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Additional Information

2	Resource recovery from sewage through an innovative
3	anaerobic-based water resource recovery facility
4	(WRRF)
5	A. Seco*, S. Aparicio*, J. González-Camejo**, A. Jiménez-Benítez**, O.
6	Mateo**, J.F. Mora**, G. Noriega-Hevia**, P. Sanchis-Perucho*, R. Serna-
7	García*, N. Zamorano-López*, J.B. Giménez*, A. Ruiz-Martínez**, D.
8	Aguado**, R. Barat**, L. Borrás*, A. Bouzas*, N. Martí*, M. Pachés**, J.
9	Ribes*, A. Robles*, M.V. Ruano*, J. Serralta** and J. Ferrer**
10 11	* CALAGUA – Unidad Mixta UV-UPV, Departament d'Enginyeria Química, Universitat de València, Avinguda de la Universitat s/n, 46100 Burjassot, Valencia, Spain
12	** CALAGUA – Unidad Mixta UV-UPV, Institut Universitari d'Investigació d'Enginyeria de
13	l'Aigua i Medi Ambient – IIAMA, Universitat Politècnica de Valencia, Camí de Vera s/n, 46022
14	Valencia, Spain
15	¹ E-mail: angel.robles@uv.es
16	
17	Abstract
18	This research work proposes an innovative water resource recovery
19	facility (WRRF) for the recovery of energy, nutrients and reclaimed
20	water from sewage, which represents a promising approach towards
21	enhanced circular economy scenarios. To this aim, anaerobic
22	technology, microalgae cultivation, and membrane technology were
23	combined in a dedicated platform. The proposed platform produces a

high-quality solid- and coliform-free effluent that can be directly discharged to receiving water bodies identified as sensitive areas. Specifically, the content of organic matter, nitrogen and phosphorus in the effluent was 45 mg COD·L⁻¹, 14.9 mg N·L⁻¹ and 0.5 mg P·L⁻¹, respectively. Harvested solar energy and carbon dioxide biofixation in the form of microalgae biomass allowed achieving remarkable methane yields (399 STP L CH₄·kg⁻¹ COD_{inf}) equivalent to theoretical electricity productions of around 0.52 kWh per m³ of wastewater entering the WRRF. Furthermore, 26.6% of total nitrogen influent load was recovered as ammonium sulphate, while nitrogen and phosphorus were recovered in the biosolids produced (650 \pm 77 mg N·L⁻¹ and 121.0 \pm 7.2 mg P·L⁻¹).

Keywords

Anaerobic membrane bioreactor (AnMBR); anaerobic digestion (AD); membrane photobioreactor (MPBR); resource recovery; sewage; water resource recovery facility (WRRF).

1. Introduction

Sewage treatment is usually based on energy-intensive aerobic processes where the energy input for organic matter oxidation accounts up to 50% of the total energy demand of the wastewater treatment plant (WWTP) (Sid *et al.*, 2017). In addition, aerobic processes present a limited chance for nutrient recovery since nitrogen is usually released

1 to the atmosphere via denitrification and phosphorus is usually stored as a metal salt 2 within the sludge, preventing its possible reuse. Therefore, within an enhanced circular 3 economy perspective, sewage treatment needs to shift towards new cost-effective, green 4 alternatives allowing to maximise resource recovery from sewage, e.g. energy, nutrients, 5 reclaimed water, and biosolids. 6 A new sewage treatment paradigm based on the so-called water resource recovery facility 7 (WRRF) concept has emerged for waste-to-resource recovery within the scientific 8 community (see e.g. Batstone et al., 2015). Within this paradigm, sewage is no longer 9 considered as a waste but as a source of raw valuable resources, resulting in 10 environmental and economic benefits (Puyol et al., 2017). To this aim, different platforms 11 for resource recovery have been defined. For instance, Batstone et al. (2015) proposed 12 two platforms mainly consisting in the following: (i) low energy mainstream, based on 13 low strength anaerobic treatment; and (ii) uptake-release-recover, where nutrients and 14 carbon are assimilated during biological uptake through either assimilation (i.e. growth) 15 or accumulation by phototrophic or heterotrophic organism, followed by anaerobic 16 digestion (AD) of this biomass and nutrients and carbon recovery from the produced 17 digestate. 18 The anaerobic membrane bioreactor (AnMBR) has emerged as a promising energy-19 effective technology for mainstream anaerobic treatment of low-strength wastewater 20 (Pretel et al., 2016). AnMBRs have an intrinsic advantage compared to conventional 21 anaerobic digestion: the use of membranes for decoupling the sludge retention time (SRT) 22 from the hydraulic retention time (HRT). Therefore, AnMBRs can treat high flow rates 23 with relatively low footprints since biomass washout is avoided by membrane filtration.

Moreover, the reduced growth rates of anaerobic organisms at low temperature is offset

- 1 by the biomass retention, promoting the application of anaerobic biotechnology to a wider
- 2 range of environmental conditions (Giménez et al., 2012).
- 3 Some bottlenecks that prevent the widespread application of AnMBR still remain, such
- 4 as the loss of dissolved methane in the effluent (which increases as the operating
- 5 temperature decreases) and the competition between sulphate-reducing organisms (SRO)
- 6 and methanogens for the available substrate when treating wastewaters with low organic
- 7 matter to sulphate ratios (COD:SO₄-S) (Giménez et al., 2012). According to this, Pretel
- 8 et al. (2016) showed that the anaerobic treatment of sulphate-rich wastewater at ambient
- 9 temperature could be enhanced including a primary settling stage prior to an AnMBR.
- 10 This combination results in a WRRF where methane is produced in a sidestream AD,
- where the operating temperature of the unit can be increased using the heat generated in
- 12 a combined heat and power (CHP) system fuelled with the biogas produced in the system.
- Moreover, when treating sulphate-rich wastewaters, since the COD:SO₄-S ratio entering
- 14 the sidestream AD is much higher than the one entering the mainstream AnMBR, the
- 15 growth of methanogens is favoured, increasing therefore the methane production of the
- whole WRRF. In this treatment scheme, methane is not produced in the mainstream
- 17 AnMBR, which operates at ambient temperature and high flow rates, and it is possible to
- drastically reduce the loss of methane dissolved in the WRRF effluent
- 19 The methane dissolved in the effluent of an anaerobic process should still be recovered
- 20 for further enhancing the environmental and economic feasibility of the WRRF. To this
- 21 aim, vacuum degasification non-porous membranes have been reported as a promising
- 22 technology to replace traditional methods for dissolved methane recovery (Cookney et
- 23 al., 2016). These membranes allow direct demethanisation of anaerobic streams with
- positive energy balances of the separation process (Cookney *et al.*, 2016).

1 As for water reclamation, AnMBR equipped with ultrafiltration membranes produces a 2 high quality permeate that is (partially) disinfected (Bair et al., 2015). Moreover, this 3 effluent contains certain nitrogen and phosphorus concentrations (Giménez et al., 2011) 4 thus representing a valuable water source for fertigation purposes. However, when 5 fertigation is not possible, these concentrations of nutrients could prevent AnMBR 6 effluent from direct emission to different receiving water bodies. In such cases, these 7 nutrients can be recovered by different techniques, such as the cultivation of phototrophic 8 organisms (Viruela et al., 2016). 9 Within the uptake-release-recover concept, membrane photobioreactor (MPBR) for 10 autotrophic microalgae cultivation has been reported as an interesting approach for 11 nutrient recovery (Bilad et al., 2014), presenting lower footprints than other microalgae 12 cultivation systems (Viruela et al., 2016). Autotrophic microalgae use light energy, 13 inorganic carbon and high amounts of inorganic compounds such as ammonium (NH₄) and phosphate (PO_4^{3-}) for growth. Hence, nutrients and solar energy are harvested in the 14 15 form of microalgae biomass while biofixing carbon dioxide. 16 The microalgae harvested from an MPBR can be used as carbon source in the sidestream 17 AD system, enhancing the energy balance of the WRRF, whilst the produced AD effluent 18 can be used for nutrient valorisation (Sialve et al., 2009). However, anaerobic digestion 19 of microalgae presents several drawbacks, such as: (i) low biodegradability of the highly-20 recalcitrant microalgae cell walls; (ii) low carbon to nitrogen ratio (C:N) that results in 21 high levels of free ammonia, which can inhibit the anaerobic process; and (iii) the need 22 of cost-effective microalgae harvesting systems since biogas production from microalgae

depends on biomass concentration (Giménez et al., 2018).

1 One alternative to improve the digestibility of microalgae and prevent the possible 2 inhibition of the process by free ammonia is the anaerobic digestion of this microalgae 3 biomass with carbon-rich substrates available in municipalities (e.g. food waste or sewage 4 sludge, among others). During the process, a high concentration of nutrients such as 5 nitrogen can be released and recovered in the form of commercial products such as 6 ammonium sulphate using, for instance, absorption-desorption, ion exchange with 7 zeolites or synthetic resins or membrane contactors. The latter stands out as a promising 8 recovery technology since it has been reported to achieve low energy requirements and 9 high efficiency recovery yields (Norddahl et al., 2006). 10 All the above is proof that several attempts have been made by different authors to 11 transform the classical WWTPs into more energy and environmental efficient facilities. 12 Shifting from aerobic for anaerobic processes (Pretel et al., 2016), recovering nitrogen as 13 ammonium sulphate using membrane contactors (Norddahl et al., 2006), or recovering 14 phosphorus as struvite (Martí et al., 2017). However, to the best of authors' knowledge, 15 there are no studies so far evaluating the resource recovery from wastewater streams in 16 real conditions as a holistic approach. 17 The mainstream of the proposed WRRF platform consists of a primary settling step, an 18 AnMBR as secondary treatment, and a MPBR as tertiary treatment. The combination of 19 AnMBR and MPBR transforms the sewage into microalgae biomass (a source of energy 20 and nutrients) and reclaimed water. The sidestream of this platform consists of an 21 additional AnMBR, a non-porous degassing membrane, and a membrane contactor. The 22 anaerobic digestion of sewage sludge and harvested microalgae biomass enhances biogas 23 production. The non-porous degassing membrane and the membrane contactor enable to 24 recover the dissolved methane and nitrogen, respectively. This treatment platform is 25 proposed for treating sulphate-rich sewage. However, when treating wastewaters with a

- 1 low sulphate content, this platform would be significantly simplified since previous
- 2 studies have demonstrated that the combination of AnMBR and MPBR is an interesting
- 3 approach for resource recovery from sewage (see e.g. Pretel et al., 2015; González-
- 4 Camejo et al., 2017).
- 5 The objective of this work is to provide a proof of concept and evaluation of the technical
- 6 feasibility of the proposed novel WRRF platform proposed for the recovery of reclaimed
- 7 water, nutrients and energy from (sulphate-rich) sewage, based on a circular economy
- 8 perspective.

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2. Methods

- 11 2.1. Water Resource Recovery Facility (WRRF) platform
- 12 Figure 1 shows the flow diagram of the WRRF platform proposed in this study, which is
- 13 located in the "Conca del Carraixet" WWTP (Valencia, Spain).
- Regarding the mainstream, the raw sewage pre-treatment consists of screening, degritter,
- and grease removal, after which the wastewater (sampling point #0) is introduced to a
- 16 gravity-based primary clarifier for continuous removal of solids. Effluent from this
- 17 primary clarifier is fed to a secondary treatment consisting in an AnMBR unit, where
- soluble organics are biologically removed and solids are physically retained. Moreover,
- 19 the nitrogen and phosphorus content in the organic forms are mineralised, becoming
- available for recovery in the MPBR pilot unit.
- 21 The solids-free permeate from the AnMBR is fed to a tertiary treatment consisting in a
- 22 MPBR for microalgae cultivation, where solar energy is harvested, inorganic nutrients

- 1 are biologically assimilated, and carbon dioxide is biofixed as microalgae biomass. In
- 2 addition, the MPBR system produces reclaimed water.
- 3 Concerning the sidestream, the sewage sludge and the harvested microalgae biomass are
- 4 concentrated and fed to an AD process based on AnMBR technology (AnMBR_{AD}, i.e.
- 5 sidestream AnMBR). This system valorises the organic matter in the form of biogas.
- 6 Moreover, a nutrient-rich permeate and biosolids are produced, which could be used for
- 7 nutrient valorisation. Specifically, the biosolids can be used for agricultural purposes,
- 8 while the produced permeate is firstly treated in a non-porous membrane for dissolved
- 9 methane recovery, and subsequently introduced to a membrane contactor for nitrogen
- 10 recovery.

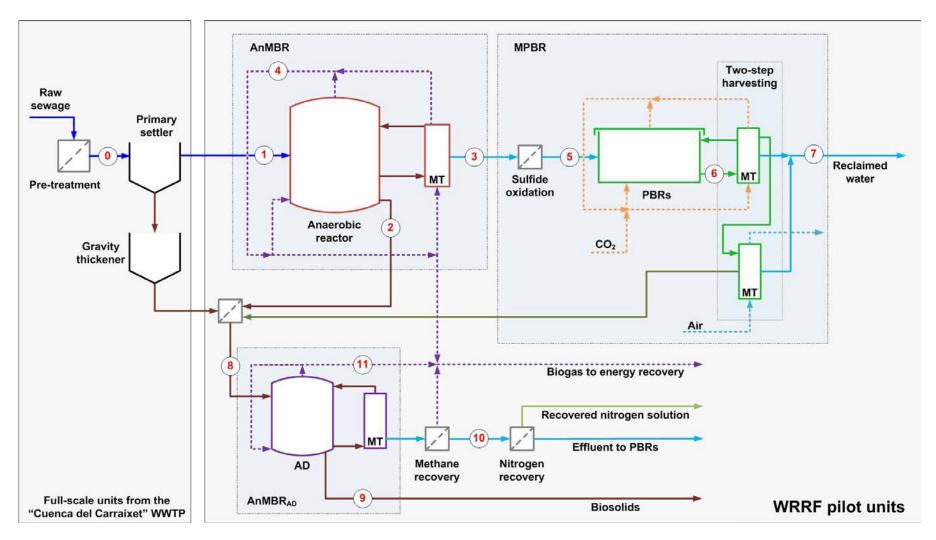


Figure 1. Layout of the WRRF including the sampling points (1-11). AD: anaerobic digester; AnMBR: anaerobic membrane bioreactor; AnMBR_{AD}: sidestream anaerobic membrane bioreactor; MPBR: membrane photobioreactor; MT: membrane tank; PBR: photobioreactor.

2.2.Description of the pilot units

2.2.1. AnMBR pilot unit

- 3 The AnMBR pilot unit mainly consists of an anaerobic reactor with a total volume of
- 4 1300 L (900 L working volume) connected to two membrane tanks, each one with a total
- 5 volume of 800 L (600 L working volume). Each membrane tank is equipped with one
- 6 industrial-scale hollow-fibre ultrafiltration membrane unit composed of 9 membrane
- 7 bundles (PURON[®] KMS PUR-PSH31, 0.03 μm pores) with a total filtration area of 31
- 8 m². Gas-assisted membrane-scouring was used to minimise cake layer formation.

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2.2.2. MPBR pilot unit

- 11 The MPBR plant mainly consists of three 1.25-m height, 2-m width and 0.1-m depth
- methacrylate flat-plate photobioreactors (PBRs) with a total volume of 750 L (maximum
- working volume of 705 L). The PBRs are connected to a two-step harvesting system.
- 14 Each filtration step consists in a membrane tank of 14 L that includes a hollow-fibre
- 15 membrane bundle with a filtration area of 3.44 m². This bundle was obtained by
- modifying an industrial-scale hollow-fibre ultrafiltration membrane unit (PURON® KMS)
- 17 PUR-PSH31, 0.03 µm pores). The PBRs were continuously stirred by gas sparging,
- enabling proper mixing of the culture and preventing wall fouling. Membrane scouring
- by gas sparging was used to minimise cake layer formation in both filtration steps.

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2.2.3. AnMBR_{AD} pilot unit

- The AnMBR_{AD} plant consists of an AD with a total volume of 1000 L (maximum working
- volume of 900 L) and a 1-L membrane tank fitted with a 0.42-m² hollow-fibre

ultrafiltration membrane unit (PURON® KMS, 0.03 µm pores). An equalisation tank of 1 2 125 L is used to mix the different co-substrates prior to being fed to the system. To 3 improve the mixing conditions in the AD and to favour the stripping of the produced 4

of the digester. Biogas-assisted membrane-scouring was used to minimise cake layer

gases from the liquid phase, a fraction of the produced biogas was recycled to the bottom

6 formation.

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2.2.4. Dissolved methane and nitrogen membrane-based recovery systems

The proposed WRRF platform was equipped with a polydimethylsiloxane (PDMS) membrane module provided by PermSelect® (MedArray Inc., USA) with a total filtration area of 2.1 m², which was used as a final polishing step for desorption and recovery of dissolved methane. Furthermore, a microporous polypropylene (PP) membrane contactor of 1.4 m² provided by Liqui-Cel[®] (model 2.5 x 8 Extra Flow X50) was used for the recovery of free ammonia as ammonium sulphate.

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2.3. Operating conditions of the pilot units

The operational conditions of the AnMBR, MPBR and AnMBR_{AD} pilot units within the experimental period of this work are shown in Table 1. The pilot units were operated continuously for 3 months. This study shows the data obtained under steady-state conditions. Steady-state conditions were related to stable suspended solids concentrations in the bioreactors. A 30-day period of steady-state data is shown in this paper. Table 2 shows the average characteristics of the different streams of the WRRF (Figure 1) during the steady-state period.

The AnMBR and the MPBR units were operated outdoors at ambient temperature. In addition, the MPBR system was operated at variable light intensity due to the dynamics of the environmental conditions. It is worth to point out that due to operating volume restrictions, only a fraction of the produced AnMBR effluent was fed to the MPBR. The pH of the MPBR was controlled at 7.5 by the addition of pure CO₂ into the aeration system to avoid undesirable chemical processes such as phosphate precipitation and free ammonia stripping.

Table 1. Operating conditions of the AnMBR, MPBR and AnMBR_{AD} pilot units. SRT: Sludge retention
 time; HRT: hydraulic retention time; PAR: photoactive Radiation; OLR: organic loading rate.

	SRT	HRT	Temperature	Working volume	Light PAR	OLR
	(d)	(d)	(°C)	(L)	(μmol·m ⁻² s ⁻¹)	(g COD·L ⁻¹ ·d ⁻¹)
AnMBR	70.0 ± 3.7	1.1 ± 0.2	24.9 ± 0.5	2100	-	0.22 ± 0.07
MPBR	5.4 ± 0.3	2.1 ± 0.1	23.8 ± 1.1	705	273 ± 118	0.04 ± 0.01
AnMBR _{AD}	69.7 ± 0.3	30.0 ± 0.3	55.0 ± 0.9	500	-	0.56 ± 0.05

Primary sludge coming from the full-scale WWTP thickener, digestate from the AnMBR pilot unit, and microalgae biomass harvested from the MPBR pilot unit were digested in the AnMBR_{AD} pilot unit. The final composition of the blending fed to the AnMBR_{AD} was the following: 34, 13, and 53% of total solids for primary sludge, AnMBR digestate, and harvested microalgae, respectively. The contribution of each tributary stream was chosen based on experimental data from previous research and new simulation data using an extended version of the mathematical model BNRM2 (data not shown) (Barat *et al.*, 2013).

- 1 As for the dissolved methane recovery system, the PDMS membrane was operated by
- 2 shell side, collecting the permeate gas into the lumen side. Vacuum was used to generate
- 3 the driving force, resulting in a transmembrane pressure of 0.8 bars in order to maximise
- 4 partial pressure gradient, thus improving methane recovery.
- 5 Concerning the nitrogen recovery system, the PP membrane contactor was operated also
- 6 by shell side, recovering the nitrogen in the lumen side in the form of ammonium sulphate.
- 7 To this aim, the nitrogen was concentrated in a sulphate acid solution of 0.05 M at a pH
- 8 of up to 9. This solution was circulated through the inner section of the membranes at a
- 9 flow rate of around 0.2 L·min⁻¹.

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2.4.Analytical methods

- 12 In order to evaluate the biological process performance, samples were collected three
- times a week from the sampling points numbered in Figure 1. Total Solids (TS), Volatile
- 14 Solids (VS), Total Suspended Solids (TSS), Volatile Suspended Solids (VSS), Total and
- 15 Soluble COD (TCOD and SCOD, respectively), Total Nitrogen (TN), Soluble Nitrogen
- 16 (SN), Ammonium (NH₄-N), Nitrite (NO₂-N), Nitrate (NO₃-N), Total Phosphorus (TP),
- 17 Soluble Phosphorus (SP), Phosphate (PO₄-P), Sulphide (S²⁻) and Sulphate (SO₂⁻⁴) were
- determined according to Standard Methods (APHA et al., 2012). Volatile Fatty Acids
- 19 (VFA) and Alkalinity (Alk) were measured by titration in accordance to the methodology
- proposed by the South African Water Research Commission (Moosbrugger *et al.*, 1992).
- 21 The presence of Escherichia coli and other coliform pathogens in permeates was
- quantitatively determined through positive β -glucorinidase assay using membrane filters,
- following the UNE-EN ISO 9308-1:2014 standard method.

- 1 The methane fraction of the biogas was measured three times a week using a gas
- 2 chromatograph equipped with a Flame Ionisation Detector (GC-FID, Thermo Scientific).
- 3 1 mL of biogas was collected by a gas-tight syringe and injected into a 15 m \times 0.53 mm
- 4 × 1 μm TRACER column (Teknokroma) which was maintained at 40 °C. The carrier gas
- 5 was helium at a flow-rate of 40 mL·min⁻¹. CH₄ pure gas (99.9995%) was used as standard.

7

2.5.Process performance indicators

- 8 The removal or recovery rate and the removal or recovery efficiency for a given
- 9 compound was calculated using Eq. 1 and Eq. 2, respectively. The volumetric microalgae
- 10 biomass productivity was calculated using Eq. 3.

Removal or recovery rate
$$(RR) = \frac{Q \cdot (C_I - C_F)}{V_R}$$
 (Eq.1)

Removal or recovery efficiency (RE) =
$$\frac{(C_I - C_F)}{C_I} \cdot 100$$
 (Eq.2)

$$Biomass \ productivity \ (BP) = \frac{Q_W \cdot X_{VSS}}{V_R} \tag{Eq.3}$$

- 11 where, C_I and C_F are the concentrations of a given compound in the influent and the
- effluent (g·m⁻³), respectively, Q is the treatment flow rate (m³·d⁻¹), V_R is the volume of
- 13 the reactor (m³), Q_w is the flow rate of wasted biomass (m³·d⁻¹) and X_{VSS} is the
- 14 concentration of volatile suspended solids in the reactor (g VSS·m⁻³).

- 16 The carbon dioxide biofixation ratio in the MPBR was calculated using the equation
- described by De Morais and Costa (2007):

$$R_{CO_2} = m_{cbm} P(\frac{M_{CO_2}}{M_C})$$
 (Eq.4)

- 1 where, m_{cbm} is the fraction of carbon in microalgae biomass (w/w), which was calculated
- 2 from the biomass composition obtained by atomic spectroscopy, P is the biomass
- 3 productivity $(g \cdot L^{-1} \cdot d^{-1})$, and M_{CO2} and M_{C} are the molecular weights of CO_2 and carbon
- 4 (C), respectively.

- 6 The anaerobic process efficiency was evaluated in terms of biodegradability percentage
- 7 and methane yield using Eq.5 and Eq.6, respectively.

% Biodegradability =
$$\frac{CH_4 - COD + H_2S - COD}{COD_{influent}} \cdot 100$$
 (Eq.5)

$$Y^{CH_4} = \frac{CH_4 - V}{COD_{influent}} \cdot 100$$
 (Eq.6)

- 8 where, $COD_{influent}$ (g $COD \cdot d^{-1}$) is the COD of the influent, $CH_4 COD$ is the COD of the
- 9 produced methane (biogas methane and methane dissolved in the effluent) (g COD·d⁻¹),
- 10 H₂S COD is the COD consumed by SRO for sulphate reduction (g COD·d⁻¹) and CH₄
- 11 V is the production of methane (biogas methane and methane dissolved in the effluent)
- 12 (L).

13

- 14 Mass balances were carried out for COD, nitrogen and phosphorus. Appendix 1 shows
- 15 the data used on mass balance calculations.

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17 2.6. Energy and economic balance of the WRRF

In order to assess the performance of the proposed WRRF platform, mass, energy and economic balances were performed. The energy and economic balance has been carried out following the model proposed by Pretel at al. (2016). The following items were considered: pumping requirements, mixing, membrane scouring, AnMBR_{AD} heating needs, energy recovery from methane, and operating and maintenance of the membrane modules (reagents for membrane cleaning and replacements). Appendix 2 shows the main assumptions considered for energy and economic balance calculations.

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3. Results and Discussion

10 The performance of the proposed WRRF platform was evaluated in terms of reclaimed 11 water production, nutrient recovery and energy recovery. The characteristics of the 12 sewage used in this study (sampling point #0 in Figure 1) as well as the characteristics of 13 the different streams of the WRRF (sampling points #1 to #11 in Figure 1) are shown in 14 Table 2. 15 It is important to highlight the significant sulphate concentration in the influent (305.6 \pm 16 45.5 mg SO₄·L⁻¹) in comparison with typical domestic wastewaters (around 90 mg SO₄·L⁻ 17 1). This high sulphate influent concentration, typical in some geographical areas like the 18 one of this study, resulted in a low COD:SO₄-S ratio in the mainstream, favouring the 19 proliferation of SRO (Giménez et al., 2012), thus degrading the soluble COD via 20 sulphate-reducing processes and hampering the methanogens development. To mitigate 21 this issue, a primary clarifier was incorporated in the layout of the system for maximising 22 energy recovery through the anaerobic partition and digestion of particulate organics in 23 the sidestream, favouring methanogens growth in the sidestream due to an increased 24 COD:SO₄-S ratio.

3.1.Reclaimed water production

- 3 As Figure 1 shows, water recovery was carried out in three consecutive steps within the
- 4 mainstream.
- 5 In the primary treatment, particulate organics were partially removed through classical
- 6 gravity clarification, reducing the concentration of TSS entering the AnMBR from 238
- 7 to 83 mg·L⁻¹ (see sampling point #0 and #1 in Table 2). This reduced solid load to the
- 8 AnMBR allows reducing the footprint of the system since smaller reaction volumes can
- 9 be projected for a given treatment flow rate. On the other hand, when operating at low
- mixed liquor suspended solids (MLSS) concentrations, both OPEX and CAPEX can be
- 11 reduced because of: (i) a reduction in the membrane fouling propensity; and (ii) the
- 12 possibility of increasing the operating transmembrane flux, decreasing the required
- 13 membrane filtration area.
- 14 The particulate organics recovered as primary sludge were valorised in the sidestream
- AD, where the high COD:SO₄-S ratio hinders the growth of SRO. In fact, COD:SO₄-S
- ratio in the influent of the sidestream AnMBR_{AD} was 164.1 kg COD·kg⁻¹ S. As a result,
- 17 the lower proportion of SO₄ limited the growth of SRO.
- During the secondary treatment (AnMBR step), solids were physically retained, organic
- 19 nutrients were mineralised and soluble organics were biologically removed via sulphate-
- 20 reducing processes, since after the primary settling step, the COD:SO₄-S ratio in the
- 21 mainstream decreased to around 2.4 kg COD·kg⁻¹ S. This low COD:SO₄-S ratio resulted
- 22 in a negligible methane production in the AnMBR unit because sulphate-reducing
- 23 organisms outcompeted methanogens for the available substrate. Therefore, this
- 24 degradation of organics in the mainstream via sulphate-reducing processes can be

- 1 considered, in combination with the previous primary settling step, an attractive approach
- 2 when treating sulphate-rich wastewaters.
- 3 The reduced COD:SO₄-S ratio in the influent to the AnMBR unit avoided methane
- 4 production in the mainline. The absence of dissolved methane in the effluent from the
- 5 AnMBR avoids any global warming potential impact associated to emissions to the
- 6 atmosphere of this compound mainstream. On the other hand, it is important to note that
- 7 most of the particulate organic matter was valorised via methanisation in the sidestream
- 8 AD system, where the treatment flow would allow increasing the operating temperature
- 9 using the heat energy generated in a CHP system using the producing biogas as fuel.
- Additionally, sulphate allowed the oxidation of soluble organic matter in the mainstream,
- 11 avoiding therefore the energy input for organic matter removal required in aerobic
- processes. However, the presence of sulphide in the effluent entails some drawbacks that
- can hinder downstream operations, such as microalgae cultivation (González-Camejo et
- 14 al., 2017). In order to avoid these possible drawbacks, a sulphide oxidation step was
- included in the WRRF after the AnMBR unit (Figure 1).
- 16 The effluent of the AnMBR unit (see sampling point #3 in Table 2) featured negligible
- 17 suspended solids and low COD concentrations. Moreover, COD levels were further
- reduced after the sulphide oxidation step (see sampling point #5 in Table 2). However,
- 19 the direct discharge of the effluent from the AnMBR to different receiving water bodies
- 20 is not always possible since it contains significant amounts of nitrogen and phosphorus.
- 21 Nonetheless, this nutrient-rich effluent from the AnMBR system is a suitable growth
- 22 medium for microalgae cultivation in a tertiary treatment based on MPBR technology.
- 23 Finally, within the tertiary treatment (MPBR step), nutrients were removed via
- 24 microalgae cultivation. The MPBR unit showed nitrogen and phosphorus removal

- 1 efficiencies of 66.7% and 85.7%, respectively, obtaining an effluent with a nutrient
- 2 content lower than the requirements established in the European Directive 91/271/CEE
- 3 for discharges to sensitive areas from urban WWTPs with treatment capacities between
- 4 10,000 and 100,000 PE (see sampling point #7 in Table 2). Moreover, as commented
- 5 before, microalgae cultivation enabled not only nutrient uptake but also solar energy
- 6 harvesting and carbon dioxide biofixation in the form of new microalgal biomass which
- 7 served as feedstock for the sidestream AnMBR_{AD} unit.

Table 2. Characterisation of the different WRRF streams during the steady-state period

Sampling point	0	1	2	3	4	5	6	7	8	9	10	11
pН	n.a.	7.9 ± 0.2	7.4 ± 0.1	n.a.	n.a.	n.a	7.4 ± 0.2	7.4 ± 0.2	n.a.	7.5 ± 0.1	n.a.	n.a.
ORP (mV)	n.a.	n.a	-467 ± 18	n.a	n.a	n.a	n.a	n.a	n.a	-522 ± 12	n.a	n.a
$\begin{array}{c} COD \\ (mg\ COD \cdot L^{\text{-}1}) \end{array}$	444 ± 42	244 ± 36	4902 ± 96	n.a	n.a	81 ± 14	1434 ± 154	45 ± 10	16737 ± 1052	17310 ± 401	n.a	n.a
$SCOD\ (mgCOD{\cdot}L^{\text{-}1})$	n.a.	98 ± 7	n.a	144 ± 11	n.a	81 ± 14	n.a	45 ± 10	1597 ± 283	n.a	1169 ± 64	n.a
$TS \pmod{mgTS \cdot L^{-1}}$	1435 ± 78	n.a	5278 ± 108	n.a	n.a	n.a	n.a	n.a	11872 ± 1048	14013 ± 929	n.a	n.a
VS (%)	78.1 ± 5.7	n.a	59.3 ± 0.8	n.a	n.a	n.a	n.a	n.a	73.5 ± 4.3	67.7 ± 2.3	n.a	n.a
$ TSS \\ (mgTSS \cdot L^{-1}) $	238 ± 28	83 ± 8	4213 ± 118	n.a	n.a	n.a	786 ± 69	n.a	9797 ± 1005	12322 ± 202	n.a	n.a
VSS (%)	81.0 ± 6.1	80.7 ± 5.7	59.3 ± 0.2	n.a	n.a	n.a	93.8 ± 2.2	n.a	75.8 ± 7.1	69.6 ± 1.3	n.a	n.a
VFA (mgHAc·L ⁻¹)	n.a.	1.9 ± 0.1	n.a	0.6 ± 0.1	n.a	n.a	n.a	n.a	756 ± 171	n.a	523 ± 35	n.a
$\begin{array}{c} Alk\\ (mgCaCO_3 \cdot L^{-1}) \end{array}$	n.a.	469 ± 50	n.a	523 ± 35	n.a	n.a	n.a	n.a	417 ± 121	n.a	1906 ± 67	n.a
CH ₄ (%)	n.a	n.a	n.a	n.a	<d.l< td=""><td>n.a</td><td>n.a</td><td>n.a</td><td>n.a</td><td>n.a</td><td>n.a</td><td>62 ± 5</td></d.l<>	n.a	n.a	n.a	n.a	n.a	n.a	62 ± 5
$TN (mgN \cdot L^{-1})$	49.9 ± 8.4	45.6 ± 5.9	332 ± 14	44.9 ± 5.8	n.a	n.a	92 ± 8	14.9 ± 1.2	547 ± 105	650 ± 77	508 ± 20	n.a
$SN (mgN \cdot L^{-1})$	n.a.	41.0 ± 4.8	n.a	44.9 ± 5.8	n.a	44.8 ± 5.3	n.a	14.9 ± 1.2	154.2 ± 44.9	n.a	508 ± 20	n.a
$TP (mgP \cdot L^{-1})$	8.3 ± 1.9	5.7 ± 1.9	85.0 ± 1.6	3.9 ± 0.5	n.a	n.a	7.7 ± 1.1	0.5 ± 0.1	145.9 ± 18.9	121.0 ± 7.2	17.7 ± 1.4	n.a
PO_4 -P (mgP·L ⁻¹)	n.a	3.3 ± 1.2	n.a	3.9 ± 0.5	n.a	3.8 ± 1.1	n.a	0.5 ± 0.1	37.3 ± 8.8	n.a	17.7 ± 1.4	n.a
$S^{2\text{-}}\left(mgS\!\cdot\!L^{\text{-}1}\right)$	n.a	n.a	n.a	52.3 ± 4.8	n.a	n.a	n.a	n.a	n.a	n.a	33.4 ± 1.7	n.a
$SO_4(mgSO_4{\cdot}L^{\text{-}1})$	n.a.	305.6 ± 45.5	n.a	132.7 ± 31.1	n.a	n.a	n.a	n.a	129.3 ± 32.4	n.a	n.a	n.a

Average and standard deviations of pH, Oxidation Redox Potential (ORP), Chemical Oxygen Demand (COD), Soluble COD (SCOD), Total Solids (TS), Volatile Solids (VS), Total Suspended Solids (TS), Volatile Suspended Solids (VSS), Volatile Fatty Acids (VFA), Alkalinity (Alk), CH₄, and TN, SN, TP, PO₄-P, S²⁻ and SO₄²⁻ concentrations in the sampling points of the proposed WRRF (See Figure 1). n.a.: not available. D.L: Detection Limit.

Apart from the requirements established in the European Directive 91/271/CEE for COD, solids and nutrients, the concentration of pathogens in the effluent from the proposed WRRF needs to be monitored based on the subsequent use of the produced water. For instance, non-faecal coliform colony-forming units (cfu) per 100 mL can be discharged to the environment according to the Spanish water quality regulation. As a result of using ultrafiltration membrane units with a mean pore size of 0.03 µm, nor E.coli cfu per 100 mL neither helminthic eggs were detected in the final treated water. Reclaimed water was therefore produced in the proposed WRRF platform, which could be used for different purposes, i.e. agricultural irrigation, aquifer recharge, urban or industrial uses, recreational areas.

3.2. Nutrient recovery

After mineralisation of the organic forms in the AnMBR unit, inorganic nutrient uptake occurred in the MPBR unit. Specifically, nitrogen uptake rate by microalgae resulted in 20 ± 3 mg N·L⁻¹·d⁻¹, while the nitrogen content in the microalgae biomass was 124 ± 25 mg N·g⁻¹ VSS. On the other hand, both the harvested microalgae biomass and the sewage sludge were fed to the sidestream AnMBR_{AD} unit, where the organic forms of nitrogen and phosphorus were also mineralised. This mineralisation led to ammonium concentrations in the AnMBR_{AD} permeate of 508 ± 20 mg N·L⁻¹.

Based on this nitrogen content, a membrane contactor was used for ammonium recovery in the form of ammonium sulphate, removing 100% of the ammonium content in the permeate from the AnMBR_{AD}. These results are similar to those obtained by Norddahl et al. (2006) at a pH over 9. However, maximum nitrogen recovery efficiencies of 83% were achieved. Ammonia stripping was identified as the main reason for this ammonium loss.

- 1 Nevertheless, these losses could be easily minimised at industrial scale by working in a
- 2 closed system.
- 3 Regarding phosphorus, phosphorus uptake rate in the MPBR unit resulted in 2.2 ± 0.6 mg
- 4 PO₄-P·L⁻¹·d⁻¹, while the phosphorus content in the microalgae biomass was 10.4 ± 4.2
- 5 mg P·g⁻¹ VSS. The phosphorus loading rate of the AnMBR_{AD} was 57% higher than the
- 6 amount determined in both permeate $(17.7 \pm 1.4 \text{ mg PO}_4\text{-P}\cdot\text{L}^{-1})$ and waste streams. These
- 7 results suggest that uncontrolled chemical precipitation occurred in the AnMBR_{AD}.
- 8 Further research is therefore needed in order to prevent this uncontrolled precipitation
- 9 and to improve the recovery of phosphorus in the sidestream AD system.
- Nutrients were also recovered in the biosolids fraction. The production of biosolids in the
- WRRF was about 0.205 kg VSS per m³ of treated water (0.461 kg VSS·kg⁻¹ COD). The
- nutrient content in the biosolids accounted for 650 ± 77 mg N·L⁻¹ and 121.0 ± 7.2 mg
- 13 P·L⁻¹ (see sampling point #9 in Table 2). These levels of nutrients make these biosolids
- eligible to be used as fertiliser. For instance, the recent literature review of Sharma *et al.*
- 15 (2017) highlights the benefits of using nutrient-rich biosolids (a source of carbon and
- inorganic nutrients) as fertiliser in the Mediterranean area, where a carbon deficiency in
- soil is commonly found. Moreover, the operating temperature set in the AnMBR_{AD} (55°C)
- 18 is reported to be effective in terms of pathogen removal (Carrington et al., 2001).
- 19 However, further research is needed to evaluate the potential of the produced biosolids
- 20 for direct farmland application or composting.

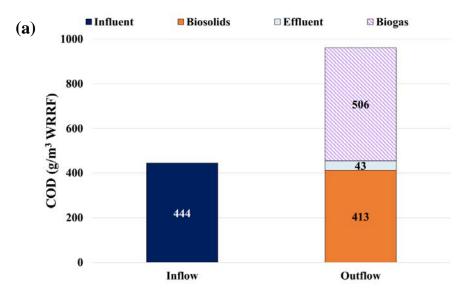
3.3.Energy recovery

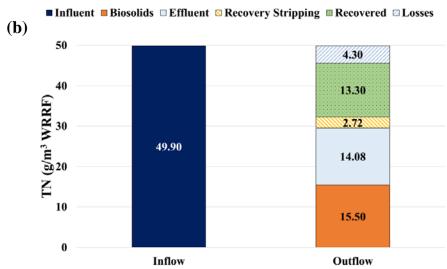
- 23 As previously mentioned, a negligible methane production was observed in the
- 24 mainstream since SRO outcompeted methanogens in the AnMBR unit. Since sulphate-

- 1 reducers consume 2 mg of biodegradable organic matter per mg of SO₄-S (Giménez et
- 2 al., 2012), a 57% of reduction of sulphate in the effluent was observed. Thus, it can be
- 3 assumed that sulphate reducing was the major pathway for organic matter removal.
- 4 Nevertheless, due to the biofixation of CO₂ in the form of microalgae biomass, an overall
- 5 methane yield of 399 STP L CH₄·kg⁻¹ COD_{inf} was achieved in the proposed WRRF
- 6 platform under the evaluated operating conditions. Specifically, methane production in
- 7 the AnMBR_{AD} unit resulted in 85.5 L_{CH4}·d⁻¹ on average. The methane content in the
- 8 produced biogas was $62 \pm 5\%$. This methane production represents a theoretical
- 9 electricity production in a CHP system of around 0.44 kWh per m³ of wastewater entering
- 10 the WRRF.
- 11 Additionally, the membrane-based system for dissolved methane recovery from the
- effluent of the AnMBR_{AD} reached an average methane recovery of 96%, reducing the loss
- of methane dissolved in the AnMBR_{AD} effluent to levels below 0.34 mg CH₄·L⁻¹.

- 3.4.Mass, energy and economic balance
- 16 Figure 2 shows the overall COD, nitrogen and phosphorus mass balances of the evaluated
- WRRF platform. Total COD output was 116% higher than the COD input (see Figure 2a)
- due to CO₂ biofixation through microalgae growth being around 0.31 g COD·L⁻¹·d⁻¹.
- Overall, the influent COD removal efficiency resulted in 90.4%. Figure 2b shows a
- significant efficiency for nitrogen recovery in the form of ammonium sulphate of 26.7%
- of total N in the influent. However, an 8.6% is lost due to stripping processes and the use
- of NO₃-N as electron acceptor during anaerobic digestion in the AnMBR_{AD}. As for the
- phosphorus mass balance, 34.8% of TP was recovered in the biosolids (see Figure 2c).

- 1 However, this figure illustrates that 55.9% of the total phosphorus content entering the
- 2 WRRF was likely to be chemically precipitated in the system.
- 3 The anaerobic digestion of sewage sludge and microalgal biomass in the AnMBR_{AD}
- 4 enhanced energy recovery in the form of biogas. Moreover, the dissolved methane loss
- 5 was minimised. The obtained digestate represented a valuable source of biosolids. These
- 6 biosolids can be used for agricultural practices, representing a promising approach
- 7 towards circular economy scenarios. Further information about mass balances is shown
- 8 in Appendix 1





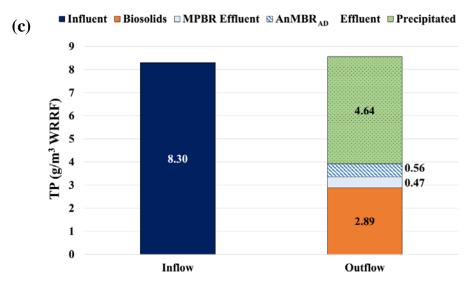
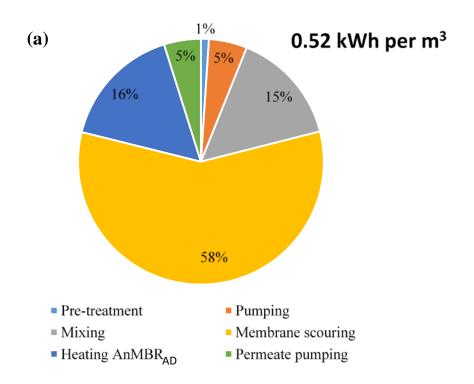


Figure 2: WRRF mass balances for: (a) COD, (b) TN and (c) TP.

- 1 As Figure 3a shows, the most energy-demanding process is the membrane scouring by
- 2 biogas sparging, which represents about 58% of the total energy requirements. This
- 3 highlights the importance of optimising the filtration performance in each membrane
- 4 filtration process to enhance the feasibility of the proposed platform.
- 5 As Figure 3b, the operating and maintenance costs of the membrane units are the most
- 6 relevant items, reaching about the 78 % of total cost. It is important to highlight that
- 7 capital expenses were not considered in this study. It also must be pointed out that
- 8 nutrients recovery as commercial products, such as ammonium sulphate, struvite and
- 9 amendments, were not considered in this economic study, that could have positive
- 10 impacts on economic balance.
- 11 Energy balance calculations resulted in a WRRF total energy demand of about 0.52 kWh
- per m³ of treated water, from which 16% was related to heat requirements (i.e. 0.08 kWh
- per m³). When biogas is used for energy recovery in a CHP system, 0.44 kWh per m³ can
- be recovered as electricity. Hence, WRRF electricity requirements would be covered by
- 15 the electricity recovered from the produced biogas. Regarding heat requirements, the
- recovered heat represents 0.03 kWh per m³. Thus, WRRF heat energy requirements would
- be 0.053 kWh per m³. Finally, this energy recovery entails a cost reduction from €0.097
- 18 to €0.036 per m³ of treated water, which corresponds to a saving cost of €0.061 per m³.
- 19 The remaining costs are mainly due to membrane maintenance expenses.



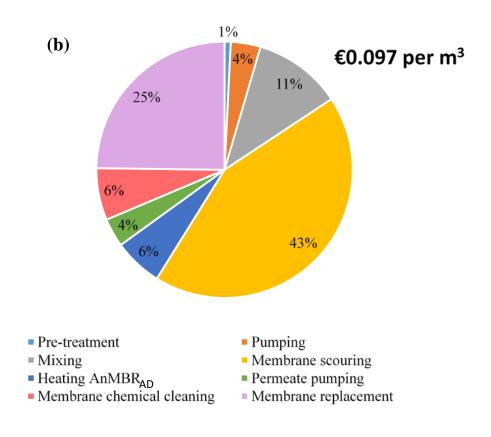


Figure 3. (a) Energy and (b) economic balance of the WRRF platform

1 *3.5.Overall discussion*

- 2 The proposed WRRF pursues the recovery of reclaimed water, nutrients and energy
- 3 contained in the wastewater. Conventional WWTPs are energy-intensive platforms where
- 4 energy consumption for aeration represents above 40% of total WWTP energy demand
- 5 (see e.g. Sid et al., 2017).
- 6 In the last few years, the process of activated sludge has been modified in order to reduce
- 7 operating costs and to add technologies capable of carrying out the recovery of nutrients,
- 8 mainly phosphorus. Sid et al. (2017) implemented a control of the aeration to decrease
- 9 the cost of nitrification-denitrification processes (e.g. Ludzack-Ettinger process)
- achieving a reduction of 10%, resulting in an overall cost of €0.1254 per m³. In contrast,
- 11 the energy consumption of the proposed WRRF is €0.0311 per m³.
- 12 Regarding the recovery of phosphorus, slight modifications have been made in
- 13 conventional WWTPs in order to replace chemical removal by struvite precipitation
- 14 (Martí et al., 2017). Although this WRRF was designed to recover phosphorus, the
- uncontrolled precipitation of phosphate prevented the application of struvite precipitation
- processes. Nevertheless, an important fraction of phosphorus was recovered as biosolids
- 17 $(121.0 \pm 7.2 \text{ mg P} \cdot \text{L}^{-1}).$
- 18 Nitrification-denitrification is the traditional process to remove nitrogen in activated
- 19 sludge. Besides being an energy-intensive process, it does not allow the recovery of
- 20 nitrogen since it is released to the atmosphere in gaseous form. New technologies for
- 21 nitrogen recovery have appeared but their coupling to a conventional WWTP is not easy,
- 22 e.g. zeolites or electrochemical techniques. Regarding this, ammonia absorption-
- 23 desorption is an alternative method, reaching removal values between 65-75% (Morales

- 1 et al., 2013). In contrast, this WRRF layout allows to obtain considerable levels of
- 2 nitrogen removal (100%) and recovery, specifically 83% through membrane contactor.
- 3 The proposed WRRF implies a new configuration for the holistic approach of the
- 4 wastewater treatment. Although it presents some advantages versus traditional WWTPs
- 5 such as less energy costs and resource recovery, further research is needed to optimise it.
- 6 Other researchers such as Batstone et al. (2015) have simulated new WRRF platforms
- 7 looking to move forward with the paradigm shift. In any case, real data from these novel
- 8 WRRF configurations are still lacking.

10

4. Conclusions

- 11 An alternative WRRF platform for sewage treatment has been presented and its
- performance has been evaluated. Results constitute the proof-of-concept of the system,
- exploring the feasibility of combining anaerobic technology, microalgae cultivation and
- 14 membrane technology for resource recovery from sewage. The implementation of this
- 15 alternative treatment solution allowed:
- The production of a stream with a content of 45 mg COD·L⁻¹, 14.9 mg N·L⁻¹ and 0.5
- 17 mg P·L⁻¹ which can be discharged to the environmental water flows according to
- 18 European Directive 91/271/CEE.
- Different resources are recovery in the WRRF platform such as ammonia
- sulphate (28% of the incoming N) and biosolids with a content of 30% of the incoming
- 21 N and 34% of the incoming P.

- 1 The energy demand of the WRRF was 0.52 kWh per m³ of water treated and the
- 2 energy production of the WRRF was 0.47 kWh per m³. WRRF total energy requirements
- 3 could be covered, which represents a cost saving of €0.061 per m³.

5

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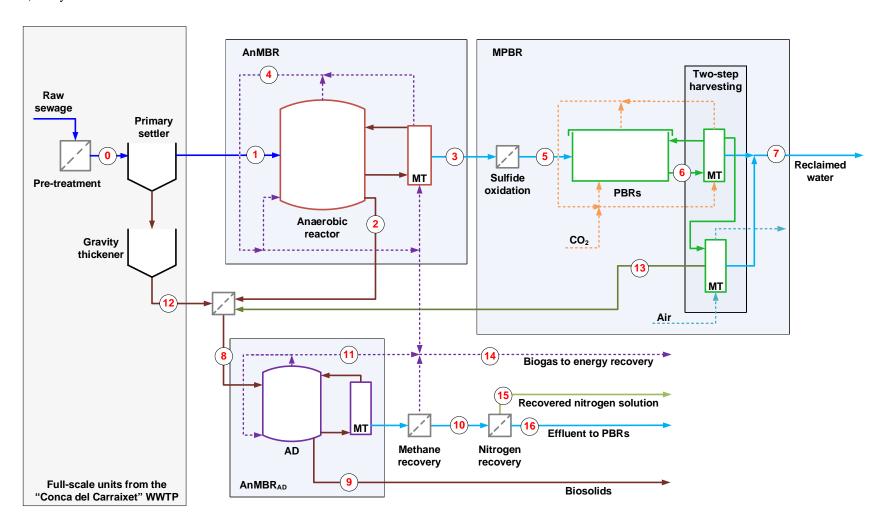
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Appendix 1. Extended mass balances.

1) Layout of the WRRF.



2) COD mass balance.

Sampling point	Liquid streams flow- rate (L·d ⁻¹)	Biogas streams flow-rate (L STP·d ⁻¹)	COD (mg COD·L ⁻¹)	CH ₄ (%)	COD (g COD·d ⁻¹)	COD (g COD·m³ treated)
0 (*)	300.79	-	444	-	133.55	444
1	298.14	-	244	-	72.75	242
2	4.81	-	4902	-	23.75	79
3	293.33	-	144	-	42.24	140
5	293.33	-	81	-	23.76	79
7 (**)	284.12	-	45	-	12.79	43
8	16.67	-	16737	-	279.01	928
9 (**)	7.17	-	17310	-	124.18	413
10	9.50	-	1169	-	11.10	37
11	-	85.50	-	62.0%	151.37	503
12	2.65	-	21	-	62.77	209
13	9.21	-	18897	-	174.04	579
14 (**)	-	85.50	-	62.0%	152.07	506
16	9,50	-	1169	-	11.10	37

Average of chemical oxygen demand (COD) concentration in the sampling points of the proposed WRRF (see Figure 1)

^(*) WRRF platform inflow stream

^(**) WRRF platform outflow streams

3) TN mass balance.

Sampling point	Liquid streams flow-rate (L·d ⁻¹)	TN (mg N·L·¹)	TN (g TN·d ⁻¹)	TN (g TN·m³ treated)
0 (*)	300.79	49.9	15.01	49.90
1	298.14	45.6	13.58	45.15
2	4.81	332	1.60	5.32
3	293.33	44.9	13.17	43.78
5	293.33	44.8	13.14	43.68
7 (**)	284.12	14.9	4.24	14.08
8	16.67	547	9.12	30.32
9 (**)	7.17	650	4.66	15.50
10	9.50	508	4.82	16.12
12	2.65	660	1.75	5.82
13	9.21	705	6.49	21.58
15 (**)	-	-	4.00	13.30
16	9,50	0	0.00	0.00
Stripping losses at recovery (***)	-	-	0.82	2.72
Nitrogen losses at WRRF platform	-	-	1.30	4.30

Average of total nitrogen (TN) concentration in the sampling points of the proposed WRRF (see Figure 1)

^(*) WRRF platform inflow stream

^(**) WRRF platform outflow streams

^(***) Estimated WRRF platform outflow stream.

4) TP mass balance.

Sampling point	Liquid streams flow-rate (L·d ⁻¹)	TP (mg P·L·¹)	TP (g TP·d·¹)	TP (g TP·m ⁻³ treated)
0 (*)	300.79	8.3	2.50	8.30
1	298.14	5.7	1.70	5.65
2	4.81	85.0	0.41	1.36
3	293.33	3.9	1.14	3.79
5	293.33	3.8	1.11	3.69
7 (**)	284.12	0.5	0.14	0.47
8	16.67	145.9	2.43	8.08
9 (**)	7.17	121.0	0.87	2.89
10	9.50	17.7	0.17	0.56
12	2.65	385	1.02	3.39
13	9.21	91.4	0.84	2.79
16	9,50	17.7	0.17	0.56
recipitation losses t AnMBR _{AD} (***)	-	-	1.40	4.64

Average of total phosphorus (TP) concentration in the sampling points of the proposed WRRF (see Figure 1)

^(*) WRRF platform inflow stream

^(**) WRRF platform outflow streams

^(***) Estimated.

Seco et al. Resource recovery from sewage through an innovative anaerobic-based water resource recovery facility (WRRF)

Appendix 2. Assumptions for energy and economic balance calculations.

The energy requirements related to pumps and blowers (adiabatic compression), were calculated by means of the corresponding theoretical equations (Eqs. (2.1) - (2.3)).

$$P_B(W) = \frac{M \cdot R \cdot T_{gas}}{(\gamma - 1) \cdot \eta_B} \left[\left(\frac{p_I}{p_A} \right)^{\frac{\gamma - 1}{\gamma}} - 1 \right]$$
 (Eq. 2.1)

where P_B is the blower power requirement, M (mol s⁻¹) is the molar flow rate of biogas, R (J mol⁻¹ K⁻¹) is the the universal constant of gases, γ is the heat capacity ratio, η_B is the blower efficiency, and p_I and p_A (Pa) are the impulsion and aspiration pressure, respectively. A blower efficiency of 0.75 was considered.

$$P_{g}(W) = q_{I} \cdot \rho_{liquor} \cdot g$$

$$\cdot \frac{\left\{ \left[\left(\frac{(L + L_{eq}) \cdot f \cdot v^{2}}{D \cdot 2 \cdot g} \right)_{A} + \left(\frac{(L + L_{eq}) \cdot f \cdot v^{2}}{D \cdot 2 \cdot g} \right)_{I} \right] + [z_{1} - z_{2}] \right\}}{\eta_{g}} \qquad (Eq. 2.2)$$

where P_g is the power requirement by general pump, q_I (m³ s⁻¹) is the impulsion volumetric flow rate, ρ_{liquor} (kg m⁻³) is the liquor density, g (m s⁻²) is the acceleration of gravity, L and L_{eq} (m) are the pipe length and equivalent pipe length, respectively, v (m s⁻¹) is the liquor velocity, f is the friction factor, D is the pipe diameter, (z₁-z₂) (m) is the height difference, and η_g is the pump efficiency. A general pump efficiency of 0.6 was considered.

$$P_P(W) = \frac{q_P \cdot TMP_P}{n_P} \tag{Eq. 2.3}$$

where P_P is the permeate pump power requirement during filtration, q_P (m³ s⁻¹) is the pump volumetric flow rate, TMP_P (Pa) is the transmembrane pressure in the filtration process, and η_P is the permeate pump efficiency. A permeate pump efficiency of 0.6 was considered.

On the other hand, the electricity production from the generated biogas (W_{Biogas}) was estimated using the following equation:

$$W_{Biogas}(kW) = \frac{V_{Biogas} \cdot \% \ CH_4 \cdot CV_{CH4} \cdot \eta_{CHP}}{1000 \cdot 24 \cdot 3600}$$
 (Eq. 2.4)

where V_{Biogas} (L d⁻¹) is the biogas volume, %CH₄ is the methane content in collected biogas, CV_{CH4} (kJ m⁻³) is the methane calorific power, and η_{CHP} is the methane electricity

Seco et al. Resource recovery from sewage through an innovative anaerobic-based water resource recovery facility (WRRF)

conversion efficiency by the CHP system. A biogas electricity conversion efficiency of 0.35 was considered.

Regarding heat energy demands in the AnMBR_{AD} reactor, total heat (Q_{TOT}) demand was calculated as the sum of the energy required to heat the inflow (Q_{Req} , Eq. 2.5), and the heat dissipated though the walls of the reactor (Q_{Diss} , Eq. 2.6).

$$Q_{Req}\left(\frac{kcal}{h}\right) = c_P \cdot q \cdot \rho_{sludge} \cdot \left(T_{fixed} - T_{inflow}\right) \tag{Eq. 2.5}$$

where c_P (kcal kg⁻¹ K⁻¹) is the specific heat, q (m³ h⁻¹) is the intlet flow rate, ρ_{sludge} (kg m⁻³) is the sludge density and (T_{fixed} - T_{inflow}) (K) is the difference in temperature between the inflow, and the temperature fixed in the biological process.

$$Q_{Diss}\left(\frac{kcal}{h}\right) = U \cdot S \cdot \Delta T \tag{Eq. 2.6}$$

where U (kcal h^{-1} m⁻² K⁻¹) is the overall heat transfer coefficient obtained by Eq. 2.7, S (m²) is the reactor surface, and ΔT (K) is the difference in temperature between reactor inside and outside.

$$U\left(\frac{kcal}{h \cdot m^2 \cdot K}\right) = \frac{1}{\frac{\delta_{reactor}}{K_{reactor}} + \frac{1}{h_{air}}}$$
 (Eq. 2.7)

where $\delta_{reactor}$ (m) is the reactor thickness, $K_{reactor}$ (kcal·h⁻¹ m⁻¹ K⁻¹) is the reactor material conductivity, and h_{air} (kcal·h⁻¹ m⁻¹ K⁻¹) is the air convective heat transfer coefficient.

Heat recovery from methane capture was calculated using the following equation:

$$Q_{biogas}\left(\frac{Kcal}{h}\right)$$

$$=\frac{V_{biogas}\cdot(\%CH_{4}\cdot CV_{CH4})\cdot\%heat\ efficiency\ CHP}{1000\cdot24\cdot4.187}$$

$$\cdot\%Heat\ exchanger \qquad (Eq. 2.8)$$

where V_{biogas} is the biogas volume (L·d⁻¹), %CH₄ is the methane richness (%), CV_{CH4} is the methane calorific power (kJ·m⁻³), %heat efficiency CHP is the heat efficiency of the CHP system (%), and %heat exchanger is the heat exchanger efficiency (%). A biogas heat conversion efficiency of 45% was considered. A heat exchanger efficiency of 65% was considered.

Table S2.1 shows the unit costs used to calculate the operating and maintenance expenses of the proposed treatment systems. A useful membrane lifetime of 20, 7 and 7 years was assumed for AnMBR, MPBR and AnMBR_{AD} membranes, according to the total chlorine contact specified by the manufacturer (see Table S2.1) and the membrane chemical cleaning frequency laid down. The energy requirement related to the pre-treatment (rotofilter) was acquired from a catalogue for full-scale implementation (AGUA TÉCNICA, 2012).

Table S2.1. Unit costs used to evaluate operating and maintenance expenses in the proposed WRRF treatment scheme.

Ultrafiltration hollow-fibre membrane, (maximum chloride contact of 500,000 ppm·h cumulative), €per m ²	35
Energy, €per kWh	0.07
Sodium hypochlorite, (NaOCl Cl active 5% PRS-CODEX), €per L	11
Citric acid (Citric acid 1-hidrate PRS-CODEX), €per kg	23.6

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