF, RO and reversible electrodialysis, similar to the OPEX range in this study (€0.034-0.084·m^{-3}). Clem and Mendonça (2022) assessed a combination of ozone and UF for tertiary treatment of wastewater and obtained an OPEX range of €0.280-0.862·m^{-3}, which is significantly higher than operating costs in this study. Hernández-Chover et al. (2022) reported an operating cost of UF treatment for water reclamation as €0.184·m^{-3}. However, it is important to highlight that the OPEX results are strongly influenced by the items considered in the

assessment (e.g., inclusion of labor cost), treatment capacity and the price of energy, so that comparisons between studies should be carefully conducted.

9.3.2.2.3. Total annualized cost

Figure 9.5c shows that the TAC varied from €0.068·m^{-3} (Sc) to €0.110·m^{-3} in summer (Sb) while in winter it varied between €0.059·m^{-3} (Wc) and €0.083·m^{-3} (Wb). Summer TAC was an average of €0.087±0.018·m^{-3} and 0.070±0.010·m^{-3} in winter. OPEX was the largest TAC cost, accounting for 70±5 % in summer and 63±4 % in winter. Improving the economic viability of the technology thus involves setting the aeration to sustain significant transmembrane fluxes and reduced fouling to reduce the energy requirements for membrane scouring.

The Spanish government recently issued a new regulation to boost water reuse (Royal Decree 4/2023, 2023) that provides an exception to the discharge fee to surface water when treated wastewater is effectively reused. The potential benefits of this measure was already pointed out in Jiménez-Benítez et al. (2020). The Spanish regulation sets a basic discharge fee of €0.01751·m^{-3} and a coefficient of increase or decrease proportional to the discharged water quality. Since tertiary UF treatment produces reclaimed class A water (European Comission, 2020), the savings in the discharge fee could reduce OPEX up to an average of €0.044±0.017·m^{-3} in summer and €0.026±0.009·m^{-3} in winter, while the TAC would be between €0.069±0.018·m^{-3} and €0.052±0.010·m^{-3}, respectively.

The nutrient content in the effluent provides additional economic benefits since it can replace the mineral fertilizers in agriculture. Considering a nutrient concentration of 9 mg N·L^{-1} and 0.9 mg P·L^{-1} in the effluent and a fertilizer price for urea ($CO(NH_2)_2$) and superphosphate (P_2O_5) of €0.295 and €0.435 per kg, respectively, the WWTP would discharge nutrients worth €34728 per year. An optimal use of reclaimed water must therefore include the evaluation of the nutrient content since the agricultural use of these substances would imply a higher economic value for the reused water. Furthermore, lower treatment cost associated with the nutrient removal in the WWTP would be expected if their recovery or reuse is included in a comprehensive fertigation system, since their removal from the wastewater would no longer be desirable. This approach aligns with current European policies which aim to boost circular economy in the water sector, such as such as the Circular Economy Action Plan (European Commission, 2020c).

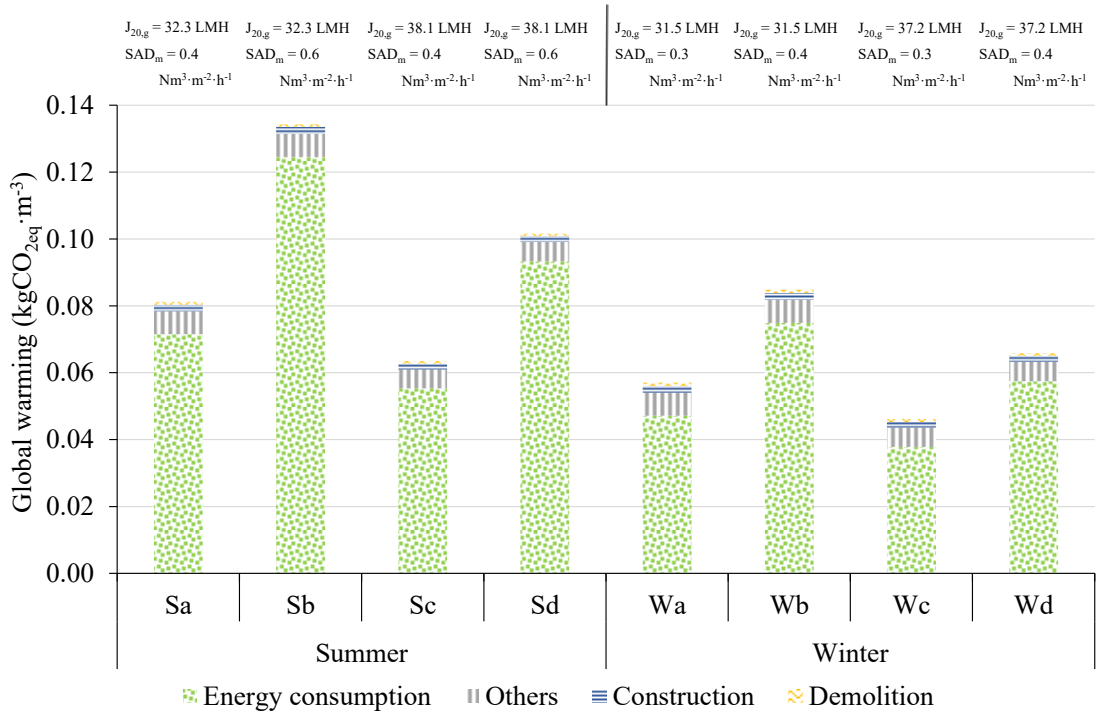
9.3.2.3. Life cycle assessment

The LCA outcome is shown in Figure 9.6, where results for global warming potential, water consumption, fossil resource scarcity and mineral resource scarcity are depicted. Major direct environmental burdens were associated to operation phase, with 95-98 % for global warming (0.04-0.13 kg CO_{2eq}·m-3), water consumption ($4.9\cdot10^{-4}$-$1.3\cdot10^{-3}$ $m^3\cdot m^{-3}$) and fossil fuel scarcity (0.01-0.03 kg $Oil_{eq}\cdot m^{-3}$) and slightly lower for mineral resource scarcity, 90-92 % ($7.6\cdot10^{-5}$-$2.2\cdot10^{-4}$ kg $Cu_{eq}\cdot m^{-3}$). Construction phase showed low influence, being only significant for mineral resource scarcity (8-10 %) mainly related to membrane acquisition (> 90 % of construction phase in this category). Environmental benefits associated with dismantling phase were negligible (<3 %).

Within operation phase, energy consumption showed the highest influence for all categories evaluated. Specifically, energy represents between 87-95 for global warming potential, 69-86 % for water consumption, 84-94 % for fossil fuel scarcity and 64-75 for mineral resource scarcity.

For comparison, assuming a tertiary treatment system for a similar flow rate using 12 UV radiation lamps with an energy consumption of 0.04-0.13 $kWh\cdot m^{-3}$ (according to (Collivignarelli et al., 2021) would present global warming potential of 0.01-0.05 kg $CO_{2eq}\cdot m^{-3}$, a water consumption impact of $1.3\cdot10^{-4}$-$4.2\cdot10^{-4}$ $m^3\cdot m^{-3}$, a fossil resource depletion of $3.4\cdot10^{-3}$-$1.1\cdot10^{-2}$ kg $Oil_{eq}\cdot m^{-3}$ and mineral resources depletion of $1.9\cdot10^{-5}$-$6.1\cdot10^{-5}$ kg $Cu_{eq}\cdot m^{-3}$.

Therefore, improving the environmental performance of the application of UF membranes for tertiary treatment necessarily involves reducing energy consumption, as specified in section with high flows and, therefore, reduce the needs of membrane and its aeration.



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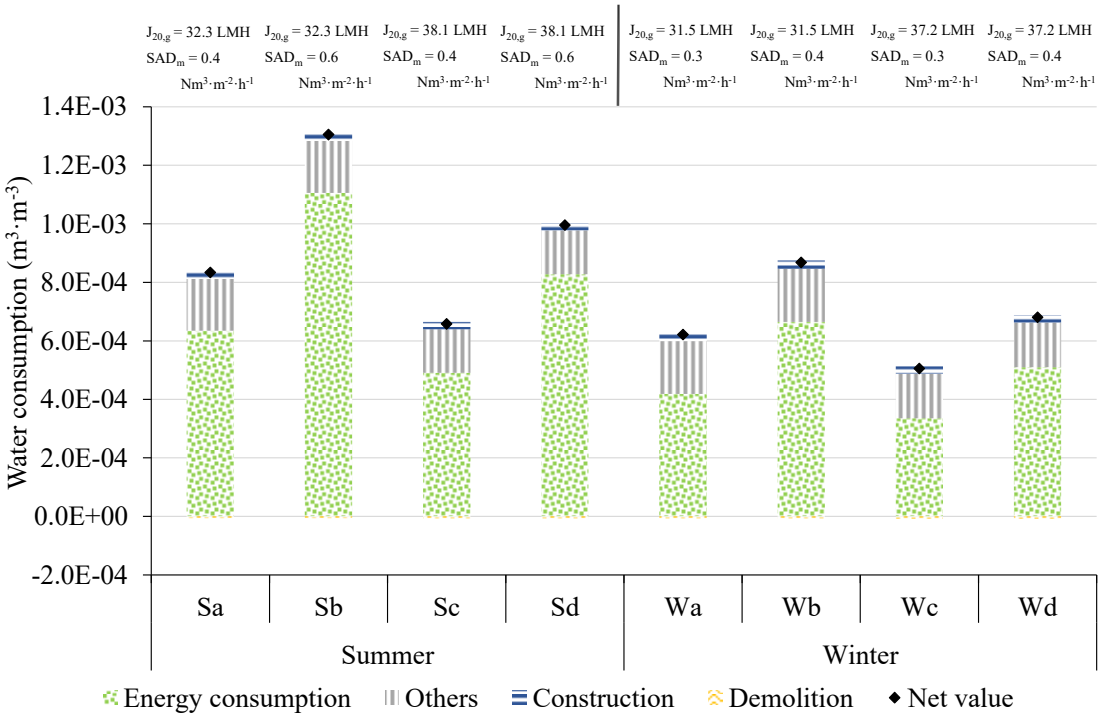
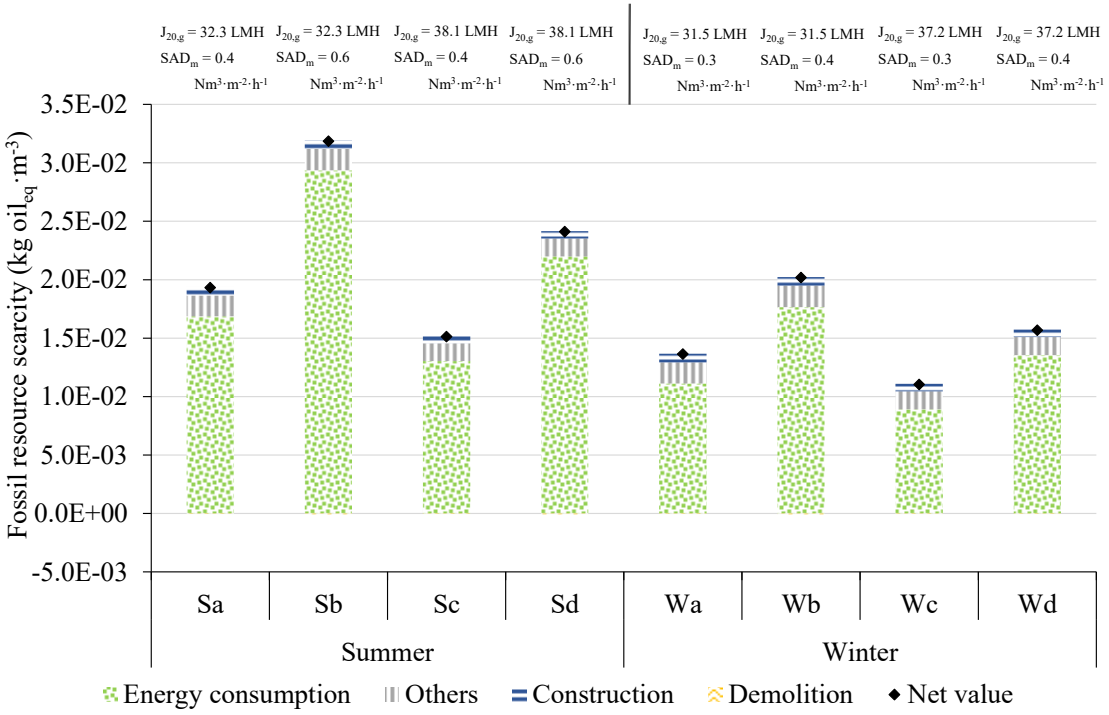


b)

Figure 9.6. Results of environmental assessment in both scenarios for a) global warming potential (GWP), b) water consumption (WC); c) fossil resource scarcity (FRS); and d) mineral resource scarcity (MRS) (continue).



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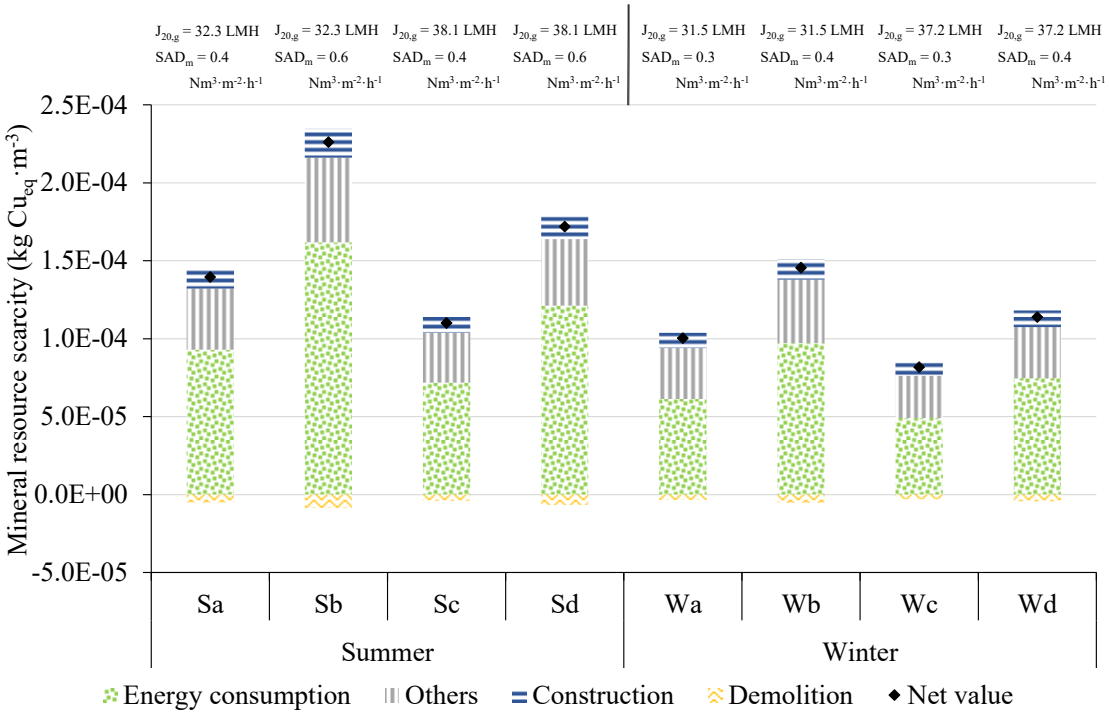

d)

Figure 9.6. Results of environmental assessment in both scenarios for a) global warming potential (GWP), b) water consumption (WC); c) fossil resource scarcity (FRS); and d) mineral resource scarcity (MRS).

9.3.3. Effluent quality

The current legal framework for water reuse in Spain includes Regulation (EU) 2020/741 on the minimum requirements for water reuse (EU-WR) together with the Spanish Royal Decree 1620/2007. The EU-WR simply deals with agricultural reuse, while the S-RD also includes others (e.g., urban, industrial, recreational, and environmental uses). As the EU-WR has just started to be applied, the current Spanish legislation will soon be updated to adapt to EU regulations.

Both rules classify different water qualities according to the irrigation method and water-crop contact. Both regulations are stricter when edible parts of the crop are in direct contact with reclaimed water, while water reuse for non-food crops is less restricted, following a fit-for-reuse approach (Radini et al., 2023). In this study, effluent quality was compared to the more stringent quality requirements of both regulations: "Crop irrigation with a water application system that allows direct contact of the reclaimed water with the edible parts for fresh human consumption" in the S-RD; and Quality Class A in the EU-WR suitable for "All food crops consumed raw in which the edible part is in direct contact with reclaimed water and root crops that are consumed raw".

Table 9.4 shows the obtained microbiological and physic-chemical results for the effluent of the UF prototype together with the requirements of the EU-WR and S-RD. The sampling campaign comprised a total of 10 samples collected during the operation in spring. The analysis did not detect any *E. coli* in the effluent immediately after the UF membranes. This is in line with other studies that described UF membranes as an effective barrier to pathogens thanks to their pore size. Ferreiro et al., (2021) applied continuous UF (pore size of 0.02 μm) with and without a self-cleaning system to the effluent of the Galindo WWTP (Basque Country, Spain) and obtained 0.0±0.0 and 0.7±1.01 CFU 100 mL^{-1} for *E. coli*, respectively. Foglia et al. (2020) studied a treatment scheme based on UASB followed by AnMBR equipped with hollow-fiber UF modules (pore size of 0.03 μm) for the treatment of WWTP effluent in Falconara Marittima (Italy) and reported 1 CFU·100 mL^{-1} in the permeate, while Yang et al., (2021) also confirmed UF (pore size of 0.02 mm) as a suitable method of meeting EU-WR standards for class A water quality.

However, other studies did find *E. coli* in UF permeate; Peña et al., (2019) analyzed the permeate of a AnMBR equipped with UF membranes with a result of 42±48 CFU·100 mL^{-1}. As in this study the permeate was stored in an open-air tank from which the samples were taken, bacteria re-growth was likely. Also, pore size was higher, 0.045 □m. Pérez et al., (2022) combined UF (pore

size of 0.02 µm) and RO for water reclamation from secondary effluent at Vuelta Ostrera WWTP (Cantabria, Spain). *E. coli* in the permeate was in the range 0-649 CFU·100 mL^{-1}, with an average of 17 CFU·100 mL^{-1}. In this case, the UF permeate was stored in a tank that fed the RO or was used for backwashing and chemical cleaning. Although the study did not specify how the samples for microbiological analysis were collected, they were possibly obtained from an open-air storage tank. Bray et al. (2021) studied 200 kDa and 400 kDa UF membranes for disinfecting effluents at Gdansk-Wshód and Gdynia-Degórze WWPs (Poland) and reported concentrations of fecal coliforms, which includes *E. coli* in the range 5-20 CFU·mL^{-1} for 200 kDa membranes and 70-180 CFU·mL^{-1} for 400 kDa However, when explaining the presence of microorganisms in the permeate they hypothesized that it could have been due to leaks or breaks in the membrane. The above studies were mostly carried out under S-RD and not EU-WR requirements for qualities A and B. These findings highlight the importance of correct sampling when measuring pathogens, as well as efficient membrane operation and maintenance. It should also be remembered that EU regulations require appropriate water quality at the point of supply to the consumer, so that if bacteria re-growth is common in storage tanks, this is an issue that reclaimed water managers would have to deal with if this technology is implemented.

Regarding organic matter, BOD_5 is included as a parameter only in EU-WR and only for Quality Class A. while for the other quality classes, BOD_5 should not be over the limit imposed by the WWTD, i.e., 25 mg·L-1. Average BOD_5 was 4.2±2.8 mg·L^{-1}, which allows unrestricted use of effluents (BOD_5 < 10 mg·L^{-1}). This would also accomplish with the updated limits expected in the incoming Wastewater Treatment Directive (European Commission, 2022b). As UF membranes retain particulate organic matter, only soluble DBO_5 passes through, which significantly improves the permeate quality. For instance, Ferreiro et al., (2021) obtained a BOD_5 of 4±1 mg·L^{-1} and 2.9 mg·L^{-1} before and after UF, respectively. As conventional efficiently operated aerobic wastewater treatments tend to have low BOD_5 in their effluents, usually below 10 mg L^{-1} (Tchobanoglous et al., 2014), this parameter should not be a bottleneck for water reuse.

TSS and turbidity also showed values below those established in the EU-WR and S-RD (Table 9.4). Again, the presence of UF membrane guarantees good quality permeate in relation to these parameters, thanks to the control produced by a given pore size. Ferreiro et al., (2021) reported a reduction of TSS and turbidity from 3.4±2.5 mg·L^{-1} and 9.5±1.7 NTU (secondary effluent) to 0.06 mg·L^{-1} and 0.95 NTU, respectively. Pérez et al., (2022) obtained results for TSS and turbidity in the secondary effluent in the range of 0-30 mg·L^{-1} (average 10.7 mg·L^{-1}) and

1.12-26.3 NTU (average 6.62 NTU), respectively, whereas UF reduced them to the 0-6 $mg \cdot L^{-1}$ range (average 1.16 $mg \cdot L^{-1}$) and 0.00-2.98 NTU (average 0.25 NTU).

Table 9.4. Microbial and physicochemical characterization of the effluent.

Parameter	Unit	Effluent Mean±SD	Regulation (EU) 2020/741	Spanish royal decree 1620/2007
E. coli	$CFU \cdot 100\ mL^{-1}$	0 ± 0	≤ 10	≤ 100
BOD_5	$mg \cdot L^{-1}$	4.2 ± 2.8	≤ 10	n.a.
SS	$mg \cdot L^{-1}$	3 ± 2	≤ 10	≤ 20
Turbidity	NTU	1.0 ± 0.6	≤ 5	≤ 10
Conductivity	$dS \cdot m^{-1}$	1.7 ± 0.3	n.a.	≤ 3.0
Metals, transition metals and metalloids				
B	$mg \cdot L^{-1}$	1.41E-01 ± 4.08E-02	n.a.	≤ 0.5
As	$mg \cdot L^{-1}$	8.26E-04 ± 1.14E-04	n.a.	≤ 0.1
Be	$mg \cdot L^{-1}$	2.90E-05 ± 4.25E-05	n.a.	≤ 0.1
Cd	$mg \cdot L^{-1}$	7.66E-05 ± 5.97E-05	n.a.	≤ 0.01
Co	$mg \cdot L^{-1}$	2.96E-04 ± 1.16E-04	n.a.	≤ 0.05
Cr	$mg \cdot L^{-1}$	2.70E-03 ± 3.87E-03	n.a.	≤ 0.1
Cu	$mg \cdot L^{-1}$	3.03E-02 ± 2.90E-02	n.a.	≤ 0.2
Mn	$mg \cdot L^{-1}$	1.61E-02 ± 8.78E-03	n.a.	≤ 0.2
Mo	$mg \cdot L^{-1}$	2.43E-03 ± 9.87E-04	n.a.	≤ 0.01
Ni	$mg \cdot L^{-1}$	3.70E-02 ± 3.90E-02	n.a.	≤ 0.2
Se	$mg \cdot L^{-1}$	9.64E-04 ± 4.74E-04	n.a.	≤ 0.02
V	$mg \cdot L^{-1}$	7.07E-04 ± 3.75E-04	n.a.	≤ 0.1

Only the concentration thresholds for metals, transition metals and metalloids (MET) are explicitly included in the S-RD, although the EU-WR also refers to them as elements to be considered if there are significant risks to human health and the environment. These risks should be evaluated by the mandatory water reuse risk management plans (WRRMPs) (Radini et al., 2023). As shown in Table 9.4, MET concentrations clearly satisfy the maximum admissible concentrations, so that no significant risks from metal pollution would be expected if this wastewater is reused. These results are consistent with locations with a low or negligible industrial contribution to WWTP influents. In the same way, conductivity only appears as a limiting parameter in the S-RD, with a maximum value of 3.0 $dS \cdot m^{-1}$, which is significantly higher than permeate conductivity in this study (1.7±0.3 $dS \cdot m^{-1}$).

Overall, the results obtained in this study demonstrate that tertiary UF treatment produces a high-quality effluent capable of accomplishing the stringent microbiological and physis-chemical requirements set by the Spanish and European regulations. It is important to highlight the differences between the EU-RW and the S-RD quality requirements for similar agricultural water reuse (Quality 2.1 and Quality Class A) in terms of *E. coli*, DBO_5, SS and turbidity, which would require an additional effort for its adoption into the Spanish legal framework, especially for *E. coli*. On the other hand, it is unclear whether its adoption in Spain will maintain the limits for parameters such as metals and conductivity, or if they will be evaluated according to the fit-for-reuse approach, i.e., only if there is a significant risk according to WRRMPs (Radini et al., 2023). The consequences of the legal modification should be considered and addressed by regulators and policy makers to guarantee economically and environmentally sustainable quality of reclaimed water to promote water reuse and circular water management (Foglia et al., 2023).

9.4. Conclusions

An UF demonstration plant for tertiary treatment was operated for water reclamation. $J_{20,g}$ between 28 and 39 LMH and SAD_m in the 0.2-1.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ were applied during winter and summer ambient conditions and fouling rates were monitored to assess the treatment performance. The obtained results recommend operating with different conditions during the year to prevent excessive fouling propensity: $J_{20,g}$ of 28-33 LMH and SAD_m of 0.4-0.6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ in summer and $J_{20,g}$ of 33-39 LMH and SAD_m of 0.3-0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ in winter.

Based on demonstration plant data, an up scaling to an industrial UF facility was projected and its energy, economic and environmental profile was evaluated. On average, energy consumption of 0.239±0.083 $kWh \cdot m^{-3}$ in summer and 0.151±0.044 $kWh \cdot m^{-3}$ in winter were obtained being blowers the main contributors (82 % in summer and 95 % in winter). With regards to CAPEX, it was calculated as €0.026±0.002$\cdot m^{-3}$ in both scenarios and membrane acquisitions accounted for the main contribution, 62-66 % in summer and 63-67 % in winter conditions. The expenditure associated to energy consumption was de major OPEX contributor (between 67-76 %), and total operating cost were calculated as €0.062±0.017$\cdot m^{-3}$ and €0.044±0.009$\cdot m^{-3}$ for summer and winter, respectively. From the results of CAPEX and OPEX, a TAC of €0.087±0.018$\cdot m^{-3}$ in summer and €0.070±0.010$\cdot m^{-3}$ in winter was obtained.

LCA results showed operation phase as major contributor to the environmental burdens, being only significant in mineral resource scarcity category (8-10 %). The environmental impacts of UF

are mainly related to energy consumption. Therefore, optimizing the filtration performance focused on diminishing energy requirements would improve the environmental sustainability of the treatment.

Finally, the analysis of the effluent confirmed tertiary treatment based on UF technology as a suitable system to obtain a high-quality effluent which accomplished the most stringent requirements for agriculture water reuse stated in both Spanish and European Regulation on water reclamation for agriculture.

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9.5. Supplementary material

9.5.1. Energy assessment

The operating data obtained from the UF prototype were used to project an industrial system for a wastewater treatment plant with a treatment flow of 14000 $m^3 \cdot d^{-1}$. The instrumentation implemented in the UF prototype system allowed the acquisition, among other, of the following data: membrane tank influent flow (Q_{inf}), temperature (T), transmembrane pressures (TMP) and fluxes (J), turbidity (Turb) and air flows for membrane scouring (Q_{air}). Based on the data collected, different key performance indicators were calculated to assess the performance of the system, i.e., J_{20} and the sparging air demand per m^2 of membrane (SAD_m).

Over-dimensioned equipment in the prototype plant prevented direct measure of energy requirements of the UF tertiary treatment. Therefore, theoretical energy consumption of pumps and blowers were calculated as follows (see Equation 9.1S, Equation 9.2S and Equation 9.4S), based on the equations proposed by Judd & Judd (2006) and the energy model described by (Pretel et al., 2016d). This energy model has been already applied to similar UF-membrane system applied to microalgae cultivation (González-Camejo et al., 2020). Adiabatic indexes, and blowers and pumps efficiencies were estimated based on standard values for these components.

$$P_B = \frac{(M \cdot R \cdot T_{gas})}{(\alpha - 1) \cdot \eta_{blower}} \left[\left(\frac{P_2}{P_1} \right)^{\frac{\alpha - 1}{\alpha}} - 1 \right] \qquad \text{Equation 9.1S}$$

where P_B ($J \cdot s^{-1}$) is the blower power requirement (adiabatic compression), M ($mol \cdot s^{-1}$) is the molar flow rate of biogas, R ($J \cdot mol^{-1} \cdot K^{-1}$) is the gas constant for biogas, P_1 (atm) is the absolute inlet pressure, P_2 (atm) is the absolute outlet pressure, T_{gas} (K) is the biogas temperature, α is the adiabatic index and η_{blower} is the blower efficiency.

$$P_g = q_{imp} \cdot \rho_{liquor} \cdot g \cdot \frac{\left\{ \left[\left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{asp} + \left(\frac{(L + L_{eq}) \cdot f \cdot V^2}{D \cdot 2 \cdot g} \right)_{imp} \right] + [Z_1 - Z_2] \right\}}{\eta_{pump}} \qquad \text{Equation 9.2S}$$

where P_g is the power requirement by a general pump ($J \cdot s^{-1}$), considering both pump aspiration and pump impulsion section, calculated from the impulsion volumetric flow rate ($q_{imp.}$ in $m^3 \cdot s^{-1}$), liquor density (ρ_{liquor} in $kg \cdot m^{-3}$), acceleration of gravity (g in $m \cdot s^{-1}$), pipe length (L in m),

equivalent pipe length of accidental pressure drops (L_{eq} in m), velocity (V in $m \cdot s^{-1}$), friction factor (f, dimensionless), diameter (D in m), difference in height (Z_1-Z_2, in m) and pump efficiency (η_{pump}). The friction factor (f) was calculated by means of Swamee-Jain expression (Equation 9.3S):

$$f = \frac{0.25}{\left[\log_{10}\left(\frac{k/D}{3.7} + \frac{5.74}{Re^{0.9}}\right)\right]^2}$$ Equation 9.3S

where Re is the Reynolds number, k (m) is the pipe roughness, and D (m) is the pipe diameter.

Equation 9.2S was modified obtaining Equation 9.4S to calculate power requirements for permeate pumping:

$$P_{stage} = \frac{q_{stage} \cdot TMP_{stage}}{\eta_{pump}}$$ Equation 9.4S

where P_{stage} is the power requirement during filtration and back-flushing ($J \cdot s^{-1}$) calculated from average transmembrane pressure (TMP_{stage} in Pa), pumped volumetric flow rate (q_{stage} in $m^3 \cdot s^{-1}$), and pump efficiency (η_{pump}).

Finally, total energy consumption (TEC) was calculated by means on Equation 9.5S by addition of the power requirements of each individual equipment (P_i in kW) per treatment flow (Q_{treat} in $m^3 \cdot h^{-1}$):

$$TEC = \frac{\sum P_i}{Q_{treat}}$$ Equation 9.5S

with TEC in $kWh \cdot m^{-3}$.

9.5.2. Short-term filtration process profile

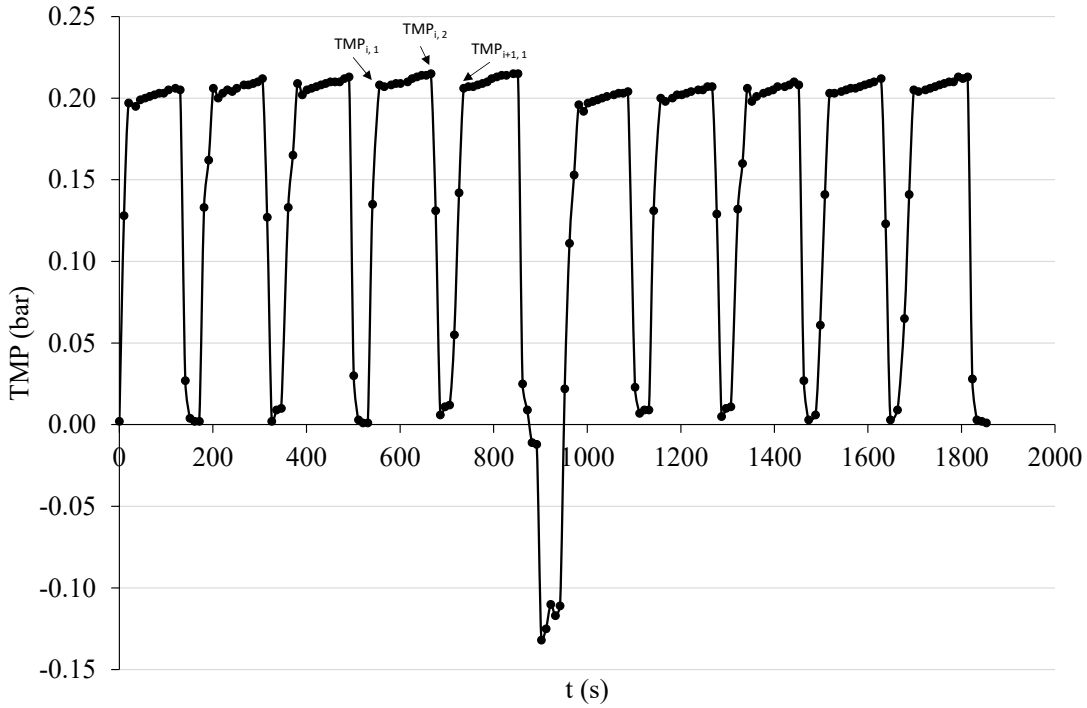

Figure 9.1S. Evolution of TMP in the short term.

9.5.3. Unit costs and correlations functions

Table 9.1S. Unit costs and correlation functions used to calculate CAPEX and OPEX.

Unit costs and correlation function of capital and operating expenses	
Pipes, (final cost is function of diameter) €·m^{-1}	8-220
Polypropylene, €·m^{-2}	9.6
Ultrafiltration hollow-fiber membrane, €·m^{-2}	Membrane = -2.985·ln A^{a} (m^2) + 68.159
Average energy cost, €·kWh^{-1}	0.2
Sodium hypochlorite, €·l^{-1}	0.88
Citric acid, €·kg^{-1}	1.06
Blower, €·unit^{-1}	Blower = 2152.4·ln Q^{b} (m^3/h) -7849.6
Pumps, €·unit^{-1}	Pump = -0.6582·Q$^{2\ b}$ (m^3/h) + 263.54·Q^{b} (m^3/h) + 1005.4

[a] Membrane surface
[b] Air/water flow

9.5.4. Life cycle inventory

Table 9.2S. Materials and processes selected for the life cycle inventory (LCI).

Construction materials, $kg \cdot m^{-3}$	Chromium steel *(1 kg Steel, chromium steel 18/8 {GLO}| market for | APOS, S (Ecoinvent 3))* Cast iron (*1 kg Cast iron {GLO}| market for | APOS, S (Ecoinvent 3))* Polyester resin (*1 kg Polyester resin, unsaturated {GLO}| market for | APOS, S (Ecoinvent 3))* Polypropylene (*1 kg Polypropylene, granulate {GLO}| market for | APOS, S (Ecoinvent 3))* PVC *(1 kg PVC pipe E (Industry data 2.0))* Transport *(1 tkm Transport, freight, lorry 16-32 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3)*	
Energy consumption, $kWh \cdot m^{-3}$, *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Energy avoided (energy recovery from methane), $kWh \cdot m^{-3}$ *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Reagent consumption, $kg \cdot m^{-3}$	Polyelectrolyte *(1 kg Polyacrylamide {GLO}| market for | APOS, S (Ecoinvent 3))* NaOCl *(1 kg Sodium hypochlorite, without water, in 15% solution state {GLO}| market for | APOS, S (Ecoinvent 3))* Citric acid *(1 kg Citric acid {GLO}| market for | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
Discharge to water, $kg \cdot m^{-3}$	Total nitrogen, N_T *(1 kg emissions N_T to water)* Total phosphorous, P_T*(1 kg emissions PT to water)*	
Discharge to soil, $kg \cdot m^{-3}$	Disposal to agriculture *(1 kg Solid manure loading and spreading, by hydraulic loader and spreader {GLO}| market for | APOS, S (Ecoinvent 3))*	NH^{+}_4 to soil *(1 kg emissions NH^{+}_4 to soil through sludge spreading)*
		PO^{-3}_4 to soil *(1 kg emissions PO^{-3}_4 to soil through sludge spreading)*
		N-based fertilizer (*1 kg Nitrogen fertiliser, as N {GLO}| market for | APOS, S)*
		P-based fertilizer (*1 kg Phosphate fertilizer, as P2O5 {GLO}| market for | APOS, S (Ecoinvent 3))*
		N_2O to air *(1 kg emissions N_2O to air through sludge spreading)*
	Disposal to landfill *(1 kg Municipal solid waste {RoW}| treatment of, sanitary landfill | APOS, S (Ecoinvent 3))* Transport *(1 tkm Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
UV tertiary treatment, unit	*1 p Ultraviolet lamp {GLO}| market for | APOS, S (Ecoinvent 3)*	

9.5.5. Contributions to energy consumption

Table 9.3S. Total energy consumption (TEC) and processes contribution to TEC of the projected full-scale UF unit.

		kWh·m^{-3}				
		Feeding pump	**Permeate pump (filtration)**	**Permeate pump (backwash)**	**Blower**	**Total**
Summer	*Sa*	0.010	0.007	0.001	0.181	0.198
	Sb	0.010	0.007	0.001	0.327	0.345
	Sc	0.010	0.009	0.000	0.134	0.153
	Sd	0.010	0.009	0.000	0.239	0.258
Winter	*Wa*	0.010	0.007	0.001	0.113	0.131
	Wb	0.010	0.007	0.001	0.190	0.207
	Wc	0.010	0.009	0.000	0.085	0.105
	Wd	0.010	0.009	0.000	0.140	0.159
		%				
		Feeding pump	**Permeate pump (filtration)**	**Permeate pump (backwash)**	**Blower**	**Total**
Summer	*Sa*	4.8	3.7	0.3	91.1	100.0
	Sb	2.8	2.1	0.2	94.9	100.0
	Sc	6.3	6.0	0.3	87.4	100.0
	Sd	3.7	3.6	0.2	92.5	100.0
Winter	*Wa*	7.3	5.7	0.4	86.5	100.0
	Wb	4.6	3.6	0.3	91.5	100.0
	Wc	9.2	8.8	0.5	81.5	100.0
	Wd	6.0	5.8	0.3	87.8	100.0

9.5.6. CAPEX allocation

Table 9.4S. CAPEX allocation of the projected full-scale UF unit.

		€·m^{-3}						
		MT	**Membranes**	**Permeate pump**	**Feeding pump**	**Membrane blower**	**Pipes**	**Total**
Summer	*Sa*	≈0.000	0.017	0.003	0.003	0.002	≈0.000	0.027
	Sb	≈0.000	0.017	0.003	0.003	0.002	≈0.000	0.027
	Sc	≈0.000	0.015	0.003	0.003	0.002	≈0.000	0.024
	Sd	≈0.000	0.015	0.003	0.003	0.002	≈0.000	0.024
Winter	*Wa*	≈0.000	0.018	0.003	0.003	0.002	≈0.000	0.027
	Wb	≈0.000	0.018	0.003	0.003	0.002	≈0.000	0.027
	Wc	≈0.000	0.015	0.003	0.003	0.002	≈0.000	0.024
	Wd	≈0.000	0.015	0.003	0.003	0.002	≈0.000	0.024
		%						
		MT	**Membranes**	**Permeate pump**	**Feeding pump**	**Membrane blower**	**Pipes**	**Total**
Summer	*Sa*	0.3	65.9	12.1	12.4	8.6	0.8	100.0
	Sb	0.3	65.6	12.1	12.3	8.8	0.9	100.0
	Sc	0.3	62.4	13.4	13.7	9.3	0.8	100.0
	Sd	0.3	62.2	13.3	13.6	9.6	0.9	100.0
Winter	*Wa*	0.3	66.5	12.0	12.2	8.3	0.8	100.0
	Wb	0.3	66.4	11.9	12.2	8.5	0.8	100.0
	Wc	0.3	63.1	13.2	13.5	9.0	0.8	100.0
	Wd	0.3	62.9	13.2	13.5	9.2	0.8	100.0

9.5.7. OPEX allocation

Table 9.5S. OPEX allocation of the projected full-scale UF unit.

		€·m^{-3}							
		Energy consumption	**Chemical cleaning**	**Membrane replacement**	**Permeate pump replacement**	**Feeding pump replacement**	**Blower replacement**	**Equipment maintenance**	**Total**
Summer	*Sa*	0.040	0.001	0.008	0.002	0.002	0.002	≈0.000	0.054
	Sb	0.069	0.001	0.008	0.002	0.002	0.002	≈0.000	0.084
	Sc	0.031	0.001	0.006	0.002	0.002	0.002	≈0.000	0.044
	Sd	0.052	0.001	0.006	0.002	0.002	0.002	≈0.000	0.065
Winter	*Wa*	0.026	0.001	0.008	0.002	0.002	0.002	≈0.000	0.041
	Wb	0.041	0.001	0.008	0.002	0.002	0.002	≈0.000	0.056
	Wc	0.021	0.001	0.007	0.002	0.002	0.002	≈0.000	0.034
	Wd	0.032	0.001	0.007	0.002	0.002	0.002	≈0.000	0.045
		%							
		Energy consumption	**Chemical cleaning**	**Membrane replacement**	**Permeate pump replacement**	**Feeding pump replacement**	**Blower replacement**	**Equipment maintenance**	**Total**
Summer	*Sa*	73.0	2.2	13.8	3.1	3.1	4.1	0.7	100.0
	Sb	82.4	1.4	9.0	2.0	2.0	2.7	0.5	100.0
	Sc	69.7	2.3	14.7	3.8	3.9	4.9	0.9	100.0
	Sd	79.4	1.5	9.9	2.6	2.6	3.5	0.6	100.0
Winter	*Wa*	63.9	3.0	18.8	4.1	4.1	5.3	0.9	100.0
	Wb	73.7	2.2	13.6	3.0	3.0	3.9	0.7	100.0
	Wc	60.9	3.0	19.2	4.8	4.9	6.2	1.1	100.0
	Wd	70.2	2.2	14.5	3.7	3.7	4.8	0.8	100.0

10. General discussion

10.1. Research work motivation

This PhD Thesis aims at contributing to evaluate the environmental and economic feasibility of different ultrafiltration (UF)-based urban wastewater (UWW) treatment processes. This general objective has resulted in the development of several studies and the publication of several scientific articles that explore the advantages, potentialities, and challenges of the processes evaluated to boost the sustainability of UWW treatment. Specifically, the performance of UF membranes has been studied at different treatment steps: primary, secondary (combined with anaerobic treatment in the so-called anaerobic membrane bioreactor – AnMBR) and tertiary treatment. These technological solutions have shown their capacity to improve the recovery and valorization of resources from UWW (mainly reclaimed water, energy, and nutrients), while guaranteeing quality standards in effluent and waste sludge. Among the strengths of the studies conducted, it is worth to highlight: 1) the use of data obtained from demonstration plants equipped with industrial devices; and 2) the dimensioning of the evaluated systems at industrial level using the data from the demonstration scale set-up and operation. Hence, the obtained results are expected to be valuable for the technical and research community to address the potential application of the evaluated processes in large facilities.

10.2. Environmental and economic trends of combining wastewater treatment (aerobic process and/or AnMBR) with fertigation under a circular economy perspective.

Aimed at investigating whether AnMBR technology for reclaiming water for fertigation faces regulatory barriers in the EU, the European Commission (EC) approved the first pilot Innovation Deal (ID) entitled "Sustainable wastewater treatment combining anaerobic membrane technology and water reuse" in 2016. The ID consortium consisted of 14 partners from 5 European member states (Spain, Portugal, Malta, Italy and France) and included a comprehensive group of stakeholders involved in water management: national and regional authorities, universities, research centers, innovators and end-users. Three EC Directorate-Generals (Research and Innovation, Agriculture and Rural Development and Environment) were also represented.

The tasks carried out within the ID included the analysis of several case studies, two of which are presented in this PhD Thesis. These case studies are in the Mediterranean Region (Italy and Spain) and compare the application of combined AnMBR technology and fertigation with conventional configurations of WWTPs with aerobic systems, irrigation with freshwater and mineral

fertilization. The applied methodology included a water balance, a nutrient balance, an economic balance, and a CO_2 emissions assessment.

Results showed that reclaimed water is a reliable resource that can be associated with a relevant renewable nutrient supply (up to 72% of N and 39% of P needs in Spanish case study, and up to 100 % of both nutrients in Italian case study). It is independent of seasonal droughts and weather variations and thus acts as an alternative resource to alleviate pressure on freshwater sources, while reducing the economic and environmental costs of mineral fertilizers (up to 171 k€·year^{-1}). However, a risk management plan is needed to ensure environmental and health safety when reclaimed water is used for both irrigation and fertigation. Results also showed the positive economic balance that can be obtained in different water reuse scenarios (up to 376 k€·year^{-1}). This savings can be used for drawing up, updating and implementing the Water Re-use Risk Management Plans under the responsibility of the new Reclaimed Water Manager, as stated by the Regulation (EU) 2020/741 on minimum requirements for water reuse. (European Comission, 2020).

Since AnMBR technology can also recover energy from the wastewater flow, its application presents an optimal scenario according to Circular Economy principles and environmental sustainability. In this regard, the combination of AnMBR technology and fertigation can significantly reduce CO_2 emissions (up to 898.9 tCO_2·year^{-1}) and reduce wastewater treatment operating cost (up to 63% less than aerobic processes). Last but not least, to comply with the current legislation, future combined AnMBR and fertigation demonstration projects need to be carried out in non-sensitive areas, following the ID conclusions. These trials should include environmental and health risk assessments, agronomic considerations, and social and economic analyses to obtain comprehensive information of the proposed water management scheme.

10.3. Economic and environmental effect of implementing UF membranes as primary treatment.

In this PhD Thesis, the implementation of UF membranes for treating the supernatant from the primary settler of 'Carraixet' WWTP (Valencia, Spain) has been investigated in terms of environmental and economic performance by means of treatment simulation, LCA and LCC. This implementation aimed to divert all the particulate (and colloidal) organic matter to the side-stream AD process, while reducing energy requirements for organic matter removal in the mainstream activated sludge process (i.e., avoiding energy for organic matter oxidation).

A first subset of simulations demonstrated that the UF membranes produce a lack of organic matter which prevent the aerobic process from denitrifying to remove enough nitrogen and, therefore, total nitrogen in the effluent (22.9 mg $N \cdot L^{-1}$) clearly exceeded the value obtained in operation-as-usual scenario (9.7 mg $N \cdot L^{-1}$). Therefore, a new set of simulations was planned to include a bypass stream before the entrance Based on the previous results, it was decided to simulate with high SRT (13 d) and low DO (0.7 $mg \cdot L^{-1}$) to favor X_{NOB} wash-out for different bypass flows, specifically 15, 20 and 25 % of the influent flow. With a 25 % of bypass a similar N_T concentration than in the conventional treatment could be reached.

Results showed that NED in conventional scheme was -0.328 kWh·kg COD_{rem}^{-1} and -0.381 kWh·kg COD_{rem}^{-1} in UF-alternative. Therefore, the latter scheme implies energy savings of 0.053 kWh·kg COD_{rem}^{-1}. OPEX in conventional scheme accounts for €-0.137 kg COD_{rem}^{-1}, while UF-alternative showed lower values for operating expenditure: €-0.150 kg COD_{rem}^{-1}. This represents savings of €0.013 kg COD_{rem}^{-1} compared to conventional treatment. The main contributors to OPEX in the conventional layout were reactor aeration (€0.116 kg COD_{rem}^{-1}) followed by chemical phosphorus precipitation (€0.024 kg COD_{rem}^{-1}), while UF-alternative appeared with two significant associated costs: €0.076 kg COD_{rem}^{-1} for membrane scouring and €0.025 kg COD_{rem}^{-1} for UF membranes replacement. UF-alternative also kept reactor aeration as a significant expenditure, but it was reduced to €0.061 kg COD_{rem}^{-1}.

Regarding CAPEX, UF membranes represents the main cost: €0.099 kg COD_{rem}^{-1} and 94.7 % of CAPEX. TAC in conventional treatment is €0.092 kg COD_{rem}^{-1} lower than in UF-alternative. Considering that energy balance and OPEX in UF-alternative are favorable compared to conventional layout, CAPEX shows that membrane cost is the main bottleneck for the implementation of membrane-based alternative, since operating savings are not able to offset membrane investment of €0.099 kg COD_{rem}^{-1} and membrane replacement cost of €0.025 kg COD_{rem}^{-1}. However, an increase in the durability of the membranes, a decrease in the cost of the membrane, an increase in the cost of energy, and an improved in transmembrane flux would improve the economic feasibility of the UF-membrane alternative.

Concerning global warming potential, estimation of N_2O emissions associated to nitrification-denitrification process appeared as main contributor to equivalent CO_2 emissions, accounting for 91 % and 87 % in conventional treatment and UF-alternative, respectively. Overall, UF-alternative allowed reducing CO_2 emissions compared to conventional treatment due to the

reduction of the total nitrogen load entering the aerobic reactor (and therefore the amount of nitrogen that eventually form N_2O during nitrification-denitrification process).

Direct emission of phosphorus to water bodies appeared as major contributor to freshwater eutrophication (98 %). Similarly, nitrogen emissions to water with the effluent represented the main contribution to marine eutrophication with approx. 97-98 % of total impact in both scenarios. Results of both eutrophication impact categories (marine and freshwater eutrophication) confirmed nutrient recovery and reuse as crucial processes to improve environmental performance of UWW treatment.

Finally results of fossil resource scarcity illustrates the opportunities that UWW represents to face global resource depletion. Energy and nutrient recovery embedded in it can contribute to preserve conventional sources and turn UWW into a supply stream for social and economic purposes. Therefore, it could be expected that environmental performance of the water treatment would improve by taking advantage of these resources, e.g., by means of fertigation of agriculture periurban crop fields (Jiménez-Benítez et al., 2020).

10.4. Biological performance of AnMBR technology for UWW treatment

The long-term biological performance of an AnMBR demo-plant was evaluated. As already mentioned, AnMBR technology shows some advantages over conventional activated sludge for UWW treatment that makes it a suitable candidate for renewing the UWW treatment approach and contributing to the economic, environmental, and social goals of water management, with Circular Economy playing an active role.

The size of the evaluated demo-plant, which treated real UWW for more than 2.5 years, allowed obtaining a robust source of performance data. It is important to highlight the variable quality and the relatively high chemical oxygen demand (COD) and biological oxygen demand (BOD) concentrations in the influent due to the contributions of nearby dairy and wine industries. Sludge retention time (SRT) was maintained at 70 days and the plant was operated at two different hydraulic retention time (HRT) (25-26 and 41 hours). Temperature varied freely between 10 and 27 ºC (ambient temperature operation), while organic loading rate (OLR) was in the range between 0.60±0.20 and 1.28±0.38 kg $COD \cdot m^{-3} \cdot d^{-1}$.

COD removal accounted for an average of 87.2±6.1 % of the influent COD and the membranes were able to virtually retain all particulates. Among the evaluated scenarios, the highest methane yield (Y^{CH_4}) value (169.0±95.1 STP L $CH_4 \cdot kg^{-1}$ COD) was obtained with a combination of high T (27±1 ºC), HRT (41±1 h) and COD:SO_4^{2-}-S ratio (8.70±3.69 COD·g^{-1} SO_4^{2-}-S). While a high temperature enhanced the hydrolysis rate, methane production increased with the COD available for methanogenic archaea (MA). The obtained lowest Y^{CH_4} values (75.2±16.9 and 70.2±36.0 STP L CH4·kg^{-1} COD) can also be explained by the lower temperature, HRT and COD:SO_4^{2-}-S ratio of the corresponding scenarios. Therefore, in sulphate rich wastewater, the role of sulfate-reducing bacteria (SRB) in reducing the effluent-soluble COD when HRT is reduced must be considered, since SRB has higher degradation rates than MA, especially at lower temperatures.

With regards to waste-sludge production (WSP) results showed a complex interaction between operational conditions and influent characteristics. The highest WSP (0.166±0.074 kgVSS·kg^{-1} COD_{rem}) was obtained with low operating temperature (19±1 °C) since this operating condition hindered biodegradability and therefore raised sludge production. The lowest WSP among the studied scenarios (0.121±0.067 and 0.103 ±0.044 kg VSS·kg^{-1} COD_{rem}) were obtained at high temperature (27±1 °C and 24±2 °C, respectively) and COD:SO_4^{2-}-S ratio (8.70±3.69 g COD·g^{-1} SO_4^{2-}-S and 8.15±4.44 g COD·g^{-1} SO_4^{2-}-S). The combination of both variables enhanced MA organic matter biodegradation and reduced sludge production. It is important to highlight that, besides the complex interaction of factors that affect the WSP, it can be as low as 0.103±0.044 kgVSS·kg^{-1} COD_{rem}).

Therefore, temperature, HRT and COD:SO_4^{2-}-S ratio jointly with SRT are crucial parameters to take into account in maximizing AnMBR's technical, economic and environmental performance. However, since temperature and influent COD:SO_4^{2-}-S ratio depend on ambient conditions, HRT could be considered the main design parameter jointly with SRT.

10.5. Filtration performance, energy consumption and GHG emissions of AnMBR technology for UWW treatment.

During the development of this PhD Thesis, the long-term operation of an AnMBR demo-plant was also evaluated focusing on filtration performance, energy balance and GHG emissions.

Membrane filtration and scouring took place within the following parameter range: the gross 20ºC-standardized transmembrane flux (J_{20}) was modified between 15 and 25 LMH and the specific gas demand per m^2 of membrane (SGD_m) was between 0.10 and 0.40 $Nm^3 \cdot m^{-2} \cdot h^{-1}$, resulting in a specific gas demand per permeate volume (SGD_P) of between 10 and 20 $Nm^3 \cdot m^{-3}$.

Results showed the complex interaction between J_{20}, SGD_m, total suspended solids (TSS) in the reactor, transmembrane pressure (TMP) and the evolution of fouling. Operation at high J_{20} with low fouling propensities can be achieved with high SGD_m and moderate mixed liquor TSS concentrations. The results showed that the system was able to conduct filtration at a moderate-high J_{20} (15–20 LMH) with competitive SGD_m (at an SGD_P of 10–20 $Nm^3 \cdot m^{-3}$) and moderate TSS (8–13 $g \cdot L^{-1}$), although shifts between supra-critical and subcritical filtration were seen to occur with small changes in the operating conditions. Operating at sub-critical filtration conditions (recommended to avoid quick TMP increase) was possible with TSS concentrations below 10 $g \cdot L^{-1}$, maximum J_{20} of 16 LMH and SGD_m of 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to 15 $Nm^3 \cdot m^{-3}$). Fouling control could be achieved even at high TMP (470–500 mbar). Finally, it should be noted that the demo plant accomplished a successful long-term operation without chemical cleaning requirements, the longest operation period being 233 days, although the results indicated that longer operating periods could be reached. Irrecoverable fouling was not detected since chemical cleaning returned the TMP to the initial values or even lower.

With regards to energy balance, the main contributions to total energy consumption (TEC) were associated with the blower requirements for membrane scouring (55.2 %), followed by reactor mixing (34.4 %). Overall, average TEC accounts for 0.192 ± 0.070 $kWh \cdot m^{-3}$ while average total energy recovery (TER) (considering 0 % of methane recovery) was 0.361 ± 0.271 $kWh \cdot m^{-3}$. Dissolved methane produced more energy than that used in its recovery, although its contribution to TER was relatively low (between 0.02 $kWh \cdot m^{-3}$ and 0.04 $kWh \cdot m^{-3}$ for dissolved methane recovery ratios of 50% and 70 %, respectively). However, this recovery definitively reduced GHG emissions. These results emphasize the importance of optimizing the parameters governing the reactor mixing requirements and filtration, especially regarding SGD_m. Increasing sludge recycling would reduce TSS concentration in the membrane tank and help to reduce SGD_m necessities.

When no methane recovery was considered, the contribution of dissolved methane (D-CH_4) to total emissions accounted for 0.5–0.6 $kgCO_{2eq} \cdot m^{-3}$ during almost the entire period of plant operation. Recovering 50 % of the methane (economic optimum according to Sanchis-Perucho et

al. (2020)) would reduce the equivalent CO_2 emissions to 0.1–0.3 $kgCO_{2eq}·m^{-3}$. Operating with 70 % methane recovery (environmental optimum according to Sanchis-Perucho et al. (2020)) kept the emissions below 0.2 $kgCO_{2eq}·m^{-3}$. By way of comparison, Rodriguez-Garcia et al. (2011) estimated emissions of around 0.3 $kgCO_{2eq}·m^{-3}$ for CAS facilities with organic matter and nutrient removal discharging in sensitive areas, while Chen (2019) estimated small-scale MBR (351 $m^3·d^{-1}$ treatment capacity) emissions to be around 0.7 $kgCO_{2eq}·m^{-3}$.

Finally, it is important to highlight the additional benefits that recovering nutrients would entail, since N and P production from traditional sources require high power consumption and GHG emissions. McCarty et al. (2011) reported 19.3 kWh per kg N and 2.11 kWh per kg P. According to these values, considering the average N and P content in the effluent (45.7 ± 10.4 mg $N·L^{-1}$ and 7.8 ± 2.0 mg $P·L^{-1}$, data not shown) and a GHG emission factor for Spanish energy mix generation of 0.14 kg CO_2·kWh, fertigation would allow savings of 0.126 kg $CO_{2eq}·m^{-3}$, which would help the combination of AnMBR+fertigation to achieve carbon neutrality if 70 % of the dissolved methane was recovered.

Overall, the AnMBR demo plant showed a suitable filtration performance, significant energy productions and low CO_2 emissions when recovering dissolved methane. These results indicate that AnMBR combined with degassing membranes represents a robust alternative to the conventional UWW treatment.

10.6. Economic sustainability of a full-scale AnMBR for UWW treatment.

Based on the results from the AnMBR demo-plant, this PhD Thesis deals with an upscaling and economic sustainability evaluation of a projected full-scale AnMBR (treatment flow 16000 $m^3·d^{-1}$) by means of life cycle costing (LCC) methodology selecting 1 kg of COD removed as functional unit. LCC was applied to 5 selected scenarios to calculate the capital and operating expenses (CAPEX and OPEX, respectively) and the total annual costs (TAC), Moreover, a sensitivity analysis (SA) and an uncertainty analysis (UA) were carried out to determine the most significant parameters and the related uncertainty in the obtained results.

First, an energy balance was carried, evaluating the TEC, TER and net energy demand (NED) of the projected full-scale AnMBR plant. Results showed that the main contributors to TEC in all

the evaluated scenarios were reactor mixing (34–57%) and membrane scouring (22–48%). Favorable energy balances were achieved in all the scenarios evaluated. NED varied between -0.210 kWh·kgCOD$_{rem}^{-1}$ and -0.645 kWh·kgCOD$_{rem}^{-1}$. Reactor mixing (0.056–0.124 kWh·kgCOD$_{rem}^{-1}$) and membrane scouring (0.048–0.120 kWh·kgCOD$_{rem}^{-1}$) showed the highest energy consumption. The main drivers of the NED results are the OLR and COD removal capacity, HRT, temperature, and the COD:SO_4^{2-}-S ratio. Optimizing HRT to reduce reactor mixing expenditures and designing a comprehensive filtration strategy able to maintain low TMP are also crucial to reducing TEC.

The major contributors to CAPEX were UF membranes (€0.029–0.073 kgCOD$_{rem}^{-1}$; 31–49%), reciprocating engines acquisition (€0.012–0.023 kgCOD$_{rem}^{-1}$; 8–24%), and reactor building (€0.007–0.014 kgCOD$_{rem}^{-1}$; 8–13%). OPEX was mainly influenced by equipment energy needs (€0.042–0.069 kgCOD$_{rem}^{-1}$; 41–46%), UF membrane replacement (€0.011–0.028 kgCOD$_{rem}^{-1}$; 9–17%) and discharge fee (€0.010–0.020 kgCOD$_{rem}^{-1}$; 9–12%). Combined heat and power (CHP) systems maintenance and replacement was also a significant operating cost (€0.009–0.017 kgCOD$_{rem}^{-1}$; 6–14%), although it finally provided revenues through energy sale (€0.120–0.222 kgCOD$_{rem}^{-1}$). The TAC varied between -€0.003 and €0.188 kgCOD$_{rem}^{-1}$, with CAPEX as the main cost.

The results obtained from LCC assessment combined with SA and UA showed that AnMBR represents a potential alternative to conventional aerobic treatments. Its definitive impulse will go through, among other aspects, reinforcing its economic sustainability, for which it is necessary to optimize operational aspects (e.g., HRT, transmembrane flux, reactor mixing and membrane sparging) and technological developments that allow increasing membrane lifespan and improve CHP power efficiency. Finally, other aspects that would also result in improving its economic performance have to do with nutrient recovery present in its effluent and favorable pricing for the sale of electricity produced from the methane generated.

10.7. Environmental sustainability of a full-scale AnMBR for UWW treatment.

Based on the results from the AnMBR demo-plant, this PhD Thesis also deals with an environmental sustainability evaluation based on the life cycle assessment (LCA) methodology of a projected full-scale AnMBR (treatment flow 16000 $m^3 \cdot d^{-1}$). LCA followed ISO 14040:2006

recommendations and included construction, operation, and dismantling phases. 1 kg of COD removed was selected as functional unit and a comprehensive life cycle inventory was constructed for each phase by means of Ecoinvent data base. LCA was conducted based on the ReCiPe 2016 method and a hierarchist approach. Finally, LCA was calculated for 5 operating scenarios and SA and UA were carried out.

Energy balance calculated for a previous study included in this PhD Thesis (Jiménez-Benítez et al., 2023) was expanded to include a higher recovery rate (80 %), according to the environmental optimum proposed by Sanchis-Perucho et al. (2020). All the evaluated scenarios performed as energy producers, presenting NED in the -0.254:-0.668 $kWh \cdot kgCOD_{rem}^{-1}$ range. AnMBR, thus, appears as a potential alternative to conventional treatments at moderate temperatures for medium-high strength UWW from the energy perspective.

With regards to global warming potential, the operating phase was the main contributor to this impact category, representing 87-92 % of the direct environmental impact Most of the direct environmental load in this category was related to fugitive methane emissions due to downstream stripping of dissolved methane in the effluent (40-55 %). Reduction of AnMBR carbon footprint therefore necessarily involves improving dissolved methane recovery. In turn, recovering the dissolved methane to produce energy would improve the energy balance. Overall, results for global warming potential were in the -0.223:0.020 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ range. Rodriguez-Garcia et al. (2011) evaluated the operation of different WWTPs, obtaining values between 0.206 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ and 0.551 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ for facilities without nutrient removal processes and discharging to non-sensitive areas and. between 0.413 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ and 0.688 $kgCO_{2eq} \cdot kgCOD_{rem}^{-1}$ for conventional activated sludge with nutrient removal and discharging to sensitive areas. It can thus be concluded that AnMBR can be regarded as an alternative treatment to reduce GHG emissions in sewage management when dissolved methane in the effluent is captured.

Mineral resource scarcity was mainly influenced by NED and nutrient recovery through sludge spreading due to avoiding production of P-based and especially N-based fertilizers. It should also be mentioned that the construction phase composed between 51 % and 71 % of the direct mineral resource scarcity life cycle impact, when depletion offsets due to energy, nutrient recovery and demolition and recycling were not considered. Exclusion of construction phase from LCA should be therefore carefully evaluated. The environmental impact on fossil resource scarcity followed a similar pattern to the global warming potential. NED was thus the most important factor in the

fossil resource scarcity impact category. Even though the operation phase was the major contributor to the fossil resource scarcity input, the construction phase burden forms 18-27 % of the direct load. It is also remarkable that both net mineral and fossil resource depletion obtained negative loads in all the scenarios evaluated, which indicates that AnMBR contributes to preserving these resources.

Effluent characterization was the main factor affecting both freshwater and marine eutrophication due to its nitrogen and phosphorus content. Finally, it is important to highlight that, although the nutrient concentrations of the effluents from anaerobic treatments present a potential for eutrophication to be considered, it is also true that they represent an opportunity to recover these resources in considerable quantities, which entails obvious environmental and economic benefits.

With regards to water consumption, the results indicated that reducing NED through biogas and dissolved methane recovery had a positive effect on the environmental loads. On the basis of the results, it could be also stated that taking advantage of nutrient content and thus avoiding P-based and especially N-based fertilizer production also contributed to a relevant reduction of the total impact in this category. It is important to highlight that water reuse was not considered. Including this practice in the LCA boundaries would prevent freshwater extraction, thus improving this impact category. The construction phase weight in the direct water consumption category made up 21-28 % of its total life-cycle impact Demolition also had a significant effect and contributed to reducing the water consumption burden in all the scenarios.

Finally, SA and UA showed energy recovery, heavy metals in sludge, dissolved methane in the effluent, and effluent nutrient content as the most important aspects affecting LCA outcome. Moreover, CHP power efficiency, dissolved methane recovery potential, filtration productivity, membrane scouring, reactor mixing, HRT and SRT appeared most influencing parameters. These results showed that actions leading to increase the recovery and valorization of dissolved methane and/or the recovery of nutrients through, for instance, fertigation, improve the environmental performance of AnMBR for UWW treatment.

10.8. Economic and environmental effect of implementing UF membranes as tertiary treatment.

This PhD Thesis has also evaluated the feasibility of using UF units for water reclamation in terms of energy balance, environmental (LCA) and economic (LCC) performance and reclaimed water quality.

Results of filtration performance of the UF pilot plant suggested that the UF system should be operated under different conditions in summer and winter to prevent excessive fouling, i.e., at J of around 28-33 LMH and SAD_m of 0.4-0.6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ in summer and around J of 33-39 LMH and SAD_m of 0.3-0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ in winter.

Energy assessment of the up-scaled projected UF treatment (14000 $m^3 \cdot d^{-1}$) showed that the TEC under summer operating conditions was 0.239±0.083 $kWh \cdot m^{-3}$ on average while results for winter period was 0.151±0.044 $kWh \cdot m^{-3}$. Membrane blowers account for 87-92 % of the TEC, while feeding pumping and permeate pumping contributed with 3-9 % of and 2-9 %, respectively. Therefore, efforts to reduce energy expenditure should be focused on diminishing membrane scouring needs. To this aim, an appropriate filtration strategy should be adopted to guarantee significant transmembrane fluxes, which eventually reduce membrane surface needs.

With regards to CAPEX, membrane acquisition appeared as the main contributor, accounting for 62-66 % in summer and 63-67 % in winter scenario. The lower $J_{20,g}$ the higher CAPEX, since membrane requirements depend on filtration productivity. On average, summer scenarios showed a CAPEX of €0.025±0.001$\cdot m^{-3}$ and winter scenario €0.026±0.001$\cdot m^{-3}$, which represents no-significant differences. Expressed in costs, the largest investment corresponds to the membranes, €0.016±0.001$\cdot m^{-3}$ and €0.017±0.001$\cdot m^{-3}$ (summer and winter, respectively). Permeate pumps and feeding pumps account for €0.003±0.000$\cdot m^{-3}$ both in summer and winter scenarios (13±1 % each), while blowers' contribution was calculated in €0.002±0.000$\cdot m^{-3}$ (9 %).

OPEX varied in summer scenario between in the €0.044-0.084$\cdot m^{-3}$ range, while in winter scenario operating expenditure were between €0.034 and €0.056$\cdot m^{-3}$. The main contributor to OPEX was TEC, with an average cost of €0.048±0.017$\cdot m^{-3}$ in summer and €0.030±0.009$\cdot m^{-3}$ in winter. These costs represent 76±6 % and 67±+6%, respectively. UF membrane replacement was also a

significant OPEX cost. It accounted for €0.007±0.001·m^{-3} in summer and winter (12±3 % and 17±3 %, respectively).

TAC varied between €0.068 and €0.110·m^{-3} in summer while in winter it varied between €0.059·m^{-3} and €0.083·m^{-3}. On average, TAC in summer conditions was €0.087±0.018·m^{-3} and 0.070±0.010·m^{-3} in winter scenario. Main contributor to TAC was OPEX, which account for 70±5 % in summer and 63±4 % in winter scenario.

LCA outcome showed operation as the main influencing phase for global warming potential, water consumption, fossil resource scarcity and mineral resource scarcity (90-98 %). Construction phase was significant only for the latter category (8-10 %). Within operation phase, energy consumption showed the highest influence (64-95 %). Therefore, improving the environmental performance of the application of UF membranes for tertiary treatment necessarily involves filtration optimization focused on reducing energy consumption.

Finally, results demonstrate that UF process as tertiary treatment allows producing a high-quality effluent capable of accomplishing the stringent microbiological and physis-chemical requirements set by the Regulation (EU) 2020/741 and Spanish Royal Decree 1620/2007.

10.9. Boosting feasible alternatives to achieve sustainable UWW treatments.

Membrane technology and the anaerobic treatments for wastewater are contributing to improving the sustainability of the wastewater management (Faragò et al., 2021). In line with Circular Economy principles, UF membranes and anaerobic treatment applied to wastewater treatment allow boosting resource recovery, leveraging its energy and nutrient content and favoring water with positive environmental and economic impacts reuse. However, some aspects still represent significant challenges that need to be addressed.

Energy balance appears as the main influencing factor for both economic and environmental feasibility of UF membrane and AnMBR. Since AD can contribute to counterbalance energy consumption, promoting methane production and valorization through anaerobic processes represents an opportunity to enhance the sustainability of UWW treatment. In this sense, co-

digestion of UWW, agrifood industry waste and food waste shows promising synergies (Liu et al., 2023; Moñino et al., 2017).

Fouling mitigation implies high energy consumption for membrane scouring. Fouling also reduces membrane lifespan when membrane cleanings increase. Therefore, fouling negatively affects economic and environmental performance of membrane technologies. An adequate design of the filtration strategy would reduce these negative effects (Shahid et al., 2020).

Dissolved methane in the AnMBR effluent, if it is not properly recovered or removed, may hinder the adoption of AnMBR as a core technology in treatment facilities, due to the potential greenhouse effect that its release into the atmosphere would entail. Fortunately, technologies such as degassing membranes or and spray aeration towers have shown their potential to address this issue (Henares et al., 2017).

The AnMBR effluent also has a high nutrient content, the discharge of which into sensitive water bodies could cause eutrophication problems. However, it is precisely this nutrient content that gives added value to this stream, given the economic and environmental potential that its reuse implies and, therefore, the reduction of dependence on industrial fertilizers (Ye et al., 2020).

Membrane cost significantly contributes to CAPEX and OPEX (trough membrane replacement necessities). A reduction of this expenditure, coming from improving the filtration performance and therefore diminishing membrane requirements or extending its lifespan; or due to a membrane cost reduction, would facilitate the adoption of membrane technology for UWW treatment and water reclamation (Das et al., 2022; Judd, 2017).

Finally, this PhD Thesis has demonstrated the potential of UF membranes and its combination with AD to contribute to UWW treatment sustainability. Overcoming the aforementioned challenges would lay the foundations for a transition of current treatment systems towards processes that are more respectful of the environment and less burdensome for the economy.

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11. General conclusions

This PhD Thesis aimed to elucidate whether the implementation of ultrafiltration (UF) membrane technology at primary (direct membrane filtration, DMF), secondary (combined with anaerobic digestion in the so-called anaerobic membrane bioreactor, AnMBR), or tertiary (tertiary UF filtration) treatment steps presents a favorable environmental and economic performance to contribute to advance sustainability within urban wastewater (UWW) treatment field. The main features studied in this PhD Thesis focused on modelling and designing full-scale systems based on the technological solutions studied from the application of life cycle assessment and life cycle costing methodologies. From this, the following key conclusions can be drawn:

AnMBR, reclaimed water and fertigation: two case studies in Italy and Spain to assess economic and technological feasibility and CO_2 emissions within the EU Innovation Deal initiative.

- AnMBR technology appears as a feasible technology for valorizing urban wastewater and its combination with fertigation can contribute to significant reductions of CO_2 emissions, up to 898.9 $tCO_2 \cdot year^{-1}$ in the studied scenarios.
- Reclaimed water can be considered a reliable water and nutrient supply source with positive economic balance for water and nutrients reuse for agriculture, with savings up to 376 $k€ \cdot year^{-1}$ and nutrients supply up to 100 % of nitrogen and phosphorus needs.
- Water reuse risk management plans are needed to ensure environmental and health safety.

Ultrafiltration after primary settler to enhance organic carbon valorization: energy, economic and environmental assessment.

- Combining primary settling and UF-based direct membrane filtration (DMF) can be effectively applied to derive organic carbon from the water to the sludge line, although it can prevent nitrification-denitrification in a downstream activated sludge system, thus compromising effluent quality in terms of nitrogen concentration. A bypass stream between primary settling and UF-based DMF can be used to ensure carbon requirements for nitrification-denitrification in downstream activated sludge systems.

- Promoting partial nitrification reduces carbon requirements for denitrification, which allows increasing the derivation of organics to the sludge line while meeting nitrogen effluent discharge requirements. To this aim, nitrite-oxidizing bacteria (X_{NOB}) must be washed out from the activated sludge system. Model predicted data showed that operating at high SRT (13 d) and low dissolved oxygen concentration (0.7 $mg \cdot L^{-1}$) allows removing X_{NOB} while ammonia-oxidizing bacteria remain in the reactor.
- Combining primary settling and UF-based DMF may allow increasing energy recovery from UWW by 20.4 %, resulting in energy savings of 0.053 kWh per kg of COD_{rem} compared to a conventional WWTP scheme.
- This alternative treatment showed OPEX savings of €0.013 kg COD_{rem}^{-1} compared to conventional scheme. Main contributors to OPEX were €0.076 kg COD_{rem}^{-1} for membrane scouring, €0.061 kg COD_{rem}^{-1} for reactor aeration and €0.025 kg COD_{rem}^{-1} for UF membranes replacement. UF membranes represents the main cost of CAPEX: €0.099 kg COD_{rem}^{-1} and 94.7 %. TAC in conventional scheme is €0.092 kg COD_{rem}^{-1} lower than in UF-alternative.
- Environmental impacts slightly decreased for all impact categories when comparing the alternative solution to a conventional WWTP.

A semi-industrial scale AnMBR for municipal wastewater treatment at ambient temperature: performance of the biological process

- Adequate long-term performances of AnMBR technology are reached at ambient temperature with effluent chemical oxygen demand (COD) concentrations below discharge effluent standards between 79±18 $mg \cdot L^{-1}$ and 121±15 $mg \cdot L^{-1}$ (e.g., average COD removal efficiencies above 87 % and solids free).
- A reduction of waste sludge between 9 and 43 % compared to conventional activated sludge (CAS) process can be achieved.
- Adequate methane productions with methane yield ranged from 70 to 169 STP L $CH_4 \cdot kg^{-1}$ COD_{inf} (corresponding to 198-727 STP L $CH_4 \cdot kg^{-1}$ VSS_{inf}) were obtained, higher than methane yields of 0.19-0.24 $Nm^3 \cdot kg^{-1}$ VSS_{inf} reported for CAS and AD of wasted sludge (Di Capua et al., 2020).

- Temperature, hydraulic retention time (HRT) and influent COD:SO_4^{2-}-S ratio (jointly with sludge retention time (SRT), not tested in this study), are crucial parameters affecting AnMBR performance. Low temperature was associated with lower COD degradation and methane production. The opposite effect was obtained with high HRT and COD:SO_4^{2-}-S ratio, since it favored methanogenic archaea metabolism.

A semi-industrial AnMBR plant for urban wastewater treatment at ambient temperature: analysis of the filtration process, energy balance and quantification of GHG emissions.

- AnMBR can operate as net energy producer (energy surplus of up to 0.169 $kWh \cdot m^{-3}$ in average) operating at competitive 20ºC-standardized transmembrane flux (J_{20}), up to 15-20 LMH, and low specific gas demand per m^2 of membrane (SGD_m) values (0.1-0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$), while achieving low fouling propensities and long operating periods between membrane chemical cleanings (up to 233 days).
- Average energy consumption was estimated at 0.192±0.070 $kWh \cdot m^{-3}$, with most of the power consumed by membrane scouring (55 %) and reactor mixing (34 %). The achieved total energy recoveries (TER) varied between 0.361±0.271 and 0.397±0.377 $kWh \cdot m^{-3}$ when energy recovery from dissolved methane was set to 0 % and 70 %, respectively. Net energy demand (NED) during the studied period ranged between -0.169±0.341 (0% of dissolved methane recovered) and -0.205±0.447 $kWh \cdot m^{-3}$ (70 % of dissolved methane recovered), which is significantly lower than CAS without nutrient removal (0.01-0.31 $kWh \cdot m^{-3}$).
- Dissolved methane recovery appears as the key challenge to be overcome for achieving low GHG emissions with AnMBR. In this study, recoveries of 50 % reduced emissions from 0.5-0.6 to 0.1-0.3; and recoveries of 70 %, below 0.2 $kgCO_2 \cdot m^{-3}$, lower than CAS (0.3 $kgCO_{2eq} \cdot m^{-3}$). Further reductions could be possible by increasing membrane surface or implementing spray aeration towers.

A Life cycle costing of AnMBR technology for urban wastewater treatment: a case study based on a demo-scale AnMBR system.

- Energy balance and life cycle costing (LCC) of the projected AnMBR showed promising results.
- Net energy productions of up to 0.645 kWh·kgCOD_{rem}^{-1} were achieved.
- Capital expenses (CAPEX) varied between €0.080 and 0.147 per kg of COD_{rem}, with UF membranes (€0.029-0.073 kgCOD_{rem}^{-1}; 31-49 %), reciprocating engines acquisition (€0.012-0.023 kgCOD_{rem}^{-1}; 8-24 %), blowers (€0.012-0.020 kgCOD_{rem}^{-1}; 13-15 %) and reactor building (€0.007-0.014 kgCOD_{rem}^{-1}; 8-13 %) as main contributors.
- Operating expenses (OPEX) varied from €-0.099 to 0.041 per kg of COD_{rem}, mainly influenced by equipment energy needs (€0.042-0.069 kgCOD_{rem}^{-1}; 41-46 %), UF membrane replacement (€0.011-0.028 kgCOD_{rem}^{-1}; 9-17 %), reciprocating engines operation (0.009-0.017 kgCOD_{rem}^{-1}; 5-14 %) and discharge fee (€0.010-0.020 kgCOD_{rem}^{-1}; 9-12 %)
- TAC between €-0.003 and 0.188 kgCOD_{rem}^{-1} were obtained.
- Optimizing operational aspects (e.g., HRT, J_{20}, reactor mixing or gas sparging for membrane scouring) and technological developments allowing to increase membrane lifespan and improve CHP power efficiency would improve the economic performance of AnMBR technology.

Life cycle assessment of AnMBR technology for urban wastewater treatment: a case study based on a demo-scale AnMBR system.

- Life cycle assessment (LCA) outcome showed positive environmental performance of AnMBR for urban wastewater (UWW) treatment.
- Energy and nutrients recovery were identified as key factors for improving the environmental performance of AnMBR for UWW treatment at full scale. Emissions of

methane and N_2O, and nitrogen and phosphorus release to soil and water bodies are major drivers for environmental impacts, jointly with heavy metal content in wasted sludge.

- Degassing membranes for dissolved methane capture appear as a key technology to enhance AnMBR environmental performance, while post-treatment technologies for nutrient recovery/removal (e.g., microalgae cultivation, anammox, ion exchange, membrane contactors, fertigation etc.) could be necessary for reducing marine and freshwater eutrophication.
- Construction phase contribution to overall LCA was relevant in some scenarios and/or impact categories. Therefore, its exclusion from the LCA system boundary should be carefully evaluated.
- Improvement of the environmental performance of AnMBR technology involves increasing energy recovery efficiency, optimizing filtration process performance (e.g., J_{20} and membrane scouring), reactor mixing, HRT and SRT.

Effect of seasonal dynamics on water reclamation from ultrafiltration membrane-based tertiary urban wastewater treatment: energy expenditure, cost, environmental and effluent quality assessment.

- TEC of UF-based tertiary treatment of UWW resulted in 0.239±0.083 and 0.151±0.044 $kWh \cdot m^{-3}$ in average for summer and winter periods, respectively, with membrane blowers (i.e., gas sparging for membrane scouring) as main contributor (87-92 %).
- CAPEX resulted in €0.025±0.001 and €0.026±0.001 per m^3 in summer and winter, respectively (negligible differences were identified according to seasonal dynamics), being membrane acquisition the main contributor (62-67 %). OPEX ranged between €0.044 and 0.084 per m^3 in summer, while it varied between €0.034 and €0.056 per m^3 in winter. The main contributor to OPEX was TEC: 76±6 % (summer) and 67±+6% (winter).
- TAC resulted in €0.087±0.018 and 0.070±0.010 per m^3 in summer and winter, respectively. Main contributor to TAC was OPEX, which accounted for 70±5 % in summer and 63±4 % in winter scenario.

- Results demonstrated that UF-based tertiary treatment allows producing a high-quality effluent capable of accomplishing the stringent microbiological and physis-chemical requirements set by the Regulation (EU) 2020/741 and the Spanish Royal Decree 1620/2007.

12. Appendix I: Resumen extendido

Introducción

La emergencia climática y ambiental que enfrenta la sociedad supone un reto que debe ser abordado desde diferentes ámbitos y perspectivas. Entre las posibles opciones de actuación, la recuperación de los recursos presentes en las aguas residuales se considera una estrategia prometedora que respeta los fundamentos de la Economía Circular. Al amparo de este modelo económico, se requiere de un cambio conceptual que modifique la percepción de las aguas residuales urbanas, pasando de ser consideradas un residuo a contemplarse como una fuente de recursos.

El recurso más obvio presente en las aguas residuales es el agua misma. El agua regenerada y su reutilización muestran algunas ventajas significativas para abordar los problemas del cambio climático y la sequía:

- Garantía de disponibilidad: en aquellos lugares en los que suministro de agua potable es la máxima prioridad dentro de los sistemas de gestión de recursos hídricos, por ejemplo, en España, las aguas residuales urbanas muestran un flujo regular hacia la EDAR. Tras el tratamiento, el efluente está disponible durante todo el año y contribuye a proporcionar un suministro fiable de agua con menores riesgos en caso de escasez o sequía.
- Menor coste que otras alternativas: la reutilización del agua ya se considera una medida de menor coste para aumentar los recursos hídricos disponibles que la desalinización o la transferencia de agua entre cuencas fluviales bajo ciertas condiciones.
- Prevención de conflictos regionales: la reutilización del agua, como estrategia para conservar las fuentes convencionales de agua, también puede contribuir a reducir la competencia por los recursos y aliviar los conflictos hídricos.
- Seguridad alimentaria: una mayor disponibilidad de agua ayuda a garantizar la producción agrícola y, por tanto, la seguridad alimentaria. En este sentido, la agricultura periurbana, entendida como los terrenos de cultivo ubicados en los entornos urbanos, puede beneficiarse enormemente de la regeneración y reutilización de agua, al estar cerca de las plantas de tratamiento.
- Empleo y comercio local: el agua representa un insumo esencial para la actividad económica y por tanto para la creación de empleo. El aumento de los recursos hídricos y la promoción de la innovación tecnológica actuarían como motor de desarrollo para elevar los indicadores de empleo y sostenibilidad social.

- Preservación de los recursos hídricos convencionales: la reutilización del agua reduce las extracciones de fuentes convencionales, por ejemplo, aguas subterráneas, ríos o lagos. Por tanto, el agua regenerada contribuye a reducir el agotamiento de las reservas de agua dulce y favorece la restauración de los ecosistemas acuáticos.
- Calidad adecuada según el propósito: la tecnología de regeneración de aguas permite producir calidades adecuadas según el propósito de uso. Este enfoque permite ajustar los costes de producción al destino del agua y, por tanto, mejorar la sostenibilidad del sistema de regeneración.
- Beneficios ambientales: la reutilización del agua presenta beneficios ambientales, entre los que se pueden mencionar: i) reducción de vertidos a masas de agua; ii) cuando se considera la fertirrigación se reduce la producción de fertilizantes minerales y con ello sus impactos ambientales asociados; y iii) menor huella de carbono que otras alternativas, por ejemplo, la desalinización.

Sin embargo, la reutilización de aguas también presenta algunos retos y barreras:

- Heterogeneidad regulatoria, falta de ella o exceso de regulación: una regulación adecuada es un requisito fundamental para garantizar prácticas seguras (tanto en términos de salud como de medio ambiente) de reutilización del agua. La heterogeneidad, la ausencia o la sobrerregulación impiden la seguridad jurídica necesaria o dificultan la expansión de la reutilización del agua
- Riesgo ambiental y para la salud: la reutilización no planificada del agua representa un riesgo ambiental y para la salud debido a la presencia de sustancias químicas y patógenos. Para abordar este problema, un enfoque multibarrera, una aplicación adecuada del agua y una gestión de riesgos coherente permiten aprovechar los beneficios del agua regenerada.
- Aceptación social: el reconocimiento de los beneficios y la seguridad de la reutilización del agua en la agricultura por parte de los usuarios finales y los ciudadanos todavía representa un desafío. Las razones de este rechazo podrían incluir: falta de conocimiento sobre la regulación y las ventajas de esta práctica; bajo nivel de participación de agricultores y responsables políticos en la toma de decisiones; percepción de costes relacionados con el tratamiento; y preocupación por sus efectos en el suelo o los cultivos.

Este cambio de percepción encaminado al impulso de la regeneración y reutilización debe ir acompañado por una adecuada innovación tecnológica, permitiendo un mejor aprovechamiento

de estos recursos, garantizando el correcto tratamiento de las aguas residuales, y asegurando la salud de la población y la conservación de los ecosistemas. La incorporación de la tecnología de membranas de ultrafiltración (UF) a las instalaciones de depuración de las aguas residuales urbanas, bien como etapa complementaria en los esquemas de tratamiento convencionales o como parte de procesos más novedosos (por ejemplo, combinada con un reactor anaerobio conformando un biorreactor anaerobio de membranas; AnMBR, del inglés *anaerobic membrane bioreactor*), se presenta como una alternativa prometedora desde un punto de vista económico y medioambiental. Su incorporación como unidad de tratamiento primario permite incrementar la cantidad de materia orgánica que se deriva al digestor anaerobio de la línea de fangos, reduciendo por tanto la cantidad de materia orgánica que ha de ser tratada en el reactor biológico. Formando parte de un sistema AnMBR, se favorece la producción de un efluente de elevada calidad, la valorización de la materia orgánica en forma de biogás, y la recuperación de nutrientes. Como tratamiento terciario, se mejora la calidad del agua tratada, alcanzando adecuados niveles de desinfección de la misma, lo cual favorece su reutilización. Adicionalmente, estos sistemas de filtración reducen los requisitos de espacio en comparación con otros elementos de tratamiento convencional con función similar, tal y como son los decantadores o los sistemas de desinfección compuestos por filtros de arena y lámparas ultravioletas.

Sin embargo, la aplicación de membranas de ultrafiltración para el tratamiento de aguas residuales urbanas presenta algunos retos que deben ser abordados y superados para impulsar su adopción en las plantas de tratamiento, entre los que destacan: los costes de adquisición y mantenimiento/reposición de las membranas; el ensuciamiento de las membranas derivado del proceso de filtración y la consecuente reducción de la eficiencia del proceso; la recuperación del metano disuelto en el efluente de sistemas AnMBR; y la recuperación de los nutrientes presentes en el efluente de sistemas AnMBR.

Esta tesis doctoral tiene como objetivo general evaluar la viabilidad técnica, económica y medioambiental de la aplicación de membranas de ultrafiltración en diferentes etapas de tratamiento de aguas residuales urbanas: tratamiento primario (filtración del efluente de decantación primaria), secundario (AnMBR) y terciario (filtración del efluente de decantación secundaria). Este objetivo general se concreta en las siguientes tareas: 1) evaluar la viabilidad técnica de los procesos de tratamiento evaluados; 2) identificar estrategias y condiciones de operación que favorezcan la optimización de los mismos desde un punto de vista económico y medioambiental; 3) estimar los costes de inversión y operación de los dichos sistemas; y 4) evaluar el impacto ambiental derivado de la implementación de estos sistemas. Para ello, en esta

tesis doctoral se utilizan datos obtenidos en tres plantas a escala demostración, los cuales permiten estudiar su escalado a nivel industrial y su posterior evaluación ambiental y económica mediante las herramientas de análisis de ciclo de vida y de costes desarrolladas.

Esta tesis doctoral se ha desarrollado en el marco de la iniciativa europea sobre innovación *Innovation Deal on sustainable wastewater treatment combining anaerobic membrane technology and water reuse* (2016-2018) y tres proyectos de investigación: 1) *Membrane for Energy and Water Recovery-LIFE MEMORY* (LIFE13 ENV/ES/001353) (2014-2018) financiado por Life+ Programme; 2) *Application of membrane technology to enhance the transformation of WWTPs in resource recovery facilities-MEM4REC* (CTM2017-86751-C2-2-R-AR) (2018-2021), financiado por la Agencia Estatal de Investigación-AEI; y 3) *Caracterización de aguas y lodos residuales urbanos e industriales y evaluación de la tecnología adecuada para la recuperación de microplásticos presentes en los mismos-MICROPLAST* (INNEST00/19/058) (2019-2020), financiado por la Agencia Valenciana de la Innovación-AVI.

Las principales resultados y conclusiones obtenidos en esta tesis doctoral se muestran a continuación:

Resultados ambientales y económicos de combinar el tratamiento de aguas residuales (proceso aerobio y/o AnMBR) con fertirrigación bajo una perspectiva de economía circular.

Los resultados en este apartado mostraron que el agua regenerada es un recurso fiable que puede asociarse con un suministro relevante de nutrientes (en el estudio realizado, hasta el 72 % de las necesidades de nitrógeno y el 39 % de las necesidades de fósforo en el estudio de caso español, y hasta el 100 % de ambos nutrientes en el estudio de caso italiano). Este suministro es independiente de las sequías estacionales y de las variaciones climáticas y, por tanto, actúa como un recurso alternativo para aliviar la presión sobre las fuentes de agua dulce, al tiempo que reduce los costes económicos y ambientales de los fertilizantes minerales (hasta 171 k€·año^{-1}). Sin embargo, se necesita un plan de gestión de riesgos para garantizar la seguridad ambiental y sanitaria cuando se utiliza agua regenerada tanto para riego como para fertirrigación. Los resultados también mostraron el saldo económico positivo que se puede obtener en diferentes escenarios de reutilización del agua (hasta 376 k€·año^{-1}). Este ahorro se podría destinar a la elaboración, actualización e implementación de los Planes de Gestión de Riesgos de Reutilización

del Agua bajo la responsabilidad del nuevo Gestor de Agua Recuperada, tal y como recoge el Reglamento (UE) 2020/741 sobre requisitos mínimos para la reutilización del agua.

Además, dado que la tecnología AnMBR también puede recuperar energía del flujo de aguas residuales, su aplicación presenta un escenario óptimo según los principios de la Economía Circular y la sostenibilidad ambiental. En este sentido, la combinación de la tecnología AnMBR y la fertirrigación puede reducir significativamente las emisiones de CO_2 (hasta 898.9 $tCO_2 \cdot año^{-1}$) y reducir el coste de operación del tratamiento de aguas (hasta un 63% menos que los procesos aerobios). Por último, pero no menos importante, son necesarios proyectos a escala demostración de la combinación AnMBR y fertirrigación que permitan avanzar en las evaluaciones de riesgos ambientales y de salud, consideraciones agronómicas y análisis sociales y económicos para obtener información completa que permita elaborar los planes de gestión de riegos de manera solvente y robusta.

Efecto económico y ambiental de la implementación de membranas UF como tratamiento primario.

Los resultados mostraron que la demanda neta de energía (DNE) en el esquema convencional de tratamiento fue de -0.328 $kWh \cdot kgDQO_{rem}^{-1}$ y de -0.381 $kWh \cdot kgDQO_{rem}^{-1}$ en la alternativa con UF tras el decantador primario. Por tanto, este último esquema implica un ahorro energético de 0.053 $kWh \cdot kgDQO_{rem}^{-1}$. El coste de operación (OPEX) en el esquema convencional representa €-0.137 $kgDQO_{rem}^{-1}$, mientras que el esquema alternativo mostró valores más bajos para los costes de operación: €-0,150 $kgDQO_{rem}^{-1}$. Esto supone un ahorro de €0.013 $kgDQO_{rem}^{-1}$ respecto del tratamiento convencional. Los principales contribuyentes al OPEX en el diseño convencional fueron la aireación del reactor (€0.116 $kgDQO_{rem}^{-1}$) seguida de la precipitación química de fósforo (€0.024 $kgDQO_{rem}^{-1}$), mientras que la alternativa basada en UF presentó dos costes asociados importantes: €0.076 $kgDQO_{rem}^{-1}$ por agitación de membranas y €0.025 $kgDQO_{rem}^{-1}$ por sustitución de membranas de UF. La alternativa con UF también mantuvo la aireación del reactor como un gasto significativo, pero se redujo a €0.061 euros por kg de DQO eliminada.

En cuanto al coste de inversión (CAPEX), las membranas de UF representan el principal coste: €0.099 $kgDQO_{rem}^{-1}$ y el 94,7 % del. El coste total anualizado (CTA) del tratamiento convencional es €0,092 $kgDQO_{rem}^{-1}$ inferior que en la alternativa con UF. Teniendo en cuenta que el balance energético y el OPEX en la alternativa son favorables en comparación con el diseño convencional, el CAPEX muestra que el coste de la membrana es el principal cuello de botella para la

implementación de la alternativa basada en membranas, ya que los ahorros en operación no pueden compensar la inversión en membrana de €0.099 $kgDQO_{rem}^{-1}$ y el coste de reposición de membranas de €0,025 $kgDQO_{rem}^{-1}$. Sin embargo, un aumento en la durabilidad de las membranas, una disminución en su coste, un aumento en el coste de la energía y una mejora en el flujo transmembrana mejorarían la viabilidad económica de la alternativa mediante membranas de UF.

En cuanto al potencial de calentamiento global, la estimación de las emisiones de N_2O asociadas al proceso de nitrificación-desnitrificación representa el principal contribuyente a las emisiones equivalentes de CO_2 (91 % y 87 % en el tratamiento convencional y el alternativo con UF, respectivamente). En general, la alternativa con UF permitió reducir las emisiones de CO_2 en comparación con el tratamiento convencional debido a la reducción de la carga total de nitrógeno que ingresa al reactor aeróbico (y por lo tanto la cantidad de nitrógeno que eventualmente forma N_2O durante el proceso de nitrificación-desnitrificación).

La emisión directa de fósforo a las masas de agua resultó ser el principal factor que contribuye a la eutrofización del agua dulce (98 %). Del mismo modo, las emisiones de nitrógeno con el efluente al agua representaron la principal contribución a la eutrofización marina (97-98 %). Los resultados de ambas categorías de impacto (eutrofización marina y de agua dulce) confirmaron que la recuperación y reutilización de nutrientes son procesos cruciales para mejorar la sostenibilidad ambiental del tratamiento de aguas residuales urbanas.

Finalmente, los resultados de la escasez de recursos fósiles ilustran las oportunidades que representan las aguas residuales urbanas para enfrentar el agotamiento global de los recursos. La recuperación de energía y nutrientes puede contribuir a preservar las fuentes convencionales y convertir las aguas residuales en una fuente de recursos. Por lo tanto, se podría esperar que la sostenibilidad ambiental del tratamiento de agua mejorara al aprovecharlos, por ejemplo, mediante fertirrigación.

Rendimiento del tratamiento biológico de la tecnología AnMBR para la depuración de aguas residuales urbanas.

El tratamiento biológico mediante tecnología AnMBR permitió un promedio de eliminación de DQO del 87.2 ± 6.1 % y las membranas fueron capaces de retener prácticamente todas las partículas. Entre los escenarios evaluados, el mayor rendimiento de producción de metano (Y^{CH_4})

(169.0±95.1 STP L CH_4·kg^{-1} DQO) se obtuvo con una combinación de temperatura ambiente alta (27±1 ºC), tiempo de retención hidráulico (TRH) de 41 ±1 h y una relación DQO:SO_4^{2-}-S de 870±369 DQO·g^{-1} SO_4^{2-}-S. Si bien una temperatura alta mejorara la tasa de hidrólisis, la producción de metano aumentó con la DQO disponible para las arqueas metanogénicas (MA). Los valores obtenidos de Y^{CH_4} más bajos (75.2±16.9 y 70.2±36.0 STP L CH_4·kg^{-1} DQO) también pueden explicarse por la menor temperatura, TRH y relación DQO:SO_4^{2-}-S de sus escenarios de operación. Por lo tanto, en aguas residuales ricas en sulfatos, resulta muy relevante el papel de las bacterias reductoras de sulfato (BRS) en la eliminación de DQO soluble cuando se reduce la HRT, ya que las BRS tienen tasas de degradación más altas que el MA, especialmente a temperaturas más bajas y compiten eficientemente por el mismo sustrato.

Con respecto a la producción de lodos residuales (PLR), los resultados mostraron una interacción compleja entre las condiciones de operación y las características del afluente. La PLR más alta (0.166±0.074 kgVSS·kg^{-1} DQO_{rem}) se obtuvo con una temperatura de operación baja (19±1 °C), ya que menores temperaturas reducen la biodegradabilidad. Las PLR más bajas entre los escenarios estudiados (0.121±0.067 y 0.103 ±0.044 kg VSS·kg^{-1} DQO_{rem}) se obtuvieron a alta temperatura (27±1 °C y 24±2 °C, respectivamente) y relaciones DQO:SO_4^{2-}-S (8.70±3.69 DQO·g^{-1} SO_4^{2-}-S y 8.15±4.44 DQO·g^{-1} SO_4^{2-}-S). La combinación de ambas variables mejoró la biodegradación de la materia orgánica por parte de las MA y redujo la producción de lodos. Es importante resaltar que, además de la compleja interacción de factores que afectan la PLR, ésta puede ser tan baja como 0.103±0.044 kgVSS·kg^{-1} DQO_{rem}.

Por lo tanto, la temperatura, el TRH y la relación DQO:SO_4^{2-}-S, junto con el tiempo de retención celular (TRC), son parámetros cruciales a tener en cuenta para maximizar el rendimiento técnico, económico y ambiental de la tecnología AnMBR. Sin embargo, dado que la temperatura y la relación DQO:SO_4^{2-}-S del afluente dependen de las condiciones ambientales, el TRH podría considerarse el principal parámetro de diseño junto con la TRC.

Rendimiento de la filtración, consumo de energía y emisiones de gases de efecto invernadero de la tecnología AnMBR para el tratamiento de aguas residuales urbanas.

Los resultados obtenidos mostraron una interacción compleja entre el flujo transmembrana normalizado a 20°C (J_{20}), la demanda de gas para agitación de las membranas (DGA_m), la

concentración de sólidos suspendidos totales (SST) en el reactor, la presión transmembrana (PTM) y la evolución del ensuciamiento. La operación a elevados J_{20} con una baja propensión al ensuciamiento se puede lograr con altas DGA_m y moderados SST. Los resultados mostraron que el sistema pudo realizar el proceso de filtración a un J_{20} moderado-alto (15–20 LMH) con un DGA_m competitivo (correspondiente a un DGA por volumen de permeado de 10–20 $Nm^3 \cdot m^{-3}$) y TSS moderado (8–13 $g \cdot L^{-1}$), aunque se observó que se producían transiciones entre regímenes de filtración supracrítica y subcrítica con pequeños cambios en las condiciones de operación. Operar en condiciones de filtración subcríticas (recomendadas para evitar un rápido aumento de PTM) fue posible con concentraciones de SST inferiores a 10 $g \cdot L^{-1}$, J_{20} máximo de 16 LMH y DGA_m de 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (correspondiente a 15 $Nm^3 \cdot m^{-3}$). El control del ensuciamiento se podría lograrse incluso con una PTM alta (470–500 mbar). Finalmente, cabe señalar que la planta de demostración logró operar a largo plazo sin requisitos de limpieza química, siendo el período de operación más largo de 233 días, aunque los resultados indicaron que se podían alcanzar períodos de operación más largos. No se detectó ensuciamiento irrecuperable ya que la limpieza química devolvió la PTM a los valores iniciales o incluso inferiores.

En cuanto al balance energético, las principales contribuciones al consumo total de energía (CTE) estuvieron asociadas a las necesidades de las soplantes para la agitación de las membranas (55.2 %), seguidas por la mezcla del reactor (34.4 %). En general, el CTE promedio fue de 0.192 ± 0.070 $kWh \cdot m^{-3}$, mientras que la recuperación de energía total (RTE) promedio (considerando 0 % de recuperación de metano) fue de 0.361 ± 0.271 $kWh \cdot m^{-3}$. El metano disuelto produjo más energía que la utilizada en su recuperación, aunque su contribución a la RTE fue relativamente bajo (entre 0.02 $kWh \cdot m^{-3}$ y 0.04 $kWh \cdot m^{-3}$ para recuperaciones de metano disuelto del 50% y 70 %, respectivamente). Sin embargo, esta recuperación redujo significativamente las emisiones de gases de efecto invernadero (GEI). Estos resultados enfatizan la importancia de optimizar los parámetros que rigen la mezcla en el reactor y el proceso de filtración, especialmente en lo que respecta a la DGA_m. Además, el aumento de la corriente de recirculación de fangos reduciría la concentración de SST en el tanque de membrana y ayudaría a reducir las necesidades de DGA_m.

Cuando no se consideró la recuperación de metano del efluente, la contribución del metano disuelto (D-CH_4) a las emisiones totales representó entre 0.5 y 0.6 $kgCO_{2eq} \cdot m^{-3}$ durante casi todo el período de operación de la planta. Recuperar el 50 % del reduciría las emisiones de CO_2 equivalentes a 0.1-0.3 $kgCO_{2eq} \cdot m^{-3}$. Operar con una recuperación de metano del 70 % mantendría las emisiones por debajo de 0.2 $kgCO_{2eq} \cdot m^{-3}$. Los tratamientos aerobios presentan unas emisiones

superiores: alrededor de 0.3 $kgCO_{2eq}·m^{-3}$ para instalaciones de fangos activados convencionales con eliminación de materia orgánica y nutrientes en áreas sensibles, o 0.7 $kgCO_{2eq}·m^{-3}$ en el caso de biorreactores aerobios de membrana a pequeña escala.

En general, la planta de demostración AnMBR mostró un rendimiento de la filtración adecuado, importantes producciones de energía y bajas emisiones de CO_2 al recuperar metano disuelto. Estos resultados indican que AnMBR combinado con membranas desgasificadoras supone una alternativa sólida al tratamiento convencional de aguas residuales urbanas.

Sostenibilidad económica de un AnMBR a gran escala para el tratamiento de agua residual urbana.

Los resultados mostraron que los principales contribuyentes al CTE en todos los escenarios evaluados fueron la mezcla del reactor (34–57%) y la agitación de las membranas (22–48%). Se lograron balances energéticos favorables en todos los escenarios evaluados. La DNE varió entre -0.210 $kWh·kgDQO_{rem}^{-1}$ y -0.645 $kWh·kgDQO_{rem}^{-1}$. La mezcla del reactor (0.056–0.124 $kWh·kgDQO_{rem}^{-1}$) y el lavado de membranas (0,048–0,120 $kWh·kgDQO_{rem}^{-1}$) mostraron el mayor consumo de energía. Los principales impulsores de los resultados de la DNE fueron la carga orgánica afluente, el rendimiento de eliminación DQO, el TRH, la temperatura y la relación DQO:SO_4^{2-}-S. Optimizar el TRH para reducir los consumos asociados a la mezcla del reactor y diseñar una estrategia de filtración integral capaz de mantener una PTM baja también son cruciales para reducir el CTE.

Los principales contribuyentes al CAPEX fueron las membranas de UF (€0.029-0.073 $kgDQO_{rem}^{-1}$; 31-49%), la adquisición de motores de cogeneración (€0.012-0.023 $kgDQO_{rem}^{-1}$; 8-24%) y la construcción de reactores (€0,007-0,014 $kgDQO_{rem}^{-1}$; 8-13%). El OPEX se vio influenciado principalmente por las necesidades energéticas de los equipos (€0.042–0.069 $kgDQO_{rem}^{-1}$; 41–46%), el reemplazo de la membrana de UF (€0.011–0.028 $kgDQO_{rem}^{-1}$; 9–17%) y canon de vertido (€0,010–0,020 $kgDQO_{rem}^{-1}$; 9-12%). El mantenimiento y la sustitución de los sistemas de cogeneración también supuso un coste de operación importante (€0.009-0.017 $kgDQO_{rem}^{-1}$; 6-14%), aunque finalmente generó ingresos a través de la venta de energía (€0.120-0.222 $kgDQO_{rem}^{-1}$). El CTA osciló entre €-0,003 y €0,188 $kgDQO_{rem}^{-1}$, siendo el CAPEX el coste principal.

Los resultados obtenidos de la evaluación de ciclo de costes combinada con el análisis de sensibilidad y de incertidumbre mostraron que la tecnología AnMBR representa una alternativa potencial a los tratamientos aerobios convencionales. Su impulso definitivo pasará, entre otros aspectos, por reforzar su sostenibilidad económica, para lo que es necesario optimizar aspectos de su operación (p. ej., el TRH, el flujo transmembrana, la mezcla de los reactores y la agitación de las membranas) y desarrollos tecnológicos que permitan aumentar la vida útil de las membranas y mejorar la eficiencia energética en la cogeneración. Finalmente, otros aspectos que también redundarían en mejorar su sostenibilidad económica tienen que ver con la recuperación de nutrientes presentes en sus efluentes y precios favorables para la venta de la electricidad producida a partir del metano generado.

Sostenibilidad ambiental de un AnMBR a gran escala para el tratamiento de UWW.

Con respecto al potencial de calentamiento global, la fase de operación fue la que más contribuyó a esta categoría de impacto, representando entre el 87% y el 92% del impacto ambiental directo. La mayor parte de la carga ambiental directa en esta categoría estuvo relacionada con emisiones de metano procedentes del efluente (40-55 %). Por lo tanto, la reducción de la huella de carbono de la tecnología AnMBR implica necesariamente mejorar la recuperación de este metano disuelto. A su vez, recuperar el metano disuelto para producir energía mejoraría el balance energético. En general, los resultados para el potencial de calentamiento global estuvieron en el rango -0.223:0.020 $kgCO_{2eq}{\cdot}kgDQO_{rem}^{-1}$. A modo de comparación, otros estudios evaluaron el funcionamiento de diferentes EDAR, obteniendo valores entre 0.206 $kgCO_{2eq}{\cdot}kgDQO_{rem}^{-1}$ y 0.551 $kgCO_{2eq}{\cdot}kgDQO_{rem}^{-1}$para instalaciones sin procesos de eliminación de nutrientes y vertidos a áreas no sensibles, y entre 0.413 $kgCO_{2eq}{\cdot}kgDQO_{rem}^{-1}$ y 0.688 $kgCO_{2eq}{\cdot}kgDQO_{rem}^{-1}$ para lodos activados convencionales con eliminación de nutrientes y descarga a zonas sensibles. Por tanto, se puede concluir que la tecnología AnMBR puede considerarse como un tratamiento alternativo para reducir las emisiones de GEI en la gestión de aguas residuales cuando se captura el metano disuelto en el efluente.

La categoría de escasez de recursos minerales estuvo influenciada principalmente por la DNE y la recuperación de nutrientes a través de la dispersión de lodos debido a que se evita la producción de fertilizantes a base de fósforo y especialmente a base de nitrógeno. También cabe mencionar que la fase de construcción representó entre el 51 % y el 71 % del impacto directo del ciclo de vida de la escasez de recursos minerales, cuando no se consideraron las compensaciones por

energía, la recuperación de nutrientes el reciclaje tras la vida útil. Por lo tanto, se debe evaluar cuidadosamente la exclusión de la fase de construcción del análisis de ciclo de vida (ACV). El impacto ambiental sobre la escasez de recursos fósiles siguió un patrón similar al potencial de calentamiento global. Por tanto, la DNE fue el factor más importante en esta categoría de impacto. Aunque la fase de operación fue la que más contribuyó a la escasez de recursos fósiles, la carga de la fase de construcción representa entre el 18 y el 27 % de la carga directa. También es destacable que tanto el agotamiento neto de los recursos minerales como fósiles obtuvieron cargas negativas en todos los escenarios evaluados, lo que indica que AnMBR contribuye a preservar estos recursos.

Las características de los efluentes fue el principal factor que influyó en la eutrofización tanto de agua dulce como marina debido a su contenido de nitrógeno y fósforo. Es importante destacar que, si bien las concentraciones de nutrientes de los efluentes de los tratamientos anaeróbicos presentan un potencial de eutrofización a considerar, también es cierto que representan una oportunidad para recuperar estos recursos en cantidades considerables, lo que conlleva evidentes beneficios ambientales y económicos.

Con respecto al consumo de agua, los resultados indicaron que la reducción de la DNE mediante la recuperación de biogás y metano disuelto tuvo un efecto positivo en las cargas ambientales. Sobre la base de los resultados, también se podría afirmar que aprovechar el contenido de nutrientes y evitar así la producción de fertilizantes a base de fósforo y N también contribuyó a una reducción relevante del impacto total en esta categoría. Es importante resaltar que no se consideró la reutilización del agua. Incluir esta práctica en los límites del ACV evitaría la extracción de agua dulce, mejorando así esta categoría de impacto. El peso de la fase de construcción en la categoría de consumo directo de agua representó entre el 21% y el 28% del impacto total del ciclo de vida. La demolición también tuvo un efecto significativo y contribuyó a reducir la carga del consumo de agua en todos los escenarios.

Finalmente, los análisis de sensibilidad e incertidumbre mostraron la recuperación de energía, los metales pesados en los lodos, el metano disuelto en el efluente y el contenido de nutrientes del efluente como los aspectos más importantes que afectan el resultado del ACV. Además, los parámetros más influyentes fueron la eficiencia energética de la cogeneración, el potencial de recuperación de metano disuelto, la productividad de la filtración, la agitación de las membranas, la mezcla del reactor, el TRH y el TRC. Estos resultados mostraron que las acciones que conducen a aumentar la recuperación y valorización del metano disuelto y/o la recuperación de nutrientes

mediante, por ejemplo, fertirrigación, mejoran la sostenibilidad ambiental de la tecnología AnMBR para el tratamiento de aguas residuales urbanas.

Efecto económico y ambiental de la implementación de membranas de UF como tratamiento terciario.

Los resultados de filtración de la planta piloto de UF sugirieron que el sistema de UF debería funcionar en condiciones diferentes en verano e invierno para evitar un ensuciamiento excesivo, es decir, a flujos transmembrana (J) de alrededor de 28-33 LMH y demandas de aire para agitación de las membrabas (DAA_m) de 0.4-0.6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ en verano y J alrededor de 33-39 LMH y DAA_m de 0.3-0,4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ en invierno.

La evaluación energética del tratamiento de UF mostró que el CTE en condiciones de funcionamiento de verano fue de 0.239±0.083 $kWh \cdot m^{-3}$ en promedio, mientras que los resultados para el período de invierno fueron de 0.151±0.044 $kWh \cdot m^{-3}$. Las soplantes de las membranas representan el 87-92 % del TEC, mientras que el bombeo de alimentación y el bombeo de permeado contribuyeron con el 3-9 % y el 2-9 %, respectivamente. Por lo tanto, los esfuerzos para reducir el gasto energético deben centrarse en disminuir las necesidades de agitación de las membranas. Para ello, se debe adoptar una estrategia de filtración adecuada que garantice flujos transmembrana significativos que, eventualmente, reduzcan las necesidades de membrana.

En cuanto al CAPEX, la adquisición de membranas apareció como el principal contribuyente, representando el 62-66 % en el escenario de verano y el 63-67 % en el escenario de invierno. Cuanto menor sea el J mayor será el CAPEX, ya que los requisitos de la membrana dependen de la productividad de la filtración. De media, los escenarios de verano mostraron un CAPEX de €0.025±0.001$\cdot m^{-3}$ y el escenario de invierno de €0.026±0.001$\cdot m^{-3}$, lo que no supone diferencias significativas. Expresada en costes, la mayor inversión corresponde a las membranas, €0.016±0.001$\cdot m^{-3}$ y €0.017±0.001$\cdot m^{-3}$ (verano e invierno, respectivamente). Las bombas de permeado y las bombas de alimentación suponen €0,003±0,000$\cdot m^{-3}$ tanto en el escenario de verano como en invierno (13±1 % cada una), mientras que la contribución de las soplantes se calculó en €0.002±0,000$\cdot m^{-3}$ (9 %).

El CTA varió entre €0.068 y 0.110$\cdot m^{-3}$ en verano mientras que en invierno osciló entre €0.059 y €0.083 m^{-3}. De media, el CTA en condiciones de verano fue de €0.087±0.018 m^{-3} y de

€0.070±0.010 m^{-3} en escenario de invierno. El principal contribuyente al CTA fue el OPEX, que representa el 70 ± 5 % en el escenario de verano y el 63 ± 4 % en el escenario de invierno.

El resultado del ACV mostró que la operación es la principal fase que influye en el potencial de calentamiento global, el consumo de agua, la escasez de recursos fósiles y la escasez de recursos minerales (90-98%). La fase de construcción fue significativa sólo para la última categoría (8-10 %). Dentro de la fase de operación, la mayor influencia corresponde al consumo energético (64-95 %). Por tanto, mejorar la sostenibilidad ambiental de la aplicación de membranas de UF para tratamiento terciario pasa necesariamente por una optimización de la filtración enfocada a reducirlo.

Los resultados demuestran que el proceso de UF como tratamiento terciario permite producir un efluente de alta calidad capaz de cumplir con los estrictos requisitos microbiológicos y fisicoquímicos establecidos por el Reglamento (UE) 2020/741 y el Real Decreto 1620/2007.

Los resultados obtenidos en esta tesis revelan que los procesos de membranas evaluados pueden suponer ventajas económicas y ambientales frente a otros sistemas de tratamiento convencional, aunque la magnitud de los beneficios obtenidos varía en función del tipo de si la membrana actúa como tratamiento, como parte de un AnMBR o como tratamiento terciario.

13. Appendix II: Abbreviations

Abbreviations

AD: anaerobic digestion

AnMBR: anaerobic membrane bioreactor

ARB: antibiotics-resistant bacteria

ARG: antibiotics-resistant genes

BMP: biomethane potential

BOD_5: biological oxygen demand

CAPEX: capital expenditure

CAS: conventional activated sludge

CE: circular economy

CHP: combined heat and power

CIP: clean-in-place tank

COD: chemical oxygen demand

DM: degassing membranes

DMF: direct membrane filtration

DO: dissolved oxygen

EPS: extracellular polymeric substances

FEW: freshwater eutrophication

FR: fouling rate

FRS: fossil resource scarcity

GHG: greenhouse gas

GWP: global warming potential

HRT: hydraulic retention time

HS-S: sulfide

J_{20}: 20ºC-standardized transmembrane flux

$J_{20,g}$: gross 20ºC-standardized transmembrane flux

LCA: life cycle assessment

LCC; life cycle costing

MA: methanogenic archaea

MBR: membrane bioreactor

ME: marine eutrophication

MF: microfiltration

MLTSS: mixed liquor total suspended solids

MT: membrane tank

MWW: municipal wastewater

N: nitrogen

NED: net energy demand

OLR: organic loading rate

OPEX: operational expenditures

P: phosphorus

RO: reverse osmosis

SA: sensitivity analyses

SAD_m: sparging air demand per m^2 of membrane

SGD_P: sparging gas demand per m^3 of permeate

SMP: soluble microbial products

SO_4^{2-}-S: sulfate

SRB: sulfate-reducing bacteria

SRT: sludge retention time

STP: standard temperature and pressure

TAC: total annual cost

TEC: total energy consumption

TER: total energy recovery

TMP: transmembrane pressure

TSS: total suspended solids

UA: uncertainty analyses

UF: ultrafiltration

UV: ultraviolet

UWW: urban wastewater

VFA: volatile fatty acids

VSS: volatile suspended solids

WSP: waste-sludge production

WWRF: water resource recovery facilities

WWTP: wastewater treatment plant

X_{AOB}: ammonia oxidizing bacteria

X_{NOB}: nitrite oxidizing bacteria

Y^{CH_4}: methane yield

14. Appendix III: Scientific articles

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AnMBR, reclaimed water and fertigation: Two case studies in Italy and Spain to assess economic and technological feasibility and CO_2 emissions within the EU Innovation Deal initiative

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ABSTRACT

The use of anaerobic membrane bioreactor (AnMBR) technology on urban wastewater can help to alleviate droughts, by reusing the water and nutrients embedded in the effluent in agriculture (fertigation) in line with Circular Economy principles. The combination of AnMBR and fertigation reduces CO_2 emissions due to the organic matter valorization and the partial avoidance of mineral fertilizer requirements. However, both AnMBR and fertigation still face technological and regulatory barriers that need to be overcome. These bottlenecks were tackled within the first Innovation Deal approved by the European Commission in 2016, and gave rise to several case studies on water reuse systems. The results of the Oliva Wastewater Treatment Plant (Spain) and Peschiera-Borromeo Wastewater Treatment Plant (Italy) showed that reclaimed water can be considered as a reliable water and nutrient source, return a positive economic balance (up to 376 k€·year^{-1}) and provide significant reductions and savings in CO_2 emissions (up to −898.9 tCO_2·year^{-1}). According to the new EU regulation, a new key player known as the *Reclaimed Water Manager*, was also proposed to be in charge of supplying reclaimed water with appropriate quantity and quality to end-users. This new agent would also be responsible for drawing up and implementing a Water Reuse Risk Management Plan in cooperation with the parties involved. Applying AnMBR technology to water reuse thus shows potential for contributing to catchment-scale Circular Economy while preserving natural water bodies, reducing the carbon footprint and creating new business opportunities. However, to take full advantage of its benefits demonstration projects would need to be carried out and favorable and harmonized regulations among the EU States would need to be adopted.

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1. Introduction

The worldwide population growth has caused and increasing demand for food and thus greater water extraction and higher consumption of fertilizers (Hussain et al., 2019). According to Ledger et al. (2012) as an additional pressure the current climate change conditions are expected to affect rain patterns and cause water scarcity in many regions. This situation calls for better efficiency in the use of water resources.

In this context, Circular Economy (CE) principles are gaining momentum as part of the solution (Puyol et al., 2017). Their adoption in the water value chain is shifting the conventional wastewater treatment plant (WWTP) model to the water resource recovery facility (WRRF) concept. This can help to achieve economic and environmental goals within the water sector whilst

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producing valuable goods from a renewable source, including reclaimed water, nutrients, energy, etc.

To promote this paradigm shift, anaerobic membrane bioreactors (AnMBR) have been proposed as potential substitutes for conventional WWTPs, which are generally based on aerobic processes. AnMBR technology combines the advantages of membrane filtration with those of anaerobic processes: effluent quality, organic matter valorization, reduced carbon footprint, low biosolids production and on-site stabilization and nutrients recovery potential. AnMBR applications for industrial wastewater are already well-established (Dereli et al., 2012) and promising results for urban wastewater treatment have been reported: Giménez et al. (2011) showed almost 90% COD removal and a negligible fouling rate when operating a submerged pilot-scale anaerobic membrane bioreactor with a 10 LMH transmembrane flux and 22 g L^{-1} suspended solids in the reactor treating real urban wastewater; Ozgun et al. (2013) reported on the membranes' contribution to maintaining high treatment efficiency despite temperature fluctuations due to the retention of particulate COD and biomass in the reactor; Foglia et al. (2019) demonstrated the ability of a combination of UASB and AnMBR to treat high-salinity low-loaded municipal wastewater in the long term with a reduced methane production but without membrane fouling and producing a high-quality effluent, whereas Pretel et al. (2016b) compared different aerobic-based WWTP to AnMBR coupled to a post-treatment for nutrient removal, observing that AnMBR combined with a conventional activated (CAS) sludge-based post-treatment reduced the environmental impact; finally, Becker et al. (2017) showed AnMBR's energy recovery potential when treating joint wastewater and food waste.

However, some issues still need to be addressed in order to improve AnMBR technical, economic and environmental feasibility: the sensitivity and control of process dynamics have been pointed out by Robles et al. (2018); Velasco et al. (2018) focused on the release of dissolved methane with the effluent; several authors (e.g. Aslam et al., 2018; Judd, 2017; Maaz et al., 2019) coincide in considering membrane fouling as one of the major bottlenecks; and Smith et al., 2012 underlined eutrophication potential due to the nutrients content of its effluent. However, it is precisely the presence of N and P in the AnMBR effluent, together with the high quality of the water, that makes their application for fertigation an attractive approach for resource recovery.

Fertigation (simultaneous reuse of water and nutrients) helps to conserve freshwater sources and reduces the energy consumed for industrial ammonia-based fertilizer production (19.3 kWh·kg N^{-1} by the Haber-Bosh process, according to McCarty et al., 2011) and P extraction (2.11 kWh·kg P^{-1} according to Gellings and Parmenter, 2004). Financial benefits for farmers can also be expected from savings in mineral fertilizers (Scheierling et al., 2011). P was included in the EU's Critical Raw Materials list in 2017 (European Commission, 2017), which justifies strategies for reducing European dependency on nutrient importation from a food security perspective as well as from an environmental point of view.

Several documents, regulation proposals, reports and scientific papers have been published on the benefits of water reclamation and its related issues (Alcalde-Sanz and Gawlik, 2017; General Secretariat of the Council, 2019; WWAP, 2017). As stated by Condom et al. (2012), 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 feasibility and sustainability of water reuse projects and plans The EU has already recognized that the use of reclaimed water has a smaller environmental impact than other alternatives, i.e. water transfers or desalination, while it also has financial and social benefits (European Commission-DG ENV, 2012). The main current concerns are related to microbial risks to public health and the environment risks. These are generally addressed by a multibarrier approach in wastewater treatment/reclamation and the development and implementation of water reuse risk management plans (WRRMP).

Based on this background, and with the aim of investigating whether the AnMBR technology for reclaiming water for fertigation faces regulatory barriers in the EU, the European Commission (EC) approved the first pilot Innovation Deal (ID) entitled Sustainable wastewater treatment combining anaerobic membrane technology and water reuse in 2016. The ID consortium consisted of 14 partners from 5 European member states (Portugal, Spain, Malta, Italy and France) and included a comprehensive group of stakeholders involved in water management: national and regional authorities, universities, research centers, innovators and end-users. Three EC Directorate-Generals (Research and Innovation, Agriculture and Rural Development and Environment) were also represented.

The tasks carried out within the ID included the analysis of several case studies, two of which are presented in this study. These cases are located in the Mediterranean Region (Italy and Spain) and compare the application of combined AnMBR technology and fertigation with conventional configurations of WWTPs with aerobic systems, irrigation with freshwater and mineral fertilization. To the best of authors' knowledge, this has not been reported on before. The work carried out also included the development of the novel concept of the Reclaimed Water Manager (RWM), a new key player in accordance with the EU regulation, who will be in charge of operating water reclamation and reuse systems and responsible for drawing up and applying the WRRMP. Both case studies examined different scenarios and compared them to the current case. An economic study was performed for the RWM and the levels of wastewater resource recovery and CO_2 emissions were assessed for all configurations.

2. Material and methods

Each case study encompassed different scenarios in which at least one was AnMBR technology. A four-step assessment methodology was developed to analyze them: (i) water balance, (ii) nutrient balance, (iii) economic analysis and (iv) CO_2 emission estimations. Several reference values were used (see below in this section), based on the literature and on data provided by stakeholders (e.g. efficiency factors, water and nutrients needs, energy needs, energy prices, etc.). Further studies must find the most appropriate variables for each specific case in order to increase the accuracy of the results. Information on the end-users' perception on water reuse for agriculture was also gathered and analyzed.

2.1. Water balance

In the first step, the *net crop requirements* or *net provision* ($m^3 \cdot ha^{-1} \cdot year^{-1}$) is calculated, considering the type of crop and the location of the site, since meteorological (effective precipitation) and soil data (available water stored in the soil) exert a considerable influence on water availability. The second step is a water balance applied to the irrigation network in order to calculate the *gross irrigation volume* (V_B in $hm^3 \cdot year^{-1}$) that will be extracted from the water source, since transport, distribution and irrigation networks suffer water losses, known as transport network return flows (r_t), distribution network return flows (r_d) and irrigation return flows (r_a). There is also a water loss associated with evaporation throughout the irrigation network: evaporation in the transport network (p_t), in the distribution network (p_d) and in the irrigation volume applied to the land (p_a) (see Fig. 1).

Evaporation water losses are determined experimentally throughout the year (measuring campaigns by the Júcar River Basin

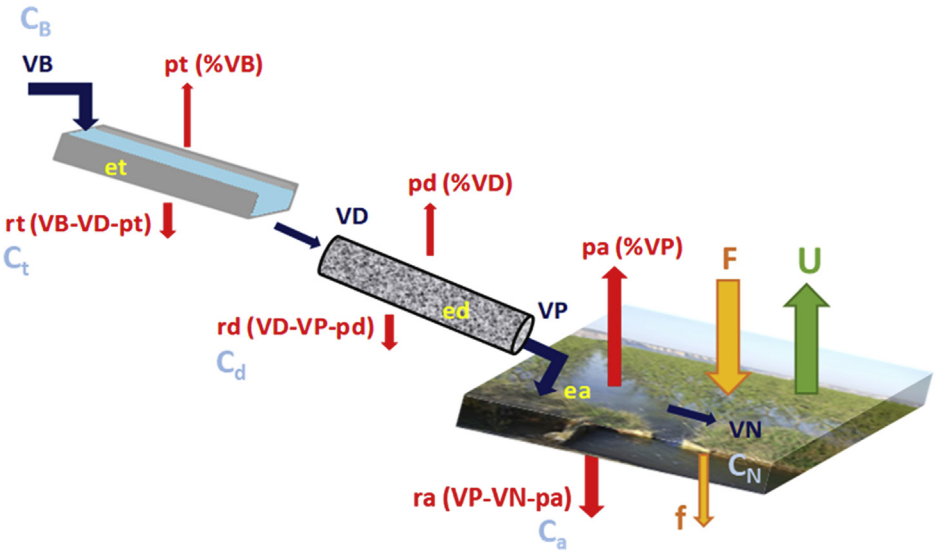

Fig. 1. Scheme of water and nutrient balances applied to the transport, distribution and irrigation network.

Authority) and return flows are calculated as follows:

$$r_t = V_B - V_D - p_t \quad \text{(Eq. 1)}$$

$$r_d = V_D - V_P - p_d \quad \text{(Eq. 2)}$$

$$r_a = V_P - V_N - p_a \quad \text{(Eq. 3)}$$

where V_D ($hm^3 \cdot year^{-1}$) is the irrigation influent water volume to the distribution network, V_P ($hm^3 \cdot year^{-1}$) is the volume applied to cropland and V_N ($hm^3 \cdot year^{-1}$) is the net irrigation volume.

Taking into account the water losses in the water flow sections, an efficiency percentage was obtained for the transport (e_t), for the distribution network (e_d) and for the irrigation method (e_a) (Eqs. (4)–(6)). These efficiency factors were used for all conversions from gross to net irrigation volumes.

$$V_D = V_B \cdot e_t \quad \text{(Eq. 4)}$$

$$V_P = V_D \cdot e_d \quad \text{(Eq. 5)}$$

$$V_N = V_P \cdot e_a \quad \text{(Eq. 6)}$$

2.2. Nutrient balance

During the nutrient balance step, the total amount of nutrients in the irrigation water is calculated together with the complementary mineral fertilizers needed to fully provide the crops with the optimal nutrients supply. The balance also evaluates the total amount of nutrients released into water bodies and is applied taking into account that water losses by evaporation give rise to higher nutrient concentrations throughout the water network (Eq. (7), Fig. 1).

$$V_B \cdot C_B + F = r_t \cdot C_t + r_d \cdot C_d + r_a \cdot C_a + f + U \quad \text{(Eq. 7)}$$

where C represents nutrient concentration (N or P), F is the fertilizer surplus, r is the return flow. Subscripts B, t, d and a indicate the water flow section in which they are identified, while f considers mineral fertilizer losses and U represents the crop's nutrient uptake.

The following assumptions were made:

- $V_P \cdot C_d + F = U + r_a \cdot C_a + f$: total nutrients provided by net irrigation water ($V_P \cdot C_d$) and mineral fertilizers (F) encompasses the crop's nutrient uptake (U), nutrient losses related to return flow in cropland ($r_a \cdot C_a$) plus mineral fertilizer losses (f).
- $C_N = C_a$: nutrient concentration in the irrigation return flow (r_a) is equal to nutrient concentration in the net irrigation water volume required by the crop (V_N).

Other agronomic considerations, e.g. micronutrients, soil quality, etc., which could change crop nutrient uptakes were outside the scope of this study.

2.3. Economic balance

Fig. 2 shows the different water, nutrient and money flows in a water re-use system to which the economic balance was applied to evaluate the feasibility of AnMBR for fertigation. As proposed by the authors and in line with the new EU regulation proposal (General Secretariat of the Council, 2019), the Reclaimed Water Manager was included. The RWM is the managing body responsible for operating, controlling and supervising the re-use system and would be in charge of drafting and implementing the Water Reuse Risk Management Plan in collaboration with the relevant parties.

The different flows shown in Fig. 2 are described as follows:

- Nutrient-rich wastewater reaches the WWTP via the sewer system. Based on the "*polluter pays principle*", there is a parallel money (tax/tariff) flow from the users (polluters) to the WWTP.
- Treated water and nutrients enter the Reclaimed Water Treatment Plant (RWTP) for additional treatment (e.g. further filtration, disinfection and/or advanced oxidation processes) to produce good quality reclaimed water.
- The water to be reused is pumped into the irrigation transport/distribution network. Farmers can also use other water sources, such as groundwater and/or surface water, which have to be paid for.

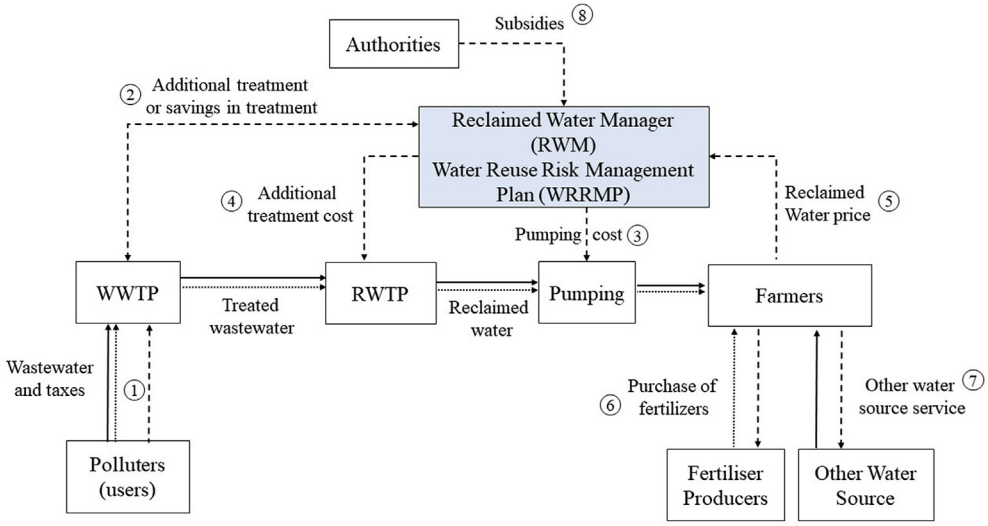

Fig. 2. Water (full line), nutrients (dotted line) and money flows (dashed line) for an agricultural water reuse system.

- According to the crop requirements, farmers might need to add mineral industrial fertilizers, which creates a money flow to fertilizer producers. If enough nutrients are present in the reclaimed water, this money flow can be reduced or even disappear.
- The farmers would pay the RWM for controlling the water re-use system and providing enough good-quality water. This opens a new market niche and business opportunity. The RWM would pay the RWTP for the water and nutrients and for using the pumping system. The RWM could also pay the WWTP for any additional treatment needed. In cases where savings can be made in wastewater treatment (WWT) (e.g. avoiding or reducing nutrient removal) or in discharge fees to surface or marine water due to water re-use, the WWTP might pay the RWM.
- The authorities could subsidize the system through the RWM, based on environmental, social and territorial considerations.

The total cost associated with running the WWT and providing reclaimed water for fertigation (C_{Total}, €·year^{-1}) was obtained as follows:

$$C_{Total} = C_{WWT} - C_{Energy\ recovery} + C_{Chemical\ precipitation} + C_{treat_for_reuse} + C_{Pumping} + C_{Fertilizers} + C_{Discharged\ fee} \quad \text{(Eq. 8)}$$

where C_{WWT} is the cost of treating wastewater to meet Urban Wastewater Treatment Directive (UWWTD) (91/271/EC) requirements, $C_{Energy\ recovery}$ is the saving associated with the energy recovered as methane during wastewater treatment, $C_{treat_for_reuse}$ is the additional treatment cost of reclaimed water reuse, $C_{Discharge\ fee}$ is the cost of discharging treated wastewater into water bodies, $C_{Pumping}$ is the energy cost for pumping a natural source of water (surface water or groundwater) or reclaimed water and $C_{Fertilizers}$ is the cost of buying fertilizers.

The cost balance of the RWM (B-RWM) was calculated by adding the cost flows in Fig. 2:

$$\text{B-RWM} = \pm 2\,(€) + 5(€) - 3\,(€) - 4\,(€) + 8\,(€) \quad \text{(Eq. 9)}$$

Where Flow 8 (subsidies) was assumed to be zero. The current price paid by polluters and farmers (current scenario, no water re-use) was kept constant in all the scenarios studied. Polluter prices were calculated according to the individual characteristics of each case, and the farmer's price was calculated as follows:

$$\text{Farmer's price} = C_{Fertilizers} + C_{Pumping} \quad \text{(Eq. 10)}$$

The flow from WWTP to the RWM (Flow 2) was calculated as the difference between the (constant) paid polluter price and the real cost. Savings were made in wastewater treatment or by the absence of chemical precipitation. The flow from farmers to the RWM (Flow 5) was calculated in the same way: as the difference between the farmer's price, which was to remain constant, and the real farming costs. Since the purchase of mineral fertilizer was partially or totally avoided, the RWM could profit from the associated savings.

2.3.1. Unit costs and energy consumption

Commercial fertilizer prices and energy consumption in Spain was estimated as shown in Table 1, assuming a WWTP energy cost of 0.125 €·kWh^{-1}. Energy consumption of 0.26 kWh·m^{-3} was considered for the CAS system with nutrient removal in the Italian case (value provided by system experts), together with an energy cost of 0.14 €·kWh^{-1}. Costs of 4.35 €·kgP$^{-1}_{removed}$ were considered for the consumption of chemical reagents during P removal (data provided by EPSAR, Spanish public body in charge of WWT in Valencia) and 0.006 €·m^{-3} for chemical membrane cleaning related to AnMBR technology (Pretel et al., 2016a). A pumping energy cost of 0.137 €·kWh^{-1}was considered for a pumping height of 60 m and 9.5 m for groundwater and reclaimed water, respectively.

It should be noted that: (i) the discharge fees applied in Valencia ($C_{Discharged\ fee}$) are on average 0.004 €·m^{-3} and 0.0135 €·m^{-3} for discharging water into coastal and inland water bodies, respectively (data provided by Júcar River Basin Authority and Regional Government of Valencia). (ii) Italy does not apply discharge fees ($C_{Discharged\ fee} = 0$). (iii) C_{WWT} already includes the cost of P chemical precipitation ($C_{Chemical\ precipitation}$) and disinfection ($C_{treat_for_reuse}$) in Peschiera Borromeo, due to regional regulations. (iv) C_{WWT} includes the cost of chemicals for membrane cleaning.

2.4. Carbon dioxide emissions

CO_2 assessment included CO_2 emissions related to WWT energy consumption ($CO_{2treatment}$), pumping ($CO_{2pumping}$) and fertilizer

Table 1
Cost of most usual mineral fertilizers and unitary treatment energy needs and energy reuse of WWT.

Mineral Fertilizer				Cost (€·kg^{-1})
Complex 39N–11P–0K				0.39
Urea 46N–0P–0K				0.31
DURAMON 26N–46P–0K				0.32
Lime superphosphate 0N–20P–0K				0.15
Complex 12N–61P–0K				0.66
	Units	Extended aeration (includes nitrogen removal)	AnMBR (without N.R.)	References
Wastewater Treatment	kWh·m^{-3}	[0.35–0.42]	0.269	EPSAR (based on Valencian Community average) and Pretel et al., 2013
Additional treatment for disinfection	kWh·m^{-3}	0.036	–	EPSAR (based on Valencian Community average)
Energy recovery	kWh·m^{-3}	–	[0.12 –0.24]	Pretel et al. (2016a)
Pumping from WWTP	kWh·m^{-3}·h^{-1}	0.004	0.004	EPSAR (based on Valencian Community average)

production ($CO_{2fertilizers}$) as well as the CO_2 emissions avoided by energy recovered as biogas ($CO_{2biogas}$). The WWT-related CO_2 emissions were based on the energy consumption given in Section 2.4. Considering a European average electricity specific emission factor of 0.2958 kg CO_2·kWh^{-1} (EEA, 2018). The energy required for fertilizer production was assumed to be 19.3 kWh·kg N^{-1} by the Haber-Bosh Process (McCarty et al., 2011) and 2.11 kWh·kg P^{-1} (Gellings and Parmenter, 2004).

Total CO_2 emissions (tCO_2·year^{-1}) were calculated as follows:

$$\text{Total } CO_2 \text{ emissions} = CO_{2treatment} + CO_{2pumping} + CO_{2fertilizers} - CO_{2biogas} \quad \text{(Eq. 11)}$$

2.5. End-users' perception

One of the social bottlenecks in agricultural water reuse identified in the Innovation Deal is the lack of current knowledge on the public perception and social acceptance of the reuse concept. A survey was therefore carried out to collect the end-users' feedback. 15 irrigators from the Mediterranean coast in Spain (Catalonia, Valencia, Alicante and Murcia) and one from Murviel Lès Montpellier in France collaborated in the survey and responded to questions related to crops, water use, water source, water quality, economics of water reuse, water supply problems and their attitude to using reclaimed wastewater for their crops.

2.6. Case studies description

The ID consortium carried out four case studies as part of the tasks involved in the project. This paper presents only the representative cases of Oliva (Spain) and Peschiera-Borromeo (Italy) WWTPs, as they could be replicated in many similar sites in both European countries, where the proposed technology could enhance agriculture sustainability with water reclamation and nutrient recovery and re-use.

2.6.1. Oliva WWTP

With a WWT flow (V_T) of 5.5 hm^3 year^{-1} (39,500 population equivalent (p.e.)), Oliva WWTP has an extended aeration (EA) process without P removal by precipitation. The WWTP is in Oliva (Valencia, Spain) and its effluent is discharged into the Mediterranean Sea. It is surrounded by 582 ha of agricultural land, supplied with 3.4 hm^3 year^{-1} by ground water extraction and flooding irrigation. This land is a non-sensitive area, according to UWWTD, but a vulnerable zone according to the Nitrates Directive (NiD) (91/676/EC). The polluter price in this case included $C_{Discharged\ fee}$ together with the cost of the treatment.

2.6.1.1. Water and nutrients needs. Citrus fruit is the most abundant crop in the study area. Citrus trees need water the whole year round with a net requirement of 3145 m^3 ha^{-1}, according to the data provided by the Jucar River Basin Authority (CHJ). Citrus N and P needs were considered as follows: 260 kg N·ha^{-1}·year^{-1} and 80 kg P·ha^{-1}·year^{-1} for flood irrigation and 220 kg N·ha^{-1}·year^{-1} and 70 kg P·ha^{-1}·year^{-1} for drip irrigation (López Bellido et al., 2010; Valencian Ministry of Agriculture, 2018; Environment; Condom et al., 2012).

2.6.1.2. Analyzed scenarios. The scenarios studied for Oliva WWTP are shown in Fig. 3. SI (a) represents the current situation, in which all the treated wastewater is discharged into the environment, while all the water needed for irrigation is extracted from the ground. SI includes pumping costs as well as the continuous impact on the availability of fresh water.

EA energy consumption is in the range of 0.35–0.42 kWh·m^{-3} (Table 1). In the case of the Oliva WWTP, a value of 0.375 kWh·m^{-3} was considered due to the treatment flow and the extent of N removal. Although SI meets the UWWTD requirements, it is still far from complying with the CE objectives, since water, energy and nutrients are not re-used. Fertigation could be used for agricultural water and nutrient demands, as shown in Scenarios II, III and IV (Fig. 3), in which no groundwater is extracted, with consequent savings. SII includes fertigation after wastewater treatment, with reclaimed water that contains concentrations of N and P of 15 and 6 mg L^{-1}, respectively. In SIII it is assumed that 100% of WWTP inflow is treated with AnMBR technology instead of EA, providing a nutrient-rich effluent with concentrations similar to the usual values in WWTPs influent (50 mg N·L^{-1} and 8 mg P·L^{-1} according to EPSAR) and biogas generated by anaerobic digestion of organic matter in wastewater. Although the Oliva WWTP discharges into a non-sensitive area, an SIV was evaluated (Fig. 3) in which wastewater is treated by two parallel lines (AnMBR and EA) to reduce the total nutrients released into the environment.

2.6.2. Peschiera Borromeo WWTP

The Peschiera Borromeo WWTP is currently treating an average inflow rate of 78.84 hm^3 year^{-1} (566,000 p.e.) by two water lines that receive wastewater from the Milan metropolitan area. Line 1 consists of a CAS process followed by tertiary treatment (disinfection with peracetic acid) while line 2 consists of an upflow biological filtration for nutrient removal and UV disinfection. This WWTP is situated in the municipality of Peschiera Borromeo (Lombardy, Italy) and discharged into the Lambro River, which was declared a sensitive area by the UWWTD and a vulnerable zone by the NiD. It should be noted that the Italian regulations set the total nitrogen (N_T) and total phosphorus (P_T) standard for water re-use at 15 and 2 mg L^{-1}, respectively, so that the WWTPs must remove the nutrients before water reclamation. This is one of the major economic barriers to possible fertigation and in this case study the polluter price includes the cost of chemical precipitation and disinfection.

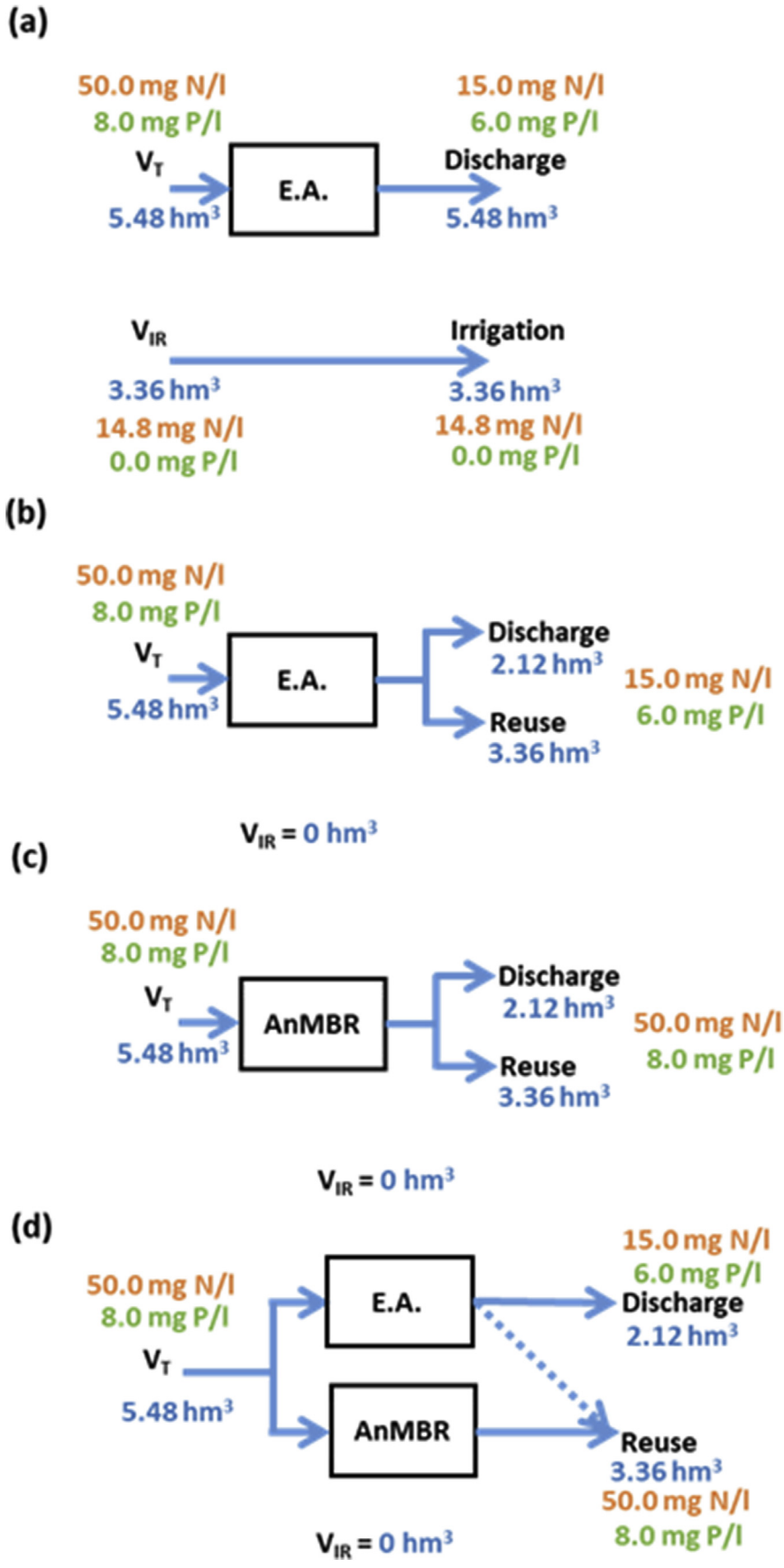

Fig. 3. Scenarios in Oliva WWTP case study: (a) SI, (b) SII, (c) SIII and (d) SIV.

2.6.2.1. Water and nutrients needs. The surrounding agricultural land which could potentially be irrigated with reclaimed water has an extension of 1,500 ha and its water demand is 12.03 hm^3 $year^{-1}$. This demand could be covered either by surface water or by a flow rate from the WWTP's water line 2. This plant is in a peri-urban agricultural park (Parco Agricolo Sud Milano) whose main crop is tomatoes. The average water requirements are 7.318 m^3 ha^{-1} from April to September (183 days). Tomato N and P needs for drip irrigation systems are 160 kg $N \cdot ha^{-1} \cdot year^{-1}$ and 20 kg $P \cdot ha^{-1} \cdot year^{-1}$ (Valencian Ministry of Agriculture; Environment; Condom et al., 2012).

2.6.2.2. Analyzed scenarios. The scenarios studied in the Peschiera Borromeo WWTP are depicted in Fig. 4. SI represents the current situation, in which all the treated wastewater is discharged into the environment, while all the irrigation water is extracted from a fresh water source, affecting both the resource and the surrounding ecosystem. Although a large amount of energy is required for WWT, SI is the common situation in many parts of Italy due to the restrictive legislation, long distances, unfavorable slope between the WWTPs and irrigation areas, and the high monitoring and distribution costs (Ventura et al., 2019).

In SII a fraction of water flow treated in the CAS process bypasses the nutrient removal process and is used for fertigation. This treatment scheme means that the bypass flow can be modified or even cancelled according to the demand for nutrients. The possibility of mixing treated water is also included (dashed line). SIII is proposed with the aim of maximizing energy and nutrient recovery, complying with CE principles. In this scenario, a fraction of the influent is treated by AnMBR, which could reduce the total WWT energy consumption through biogas production, while generating a nutrient-rich effluent that would notably reduce fertilizer costs. In this scenario and in SII, a modification of the Italian regulations is assumed to allow either land application as a wastewater post-treatment, or the use of reclaimed water as a fertilizer product.

In SIV all wastewater is treated through a CAS system with nutrient removal and water is re-used for agriculture. SIV represents the current situation of some Italian WWTPs, such as Milano San Rocco and Milano Nosedo, where farmers supply part of the effluent for irrigation together with their own artificial fertilizers.

3. Results and discussion

This section presents and discusses the main results obtained for the two case studies analyzed.

3.1. Efficiency factors

Preliminary studies (not shown) allowed for the calculation of efficiency factors for conversion between gross and net irrigation volumes, which were applied in this study and are shown in Table 2. Peschiera Borromeo is more efficient since the distribution network consists of pipelines and the irrigation method used is the drip system. In the case of Oliva, the pipeline distribution and drip irrigation systems were included as an optimized version of each scenario, assuming transport, distribution and application efficiencies of 95%, 97% and 97%, respectively.

3.2. Oliva WWTP

3.2.1. Water balance

The current situation is represented by SI. In this scenario, 3.36 hm^3 y^{-1} of groundwater is used for irrigation (Fig. 3), which intensifies natural resource depletion. This consumption can be avoided by using reclaimed water for irrigation, which would improve the conservation of water bodies. In Scenarios II and III, 61.3% of the wastewater treated in the Oliva WWTP is reused.

A total supplied water efficiency of 54.5% was calculated (Table 2), showing that there are high water losses throughout the irrigation network due to the return flows and the irrigation method used (flooding). The application of an optimized irrigation system (pipeline irrigation network and drip feed) would significantly reduce water losses from 1.32 hm^3 $year^{-1}$ to 0.18 hm^3 $year^{-1}$ with the consequent increase in network efficiency.

The results show that irrigation system optimization is a key factor in minimizing water losses. As expected, the water balance calculated for the four scenarios in Oliva WWTP with an optimized irrigation network showed a smaller water re-use flow of 2.05 hm^3 $year^{-1}$.

3.2.2. Nutrient balance

Used as the baseline, SI discharges treated wastewater into coastal waters, releasing nutrients into the environment. In this scenario, the total crop nutrient demand is met by mineral fertilization.

The smaller discharge flow in SII reduces nutrient losses to the environment: 42.2% less N and 35.4% less P (Fig. 5). At the same time, SII recovers the P contained in the reclaimed wastewater: 20.6 kg $P \cdot ha^{-1} \cdot year^{-1}$, reducing mineral P addition by 29.4% (Fig. 5a).

The application of AnMBR technology in SIII reduces P losses by 18.9% (SIII compared to SI, Fig. 5a) while N discharged into the environment increases from 118.7 t $year^{-1}$ in SI to 178.6 t $year^{-1}$ in SIII. The reason behind this result lies in the fact that the N content in the AnMBR effluent is higher (from 15 to 50 mg L^{-1}) than for P concentration (from 6 to 8 mg L^{-1}, Fig. 3), so that the reduction in discharged flow in SIII compensates for the increase in the effluent's P concentration, whereas in the case of N its higher AnMBR effluent concentration causes higher N losses, even though the discharge flow is smaller. In any case, SIII can reduce the N and P added by mineral fertilizers by up to 71.6% and 39.2%, respectively (Fig. 5a). SIV has the same added mineral needs as SIII and reduces nutrient losses with respect to Scenarios I and III.

It should be noted that the application of an optimized irrigation system under all circumstances not only reduces water losses, but also nutrient losses and therefore the amount of N and P released through the network was lower in the four optimized scenarios (Fig. 5). This entails an extra environmental benefit for groundwater bodies.

3.2.3. Economic balance

The results obtained from the economic analysis are shown in Table 3. The operating costs of AnMBR wastewater treatment are 63.4% lower in SIII and 39% lower in SIV. The coastal body discharge fee in Scenarios II to IV is reduced in proportion to the amount of water reclaimed. Disinfection costs only apply in SII, since membranes are used in Scenarios III and IV. A pumping cost is required in all the scenarios, but groundwater extraction pumping costs disappear in SII to SIV. Fertilizer costs are also reduced due to fertigation, especially when the reclaimed wastewater is obtained from AnMBR.

As explained above, the authors propose to maintain the total cost for polluters and farmers in all cases equal to that of SI (278.9 and 202.0 k€ $\cdot$ $year^{-1}$, respectively). A flow from the WWTP to the RWM would be possible in the water re-use scenarios, since WWTP cost would actually be smaller due to lower discharge fees and C_{WWT}. Similarly, due to savings in fertilizers and groundwater pumping, part of the farmers' expenses could be transferred to the RWM, who would have a total income of 0.029 (SII), 0.099 (SIII) and 0.080 (SIV) € $\cdot$ m^{-3}_{reused} (Table 3).

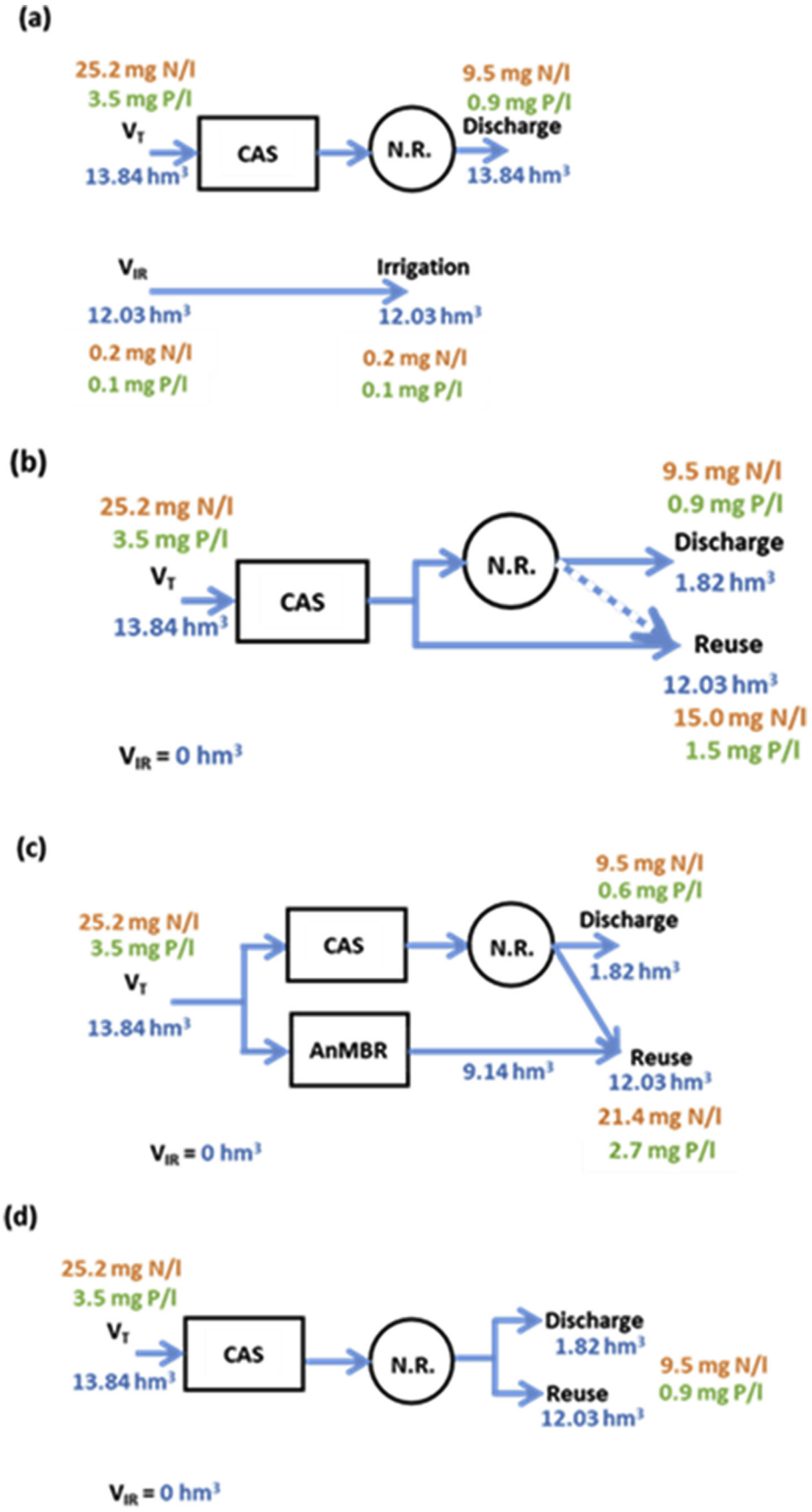

Fig. 4. Scenarios in Peschiera Borromeo WWTP case study: (a) SI, (b) SII, (c) SIII, (d) SIV.

Table 2
Return flows and supplied water efficiency for both Oliva and Peschiera Borromeo case studies.

Irrigation system	(%)	Oliva[a]	Peschiera Borromeo[b]
Transport network	Water loss by evaporation (p_t)	0.3	0.0
	Water loss through return flow (r_t)	14.7	3.0
	Efficiency (e_t)	85.0	97.0
Distribution network	Water loss by evaporation (p_d)	0.0	0.0
	Water loss through return flow (r_d)	15.0	3.0
	Efficiency (e_d)	85.0	97.0
Irrigation method	Water loss by evaporation (p_a)	8.1	2.0
	Water loss through return flow (r_a)	16.5	1.0
	Efficiency (e_a)	75.4	97.0

[a] Percentages calculated from the data supplied by Júcar River Basin Authority.
[b] Percentages calculated for a pipeline network.

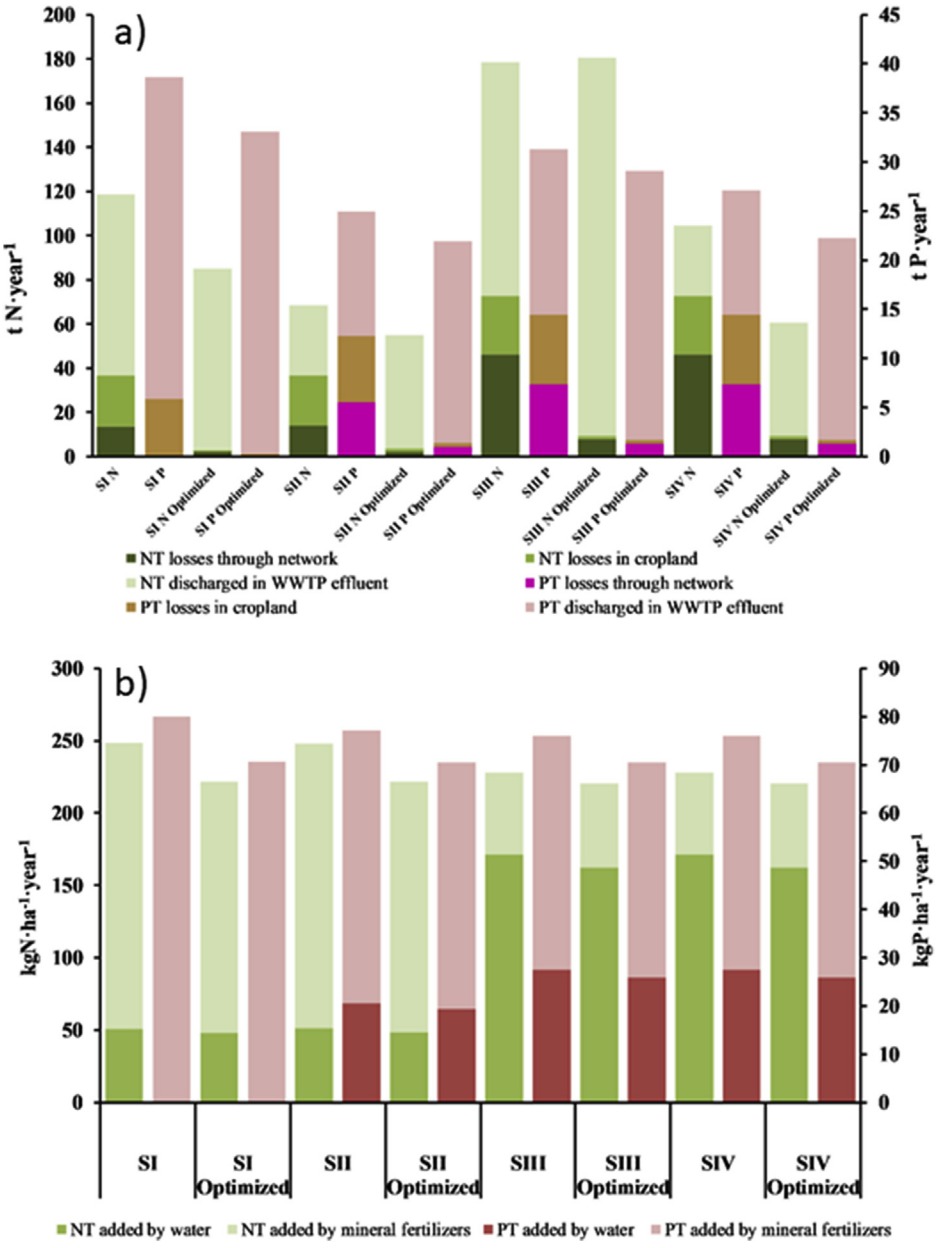

Fig. 5. a) Nutrients discharged into the environment in the four scenarios proposed in Oliva WWTP; b) Results of nutrient balance applied to cropland in the four scenarios proposed in Oliva WWTP.

Table 3
Economic results from the four scenarios studied in Oliva WWTP.

Costs	Units	SI	SI opt	SII	SIIopt	SIII	SIIIopt	SIV	SIVopt
Wastewater treatment C_{WWT}	k€·year^{-1}	257.0	257.0	257.0	257.0	93.9	93.9	156.8	195.7
Discharge fee $C_{Discharged\ fee}$	k€·year^{-1}	21.9	21.9	8.5	13.7	8.5	13.7	8.5	13.7
WWTP cost	k€·year^{-1}	*278.9*	*278.9*	*265.5*	*270.7*	*102.4*	*107.6*	*165.3*	*209.4*
Extra treatment for reuse $C_{rteat_for_reuse}$ (4)	k€·year^{-1}	0.0	0.0	15.0	9.0	0.0	0.0	0.0	0.0
Pumping for reuse $C_{Pumping\text{-}WWT}$ (3)	k€·year^{-1}	0.0	0.0	16.0	9.7	16.0	9.7	16.0	9.7
WWTP + RWTP cost	k€·year^{-1}	*278.9*	*278.9*	*296.5*	*289.4*	*118.4*	*117.3*	*181.3*	*219.1*
Fertilisers $C_{Fertilizers}$	k€·year^{-1}	92.0	81.1	87.0	77.2	31.0	31.1	31.0	31.1
Pumping groundwater $C_{Pumping}$	k€·year^{-1}	110.0	67.3	0.0	0.0	0.0	0.0	0.0	0.0
Farming cost	k€·year^{-1}	*202.0*	*148.4*	*87.0*	*77.2*	*31.0*	*31.1*	*31.0*	*31.1*
Total C_{Total}	k€·year^{-1}	**480.9**	**427.3**	**383.5**	**366.6**	**149.4**	**148.4**	**212.3**	**250.2**
Flow WWTP to manager (2)	k€·year^{-1}	0.0	0.0	13.4	8.2	176.6	171.3	113.7	69.5
Flow farmer to manager (5)	k€·year^{-1}	0.0	0.0	115.0	71.2	171.0	117.4	171.0	117.4
RWM Balance	k€·year^{-1}	0.0	0.0	97.4	60.5	331.6	279.0	268.7	177.1

The results show the economic viability of AnMBR technology, since the total cost was reduced from 480.9 k€·year^{-1} (SI) to 149.4 k€·year^{-1} (SIII) or 212.3 k€·year^{-1} (SIV). At the same time, Scenarios III and IV do not only meet the UWWTD and NiD requirements, but also reduce the costs associated with fertilizers and pumping. This means that the RWM budget can be devoted to covering the reclaimed water management expenses (including the WRRMP).

The economic balance for the four scenarios shows that, while the wastewater treatment costs with and without optimized irrigation networks remain constant for Scenarios I to III, discharge fees increase and the available money flow from the WWTP to the RWM is reduced. The same wastewater flow needs to be treated per year but a higher flow of treated water is discharged since a smaller water flow is reclaimed. On the other hand, as the reclaimed water flow is only 61% of that in the non-optimized scenarios, disinfection and pumping costs and mineral fertilization need to be reduced (Table 3). Water re-use costs are therefore reduced due to the smaller water flow needed by a more efficient system.

In SI, where treated water is not reclaimed, groundwater pumping costs also decreased for the same reason. A lower farmer's price was thus obtained for the optimized scenarios (148.4 k€·year^{-1}) and the following total RWM income reduction was calculated: 37.7% in SII, 15.9% in SIII and 34.1% in SIV. The reasons for this are the downscaled re-use system, while wastewater treatment costs remain the same (except for SIV, where two different technologies with two different costs are involved), together with the decrease in the farmer's price due to the lower cost of optimized SI.

An optimized irrigation system saves water ready for re-use and reduces its cost, with a lower RWM budget. However, certain strategies could be applied to increase the RWM economic balance, including subsidies, changes in discharge fees, extension of the irrigated area, etc.

3.2.4. Carbon dioxide emissions

Carbon dioxide emissions are associated with the energy cost of mineral fertilizer production, wastewater treatment, disinfection, pumping and energy recovered as biogas. Water re-use by conventional treatment technologies (SII) would reduce emissions by between 6.0 and 15.2% CO_2, depending on whether optimized or non-optimized scenarios are considered. AnMBR technology (SIII) would provide the biggest environmental improvement in terms of CO_2 emissions, since it would reduce them by between 74.7% and 75.3% CO_2 as a consequence of the high N content in the reclaimed water and the biogas generated. An improvement is also observed when comparing Scenarios II to IV, with reclaimed water, to SI, where no water is reclaimed.

As expected, the optimized Scenarios I to III show an improvement in CO_2 emissions, due to reduced fertilizer use, pumping and disinfection (when applied) (Fig. 6). The greatest improvement, when optimized, is obtained for SI (11%). In SIV, given the fact that the amount of reused nutrients is equal to that in SIII, the CO_2 emissions associated with fertilizer production are reduced by the same proportion. The high energy consumption in the EA process and the absence of energy recovery in this treatment produce higher CO_2 emissions than SIII. This scenario, in which AnMBR is combined with an optimized irrigation system, provided the highest environmental benefit in terms of total CO_2 emissions (reduction of up to 1153 t CO_2·year^{-1}).

The study carried out in Oliva WWTP (Spain) showed that in this case AnMBR is suitable for the combined purpose of wastewater treatment and fertigation, since it maximizes energy and nutrient recovery and reduces both costs and CO_2 emissions.

The positive impact of AnMBR would be still higher, if groundwater conservation could be quantified. Applying this technology in vulnerable zones requires risk assessment plans, including nutrient balances. Methods such as drip irrigation are recommended, since they generate smaller return flows than flood irrigation.

3.3. Peschiera Borromeo WWTP

3.3.1. Water balance

In the present situation (SI) 12.03 hm^3 year^{-1} of fresh water is used for irrigation (Fig. 4), depleting natural resources. Fertigation (Scenarios II and III) or irrigation (SIV) would improve the conservation of water bodies. In these scenarios, 87% of the wastewater treated in the Peschiera Borromeo WWTP would be re-used. The irrigation network in the surrounding area was designed with pipelines and the present irrigation method is by drip, so that the results are those of an optimized irrigation system.

3.3.2. Nutrient balance

SI is the situation in which the highest amount of nutrients is discharged into the environment (Fig. 7a), which was expected in a situation that does not re-use treated wastewater. Scenarios II to IV show the expected lower nutrient discharge into the environment, with a 7.6 times smaller discharged water flow in all cases. Total N losses fall by 77.0% (SII), 73.5% (SIII) and 79.9% (SIV) with respect to SI. P losses are 79.9%, 74.3% and 82.7% less for Scenarios II, III and IV, respectively and with respect to SI. The biggest difference between the scenarios lies in the nutrient losses through the network, which are greater in SIII, with the highest nutrient content of reclaimed water.

In SI almost all the required nutrients are supplied by mineral fertilizers, whereas higher percentages of nutrients are provided by

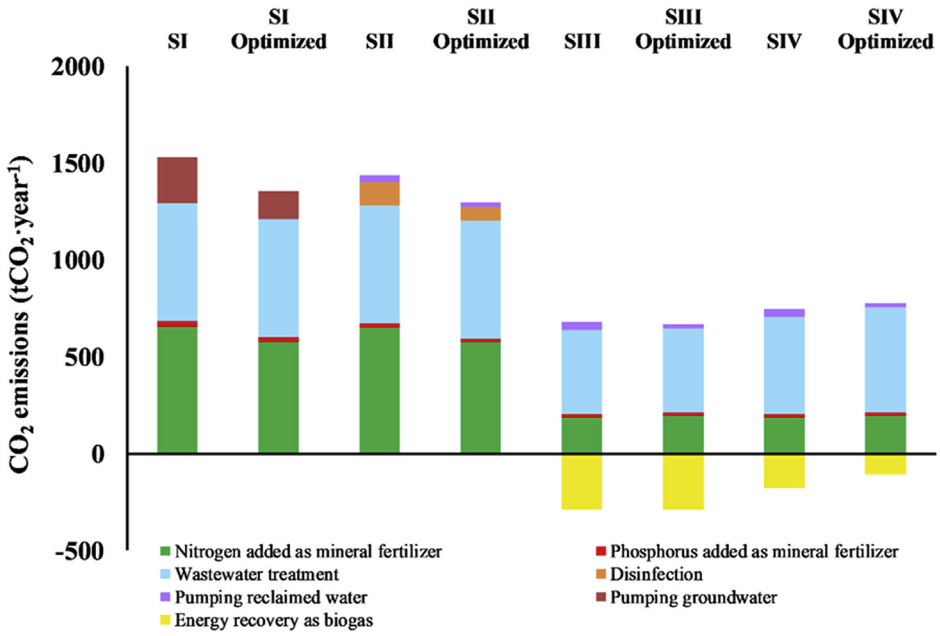

Fig. 6. CO_2 emissions for the four scenarios studied in Oliva WWTP.

the reclaimed water in Scenarios II, III and IV (70%, 100% and 44%, respectively) (Fig. 7b). SIII illustrates the flexibility that AnMBR technology gives to the system. In fact, water quality can be modified in terms of nutrient content, by mixing the reclaimed water with the effluent from the nutrient removal process, in such a way that no mineral fertilizers are needed.

Since there is no clear advantage regarding nutrient losses for any of the three water re-use scenarios, the economic and CO_2 emissions analyses become decisive.

3.3.3. Economic balance

Table 4 shows the results of the economic analysis. Disinfection and nutrient removal are considered part of the wastewater treatment cost. In fact, they are not an additional treatment because, according to the Italian regulations, nutrients must be removed from the final effluent up to the allowed quality standard. This cost in SI (512.9 k€·year^{-1}) was established as the price polluters pay in all scenarios. Treatment costs are lower in Scenarios II and III than in SI (6.1% and 40.1%, respectively). Since irrigation water has to be pumped either from surface water bodies or from WWTP effluent, all water scenarios include water pumping costs. Fertilizer costs decrease in the situations with water re-use, being zero when AnMBR technology is applied, which brings considerable savings in farming costs. Water re-use scenarios provide a relevant money flow from farmers to the RWM, especially in Scenarios II and III, due to the higher contribution of the WWTP effluent to fertilization needs.

Although SIV gives rise to the lowest nutrient losses to the environment (see Fig. 7a), it results in the highest total cost of all re-use scenarios. This is a direct consequence of removing all the nutrients from the influent, which means a higher quantity of mineral fertilizers is needed. This, in turn, results in fewer economic resources available for the RWM and shows that the current situation in some Italian WWTPs is not the optimal. AnMBR technology (SIII) could reduce the total cost of the water re-use system to 388.1 k€·year^{-1} due to the following factors: (i) there is no need for mineral fertilizers and (ii) treatment cost is 40.1% lower than SI, because of the lower energy consumption of the biological process and the reduced disinfection and chemical precipitation costs.

3.3.4. Carbon dioxide emissions

The highest CO_2 emissions were those of the current SI, followed by SIV, which represents the most frequently adopted alternative in the area (see Fig. 8). Water re-use with CAS treatment (SII) reduced CO_2 emissions by 46.4% with respect to the present situation, while the smallest carbon footprint was obtained for SIII, with AnMBR technology (−898.9 $tCO_2 \cdot year^{-1}$). On the one hand, the possible energy recovery as biogas is substantially higher with this wastewater treatment than with CAS, while on the other the avoidance of mineral fertilizers also prevented a major CO_2 emissions source. These results show that the AnMBR/fertigation combination can considerably reduce the carbon footprint and the financial costs in comparison with conventional treatments such as the CAS system, provided that a high percentage of the influent can be treated by AnMBR (in this case 86.8%) and application to farmland can be considered as a post-treatment.

3.4. End-user acceptance

The survey was disseminated among irrigators whose average annual water usage is 5000 m^3/ha. The responses revealed that 87.5% of those surveyed have at some point if not currently irrigated their crops with reclaimed water and 64% of these cases declared that they achieved an economic benefit in doing so. The study showed that scarcity problems are more frequent or are more important to the farmers surveyed than water quality problems. Finally, one of the main results of this study was that all (100%) the irrigators surveyed claimed that they would be willing to use water from a wastewater treatment plant if they could be sure it is of high quality, which is consistent with their acknowledgement that reclaimed water guarantees the water supply.

4. Conclusions

Reclaimed water is a reliable resource that can be associated with a relevant renewable nutrient supply. It is independent of

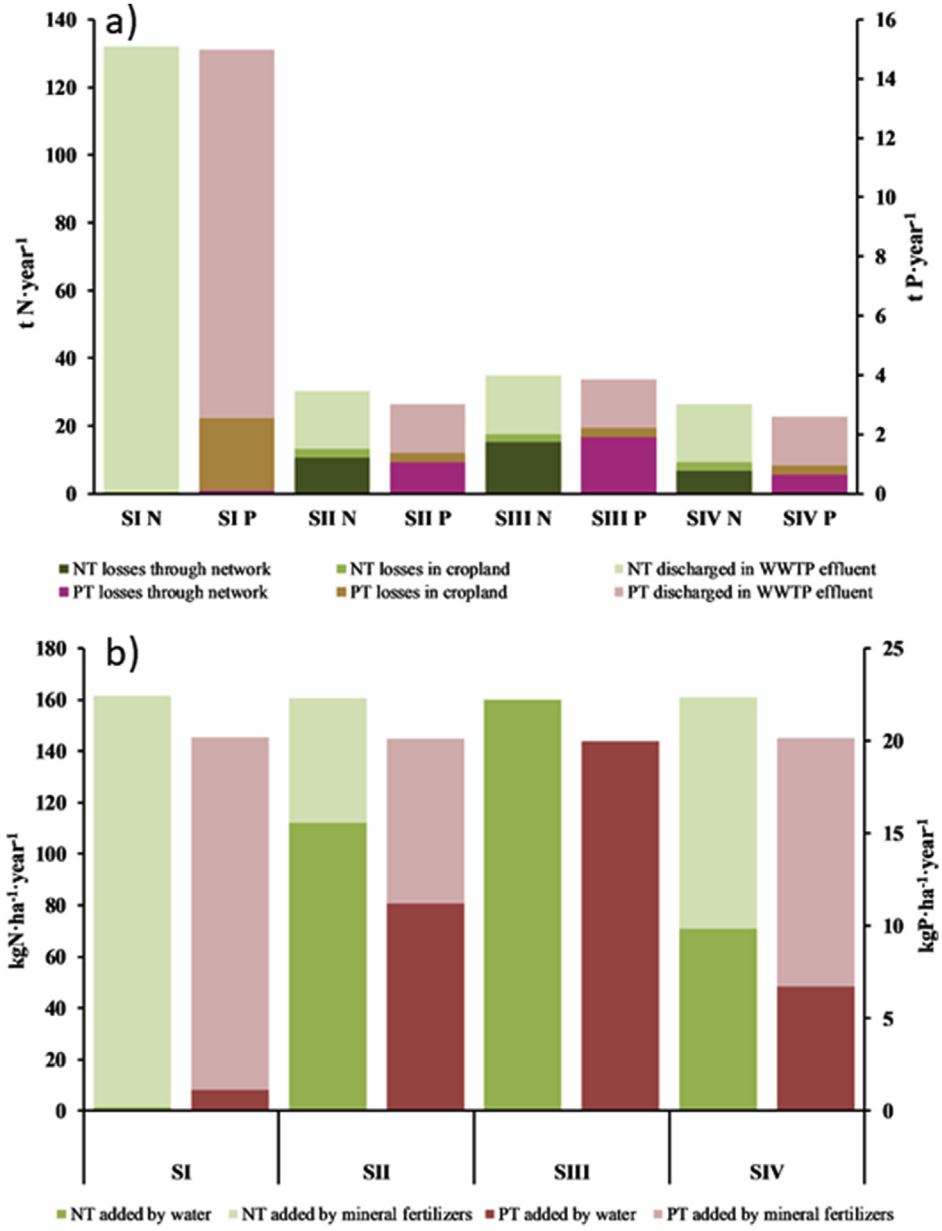

Fig. 7. a) Nutrients discharged into the environment in the four scenarios studied in Peschiera Borromeo WWTP; b) Nutrients balance applied to cropland in the four scenarios studied in Peschiera Borromeo WWTP.

Table 4
Economic results from the four scenarios studied in Peschiera Borromeo WWTP.

Costs	Units	Scenario I	Scenario II	Scenario III	Scenario IV
Wastewater treatment C_{WWT}	k€·year^{-1}	407.0	407.0	261.0	407.0
Disinfection $C_{Disinfection}$	k€·year^{-1}	69.8	69.8	23.7	69.8
Chemical precipitation	k€·year^{-1}	36.1	4.7	22.6	36.1
WWTP cost	k€·year^{-1}	*512.9*	*481.5*	*307.3*	*512.9*
Pumping for reuse $C_{Pumping\text{-}WWT}$	k€·year^{-1}	0.0	80.8	80.8	80.8
RWTP cost	k€·year^{-1}	*512.9*	*562.3*	*388.1*	*593.7*
Fertilisers $C_{Fertilizers}$	k€·year^{-1}	170.8	53.2	0.0	97.3
Pumping surface water $C_{Pumping}$	k€·year^{-1}	80.8	0.0	0.0	0.0
Farming cost	k€·year^{-1}	*251.6*	*53.2*	*0.0*	*97.3*
Total C_{Total}	k€·year^{-1}	**764.5**	**615.5**	**388.1**	**691.0**
Flow WWTP to manager	k€·year^{-1}	0.0	31.4	205.6	0.0
Flow farmer to manager	k€·year^{-1}	0.0	198.5	251.7	154.4
RWM Balance	k€·year^{-1}	0.0	149.0	376.4	73.6

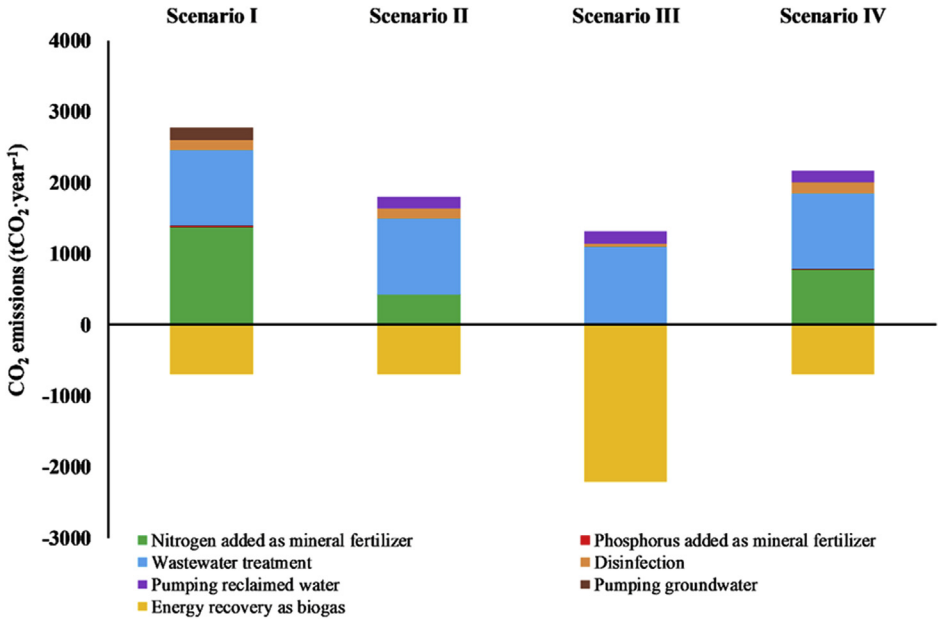

Figure 8. CO_2 emissions for the four scenarios studied in Peschiera Borromeo WWTP.

seasonal droughts and weather variations and thus acts as an alternative resource to alleviate pressure on freshwater sources, while reducing the economic and environmental costs of mineral fertilizers. However, a risk management plan is needed to ensure environmental and health safety when reclaimed water is used for both irrigation and fertigation.

This work has shown the positive economic balance that can be obtained in different water reuse scenarios. The authors propose that savings can be used for drawing up, updating and implementing the Water Re-use Risk Management Plans under the responsibility of the new *Reclaimed Water Manager*.

Since AnMBR technology can also recover energy from the wastewater flow, its application presents an optimal scenario according to Circular Economy principles and environmental sustainability. In this regard, the combination of AnMBR technology and fertigation can significantly reduce CO_2 emissions.

Lastly, in order to comply with the current legislation, future combined AnMBR and fertigation demonstration projects need to be carried out in non-sensitive areas, following the ID conclusions. These trials should include environmental and health risk assessments, agronomic studies and social and economic analyses to obtain comprehensive information of the proposed water management scheme.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Antonio Jiménez-Benítez: Writing - original draft, Investigation, Methodology, Formal analysis, Writing - review & editing. **Francisco Javier Ferrer:** Investigation, Methodology, Resources. **Silvia Greses:** Investigation, Formal analysis. **Ana Ruiz-Martínez:** Writing - original draft, Formal analysis. **Francesco Fatone:** Resources, Investigation, Validation. **Anna Laura Eusebi:** Resources, Investigation, Validation. **Nieves Mondéjar:** Investigation, Methodology, Resources. **José Ferrer:** Methodology, Formal analysis, Investigation, Validation. **Aurora Seco:** Conceptualization, Investigation, Methodology, Supervision, Writing - review & editing.

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A semi-industrial scale AnMBR for municipal wastewater treatment at ambient temperature: performance of the biological process

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ABSTRACT

A semi-industrial scale AnMBR plant was operated for more than 600 days to evaluate the long-term operation of this technology at ambient temperature (ranging from 10 to 27 °C), variable hydraulic retention times (HRT) (from 25 to 41 h) and influent loads (mostly between 15 and 45 kg COD·d^{-1}). The plant was fed with sulfate-rich high-loaded municipal wastewater from the pre-treatment of a full-scale WWTP. The results showed promising AnMBR performance as the core technology for wastewater treatment, obtaining an average 87.2 ± 6.1 % COD removal during long-term operation, with 40 % of the data over 90%. Five periods were considered to evaluate the effect of HRT, influent characteristics, COD/SO_4^{2-}-S ratio and temperature on the biological process. In the selected periods, methane yields varied from 70.2±36.0 to 169.0±95.1 STP L CH_4·kg^{-1} COD_{inf}, depending on the influent sulfate concentration, and wasting sludge production was reduced by between 8 % and 42 % compared to conventional activated sludge systems. The effluent exhibited a significant nutrient recovery potential. Temperature, HRT, SRT and influent COD/SO_4^{2-}-S ratio were corroborated as crucial parameters to consider in maximizing AnMBR performance.

1. Introduction

Anaerobic treatment of municipal wastewater (MWW) has several advantages over conventional aerobic processes, as has been widely recognized over the years. Its main advantages are: low sludge production due to low anaerobic biomass yield (Giménez et al., 2011); reduced energy consumption because no aeration is required (Lee et al., 2017); methane is produced from organic matter valorization, which improves the technology's energy balance (Krzeminski et al., 2017); low greenhouse gas emissions when methane is recovered from both biogas and effluent streams (Jiménez-Benítez et al., 2020b); and potential recovery potential of nutrients content (Batstone and Virdis, 2014). Moreover, the limitations associated with anaerobic processes applied to low-organic strength wastewaters (e.g. low growth rate of anaerobic biomass at ambient temperature or poor sludge settling properties) can be overcome by combining membrane technology and anaerobic reactors in the so-called anaerobic membrane bioreactor (AnMBR) (Ozgun et al., 2013), where the filtration process allows complete physical biomass retention. Hydraulic retention time (HRT) and sludge retention time (SRT) can thus be decoupled, which means high-rate reactors can be designed and high-quality effluent suitable for water reclamation can be achieved with the appropriate membrane pore-size (Jiménez-Benítez et al., 2020a). Eventually, membrane modules are more compact than the equivalent secondary settlers + disinfection units (e.g. UV process), which helps to reduce the layout footprint of wastewater treatment facilities and permits existing space-constrained treatment plants to be retrofitted (Xiao et al., 2019).

A high concentration of nutrients (mainly nitrogen and phosphorus,

Abbreviations: AnMR, anaerobic membrane bioreactor; BMP, biomethane potential; BOD_5, biological oxygen demand; CAS, conventional activated sludge; CE, circular economy; COD, chemical oxygen demand; GHG, greenhouse gas; HRT, hydraulic retention time; MA, methanogens; MWW, municipal wastewater; N, nitrogen; OLR, organic loading rate; P, phosphorus; SRB, sulfate-reducing bacteria; SRT, sludge retention time; STP, standard temperature and pressure; TSS, total suspended solids; VFA, volatile fatty acids; VSS, volatile suspended solids; WSP, waste-sludge production; WWTP, wastewater treatment plant; Y^{CH_4}, methane yield.

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but also magnesium, potassium, etc.) in the effluent of anaerobic processes has traditionally been considered a significant environmental risk to aquatic ecosystems (eutrophication) and potable water resources when these streams are discharged (Pretel et al., 2016; Rodriguez-Garcia et al., 2011). This being true, it is also possible to consider it as an opportunity to introduce the fundamentals of the circular economy (CE) to wastewater treatment, since anaerobic treatments can be used to recover nutrients. Recovery plus recycling would contribute to closing both the materials and energy loops, and several processes and technologies are already available or being investigated to take advantage of these essential resources for agriculture and industry (Robles et al., 2020a). Including these resources in a comprehensive nutrient management system is becoming more important, as shown, among others, by the interest shown by the European Union in this matter (European Commission, 2015). At the present time, most efforts focus on the P since this element and phosphorite were included in European Critical Raw Materials lists in 2017 and 2014, respectively. However, N recovery is also attracting attention because of the high energy consumption and associated greenhouse gas emissions from its fixation from atmospheric N_2. Nutrient recovery thus helps to reduce both the dependence on their production and/or imports, which is even more important for those that are exhaustible such as phosphorus and potassium, and greenhouse gases (GHG) associated with mineral fertilizers, whose production results in significant energy expenditure (McCarty et al., 2011) and has an environmental impact.

Biosolids production has also been identified as a key issue in wastewater treatment; aerobic processes are characterized by high sludge production associated with a high biomass yield of aerobic metabolism, which involves a significant management cost that affects the economic and environmental performance of sewage treatments. Additionally, the regulation of waste sewage sludge, which is a challenge in wastewater treatment processes, is also expected to be tightened (Collivignarelli et al., 2019). AnMBR can help to overcome these problems in two ways: firstly, the anaerobic process mineralizes organic matter with reduced anabolism and low sludge production. By a way of example, Foladori et al. (2015) reported a sludge production in aerobic WWTPs of 0.25-0.35 kg VSS per kg COD removed (unstabilized sludge) and around 0.18 kg VSS per kg COD removed for stabilized sludge, respectively, while Robles et al. (2020) reported a stabilized sludge production in an AnMBR demonstration plant treating MWW of 0.148 kg VSS per kg COD removed. Secondly, biomass retention by membrane modules allows increasing SRT, which contributes to achieve on-site sludge stabilization and reduces or even avoids further sludge treatment.

A final consideration is that anaerobic removal of organic matter generates methane, a storable energy carrier suitable to produce renewable heat and power. Boosting anaerobic processes through AnMBR may thus improve the energy balance of existing wastewater treatment facilities. Using it as the core technology would take full advantage of the organic matter and the 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, despite these indisputable benefits, AnMBR technology presents three fundamental challenges: i) competition between sulfate-reducing bacteria (SRB) and methanogens (MA) for the same substrates reduces methane yields when treating influents with high sulfate contents (Durán et al., 2020); ii) reduction of sulfate by SRB produces sulfide and hydrogen sulfide, which eventually could entail inhibition in anaerobic metabolism and needs to be considered in energy recovery and water reclamation processes; and iii) dissolved methane losses through the effluent reduce energy recovery and, especially, increase GHG emissions (Jiménez-Benítez et al., 2020b). In this sense, co-digestion (e.g. the organic fraction of municipal waste, food waste, etc.) has been explored to increase influent organic load and thus increase methane production (Karki et al., 2021; Moñino et al., 2017), while several promising technologies for dissolved methane recovery (e.g. PDMS membranes) are also being studied (Sanchis-Perucho et al., 2020; Crone et al., 2016).

AnMBR technology is thus a clear candidate for renewing the wastewater treatment approach and would contribute to the economic, environmental and social goals of water management, with CE playing an active role. Persevering with a business-as-usual scenario based on conventional technologies, which have been shown to be effective but not efficient, would entail ignoring the current global context: climate change, energy transition, resource depletion, and environment degradation.

This work describes the results of operating of an AnMBR semi-industrial plant (demonstration scale, TRL of 6) for MWW treatment at the Alcázar de San Juan WWTP (Alcázar de San Juan, Ciudad Real, Spain). The size of the prototype, together with being fed with real MWW and operated for more than 2.5 years, means it can be considered a reliable and robust source of performance data. The main objective of the study was to contrast AnMBR with conventional wastewater treatments and demonstrate its potential benefits.

2. Materials and methods

2.1. AnMBR description and operation

Fig. 1 shows the process and instrumentation diagram (P&ID) of the semi-industrial AnMBR plant operated in this work. This plant mainly consists of a 40 m^3 (c.a. 34.4 m^3 working volume + 5.6 m^3 headspace) anaerobic reactor connected to three 0.8 m^3 membrane tanks (c.a. 0.7 m^3 working volume + 0.1 m^3 headspace). Each membrane tank was fitted with one ultrafiltration membrane system (PURON® PSH41, KMS, 0.03-μm pore size, total filtration area of 41 m^2), giving a total filtration area of 123 m^2. A sieve screw (RT, 1.5-mm screen size), an equalization tank (1.1 m^3) and a clean-in-place tank (0.37 m^3) were the other main elements in the plant. Total sludge volume was recorded considering the working volumes of both the anaerobic reactor and membrane tanks, accounting for 36.32±1.63 m^3 during the operating period. The AnMBR was equipped with a 2.1 m^2 degassing membrane unit for dissolved methane recovery, consisting of a polydimethylsiloxane (PDMS) hollow-fiber commercial module provided by PermSelect®, MedArray Inc. USA. The plant was fed with effluent from the pre-treatment step (sand and grease removal) of the Alcázar de San Juan WWTP (Alcázar de San Juan, Ciudad Real, Spain). Further details of this AnMBR system can be found in Robles et al. (2020).

Regarding filtration process, the system was able to operate with transmembrane flux in the range 14-21 LMH, with moderate sparging gas demand (11-14 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) and significant total suspended solids (TSS) (8-13 $g \cdot L^{-1}$). Transmembrane pressures varied between 87 and 490 mbar and fouling rates between 1.7 and 16.6 $mbar \cdot d^{-1}$. Only two chemical cleanings of membrane were needed during the considered experimental period.

The semi-industrial plant performance was assessed from two points of view: (i) considering the 610 days of operations included in this work; and (ii) selecting 5 periods from the overall operating period. It is important to highlight the variable influent quality and the relatively high COD and BOD concentrations due to the contributions of several nearby dairy and wine industries. During the operation period and based on data obtained in previous AnMBR pilot plant studies (Giménez et al., 2014, 2012; Seco et al., 2018), SRT was maintained at 70 days and the plant was operated at two different HRT (25-26 and 41 hours). Temperature varied freely between 10 and 27 °C (ambient temperature operation), while OLR was in the range between 0.60±0.20 and 1.28 ±0.38 kg $COD \cdot m^{-3} \cdot d^{-1}$. Table 1 shows the characterization of the influent and effluent streams together with the main operating conditions during the five selected periods.

2.2. AnMBR monitoring

The following parameters were regularly monitored in the influent:

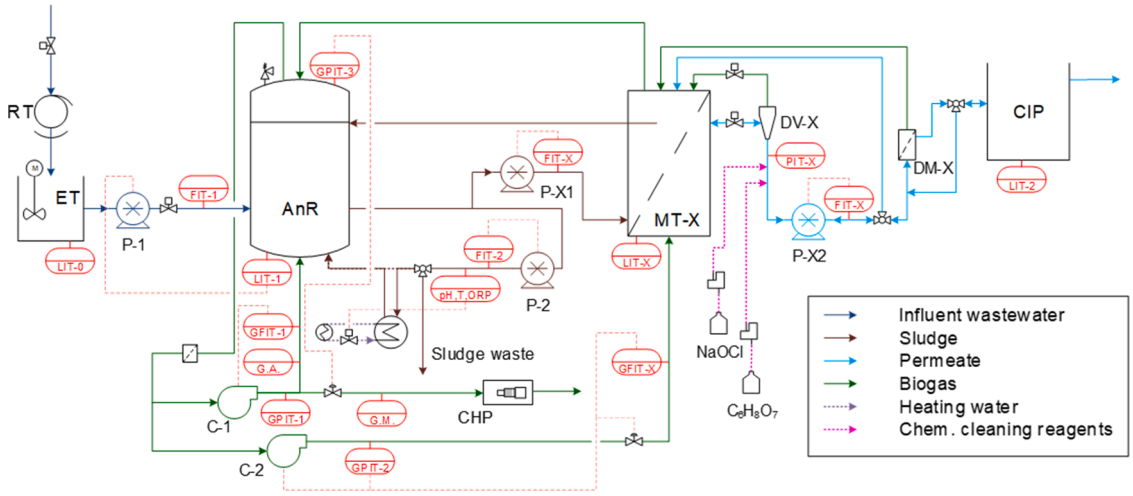

Fig. 1. Process and Instrumentation Diagram (P&ID).

Table 1
Influent and effluent characterization and operation conditions during the studied periods.

KEY REACTOR PARAMETERS	Units	I	II	III	IV	V
SRT	d	70±1	68±2	70±2	71±2	70±0
HRT	h	41±1	25±1	26±2	26±2	41±13
T	°C	27±1	24±2	19±1	27±1	18±2
INFLUENT	**Units**	**I**	**II**	**III**	**IV**	**V**
$Q_{treatment}$	$L{\cdot}d^{-1}$	21228±591	33016±2773	33243±3118	34207±1478	20984±7482
TSS	$mg{\cdot}L^{-1}$	462±188	418±293	332±145	354±116	430±120
VSS	%	64.6±5.9	82.0±11.5	63.1±14.6	75.5±9.6	80.1±13.0
COD	$mg{\cdot}L^{-1}$	1285±429	1403±532	896±201	755±224	1038±238
OLR	kg $COD{\cdot}m^{-3}{\cdot}d^{-1}$	0.75±0.25	1.28±0.38	0.82±0.19	0.71±0.23	0.60±0.20
BOD_5	$mg{\cdot}L^{-1}$	772±277	911±288	569±122	451±154	581±180
N_T	$mg\ N{\cdot}L^{-1}$	52.0±13.3	54.7±16.3	46.8±6.9	46.0±11.2	35.6±8.0
P_T	$mg\ P{\cdot}L^{-1}$	10.1±2.8	8.6±2.1	8.5±1.2	6.8±1.3	7.6±1.1
PO_4^{3-}-P	$mg\ P{\cdot}L^{-1}$	2.5±0.0	5.8±2.1	6.0±0.9	5.3±1.3	3.9±1.7
S^{2-}	$mg\ S{\cdot}L^{-1}$	37.9±26.7	59.6±19.5	47.0±38.7	30.7±49.2	38.3±14.5
SO_4^{2-}-S	$mg\ S{\cdot}L^{-1}$	147.7±13.3	172.2±28.5	125.4±47.6	157.3±46.9	149.7±26.9
$COD/\ SO_4^{2-}$-S	$g\ COD{\cdot}g^{-1}$-S	8.70±3.69	8.15±4.44	7.14±4.31	4.80±2.86	6.94±2.83
EFFLUENT	**Units**	**I**	**II**	**III**	**IV**	**V**
COD	$mg{\cdot}L^{-1}$	91±31	116±24	121±15	80±17	79±18
N_T	$mg\ N{\cdot}L^{-1}$	47.9±6.2	54.0±14.6	47.3±4.3	52.5±10.2	37.0±11.1
P_T	$mg\ P{\cdot}L^{-1}$	8.8±1.9	9.9±1.9	6.9±2.8	6.3±0.5	7.8±1.2
S^{2-}	$mg\ S{\cdot}L^{-1}$	174.3±28.7	207.5±73.2	233.1±66.3	183.2±27.0	166.2±27.9
VFA	$mg\ COD{\cdot}L^{-1}$	8.8±13.7	21.1±45.0	41.3±27.6	0.0±0.0	5.4±10.9
CH_4	$mg{\cdot}L^{-1}$	16.0±0.5	17.1±0.8	17.9±0.3	16.3±0.5	18.7±0.6
REACTOR	**Units**	**I**	**II**	**III**	**IV**	**V**
TSS	$g{\cdot}L^{-1}$	8.4±0.5	12.6±0.5	11.3±1.0	10.4±0.8	8.3±1.2
VSS	%	70.3±5.5	68.1±1.8	72.8±3.8	65.7±2.7	68.9±5.9
pH		7.1±0.1	7.0±0.1	7.0±0.1	7.0±0.1	7.0±0.3
Q_{WS}	$L{\cdot}d^{-1}$	522±5	511±107	521±5	510±18	520±4
COD	$g{\cdot}L^{-1}$	11.4±0.6	15.8±1.1	14.7±2.0	13.2±0.5	13.0±0.4
Q_{biogas}	$L\ STP{\cdot}d^{-1}$	5396±4435	5558±3204	1901±597	1359±999	2241±924
$\%CH_4$	%	77±2	77±2	72±2	77±2	76±1

total COD and BOD_5, sulfate (SO_4^{2-}-S), sulfide (HS^--S), total nitrogen (N_T), total phosphorus (P_T), nitrate (NO_3^--N), phosphate (PO_4^{3-}-P), TSS, and volatile suspended solids (VSS). Effluent characterization comprised total COD, N_T, P_T and HS-S, and volatile fatty acids (VFA). Finally, reactor monitoring was based on TSS, VSS, COD, pH, temperature and methane content in the biogas (% CH_4). Dissolved methane in the effluent was calculated by Henry's law and assuming saturation conditions. pH, temperature and biogas flow rate were registered online. 24-hour-composite samples were taken from the influent twice a week for all measurements except for COD, which was analyzed daily. Grab samples were taken twice a week from the effluent and mixed liquor.

2.3. Analytical methods

TSS, VSS, COD, BOD_5, sulfate, sulfide, and nutrients were determined according to Standard Methods (APHA, 2005). VFA concentration was determined by titration according to the method proposed by Moosbrugger et al. (1992).

2.4. Data processing and calculations

Different key performance indicators were calculated to evaluate the performance of the system as shown in Robles et al., (2020), *i.e.* methane

yield taking into account the methane dissolved in the permeate, COD removal rates, and biosolids production.

3. Results and discussion

The performance of the demonstration plant was evaluated for 610 days. Five periods were further characterized to assess the robustness of the AnMBR-technology against the disturbances in the main operating conditions derived from the natural variations throughout the year.

3.1. Organic removal efficiency

Fig. 2 shows the 15-day moving average for the influent organic load along with its 95% confidence bands, the daily average temperature, and both the removed and biodegraded COD. The influent organic load was highly variable, mainly due to the aforementioned variable industrial discharges. Biodegraded COD accounted for the COD that was biologically removed by SRB or was transformed into methane by MA, whereas the removed COD also considered the physically retained COD (by the ultrafiltration process) besides the biodegraded COD. Conversely, the influent sulfate-concentration variation was moderate, resulting in different COD/SO_4^{2-}-S ratios for each period (see Table 1). The temperature followed the natural variations in mid-latitude temperate climates, with strong variations between day to night (data not shown) and summer to winter. The daily-average temperature difference from winter to summer was higher than 17 °C.

The HRT was varied to simulate population shifts during summer and winter periods as a result of population density changes during vacation periods. Initially, HRT was set to 41 hours in period P1, in accordance with the low population-density in coastal areas before the summer. HRT was almost halved from P1 to P2 (25±1 h) and P4 (26±2 h) to simulate the intensification of water usage by the massive influx of tourists in coastal areas during vacation periods. Similarly, P3 y P5 were operated at the same low temperature (19±1 and 18±1 °C, respectively) but different HRT (25±1 h in P3 and 41±13 h in P5) to simulate population variation during the winter period. HRT was kept practically constant from period P2 to period P4 (around 25 h), representing densely populated all-year-long areas. However, as shown in Table 1, the OLR decreased (1.28±0.38 kg COD·m^{-3}·d^{-1} in P2, 0.82±0.19 kg COD·m^{-3}·d^{-1} in P3 and 0.71±0.23 kg COD·m^{-3}·d^{-1} in P4) as a result of the drop in the influent COD concentrations (1403±532 mg·L^{-1} in P2, 896±201 mg·L^{-1} in P3 and 755±224 mg·L^{-1} in P4), which was attributed to changes in the activity of the industries that discharge into the sewer system. The sulfate concentration also decreased from period P2 (172.2±28.5 mg S·L^{-1}) to period P3 (125.4±47.6 mg S·L^{-1}), albeit the COD reduction was more significant, resulting in a lower COD/SO_4^{2-}-S ratio. Conversely, the sulfate concentration in period P4 (157.3±46.9 mg S·L^{-1}) was higher than in period P3 despite the fact that COD concentration continued to fall, both contributing to a significant reduction in the COD/SO_4^{2-}-S ratio. SO_4^{2-}-S concentration in period P5 (149.7 ±26.9 mg S·L^{-1}) remaining similar to P4. The increased influent COD concentration in period P5 (1038±238 mg·L^{-1}) thus resulted in a higher COD/SO_4^{2-}-S ratio. Despite the higher COD concentration, OLR was lower in period P5 (0.60±0.20 kg COD·m^{-3}·d^{-1}) than in period P4 as a result of the higher HRT in period P5.

Fig. 2 shows that COD removal accounted for an average of 87.2 ± 6.1 % of the influent COD. 87 % of the COD removal data was higher than 80 %, whilst almost 40 % of the COD removal data was over 90 %. The membranes were able to virtually retain all particulates. However, the occasional unbalanced performance of the different stages in anaerobic degradation due to low temperatures or organic load peaks accumulated intermediate soluble organics that were able to pass through the membrane, thus increasing the effluent COD and reducing COD removal.

COD was not completely biodegraded by SRB or by MA. Conversely, a fraction of the removed COD was simply retained within the system due to membrane filtration, helping to increase the solids concentration and therefore waste-sludge production. Regarding the biologically removed COD, sulfate concentration was virtually zero in the effluent, indicating that SRB completely reduced influent sulfate (2 grams of COD were consumed per gram of SO_4^{2-}-S reduced). The analysis of the variance ANOVA of the COD consumed by SRB (expressed as OLR-SRB, i.e., the influent OLR which is consumed by SRB) found a significant difference between the mean values in the different periods (P-value = 0.0) (see complementary material). Nevertheless, Fischer's least significant difference (LSD) method for the pairwise comparison of mean values in the different periods were fairly similar in the mean values for the OLR-SRB among periods P1 and P5, and between periods P2 and P4. While P3 had an intermediate mean value.

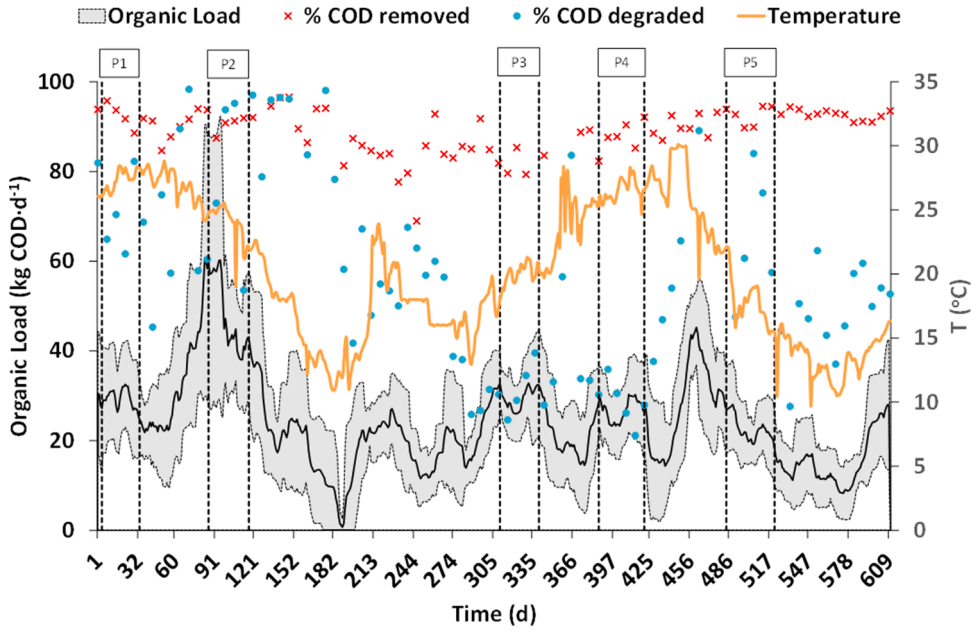

Fig. 2. Operating conditions during the experimental period.

The remaining COD was theoretically available for MA (i.e., OLR-MA) in the proportion shown in Fig. 3. As can be seen in this figure, the OLR in period P2 was much higher than the others, as a result of the strengthened influent wastewater organic load. During P2, 24.5 % of the OLR was consumed by SBR (OLR-SBR) while 75.5 % remained available for MA (OLR-MA). On the other hand, P4 showed the lowest OLR-MA (58.3 %). The P-value for the analysis of the variance of the OLR-MA (see complementary material) in each period was lower than 0.05 (P-value = 0.0) again indicating a significant difference between the mean values in the different periods. In this case, Fischer's LSD method found significant similarity between mean values of P1 and P3, and between P4 and P5. P2 had the highest mean value for OLR available for MA due to the high treatment flow and influent COD concentration.

The system's response to organic-load peaks entailed an accumulation of VFA, likely due to the faster response of acid-generating than acid-consuming organisms, although the system was able quickly restore the equilibrium. The VFA effluent concentrations were low in the periods with longer HRT (P1 and P5) and moderate organic overloads. Also, higher operating temperatures were associated with lower VFA concentrations (P1, P2 and P4). In contrast, the combination of short HRT and lower temperature of P3 led to more severe organic overloading and hindered accumulated VFA consumption, resulting in the highest VFA effluent concentrations (see Table 1). This result is attributed to the more severe impairment of acid consuming organisms, which seemed to be more affected by lower temperatures than acid-producing organisms.

3.2. Methane production

Table 2 shows the percentages of the COD removed by filtration and biodegradation and biodegraded COD. As aforementioned, the COD removed was higher in periods P1 (93.1±6.3 %) and P5 (92.5±3.5 %), when HRT was higher (41±1 and 41±13 h, respectively), indicating that the extended contact time between soluble organics and anaerobic microorganisms enabled MA to further consume the organic matter and produce CH_4 and CO_2. The absence of intermediate products indicates that the intermediate anaerobic degradation steps were properly balanced, hydrolysis and solubilization being the limiting steps.

The percentage of biodegraded COD in P1 and P2 was higher than the rest of the periods (71.3±27.7 % and 74.4±14.5 %, respectively) due to their high influent COD concentration (1285±429 mg COD·L^{-1} in P1 and 1403±532 mg COD·L^{-1} in P2) and temperature (27±1 °C and 24±2 °C, respectively), which favored bacterial metabolism and hence organic matter consumption. P4, with 63.4±26.2 % of biodegraded COD, also presents a high operating temperature (27±1 °C) but, also the lowest influent COD concentration (755±224 mg COD·L^{-1}). The percentage of biodegraded COD in P4 is similar to those obtained in P3 (61.0±16.2 %) and P5 (61.4±13.0 %), operated at low temperatures (18±2 °C and 19 ±1 °C, respectively), but with higher influent COD (896±201 mg COD·L^{-1} in P3 and 1038±238 mg COD·L^{-1} in P5).

Fig. 4 depicts the time evolution of the 15-day moving average biogas production along with its 95 % confidence bands. As can be seen, the biogas production trend was directly related to the fraction of COD load available for MA (OL-MA), although it was also influenced by temperature, which limited the production of substrates for SRB and MA as a result of the reduction in the hydrolysis and solubilization rates.

The capacity of the system to recover energy was evaluated in terms of methane yield (Y^{CH_4}), which stands for the amount of methane produced per unit of influent COD. The biomethane potential (BMP) representing the fraction of the influent COD that ended up as methane (by comparing Y^{CH_4} to the theoretical maximal methane production, i.e., 350 STP L CH_4·kg^{-1} COD) was also calculated. The resulting Y^{CH_4} and BMP in the different periods are shown in Table 2. However, only the results regarding Y^{CH_4} will be discussed, since the BMP results followed the same trend.

The highest Y^{CH_4} value was obtained in P1 (169.0±95.1 STP L CH_4·kg^{-1} COD) with a combination of high T (27±1 °C) and HRT (41±1 h). Its influent COD/SO_4^{2-}-S ratio was also the most favorable for methane production (8.70±3.69 COD·g^{-1} SO_4^{2-}-S). While a high temperature enhanced the hydrolysis rate, which is considered the limiting stage in anaerobic digestion, methane production increased with the COD available for MA, for which P1 had the highest percentage of influent COD available for MA (77.0 %, see Fig. 3). In contrast, the slight temperature drop and reduced HRT reduction between periods P1 and P2 (24±2 °C and 25±1 h, respectively) helped to reduce the Y^{CH_4} to 109.6±70.0 STP L CH_4·kg^{-1} COD, even when Fischer's LSD method showed that the amount of COD for MA in P2 was the highest (35 kg·d^{-1}). It is important to highlight that an error of 18.5 % was obtained when COD mass balance was applied to P2 (see Fig. 5 and discussed in section 3.5). This COD loss could be attributed to underestimated biogas production. As mentioned above, dissolved methane in the effluent was

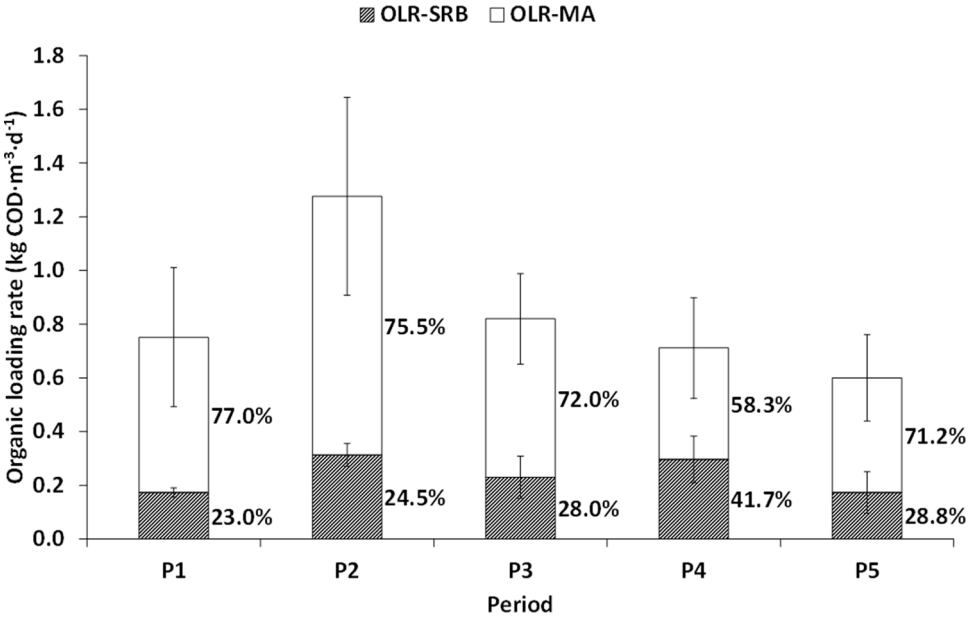

Fig. 3. Organic loading rate consumed by SRB and available for MA.

Table 2
Percentage of COD removed and biodegraded, methane yield (Y^{CH_4}), biomethane potential (BMP) per influent COD and waste sludge production (WSP) per kg of influent and removed COD.

Period	SRT (d) Mean ±SD	HRT (h) Mean ±SD	T (°C) Mean ±SD	COD_{rem} (%) Mean ±SD	$COD_{biodegraded}$ (%) Mean±SD	(STP L $CH_4 \cdot kg^{-1}$ COD) Mean±SD	BMP (%) Mean±SD	WSP (kg VSS·kg^{-1} COD_{inf}) Mean±SD	WSP (kg VSS·kg^{-1} COD_{rem}) Mean±SD
P1	70±1	41±1	27±1	93.1±6.3	71.3±27.7	169.0±95.1	48.3 ±28.3	0.112±0.045	0.121±0.067
P2	68±2	25±1	24±2	91.9±2.7	74.4±14.5	109.6±70.0	31.3 ±22.1	0.095±0.032	0.103±0.044
P3	70±2	26±2	19±1	86.7±4.7	61.0±16.2	75.2±16.9	21.5±4.8	0.144±0.050	0.166±0.074
P4	71±2	26±2	27±1	89.5±4.9	63.4±26.2	70.2±36.0	20.1 ±10.8	0.135±0.055	0.150±0.079
P5	70±0	41±13	18±2	92.5±3.5	61.4±13.0	102.3±63.8	29.2 ±18.2	0.137±0.061	0.148±0.072

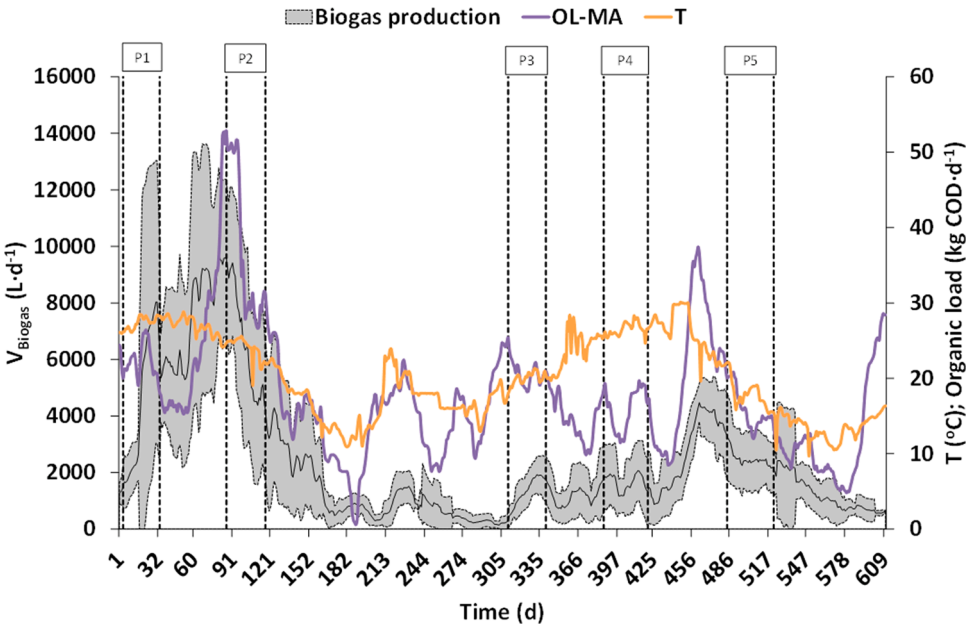

Fig. 4. Time evolution of the biogas production trend, temperature and organic load available for MA.

calculated assuming saturation conditions, although there could have been oversaturation episodes (Pauss et al., 1990).

P3 and P4 present the lowest Y^{CH_4} values (75.2±16.9 and 70.2±36.0 STP L $CH_4 \cdot kg^{-1}$ COD, respectively). These results can also be explained by the temperature, HRT and COD/SO_4^{2-}-S ratio. Despite P3 being operated with a moderate influent COD/SO_4^{2-}-S ratio (7.14±4.31 g COD·g^{-1} SO_4^{2-}-S) and available organic matter (72.0 %), influent COD could not offset its low operating temperature (19±1 °C) and HRT (26±2 h), which hindered hydrolysis and possibly also methane production. In P4, the operating temperature was favorable (27±1 °C) and HRT remained at 26±2 h, but the COD/SO_4^{2-}-S ratio was significantly lower than in the other periods (4.80±2.86 g COD·g^{-1} SO_4^{2-}-S), which justifies the lower Y^{CH_4}.

Finally, similar Y^{CH_4} values were obtained for P2 and P5 (109.6 ±70.0 and 102.3±63.8 STP L $CH_4 \cdot kg^{-1}$ COD, respectively). In this case, P2 had high temperature (24±2 °C) and high COD/SO_4^{2-}-S ratio (8.15 ±4.44 g COD·g^{-1} SO_4^{2-}-S) but lower HRT (25±1 h). P5 operated at high HRT (41±13 h), a moderate influent COD/SO_4^{2-}-S ratio (6.94±2.83 g COD·g^{-1} SO_4^{2-}-S) but the lowest temperature (18±2 °C). Considering that influent quality cannot be modified, HRT thus becomes the main design parameter for maximizing methane production in low winter temperatures.

It should be remembered that influent sulfate concentration depends on the region in which the wastewater is generated, since it is associated with the concentration of sulfate in water sources as well as the possible contribution of factories that discharge sulfur to the sewage system. Since the COD consumed by SRB is biodegradable and should have undergone at least the first stages of anaerobic degradation (i.e., hydrolysis and solubilization) to be available for SRB, it would also have been available for MA. A sulfate-free influent would thus have yielded a much higher Y^{CH_4} and BMP would have been similar to the percentage of biodegraded COD. As way of example, methane yields without considering the COD consumed by SRB in the studied periods, $Y_{SO_4}^{CH_4}$, would rise to 87.4-191.0 STP L $CH_4 \cdot kg^{-1}$ COD. However, in sulphate-rich wastewater, the role of SRB in reducing the effluent-soluble COD when HRT is reduced must be considered, since SRB has higher degradation rates than MA, especially at lower temperatures.

3.3. Biosolids

The waste-sludge production (WSP) expressed in kg VSS per kg of influent COD and per kg of removed COD is shown in Table 2. The reported values are the outcome of a complex interaction between operational conditions and influent characteristics. The fraction and

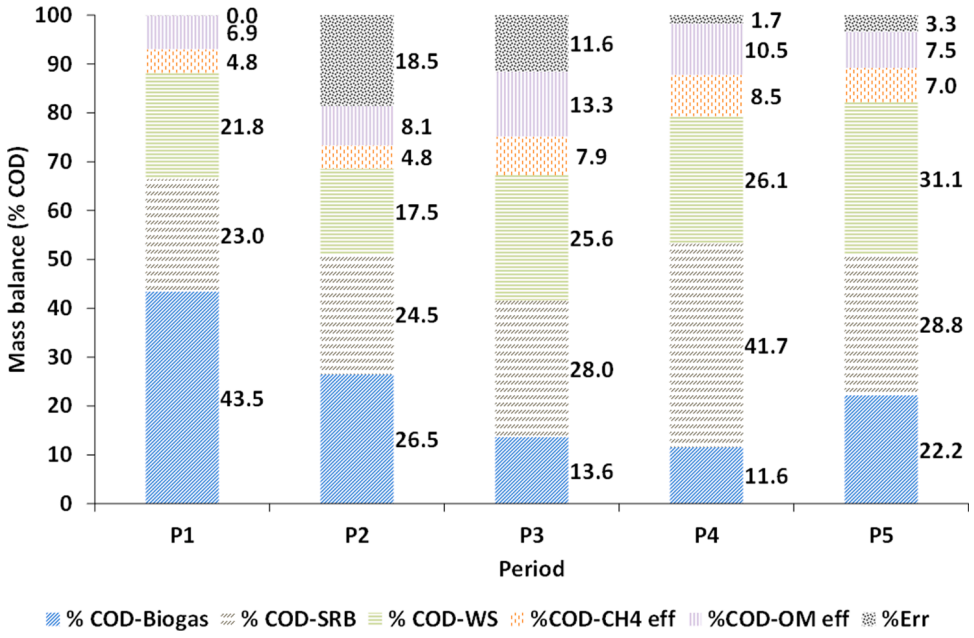

Fig. 5. COD mass balance.

composition of particulates in the influent and their fate within the system, which depends on the operating conditions (temperature, HRT and especially SRT), influence the accumulation and retention of biosolids and therefore increased WSP. A greater influx of solids into the system entails greater accumulation and retention unless the operating conditions favor hydrolysis and solubilization. On the other hand hydrolysis is widely accepted as the limiting step in anaerobic degradation and is severely affected by temperature drops (Batstone, 2006). Although temperature also affects the following steps of the anaerobic degradation, results indicated that intermediates accumulation can be mitigated as long as HRT is high enough. In the same way, WSP can also be reduced by increasing system SRT, thus extending the time for further hydrolysis and solubilization of the accumulated solids.

As can be seen in Table 2, P3 had the highest WSP (0.166±0.074 kg VSS·kg^{-1} COD$_{rem}$) mainly due to the low temperature (19±1 °C). The operating conditions hindered biodegradability and therefore raised sludge production. P4 and P5 also had high WSP (0.150±0.079 kg VSS·kg^{-1} COD$_{rem}$ and 0.148±0.072 kg VSS·kg^{-1} COD$_{rem}$, respectively) although lower than P1. The high production in P5 can be attributed to the lowest operating temperature (18±2 °C). However, despite temperature in P4 being the highest (27±1 °C) the significant WSP in this period was associated with its low COD/SO_4^{2-}-S (4.80±2.86 g COD·g^{-1} SO_4^{2-}-S). In this period, the percentage of influent COD consumed by SRB accounted for 41.7 % while in the other periods was below 29 %. Since the SRB biomass yield was significantly higher than MA, the associated solids production was also higher, as was sludge production in this period (Durán et al., 2020).

Finally, P1 and P2 had the lowest WSP (0.121±0.067 and 0.103 ±0.044 kg VSS·kg^{-1} COD$_{rem}$, respectively). Both were operated at high temperature (27±1 °C in P1 and 24±2 °C in P2) and had the highest COD/SO_4^{2-}-S ratio (8.70±3.69 g COD·g^{-1} SO_4^{2-}-S and 8.15±4.44 g COD·g^{-1} SO_4^{2-}-S, respectively). The combination of both variables enhanced MA organic matter biodegradation and reduced sludge production. P2 was also characterized by the highest percentage of influent VSS, COD and BOD_5, indicating the highest influent wastewater biodegradability and that the operating conditions were appropriate for the maximum biosolids degradation. It is important to highlight that, besides the complex interaction of factors that affect the WSP, it can be as low as 0.103±0.044 kg VSS·kg^{-1} COD$_{rem}$.

As a way of comparison, Foladori et al., (2015) reported 0.23-0.35 kg VSS·kg^{-1} COD$_{rem}$ (SRT of around 10-20 days) and 0.18 kg VSS·kg^{-1} COD$_{rem}$ for aerobic treatment without and with sludge stabilization, respectively. According to these values, all the selected periods had lower WSP than the aerobic treatments with percentage reductions between 8 % (P3) and 43 % (P2). Increasing SRT over 70 days could be expected to lead to lower WSP and additional biosolids stabilization, although solids concentration in the reactor would also rise and affect processes such as membrane filtration in the operating stage or increase reactor capacities' requirements in the design stage. In this regard, Ji et al., (2020) obtained a WSP of 0.09 kg VSS·kg^{-1} COD$_{rem}$ operating two different AnMBR with different pore size membranes for 120 days at 25°C. In this work no excess sludge was wasted during long-term operations except for the sludge samples. This value confirms that it is possible to achieve low WSP with high SRTs. On the other hand, with the same technology Seco et al., (2018) reported a WSP of 0.218 kg VSS·kg^{-1} COD$_{rem}$ with an SRT of 140 days and 27°C. Design and operating conditions thus need to be carefully selected taking influent characteristics into account and especially the particulate COD and COD/SO_4^{2-}-S ratio.

3.4. Nutrients recovery potential

Anaerobic treatments have low nutrients requirements per COD removed, as can be seen from the effluent characterization shown in Table 1, which confirms that AnMBR is promising technology for turning wastewater into a source of nutrients. N_T and P_T concentrations in the effluent accounted for 37.0-54.0 mgN·L^{-1} and 6.3-9.9 mgP·L^{-1}, respectively. Considering the treatment capacity of "Alcázar de San Juan" WWTP (16000 m^3·d^{-1}), these concentrations represent 216 – 315 t N·year^{-1} and 37 – 58 t P·year^{-1}. It should be noted that over 90 % of European phosphorus applications are import-dependent, so that wastewater can play an active role as a P supplier by combining AnMBR and CE fundamentals (Rosemarin et al., 2020). Selecting urea ($CO(NH_2)_2$) and P_2O_5 as representative N-based and P-based fertilizers, respectively, nutrients equivalent to 463-676 t Urea·year^{-1} and 84-132 t P_2O_5·year^{-1} could be provided by effluent. If a price of €273.6 per t of urea (bib31) and €783.0 per t of P_2O_5 (bib30) is assumed, the mineral-fertilizer equivalent-value of the nutrients in the discharge flow would be between k€162 and k€230 per year, and would pay for 10-15%

of water treatment costs at €0.269 per m^3 for AnMBR (Jiménez-Benítez et al., 2020a).

To reduce production costs and impact, an alternative embedded nutrient strategy consists of direct reuse (fertigation) instead of producing fertilizers from effluent, which makes AnMBR technology especially suitable for decentralized plants in rural areas where the effluent can be discharged directly to farmland. It is important to highlight that apart from a nutrient-rich stream the combination of an anaerobic process with UF membrane implemented in an AnMBR produces an effluent free of solids and pathogens (Krzeminski et al., 2017), which makes it especially appropriate for fertigation. For example, "Alcázar de San Juan" WWTP could cover an annual winter wheat crop (productivity 6.7 t grain·ha^{-1}) of 1080-1575 ha attending to N needs (0.2 t·ha^{-1}) and 1527-2400 ha if P_2O_5 requirements are considered (0.055 t·ha^{-1}) (Roy et al., 2006).

3.5. COD mass balance and overall performance considerations

Fig. 5 shows the COD mass balance in the periods studied. Each sector of the circles depicts the fraction of the influent COD (i.e., COD biodegraded by SRB, COD associated with biogas, COD from dissolved methane, COD wasted with sludge, COD in the effluent and mass balance error).

P1 shows no mass balance error but periods P2 to P5 have positive values in all cases indicating that the influent COD is higher than COD leaving the system through biogas, effluent, wasted sludge and COD biodegraded by SRB. The errors in P4 (1.7 %) and P5 (3.3 %) are negligible, while P2 and P3 show significant errors (18.5 % and 11.6 %, respectively). As all the parameters involved in COD mass balance (e.g., flow rates, COD concentration, biogas production, waste sludge, etc.) are based on analytical and/or on-line measurements except for dissolved methane (calculated as stated in Section 2.2), it is hypothesized that in these two periods dissolved methane in the effluent have been underestimated. As mentioned above, dissolved methane is calculated considering saturation conditions based on Henry's law, although oversaturation could take place in these periods due to methane bubbles passing through the membranes or poor reactor mixing.

Fig. 5 confirms the influence of operating at high T, HRT and COD/SO_4^{2-}-S ratio to enhance methane production, with P1, P2 and P5 showing the highest percentages of biogas associated to COD. P1, operated at the highest temperature, HRT and COD/SO_4^{2-}-S ratio (27±1 °C, 41±1 h and 8.70±3.69 g COD·g^{-1} SO_4^{2-}-S, respectively), shows a percentage of 43.5 % of biogas-associated COD, the highest value. P2 was operated at high temperature (24±2 °C) and a favorable COD/SO_4^{2-}-S ratio (8.15±4.44 g COD·g^{-1} S). Both conditions could offset the lower HRT (25±1 h) and kept biogas production at significant percentages (26.5 %). If the COD mass balance error (18.5 %) is attributed to underestimated methane production, the biogas percentage could rise to 45 %, as in P1. P5 also presented significant biogas production (22.2 %). This period operated at high HRT (41±1 h) and moderate influent COD/SO_4^{2-}-S ratio (6.94±2.83 g COD·g^{-1} SO_4^{2-}-S), but the lowest T (18 ±2 °C), which hindered methane production. Since a negligible error was obtained for this period, potential increased biogas production due to underestimated dissolved methane cannot be considered.

The lowest percentages of COD associated with biogas were obtained in P3 (13.6 %) and P4 (11.6 %). P3 operated at a low temperature (19±1 °C) and HRT (26±2 h) and therefore with low biogas production despite a favorable influent COD/SO_4^{2-}-S ratio (7.14±4.31 g COD·g^{-1} SO_4^{2-}-S), similar to P5. However, if mass balance error is considered (11.6 %), biogas-associated COD could rise to 25.2 %. P4, with low HRT (26±2 h) and the lowest influent COD/SO_4^{2-}-S ratio (4.80±2.86 g COD·g^{-1} SO_4^{2-}-S) had the lowest COD percentage associated with methane production (11.6 %), despite its favorable operating temperature (27±1 °C).

For the COD associated with waste sludge, Fig. 5 shows that P5 had the highest percentage (31.1 %). This period operated at the lowest T (18±2 °C), which hindered particulate degradation, and the lowest OLR (0.60±0.20 kg COD·m^{-3}·d^{-1}). Since OLR before P5 was higher than in this period (see Fig. 2), the sludge wasted during P5 could have been influenced by accumulated VSS. P2 had a percentage of 17.2 %, due to its favorable operating temperature (24±2 °C) and the highest OLR (1.28±0.38 kg COD·m^{-3}·d^{-1}). P3 and P4 show a similar percentage associated with waste sludge (25.6 % and 26.1 %, respectively), despite operating at very different temperatures (19±1 °C in P3 and 27±1 in P4 °C). This can be attributed to the higher COD percentage consumed by SRB (41.7 %) in P4. As these bacteria have a higher growth yield than MA and therefore increase solids production in the reactor.

The results show an overall promising performance of AnMBR technology for treating MWW at ambient temperature. The COD removal capacity allows compliance with treatment regulation (e.g., MWWTD); methane yields indicate its potential for organic matter valorization and energy recovery; and nutrient content in the effluent indicates its capacity for turning wastewater into a valuable source of nutrients. Low waste-sludge production means AnMBR can be considered a robust candidate as the new core technology to boost the transition of wastewater treatment plants towards the new resource recovery facilities, especially suitable for small and decentralized plants in which anaerobic processes are not currently applied.

Temperature, HRT, SRT and COD/SO_4^{2-}-S ratio are crucial parameters to improve technological, economic and environmental performance of AnMBR technology for wastewater treatment, HRT and SRT being the only ones which do not depend on ambient conditions. Both HRT and SRT thus appear as the main design parameter. It is important to highlight the benefits that the implementation and optimization of dissolved methane recovery technologies (e.g., hollow-fiber membranes) can provide to AnMBR: dissolved methane will improve the wastewater treatment energy balances while the carbon footprint associated with greenhouse gas emissions is minimized.

4. Conclusions

An industrial AnMBR prototype was operated for 610 days during which an average of 87.2 ± 6.1 % of influent COD removal was achieved, with 87 % of the data over 80 % and almost 40 % over 90 %. UF membranes contributed to wastewater treatment by retaining almost all the particulates. In addition, in the long-term operations, 5 periods were selected to evaluate the performance of the AnMBR. In these periods, a Y^{CH_4} between 70.2 and 169.0 STP L CH_4·kg^{-1} COD were achieved and methane percentages in the biogas varied from 75 to 77 %. Sludge production ranged between 0.103±0.044 and 0.166±0.074 kg VSS·kg^{-1} COD_{rem}, which represents a reduction of 9 to 43 % compared to CAS. A significant nutrient recovery potential was also identified.

Finally, temperature, HRT and COD/SO_4^{2-}-S ratio jointly with SRT are crucial parameters to take into account in maximizing AnMBR's technical, economic and environmental performance. However, since temperature and influent COD/SO_4^{2-}-S depend on ambient conditions, HRT could be considered the main design parameter jointly with SRT.

Declaration of Competing Interest

None.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2022.118249.

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A semi-industrial AnMBR plant for urban wastewater treatment at ambient temperature: Analysis of the filtration process, energy balance and quantification of GHG emissions

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ABSTRACT

A semi-industrial scale AnMBR urban wastewater treatment plant was operated for 580 days at ambient temperature (ranging from 10 to 30 °C) to assess its long-term filtration performance, energy balance and GHG emissions. The applied 20ºC-standardized transmembrane flux (J_{20}) was varied between 15 and 25 LMH and the specific gas demand per m^2 of membrane (SGD_m) was modified between 0.10 and 0.40 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to a specific gas demand per permeate volume (SGD_P) between 10 and 20 $Nm^3 \cdot m^{-3}$). The filtration strategy allowed successful long-term operations without any chemical cleaning requirements and little fouling for 233 days. The plant operated as a net energy producer for more than 50 % of the experimental period, with an average net energy demand of − 0.169 ± 0.341, − 0.190 ± 0.376 and − 0.205 ± 0.447 $kWh \cdot m^{-3}$, considering 0 %, 50 % and 70 % of dissolved methane recovery, respectively. Finally, demethanization of AnMBR effluent is needed to achieve an environmentally sustainable operation of the technology. Therefore, the combination of AnMBR with degassing membranes appears as a suitable alternative to conventional wastewater treatment.

1. Introduction

Although conventional wastewater treatment is usually based on aerobic processes (with conventional activated sludge (CAS) or an aerobic membrane bioreactor (MBR)), anaerobic membrane bioreactors (AnMBRs) are receiving increasing attention due to their advantages [1] including: i) organic matter valorization through methane production during anaerobic digestion; ii) a high-quality solid-free effluent production; iii) nutrients are conserved for recovery from the effluent; iv) operations at high sludge retention time (SRT), since membranes decouple hydraulic retention time (HRT) and SRT; v) they can treat low-loaded streams at ambient temperature since the membranes guarantee solids retention and avoid biomass washout; vi) improved stabilization of biosolids and significant reduction in sludge production when long SRTs are applied; and vii) they make reduced layouts possible thanks to the settlers being substituted for compact membrane units.

AnMBR technology has been considered for the treatment of different industrial wastewater streams, such as textile, petrochemical, brewing or pharmaceutical industry effluents with satisfactory results [13,5]. However, despite the potential benefits of AnMBR plants for treating urban wastewater (UWW), they still face bottlenecks that need to be overcome, including: i) membrane fouling; ii) eutrophication potential of the effluent when it is discharged in sensitive zones; iii) dissolved methane in the effluent [39]; and iv) competition between sulfate-reducing bacteria (SRB) and methanogens, which can affect methane yield and generate corrosive sulfide [7].

Abbreviations: AnMBR, anaerobic membrane bioreactor; BOD_5, biological oxygen demand; CAS, conventional activated sludge; COD, chemical oxygen demand; GHG, greenhouse gas; HRT, hydraulic retention time; J_{20}, 20ºC-standardized transmembrane flux; N, nitrogen; MBR, membrane bioreactor; P, phosphorus; SGD_m, sparging gas demand per m^2 of membrane; SGD_P, sparging gas demand per m^3 of permeate; SRB, sulfate-reducing bacteria; SRT, sludge retention time; TSS, total suspended solids; UWW, urban wastewater; VSS, volatile suspended solids.

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Membrane fouling has been extensively identified as a key issue to be addressed before AnMBRs can be widely used for UWW [19]. This phenomenon is governed by different interrelated factors. For instance, operating at high temperatures reduces the viscosity of the mixed liquor, which favors filtration but also involves the release of extracellular polymeric substances (EPS), which are related to fouling [21]. Although anti-fouling strategies based on gas sparging have been reported as effective, they increase the filtration energy demand and introduce shear forces. Increased SRT can also reduce membrane fouling since it consumes EPS and SMP. However, high SRT increases the solids concentration and hinders the progress of filtration.

The loss of part of the methane production as gas dissolved in the effluent must also be addressed to reduce fugitive emissions, identified as one of the greatest contributions to GHG of this technology [38], and to improve the energy performance of the facilities by recovering part of the methane produced [41]. Among other technologies, degasification membranes [35] and spray aeration towers are thus being studied for this aim.

AnMBR plants have been reported to have a lower energy consumption than CAS and MBR; Lazarova et al. [20] reported energy demands of 0.25–0.60 $kWh{\cdot}m^{-3}$ for CAS and 0.50–2.50 $kWh{\cdot}m^{-3}$ for MBR. Pretel et al. [25] reported AnMBR net energy production of 0.17–0.19 $kWh{\cdot}m^{-3}$ without considering dissolved methane recovery, while Jiménez-Benítez et al. [16] reported net energy production of up to 0.47 $kWh{\cdot}m^{-3}$ when treating high-loaded UWW at mild temperatures when dissolved methane was considered and sulfate content in the influent was 112–172 mg $S\text{-}SO_4{\cdot}L^{-1}$. The recovery of organic matter through biogas production makes it possible to partially offset the indirect GHG emissions derived from its energy demand. Also, anaerobic processes do not emit nitrous oxide (N_2O), as do aerobic nitrification/denitrification processes [32]. This is an important feature since the global warming potential of this gas has been estimated at 298 $kgCO_{2eq}$ per kg of N_2O [34].

For all the above reasons, AnMBRs appear as a promising technology for the so-called *water resource recovery facility* concept [10], as opposed to the conventional urban wastewater treatment approach, which considers wastewater as a source of pollution.

This paper evaluates the performance of a semi-industrial AnMBR plant in terms of filtration process performance, energy balance and GHG emissions. The plant was fed with effluent from the pre-treatment of the "Alcázar de San Juan" wastewater treatment plant (Ciudad Real, Spain). The major novelties of this study include i) long-term operation of 580 days; and ii) technical characteristics and operation (real UWW, reactor volume of volume 40 m^3, commercial hollow-fiber membranes, industrial equipment, ambient conditions, etc.). These characteristics allowed obtaining robust data for scaling-up AnMBR technology to full-scale facilities, thus providing of useful information in terms of membrane performance, energy balance, and GHG emissions for the design and full-scale operation of AnMBR systems. The goal of the research was to determine the potential of AnMBR over conventional treatments by assessing its robustness against variations in the main operating conditions (temperature, organic load, treatment flow, etc).

2. Materials and methods

2.1. AnMBR description

The semi-industrial AnMBR plant implemented in the study consisted mainly of the following components: an anaerobic reactor (40-m^3 total volume; 34.4-m^3 working volume); three membrane tanks (MT_A, MT_B and MT_C; 0.8-m^3 total volume each; 0.7-m^3 working volume each) equipped with an ultrafiltration membrane system (PURON® PSH41, KMS, 0.03-μm pore size, 41-m^2 filtration area per tank); a sieve screw (1.5-mm screen size); an equalization tank (1.1 m^3); and a clean-in-place (CIP) tank (0.37 m^3). By including three identical MTs in the system, the effect of applying different J_{20} and gas-assisted membrane scouring intensities on membrane fouling can be tested in parallel treating the same feed at the same ambient conditions. Further details of this system can be found elsewhere in [31] and in supplementary materials. The plant was also equipped with a degassing membrane unit (2.1-m^2 filtration area) for dissolved methane recovery (PermSelect®, MedArray Inc. USA), using vacuum pressure as the driving force (for further details see Sanchis-Perucho et al. [36]).

The AnMBR was fed with effluent from the pre-treatment of the "Alcázar de San Juan" wastewater treatment plant (Ciudad Real, Spain), consisting of screening, grit and grease removal. The average influent quality is shown in Table 1. Since the dairy and wine industries are important elements in the local economy, the influent wastewater was characterized by high concentrations of COD (1071 ± 467 $mg{\cdot}L^{-1}$), BOD_5 (710 ± 274 $mg{\cdot}L^{-1}$) and sulfate (140.3 ± 42.5 mg $SO_4\text{-}S{\cdot}L^{-1}$).

2.2. AnMBR operation

The evolution of HRT, temperature, and organic load available for methanogenesis (OL-MA) during the 580-days experimental operation can be found elsewhere in Robles et al. [29] and in supplementary materials. Temperature varied freely in the 10–30 °C range according to ambient conditions and season and SRT was set to 70 d. Several periods with different average HRT were operated as follows: 40 h (days 1–32); 25 h (days 33–122); 46 h (days 123–170); 38 h (days 171–291); 26 h (day 292–500) and 70 h (days 501–580). The different HRT simulated population changes during the summer and winter periods. Eventually, HRT was stablished at 70 h in a final experimental stage in order to assess the treatment process performance at high HRT.

The number of membrane tanks in operation depended on the selected HRT, while OL-MA was affected by economic activity, mainly dairy and wine, the latter with a markedly seasonal contribution. The influence of the grape harvest and winemaking can be noted between August and October between days 60 (August) and 100 (October) and between days 430 (September) and 480 (October).

Membrane filtration and scouring took place within the following parameter range: the gross 20ºC-standardized transmembrane flux (J_{20}) was modified between 15 and 25 LMH and the specific gas demand per m^2 of membrane (SGD_m) was between 0.10 and 0.40 $Nm^3{\cdot}m^{-2}{\cdot}h^{-1}$, resulting in a specific gas demand per permeate volume (SGD_P) of between 10 and 20 $Nm^3{\cdot}m^{-3}$, (for further details of membrane operation see Robles et al. [30]).

Membranes were operated according to a specific schedule involving a combination of different individual stages taken from a basic 300 s-filtration: 30 s-relaxation cycle. Besides classical membrane operating stages (filtration, relaxation, and back-flush), degasification and ventilation stages were also considered in the membrane operation. Degasification aims to recover the accumulated biogas in top of the dead-end, hollow-fiber membranes, since this accumulation of biogas reduces the effective filtration area. The degasification stage consists of a period of high flow-rate filtration that is carried out to enhance the filtration process efficiency by removing the accumulated biogas. On the other hand, during ventilation, permeate is pumped from the CIP tank to the membrane tank through the degasification vessel instead of through the membrane. The aim of the ventilation stage is to recover the accumulated biogas in the degasification vessel due to the extraction of gas

Table 1
Average influent wastewater characteristics.

Parameter	*Unit*	*Mean ± SD*
TSS	$mg\ TSS{\cdot}L^{-1}$	405 ± 215
Total COD	$mg\ COD{\cdot}L^{-1}$	1071 ± 467
BOD_5	$mg\ COD{\cdot}L^{-1}$	710 ± 274
Total Nitrogen	$mg\ N{\cdot}L^{-1}$	48.2 ± 11.7
Total Phosphorus	$mg\ P{\cdot}L^{-1}$	8.4 ± 2.4
Sulfate	$mg\ SO_4\text{-}S{\cdot}L^{-1}$	140.3 ± 42.5

bubbles during filtration.

The chemical membrane cleaning included a first washing phase with sodium hypochlorite (2000 ppm) for 8 h followed by a second washing phase with citric acid (2000 ppm) for another 8 h.

The size of the AnMBR plant and the type and quality of the wastewater (real urban wastewater) involved in this study can contribute to determine the conditions for the technical, economic and environmental feasibility of these technologies as an alternative to conventional wastewater treatment.

2.3. Streams characterization and analytical methods

The following parameters were regularly monitored in the influent: total chemical oxygen demand (COD), biological oxygen demand (BOD_5), sulfate, sulfide, total suspended solids (TSS), and volatile suspended solids (VSS). Effluent quality was mainly monitored through COD, sulfide and volatile fatty acids content. Reactor monitoring was based on TSS, VSS, COD, pH and methane content in the generated biogas (% CH_4). TSS, VSS, COD, BOD_5, sulfate and sulfide were determined according to standard methods [2]. VFA concentration was determined by titration according to the method proposed by [27]). Dissolved methane in the effluent was determined according to Giménez et al. [9].

2.4. Data processing and calculations

The instrumentation implemented in the prototype acquired the following data, among others: reactor influent flow, reactor temperature, transmembrane pressures and fluxes, biogas flows for reactor mixing and membrane scouring, mixed liquor feeding flow to the membrane tanks, and biogas production and quality. Different key performance indicators were calculated to assess the performance of the system, i.e. J_{20}, specific energy consumption (EC), total energy recovery (TER), net energy demand (NED) and greenhouse gas (GHG) emissions.

It is important to highlight that a fraction of the influent organic matter is consumed by SRB, which meant it was not available for biogas production by methanogenic organisms. Specifically, 2 g of influent COD were consumed by SRB for each gram of sulfate (stoichiometrically).

Over-dimensioned equipment in the prototype plant prevented the direct measurement of electrical power consumption from being representative of the energy requirements of AnMBR technology. The power required by the equipment (pumps and blowers) was therefore calculated theoretically using the equations proposed by Judd and Judd [17] and Pretel et al. [26]. Adiabatic indexes, and blower and pump efficiencies were estimated based on standard values for these components.

Two sources of GHG emissions were considered: i) indirect CO_2 emissions related to the NED of the system ($CO_{2_{NED}}$) and ii) direct equivalent CO_2 emissions from dissolved methane in the effluent (CO_{2D-CH_4}). Both sources are considered as the main GHG contributors during the operation of the demo plant. It should be noted that, according to the IPCC2013 application guides, since they are included in a short-term carbon cycle, CO_2 emissions of biogenic origin produced during anaerobic digestion can be excluded from GHG calculations, as well as those associated with the subsequent combustion of biogas in a CHP unit. Three different values were calculated for CO_{2D-CH_4} and therefore for total CO_2 emissions: i) without dissolved methane recovery; ii) with 50 % of dissolved methane recovery and iii) with 70 % of dissolved methane recovery. Percentages of 50 % and 70 % were assumed based on the optimum economic and environmental results from previous studies, respectively [35].

Details on data calculations are provided as supplementary material.

3. Results and discussion

3.1. Filtration process performance

Fig. 1 gives the performance of the filtration process during the complete experimental period for the MT_A, MT_B and MT_C membrane tanks (average values can be seen in table S1 of the supplementary material). The black dashed bars indicate short maintenance stops (days 94 and 442) and gray dotted-dashed bars indicate membrane chemical cleanings (days 128 and 363). A conservative maximum TMP security value of 500 mbar was selected based on the supplier's information. Finally, the TSS concentration depicted in Fig. 1 corresponds to the reactor mixed liquor (i.e. MLSS), thus representing the TSS

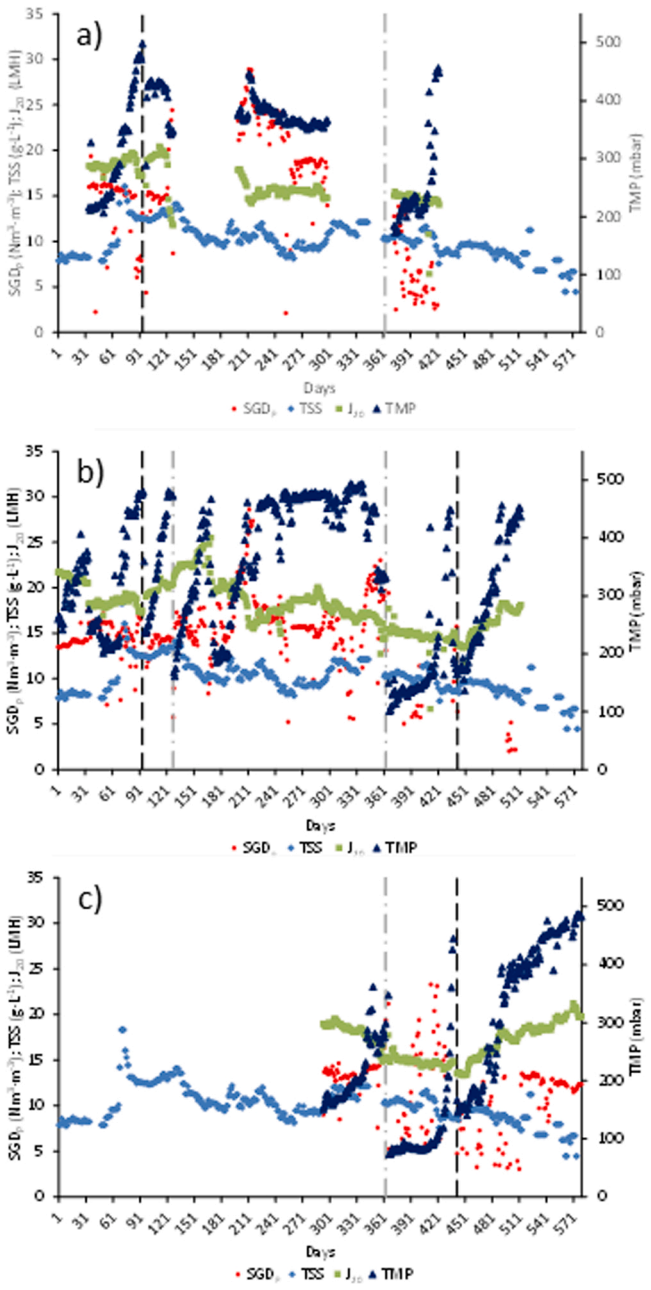

Fig. 1. Evolution of sparging gas demand per m^3 of permeate (SGD_P), gross 20°C-standardized transmembrane flux (J_{20}), transmembrane pressure during filtration stage (TMP) and total suspended solids (TSS) in a) MT_A; b) MT_B and c) MT_C. (Maintenance stop: black dashed line; Membrane chemical cleanings: gray dotted-dashed lines).

concentration of the mixed liquor entering the three MTs.

As can be seen in Fig. 1, J_{20} was maintained in similar values for all MTs: 14–20, 15–23 and 15–20 LMH for MT_A, MT_B, and MT_C, respectively. These J_{20} are somewhat higher than previous long-term AnMBR studies [15,24]. The J_{20} applied were also competitive against MBR for urban wastewater treatment. For instance, Xiao et al. [42] evaluated 19 large-scale Chinese MBRs and reported fluxes in the range of 15–25 LMH (19.6 LMH on average).

Regarding SGD_m, MT_A was operated between 0.1 and 0.4 $Nm^3 \cdot m^{-2} \cdot h^{-1}$; MT_B mostly at 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$, although higher (0.35–0.40 $Nm^3 \cdot m^{-2} \cdot h^{-1}$; days 130–200) and lower (0.1–0.2 $Nm^3 \cdot m^{-2} \cdot h^{-1}$; days 360–500) values were also evaluated; and MT_C at 0.25 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ at days 294–360 and 510–580 and in the range of 0.1–0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ between days 360 and 510. The modifications in the applied SGD_m aimed to assess gas sparging intensity for fouling control.

In this operation, SGD_P varied between 7 and 23 $Nm^3 \cdot m^{-3}$ for MT_A and between 14 and 16 $Nm^3 \cdot m^{-3}$ for MT_B, with short periods above 20 $Nm^3 \cdot m^{-3}$ (e.g. days 200–216) and below 10 $Nm^3 \cdot m^{-3}$ (days 383–401). Fig. 1c gives the first period of MT_C, with SGD_P at 13–14 $Nm^3 \cdot m^{-3}$ (days 294–360), a second highly variable period between 5 and 22 $Nm^3 \cdot m^{-3}$ and a third period with SGD_P steady at around 12–13 $Nm^3 \cdot m^{-3}$.

Fig. 1 allows the comparison of the filtration performance of the three MTs, attending to the differences in their operation. Between days 31 and 91, MT_A and MT_B (see Fig. 1a and b) operated at similar J_{20} (16–20 LMH) and SGD_P (15–16 $Nm^3 \cdot m^{-3}$) and both tanks started from a close TMP, in the 210–220 mbar range. It can be observed that the behavior of both MTs was very similar, reaching in both cases the limit value of ≈ 500 mbar at the same time (day 91). It can also be observed a sharp increase in TSS on day 45, reaching a concentration of 15 $g \cdot L^{-1}$ around day 75, which coincides with the sharp TMP increase recorded in both tanks. Therefore, it can be concluded that the membranes were operated at supra-critical filtration conditions due to the combination of the J_{20}, SGD_P and TSS values within the experimental period.

After the maintenance stop conducted between days 91 and 94, both MT_A and MT_B partly reduced their TMP without applying any membrane chemical cleaning, thus the observed recovery was associated with reversible fouling that was removed as a result of prolonged air-assisted membrane scouring without filtration.

After restarting filtration, the behavior of both tanks differed. In the case of MT_A, there was an abrupt rise in TMP after only three days of operation, reaching a stable TMP around 420 mbar. From day 121, the J_{20} was reduced from 20 LMH to 12 LMH, and this caused a drop in the TMP from 420 to 330 mbar, which represents a reduction of 21 %. This TMP profile would indicate that reducing J_{20} turned the filtration process from supra-critical/critical (since TMP first raised and then could be maintained at 420 mbar) to sub-critical conditions. In MT_B, the filtration process caused a constant increase in TMP until reaching the maximum threshold value (i.e. 500 mbar) set for conducting a membrane chemical cleaning. On day 128, the membrane in MT_B was chemically cleaned, which reduced TMP to 161 mbar. A reduction of 43 % in membrane total resistance ($R_T^{\}$) when comparing the R_T achieved after the first maintenance stop on days 91–94 (i.e. 14.1 $LMH \cdot mbar^{-1}$) and the chemical cleaning on day 128 (i.e. 8.1 $LMH \cdot mbar^{-1}$) would indicate the amount of irreversible fouling accumulated on the MT_B membrane when the maintenance stop was conducted (between days 91 and 94). Eventually, filtration performance of MT_B between days 128 and 221 confirms that the combination of J_{20} above 20 LMH, SGD_P below 17 $Nm^3 \cdot m^{-3}$ and TSS above 10 $g \cdot L^{-1}$ led to significant fouling propensities, increasing TMP from 150 to 190–470 in 35–40 days.

Different profiles between MT_A and MT_B regarding J_{20}, SGD_P and TMP can be observed from day 200 and 295. Specifically, the reduction in J_{20} from 17.5 to 15–16 LMH applied to the MT_A managed to reduce the TMP from the maximum of 450 mbar to 360 mbar (day 245). Moreover, the reduction recorded in the TSS concentration from 11 to 9 $g \cdot L^{-1}$ also favored reducing the SGD_m from 0.37 to 0.29 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to a SGD_P of 23 and 19 $Nm^3 \cdot m^{-3}$, respectively), resulting in a slight decrease in TMP, thus confirming that filtration was carried out under sub-critical conditions with this combination of J_{20}, SGD_m and TSS concentration. In this same period, MT_B was operated with a rising J_{20} from 15.5 LMH to 21 LMH and a SGD_m of 0.29 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (SGD_P of 19 $Nm^3 \cdot m^{-3}$) in average. These operating conditions resulted in a sharp TMP increase from 320 mbar to 460 mbar (day 225). Despite this increase, the reduction in TSS concentration to 10 $g \cdot L^{-1}$ allowed to offset the effect of increasing J_{20} on TMP thus managing to remain stable TMP values of around 470 mbar, which indicated that sub-critical filtration conditions were kept. It is also worth to highlight the drop in TMP that occurred between days 290 and 300 due to a decrease in J_{20} from 21 LMH to 16 LMH and the subsequent TMP increase observed when the TSS concentration increased from day 300. Thus, slight changes on J_{20} and/or SGD_m quickly allowed compensating the effect of TSS concentration on TMP, highlighting the potential of gas-assisted membrane scouring as physical fouling control strategy to keep sub-critical/critical filtration conditions thereby controlling membrane fouling propensity.

Between days 300 and 363, when a second membrane chemical cleaning was conducted, MT_B and MT_C were operated in parallel. Fig. 1b shows how MT_B experimented a TMP reduction due to increasing the SGD_m up to 0.35 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to a SGD_P of 20 $Nm^3 \cdot m^{-3}$) between days 344 and 361. MT_C was operated at similar J_{20} values than the ones established in MT_B, although a constant SGD_m of 0.24 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (SGD_P of 13 $Nm^3 \cdot m^{-3}$) was established in MT_C. Thus, it can be concluded that, while it was possible to control TMP in MT_B by increasing gas sparging intensity, TMP in MT_C continuously increased between days 300 and 363. Therefore, MT_B operated at sub-critical filtration conditions (J_{20} of 15–18 LMH, SGD_P of 15–20 $Nm^3 \cdot m^{-3}$ and TSS of 12 $mg \cdot L^{-1}$) and MT_C at supra-critical filtration conditions (J_{20} of 15–18 LMH, SGD_P of 13–14 $Nm^3 \cdot m^{-3}$ and TSS of 12 $mg \cdot L^{-1}$).

On day 363, a chemical cleaning was conducted in the three MTs. In the case of MT_A, TMP turned out to similar values to the original ones (day 30). Thus, fouling was properly removed through chemical cleaning, revealing negligible levels or irremovable fouling on the membrane within the experimental period. In the case of MT_B, the R_T values achieved after this second chemical cleaning (i.e. 6.7 $LMH \cdot mbar^{-1}$) suggested that the first one (day 128) was not able to completely recovering irreversible fouling, i.e. around 1.4 $LMH \cdot mbar^{-1}$ of R_T was additionally removed compared to the results from the first chemical cleaning applied. In the case of MT_C, R_T was reduced to around 4.9 $LMH \cdot mbar^{-1}$, lower than the initial TMP on day 294 (i.e. 7.9 $LMH \cdot mbar^{-1}$), which would indicate the presence of previous fouling, since this membrane was kept submerged in the mixed liquor.

During days 361–445, the three MTs were operated in parallel. J_{20} between 14 and 15 LMH and low SGD_m (mostly 5–6 $Nm^3 \cdot m^{-2} \cdot h^{-1}$) were established for MT_A and TM_B. MT_C was operated at J_{20} of 15 LMH and higher but fluctuating SGD_P (5–21 $Nm^3 \cdot m^{-3}$). Around day 400, a sharp increase in TMP up to 460–480 mbar was observed in MT_A and MT_B driven by the low agitation, while MT_C could sustain filtration at low TMP for a few days longer (until day 420) since it was operated with higher SGD_P.

Finally, a maintenance stop was conducted on day 443. After this stop, R_T did not reach the value obtained after the previous chemical cleaning, thus some irreversible fouling (around 5 $LMH \cdot mbar^{-1}$ equivalent) may be developed from day 363–443 During this period, MT_B and MT_C were operated at similar J_{20} and SGD_m levels. Indeed, both tanks resulted in similar TMP behaviors. From day 505, only the MT_C was operated and its SGD_m was increased to 0.24 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to an SGD_P of 12.5 $Nm^3 \cdot m^{-3}$). This increase resulted in a significant reduction in TMP growth rate and therefore in fouling rate (i.e. from 4.1 $mbar \cdot d^{-1}$ to 1.4 $mbar \cdot d^{-1}$).

Therefore, Fig. 1 shows the complex interaction between J_{20}, SGD_m (as shown by SGD_P), TSS in the reactor, TMP and the evolution of fouling. Operation at high J_{20} with low fouling propensities can be

achieved with high SGD_m and low mixed liquor TSS concentrations. These results are in line with those obtained elsewhere by Mei et al. [23].

The results show that the system was able to conduct filtration at a moderate-high J_{20} (15–20 LMH) with competitive SGD_m (at an SGD_P of 10–20 $Nm^3 \cdot m^{-3}$) and moderate TSS (8–13 $g \cdot L^{-1}$), although shifts between supra-critical and subcritical filtration were seen to occur with small changes in the operating conditions. Operating at sub-critical filtration conditions (recommended to avoid quick TMP increase) was possible with TSS concentrations below 10 $g \cdot L^{-1}$, maximum J_{20} of 16 LMH and SGD_m of 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (corresponding to 15 $Nm^3 \cdot m^{-3}$). Fig. 1 also shows that fouling control can be achieved even at high TMP (e.g. between days 210 and 300 in MT_A (380–400 mbar) or between days 210 and 335 in MT_B (470–500 mbar). During these periods, the TMP presented a plateau profile, which indicated negligible fouling propensity. In this sense, the rapid response of TMP to changes in J_{20} and/or SGDm contributes to facilitate fouling control. However, operating at high TMP would imply increasing energy requirement for permeate pumping and increasing dissolved methane concentration. Therefore, to avoid these drawbacks operation at high TMP is not advisable.

As filtration performance comparison, [18] operated a pilot-scale AnMBR to treat municipal wastewater at ambient temperature (25 °C) and reported significantly lower transmembrane fluxes of approx. 3 and 4.5 LMH for HRT of 48 and 24 h, respectively, despite experiments at HRT of 24 h were conducted with lower mixed liquor suspended solids, in the 6.7–10.2 $g \cdot L^{-1}$ range (mixed liquor suspended solids at HRT of 48 h were not included in the study). Shin & Bae [37] described the performance of different pilot scale AnMBR treating domestic wastewater. These authors found that the most common transmembrane fluxes were in the 6–14 LMH range, while applied SGD_m varied mostly between 0.15 and 0.31 $Nm^3 \cdot m^{-2} \cdot h^{-1}$. Lee & Liao, [11] considered 10 LMH as typical design parameter for transmembrane flux.

Finally, it should be noted that the demo plant achieved a successful long-term operation without chemical cleaning requirements, the longest operation period being 233 days of MT_B (days 128–363), although the results indicate that longer operating periods could be achieved. No irrecoverable fouling was detected during the study since chemical cleaning returned the TMP to the initial values or even lower.

3.2. Energy assessment

Evolution of biogas production, its methane content expressed in percentage of CH_4, and removed COD (% COD_{rem}) can be found in supplementary materials. Moreover, details on the evolution of biogas production process in this demo plant during this operational period can be found in Robles et al. [29], where it could be concluded that temperature, HRT, SRT and influent $COD \cdot SO_2^{-4}$-S ratio showed crucial influence in biogas production.

As already mentioned in Section 2.2, organic load and temperature varied according to economic activities and ambient conditions, respectively. Methane production is therefore highly influenced by these factors. Periods with high temperature and high OL-MA showed higher biogas productions. Conversely, low OL-MA and low temperature led to reduced biogas production. With regards to methane content in the biogas, it remained mostly above 70 % (75 ± 3 % on average). Finally, AnMBR reached a significant COD removal efficiency (89 ± 5 % on average) (further details can be found in Fig. S3 of supplementary materials).

Fig. 2 shows the evolution of EC, TER and NED. The gaps in the figures are due to maintenance stops and failures in the online equipment required for calculations.

Fig. 2a shows an initial period between days 1–30 with high energy consumption for reactor mixing (around 0.11 $kWh \cdot m^{-1}$). On day 31, mixing was reduced to 0.04–0.08 $kWh \cdot m^{-3}$ until day 500, without affecting reactor performance. EC related to membrane scouring also varied according to the applied SGD_m and number of membrane tanks in

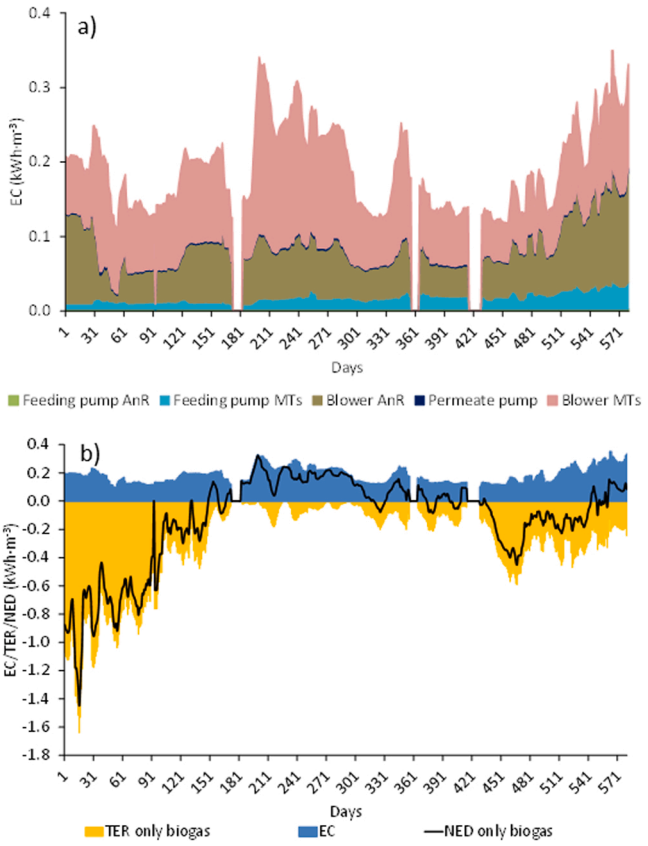

Fig. 2. Results of (a) allocation of energy consumption (EC) per equipment and (b) total energy consumption (EC), total energy recovery (TER) and net energy demand (NED) considering only the energy recovered from biogas (only biogas).

operation. Between days 1–30, only MT_B was in operation at an SGD_m of 0.3 $Nm^3 \cdot m^{-2} \cdot h^{-1}$. On day 35, MT_A started operating jointly with MT_B, leading to an increased need for sparging and higher energy consumption (1.2–1.3 $kWh \cdot m^{-3}$). MT_A and MT_B were also operated in parallel between days 194–299, when energy consumption reached the highest value (1.5–2.0 $kWh \cdot m^{-3}$), which was associated with higher applied SGD_m in MT_A (0.36–0.43 $Nm^3 \cdot m^2 \cdot h^1$). From day 300, MT_A was stopped, SGD_m in MT_B was reduced to 0.20 $Nm^3 \cdot m^2 \cdot h^1$ and MT_C started with an applied SGD_m of 0.24 $Nm^3 \cdot m^2 \cdot h^1$. Energy consumption for membrane scouring was thus reduced to 0.5–0.6 $kWh \cdot m^3$. On days 330–355, SGD_m in MT_B was set to 0.35 $Nm^3 \cdot m^2 \cdot h^1$, which led to higher energy consumption for membrane scouring. The period between days 365–500 was characterized by lower applied SGD_m (and thus SGD_P) than previously, which allowed moderate-low energy consumption despite MT_B and MT_C being operated in parallel, and MT_A was jointly operated between days 373–420. The EC associated with membrane tank feeding mostly remained below 0.02 $kWh \cdot m^{-3}$ while the energy requirement for the reactor feeding pump was negligible. After day 500, HRT was set to 70 h, so that the treatment flow was reduced, leading to higher specific energy consumption in $kWh \cdot m^{-3}$.

Fig. 2b gives the total EC, TER and NED. The TER in Fig. 2b was based on energy recovered from the biogas stream only (0 % of dissolved methane recovery). This recovered energy allowed the plant to perform as a net energy producer for over 50 % of the study period (negative values in NED). As can be seen in Fig. 2b, methane production and therefore energy recovery was boosted by a high OL-MA and temperature (see Fig. S2 in supplementary materials). In this regard, TER followed the same trend as the temperature from day 1–160, with a high energy production at 28 °C (days 1–60), which dropped steadily as the temperature dropped to 14 °C (day 160). Days 440–580 also showed

high TER values, when energy production increased when the temperature remained above 26 °C (days 400–450) and by the increase in OL-MA (days 450–490). On the other hand, days 160–440 resulted in reduced TER since this period was characterized by a relatively low temperature and OL-MA.

Table 2 gives the average results for the entire study period and shows that the main contributions to EC were associated with the blower requirements for membrane scouring (55.2 %), followed by reactor mixing (34.4 %). Conversely, membrane tanks' feeding pump, filtration pumps, and reactor feeding pump had comparatively very low energy consumption (7.8 %, 1.6 % and 1.0 %, respectively). The contribution of the degassing membrane blowers was negligible (<0.02 %). Overall, average EC accounts for 0.192 ± 0.070 kWh·m^{-3} while average TER (0 % methane recovery) was 0.361 ± 0.271 kWh·m^{-3}. Dissolved methane produced more energy than that used in its recovery, although its contribution to TER was relatively low (between 0.02 kWh·m^{-3} and 0.04 kWh·m^{-3} for dissolved methane recovery ratios of 50% and 70 %, respectively). However, this recovery definitively reduced GHG emissions, as will be explained in the following section.

These results can be compared with other studies dealing with AnMBR for wastewater treatment. Rong et al. [4] evaluated the energy balance of an AnMBR at pilot scale and a projected scaled-up facility of 10000 m^3·d^{-1}. Influent COD was 331–414 mg·L^{-1} and temperature ranged between 15 and 25 °C. These authors obtained a net electricity demand in the 0.09–0.10 kWh·m^{-3} range, with gas sparging accounting for 52.46–74.60 % and reactor mixing for 4.08–6.13 %. More specifically, Hu et al. [14] reported energy consumptions for gas sparging between 0.25 and 3.4 kWh·m^{-3}, accounting for approx. 70 % of total energy consumption, which are between 2 and 32 times the average energy consumption obtained in this study (0.106 ± 0.047 kWh·m^{-3}). Concerning degassing membranes, lower energy consumptions were achieved (between $2.3{\cdot}10^{-5}$ ± $3.4{\cdot}10^{-6}$ and $4.3{\cdot}10^{-5}$ ± $6.3{\cdot}10^{-6}$ kWh·m^{-3}, see Table 2) compared to the one reported by Velasco et al. [40], i.e. 0.01 kWh·m^{-3} for 90 % of dissolve methane recovery.

Moreover, Krzeminski et al. [19] reported 0.3–0.6 kWh·m^{-3} for CAS and 0.6–2.3 kWh·m^{-3} for MBR, including nutrient removal. In this sense, Fraia et al. [8] reported energy consumption for nitrogen removal up to 6.08 kWh per kg of influent total nitrogen (kWh·TN$_{in}$) in wastewater treatment facilities with high nitrogen removal efficiency (> 89.2 %), while energy consumption associated to phosphorus removal was considered negligible. Hence, assuming the nitrogen load of the AnMBR influent (see Table 1) the energy demand for nitrogen removal in an aerobic/anoxic process would be of approx. 0.29 kWh·m^{-3}, according to this literature value. Hence, extracting this value to the above-mentioned ranges for CAS and MBR energy demand, the energy consumption related to COD removal in CAS and MBR technology would be between 0.01 and 0.31 and 0.31–1.99 kWh·m^{-3}, respectively. These estimated range of energy requirements would be still higher than the average NED reported in this study when dissolved methane recovery is not conducted (−0.169 ± 0.341 kWh·m^{-3}; see Table 2).

Table 2
Energy consumption (EC), total energy recovery (TER) and net energy demand (NED) for 0 %, 50 % and 70 % of dissolved methane recovery (D-CH4).

	D-CH$_4$ Recovery fraction		
kWh·m^{-3}	*0%*	*50%*	*70%*
EC blower DM	-	$2.3{\cdot}10^{-5}$ ± $3.4{\cdot}10^{-6}$	$4.3{\cdot}10^{-5}$ ± $6.3{\cdot}10^{-6}$
EC blower AnR	0.066 ± 0.030		
EC blower MT	0.106 ± 0.047		
EC AnR feeding pump	0.002 ± 0.000		
EC MT feeding pump	0.015 ± 0.007		
EC permeate pump	0.003 ± 0.001		
Total EC	0.192 ± 0.070		
TER	0.361 ± 0.271	0.382 ± 0.306	0.397 ± 0.377
NED	***-0.169 ± 0.341***	***-0.190 ± 0.376***	***-0.205 ± 0.447***

These results emphasize the importance of optimizing the parameters governing the reactor mixing requirements and filtration, especially with regard to SGD$_m$. Increasing sludge recycling would reduce TSS concentration in the MTs and help to reduce SGD$_m$ necessities. Since the MT feeding pump represents only 8 % of EC and membrane scouring accounts for 55 %, this strategy could improve the AnMBR energy performance through a slight increase in the consumption of the sludge pump, which would be compensated by a larger reduction of consumption for membrane scouring. This sludge circulation could also be used to promote reactor mixing and would reduce its energy consumption (34 %).

It should be noted that the significant sulfate content in the influent favors SRB growth, so that improved AnMBR energy performance could be expected when treating low-sulfate wastewater [29].

3.3. GHG emissions

Fig. 3 shows the equivalent CO_2 emissions associated with dissolved CH_4 (D-CH_4) when no recovery is considered (gray area) and net GHG emissions (including D-CH_4 and those associated with EC) considering 0 %, 50 % and 70 % of dissolved methane recovery (orange, blue and green lines respectively).

When no methane recovery was considered, the contribution of D-CH_4 to total emissions accounted for 0.5–0.6 kgCO_{2eq}·m^{-3} during almost the entire period of plant operation. The periods in which the orange line is lower than the corresponding gray bar indicate net energy production (e.g. days 1–130, see Fig. 2a). On these days, savings in indirect CO_2 emissions related to energy production generated a *GHG credit* which reduced the total carbon footprint. This effect was also found to a lesser extent during days 430–480. Conversely, the orange line over the gray area indicates emissions related to net EC, which is added to those of dissolved methane release (days 210–340).

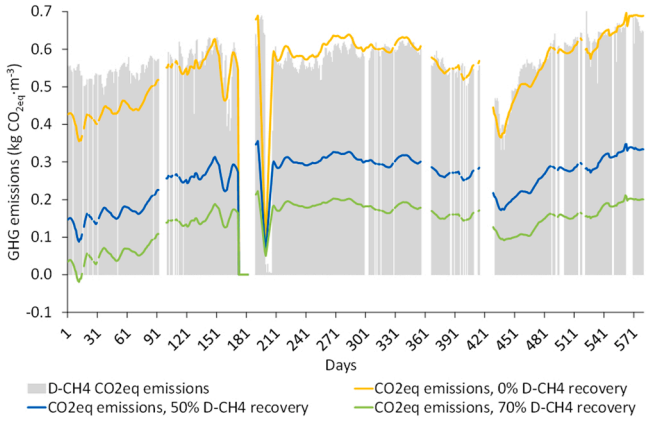

Fig. 3. Evolution of GHG emissions in the operating phase of the demonstration plant: direct CO_2 equivalent emissions of dissolved methane (gray bars), and CO_2 equivalent emissions when considering NED and dissolved methane recovery of 0 % (orange), 50 % (blue) and 70 % (green). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Net GHG emissions with 0 % of methane recovery (orange line) showed values around 0.4 $kgCO_{2eq}·m^{-3}$, when high volumes of biogas were produced and energy recovery was significantly high (days 1–130), whereas the value oscillates around 0.6 $kgCO_{2eq}·m^{-3}$ when low biogas production was obtained. According to Sanchis-Perucho et al. [35], the economic optimum for dissolved methane recovery would be 50 % and the environmental optimum 70 % approx. The results obtained with 50 % and 70 % of dissolved methane recovery (blue and green lines, respectively) show the importance of this process for the improvement of the overall AnMBR environmental performance by reducing CH_4 emissions. For instance, recovering 50 % of the methane would reduce the equivalent CO_2 emissions to 0.1–0.3 $kgCO_{2eq}·m^{-3}$. Operating with 70 % methane recovery kept the emissions below 0.2 $kgCO_{2eq}·m^{-3}$ during the entire study period. By way of comparison, Rodriguez-Garcia et al. [33] estimated emissions of around 0.3 $kgCO_{2eq}·m^{-3}$ for CAS facilities with organic matter and nutrient removal discharging in sensitive areas, while Chen [6] estimated small-scale MBR (351 $m^3·d^{-1}$ treatment capacity) emissions to be around 0.7 $kgCO_2·m^{-3}$.

The GHG emissions associated with D-CH_4 appear as the main contributor to this environmental impact (between 83.8 % and 95.7 %). Since the other contributors are derived from power consumption, the higher use of renewable energies in the energy mix will raise these percentages even further. The improvement of AnMBR will necessarily involve further research in systems of dissolved methane recovery to offset CO_2 emissions. In this regard, AnMBR combined with degassing membranes represents a promising combination, as it merges greater energy recovery with the possibility of reducing GHG emissions [12].

Increasing energy recovery would also help to widen the applicability of AnMBR technology, for example co-digestion of wastewater with other substrates (organic fraction of food waste, food industry wastewater, etc.) has been successfully explored [26]. Biogas upgrading is another strategy that takes advantage of methane; this option has recently received great attention in the UE (e.g. REPowerEU Plan) and promising environmental and economic results have been obtained [3].

Finally, it is important to highlight the additional benefits that recovering nutrients would entail, since N and P production from traditional sources require high power consumption and GHG emissions. McCarty et al. [22] reported 19.3 kWh per kg N and 2.11 kWh per kg P. According to these values, considering the average N and P content in the effluent (45.7 ± 10.4 mg $N·L^{-1}$ and 7.8 ± 2.0 mg $P·L^{-1}$, data not shown) and a GHG emission factor for Spanish energy mix generation of 0.14 kg CO_2.kWh [28], fertigation would allow savings of 0.126 kg $CO_{2eq}·m^{-3}$, which would help the combination of AnMBR+fertigation to achieve carbon neutrality if 70 % of the dissolved methane was recovered.

The results of this study suggest that AnMBR shows promise as a potential technology for reducing the energy dependence and overall environmental impacts of wastewater treatment. The combination of anaerobic treatments and membrane technology in AnMBR paves the way for turning wastewater into a source of resources. Energy, nutrients and reclaimed water could be supplied by the continuously generated sewage, while this approach complies with the fundamental principles of the CE and is fully aligned with the Sustainable Development Goals.

4. Conclusions

An industrial AnMBR wastewater treatment prototype was successfully operated for 580 days. Fouling propensity remained low and long-term operations were achieved, with a maximum continuous operation of 233 days without applying membrane chemical cleaning. The UF modules were able to operate at a high J_{20} in the range of 15–20 LMH and a competitive SGD_m of between 0.1 and 0.4 $Nm^3·m^{-2}·h^{-1}$ (which corresponded to a SGD_P of 10–20 $Nm^3·m^{-3}$). TMP showed a quick response to changes in J_{20} and SGD_m, which facilitated fouling control. Moreover, operation with TSS concentrations below 10 $g·L^{-1}$, J_{20} of 16 LMH and SGD_m of 0.3 $Nm^3·m^{-2}·h^{-1}$ (corresponding to 15 $Nm^3·m^{-3}$) would further lengthen the filtration periods between chemical cleanings.

With regards to energy assessment, the average EC was estimated at 0.192 ± 0.070 $kWh·m^{-3}$, with most of the power consumed by membrane scouring (55 %) and reactor mixing (34 %). The valorization of organic matter provided an average TER of 0.361 ± 0.271, 0.382 ± 0.306 and 0.397 ± 0.377 $kWh·m^{-3}$ with 0 %, 50 % and 70 % recovery of dissolved methane, respectively, obtaining a NED of − 0.169 ± 0.341, − 0.190 ± 0.376 and − 0.205 ± 0.447 $kWh·m^{-3}$, respectively. The increase in energy production associated with the recovery of dissolved methane is therefore low, but higher than the energy consumption of degassing membranes. Finally, dissolved methane recovery appears as the key challenge to be overcome for achieving low GHG emissions with AnMBR. In this study, recoveries of 50 % and 70 % reduced emissions from 0.5 to 0.6–0.1–0.3 and below 0.2 $kgCO_2·m^{-3}$, respectively.

Overall, the AnMBR demo plant showed a suitable filtration performance, significant energy productions and low CO_2 emissions when recovering dissolved methane. These results indicate that AnMBR combined with degassing membranes represents a robust alternative to the conventional wastewater treatment.

CRediT authorship contribution statement

A. Jiménez-Benítez: Writing – original draft, Investigation, Methodology, Formal analysis, Writing – review & editing. **A. Ruiz-Martínez:** Investigation, Methodology, Resources, Writing – review & editing. **Á. Robles:** Investigation, Methodology, Formal analysis, Validation, Writing – review & editing. **J. Serralta:** Resources, Investigation, Validation, Writing – review & editing. **J. Ribes:** Resources, Investigation, Management and coordination responsibility for the research activity planning and execution, Validation, Writing – review & editing. **F. Rogalla:** Investigation, Methodology, Resources. **A. Seco:** Definition, Methodology, Formal analysis, Investigation, Validation, Writing – review & editing, Supervision, Management and coordination responsibility for the research activity planning and execution. **J. Ferrer:** Methodology, Formal analysis, Investigation, Validation, Writing – review & editing, Supervision, Management and coordination responsibility for the research activity planning and execution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2023.109454.

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Life cycle costing of AnMBR technology for urban wastewater treatment: A case study based on a demo-scale AnMBR system

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ABSTRACT

This study aims at assessing the economic performance of a projected full-scale anaerobic membrane bioreactor (AnMBR) for urban wastewater (UWW) treatment at ambient temperature. To this aim, data from an AnMBR demonstration plant (industrial prototype, TRL 6) was used, which was operated for 3 years treating real UWW, allowing gathering a robust set of information for scaling-up to full scale. The obtained results revealed that reactor mixing (0.056–0.124 kWh·kgCOD$_{rem}^{-1}$; 34–57%), and membrane scouring (0.048–0.120 kWh·kgCOD$_{rem}^{-1}$; 22–48%) were the main contributors to the total energy demand; while net energy productions between 0.210 and 0.645 kWh·kgCOD$_{rem}^{-1}$ were achieved. Capital expenditure was highly influenced by UF membranes (€0.029–0.073 kgCOD$_{rem}^{-1}$; 31–49%), combined heat and power technology for energy recovery (€0.012–0.023 kgCOD$_{rem}^{-1}$; 8–24%), and reactor construction (€0.07–0.014 kgCOD$_{rem}^{-1}$; 8–13%); while the main contributors to operating expenditure were energy requirements (€0.042–0.069 kgCOD$_{rem}^{-1}$; 41–46%), membrane replacement (€0.011–0.028 kgCOD$_{rem}^{-1}$; 9–17%), and discharge fee (€0.010–0.020 kgCOD$_{rem}^{-1}$; 9–12%). Total annualized costs showed high variability, between €− 0.003 and 0.188 kgCOD$_{rem}^{-1}$. Results presents AnMBR as a competitive technology for UWW treatment compared to conventional aerobic technologies (e.g., CAS). Membrane fouling control; hydraulic retention time; biogas requirements for reactor mixing and membrane stirring; and energy recovery efficiency were identified as key parameters for improving economic sustainability of AnMBR technology.

1. Introduction

Water management is one of the fields in which new technologies, concepts and approaches are being actively investigated to successfully face the climate change threat, meet Circular Economy (CE) principles and accomplish Sustainable Development Goals [32]. It is therefore crucial to progress towards converting former wastewater treatment plants (WWTP) into full water resource recovery facilities in which urban wastewater (UWW) is regarded as a raw material and no longer as a waste product [13]. This paradigm shift would take advantage of the value embedded in wastewater and boost the economic framework linked to its management.

Among the possibilities for improving the economic performance of wastewater treatment facilities, the combination of anaerobic digestion (AD) and membrane bioreactors (MBR), resulting in the so-called anaerobic membrane bioreactor (AnMBR), is gaining momentum [20,27,36]. Reduced growth rates of anaerobic microorganisms can be compensated, even when operating at low reactor temperatures, since an AnMBR can decouple the sludge retention time (SRT) from the hydraulic retention time (HRT) by membrane filtration. The additional benefits of AnMBR also include:

Abbreviations: AD, anaerobic digestion; AnMBR, anaerobic membrane bioreactor; CAPEX, capital expenditure; CAS, conventional activated sludge; CE, circular economy; CHP, combined heat and power; COD, chemical oxygen demand; DM, degassing membranes; HRT, hydraulic retention time; J_{20}, 20ºC-standardized transmembrane flux; LCC, life cycle costing; MBR, membrane bioreactor; MLTSS, mixed liquor total suspended solids; NED, net energy demand; OLR, organic loading rate; OPEX, operating expenditure; SA, sensitivity analyses; SGD_P, sparging gas demand per m^3 of permeate; SO_4^{2-}-S, sulfate; SRT, sludge retention time; TAC, total annualized cost; TEC, total energy consumption; TER, total energy recovery; UA, uncertainty analyses; UWW, urban wastewater; WWTP, wastewater treatment plant.

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- **Effluent quality:** membrane pore size can ensure microbial quality in the effluent [42], favoring the production of fit-for-purpose reclaimed water.
- **Organic matter valorization:** anaerobic metabolism converts biodegradable organics into methane, which acts as an energy carrier and enhances the energy balance of wastewater treatment [44].
- **Biosolids production and stabilization:** anaerobic metabolism reduces sludge waste management needs due to the low growth yield of anaerobic biomass, while onsite sludge stabilization can be achieved by operating at a high SRT [39].
- **Nutrient recovery:** anaerobic systems conserve most of the mineralized nutrients embedded in the effluent, which favors their recovery [33] or direct reuse through fertigation [19].
- **Reduced footprint:** the MBR configuration eliminates settling units, which reduces the investment costs associated with land for new facilities and favors the retrofitting of existing plants with space constraints [47].

However, AnMBR technology for UWW treatment still faces some operating issues that need to be properly solved. The main bottlenecks for widening the implementation of AnMBR are: sensitivity to process dynamics and development of appropriate control strategies [37]; membrane fouling [16]; and loss of dissolved methane with the effluent [46], which worsen the energy balance and increase the carbon footprint.

Despite the aforementioned challenges, AnMBR has continuously improved its performance, e.g., in terms of effluent quality or maintenance of competitive transmembrane fluxes [34]. However, from an economic perspective, energy demand needs to be further optimized for enhancing its feasibility. Membrane fouling control through gas scouring has been reported as one of the main contributors to energy costs in AnMBR systems [20,29]. Chemical demands for membrane cleaning and membrane replacement also contribute to operating expenses (OPEX) of AnMBR technology [22]. Sludge handling and management is also becoming an emerging global problem [10], so that the reduced sludge production of AnMBRs, jointly with the energy produced through biodegradable organics, can offset the operating costs related to membrane operation and favor the financial framework of AnMBRs against conventional aerobic treatments.

Robust comparisons between different treatment processes and/or operating conditions still represent a complex challenge, especially in the emerging technologies, due to the wide range of assumptions considered, different influent/effluent quality and methodologies applied. In this regard, Ferrer et al. [7] proposed a method of AnMBR design and assessment and obtained a total annual cost (TAC) including CAPEX and OPEX of between €0.101 and €0.097 per m^3 for sulfate-rich UWW and between €0.097 and €0.070 per m^3 for low-sulfate UWW. Using the same method, Pretel et al. [30] calculated a TAC of €0.126–0.169 per m^3 for an AnMBR combined with CAS or MBR-based post-treatment for nutrient removal. These values are in line with those reported by Smith et al. [43] for an AnMBR treating medium- and high-strength wastewater (around €0.124 per m^3). These authors also studied the influence of sludge fate (landfill, incineration, and land application) and concluded that AnMBR costs are lower than high rate activated sludge+AD and CAS+AD when sludge is landfilled, lower than CAS+AD when it is incinerated and lower than AeMBR+AD in any scenario. Xu et al. [48] reported significant differences between AeMBR and AnMBR when water and nutrient reuse is considered: in a discharge scenario, the net present value (NPV) of an AeMBR was $11 million lower than AnMBR, but in a reuse scenario fertilizer savings would allow a NPV $41 million lower for AnMBR. Furthermore, tertiary treatments for disinfection are unlikely to be necessary when ultrafiltration membranes are used, contributing to avoiding or reducing the costs associated with conventional disinfection processes, e.g., UV radiation, chlorination, etc. In this regard, boosting water reuse can also provide revenues for water management systems that could make AnMBR economic performance more attractive.

These results confirm a promising trend, especially when they are compared with the values reported for conventional and aerobic treatments. For example, Iglesias et al. [17] analyzed the costs of full-scale MBRs and extended aeration (EA) facilities in Spain. These authors calculated an OPEX for the studied MBR in the range of €0.20–0.45 per m^3 and an average of €0.22 per m^3 in case of EA. When EA was combined with reclaimed treatments, operating costs rose to €0.31-€0.40 per m^3. Total costs for general MBR technology at more than 50% of its treating capacity was estimated to be €0.20–0.50 per m^3. On the other hand, Xiao et al. [47] reported an OPEX of $0.11–0.20 per m^3 for aerobic MBR in China. EPSAR, the Public Entity for Sewage Treatment in the *Comunitat Valenciana* (Spain), reported €0.33 per m^3 as the average OPEX for all the technologies implemented in this region, based on different aerobic treatments for different treatment capacities [5].

This work aimed to assess the economic performance of a full-scale AnMBR for UWW using Life Cycle Costing (LCC) methodology, which has been widely applied to economic appraisals considering all the costs of constructing, operating, maintaining and disposing of products, processes and services [23]. The results were obtained using the data from an AnMBR demonstration plant treating UWW under different operating conditions within the LIFE MEMORY Project [20,36]. Given the size of the demonstration plant (equipped with commercial ultra-filtration hollow-fiber membranes, industrial pumping and auxiliary elements and control and monitoring devices) and the long operating period (around 3 years with different ambient conditions), data obtained from the plant can be considered as a robust set of information for scaling-up AnMBR to full-scale facilities and favor the adoption of AnMBR and its advantages over conventional UWW treatments.

2. Materials and methods

2.1. AnMBR plant

The demonstration plant comprised a 40 m^3 anaerobic reactor (AnR) (34.4 m^3 working volume) connected to three x 0.8 m^3 membrane tanks (MT-X) (0.7 m^3 working volume), equipped with one ultrafiltration membrane system (PURON® PSH41, 0.03-μm pore size and 41 m^2 total filtration area). The total system filtration area is 123 m^2. The plant also includes a rotofilter (RF, 1.5-mm screen size), an equalization tank (ET, 1.1 m^3), a clean-in-place (CIP) tank (0.37 m^3), and degassing membranes (DM) to recover the methane dissolved in the effluent (further details of DM in Sanchis-Perucho et al. [38]).

For energy calculations, data regarding flow rate, liquid level, pressure (transmembrane, inlet and outlet for blowers, and gas), temperature and methane production was obtained from the available plant instrumentation. Data obtained from the DM unit (transmembrane pressure, flow rates, and methane recovery efficiency) were also used to estimate the dissolved methane recovery potential of the system, according to Sanchis-Perucho et al. [38]. Adiabatic indexes and blower and pump efficiencies were estimated from the standard values for this equipment. Further details of the plant can be found in Robles et al. [34]. The flow diagram and an overview of the AnMBR are provided in e-supplementary materials.

2.2. AnMBR operation

The AnMBR prototype treated effluent from the pre-treatment step (screening and sand removal) of the "Alcázar de San Juan" WWTP. After pre-treatment in the RF and homogenization in the ET, the wastewater was pumped to the AnR. The sludge was continuously recycled through the external membrane tanks (immersed configuration), where the final effluent was obtained by vacuum filtration. A fraction of the biogas produced in the AnR was constantly recirculated from the headspace to the bottom of the reactor through coarse bubble diffusers for stirring purposes. This mixing strategy favored the stripping of dissolved gases

from the liquid phase for their collection. Another fraction of the biogas produced was driven to the bottom of the membrane tanks for membrane scouring.

The membrane operating strategy followed a sequence of 300-s basic F-R cycle (280 s filtration and 20 s relaxation), 40 s of back-flush every 5 F-R cycles, 20 s of ventilation every 15 F-R cycles and 40 s of degasification every 50 F-R cycles.

The selected average operating conditions and characterization of the five scenarios evaluated in this study are provided in e-supplementary materials (see Table S1) and further details can also be found in Robles et al. [36] and Jiménez-Benítez et al. [20]. Influent quality was characterized by a high COD (755 ± 224–1403 ± 532 mg·L^{-1}) and COD: SO_4^{2-}-S ratios between 4.80 ± 2.86 and 8.70 ± 3.69 due to dairy- and wine-industry contributions. SRT of the demo-plant was set to 70 days, based on previous studies on AnMBR technology for urban wastewater treatment at ambient temperature. By achieving high concentrations of biomass in the reactor, an appropriate level of biodegradability can be ensured even at low temperatures. This has been validated in previous studies (e.g., Giménez et al. [12]). However, to prevent the excessive concentrations of suspended solids in the mixed liquor that could negatively affect the filtration process [35], HRT was set at values above 20 h. This allowed maintaining mixed liquor solids concentrations compatible with competitive filtration process performances.

Scenarios I to V were used to scale and estimate the performance of a full-scale AnMBR system that would treat the influent to the "Alcázar de San Juan" WWTP (i.e., 16,000 $m^3 \cdot d^{-1}$). From this scaling, both the energy balance and the LCC were evaluated.

Regarding the biological process, an average COD removal efficiency of 87.2 ± 6.1% was achieved while effluent COD concentration remained below its discharge limits according to European regulations (<125 mg·L^{-1}) despite treating medium-high organic load rates (OLR) (0.60 ± 0.20–1.28 ± 0.38 kg COD·m^{-3}·d^{-1}). Biogas production varied widely throughout the studied periods (1359 ± 999–5558 ± 3204 STP L·d^{-1}), but the methane percentage remained high and stable (75 ± 2–77 ± 2%).

The filtration process performance demonstrated that it was possible to operate the membranes at competitive 20ºC-standardized gross transmembrane fluxes ($J_{20\ gross}$), 15–25 L·m^{-2}·h^{-1}, while maintaining low membrane fouling propensities (0.3–4.9 mbar·d^{-1}) by applying specific gas demands per volume of permeate produced (SGD_P) of 10–20 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$ (see Jiménez-Benítez et al. [20]). These SGD_p were slightly lower than those reported for aerobic MBR by Iglesias et al. [17] (18.3–24.32 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) and to those applied by Pretel et al. [31] to an AnMBR (14–32 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$), both also equipped with hollow fiber membranes.

2.3. Energy assessment

The energy balance was calculated according to the equations proposed in Judd and Judd [21] and Pretel et al. [29] (see e-supplementary materials). The equipment considered for power energy requirements ($W_{elements}$) included rotofilter, pumps (feeding, recycling and permeating), blowers and a sludge dewatering system. Heat energy requirements (Q) were not considered in the study since the full-scale AnMBR was assumed to operate at ambient temperature, as per the demo-scale AnMBR.

Combined Heat and Power (CHP) technology was considered for energy recovery from biogas. Since none of the available CHP systems is suitable for low biogas productions, W_{biogas} was estimated from the biogas flow rate Q_{biogas} (L·d^{-1}), methane percentage in the biogas $\%_{CH4}$, and methane calorific power CV_{CH4} (kJ·m^{-3}). In this case, the widely used reciprocating engines were selected as CHP. These systems can achieve high power and overall CHP efficiencies (24–36% and 66–71%, respectively) according to Darrow et al. [4]. Eq. 1 gives the energy from the biogas recovered in terms of power (W_{biogas} in kW):

$$W_{biogas} = \frac{Q_{biogas} \bullet \%_{CH4} \bullet CV_{CH4} \bullet \%_{power\ efficiency\ CHP}}{1000 \cdot 24 \cdot 3600} \quad (1)$$

An average $\%_{power\ efficiency\ CHP}$ of 35% was selected in this study.

Table S1 (in e-supplementary materials) shows the biogas flow rate and the biogas methane percentage for each scenario. The concentration of dissolved methane in the effluent was calculated from Henry's Law. Saturation conditions were considered for all evaluated temperatures (see Giménez et al. [11]), since coarse bubble diffuser stirring favors methane stripping from the liquid phase thus avoiding oversaturation. The percentage of recovered methane was calculated following previous studies carried out in the demonstration plant [38].

2.4. Life cycle costing

The economic feasibility of the projected full-scale AnMBR was assessed by means of the experimental data provided by the prototype through LCC, which was used to calculate the total annual costs (TAC), defined as the sum of the capital and operating expenses (CAPEX and OPEX, respectively) as in Eq. 2:

$$TAC = \frac{r\ (1 + r)^t}{(1 + r)^t - 1} \cdot CAPEX + OPEX \quad (2)$$

where r is the annual discount rate and t is the depreciation period in years.

A functional unit of 1 kg of removed COD ($kgCOD_{rem}$) was selected for the economic assessment, according to the main pollutant target of AnMBR technology. A depreciation period of 20 years for building works (annual discount rate 3%) and 10 years for equipment (annual discount rate 5%) was used to calculate the CAPEX. For OPEX, the energy costs considered both biogas and dissolved methane recovery by DM. Chemicals for membrane cleaning (sodium hypochlorite and citric acid) were included in OPEX appraisals. Since chemical cleaning degrades membranes, the UF membrane lifespan was calculated considering the cleaning frequency requirement obtained during the demo-scale AnMBR operation and the permissible maximum total contact with chlorine before membrane replacement, i.e., 500,000 ppm-hours cumulative, or a maximum of 10 years, according to the supplier. The lifetime of blowers and pumps was based on the total working hours recommended by the manufacturers (50,000 and 75,000 h, respectively) or also set to a maximum of 10 years. When possible, equipment costs were estimated by correlations between capacities (mainly based on flow rates) and the unit costs provided by the manufacturers, while labor costs were not included. LCC was composed of construction and operating costs over 20 years. Dismantling was not included.

Further considerations when calculating CAPEX and OPEX can be found in the e-supplementary materials. Table 1 shows the unit costs and correlation functions used to assess CAPEX and OPEX in the proposed scenarios.

2.5. Sensitivity and uncertainty analyses

Sensitivity (SA) and uncertainty analyses (UA) were carried out to determine the most significant parameters and the related uncertainty in the obtained results. In this regard, for the purpose of SA, the Standardized Regression Coefficients (SRC) method was applied to select the most influential parameters. A threshold of 0.1 (absolute value) for the standardized regression slope (b_i) was chosen to select the significant factors. The UA was conducted by the Monte Carlo method to evaluate the propagation of uncertainty in the results, which were assessed by means of i) the 5th and 95th percentiles and ii) the empirical cumulative distribution function (eCDF). 2000 model runs were performed for both SA and UA. The sampling matrix was generated by the Latin hypercube sampling (LHS) method with a variability of 5% for the membrane and equipment cost quoted by manufacturers and 10% for the rest of

Table 1
Unit costs and correlation functions used to evaluate capital and operating expenses (CAPEX/OPEX) in the scenarios evaluated.

Unit costs and correlation function of capital and operating expenses		
Pipes, (final cost is function of diameter) €·m^{-1} [a]		8–220
Concrete wall/Floor&cover, €·m^{-3}		350/130
Steel cover, €/m^{2} [b]		51.11
Ultrafiltration hollow-fiber membrane, €·m^{-2}[c]		€·m^{-2} = −2.985·ln A (m^2) + 68.159
Energy	Energy term €·kWh^{-1} [d]	0.231–0.340
	Power term €·kW^{-1}·year^{-1} [d]	1.57–24.73
Sodium hypochlorite (NaOCl Cl active 5%), €·L^{-1}		0.72
Citric acid (Citric acid 1-hidrate), €·kg^{-1}		2.96
Polyelectrolyte, €·kg^{-1}		2.35
Wasted sludge for farming, € per t		4.81
Wasted sludge for landfill, € per t		30.05
Wasted sludge for incineration, € per t		250
Blower, €·unit^{-1} [e]		€·unit^{-1} = 970.66·Q (Nm^3·h^{-1})$^{0.6008}$
Water pumps, €·unit^{-1} [f]		€·unit^{-1} = −0.6582·Q^2 (m^3·h^{-1}) + 263.54·Q (m^3/h) + 1005.4
Sludge pumps, €·unit^{-1} [g]		€·unit^{-1} = 1071.1·ln Q (m^3·h^{-1}) + 19,643
Rotofilter, €·unit^{-1} [h]		€·unit^{-1} = 13.831·Q (m^3·h^{-1}) + 2220.8
Thick-bubble diffuser with 4" Membrane Disc Aeration Head (Barmatec S.L.), €·unit^{-1}		16.94
Dewatering system (centrifuge), €·unit^{-1} [i]		€·unit^{-1} = 7246.1·ln Q (m^3·h^{-1}) + 93,099
CHP, reciprocating engine	€·unit^{-1} [j]	€·unit^{-1} = −2·10^{-9}·kW^3 + 5·10^{-5}·kW^2-0.4842·kW+ 2708.9
	O&M €·kWh^{-1} [j]	€·kWh^{-1} = 0.0192·e$^{-0.0001·kWh}$
PermSelect® Silicone Membrane Module, €·m^{-2}		37.10

[a] Frans Bonhomme España
[b] BEDEC (iTeC)
[c] Koch Membrane System
[d] Energy and power tariff 6.1 TD. Entidad Pública de Saneamiento de Aguas Residuales de la Comunidad Valenciana (EPSAR). Public Entity for Sewage Treatment of Comunitat Valenciana
[e] Pedro Gil S.L.; Tebyc S.L.; FPZ
[f] Sugein S.L.; Tebyc S.L.
[g] Bombas Ideal S.A.
[h] Drainfilter S.L.
[i] Hutchison Hayes Separation Inc.
[j] U.S. Department of Energy: Combined Heat and Power Technology-Fact Sheet Series

parameters considered (further details can be found in e-supplementary materials). OPEX, CAPEX and TAC were used as outputs for SA and UA assessment.

3. Results and discussion

3.1. Energy assessment

Total energy consumption (TEC), total energy recovery (TER) and net energy demand (NED) of the projected full-scale AnMBR plant were evaluated for each scenario (see Table 2 and Fig. 1). These results are discussed in the following sections.

3.1.1. Total energy consumption

shows that the main contributors to the TEC in all the scenarios were reactor mixing (34–57%) and membrane scouring (22–48%). Energy inputs related to methane recovery blowers (0.3–0.5%), rotofilter (0.5–0.7%) and waste sludge pumping (0.1%) were negligible. Moderate energy inputs were required for sludge dewatering (5–9%), feed pumping (1–2%), permeate pumping (2–8%) and sludge recycling (3–4%).

Table 2
Total annualized cost (TAC) comparison of different treatment technologies for urban wastewater.

Technology	Scale	Membrane configuration	TAC (€·kgCOD$_{rem}^{-1}$)	Ref
AnMBR	Pilot+simulation	HF	0.14–0.20	Ferrer et al. [7]
ANMBR	Pilot+simulation	HF	0.18–0.43	Pretel et al. [28]
MBR	Pilot+simulation	HF	0.21	Pretel et al. [30]
MBR	Full	HF	1.74–3.33	Bertanza et al.[2]
CAS	Full	-	1.18–2.51	Bertanza et al.[2]
CAS	Pilot+simulation	-	0.14	Pretel et al. [30]
CAS	Full+simulation	-	0.93–0.95	Jafarinejad [18]
SBR	Full+simulation	-	1.07–1.13	Jafarinejad [18]
EAAS	Full+simulation	-	0.95–1.04	Jafarinejad [18]

AnMBR: anaerobic membrane bioreactor; MBR: membrane bioreactor; CAS: conventional activated sludge; SBR: sequential batch reactor; EAAS: extender aeration activated sludge; HF: hollow fiber

Scenario II required the lowest energy consumption (TEC of 0.166 kWh·kgCOD$_{rem}^{-1}$), mainly due to its comparatively low sludge dewatering, reactor mixing, and membrane scouring requirements (0.009 kWh·kgCOD$_{rem}^{-1}$, 0.056 kWh·kgCOD$_{rem}^{-1}$ and 0.080 kWh·kgCOD$_{rem}^{-1}$, respectively). These results were influenced by the highest COD removal efficiency (equivalent to 20631 kgCOD·d^{-1} at industrial scale) and the lowest HRT (25 ± 1 h) within the operating conditions evaluated in this work. On the other hand, Scenario III required the highest energy consumption (TEC of 0.273 kWh·kgCOD$_{rem}^{-1}$) due to its lower COD removal capacity (12,428 kgCOD·d^{-1} at industrial scale), which increased the energy requirements per kg of COD removed, especially for membrane scouring (0.120 kWh·kgCOD$_{rem}^{-1}$) and reactor mixing (0.097 kWh·kgCOD$_{rem}^{-1}$), even though its HRT (26 ± 2 h) was almost the same as that of Scenario II.

The permeate pumping results also highlighted the importance of maintaining not only low fouling rates (FRs), but also low TMP. For example, although Scenarios III and IV operated at low FR (0.3 and 0.5 mbar·day^{-1}, respectively), the average TMP in Scenario IV (87 mbar) was significantly lower than in Scenario III (462 mbar), resulting in a reduction of the energy consumption from 0.022 kWh·kgCOD$_{rem}^{-1}$ to 0.005 kWh·kgCOD$_{rem}^{-1}$ (77% less), associated with permeate pumping. It is important to consider that filtration performance is influenced by several inter-related biological, physical and chemical factors (e.g., temperature, shear rate, SRT, HRT, extracellular polymeric substances and soluble microbial products concentrations, operation mode, etc.) [26]. The higher temperature (27 ± 1 °C) and lower mixed liquor total suspended solids (MLTSS) concentration (10.4 ± 0.8 g·L^{-1}) in Scenario IV could have led to better sludge filtration properties compared to Scenario III (temperature 19 ± 1°C and 11.3 ± 1.0 g·L^{-1} MLTSS). Additionally, both Scenarios I and IV operated at similar temperature (27 ± 1°C) and lower MLTSS in Scenario I than in Scenario IV (8.4 ± 0.5 and 10.4 ± 0.8 g·L^{-1}, respectively), but TMP was significantly higher in Scenario I (318 mbar) than in Scenario IV (87 mbar). Energy consumption for permeate pumping in Scenario I (0.010 kWh·kgCOD$_{rem}^{-1}$) was hence double that of Scenario IV (0.005 kWh·kgCOD$_{rem}^{-1}$) even though COD removal in Scenario IV (10,815 kgCOD$_{rem}$·d^{-1}) was significantly lower than in Scenario I (19,138 kgCOD$_{rem}$·d^{-1}).

Membrane scouring has been classically identified as a key parameter of AnMBR technology [40]. Scenarios I and II performed with

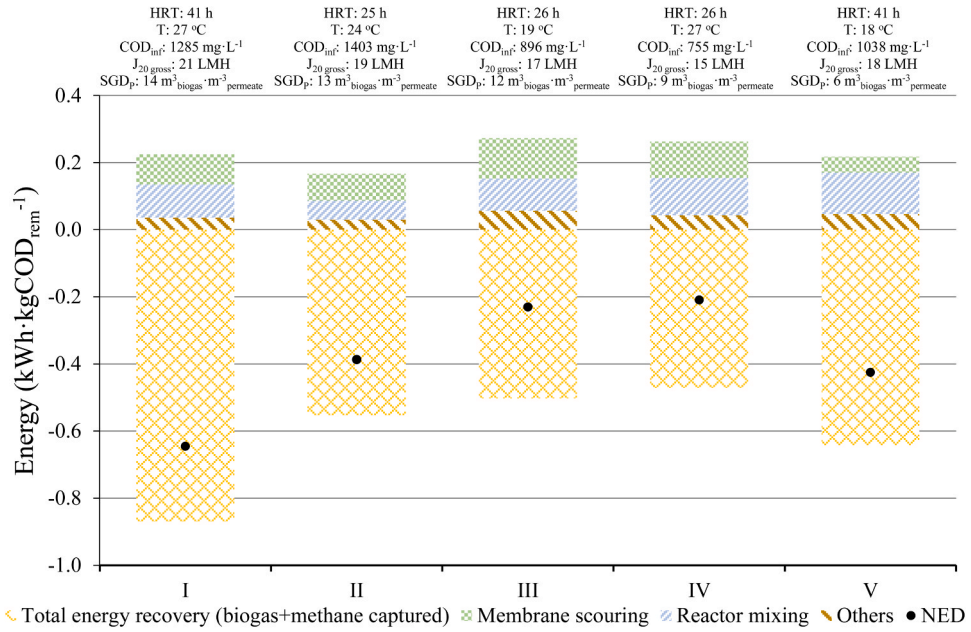

Fig. 1. Total energy consumption (TEC), total energy recovery (TER) and net energy demand (NED) for the projected full-scale AnMBR plant. "Others" refers to: rotofilter; feeding and permeate pumping; sludge waste, recycling and dewatering and methane recovery blowers.

similar high COD removal (19138 and 20631 $kgCOD_{rem}^{-1}·d^{-1}$, respectively) and were operated with similar SGD_p (14 and 13 $Nm^3_{biogas}·m^{-3}_{permeate}$, respectively). Both showed therefore low energy consumption associated with membrane scouring: 0.089 $kWh·kgCOD_{rem}^{-1}$ in Scenario I and 0.080 $kWh·kgCOD_{rem}^{-1}$ in Scenario II. On the other hand, Scenarios II and III were both operated at a similar average SGD_p of 12 and 13 $Nm^3biogas·m^{-3}_{permeate}$ respectively but had different COD removal performance (20631 and 12428 $kgCOD_{rem}·d^{-1}$, respectively). Therefore, Scenario II showed lower energy consumption regarding membrane scouring (0.080 $kWh·kgCOD_{rem}^{-1}$) than Scenario III (0.120 kWh·kgCOD$_{rem}^{-1}$). Finally, Scenario V was operated with the lowest SGD_p (6 $Nm^3_{biogas}·m^{-3}_{permeate}$) and performed with an intermediate COD removal rate (15376 $kgCOD_{rem}·d^{-1}$). Scenario V therefore had the lowest energy expenditure for membrane scouring (0.048 $kWh·kgCOD_{rem}^{-1}$).

These results confirm that the challenging issue of optimizing energy consumption in AnMBR performance depends on multiple factors. Biological conditions in the reactor should ensure high COD removal, not only for accomplishing the regulations, but also to better reach high energy efficiencies. Optimizing HRT to reduce reactor mixing expenditures and designing a comprehensive filtration strategy able to maintain low TMP are crucial to reducing TEC. This strategy should also consider appropriate combinations of $J_{20\ gross}$ and SGD_p.

3.1.2. Total energy recovery

Table 2 shows the TER when energy is recovered from both: only biogas; and biogas and dissolved methane captured from the effluent. The results confirmed the potential of AnMBR for organic matter valorization via methane production, although the energy production values varied widely. Scenarios I and V showed the highest energy recoveries (−0.827 and −0.581 $kWh·kgCOD_{rem}^{-1}$, respectively), while III (−0.429 $kWh·kgCOD_{rem}^{-1}$) and IV (−0.386 $kWh·kgCOD_{rem}^{-1}$) had lower energy productions. Scenario II showed an intermediate result (−0.513 $kWh·kgCOD_{rem}^{-1}$).

These energy outcomes were influenced by both the influent characteristics and the operating conditions (see Table S1 in e-supplementary materials). In Scenarios I and II, the high $COD:SO_4^{2-}$-S ratios (8.7 ± 3.7 and 8.2 ± 4.4) and temperatures (27 ± 1ºC and 24 ± 2ºC) resulted in organic matter conversions with methane yields (Y^{CH4}) of 169.0 ± 95.1 L STP·$kgCOD_{rem}^{-1}$ (Scenario I) and 109.6 ± 70.0 L STP·$kgCOD_{rem}^{-1}$ (Scenario II). Scenario IV showed a reduced methane yield of 70.2 ± 36.0 L STP·$kgCOD_{rem}^{-1}$, even though it operated at a high temperature (27 ± 1 ºC), due to its low $COD:SO_4^{2-}$-S ratio (4.8 ± 2.9) and OLR (0.71 ± 0.23 $kgCOD_{in}·m^{-3}·d^{-1}$). The comparison between Scenarios IV and V also depicted the importance of the $COD:SO_4^{2-}$-S ratio: both scenarios operated at similar OLR (0.71 ± 0.23 and 0.60 ± 0.20 $kgCOD_{in}·m^{-3}·d^{-1}$, respectively) but the lower $COD:SO_4^{2-}$-S ratio in Scenario IV (4.80 ± 2.86) than in Scenario V (6.94 ± 2.83) led the former to reduced methane productions (methane yield of 70.2 ± 36.0 L STP·$kgCOD_{rem}^{-1}$) despite its high operating temperature (27 ± 2ºC).

Hence, increasing the $COD:SO_4^{2-}$-S ratio or increasing the OLR available for methanogenic organisms appears to be a suitable approach to improve energy recovery. For this, additional co-substrates could be explored, such as adding food waste, as proposed by Moñino et al. [25] or Galib et al. [9]. Operating at high temperatures also favors the conversion of organic matter into methane while reducing its losses in the effluent. Both effects contribute to improving energy recovery.

Recovering dissolved methane increased TER by 0.040–0.086 $kWh·kgCOD_{rem}^{-1}$, thus improving the energy balance while reducing the carbon footprint related to fugitive CH_4 emissions. The energy demands associated with the methane recovery blowers appeared to be negligible. These results confirm that degassing membranes are an attractive technology for enhancing AnMBR performance.

3.1.3. Net energy demand

As Table 2 shows, the best NED performance was achieved in Scenario I (−0.645 $kWh·kgCOD_{rem}^{-1}$). In this case, the relatively high reactor mixing energy costs (0.099 $kWh·kgCOD_{rem}^{-1}$) due to the high HRT (41 ± 13 h) could be offset by the highest TER (−0.870 $kWh·kgCOD_{rem}^{-1}$) driven by its operation at higher ambient temperatures (27 ± 1 ºC), OLR (0.75 ± 0.25 $kgCOD·m^{-3}·d^{-1}$) and $COD:SO_4^{2-}$-S ratio (8.70 ± 3.69) than the other scenarios. Scenarios II to V also performed as net energy producers with values between − 0.210 $kWh·kgCOD_{rem}^{-1}$ (Scenario IV) and − 0.425 $kWh·kgCOD_{rem}^{-1}$ (Scenario V).

By way of comparison, Hernández-Sancho et al. [15] reported an average NED of 1.684 ± 1.428 $kWh·kgCOD_{rem}^{-1}$ based on data provided by the EPSAR for 177 WWTP in Valencia (Spain), mainly obtained from conventional aerobic-based facilities. More recently, Silva and Rosa [41] evaluated 17 Portuguese WWTPs based on extended aeration and CAS,

and found an average of 1.3 kWh·kgCOD$_{rem}^{-1}$ and a 25–75 percentile range between 0.5 and 1.8 kWh·kgCOD$_{rem}^{-1}$. Gurung et al. [14] calculated the NED for the CAS-based Finnish Mikkeli WWTP as 0.81 kWh·kgCOD$_{rem}^{-1}$, and identified AnMBR and co-digestion of sludge and organic waste as suitable technological options for retrofitting the facility. Vaccari et al. [45] assessed a total of 267 activated sludge WWTPs in Italy and obtained a range between 3.2 kWh·kgCOD$_{rem}^{-1}$ (treatment capacity < 2000 person-equivalent) and 0.85 kWh·kgCOD$_{rem}^{-1}$ (treatment capacity > 100,000 person-equivalent). A similar study conducted in Greece [3] concluded that WTTPs with above 100,000 person-equivalent treatment capacities operated at a NED of 0.35 kWh·kgCOD$_{rem}^{-1}$ and 0.95 kWh·kgCOD$_{rem}^{-1}$ below 10,000 person-equivalent. Barillon et al. [1] reported net energy demands of six full-scale MBR with two membrane configurations (flat sheet and hollow fiber). The results indicated that energy consumption was approximately within the range of 0.7–2.1 kWh·kgCOD$_{rem}^{-1}$. Pretel et al. [30] simulated an aerobic MBR treating urban wastewater and obtained net energy demands between 0.99 and 1.02 kWh·kgCOD$_{rem}^{-1}$. Finally, Longo et al. [24] gathered energy consumption data of 601 WWTP with different treatments schemes and reported average NED of 2.91, 1.30, and 0.57 kWh·kgCOD$_{rem}^{-1}$ for MBR, extended aeration, and CAS, respectively.

It is important to emphasize that the above-mentioned energy consumption from aerobic-based WWTPs also included nitrification, not occurring in an AnMBR treatment. According to Fraia et al. [8], energy consumption for nitrogen removal would be 6.08 kWh per kg of influent total nitrogen (kWh·TN$_{in}$) in conventional WWTP with high nitrogen removal efficiency (> 89.2%). Considering the influent nitrogen load in the five scenarios (35.6–54.7 mg N·L^{-1}), the aforementioned removal efficiency for this nutrient would be equivalent to an increase in energy consumption in the 0.202–0.370 kWh·kgCOD$_{rem}^{-1}$ range. NED for AnMBR with nitrogen removal would therefore still be lower than the energy requirements for conventional treatments.

The NED results obtained illustrate the potential of AnMBR systems for recovering resources from organic matter, even when treating sulfate-rich UWW, and for improving the energy efficiency of treatment facilities. The main drivers of the NED results are the OLR and COD removal capacity, HRT, temperature and the COD:SO_4^{2-}-S ratio. In this sense, a previous study focused on the biological performance of the demo-scale AnMBR [36] conducted a mass balance and concluded that the influent COD transformed into biogas was between 12% and 44%, while dissolved methane account for 5–9%. COD consumed by sulfate-reducing bacteria was calculated as 23–42% and the organic matter removed via waste sludge 18–31%. Therefore, there is still room to improve the recovery percentage and, consequently, the energy balance of AnMBR technology.

3.2. Life cycle costing

3.2.1. Capital expenditure

Fig. 2 summarizes the CAPEX results obtained for the projected full-scale AnMBR system (detailed results and relative weights can be found in the e-supplementary material).

These results showed the lowest value for Scenarios II (€0.080 kgCOD$_{rem}^{-1}$). Scenarios I (€0.096 kgCOD$_{rem}^{-1}$) and V (€0.109 kgCOD$_{rem}^{-1}$) presented similar investment cost, while Scenario IV obtained the highest CAPEX (€0.147 kgCOD$_{rem}^{-1}$).

UF membranes, anaerobic reactor building, blowers, and reciprocating engines were the main contributors to CAPEX. The reactor construction involved an investment of between €0.007 kgCOD$_{rem}^{-1}$ (Scenario II) and €0.014 kgCOD$_{rem}^{-1}$ (Scenario V), representing around 8–13% of the CAPEX. Since reactor volume is determined by the HRT selected in each scenario, there is clear room for optimization of savings in this constructive element. In this regard, Scenario II required the lowest reactor investment per kg of COD removed due to the combination of the highest COD removal (20631 kgCOD·d^{-1}) and the lowest HRT (25 ± 1 h).

In terms of equipment, the results obtained show that UF membranes were among the main contributors to CAPEX. The membrane acquisition cost varied from €0.029 to €0.073 kgCOD$_{rem}^{-1}$ (between 31% and 49% of CAPEX). Strategies for maximizing filtration process productivity (e.g., optimizing transmembrane fluxes, gas sparging intensities for membrane scouring and downtime for physical membrane cleaning) are required to reduce membrane needs and minimize CAPEX. Enhancing material effectiveness to preventing membrane fouling and therefore maintaining high fluxes; improving membrane scouring procedures; revamping module configurations; and membrane market trends (i.e., reduction of membrane acquisition costs) would also help to reduce this item.

Investment in reciprocating engines also appeared as a relevant cost.

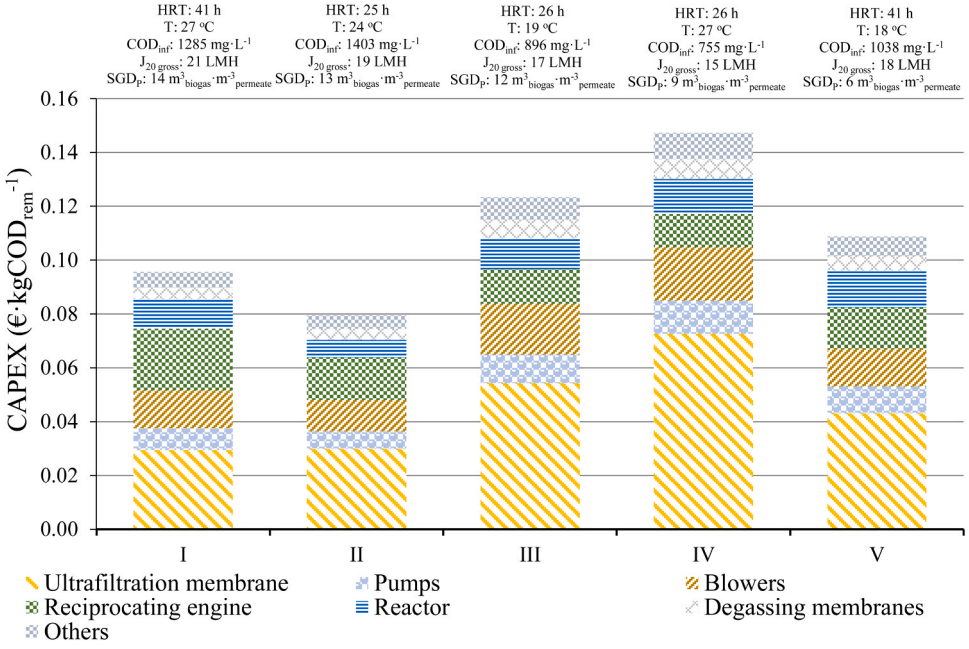

Fig. 2. Allocation of CAPEX for each scenario. "Others" refers to: pumps; degassing membranes; diffusers; pipes; sludge centrifuge; membrane tanks and rotofilter.

Scenario I showed the highest CAPEX allocation for this equipment (€0.023 $kgCOD_{rem}^{-1}$) due to the highest biogas production (5239 ± 4435 $m^3 \cdot d^{-1}$), which entails equipment with greater capacity. Scenario IV required the lowest capital expenditure for this unit (€0.012 $kgCOD_{rem}^{-1}$) because of its low biogas production (1359 ± 999 $m^3 \cdot d^{-1}$). Investment in CHP varied between 8% (Scenario IV) and 24% (Scenario I) of CAPEX.

Blowers also showed a significant contribution to CAPEX. In this case, Scenarios III and IV had the highest expenditures, with €0.019 and €0.020 $kgCOD_{rem}^{-1}$, respectively. Despite their low operating HRT (26 ± 2 h in both scenarios) and intermediate SGD_P (12 and 9 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$, respectively), their comparatively low organic matter removal resulted in a high blower cost per kg of COD removed. On the other hand, Scenario II with similar SGD_P (13 $Nm^3_{biogas} \cdot m^{-3}_{permeate}$) than Scenario III but the highest COD removal showed the lowest blower cost (€0.012 $kgCOD_{rem}^{-1}$). Investment in blowers was between 13% and 15% of CAPEX.

Lastly, pumps (8–9%), DM (4–6%), centrifuge for sludge dewatering (2–3%), rotofilter (2–3%), and pipes (1%) contributed less to CAPEX while membrane tanks (0.2–0.3%) and diffusers (0.1%) were negligible in all scenarios.

Reducing CAPEX is thus crucial to improving investment expenditure competitiveness for AnMBR compared to aerobic MBR and CAS. The strategy to reduce CAPEX involves optimizing the HRT to adjust total reaction volume, increasing the filtration productivity to reduce the membrane needs, properly selecting the CHP power to avoid oversizing and optimizing the capacities of the blowers.

3.2.2. Operating cost

As can be seen in Fig. 3, OPEX varied between €− 0.099 $kgCOD_{rem}^{-1}$ (Scenario I) and €0.041 $kgCOD_{rem}^{-1}$ (Scenario IV) (detailed results and relative weights can be found in e-supplementary material). One of the main contributors to OPEX was TEC (€0.042–0.069 $kgCOD_{rem}^{-1}$), representing 41–46% of the total operating cost, excluding energy recovery.

UF replacement was also a significant OPEX cost. In this regard, Scenario IV, with the lowest $J_{20\ gross}$ (15 LMH) required the largest membrane area and therefore showed the highest replacement cost (€0.028 $kgCOD_{rem}^{-1}$) while Scenario I with the highest $J_{20\ gross}$ (21 LMH) showed the lowest UF replacement cost (€0.011 $kgCOD_{rem}^{-1}$). This expense was between 9% and 17% of OPEX, excluding energy recovery. It is important to highlight that FR directly affected UF membrane replacement cost since membrane lifespan and the associated replacement frequency is determined by the maximum total contact with chlorine permissible during membrane cleaning. However, in this study all the scenarios required one replacement over the considered lifespan of the facility so that there were no differences in this concept.

The contribution of reciprocating engines to OPEX was high in Scenarios I (€0.017 $kgCOD_{rem}^{-1}$; 14%), II (€0.012 $kgCOD_{rem}^{-1}$; 12%) and V (€0.011 $kgCOD_{rem}^{-1}$; 9%), in which a significant amount of biogas was produced. However, overall CHP contributed with savings of €0.099–0.181 per $kgCOD_{rem}$ when CAPEX and OPEX are balanced with cost reductions due to energy recovery. Moreover, these revenues do not consider the heat recovered and the environmental benefits of avoiding methane release, which is an additional value to take into account. Further improvement in CHP efficiencies would also improve AnMBR economic and environmental performance. Revenues could also be increased if energy recovery from biogas were to be conducted in the tariff periods with the highest energy costs and not proportionally distributed throughout the day, as considered in this study. However, this optimization strategy would entail an additional infrastructure (e.g., gasometer) and economic appraisals to assess its feasibility.

The fee for effluent discharging to surface water significantly contributed to the operating costs (€0.010–0.020 $kgCOD_{rem}^{-1}$; 9–12% of OPEX), excluding energy recovery. Reducing or even removing this charge in case of water (and nutrient) recovery could be a proper governance instrument for promoting this practice, taking advantage of the economic, environmental, and social benefits of water (and nutrient) recovery. By way of example, Jiménez-Benítez et al. [19] evaluated two case studies of fertigation with AnMBR effluent in Spain and Italy, showing up to 100% in mineral fertilizers savings, and obtaining reductions of 75% (Spain) and even a carbon sink effect (Italy) when CO_2 emissions associated with savings on fertilizer production are considered. For this, appropriate reclaimed water management tools should be implemented, e.g., according to the Regulation (EU) 2020/741 on the minimum requirements for water reuse [6].

Finally, it is important to highlight the reduced OPEX associated with sludge conditioning (€0.002–0.003 $kgCOD_{rem}^{-1}$; 2%) and sludge disposal (€0.002–0.004 $kgCOD_{rem}^{-1}$; 2%), which confirm the potential benefits of

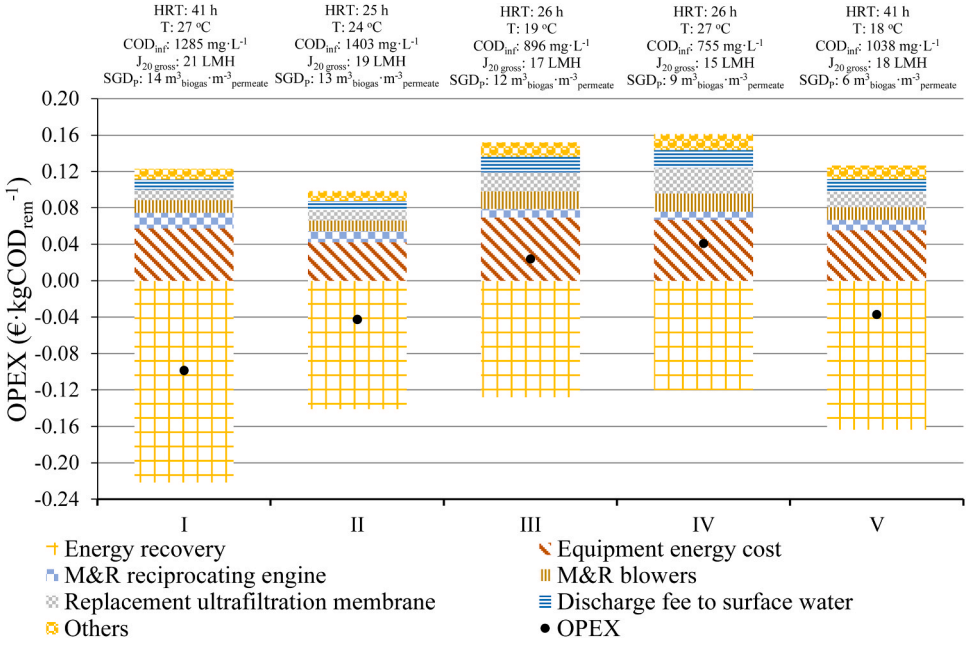

Fig. 3. Allocation of OPEX for each scenario. "Others" refers to: power cost; membrane cleaning; sludge conditioning; sludge disposal; M&R pumps; M&R CHP; M&R blowers; M&R civil works; M&R pipes and diffusers; M&R rotofilter; discharge fee to surface water; and degassing membrane replacement.

AnMBR as an alternative to aerobic treatments (e.g., CAS, EA) to address waste sludge management.

These results are in agreement with the trends found by Judd [22] in which equipment energy needs, UF membrane replacement and discharge fees are the main costs in the OPEX.

3.2.3. Total annual cost

Fig. 4 shows that TAC varied from €− 0.003 $kgCOD_{rem}^{-1}$ (Scenario I) to €0.188 $kgCOD_{rem}^{-1}$ (Scenario IV). The results from Scenario I are explained by a low CAPEX (€0.096 $kgCOD_{rem}^{-1}$) mainly due to the lower UF membrane cost (€0.029 $kgCOD_{rem}^{-1}$) resulting from applying high $J_{20\ gross}$ (21 LMH); and the lowest OPEX (€−0.099 $kgCOD_{rem}^{-1}$) thanks to its high energy recovery (€−0.222 $kgCOD_{rem}^{-1}$). On the other hand, Scenario IV had the highest CAPEX (€0.147 $kgCOD_{rem}^{-1}$) influenced by its lower $J_{20\ gross}$ (15 LMH) and therefore high UF membrane cost (€0.073 $kgCOD_{rem}^{-1}$); and OPEX (€0.041 $kgCOD_{rem}^{-1}$) influenced by its high total maintenance and replacement costs (€0.068 $kgCOD_{rem}^{-1}$) and equipment energy costs (€0.067 $kgCOD_{rem}^{-1}$).

Table 2 shows TAC results reported in bibliography for different treatment technologies for urban wastewater. It is important to highlight the difficulties involved for achieving robust comparisons due to the variety of functional units, assumptions and hypotheses, or treatment schemes within the reviewed studies. Therefore, the comments presented here should be considered as indicative. Ferrer et al. [7] modelled an AnMBR with 50,000 $m^3 \cdot d^{-1}$ of treatment capacity. These authors obtained a TAC between €0.14 $kgCOD_{rem}^{-1}$ (low-sulfate influent and considering methane captured from the effluent) and €0.20 $kgCOD_{rem}^{-1}$ (sulfate-rich influent without methane capture). Pretel et al. [28] simulated an AnMBR treating sulfate-rich and low-sulfate wastewater at 15 and 30 °C and obtained TAC in the range €0.18–0.43 $kgCOD_{rem}^{-1}$. With regards to MBR technology, Pretel et al., [30] obtained a TAC of €0.21 $kgCOD_{rem}^{-1}$, which are significantly lower than those obtained by Bertanza et al. [2] (€1.74–3.33 $kgCOD_{rem}^{-1}$). In the case of CAS, Pretel et al., [30] calculated a TAC of €0.14 $kgCOD_{rem}^{-1}$, while Bertanza et al. [2] reported much higher TAC for the same technology (€1.18–2.51 $kgCOD_{rem}^{-1}$). Lastly, Jafarinejad [18] modelled three different treatments schemes: CAS, sequencing batch reactor and extended aeration activated sludge. The author obtained TAC of €0.93–0.95, €1.07–0.13, and €0.95–1.04 $kgCOD_{rem}^{-1}$, respectively. According to these values, the AnMBR system evaluated in the present study would be competitive even in the case of Scenario IV, which showed the highest TAC, i.e., €0.188 $kgCOD_{rem}^{-1}$.

Finally, CAPEX appears as the main contributor to TAC in Scenarios II to V, which is consistent with the results reported by Smith et al. [43]. Scenario I is especially relevant, in which the high energy production offset the other operating costs and CAPEX, bringing the TAC close 0 (€−0.003 $kgCOD_{rem}^{-1}$). Optimization strategies should thus be used to address both CAPEX and OPEX, since both items showed a potential for reducing TAC and contributing to the economic sustainability of AnMBR-based wastewater treatment.

3.3. Sensitivity analysis

By way of example, the results from the SA of Scenario I are shown in Fig. 5, providing the main influencing parameters ($b_i \geq 0.1$).

3.3.1. OPEX output

Fig. 5a shows the SA results for OPEX. The positive main influencing parameters were HRT, blower cost, biogas for reactor mixing, SGD_P applied to membrane tanks and the AnMBR lifespan. Most of these were related to the biogas sparging needs to mix the reactor or stir the membranes. Reducing gas sparging intensities would thus entail significant savings in OPEX. Optimizing HRT would also reduce reactor volume and hence mixing requirements. Finally, extending the AnMBR lifespan by 10%, from 20 to 22 years, would increase the replacement cost associated with membranes and equipment and thus also the OPEX. In Scenario I, it would be needed to replace both the UF membranes and the equipment (lifespan of 10 years) once throughout the lifespan set for the AnMBR plant (i.e., 20 years). By applying an uncertainty of 10%, AnMBR lifespan would increase from 20 to 22 years in the upper boundary. Thus, new membranes and equipment would be necessary for years 21 and 22. However, the total cost of these new devices would not be distributed in a decade, but only among the two additional years, increasing therefore the total replacement cost for this scenario. In this respect, the second replacement for membranes and equipment would be on service only a 20% of their potential durability.

In relation to the parameters with negative influence, increasing the membranes' useful life and effective filtration time (i.e., operating

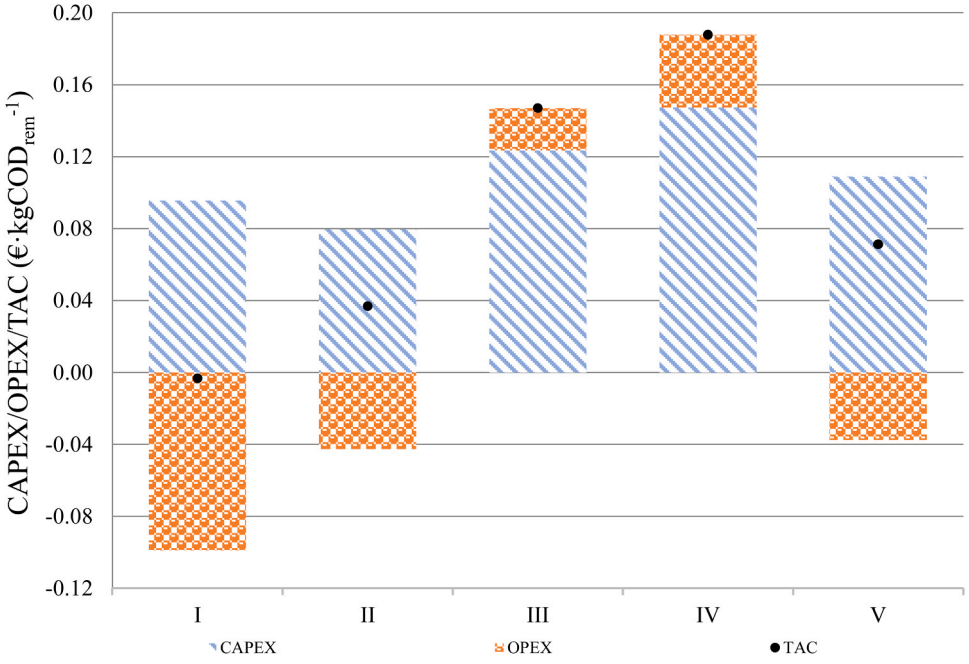

Fig. 4. Allocation of total annual cost (TAC) for each scenario.

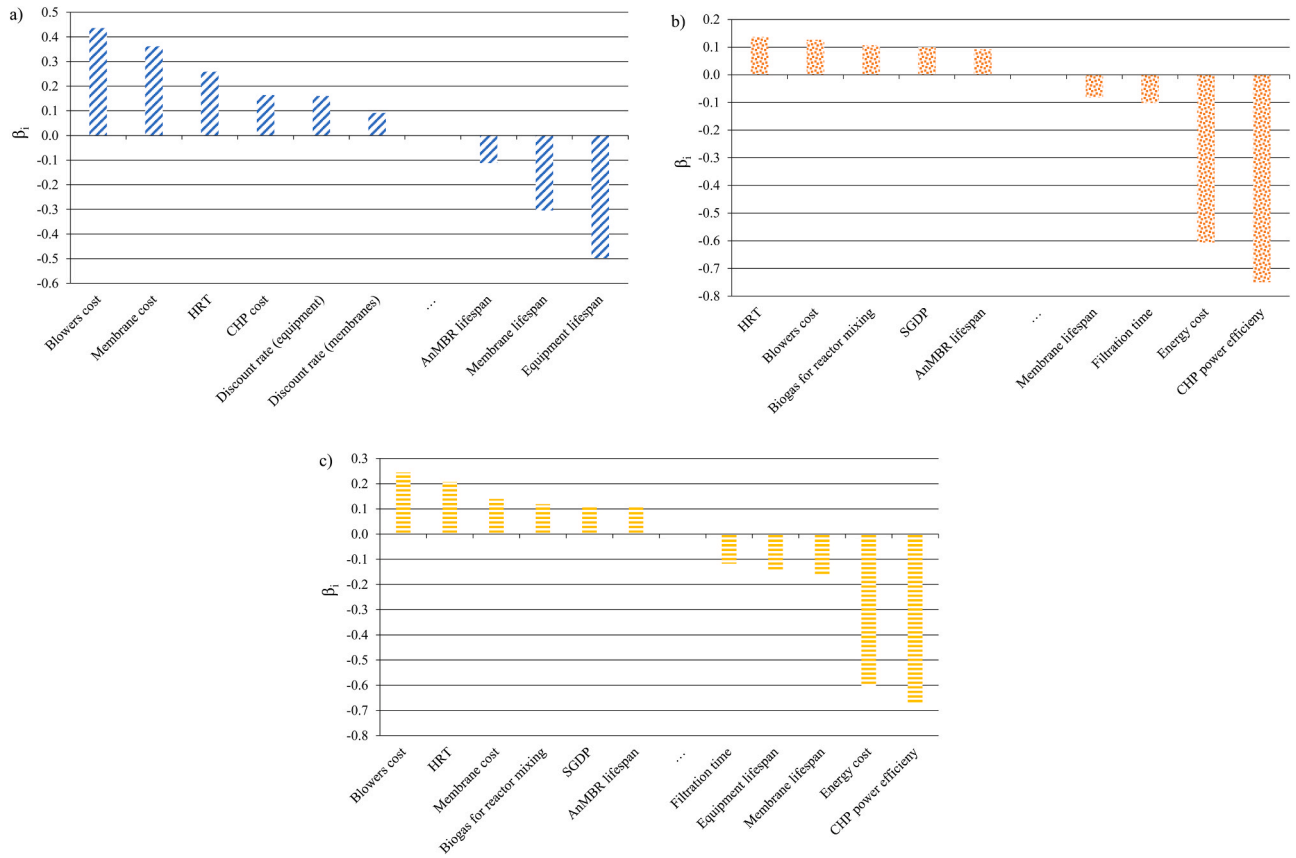

Fig. 5. Sensitivity analysis results for Scenario I. Main influential factors on model outputs: a) CAPEX, b) OPEX, and c) TAC.

period without downtime for physical cleaning of the membrane) would reduce the OPEX related to membrane replacement and filtration productivity. The largest effect would be associated with energy costs and CHP efficiency. Since Scenario I showed high energy production and performed as a net energy producer, favoring energy recovery through improved CHP power efficiency, and increasing the energy price for its sales would produce additional revenues and reduce OPEX. It is important to highlight that the effect of energy price would be the opposite in a net energy consumer scenario.

3.3.2. *CAPEX output*

The parameters with the greatest influence on CAPEX are shown in Fig. 5b. The SA results indicated that the costs associated with the blowers, membranes, size of the reactors (through the HRT) and capacity of the CHP (and its consequent cost) had a significant direct influence on CAPEX, as did the discount rates applied to the acquisition of membranes and equipment.

Therefore, reducing the gas sparging requirements, improving the filtration productivity (and therefore the necessary membrane surface), and optimizing the HRT to reduce the volume of the reactors, would be useful strategies to reduce CAPEX. Moreover, an accurate selection of CHP capacity would also be important to avoid over dimension since its influence on CAPEX was also significant.

Finally, CAPEX could also be reduced by increasing the useful lifespan of the plant, membranes and equipment. This necessarily involves adequate maintenance and optimization of the filtration operation, thus reducing the need for chemical cleaning of the membranes, which is mainly responsible for their deterioration.

3.3.3. *TAC output*

Fig. 5c shows the input parameters with a strong influence on TAC output, all of which were identified as influential parameters on CAPEX and OPEX outputs. The elements associated with gas sparging (blowers, SGD_P and reactor mixing) jointly with the equipment replacement needs derived from extending the AnMBR lifespan implied an increase in TAC. On the other hand, increasing the membrane and equipment lifespan, improving filtration productivity, and obtaining better conditions for energy recovery (e.g., through higher profits from the sale of electric power, or an improvement in the efficiency of CHP systems) would improve the overall economic assessment represented by the TAC.

Finally, by way of example, Fig. 6 shows the Monte Carlo simulation results for TAC with regard to CHP power efficiency (Fig. 6a) and energy tariff variation (Fig. 6b) in Scenario I. Fig. 6a shows that CHP power efficiencies over 38.5% would lead to a TAC below €0 $kgCOD_{rem}^{-1}$ in all simulations, which represents revenues for treating wastewater. On the contrary, cogeneration energy efficiencies lower than 31.5% obtained a positive TAC in all simulations, which indicates an economic outlay for treating the water. In those situations, with efficiencies between 31.5% and 38.5%, the TAC may be positive or negative depending on the rest of the variables, but the probability of a negative TAC increases as the efficiency of the CHP increases.

Moreover, a 10% increase in the price of the electricity sold would led to a negative TAC (see Fig. 6b), while a similar reduction of in the energy price would imply a TAC greater than €0 $kgCOD_{rem}^{-1}$. As already mentioned in Section 3.3.1, the influence of the electricity tariff on the results is marked by the fact that the scenario considered performed as a net producer of energy. Otherwise, an increase in the energy tariff would increase costs. These results highlight the importance of considering energy in AnMBR feasibility studies.

3.4. *Uncertainty analysis*

Fig. 7 gives the empirical cumulative distribution function (eCDF) and confidence interval for the OPEX, CAPEX and TAC outputs in

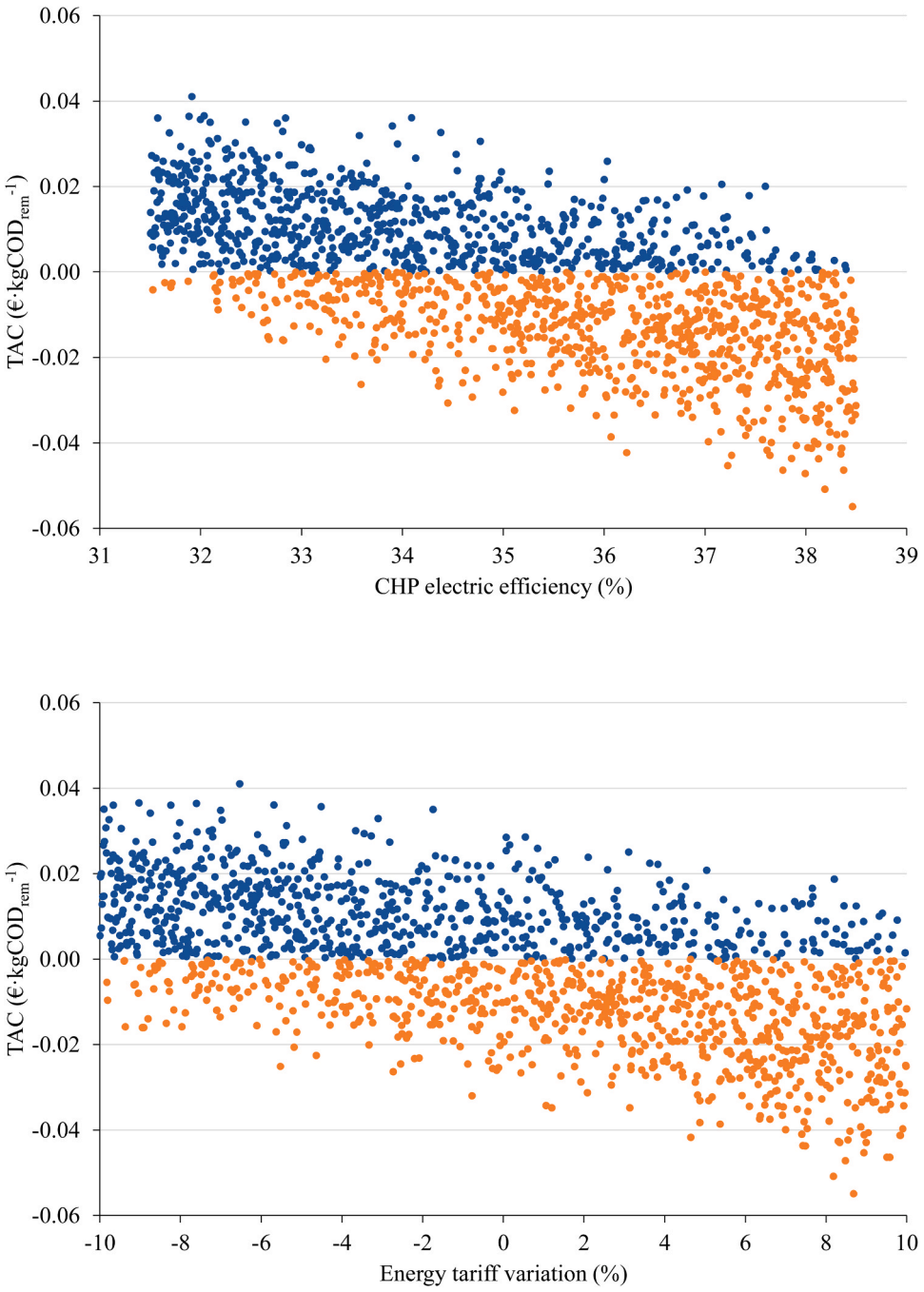

Fig. 6. Monte Carlo simulation results for TAC regarding a) CHP power efficiency (%) and b) energy tariff variation (%). Red dots indicate simulation outputs with TAC < 0 and blue dots represent simulation outputs with TAC > 0.

Scenario I. The eCDF followed a standard normal distribution in all cases and the narrow confidence bands indicated low variability of each value. Fig. 7 also shows that CAPEX remained at positive values, which is coherent with investment requirements, while OPEX sign mainly depends on energy recovery. Finally, Fig. 7c shows that the TAC could change from negative to positive results, going from a benefit scenario for water treatment to one of treatment expenditure.

Regarding the percentiles, the 5th-95th range for OPEX, CAPEX, and TAC resulted in €0.014, €0.051, and €0.052 per kgCOD$_{rem}$, respectively (see Table 3)(Table 4). These results show that the uncertainty in CAPEX is higher than in OPEX, being the former the main contributor to TAC uncertainty. Therefore, in order to reduce sources of uncertainty, efforts should be made to obtain reliable and robust investment data, while a careful design of AnMBR systems should be conducted to ensure an optimal performance. According to Section 3.3, improving data regarding UF membranes, CHP and blower cost would reduce CAPEX uncertainty. In the case of OPEX, accurate data related to reactor mixing and membrane sparging requirements and revenues through energy recovery (CHP power efficiency and energy tariff) would also improve overall UA.

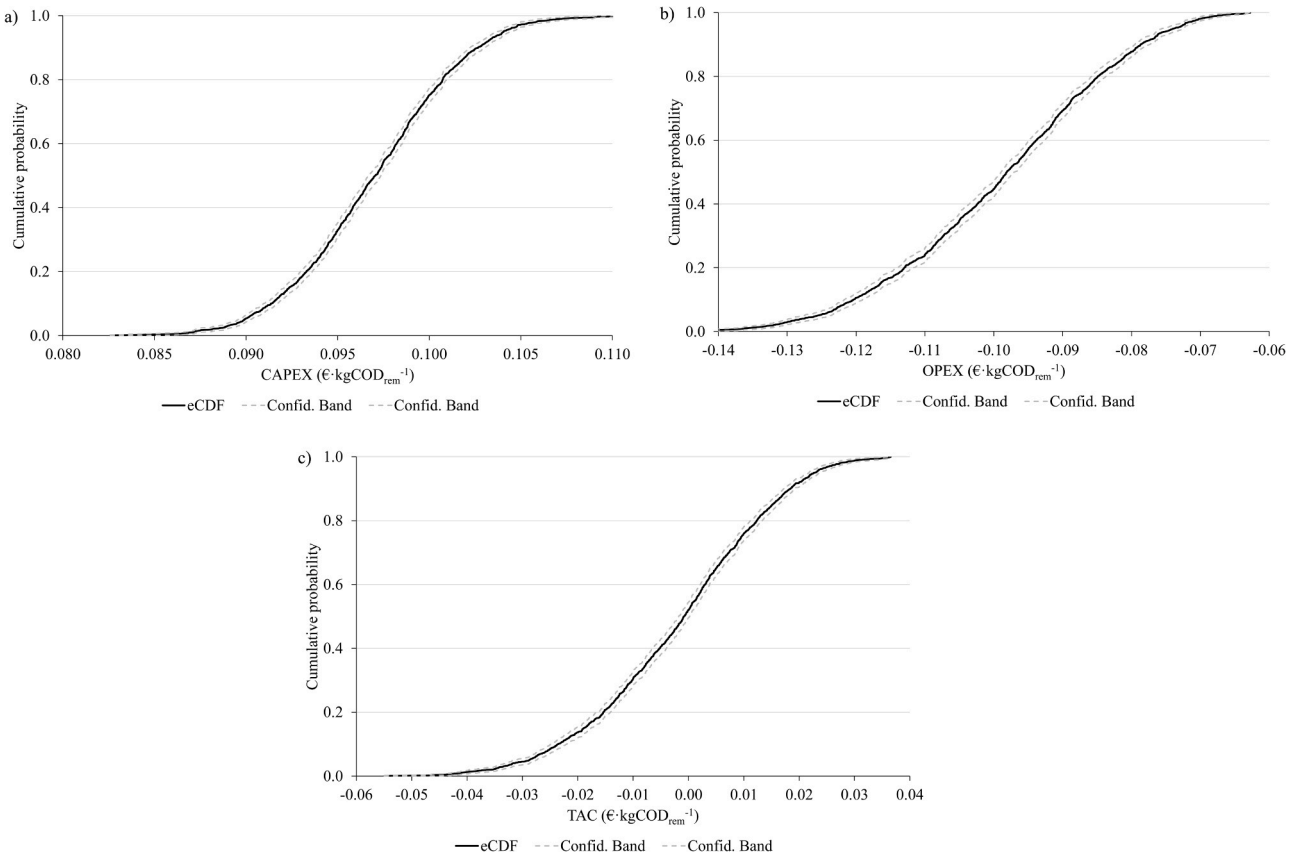

Fig. 7. Uncertainty analysis results for Scenario I. Empirical cumulative distribution function (eCDF) for: a) CAPEX, b) OPEX, and c) TAC.

Table 3
Power energy requirements, total energy recovery (TER), total energy consumption (TEC), net energy demand (NED) and % of TEC for each scenario.

Scenario	Rotofilter	Feeding pumping	Permeate pumping	Membrane scouring	Sludge wasting	Sludge recycling	Sludge dewatering	Reactor mixing	Dissolved methane vacuum blower	TER (biogas + dissolved methane capture)	TER (only biogas)	TEC	NED
						kWh·kgDQO$_{rem}^{-1}$							
I	0.001	0.002	0.010	0.089	≈ 0.000	0.006	0.016	0.099	0.001	-0.870	-0.827	0.225	-0.645
II	0.001	0.003	0.010	0.080	≈ 0.000	0.006	0.009	0.056	0.001	-0.553	-0.513	0.166	-0.387
III	0.002	0.005	0.022	0.120	≈ 0.000	0.010	0.016	0.097	0.001	-0.503	-0.429	0.273	-0.230
IV	0.002	0.006	0.005	0.108	≈ 0.000	0.011	0.018	0.112	0.001	-0.472	-0.386	0.262	-0.210
V	0.001	0.003	0.014	0.048	≈ 0.000	0.008	0.019	0.124	0.001	-0.643	-0.581	0.218	-0.425
							TEC (%)						
I	0.5	1.0	4.4	39.8	0.1	2.8	6.9	44.3	0.3				
II	0.6	1.7	6.3	48.2	0.1	3.6	5.4	33.8	0.3				
III	0.6	1.8	8.1	44.1	0.1	3.6	5.7	35.6	0.5				
IV	0.7	2.1	1.9	41.0	0.1	4.3	6.8	42.6	0.4				
V	0.6	1.3	6.3	21.9	0.1	3.6	8.9	56.8	0.5				

Table 4
Uncertainty results for Scenario I: 5th-95th percentile and 5th-95th range.

	€·kgCOD$_{rem}^{-1}$		
	CAPEX	OPEX	TAC
5th percentile	0.090	-0.125	-0.029
95th percentile	0.104	-0.074	0.023
5th-95th range	0.014	0.051	0.052

4. Conclusions

Favorable energy balances were achieved in all the scenarios evaluated. NED varied between − 0.210 kWh·kgCOD$_{rem}^{-1}$ and − 0.645 kWh·kgCOD$_{rem}^{-1}$. Reactor mixing (0.056–0.124 kWh·kgCOD$_{rem}^{-1}$) and membrane scouring (0.048–0.120 kWh·kgCOD$_{rem}^{-1}$) showed the highest energy consumption.

The major contributors to CAPEX were UF membranes (€0.029–0.073 kgCOD$_{rem}^{-1}$; 31–49%), reciprocating engines acquisition (€0.012–0.023 kgCOD$_{rem}^{-1}$; 8–24%), and reactor building (€0.007–0.014 kgCOD$_{rem}^{-1}$; 8–13%). OPEX was mainly influenced by equipment energy needs (€0.042–0.069 kgCOD$_{rem}^{-1}$; 41–46%), UF membrane replacement (€0.011–0.028 kgCOD$_{rem}^{-1}$; 9–17%) and discharge fee

(€0.010–0.020 $kgCOD_{rem}^{-1}$; 9–12%). CHP maintenance and replacement was also a significant operating cost (€0.009–0.017 $kgCOD_{rem}^{-1}$; 6–14%), although it finally provided revenues through energy sale (€0.120–0.222 $kgCOD_{rem}^{-1}$). The TAC varied between -€0.003 and €0.188 $kgCOD_{rem}^{-1}$, with CAPEX as the main cost.

The results obtained from LCC assessment combined with SA and UA showed that AnMBR represents a potential alternative to conventional aerobic treatments. Its definitive impulse will go through, among other aspects, reinforcing its economic sustainability, for which it is necessary to optimize operational aspects (e.g., HRT, transmembrane flux, reactor mixing and membrane sparging) and technological developments that allow increasing membrane lifespan and improve CHP power efficiency. Finally, other aspects that would also result in improving its economic performance have to do with nutrient recovery present in its effluent and favorable pricing for the sale of electricity produced from the methane generated.

CRediT authorship contribution statement

A. Jiménez-Benítez: Writing – original draft, Investigation, Methodology, Formal analysis, Writing – review & editing. **A. Ruiz-Martínez** Investigation, Methodology, Resources, Writing – review & editing. **J. Ferrer:** Investigation, Validation, Writing – review & editing, Supervision, Management and coordination responsibility for the research activity planning and execution. **J. Ribes:** Resources, Investigation, Management and coordination responsibility for the research activity planning and execution, Validation, Writing – review & editing. **F. Rogalla:** Investigation, Methodology, Resources. **Á. Robles:** Definition, Methodology, Formal analysis, Investigation, Validation, Writing – review & editing, Supervision, Management and coordination responsibility for the research activity planning and execution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2023.110267.

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Life cycle assessment of AnMBR technology for urban wastewater treatment: A case study based on a demo-scale AnMBR system

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ABSTRACT

This study aims at assessing the environmental performance of a projected full-scale anaerobic membrane bioreactor (AnMBR) treating urban wastewater (UWW) at ambient temperature. To this aim, data from an AnMBR demonstration plant equipped with commercially available equipment, including industrial hollow fiber and degassing membranes, was used for projecting a full-scale facility. The use of real operation data allows to obtain robust results that contribute to improve the knowledge of the environmental performance of this technology, pointing out its strengths and the challenges that still need to be addressed. Life cycle assessment (LCA) was applied by means of Ecoinvent data base and ReCiPe2016 methodology considering 1 kg of removed COD as functional unit. Additionally, sensitivity and uncertainty analysis were conducted. Energy balance showed AnMBR performing as energy producer (net energy surplus up to −0.688 kWh·kg COD_{rem}^{-1}) and carbon sink (emissions credit up to 0.223 $kgCO_{2eq}$·$kgCOD_{rem}^{-1}$). Results also showed energy recovery, heavy metals in sludge, dissolved methane in the effluent, and effluent nutrient content as the most important aspects affecting LCA outcome. Construction phase affected some impact categories significantly (e.g., 51–71% in mineral resource scarcity, 18–27% in fossil resource scarcity, 21–28% in water consumption), therefore its exclusion should be carefully evaluated. CHP efficiency, dissolved methane recovery, filtration productivity, membrane scouring, reactor mixing, HRT and SRT appeared most influencing parameters. Finally, actions leading to increase the recovery and valorization of dissolved methane and/or of nutrients through, for instance, fertigation, improve the environmental performance of AnMBR for UWW treatment.

1. Introduction

The study of the environmental performance of water management has been consolidated as a key element to promote innovation and sustainability within this sector [5]. In this context, Life Cycle Assessment (LCA) has been established as a suitable instrument for evaluating the environmental impacts of the water infrastructure (catchment, purification, distribution, sewer systems, wastewater treatment, reclamation, discharge, etc.), providing valuable information on systems design, operation of facilities and policy development [1].

Despite the unquestionable benefits of wastewater treatment (WWT), the conventional processes applied to obtain adequate effluent quality and guarantee the protection of the environment and public health are based on energy-intensive and chemical-dependent technologies [21] with a considerable associated impact. Furthermore, increased pressure on the environment is foreseen since wastewater production and the associated treatment are expected to increase [28] while the authorities are tightening the regulations on waste [4]. Therefore, sewage management strategies should focus on saving energy and chemicals. The progress towards sustainability in the wastewater management sector should also include shifting from the conventional pollutant-removal concept to the resource-recovery paradigm, which offers advantages in reclaimed water, energy and the valuable materials embedded in wastewater, in line with the principles of the Circular Economy [33].

Anaerobic membrane bioreactors (AnMBR) have been proposed as an alternative technology to conventional treatments based on aerobic processes [14,32] and could overcome some of the drawbacks

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mentioned above and contribute to enhance sewage treatment sustainability. While anaerobic microorganisms produce methane from organic matter, which acts as an energy carrier and improves the energy balance [18], they also provide a nutrient-rich effluent that can be fed to nutrient recovery processes (struvite crystallization, membrane contactors, photobioreactors…) or directly reused through fertigation, i.e., direct agriculture reuse of the effluent [16]. On the other hand, membrane technology ensures reduced downstream solids and pathogen concentrations making reclaimed water attractive for other uses (irrigation, aquifer recharge, etc.), while the compactness of membrane modules reduces a facility's footprint and thus favors the retrofitting of existing treatment plants operating in a limited space [39]. Membrane units also decouple sludge retention time (SRT) and hydraulic retention time (HRT), enabling AnMBRs to operate at low/middle temperatures since the membranes avoid microorganisms wash-out from the reactor [38].

To the best of author's knowledge, few LCAs have been carried out on AnMBRs for urban wastewater treatment, while the comparison of published studies is somewhat challenging due to the diversity of methods (ReCiPe, CML-IA, TRACI, etc.), functional units (m^3 of treated water, kg PO_4^{-3} removed, etc.) and the initial assumptions used. For example, the facility construction phase has traditionally been excluded from the LCA since its impact was considered negligible compared to those related to the operating phase [30]. However, Morera et al. [23] more recently concluded that the environmental loads of the construction phase have been underestimated due to a lack of detail when drawing up the inventories. These authors found a significant impact of the construction phase (over 5%) on most of the categories evaluated, and especially in the case of metal depletion (63%) using the ReCiPe method. Efforts are now being made to enhance LCA studies on wastewater treatment aimed at clarifying these uncertainties and improving their quality and comparability [6].

A wide implementation of AnMBR for UWW is still hampered by the phenomenon of membrane fouling, whose management may imply an increase in membrane surface requirements and energy and chemicals consumption, thus affecting the economic and environmental viability of this technology. Fouling is governed by different interrelated factors, among which the concentration and nature of the mixed liquor solids, membrane scouring intensity, SRT or operating temperature can be mentioned [19]. In this sense, high temperatures diminish mixed liquor viscosity, which boosts filtration but also could make microorganism to release extracellular polymeric substances (EPS) and/or Soluble microbial products (SMP), which increments fouling [20,31]. Fouling-prevention strategies based on gas sparging are effective, but they increase the filtration energy demand. On the other hand, increased SRT might enhance EPS and SMP consumption, although it implies increasing the solids concentration, which hinders the filtration process.

Energy demand, chemical consumption, nutrient release and the methane dissolved in the effluent have been identified as key factors in driving AnMBR environmental performance and determine its comparison with other technologies. Pretel et al. [26] estimated the environmental impact of different aerobic-based wastewater treatment plants (WWTP) (conventional activated sludge-CAS; aerobic membrane bioreactors-AeMBR) and AnMBR both alone and combined with post-treatment for nutrient removal. The results showed that AnMBR alone presented the lowest environmental impact within all the categories evaluated except eutrophication, although eutrophication loads could also be reduced if AnMBR is combined with a CAS-based post-treatment or nutrient recovery processes. Smith et al. [38] also compared AnMBR to various aerobic-based WWTPs (high rate activated sludge with anaerobic digestion-HRAS+AD; CAS and anaerobic digestion-CAS+AD; AeMBR and anaerobic digestion-AeMBR+AD). The findings showed that the methane dissolved in AnMBR effluent accounted for 75% of the global warming impact, which indicates that dissolved methane recovery is a key point to be addressed. The results also demonstrated improved AnMBR environmental performance when treating high-loaded wastewater, being comparable to HRAS+AD and lower than CAS+AD in some impact categories. Cashman et al. [3] compared decentralized AeMBR and AnMBR for different population densities and treatment flows and found that, for all the scales evaluated, AnMBR at 20 °C presented the highest net energy benefits and concluded that AnMBRs operated at lower temperatures in warm climates were a promising technology. Xu et al. [40] found that AnMBR had a lower environmental impact than AeMBR when the system boundary was expanded to include effluent reuse and concomitant nutrient recovery through crop uptake. Without this boundary expansion and in disagreement with Pretel et al. [26], AnMBR showed worse performance but, as commented in regard to Smith et al. [38], this contradiction may be due to differences in influent quality: Pretel et al. [26] evaluated medium/high-loaded wastewater, while Xu et al. [40] considered a low-loaded influent. Finally, Pretel et al. [27] assessed the economic and environmental performance of an AnMBR treating UWW and food waste. The results showed that codigestion allowed a net energy production, improved the economic sustainability (savings up to €0.023 per m^3 of treated water) and reduced overall environmental impacts of AnMBR technology.

The main goal of this study was thus to apply the LCA method to evaluate the environmental performance of a projected full-scale AnMBR system (treatment capacity of 16000 $m^3 \cdot d^{-1}$) for urban wastewater (UWW) treatment. Unlike other studies, mainly based on pilot plants and simulation data, the results were obtained using real data from an AnMBR demonstration plant treating UWW under different operating conditions (see Jiménez-Benítez et al. [14]; Robles et al. [32], (2020b)). Due to the dimensions and equipment of this facility, the results can be regarded as robust information for scaling up AnMBR technology for industrial-scale urban WWTPs.

2. Materials and methods

2.1. AnMBR demo-plant description

The demonstration plant mainly consisted of a 40 m^3 anaerobic reactor (AnR) (34.4 m^3 working volume) connected to three 0.8 m^3 membrane tanks (MT-X) of 0.7 m^3 working volume. Each MT was equipped with an ultrafiltration (UF) hollow-fiber membrane module (PURON® PSH41, 0.03-μm pore size, filtration area of 41 m^2). The plant also included a degassing membranes (DM) system to recover the methane dissolved in the effluent (PermSelect® Silicone Hollow Fiber Membrane; filtration area of 2.1 m^2). The demonstration plant also included different on-line sensors and measurement equipment to monitor and register data on flow rates, liquid levels, pressure (transmembrane, inlet and outlet for blowers, and gas), temperature and methane production and recovery. For further details, see Robles et al. (2020b) and e-supplementary materials.

2.2. AnMBR demo-plant operation

Effluent from the pre-treatment unit of the "Alcázar de San Juan" WWTP (screening and sand removal) was pumped into the anaerobic reactor after further pre-treatment in the rotofilter (RF) and homogenization in the equalization tank (ET). The mixed liquor was continuously recycled through the MTs, where the final effluent was obtained by vacuum filtration. In order to maintain appropriate mixing conditions in the anaerobic reactor, part of the biogas produced was constantly recirculated from the headspace to the bottom of the reactor through coarse bubble diffusers, while another fraction of the biogas produced was pumped to the bottom of the MTs for membrane scouring. For further details, see Robles et al. (2020b).

The demo-plant was operated during 580 days under different operating conditions from which five operating periods have been selected to design and simulate the performance of a projected full-scale AnMBR system to treat the Alcázar de San Juan WWTP flow rate (16000 $m^3 \cdot d^{-1}$). The operating periods have been named by combining the

Table 1
Materials and processes selected for the Life Cycle Inventory.

Construction materials, kg·kg COD_{rem}^{-1}	Concrete *(1 m3 Concrete, normal {RoW}| market for | APOS, S (Ecoinvent 3))*	
	Chromium steel *(1 kg Steel, chromium steel 18/8 {GLO}| market for | APOS, S (Ecoinvent 3))*	
	Cast iron *(1 kg Cast iron {GLO}| market for | APOS, S (Ecoinvent 3))*	
	Polyester resin *(1 kg Polyester resin, unsaturated {GLO}| market for | APOS, S (Ecoinvent 3))*	
	Polypropylene *(1 kg Polypropylene, granulate {GLO}| market for | APOS, S (Ecoinvent 3))*	
	PVC *(1 kg PVC pipe E (Industry data 2.0))*	
	Silicone *(1 kg Silicone product {GLO}| market for | APOS, S (Ecoinvent 3))*	
	Transport *(1 tkm Transport, freight, lorry 16–32 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3)*	
Energy consumption, kWh·kg COD_{rem}^{-1}, *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Energy avoided (energy recovery from methane), kWh·kg COD_{rem}^{-1} *(1 kWh Electricity, medium voltage {ES}| market for | APOS, S (Ecoinvent 3))*		
Reagent consumption, kg·kg COD_{rem}^{-1}	Polyelectrolyte *(1 kg Polyacrylamide {GLO}| market for | APOS, S (Ecoinvent 3))*	
	NaOCl *(1 kg Sodium hypochlorite, without water, in 15% solution state {GLO}| market for | APOS, S (Ecoinvent 3))*	
	Citric acid *(1 kg Citric acid {GLO}| market for | APOS, S (Ecoinvent 3))*	
	Transport *(1 tkm Transport, freight, lorry 3.5–7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
Discharge to air, kg·kg COD_{rem}^{-1}	Methane CH_4 *(AnMBR, 1 kg emissions of CH4 to air)*	
Discharge to water, kg·kg COD_{rem}^{-1}	Ammonium NH_4^+ *(AnMBR, 1 kg emissions NH_4^+ to water)*	
	Total phosphorous, PO_4^{-3} *(AnMBR, 1 kg emissions PO_4^{-3} to water)*	
Discharge to soil, kg·kg COD_{rem}^{-1}	Disposal to agriculture *(1 kg Solid manure loading and spreading, by hydraulic loader and spreader {GLO}| market for | APOS, S (Ecoinvent 3))*	N to soil *(AnMBR, 1 kg emissions N to soil through fertigation)*
		PO_4^{-3} to soil *(AnMBR, 1 kg emissions PO_4^{-3} to soil through fertigation)*
		N-based fertilizer *(1 kg Nitrogen fertiliser, as N {GLO}| market for | APOS, S)*
		P-based fertilizer *(1 kg Phosphate fertilizer, as P2O5 {GLO}| market for | APOS, S (Ecoinvent 3))*
		Cd to soil *(AnMBR, 1 kg emissions Cd to soil through sludge spreading)*
		Co to soil *(AnMBR, 1 kg emissions Co to soil through sludge spreading)*
		Cr III to soil *(AnMBR, 1 kg emissions Cr^{+3} to soil through sludge spreading)*
		Cu to soil *(AnMBR, 1 kg emissions Cu to soil through sludge spreading)*
		Zn to soil *(AnMBR, 1 kg emissions Zn to soil through sludge spreading)*
		Ni to soil *(AnMBR, 1 kg emissions Ni to soil through sludge spreading)*
		Pb to soil *(AnMBR, 1 kg emissions Pb to soil through sludge spreading)*
		N_2O to air *(AnMBR, 1 kg emissions N_2O to air through fertigation)*
	Disposal to landfill *(1 kg Municipal solid waste {RoW}| treatment of, sanitary landfill | APOS, S (Ecoinvent 3))*	
	Disposal to incineration *(1 kg Digester sludge {GLO}| treatment of digester sludge, municipal incineration | APOS, S (Ecoinvent 3))*	
	Transport *(1 tkm Transport, freight, lorry 3.5–7.5 metric ton, EURO6 {GLO}| market for | APOS, S (Ecoinvent 3))*	
Demolition and materials recycling, kg·kg COD_{rem}^{-1}	Steel and Iron *(1 kg Steel and iron (waste treatment) {GLO}| recycling of steel and iron | APOS, S (Ecoinvent 3))*	
	Concrete demolition *(1 kg Waste reinforced concrete {Europe without Switzerland} | treatment of waste reinforced concrete, recycling | APOS, S (Ecoinvent 3))*	
	Polyethylene treatment *(1 kg Waste polyethylene/ polypropylene product {Europe without Switzerland} | treatment of waste polyethylene/polypropylene product, collection for final disposal | APOS, S (Ecoinvent 3)*	
	PVC recycling *(1 kg PVC (waste treatment) {GLO}| recycling of PVC | APOS, S (Ecoinvent 3))*	

letter "H" to indicate the HRT followed by its average value (subindex) and the letter "T" to indicate the temperature followed by its average value (subindex): i) $H_{41}T_{27}$; ii) $H_{25}T_{24}$; iii) $H_{26}T_{19}$; iv) $H_{26}T_{27}$; and v) $H_{41}T_{18}$. Complete characterization of the operating periods evaluated in this study are provided in e-supplementary materials (see Table S1).

The influent wastewater was characterized by a high COD (755 ± 224–1403 ± 532 mg·L^{-1}) and sulfate (125.4 ± 47.6–172.2 ± 28.5 mg SO_4^{-2}-S·L^{-1}) concentrations due to dairy- and wine-industry contributions. Despite the high influent COD concentration, COD removal efficiencies of 90–92% were achieved with concentrations below the European discharge limits (125 mg·L^{-1}). Further details of the biological performance of the demo-plant can be found in Robles et al. [32].

Regarding the filtration process, membrane modules were operated at 20 °C-standardized gross transmembrane fluxes ($J_{20\ gross}$) between 15 and 21 L·m^{-2}·h^{-1} and specific gas demands per volume of permeate (SGD_P) between 6 and 14 Nm^3_{biogas}·$m^{-3}_{permeate}$. These conditions resulted in fouling rates of between 0.3 and 4.7 mbar·d^{-1}. Details on filtration process of the demo-plant can be found in Jiménez-Benítez et al. [15].

2.3. System boundary and functional unit

According to ISO 14040:2006, the LCA framework applied in this study was subdivided into four stages: (1) goal and scope definition phase, (2) inventory analysis phase, (3) impact assessment phase, and (4) interpretation phase. The life cycle inventories (LCI) of individual materials and processes were compiled using the *Ecoinvent 3.0* and *Industry data 2.0* databases accessed via SimaPro (PRé Consultants; The Netherlands) (see Table 1). A functional unit of 1 kg of removed COD (kg COD_{rem}) was selected. A total facility life span of 20 years was set.

The following system boundaries were considered:

- The flow diagram considered in this LCA begins with the rotofilter treatment step. Previous screening and sand removal systems are not included since they are common to most treatment systems like CAS.
- Construction (anaerobic reactors, membrane tanks, pipes and equipment), facility operation, and demolition phases (materials disposal and recycling) were considered. Transport of materials, chemicals and sludge (assuming a distance of 20 km for construction and operation and 30 km for demolition) were also incorporated.
- The building work and infrastructure life cycles were set to 20 years [6]. Blower and pump lifespans were estimated from the total working hours provided by the manufacturers (50000 and 75000 h, respectively). A 10-year lifetime was set on other equipment (diffuser, reciprocating engines). UF membrane lifetime was estimated according to the total chlorine contact specified by the manufacturer (500000 h). A 10-year lifespan period was set to DM.
- Neither reuse of nutrients in the effluent for fertigation purposes nor nutrient recovery or removal by other technologies were considered.
- The waste sludge fate was set as follows: 90% to agricultural application, 10% to landfill (in line with data published by the Spanish Ministry of Ecological Transition and Demographic Challenge [22]). Nitrogen and phosphorus sludge content was considered available

Table 2
Relative LCA results for midpoint impact categories in % (The largest impacts are indicated in red and the minor ones in green. Negative values indicate environmental benefits).

Impact category	$H_{41}T_{27}$	$H_{25}T_{24}$	$H_{26}T_{19}$	$H_{26}T_{27}$	$H_{41}T_{18}$
Global warming	-100	-45	1	9	-22
Stratospheric ozone depletion	-100	-64	-92	-65	-63
Ionizing radiation	-100	-61	-43	-39	-70
Ozone formation, human health	-100	-61	-53	-42	-65
Fine particulate matter formation	-100	-62	-52	-43	-66
Ozone formation, terrestrial ecosystems	-100	-61	-53	-42	-65
Terrestrial acidification	-100	-62	-55	-45	-66
Freshwater eutrophication	76	81	96	100	86
Marine eutrophication	51	54	78	100	49
Terrestrial ecotoxicity	-100	-65	-74	-53	-56
Freshwater ecotoxicity	69	62	96	100	87
Marine ecotoxicity (kg 1,4-DCB)	69	62	96	100	86
Human carcinogenic toxicity	65	58	94	100	87
Human non-carcinogenic toxicity	69	62	96	100	87
Land use	-100	-63	-81	-61	-66
Mineral resource scarcity	-100	-77	-90	-67	-45
Fossil resource scarcity	-100	-61	-50	-39	-64
Water consumption	-100	-61	-72	-60	-70

for crops, therefore equivalent avoidance of mineral fertilizer production was contemplated.
- N_2O emissions due to sludge agricultural application were included as 1.18% of total nitrogen content, in accordance with Doka [8].
- Disinfection treatment step was considered implicit in UF performance as per Ferreiro et al. [10], Foglia et al. [11] and Yang et al. [41], among others.
- According to the IPCC Guidelines for National Greenhouse Gas Inventories [9], biogenic CO_2 emissions from wastewater treatment were not taken into account to calculate greenhouse gases (GHG).
- According to Kidd et al. [17], sludge heavy metal content from municipal wastewater treatment was considered, i.e., the industrial contribution is considered to be low (see e-supplementary materials). Displacement of heavy metals in the avoided mineral fertilizers was

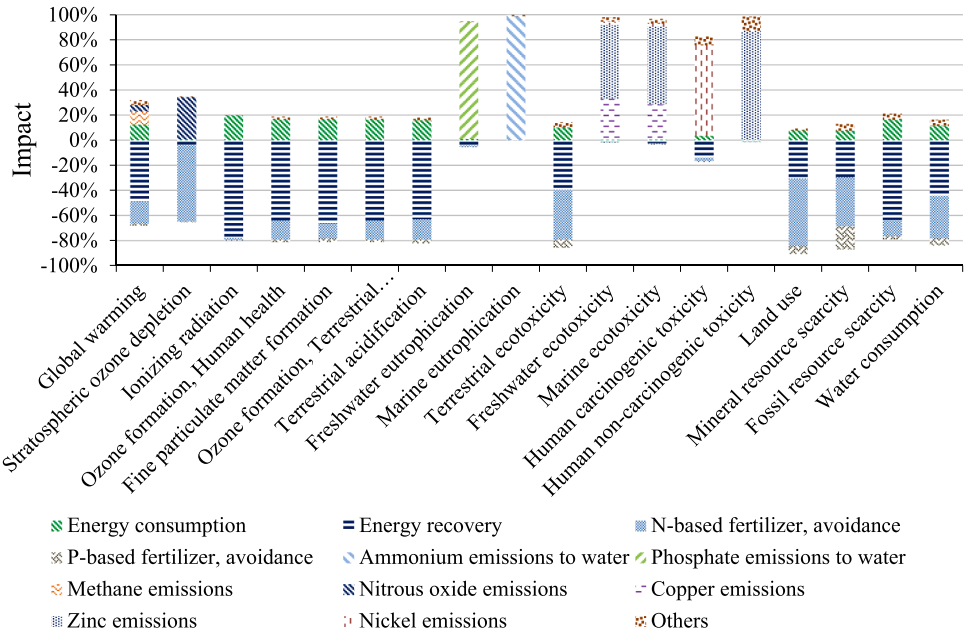

Fig. 1. Example of allocation of loads per impact category for $H_{41}T_{27}$.

not taken into account, due to a lack of information about their content in commercial fertilizer.

- The energy balance in each operating period was calculated as shown in Jiménez-Benítez et al. [15]. The material impact of DM and reciprocating engines for energy recovery were thus included. Moreover, it is considered that the energy recovered is used in its entirety, i.e., if the energy recovered exceeds the plant requirement, it is considered that the energy surplus is discharged into the electrical grid and is consumed outside the facility.
- Dissolved methane recovery by means of DM was included. Recovery percentages in the 80–84% range was considered based on the environmental optimum performance obtained by Sanchis-Perucho et al. [37].
- Fugitive methane emissions due to the stripping of non-captured dissolved methane in the effluent to the atmosphere were considered. These emissions have been estimated as the difference between the methane content in the effluent and that recovered in accordance with the previous point.
- Potential biogas treatments to adapt its quality to CHP requirements (e.g., removal of hydrogen sulfide or siloxanes) have been excluded from the scope of the analysis due to the lack of information on the concentration of these compounds.
- Emissions to air of given compounds (e.g., CO, SO_2, NO_2, non-methane volatile organic compounds) resulting from biogas combustion (through CHP) were excluded due to a lack of information.

LCA was based on the ReCiPe 2016 method and a hierarchist approach. A general discussion based on the 18 midpoint impact categories provided by ReCiPe 2016 method is offered. Moreover, global warming, mineral and fossil resource scarcity, freshwater eutrophication, marine eutrophication and water consumption were selected in this work as key process indicators and a deeper analysis of them was conducted. The endpoint areas (damage to human health; damage to ecosystems; and damage to resource availability) proposed in ReCiPe 2016 methodology were also considered in this study. The results of all midpoint and end-point impact categories can be found in e-supplementary materials.

2.4. Global sensitivity and uncertainty analysis

Sensitivity analysis (SA) and uncertainty analysis (UA) were conducted to identify influential parameters on model outputs and related uncertainty. 2000 model runs (Monte Carlo method) were performed for both SA and UA. In both cases, the Latin hypercube sampling (LHS) method was used to construct the sampling matrix, applying a variability of 5% for the membrane and equipment weight quoted by manufacturers and 10% for the rest of parameters considered (further details can be found in e-supplementary materials). The SA was carried out by means of the Standardized Regression Coefficients (SRC) method applied to all variables and constants considered for modelling the projected full-scale AnMBR system. An absolute value of 0.1 was selected as the threshold value for the standardized regression slope (b_i) for identifying influential input model factors. The UA was conducted to evaluate the propagation of uncertainty in the results, which were assessed by means of i) the 5th and 95th percentiles and ii) the empirical cumulative distribution function (eCDF). The above-mentioned midpoint impact categories (global warming, mineral and fossil resource scarcity, freshwater eutrophication, marine eutrophication and water consumption) were used as model outputs for both SA and UA.

3. Results and discussion

3.1. Energy balance

First of all, it should be noted that in a previous life cycle costing (LCC) study carried out for the same full-scale AnMBR plant [14], the energy balance was conducted considering the economic optimum for the operation of the DM unit. This optimum, calculated based on Sanchis-Perucho et al. [37], was estimated in the 47–55% range of dissolved methane recovery. Sanchis-Perucho et al. [37] also included the estimation of the environmental optimum, which raises the % of dissolved methane recovered to the 80–84% range. The present study, focused on the environmental assessment of the AnMBR technology, has been carried out based on this last recovery percentage range. However, differences in energy balance results are low and it is possible to consult both cases in the e-supplementary materials (Table S2 and Table S3).

Since the energy balance has a crucial influence on many of the impact categories evaluated, main results are summarized in this

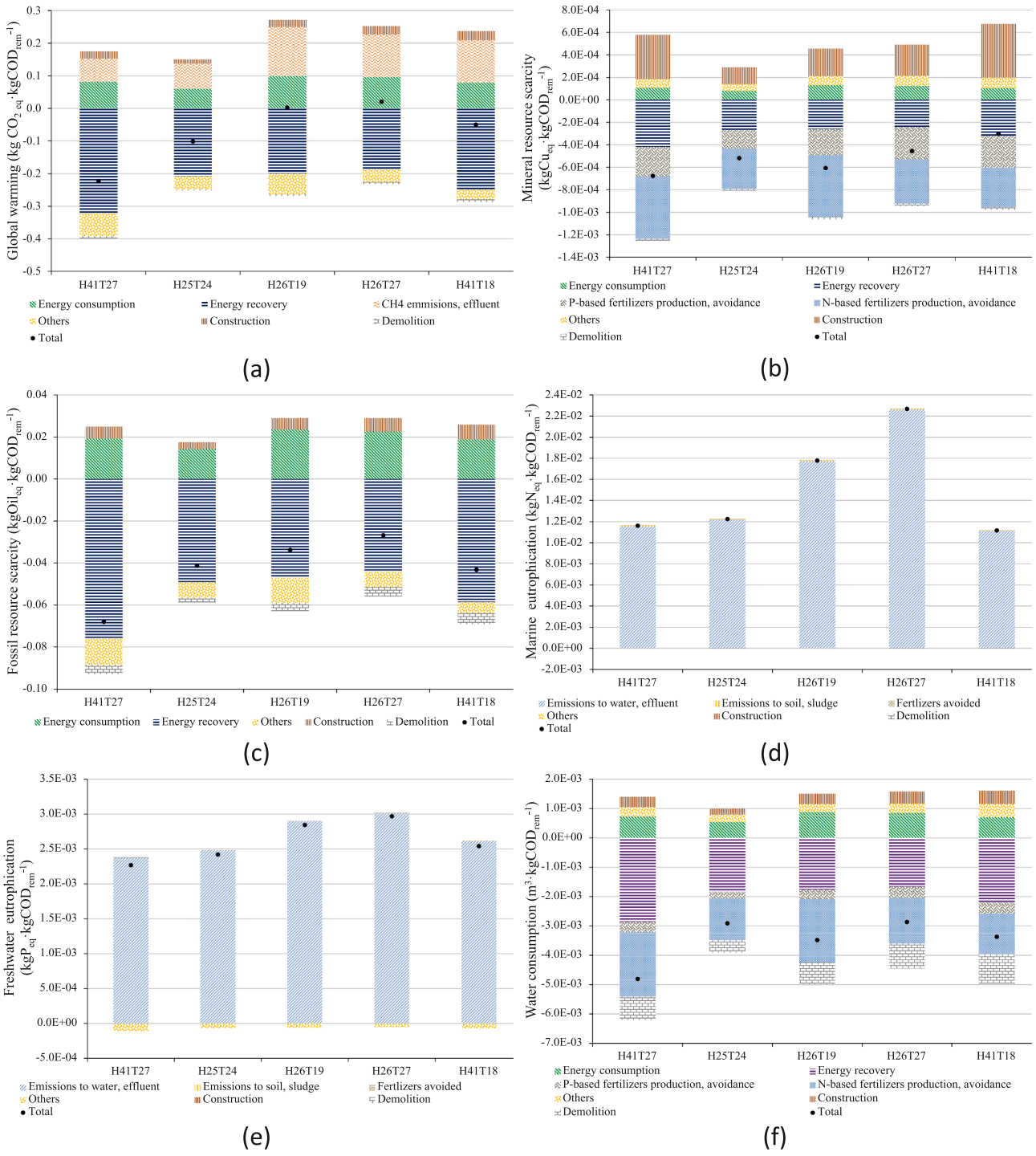

Fig. 2. Environmental impact through: (a) global warming potential, (b) mineral resource scarcity, (c) fossil resource scarcity, (d) marine eutrophication, (e) freshwater eutrophication, and (f) water consumption. Figures include allocations for operating phase jointly with impacts for construction phase and demolition phase.

section. All the evaluated operating periods performed as energy producers, especially $H_{41}T_{27}$, $H_{25}T_{24}$ and $H_{41}T_{18}$ (net energy demands (NED) of − 0.668, − 0.409 and − 0.468 kWh·kg COD_{rem}^{-1}, respectively). $H_{26}T_{27}$ showed the less favorable NED (−0.254 kWh·kg COD_{rem}^{-1}), while $H_{26}T_{19}$ obtained an intermediate value (−0.277 kWh·kg COD_{rem}^{-1}). These results were mainly associated with HRT, which affects reactor mixing requirements; membrane scouring; filtration strategy to minimize membrane fouling; reactor temperature; organic loading rate (OLR) and the COD:SO_4^{-2} ratio, which influences methane production and thus recovered energy. AnMBR, thus, appears as a potential alternative to conventional treatments at moderate temperatures for medium-high strength urban wastewater. For further details, see Table S2 in e-supplementary materials.

3.2. Life cycle assessment

3.2.1. Midpoint categories

Table 2 shows the LCA results for all the midpoint categories included in ReCiPe 2016 for the five evaluated operating periods. Overall, $H_{41}T_{27}$ resulted in the lowest overall environmental impact of

the evaluated operating periods in almost all categories (i.e., global warming, stratospheric ozone depletion, ionizing radiation, ozone formation-human health, fine particulate matter formation, ozone formation-terrestrial ecosystem, terrestrial acidification, freshwater eutrophication, terrestrial ecotoxicity, land use, mineral resource scarcity, fossil resource scarcity and water consumption). However, $H_{41}T_{27}$ also was found to have a significant impact among the studied operating periods for freshwater and marine ecotoxicity, and human carcinogenic and non-carcinogenic toxicity. Table 2 also shows that, in general, the worst environmental results were reached in $H_{26}T_{27}$.

Fig. 1 shows the effect of the most influential processes on each impact category for $H_{41}T_{27}$ (similar information related to the rest of the operating periods is provided in e-supplementary material). The main results given in Fig. 1 generalized to all the operating periods can be summarized as follows:

- **Energy recovery:** high organic loading rate jointly with methane production allowed a higher energy recovery in $H_{41}T_{27}$, $H_{25}T_{24}$ and $H_{41}T_{18}$. Improved total energy recovery offset energy consumption, thus reducing the environmental impact of the following categories: global warming, ionizing radiation, ozone formation (human health and terrestrial ecosystem), fine particulate matter formation, terrestrial acidification and ecotoxicity, mineral and fossil resources scarcities, land use and water consumption.
- **Nutrient recovery:** direct reuse of phosphorus and especially the nitrogen embedded in sludge for fertilizing purposes contributed to reducing the environmental loads associated with the avoidance of mineral fertilizer production. This influence was particularly relevant for stratospheric ozone depletion, terrestrial ecotoxicity, land use, mineral resource scarcity and water consumption. Other categories (e.g., global warming, ozone formation (human health and terrestrial ecosystem), fine particulate matter formation, terrestrial acidification, and fossil resource scarcity also reduced their impact due to nutrient recovery. It is important to highlight that Fig. 1 foresees that the environmental benefits of nitrogen recovery in agriculture through sludge spreading would compensate for the drawbacks related to the N_2O emissions derived to air (for the categories of global warming and stratospheric ozone depletion), resulting in positive net balances due to this agricultural practice.
- **Release to the environment of the effluent nutrient content:** the concentration of nutrients in the effluent is the factor that determines the eutrophication potential of the technology. However, since functional unit is defined as 1 kg of removed COD, the environmental load for each operating period was also affected by the influent COD: N and COD:P ratios and the organic matter removal efficiency. As a way of example, $H_{41}T_{27}$ and $H_{26}T_{19}$ obtained a very similar total nitrogen concentrations in the effluent (47.9 ± 6.2 and 47.3 ± 4.3 mg $N{\cdot}L^{-1}$, respectively) but COD removal in $H_{41}T_{27}$ (equivalent to 19138 $kg{\cdot}d^{-1}$ for 16000 $m^3{\cdot}d^{-1}$ of treatment flow) was significantly higher than in $H_{26}T_{19}$ (equivalent to 12428 $kg{\cdot}d^{-1}$). Therefore, the latter showed higher marine eutrophication. The eutrophication results demonstrated that nutrient recovery in a downstream post-treatment step (e.g., microalgae cultivation, ion exchange, membrane contactors, etc.) or by fertigation (i.e. nutrient-loaded water reclamation for agricultural purposes) should be considered in order to further enhance the environmental feasibility of AnMBR for UWW treatment [16] and fully leverage resource recovery from the AnMBR effluent. Indeed, reduced environmental loads in other categories influenced by avoiding mineral fertilizer production (e.g., global warming, stratospheric ozone depletion, land use, etc.) could be expected if these nutrients are recovered.
- **Direct GHG emissions due to dissolved methane:** fugitive methane released from the effluent had a significant impact on global warming potential, so that the capture of this gas is a key challenge to be addressed for AnMBR environmental improvement. As way of example, Fig. 1 shows that 40% of the global warming load in $H_{41}T_{27}$ was associated with dissolved methane emissions. Increasing its recovery would reduce this impact while improving the NED.
- **Heavy metals:** the heavy metal content considered in the sludge (see e-supplementary materials) mainly affected freshwater ecotoxicity, marine ecotoxicity, and human toxicity, both carcinogenic and non-carcinogenic. Zinc showed a significant impact on human non-carcinogenic toxicity and freshwater and marine ecotoxicity. The two latter categories were also affected by the copper concentration. Nickel appeared as the main metal influencing human carcinogenic toxicity. Fig. 1 reveals that the benefits of recovering energy and nutrients did not offset the impact of metals associated with human carcinogenic toxicity and the freshwater and marine ecotoxicity impact categories. $H_{26}T_{27}$ obtained the worst performance within these categories due to the combined effect of its moderate dry sludge production (847 $t{\cdot}year^{-1}$) and its lower COD removal performance (10815 $kg{\cdot}d^{-1}$), while $H_{25}T_{24}$ produced the lowest environmental loads for these categories due to its lower dry sludge production (632 $t{\cdot}year^{-1}$) and higher COD removal efficiency (20631 $kg{\cdot}d^{-1}$).

As mentioned above, to better analyze the LCA results from the full-scale AnMBR, the following categories were selected as key indicators of the process: global warming potential, mineral and fossil resource scarcity, freshwater and marine eutrophication, and water consumption.

3.2.1.1. Global warming potential. Fig. 2a shows the global warming potential obtained for the evaluated operating periods, considering construction, operating, and demolition phases. The operating phase was the main contributor to this impact category, representing 87–92% of the direct environmental impact (without considering emission avoidance due to energy recovery and others, e.g., avoidance of mineral fertilizer production) for all the evaluated operating periods. The construction phase weight represented between 8% and 13% of the total direct impact, values slightly higher than those reported by Morera et al. [23] (5–10%) for a large CAS WWTP using 1 m^3 of treated water as the functional unit. The main contributors to construction phase are concrete applications (44–47%) followed by PVC of pipes (32–34%).

Most of the direct environmental load in this category was related to fugitive methane emissions due to downstream stripping of dissolved methane in the effluent (40–55%). This percentage is lower to that obtained by Smith et al. [38]: 75% using as functional unit the treatment of 18,950 $m^3{\cdot}d^{-1}$). Pretel et al. [25] evaluated the environmental performance of an AnMBR for urban wastewater treatment and also identified methane emissions as a key point. Reduction of AnMBR carbon footprint therefore necessarily involves improving dissolved methane recovery. In turn, recovering the dissolved methane to produce energy would improve the energy balance. $H_{41}T_{27}$, $H_{25}T_{24}$ and $H_{41}T_{18}$ acted as a carbon sinks (−0.223, −0.101 and −0.050 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$, respectively) when considering the dissolved methane capture efficiencies applied in this study (80–84%), while $H_{26}T_{19}$ and $H_{26}T_{27}$ showed almost carbon neutral performance: 0.003 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$ and 0.020 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$. In this regard, Rodriguez-Garcia et al. [34] evaluated the operation of different WWTPs, among others, based on conventional activated sludge obtaining values between 0.206 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$ and 0.551 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$ for facilities without nutrient removal processes and discharging to non-sensitive areas. In the case of the more restrictive situation of conventional activated sludge with nutrient removal and discharging to sensitive areas, the same authors obtained emission values between 0.413 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$ and 0.688 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$. It can thus be concluded that AnMBR can be regarded as an alternative treatment to reduce GHG emissions in sewage management when dissolved methane in the effluent is captured.

Enhanced dissolved methane capture efficiency is possible from a technical perspective by increasing the DM filtration area, although this would entail an increase in investment costs, which would affect

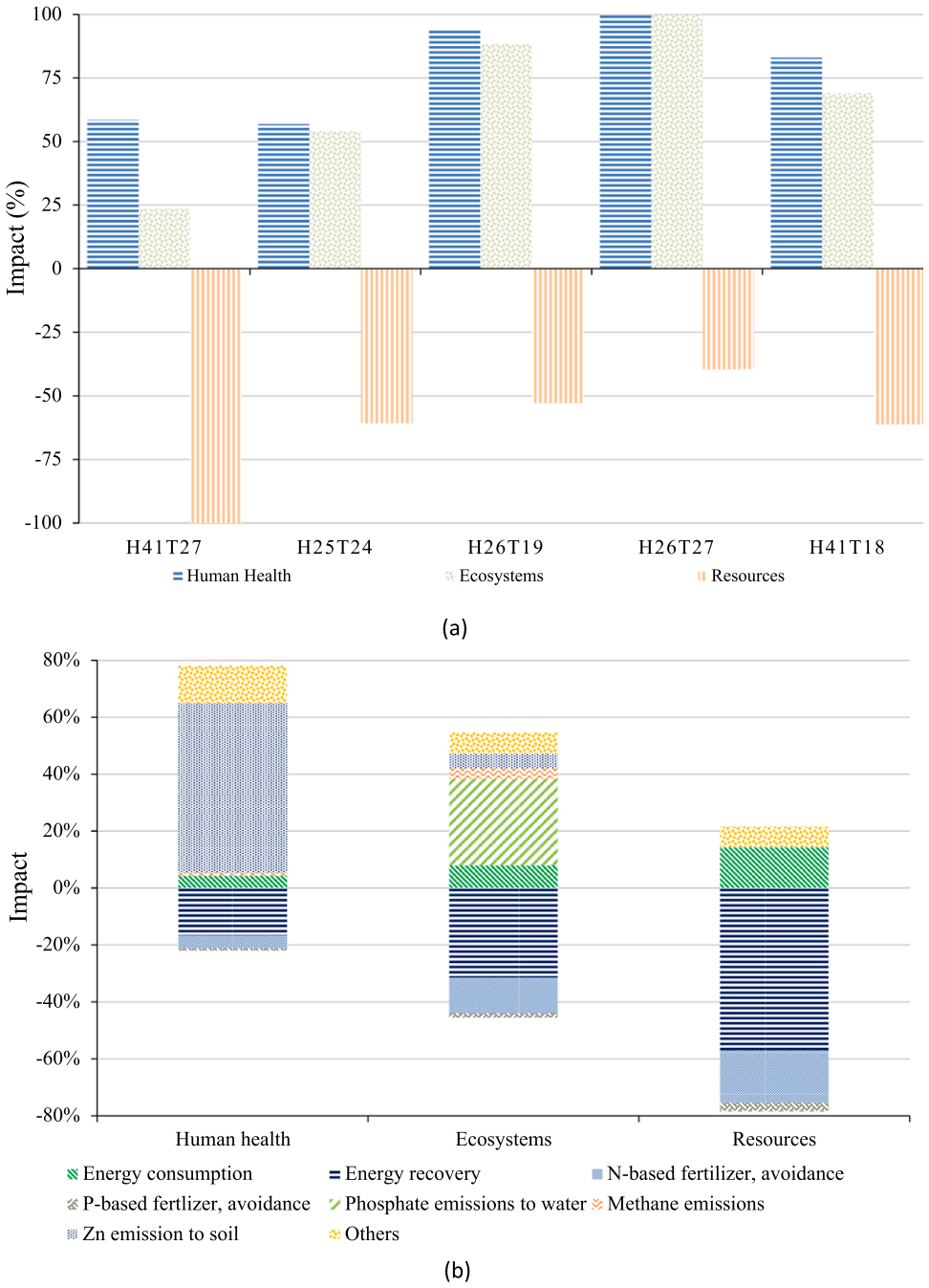

Fig. 3. (a) LCA results for endpoint impact categories. (b) Example of allocation of loads per impact category for $H_{41}T_{27}$.

AnMBR's economic feasibility [36]. Moreover, increasing DM filtration area would increment the environmental impacts associated with the use of DM materials. However, results showed that environmental loads of construction phase were significantly lower than operation phase. Therefore, it is expected that the contribution to the environmental impact associated with an increase in the DM membrane surface would be low. Finally, DM can be combined with additional technologies to prevent dissolved methane release. In this sense, Sanchis-Perucho et al. [35] reached up to 100% of methane capture by adding a degassing tower after DM with low investment cost.

On the other hand, it is important to highlight that global warming potential results strongly depended on the energy mix considered. The larger the fossil fuel burden, the higher the CO_{2eq} emissions associated with energy consumption. For example, according to the International Energy Agency [13], in 2018 the main energy sources in Norway, France and Poland were hydro (≈96%), nuclear (≈72%) and coal (≈78%), respectively. Consequently, the global warming potential impact factor in the Ecoinvent database reflects this diversity of technologies and establishes 0.0289 kg $CO_{2eq}{\cdot}kWh^{-1}$ for the Norwegian mix, 0.0533 kg $CO_{2eq}{\cdot}kWh^{-1}$ for the French mix and 1.01 kg $CO_{2eq}{\cdot}kWh^{-1}$ for the Polish one. The appropriate global warming potential comparisons between the WWT technologies should thus take location into consideration [6].

3.2.1.2. Mineral and fossil resource scarcity. Mineral resource scarcity was mainly influenced by NED and nutrient recovery through sludge spreading due to avoiding production of P-based and especially N-based fertilizers (see Fig. 2b). Hence, there is still a great potential for reducing the environmental loads in this category by reducing resource depletion if effluent nutrient content is also recovered and/or reused for agriculture. $H_{41}T_{27}$ showed the lowest environmental impact in this category

among studied operating periods due to the high energy recovery and the combination of high total nitrogen and phosphorus concentration in the mixed liquor (505.8 ± 139.4 mg $N{\cdot}L^{-1}$ and 96.8 ± 12.3 mg $P{\cdot}L^{-1}$), the highest dry sludge production (1049 $t{\cdot}year^{-1}$) and its good COD removal performance (19138 $kg{\cdot}d^{-1}$). It should also be mentioned that the construction phase composed between 51% and 71% of the direct mineral resource scarcity life cycle impact, when depletion offsets due to energy, nutrient recovery and demolition and recycling are not considered. These results are in line with those obtained by Morera et al. [23], who evaluated the construction phase contribution to mineral resource scarcity as over 60%, but considering 1 m^3 of treated water as functional unit. The major contributors to construction phase load are associated to steel chromium used (77–87%) and concrete applications (12–20%). Environmental WWT assessment of scenarios with reduced energy and material recovery may therefore present a non-negligible construction phase impact, at least for the mineral resource scarcity category.

The environmental impact on fossil resource scarcity followed a similar pattern to the global warming potential (see Fig. 2c), since energy production in Spain still depends strongly on the consumption of fossil fuels [7]. NED is thus the most important factor in the fossil resource scarcity impact category and, accordingly, $H_{41}T_{27}$ presented the lowest environmental load, while $H_{26}T_{27}$ showed the lowest fossil resource preservation. However, this analogous pattern between global warming and fossil resource scarcity may change according to the energy mix selected, as already mentioned. It is also remarkable that both net mineral and fossil resource depletion obtained negative loads in all the operating periods, which indicates that AnMBR contributes to preserving these resources by recycling and valorizing the resources embedded in wastewater and hence contributing to avoid non-renewable resources extraction from conventional sources. These results are in line with Pretel et al. [25], who also obtained negative loads for abiotic depletion category for wastewater treatments based on AnMBR. Conversely, Niero et al. [24] and Canaj et al. [2] obtained positive loads for fossil depletion and fossil and mineral resource scarcity, respectively, assessing the environmental performance of WWTP based on CAS and AD technologies, which confirms the environmental benefits of AnMBR-based treatment over conventional ones.

Even though the operation phase was the major contributor to the fossil resource scarcity input, the construction phase burden forms 18–27% of the direct load, with PVC of pipes (47–51%) and concrete applications (27–28%) as main contributors. Again, this contribution is similar to the results reported by Morera et al. [23], i.e., ≈ 20%, and confirmed that the construction phase could be important in some cases.

3.2.1.3. Freshwater and marine eutrophication. Effluent characterization was the main factor affecting both freshwater and marine eutrophication due to its nitrogen and phosphorus content (see Fig. 3d and e) in line with results obtained by Niero et al. [24], Pretel et al. [25] and Rebello et al. [29], among others. Since freshwater eutrophication is characterized in kg P_{eq}, the trends in this impact category are marked by the effluent phosphorus content and the organic matter removal efficiency in each OP. Results for freshwater eutrophication are in the $2.30{\cdot}10^{-3}$-$2.97{\cdot}10^{-3}$ kg $P_{eq}{\cdot}kgCOD_{rem}^{-1}$ range, which are higher than those reported by Canaj et al. [2] for aerobic-based WWTP ($9.55{\cdot}10^{-4}$ kg $P_{eq}{\cdot}m^{-3}$), since aerobic-based treatments allows reducing the nutrient content in the effluent.

Despite $H_{26}T_{19}$ and $H_{26}T_{27}$ showed the lowest effluent phosphorus content (6.9 ± 2.8 and 6.3 ± 0.5 mg $P{\cdot}L^{-1}$, respectively), they had the highest impact within this category due to their comparatively low COD removal (12428 $kg{\cdot}d^{-1}$ in $H_{26}T_{19}$ and 10815 $kg{\cdot}d^{-1}$ in $H_{26}T_{27}$). On the other hand, $H_{41}T_{27}$ and $H_{25}T_{24}$ showed the highest phosphorus discharge concentration (8.8 ± 1.9 and 9.9 ± 1.9 mg $P{\cdot}L^{-1}$, respectively), but their high COD removal allowed them to reduce their relative impact.

Similarly, although marine eutrophication is characterized in kg N_{eq}, the relative impact of each operating period also depended on the COD removal. Results in this category were in the $1.12{\cdot}10$–2–$2.27{\cdot}10$–2 kg N_{eq} $kgCOD_{rem}^{-1}$ range, also higher than the one obtained by Canaj et al. [2], $2.73{\cdot}10$–4 kg N_{eq} m^{-3}. This is coherent with de nitrogen content in its effluent (23.5 mg $N{\cdot}L^{-1}$), which is half that in this study (37–54 mg $N{\cdot}L^{-1}$). $H_{41}T_{18}$, which combined the lowest effluent N_T concentration (37.0 ± 11.1 mg $N{\cdot}L^{-1}$) with a moderate COD removal performance (15376 $kg{\cdot}d^{-1}$) showed the lowest marine eutrophication burden. On the other hand, despite presenting similar effluent N_T concentrations, $H_{25}T_{24}$ showed a significantly lower marine eutrophication than $H_{26}T_{27}$, since the COD removal performance of the former (20631 $kg{\cdot}d^{-1}$) was double that the obtained in the later (10815 $kg{\cdot}d^{-1}$).

The influence of the construction phase on marine eutrophication was found to be negligible, in agreement with Morera et al. [23]. These authors reported a ≈ 5% construction phase freshwater eutrophication burden, unlike the present study, in which its contribution was found to be below 1%. The importance of the N and P sludge concentration on the impact of the agriculture use of biosolids was found to be insignificant.

Finally, it is important to highlight that, although the nutrient concentrations of the effluents from anaerobic treatments present a potential for eutrophication to be considered, it is also true that they represent an opportunity to recover these resources in considerable quantities, which entails obvious environmental and economic benefits. As an example, Jiménez-Benítez et al. [16] carried out an analysis of the combination of AnMBR and fertigation applied to two case studies in Spain and Italy. The results showed that proper irrigation planning allows reducing the discharges of both N and P into the environment by up to 74%, which also means a potential of reducing the need for chemical fertilizers by up to 100%. In addition, significant economic and CO_2 savings were recorded in the AnMBR+fertigation systems compared to the base scenarios made up of a conventional WWTP and a separate irrigation system.

3.2.1.4. Water consumption. Fig. 2f shows the results of the water consumption impact category. The results indicated that reducing NED through biogas and dissolved methane recovery had a positive effect on the environmental loads. On the basis of the results, it could be also stated that taking advantage of nutrient content and thus avoiding P-based and especially N-based fertilizer production also contributed to a relevant reduction of the total impact in this category.

All the evaluated operating periods resulted in negative environmental loads (between $-4.81{\cdot}10^{-3}$ and $-2.87{\cdot}10^{-3}$ $m^3{\cdot}kgCOD_{rem}^{-1}$), which indicates a global water consumption saving. Conversely, Canaj et al. [2] obtained a water consumption of 0.38 m^3 per m^3 of treated water, but considering groundwater withdrawal for irrigation.

According to the results depicted in Fig. 1, energy recovery and avoidance of N-based fertilizer production showed a high influence in water consumption impact. Therefore, $H_{26}T_{27}$ comparatively had the worst performance in this impact category since it showed the lowest energy recovery potential and relatively low avoidance of mineral fertilizer production. The opposite results were achieved in $H_{41}T_{27}$, which had the highest methane production and fertilizing potential through sludge spreading. It is important to highlight that water reuse was not considered in this study. Including this practice in the LCA boundaries would prevent freshwater extraction, thus improving this impact category, in line with the results obtained by Cashman et al. [3] and Canaj et al. [2].

The construction phase weight in the direct water consumption category made up 21–28% of its total life-cycle impact (e.g., associated with minerals extraction and processing), without considering the load credits provided by energy recovery, avoidance of fertilizer production and material recycling after dismantling. Demolition also had a significant effect and contributed to reducing the water consumption burden in all the operating periods. Excluding the construction and demolition

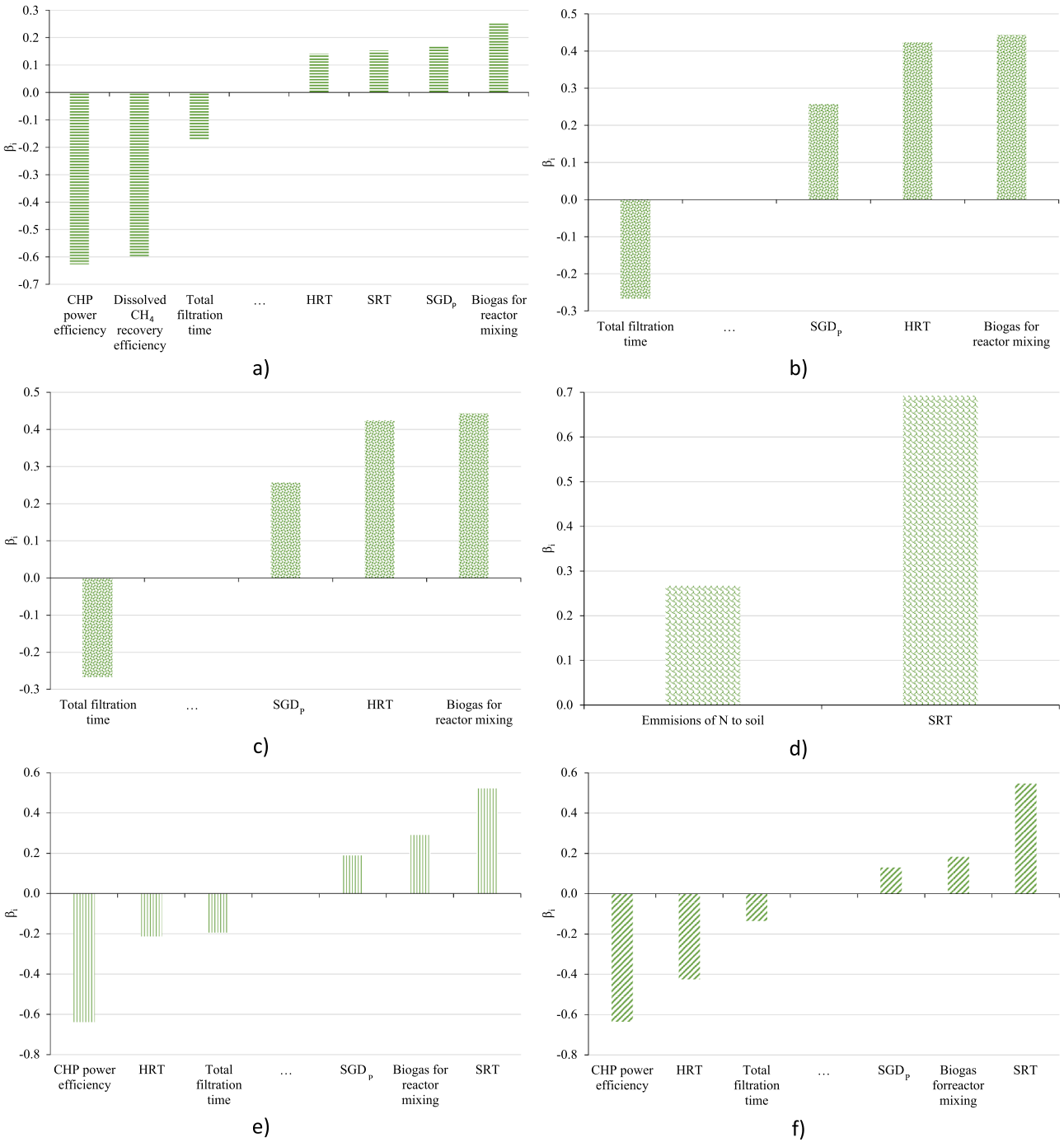

Fig. 4. Sensitivity analysis of $H_{41}T_{27}$ for (a) global warming potential, (b) mineral resource scarcity, (c) fossil resource scarcity, (d) marine eutrophication, (e) freshwater eutrophication, and (f) water consumption. "…" separates positive from negative results.

phases from the LCA boundary for this impact category should be therefore carefully evaluated so as not to affect the results.

3.2.2. *Endpoint categories*

Fig. 3a shows the results for the three endpoint impact categories included in ReCiPe 2016: human health, ecosystems, and resources.

3.2.2.1. Human health. The highest impact on human health was reached in $H_{26}T_{27}$, which is consistent with the midpoint results. In this respect, $H_{26}T_{27}$ resulted in the highest impact in terms of human carcinogenic and non-carcinogenic toxicity. As can be seen in Fig. 3b, the main factor influencing human health was the zinc emissions associated with sludge spreading. Reducing sludge production and its agricultural use or reducing its heavy metal content would address this issue. For example, a reduction of 10% in dry sludge production in $H_{26}T_{27}$ (from 847 t·year^{-1} to 762 t·year^{-1}) would reduce its human health impact by 10% in the operation phase, from $5.070{\cdot}10^{-6}$ to $4.538{\cdot}10^{-6}$ DALY (Disability-Adjusted Life Year). On the other hand, A 10% reduction in zinc concentration in sludge (from 500 mg·kg^{-1} to 450 mg·kg^{-1}) would reduce human health impact to $4.629{\cdot}10^{-6}$ DALY, which represents a reduction of 9%. In Section 3.2.2.3 it will be seen that, unlike in the human health, the reduction in sludge production has a detrimental effect on the resources category due to a lower potential use in agriculture of the nutrients present in the sludge. Finally, AnMBR

also contributes to human health by the physical disinfection provided by the UF membranes. Despite this effect is not captured by ReCiPe2016, it is clear that preventing pathogens discharge into water bodies enhances human health protection.

3.2.2.2. Ecosystems. As regards ecosystems, $H_{26}T_{27}$ presented the highest relative impact, while $H_{41}T_{27}$ had the lowest environmental burden. The biggest influence on ecosystems came from phosphorus release, methane emissions associated with effluent discharge, NED, and from zinc released to the soil in a lesser extent. In $H_{26}T_{27}$, the lowest effluent P_T concentration (6.3 ± 0.5 mg P·L^{-1}), the relative moderate dry sludge production (847 t·year^{-1}), and the low dissolved methane concentration in the effluent (2.6 ± 0.5 mg·L^{-1}, mainly due to operating at high temperature: 27 ± 1 °C) were not able to offset the combination of the lowest resulting NED (−0.254 kWh·kg COD_{rem}^{-1}) and the lowest COD removal performance (10815 kg·d^{-1}) of this OP. On the other hand, $H_{41}T_{27}$ presented the minimum impact due to a more favorable NED (−0.668 kWh·kg COD_{rem}^{-1}) and low dissolved methane release (2.6 ± 0.5 mg·L^{-1}, operating temperature of 27 °C), despite its highest dry sludge production (1049 t·year^{-1}) and high effluent P_T concentration (8.8 ± 1.9 mg P·L^{-1}).

3.2.2.3. Resources. The results obtained for the impact on resources were mainly affected by NED and nutrient recovery for agriculture, especially nitrogen and its associated avoidance of mineral fertilizer production (see Fig. 3b). Both items gave to all the operating periods evaluated a net impact reduction in this impact category, as previously mentioned for the former midpoint impact categories (mineral and fossil resource scarcities). Again, $H_{41}T_{27}$ showed the best results due to the resulting favorable NED (−0.668 kWh·kg COD_{rem}^{-1}) and high dry sludge production (1049 t·year^{-1}), suitable for nutrient recovery through sludge spreading. Conversely, $H_{26}T_{27}$ resulted in a less favorable NED (−0.254 kWh·kg COD_{rem}^{-1}), produced less sludge (847 t·year^{-1}) and removed less COD (10815 kg·d^{-1}), with the worst results for ecosystems. It is important to highlight here that sludge production and spreading has opposite effects when analyzing the impact on human health and ecosystems. This is due to the effect of different substances on different areas of protection is considered. In the case of the human health area, the impact is given by the presence of heavy metals in the sludge, therefore, an increase in the amounts destined for agricultural use means increasing the amount of metals deposited in the soil and, consequently, an increased potential for health damage. On the contrary, when the area of resources is considered, the substances of influence are nutrients, so that a greater use of sludge in agriculture reduces the needs of industrial fertilizers and, consequently, a lower consumption of resources.

3.2.3. Sensitivity analysis

By way of example, the results from the SA of $H_{41}T_{27}$ are shown in Fig. 4, providing the main influencing parameters ($b_i \geq 0.1$) for the key midpoint impact categories selected in this work. These factors are CHP electric efficiency; dissolved methane recovery efficiency; total filtration time; SGD_P; rector mixing; HRT; SRT; and emissions of nitrogen to soil.

3.2.3.1. CHP electric and dissolved CH_4 recovery efficiencies. CHP efficiency had a significant influence on global warming potential, fossil resource scarcity, freshwater eutrophication and water consumption, which is consistent with the results given in Fig. 1. A high energy recovery potential reduces CO_2 emissions associated with fossil fuel use and prevents fossil resource depletion. Same effect would produce an increase in dissolved CH_4 recovery and the consequent increase in energy production, jointly with a direct reduction in the global warming potential associated with diminishing CH_4 emissions from the effluent. Moreover, reducing the consumption of electricity produced from fossil fuels also makes it possible to improve the water consumption results,

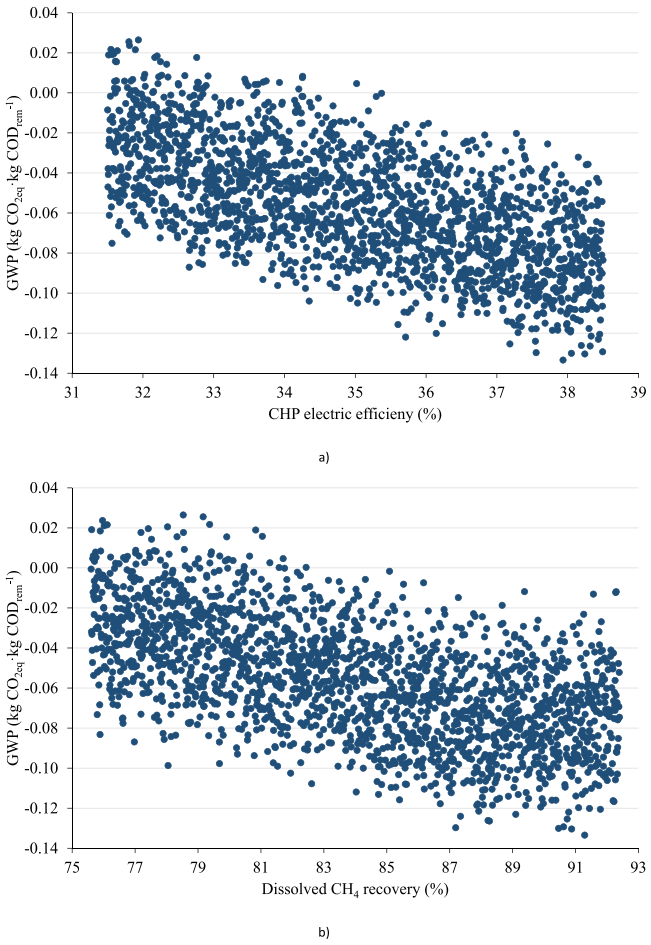

Fig. 5. Monte Carlo simulation results. Effect on global warming potential (GWP) output of: a) CHP electric efficiency and b) dissolved methane recovery efficiency for $H_{41}T_{27}$.

since the energy produced from conventional sources (i.e., coal, gas, nuclear, etc.) requires significant water consumption. The relationship between freshwater eutrophication and energy is not so obvious and could be associated with phosphorus emissions during fossil fuel extraction [12]. Therefore, the improvements associated with energy recovery go firstly through the development of CHP technology that allows increasing its efficiency and, secondly, through the optimization of the operation of methane recovery technologies.

Fig. 5 shows the Monte Carlo simulation results for global warming potential with regard to CHP power efficiency (Fig. 5a) and dissolved methane recovery efficiency (Fig. 5b) in $H_{41}T_{27}$. As already mentioned, higher CHP power efficiencies leads to reduced CO_2 emissions, since the external energy requirements from the grid are lower. The trend of the results shows emissions below − 0.4 kg $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$ with CHP efficiencies of 38.5%. Additionally, dissolved methane recovery over 83% (see Fig. 5b) would also guarantee negative net GHG emissions. Therefore, the combination of AnMBR and CHP shows a promising performance to contribute to reduce the carbon footprint of wastewater treatment.

3.2.3.2. Downtime for physical cleaning. Reducing the downtime for physical cleaning would increase filtration productivity (i.e., total filtration time), while decreasing SGD_P if keeping the same gas sparging intensity for membrane scouring. Under this premise, increasing filtration productivity would reduce energy needs and improve global warming potential, fossil resource scarcity, freshwater eutrophication and water consumption. Therefore, downtime for physical cleaning

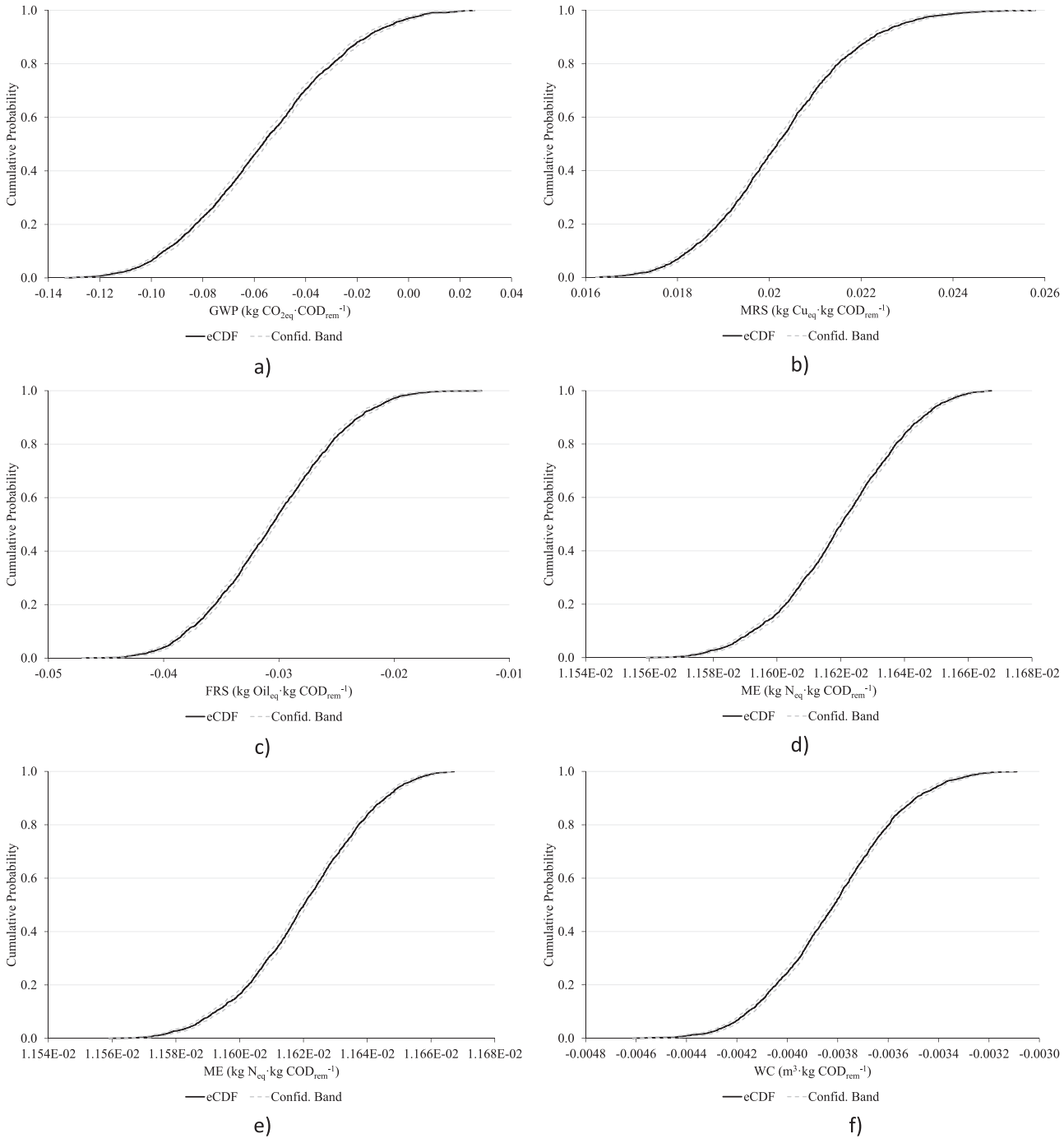

Fig. 6. Uncertainty analysis results for $H_{41}T_{27}$. Empirical cumulative probability for: (a) global warming potential (GWP), (b) mineral resource scarcity (MRS), (c) fossil resource scarcity (FRS), (d) marine eutrophication (ME), (e) freshwater eutrophication (FE), and (f) water consumption (WC).

should be optimized to improve filtration productivity without increasing TMP. Moreover, an enhanced filtration productivity would lead to a reduction in the membrane surface which would also prevent mineral resource scarcity since less material is required in their manufacture.

3.2.3.3. Biogas sparging for membrane scouring and reactor mixing. As commented in Section 3.1, as membrane scouring and reactor mixing are included among the main energy consumption drivers, increasing them also raises the environmental loads associated with the impact categories significantly influenced by the generation and use of energy: global warming potential, fossil and mineral resource scarcity, freshwater eutrophication and water consumption. Reducing energy consumption for membrane scouring requires going through the optimization of the filtration process, e.g., minimizing gas sparging at constant flux or vice versa while guaranteeing low membrane fouling propensities. On the other hand, an optimization of the reactor mixing would also contribute to minimize the environmental impact associated with this process. This optimization would be based on maintaining the minimum gas flow to avoid sedimentation of the solids in the reactor, considering the rheological and hydrodynamic conditions of the system, e.g., solids concentration, viscosity, gas rising, etc.

3.2.3.4. HRT. HRT was identified as an influential input factor on three

Table 3
Uncertainty results for $H_{41}T_{27}$: 5th-95th percentile and 5th-95th range.

	GWP[a] kg CO_{2eq}·kg^{-1} COD_{rem}	MRS[b] kg Cu_{eq}·kg COD_{rem}^{-1}	FRS[c] kg Oil_{eq}·kg COD_{rem}^{-1}	ME[d] kg N_{eq}·kg COD_{rem}^{-1}	FE[e] kg P_{eq}·kg COD_{rem}^{-1}	WC[f] m^3·kg COD_{rem}^{-1}
5th percentile	-0.126	0.017	-0.045	0.012	0.002	-0.004
95th percentile	0.019	0.026	-0.014	0.012	0.002	-0.003
5th-95th range	0.144	0.009	0.031	≈ 0.000	≈ 0.000	0.001

[a] global warming potential;
[b] mineral resource scarcity;
[c] fossil resource scarcity;
[d] marine eutrophication;
[e] freshwater eutrophication;
[f] water consumption

impact categories. HRT was positively correlated with global warming potential, mineral and fossil resource scarcity. Increasing HRT means increasing the reactor volume for a given treatment capacity. Thus, the emissions associated with material consumption (e.g., concrete) and energy consumption for reactor mixing would increase as the HRT is increased, resulting in increased environmental loads within mineral and fossil resource depletion impact categories. Hence, reducing HRT is a suitable option for improving the environmental performance in these three categories. However, reducing HRT for a given SRT would entail an increase in the mixed liquor total suspended solids (MLTSS) concentration in the reactor, which eventually could increment membrane fouling propensity for a given filtration operating mode (e.g., downtime for physical cleaning, gas sparging intensity for membrane scouring, etc.). Although fouling propensity could be controlled by modifying the filtration operating mode when raising MLTSS, it may impact other dimensions (e.g., energy consumption), thus both filtration process configuration and operation and HRT might be considered jointly for process optimization purposes.

3.2.3.5. SRT. SRT was identified to mainly affect methane production, reactor volume, wasting flow rate and nutrients concentration in the effluent. Regarding impact categories, it is positively correlated with global warming potential, fossil resource scarcity, marine and freshwater eutrophication and water consumption. Increasing SRT generally implies an increase in methane production. However, the operation of the AnMBR demo-plant was already carried out at long SRT (70 d), so the availability of additional methanizable organic matter was not significant and the potential increase in energy recovery was low. On the other hand, maintaining the MLTSS concentration by increasing the SRT implies an increase in the reactor volume. As commented above, increasing the reactor volume would result in increased consumption of construction materials and energy requirements for reactor mixing. In addition, the increase in the SRT implies a slight decrease in the waste sludge and, therefore, in the potential for nutrient recovery through biosolids agricultural use, which limits the environmental benefits associated with the displacement of the manufacture of industrial fertilizers. These combined effects explain why, in this case, an increase in SRT leads to an increase in global warming potential and fossil resource scarcity. However, it is important to highlight that in the case of facilities operating at short SRTs, increasing the SRT could imply a significant increase in methane production, even enough for the effect of increasing SRT to be the opposite of that obtained in this study, thus decreasing global warming potential as increasing SRT. Finally, an increase in the SRT means increasing the mineralization of organic nutrients, increasing the concentration of inorganic nitrogen and phosphorus that is discharged into the environment along with the effluent, although it had a negligible effect in this work due to the long SRT applied. Additional releases of nutrients to the environment would increase the environmental load within eutrophication impact categories.

3.2.3.6. Emissions of nitrogen to soil. Emissions of nitrogen to soil from sludge spreading contributes to marine eutrophication (quantified in kg of N_{eq}). On the one hand, A fraction of the nitrogen can percolate through the soil and reach groundwater, through which it is finally transported to the marine environment. On the other hand, runoff can also carry a fraction of nitrogen that will flow towards the sea through surface water. To limit this detrimental effect, the application of agricultural sludge must be carried out following good agricultural practices and adequately balancing the nutrient needs of crops and the amount of sludge to be applied.

3.2.4. Uncertainty analysis

Fig. 6 shows the empirical cumulative function (eCDF) and confidence interval for the six selected midpoint impact categories in $H_{41}T_{27}$ as representative of the study. It can be seen that eCDF follows a standard normal distribution. Table 3 shows that the lowest 5th-95th range was obtained for marine and freshwater eutrophication (both almost 0.0 kg N_{eq}·kg COD_{rem}^{-1} and kg P_{eq}·kg COD_{rem}^{-1}, respectively) followed by water consumption, with $1.0{\cdot}10^{-3}$ m^3·kg COD_{rem}^{-1}. The highest 5th-95th range was for global warming potential (0.144 kg CO_{2eq}·kg COD_{rem}^{-1}) and fossil resource scarcity (0.031 kg Oil_{eq}·kg COD_{rem}^{-1}), while the mineral resource scarcity showed intermediate values (0.009 kg Cu_{eq}·kg COD_{rem}^{-1}). These results therefore indicate that the global warming potential is subject to greater uncertainty and that it is necessary to strengthen the mechanisms to guarantee that the data used in constructing the LCI that influence this impact category are sufficiently robust.

4. Conclusions

AnMBR performed as energy producer (NED between −0.277 and −0.688 kWh·kg COD_{rem}^{-1}), being HRT, membrane scouring, filtration strategy, reactor temperature, OLR, and the COD:SO_4^{-2} ratio the main influential parameters on NED.

Energy and nutrients recovery were identified as key factors for improving the environmental performance of a full-scale AnMBR for UWW designed using real data from a demo-scale AnMBR plant. Methane and N_2O release to air, and nitrogen and phosphorus release to soil and water bodies are major drivers for environmental impacts, jointly with heavy metal content in sludge. Degassing membranes for dissolved methane capture appear as a key technology to enhance AnMBR environmental performance, while post-treatment technologies for nutrient recovery/removal (e.g., microalgae cultivation, anammox, ion exchange, membrane contactors, etc.) may be necessary for reducing marine and freshwater eutrophication.

Recovering the dissolved methane with efficiencies of 80–84% led AnMBR to act as carbon sink (emissions avoidance up to 0.223 $kgCO_{2eq}{\cdot}kgCOD_{rem}^{-1}$). Construction phase contribution to overall LCA was relevant in some operating periods and/or impact categories: 8–13% in global warming potential, 51–71% in mineral resource scarcity, 18–27% in fossil resource scarcity, 21–28% in water consumption, but negligible in marine and freshwater eutrophication: $< 1\%$. Therefore, its exclusion from the system boundary should be carefully

evaluated.

Finally, the SA confirms that there is room to improve the environmental performance of the AnMBR technology by increasing the potential for energy recovery (improvement in the efficiency of CHP technologies and optimization of dissolved methane recovery technologies), optimization of filtration, membrane scouring and mixing processes, adjustment of HRT and SRT and use of appropriate agricultural practices.

CRediT authorship contribution statement

A. Jiménez-Benítez: Writing - Original Draft, Investigation, Methodology, Formal analysis, Writing - Review & Editing. **J.R. Vázquez** Investigation, Methodology, Resources, Writing - Review & Editing. **A. Seco:** Investigation, Validation, Writing - Review & Editing, Supervision, Management and coordination responsibility for the research activity planning and execution. **J. Serralta:** Resources, Investigation, Management and coordination responsibility for the research activity planning and execution, Validation, Writing - Review & Editing. **F. Rogalla:** Investigation, Methodology, Resources. **Á. Robles:** Definition, Methodology, Formal analysis, Investigation, Validation, Writing - Review & Editing, Supervision, Management and coordination responsibility for the research activity planning and execution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2023.111141.

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