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Long-term operation of a pilot-scale anaerobic membrane bioreactor (AnMBR) treating

high salinity low loaded municipal wastewater in real environment

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1 Abstract

2 Long term operation of an anaerobic membrane bioreactor (AnMBR) treating municipal wastewater was investigated in a real seawater intrusion spot in Falconara Marittima (Central 3 Italy) on the Adriatic coastline. Changes in biological conversion and system stability were 4 determined with respect to varying organic loading rate (OLR) and high salinity conditions. At 5 an OLR of 1 kgCOD m³⁻¹d⁻¹, biogas production was around 0.39 ± 0.2 L d⁻¹. The increase of 6 the OLR to 2 kgCOD $m^{3-1}d^{-1}$ resulted in the increase of biogas production to 2.8 ± 1.5 L d^{-1} 7 (with 33.6% \pm 10.5% of CH₄) with methanol addition and to 4.11 \pm 3.1 L[·]d⁻¹ (with 29.7% \pm 8 9 11.8% of CH₄) with fermented cellulosic sludge addition. COD removal by the AnMBR was $83\% \pm 1\%$ when the effluent COD concentration was below 100 mg O₂·L⁻¹. The addition of the 10 fermented sludge affected the membrane operation; significant fouling occurred after long-term 11 filtration, where the trans-membrane pressure (TMP) reached up to 500 mbar. Citric acid 12 13 solution was applied to remove scalants and the TMP reached the initial value. High saline conditions of 1500 mgCl⁻L⁻¹ adversely affected the biogas production without deteriorating the 14 15 membrane operation. The treated effluent met the EU quality standards of the D.M. 185/2003 and the new European Commission Resolution for reuse in agriculture. 16

Keywords: AnMBR; membrane fouling; municipal wastewater; fermentation; salinity;
cellulosic sludge

19

20 Nomenclature

- 21 AD: Anaerobic Digestion
- 22 AeMBR: Aerobic Membrane Bioreactor
- 23 AnMBR: Anaerobic Membrane Bioreactor
- 24 CAS: Conventional Activated Sludge
- 25 COD: Chemical Oxygen Demand

- 1 DO: Dissolved Oxygen
- 2 EC: Electrical Conductivity
- 3 EPS: Extracellular Polymeric Substances
- 4 HRT: Hydraulic Retention Time
- 5 MLSS: Mixed Liquor Suspended Solids
- 6 MWT: Municipal Wastewater Treatment
- 7 OLR: Organic Loading Rate
- 8 ORP: Oxidation Reduction Potential
- 9 PE: Population Equivalent
- 10 sCOD: Soluble Chemical Oxygen Demand
- 11 SMA: Specific Methanogenic Activity
- 12 SRT: Sludge Retention Time
- 13 SuMBR: Submerged Membrane Bioreactor
- 14 tCOD: Total Chemical Oxygen Demand
- 15 TKN: Total Kejdahl Nitrogen
- 16 TMP: Trans Membrane Pressure
- 17 TN: Total Nitrogen
- 18 TP: Total Phosphorus
- 19 TS: Total Solids
- 20 TSS: Total Suspended Solids
- 21 UASB: Upflow Anaerobic Sludge Blanket
- 22 VFA: Volatile Fatty Acid
- 23 VS: Volatile Solids
- 24 WWTP: Wastewater Treatment Plant
- 25

1 **1. Introduction**

2 Anaerobic treatment in high-rate bioreactors has increased in number of applications during municipal wastewater treatment (MWT) in the last decade while presenting an advanced 3 technology for environmental protection and resource preservation [1,2]. The combination of 4 5 membrane and an anaerobic bioreactor (Anaerobic membrane bioreactor (AnMBR)) paved the way for a sustainable wastewater treatment with complete biomass retention, and with 6 7 additional advantages such as less sludge production, high quality effluent and net energy production as the organic matter is converted into high-value products (volatile fatty acids 8 9 (VFAs)) and energy in the form of biogas [3,4]. These advantages of anaerobic treatment 10 systems result in a decrease in operational costs compared to conventional wastewater treatment 11 plants (WWTPs) that often include aerobic processes (i.e. conventional activated sludge (CAS) or aerobic membrane bioreactor (AeMBR)) [5-7]. 12

13 Sludge bed-based technologies, such as upflow anaerobic sludge blanket (UASB) reactors, have been widely used for MWT in full-scale WWTPs especially in tropical regions [8-10]. The 14 15 AnMBRs, on the other hand, have been mainly applied for the treatment of high strength industrial wastewater [1,11]. Currently, emphasis has been given to adapt AnMBRs in MWT 16 17 to simultaneously recover energy and clean water [12,13]. In fact, AnMBRs are suitable for the 18 treatment of low-loaded wastewater due to the complete retention of slow-growing methanogens and thus have the potential to operate at higher organic loading rates (OLR) 19 [14,15]. Applied OLRs in AnMBRs range from 0.3 kgCOD^{m³⁻¹d⁻¹} to 12.5 kgCOD^{m³⁻¹d⁻¹} 20 during the treatment of municipal wastewater [4,16]. From this point of view, further research 21 is still required to optimize the application of AnMBRs in MWT with respect to different 22 feeding characteristics that can help to assess the overall performance of AnMBRs at varying 23 OLRs. 24

The application of AnMBRs is coastal regions is also another point that lacks sufficient 1 2 information in the literature for a successful WWTP operation. In coastal regions, variable 3 salinity of wastewater occurs due to seawater infiltration to sewers or introduction of saline water from industrial processes such as seafood and cheese production [12,17]. In general, the 4 salinity effect in anaerobic processes can cause two main critical operational problems. Firstly, 5 increased salinity results in deterioration of membrane filtration and fouling aspects due also to 6 7 the decrease of the biomass particle size. For instance, salinity increase from 8 20 gNa⁺.L⁻ to 20 gNa⁺.L⁻¹ was accompanied by the increase of the transmembrane pressure (TMP) up to 350 8 mbar, while a ten-fold reduction in biomass particle size resulted in a filtration resistance 9 10 increase [18]. Similarly, Yurtsever et al. [19] indicated that salinity induced large molecules, to 11 be detected as foulants in gel/cake layer; they may originate from biomass loosely bound extracellular polymeric substances (EPS). Secondly, saline conditions can suppress microbial 12 13 growth and cause the disintegration of flocs and granules that further lead prominent biomass wash-out affecting the sludge granulation [20]. 14

15 The advantages of AnMBRs in MWT are evident with respect to the necessity of low-cost energy technologies in WWTPs; however, long-term operational experiences in coastal regions 16 17 are not fully recognized. The main motivation of this study was therefore to investigate the 18 treatment efficiency of low-loaded wastewater in combined sewers by AnMBR and to 19 determine the optimal operating conditions with respect to varying OLRs by using fermented cellulosic sludge rich in VFAs. In this aspect, a pilot-scale UASB coupled with anaerobic 20 21 ultrafiltration membranes was operated with real influent in a coastal area that is considered hotspot for seawater intrusion, Falconara Marittima WWTP, Italy. The effect of seawater 22 intrusion in biological activity and membrane filtration of municipal wastewater in coastal areas 23 was further elaborated in the specifically-designed integrated treatment scheme. Since the 24

majority of first generation MBRs was implemented in northern Europe, the presented results
may offer options to a critical part of the MBR market.

3 2. Materials and methods

4 2.1. Full scale plant in Falconara area

The pilot plant is located in the WWTP of Falconara Marittima (coastal area hotspot for 5 seawater intrusion) (Figure 1) and fed with the real influent of the full-scale plant. The WWTP 6 7 of Falconara Marittima (Italy) has a design treatment capacity of 80,000 PE with nominal influent flow rate of 30,000 m³·d⁻¹. After screening, degritting and primary settling, the 8 wastewater is biologically treated with CAS that is operated with two identical parallel lines 9 10 applying the Modified Ludzack Ettinger scheme. The total volume of the biological compartment is 13,700 m³, divided into 8,860 m³ and 4,900 m³ for denitrification and 11 nitrification compartments, respectively. The aerated compartments are equipped 12 by ceramic fine bubble diffusers. The Falconara Marittima WWTP is continuously monitored 13 by online sensors (Dissolved Oxygen (DO); temperature; mixed liquor suspended solids 14 15 (MLSS) and oxidation reduction potential (ORP)) and magnetic flow meters (influent, effluent, recirculation and waste sludge). The average sludge retention time (SRT) is 16 10 days and the sludge recycle ratio (Q_{sludge recycled}/Q_{influent}) is 0.5. 17

The Falconara Marittima WWTP is controlled by the European Environment Agency (www.eea.europa.eu, 2017), since it is located in a coastal area. Since it is a hotspot for infiltrations from groundwater and marine intrusions, low-loading wastewater occurs in the WWTP influent during the dry weather conditions. The average quality of the raw influent is given in **Table 1**; where the values reveal that the influent is characterized by high chloride concentration around 334 ± 236 mg·L⁻¹ as a result of the seawater intrusion.

24 2.2. Pilot plant in Falconara Marittima

Following the preliminary treatment (screening, degritting and oils removal), pretreated influent from Falconara WWTP was sent to a pilot-scale UASB coupled with an anaerobic ultrafiltration membrane. To evaluate the long-term stability period, the experimental work was conducted for 480 days with real wastewater influent. As shown in **Figure 2**, different phases were designed by increasing the influent organic loading testing the following configurations: a) urban wastewater, b) co-treatment of urban wastewater and methanol, c) co-treatment of urban wastewater and fermentation liquid.

A steady influent flow rate of about 3 L^{-h⁻¹} of wastewater and the external addition of methanol
and fermented sludge were achieved by peristaltic pumps (Watson-Marlow, UK). The first one
was able to guarantee a flow rate of 15 L^{-h⁻¹}, the second and the third was 3 L^{-h⁻¹}-10 L^{-h⁻¹} and
15 L^{-h⁻¹}, respectively.

The UASB was a cylindrical Plexiglas reactor (16 L) with an internal diameter of 15 cm and a 12 13 total height of 136 cm. The reactor was divided into two compartments: the first was the real reaction chamber at the bottom (85 cm, 12.4 L), while the second, on the top, was a tri-phase 14 15 separator (GLS) with 21.9 cm height and was connected to a hydraulic guard which created the appropriate backpressure for the biogas release. The temperature of the UASB reactor was kept 16 constant (30°C) by applying internal and external windings with hot water at 45°C. The 17 18 produced biogas was measured by a milligas counter (Ritter, Germany). The hydraulic retention time (HRT) was maintained at 5-6 h. The up-flow velocity of the UASB reactor was maintained 19 at 1 m⁻¹. The UASB reactor was inoculated with the sludge obtained from a paper mill WWTP 20 21 in Castelfranco Veneto (Italy).

The UASB effluent was collected in a mixed reactor equipped with pH and temperature probes
and it was partially recirculated in the UASB reactor (internal recirculation) and partially sent
by gravity to the second unit (anaerobic ultrafiltration tank). The performance of anaerobic
process was monitored throughout the experimental period with respect to OLR, indicator α,

specific methanogenic activity (SMA), upflow speed and biogas production. The UASB was
followed by anaerobic hollow-fiber ultrafiltration membrane (PURON® Koch membrane
system, with 0.03 µm of nominal pore-size, a total nominal surface 0.5 m², 0.25 m height)
installed in a Plexiglas reactor (0.29 m x 0.7 m x 0.39 m) and equipped with level and TMP
sensors.

6 2.3. Experimental periods for UASB and AnMBR

Different OLRs (ranging from 1 kgCOD $m^{3-1}d^{-1}$ to 2 kgCOD $m^{3-1}d^{-1}$) were studied and 7 controlled based on the influent flow. All experimental periods, applied configurations and 8 operational parameters are summarized in Table 2. In Period 1 only raw wastewater was treated 9 by UASB at 30 °C, while the co-treatment of raw wastewater and methanol was tested in Period 10 2. Period 2 was divided in 3 sub-periods: In Period 2.1, mixture of raw wastewater and methanol 11 was treated only by UASB at 30 °C. In Period 2.2, the co-treatment of raw wastewater and 12 13 methanol was conducted at 30°C UASB + ambient AnMBR; whereas, the same configuration of Period 2.2 was applied in Period 2.3 except that the temperature of AnMBR was also kept at 14 15 30 °C. In Period 3, the wastewater was co-treated with the supernatant of fermented cellulosic sludge in UASB + AnMBR. During Period 3, the raw wastewater was filtered by dynamic 16 rotating primary unit (SALSNES FS1000) to recover cellulosic sludge [21]. The separated 17 sludge was then sent to anaerobic fermentation reactor (1400 L) operating at 30°C in 18 uncontrolled pH. The fermented flow was dewatered (BABY 2 PIERALISI) and the liquid 19 supernatant was used to increase the OLR of UASB reactor up to 2 kgCOD^{·m³⁻¹d⁻¹}. Finally, 20 during the fermentation liquid co-treatment, NaCl solution was added in the UASB reactor to 21 simulate high saline conditions in Period 4. Chloride concentration was increased gradually 22 from 200 mg L^{-1} to 500 mg L^{-1} during first 50 days and then to 1500 mg L^{-1} after 50 days and 23 the maximum of 2200 mg·L⁻¹ at the end. The main characteristics of the influents are reported 24 in Table 3. 25

1 The performance of the system was investigated in terms of organic content removal and biogas 2 production with respect to above-listed configurations (Periods 1-2-3-4). The indicator α , that 3 is defined as the ratio of partial alkalinity over total alkalinity, was measured to verify the 4 stability of the biological process. An upflow velocity of 0.7 m⁻¹ to 1 m⁻¹ was maintained to 5 keep the sludge blanket in suspension.

6 2.4. Functional characterization of AnMBR

7 Preliminary tests were carried out to optimize the AnMBR membrane pilot-scale before being coupled with UASB. The start-up conditions of the AnMBR are given in **E-Supplementary** 8 material. The critical flux of the AnMBR was studied through step-flux method and 6 steps of 9 10 10 minutes have been tested for 60 minutes. The pump was set up in order to increase the flux rate and TMP was recorded. The critical flux was 12-14 L^{-m²⁻¹h⁻¹} and for all the experimental 11 periods membrane flux was maintained below this value. Moreover, the effect of the solid 12 concentration range (0-10 mgMLSS·L⁻¹, 30-50 mgMLSS·L⁻¹, 80-100 mgMLSS·L⁻¹ and 300 13 mgMLSS[·]L⁻¹) in the membrane fouling was assessed. The effect of gas sparging in the fouling 14 15 rate was evaluated at different solid concentrations with and without gas-sparging to investigate the effect of gas bubbles in terms of membrane fouling and operative TMP values. The gas-16 sparging method adopted in these tests, using nitrogen gas (N₂) for 10 seconds off (gas off) and 17 10 seconds on (gas on), with a specific flow rate value of 2 m³·m²⁻¹h⁻¹ [22]. Moreover, gas-18 sparging frequency was studied. The results showed that the increase of the gas sparging 19 frequency increased the percentage of degassing methane; and the degassing methane from gas-20 21 sparging could be recovered (see E-Supplementary material).

For each experimental set, the temperature was measured at the beginning and at the end of the test. The temperature was normalized at 20°C using the Arrhenius equation. For each flux (J) ranging from 6 LMH to 22 LMH, the average TMP (TMP_{ave}) and the slope (dTMP/dt) were calculated (**E-Supplementary material**). The TMP reached up to 0.79 mbar⁻¹ at MLSS concentration of 300 mgMLSS·L⁻¹ without gas sparging. Differently, for the same MLSS
concentration, the TMP was maintained below 0.1 mbar·min⁻¹ by switching on the gas sparging.
Therefore, the gas sparging decreasing the fouling rate independently from the MLSS
concentrations and the preliminary tested gas sparging durations were adopted for the long
operating periods.

Granulometric characterizations of the influent of AnMBR was conducted to characterize the
particle size of influent solids. Accumulative volume percent was calculated. Diameters d₅₀ and
d₉₀ of particles were found to be 14 μm and 58 μm, respectively.

9 Membrane cleaning was performed by adding hypochlorite solution at 14% w/v (200 ppm) 10 every 45 days to remove organic fouling of the membrane; while NaOCl at a concentration of 11 $1000 \text{ mg} \text{L}^{-1}$ or citric acid (C₆H₈O₇) at 1000 mg L⁻¹ was used to restore the initial permeability 12 of the membrane. Permeability tests were carried out with tap water and TMP values were 13 measured at different steps of permeation flow rate. Following the preliminary tests, the 14 AnMBR reactor was fed with UASB effluent in different Periods (**Table 2**) and coupled with 15 UASB as mentioned earlier.

16 2.5. Analytical methods

17 Standard analyses were conducted in the influent flow, the UASB effluent and the membrane 18 permeate twice a week. All the samples were analyzed in terms of pH, chemical oxygen demand (COD), total Kejdahl nitrogen (TKN), ammonia nitrogen (NH4⁺-N), soluble COD 19 (sCOD), nitrate nitrogen (NO₃-N) and nitrite nitrogen (NO₂-N) according to Standard Methods 20 21 [23]. The sCOD was measured in the filtrate obtained after the filtration of the sample through 0.45 µm Whatman membrane filters. NO₂-N, NO₃-N were measured by ion chromatography 22 (Dionex DX120) in samples that were first filtered through 0.45 µm Whatman membrane 23 filters. 24

Moreover, in each period, anaerobic biomass was sampled from UASB reactor to investigate 1 2 SMA (data not shown) at different OLRs according to the experimental method reported by Hussain et al. [24]. Acetate (solution of 2 gCOD L^{-1} , with a ratio VS/COD of 2) was used and 3 its degree of conversion into methane was normalized considering the volatile solids (VS) and 4 expressed in m³CH₄·kgVS⁻¹d⁻¹. The CH₄ content of the biogas was analyzed by a Brüel and 5 Kjaer Multi-gas Monitor Type 1302, based on photoacoustic spectroscopy. During the periods 6 7 with methanol and fermentation liquid addition, extracellular polymeric substances (EPS) 8 were also analyzed according to the method found in Zhang et al. [25].

9 2.6. Statistical analysis

10 Principal component analysis (PCA) was applied to the dataset to identify the relationships between the applied operating conditions and system performance. The obtained loadings of 11 the variables in each principal component (see E-Supplementary material) mapped their 12 13 relationship with the respective principal component (PC). The scores of the principal components mapped the different samples in the new dimensional space of the principal 14 15 components that simplified the investigation of the different relationships between the variables. The first two principal components (PC1 and PC2) were then selected for further 16 17 interpretation of the results. More information on the PCA can be found in [26].

18 **3. Results and discussion**

19 3.1. Characterization and variability of the influent

The main characteristics of the influent for each operational period is given in **Table 3**. pH of the influent remained almost stable (7.5 - 7.8). The total alkalinity of the influent ranged from $281 \pm 92 \text{ mgCaCO}_3 \cdot \text{L}^{-1}$ to $526 \pm 110 \text{ mgCaCO}_3 \cdot \text{L}^{-1}$, with the highest concentration observed in Period 3. EC was $1464 \pm 153 \text{ ms/cm}$ and $1467 \pm 380 \text{ ms/cm}$ in Period 1 and Period 2 respectively, while the addition the fermentation liquid increased the EC to $1817 \pm 350 \text{ ms/cm}^{-1}$ ¹ in Period 3. Saline conditions in Period 4 increased further the EC up to $2787 \pm 1300 \text{ ms/cm}^{-1}$

¹. Cl⁻ concentration of the influent was $282 \pm 126 \text{ mg} \cdot \text{L}^{-1}$, $304 \pm 300 \text{ mg} \cdot \text{L}^{-1}$, $393 \pm 377 \text{ mg} \cdot \text{L}^{-1}$ 1 and $1100 \pm 618 \text{ mg L}^{-1}$ during Periods 1, 2, 3 and 4, respectively. COD concentrations were 207 2 \pm 73 mg L⁻¹, 388 \pm 80 mg L⁻¹, 375 \pm 148 mg L⁻¹ and 550 \pm 330 mg L⁻¹ in Periods 1,2,3 and 4, 3 respectively. Meanwhile, the average sCOD concentration was 50 mg⁻L⁻¹ in Period 1 and in the 4 range of 242-297 mg·L⁻¹ in Periods 2, 3 and 4. The addition of methanol and fermentation liquid 5 6 increased the soluble organic fraction of the influent. On the other hand, the soluble organic 7 fraction in the influent decreased due to the saline conditions occurred in Period 4. NH4⁺-N concentration was $22\pm6 \text{ mg L}^{-1}$, $20.5\pm6 \text{ mg L}^{-1}$, $61\pm33 \text{ mg L}^{-1}$ and $30\pm10 \text{ mg L}^{-1}$ in Periods 1, 2, 8 9 3 and 4, respectively. The highest influent TP and PO₄-P concentrations were observed in Period 3, while the highest TSS concentration was recorded in Period 4. The results are in line 10 with existing literature that were reported for typical real municipal wastewater, except for 11 parameters such as EC, TSS, Cl⁻ and SO₄²⁻ that were measured comparatively higher due to 12 seawater intrusion in the WWTP of Falconara Marittima [3,5,27]. 13

14 3.2. Start-up period of UASB

The UASB reactor was inoculated with anaerobic sludge (granular sludge TS = 2.72%, VS/TS 15 = 74%; flocculent sludge TS = 1.42%, VS/TS = 60%) taken from a paper mill WWTP in 16 17 Castelfranco Veneto, Italy. The temperature of the UASB was increased gradually to 30°C after 10 days of operation period. The reactor operated for 5 months at 30°C with OLR value of 1.05 18 ± 0.4 kgCOD m³⁻¹d⁻¹. The flow rate was maintained between at 3.38 ± 0.6 L h⁻¹. Meanwhile, α 19 was between 0.12 and 0.43, which indicated a stable biological process. The average biogas 20 production was 0.39 ± 0.2 Lbiogas d⁻¹. The COD and TSS removal efficiencies were 63% and 21 84%, respectively. In addition, 86% of P and N were released during the start-up period of 22 UASB. 23

24 **3.3. Effect of OLR in the system performance**

Following the start-up period, the UASB was first fed with the mixture of municipal wastewater 1 and methanol (as the external C source) and the OLR was increased to 2.1 ± 0.6 kgCOD m³⁻¹d⁻¹ 2 ¹ (Period 2). The flow rate was maintained at 2.98 \pm 0.3 L^{·h-1}. The variations in OLR and α 3 4 value throughout the operation period are given in **Figure 3a** and **Figure 3b**, respectively. α value remained almost stable in the beginning of Period 2, and then tended to increase up to 5 0.65. while 86% P and 88% N were released. The average biogas production increased to 2.8 \pm 6 1.5 Lbiogas d^{-1} with 33.6% \pm 10.5% of CH₄ in Period 2. The COD and TSS removal efficiencies 7 of the UASB was 70% and 48% respectively while the application of the AnMBR increased 8 the average COD and TSS removal to 85%, and > 99.99%. On the other hand, the release of P 9 10 and N in the UASB was 86% P and 88% N and then slightly decreased to 76% and 83% in the integrated UASB+AnMBR configuration, respectively. 11

In Period 3, α value was between 0.16 and 0.52 with an average of 0.29 \pm 0.1. The addition of 12 13 the fermentation liquid in the influent resulted in a peak in the biogas production; while only 9% CH₄ was measured indicating excess CO₂ production via fermentation (Figure 4). The 14 15 biogas production started to increase gradually together with the CH₄ content of the biogas. Hence, up to 10.25 Lbiogas d⁻¹ was generated with 51.9% of CH₄ (average of 4.11 ± 3.1 16 Lbiogas d^{-1} with 29.7% \pm 11.8% of CH₄). The addition of fermentation liquid as the external 17 18 carbon source increased the biogas production without affecting the overall CH₄ content of the biogas. In Period 3, the COD removal efficiencies were 42% and 83% in UASB and 19 UASB+AnMBR, respectively. The TSS removal efficiencies in UASB and UASB+AnMBR 20 were 38% and 100%, respectively, while P and N releases were 85% and 75%, respectively. 21

The application of the AnMBR in the study of Gouveia et al. [2] for the treatment of municipal wastewater under psychrophilic conditions and loading rate of 2 and 2.5 kgtCOD[·]m³⁻¹d⁻¹ resulted in effluent tCOD concentrations of 100 mg[·]L⁻¹–120 mg[·]L⁻¹. In another study by Wei et al. [28], a wide range of volumetric OLR (0.8–10 gCOD[·]L⁻¹d⁻¹) was tested in AnMBR to treat

synthetic municipal wastewater. The results showed that at steady conditions, 98% COD 1 2 removal was achieved while the application of high sludge OLR led to high methane production of over 300 mL gCOD⁻¹. Wijekoon et al. [29] tested the performance of a thermophilic AnMBR 3 at different OLRs ranging from 5 12 kgCOD^{·m³⁻¹d⁻¹} to 12 kgCOD^{·m³⁻¹d⁻¹}. The authors reported 4 an average biogas production of 15 L d⁻¹, 20 L d⁻¹ and 35 L d⁻¹ at OLRs of 5.1 ± 0.1 kgCOD m³⁻ 5 $^{1}d^{-1}$, 8.1 ± 0.3 kgCOD m³⁻¹d⁻¹ and 12.0 ± 0.2 kgCOD m³⁻¹d⁻¹, respectively, with CH₄ content of 6 about 55%–65%. In addition, the reactor showed optimum COD removal efficiencies at 8 ± 0.3 7 kgCOD m³⁻¹d⁻¹ OLR. In a recent study, the highest VFA yield (48.20 \pm 1.21 mgVFA 10 8 mgCOD_{feed}) was observed at OLR of 550 mgCOD⁻¹; however, the authors achieved less VFA 9 10 yield at the examined maximum OLR (715 mgCOD¹L⁻¹), indicating that elevated OLRs can lead to high VFA production but it is also crucial to optimize operating OLR during the 11 treatment of low strength wastewater in AnMBR [30]. In high-rate bioreactors such as UASB 12 13 and AnMBR, VFAs may not be efficiently converted to methane due to low-retention times and can accumulate in the reactor and thus can be detected in the effluent [1,31]. In operating 14 conditions at elevated OLRs, VFAs should be therefore monitored to meet the local standards 15 for discharge or reuse. 16

17 **3.4. Effect of salinity in the system performance**

18 The impact of high salinity conditions was assessed in Period 4. Chloride concentration during all operational periods are given in **Figure 5**. The red points mark the membrane cleaning days. 19 α value was stable around 0.3. Biogas production gradually decreased with increasing Cl⁻ 20 concentrations (see **Figure 4**). Although the biogas production was 1.2-1.3 L^{-d⁻¹} during in the 21 beginning of Period 4 (so-called initial low saline conditions at 200 mgCl⁻L⁻¹), it decreased to 22 0.13-0.57 L⁻¹ in the presence of 500 mgCl⁻⁻L⁻¹. When the Cl⁻ concentration was increased to 23 1500 mg·L⁻¹ (at the end of Period 4), the CH₄ content in biogas reduced by 27% compared to 24 the reported values under low saline conditions. The CH₄ content of the biogas was adversely 25

1 affected (10%-20% in the beginning of Period 4 and 5% at the end) by high saline conditions.

The system almost failed to operate at the maximum examined Cl⁻ concentration (app. 2200 mg·L⁻¹), since the biogas production was 0.08 L·d⁻¹ with 3% CH₄.

In line with the results of the current study, Aslan et al. [32] demonstrated that the COD removal 4 significantly decreased at about 20 gNa⁺·L⁻¹ when treating saline wastewater in a UASB reactor. 5 6 Reduced biogas production and COD removal were also reported by Song et al. [12] in an AnMBR operating under saline conditions 15 gNa⁺L⁻¹. A decrease in biomass production was 7 observed with the increase of salinity in the AnMBR. A NaCl shock load (increase from 5 8 gNaCl[·]L⁻¹ to 60 gNaCl[·]L⁻¹) caused a reduction of COD and TKN removal efficiencies in the 9 10 study of Yogalakshmi et al. [17]; COD and TKN removal dropped to 64% and 23% at 60 gNaCl⁻L⁻¹, respectively, while the nitrification was completely inhibited. In the study of Luo et 11 al [33], a reduction of TOC and NH₄⁺-N removals was initially observed with elevated NaCl 12 13 loading; however, microbial diversity in saline AnMBR did not change and the adaptation of microbial community to saline conditions was stated to recover AnMBR biological 14 15 performance. In a recent work of Muñoz Sierra et al. [34], the performance of UASB and AnMBR was evaluated for the treatment of highly-saline phenolic wastewater. The authors 16 highlighted the superiority of AnMBR over UASB in terms of bioreactor conversion, biomass 17 characteristics and microbial community under salinity up to 26 gNa⁺·L⁻¹ due to its greater 18 probability to maintain functionality and to respond to high salinity. In another study of the 19 same authors [18], a short-term salinity fluctuation of 18 gNa⁺·L⁻¹ to 20 gNa⁺·L⁻¹ did not affect 20 the long term operation of AnMBR. The results indicated that high saline conditions initially 21 caused a decrease of the biological performance of AnMBR; however, long term adaptation of 22 microbial community to saline conditions (i.e. halotolerant or even halophilic microorganisms) 23 is required with high biomass concentrations for the system to regain its stability. 24

25 **3.5. Effect of OLR and high salinity on membrane operation**

The variations in the TMP value is shown in Figure 6a. The red points mark the membrane 1 2 cleaning days. The TMP of the membrane was stable at around 50 mbar when the system was operated with methanol addition (Period 2, OLR of 2 kgCOD^{·m³⁻¹d⁻¹}), with gas-sparging 3 condition of 10 seconds on and 120 seconds off. The specific flux normalized at 20°C was 175 4 L'h⁻¹m²⁻¹bar⁻¹ and only NaOCl cleaning was necessary after 50 days of operation. (Figure 6b) 5 During the fermentation liquid addition (Period 3, OLR of 2 kgCOD^{·m³⁻¹d⁻¹}) the behavior was 6 first similar to the methanol co-treatment; then the TMP increased gradually after 50 days of 7 operation and reached to 500 mbar after 100 days of operation. Thus, a more intense chemical 8 cleaning was applied to restore the initial permeability (citric acid at 1000 mg·L⁻¹) on day 315. 9 10 A significant EPS production was observed when the fermentation liquid was used, leading to larger formation of the "cake" on the surface of the membrane. This was mainly due to the 11 fluctuation of the characteristics of the production of fermentation liquid. There was an increase 12 in EPS concentration from 52.8 mgEPS⁻¹ in the Period 2 to 70.8 mgEPS⁻¹ in the Period 3. 13 The latter decreased the membrane permeability a short time, higher rate pore obstruction and 14 15 therefore more intense and frequent cleaning was required.

In Period 4, the TMP remained stable at 12 mbar when the reactors were fed with the fermentation liquid together with additional NaCl to increase salinity in the system, at the concentration of 500 mgCl⁻L⁻¹. The average TMP was around 50 mbar, at Cl⁻ concentration of 1500 mgCl⁻L⁻¹, caused by the increased filtration resistance following the increased TMP due to the increased EPS concentration in Period 3. The saline conditions therefore only affected the initial TMP value and remained constant during the system operation.

The TMP of AnMBRs is highly dependent on the critical flux and the sparging rate together with other environmental and operating conditions [35]. Furthermore, an initial flux below the critical flux, prior to the introduction of peak flow, is reported to be advantageous to permeability recovery [36]. In the study of Muñoz Sierra et al. [18], increased salt concentration

was found to affect the TMP negatively (350 mbar at a flux of 4.0 Lm²·h⁻¹). The deterioration 1 2 of membrane filtration performance was attributed to the decrease of biomass particle size when 3 salinity was increased. The small particle size had a significant influence on the cake layer compaction that increased the operational values of the filtration resistance. Furthermore, 4 higher stability of process performances of AnMBR over UASB was reported to overcome high 5 salinity [34]. Elevated TMP values were also reported by Yurtsever et al. [19] with respect to 6 7 high salinity conditions. The salinity induced large molecules as foulants in gel/cake layer, that may originate from biomass loosely bound EPS. The EPS properties are highly dependent on 8 9 the operating OLR [37]; and the OLR increase is often accompanied with high EPS production.

10 **3.6. Relationship between process parameters and system performance**

11 PCA was carried out to reveal the relationships between the applied operating conditions (i.e. OLR, salinity) and system performance in terms of biogas production, CH₄ content of the 12 13 biogas, α value, TMP and Js. The PCA supported our previous discussion regarding the performance of the UASB+AnMBR system at different periods. OLR was closely grouped 14 15 together with the biogas production, CH_4 content and α value (Figure 7). This cluster showed the close relationship between these parameters mostly in Period 1 which included only the 16 17 UASB operation. The data-points of Period 2 was comparatively more equally distributed 18 between the parameters; where the negative impacts of OLR on TMP were clearly reflected in 19 our data especially in Period 3. Furthermore, data points of Period 4 were characterized by high chloride concentrations. The displayed negative correlation between the salinity and TMP 20 21 and/or Js in Period 4 was due the citric acid cleaning of the membrane following the fouling occurred in Period 3 that was previously mentioned. TMP value was close to its initial value 22 23 right after the citric acid cleaning and although TMP also slightly increased Period 4 at high chloride concentrations (see E-Supplementary material), this increase was relatively low 24 25 comparing to Period 3.

1 3.7. Assessment of the possible re-use of the effluent

Population density and economic activity lead to significant differences in the water stress levels of the basins. Water stress occurs in many areas of the EU, particularly in the Mediterranean regions and part of the Atlantic regions. The data indicated by the European Environment Agency regarding the use of water in various sectors indicate that irrigation in agriculture represents about half of the water used annually with high seasonal and geographical variations.

8 In May 2018, the European Commission presented a proposal for the reuse of treated 9 wastewater through a regulation that establishes common minimum requirements. This 10 proposal was going under revision with resolution of 12th February 2019. The proposal imposes 11 obligations that include:

Compliance with the minimum requirements. These requirements differ based on four water
quality classes defined according to the type of crop and the irrigation method.

• Monitoring of recovered water based on minimum test frequency requirements.

15 The resolution also provides that the competent authorities of the member states can impose 16 additional requirements based on a risk management plan presented by the water utility to 17 mitigate unacceptable risks to health and environment.

In Italy, at national level, D.M. 185 of 2003 establishes the minimum quality requirements forthe reclaimed wastewater and its reuse, including limits on nitrogen and phosphorus.

The obtained effluent quality was compared against the values reported in **Table 4** (minimum requirements for reuse). Analyzing the results obtained from this study, the UASB effluent is not compliant with the limits required both by D.M. 185/03 and by the new EU proposal. The permeate of the AnMBR appears of higher quality because it is free from TSS and falls into class A since ultrafiltration guarantees also disinfection and *E. coli* removal. For the restrictions posed by the D.M. 185/2003 for the reuse of the treated wastewater are mainly related to the

high concentration of chloride (High values are due to the salt water intrusion since the plant
is in a coastal area) and the nutrients concentration (N and P), both exceed the limit values. The
high nutrient content in the permeate (TN reaches a maximum value of 60 mgN·L⁻¹, TP of 6
mgP·L⁻¹), allows a potential reuse in fertigation field.

5 The national law in Italy is more restrictive compared to the new European proposal, which 6 introduces an innovative approach based on a framework for the risk management. EU 7 requirements vary according to the type of irrigated crops and according to the method of 8 irrigation, taking into account the potential risk of contamination of the products. The most 9 strict requirements are provided for class A, where reclaimed water can be in contact with the 10 edible parts of irrigated food crops.

11 **4.** Conclusions

Regarding the change of paradigm in the context of circular economy in terms of water, 12 13 nutrients and energy; site-specific optimization of an AnMBR is crucial especially in coastal regions. In this particular study, the addition of fermented cellulosic sludge to raw wastewater 14 15 increased the biogas production without affecting the overall CH₄ content of the biogas. In case of the application of the process in coastal areas, the biogas production decreased by 27% due 16 to the saline conditions when Cl concentration was up to 1500 mg L⁻¹. Moreover, the CH₄ 17 18 content of the biogas was also adversely related to the high saline conditions up to almost null value for chloride higher than 2000 mg·L⁻¹. At Cl⁻ concentration less than 1500 mgCl·L⁻¹, long 19 term adaptation of microbial community (i.e. halotolerant or even halophilic microorganisms) 20 21 may be required with high biomass concentrations for the system to regain its stability and recover the bioreactor performance in UASB+SuMBR. Concerning the membrane operation, 22 23 the increased salinity resulted only in the increase of initial TMP value where it stayed constant during the following operating days. The AnMBR effluent fell into class A since ultrafiltration 24 guarantees also disinfection and E. coli removal. 25

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10 Appendix A. Supplementary data

11 Supplementary data to this article can be found in the online version.

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1 Figure Captions

- Fig. 1. Salt water intrusion (European Environment Agency, 2017) and the location of
 Falconara Marittima WWTP.
- 4 Fig. 2. Scheme of the UASB pilot plant coupled with anaerobic ultrafiltration membranes:
- 5 Configuration with a) urban wastewater, b) co-treatment of urban wastewater and methanol, c)
- 6 co-treatment of urban wastewater and fermentation liquid.
- **Fig. 3.** Variations in a) OLR b) α value (the ratio of partial alkalinity over total alkalinity) at
- 8 different periods of the operational time.
- 9 Fig. 4. Variations in a) Biogas production b) CH₄ content of the biogas at different periods of
 10 the operational time.
- 11 Fig. 5. Variations in chloride concentration at different periods of the operational time.
- 12 **Fig. 6.** Variations in a) TMP profile b) Specific flux at 20°C in AnMBR at different periods of
- 13 the operational time. The red points mark the membrane cleaning days. Citric acid cleaning
- 14 was conducted on day 315. TMP and specific flux were not recorded between the days 315
- 15 and 432.
- 16 Fig. 7. Principal component analysis (PCA) of the operating parameters and system
- 17 performance.