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Anaerobic Membrane Bioreactor for Urban Wastewater Valorisation: Operative Strategies and Fertigation Reuse

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In European scenario, the potential source for water supply from treated wastewater is actually estimated in 1,100 Mm3/y (EU-ENV, 2015). Anaerobic processes compared with conventional aerobic ones, cause a net reduction of the operative costs and possible reuse for fertigation purposes. The tested anaerobic pilot (HRT 6 h and T 30°C) is constituted from a UASB reactor (16 L). Increment of influent organic loading rate (OLR) was studied for 1 year from 1 to 2 kgCOD/m^3/d by feeding raw wastewater (Period 1), methanol (Period 2) and fermented supernatant from cellulosic sludge (Period 3).The biogas production was assessed equal to 0.13 m^{3}biogas/kgCOD (Period 1), to 0.57 m^{3}biogas/kgCOD (Period 2) and to 0.24 m^{3}biogas/kgCOD (Period 3) with methane percentages constant around 33%. UASB effluent has not the final quality to comply limit values for water reuse and fertigation, especially for microbiological parameters. Further treatments could be necessary to achieve the removals of bacteria, such as E.Coli, recalcitrant organic traces and metals. Therefore, some advanced post treatments have been studied in this paper after UASB treatments, like UV disinfection, UV coupled with H2O2 and GAC adsorption. The innovative solution is coupling UASB reactor with anaerobic submerged sidestream AnMBR (UF hollow fiber membrane with 0.03 µm of nominal pore-size and 0.5 m^2 of surface area KOCH, Puron single bundle). Membrane cleaning was carried out using sodium hypochlorite solution (400 ppm) each 45 days to remove organic fouling and to recover the initial permeability of the membrane. The average operative flux at process temperature was equal to 8.8±1.9 L/h/m^2 and operating TMP of 44.6±8.5 mbar was detected. The removal of E.Coli was investigated in the effluent from UASB and in the permeate from AnMBR process. At clean membrane conditions, complete removal of bacteria (99±1%) was found. Furthermore, the determination of microplastics distribution was carried out both in the experimental anaerobic pilot and in the conventional full scale aerobic treatment plant. The effluent microplastics were quantified and the removal role of the different operative units was studied.

Keywords: anaerobic membrane, reuse, fertigation, microplastics

1. Introduction

Widespread water scarcity characterizes the southern European countries where main pressures from water consumption are concentrated on irrigation and domestic demand also related to touristic activities. Over the past thirty years, droughts have dramatically increased in number and intensity in the EU and, to date, at least 11% of the European population and 17% of its territory has been affected by water scarcity. The potential role of treated wastewater reuse as an alternative source of water supply, is now well developed. The current volume of reused water in Europe (EU-ENV, 2015) has been estimated at around 1,100 Mm^3/y.
Innovative anaerobic treatments of urban wastewater reuse can be a real option and in the same time they allow to save energy and produce a small amount of excess sludge. Compared with conventional aerobic technologies, they cause a net reduction of the operative costs for the lack aeration and reduce treatment cost

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up to 46%, bringing benefits both for environmental and economical sustainability. Therefore, anaerobic treatment is identified as promising technology to couple energy production and valorization of effluent. Particularly, anaerobic processes can enable synergistic application of water reuse and recovery of materials and nutrients (N, P) with economic benefits for wastewater treatment operators and users of treated wastewater.

It is also possible to product bioethanol from the wastewater by means of anaerobic fermentation (Sofia et Al, 2013). At this scope, the first Innovation Deal (ID) European program was focused on regulatory frameworks affecting water reuse for agricultural purposes identifying the AnMBR as possible solution for fertigation purpose. The Innovation Deal has been analysed the regulatory barriers that prevent a paradigm shift towards converting wastewater treatment plant into water and resource recovery facility. In addition to regulatory barriers, the installation of new plants is very often hampered by the diffusion of the "Nimby Syndrome", a protectionist attitude adopted by community group against the installation of new plants near their homes (Giuliano et Al, 2018). The ID aimed at facilitating the market application of anaerobic membrane technologies that might enable efficient recovery of energy and materials from wastewater while ensuring high level of environmental and public health protection.

In general, anaerobic processes permit to obtain elevated release of nutrients without reaching sufficient quality level for reuse in terms of bacteriological proprieties and organic matter. Therefore, tertiary treatments are needed to increase the final effluent characteristics. The combination of anaerobic processes with advanced post-treatments, like UltraViolet (UV) or UV coupling with Hydrogen Peroxide (H_2O_2) or Granular Activated Carbon (GAC) units, could improve the final effluent quality. The UV technology provides a rapid and effective inactivation of microorganisms through a physical process. The UV light has demonstrated efficacy against pathogenic organisms, including those responsible for cholera, polio, typhoid, hepatitis and other bacterial, viral and parasitic diseases (Li et al. 2017). The H_2O_2 concentrations, reported in literature, range from 0.8 to 9.0 mmol/l (from 27.2 to 306 mgH_2O_2/L). Approximately 10% of the bacterium luminescence is inhibited by 0.5 mmolH_2O_2/L and 95% is inhibited by 9.0 mmolH_2O_2/L after 30-60 min of contact time. Other authors identified the optimal H_2O_2 concentration of 20 mg/L to degrade pharmaceutical compounds present in secondary effluent of urban wastewater treatment plants (Urbano et al., 2017).

With a more innovative approach, anaerobic processes can be coupled with advanced separation system like ultrafiltration membranes. An example of selective membranes to address the CO2 capture was used to reduce GHG emissions from IGCC plant for power and hydrogen production (Sofia et Al., 2015) (Anaerobic Membrane Bioreactor (AnMBR)). The interesting on advanced filtration processes are related not only to ordinary pollutants but also to emerging toxic compounds (Eusebi et al., 2011). Some studies reported that with AnMBR, pharmaceutical compounds like Trimethoprim (Tmp) and sulfamethoxazole (Smx) are consistently removed at efficiencies of 94.2 ± 5.5%, and 67.8 ± 13.9%, respectively, while, marginal removals are obtained for carbamazepine (Cbz) (0.3 ± 19.0%) and for diclofenac (Dcf) (15.0 ± 7.2%) (Yeyuan Xiao et Al., 2017). In this scenario, membrane filtration seems to be promising solution also for microplastics (MPs) removal.

Considering the high volumes treated daily from Wastewater Treatments Plants (WWTP), a big amount of microplastics can be still released through the effluents; but WWTP, that are not specifically designed to retain MPs, can effectively remove most of these particles from the influent reducing microplastics' input in the environment. Jing Sun at Al., 2018, reported in their studies that the overall microplastics removal efficiencies of WWTPs without tertiary treatment is above 88% and the number increased to over 97% in the WWTPs with tertiary treatments. The membrane bioreactor (MBR) can remove 99.9% of MPs (from 6.9 to 0.005 MP/L), rapid sand filter 97% (from 0.7 to 0.02 MP/L), dissolved air flotation 95% (from 2.0 to 0.1 MP/L) (Julia Talvitieet Al., 2017). The efficiency of an advanced MBR technology is higher, in fact, the permeate contains only 0.4MP/L in comparison with the final effluent of the CAS process (1.0 MP/L) (Mirka Lares et. Al., 2018). This paper shows the preliminary performances of demonstrative UASB coupled with different advanced post-treatments: membrane ultrafiltration, UV disinfection and GAC adsorption.

2. Materials and methods

The WWTP of Falconara Marittima (Italy) has a design treatment capacity of 80,000 PE with nominal influent flowrate of 30,000 m^3/d. After preliminary treatments, feeding is biologically treated in a demonstrative UASB followed by the AnMBR process. The UASB reactor (16 L reactor) coupled with an Ultrafiltration (UF) hollow fiber membrane with 0.03 μm of nominal pore-size and 0.5 m^2 of surface area (KOCH, Puron single bundle) worked in submerged-sidestream AnMBR configuration for about 1 year. The quality of the raw influent is reported in Table 1. All the main parameters were analyzed according to the Standard Methods (APHA, 2005).

Table 1: Characterization of the influent to the WWTP in dry weather

Qin	pH	COD	TSS	NH4-N	TN	TP
m3/d	-	mg/l	mg/l	mg/l	mg/l	mg/l
19,525±2,488	8.1±0.2	373±148	232±110	31±6	38±7	5.1±1.5

Hydraulic Retention Time (HRT) was imposed equal to 6 hours and Temperature was maintained at 30°C both for reactor and membrane tank. The up-flow velocity of the UASB reactor was maintained at 1 m/h. The critical flux for the AnMBR was studied at different solids concentrations and the effects of gas sparging on fouling rate was evaluated carrying out each test, at a specific solid concentration with no gas and with gas-sparging, to investigate the positive effect of gas bubbles that shake up membrane fibres and provide to clean them. Gas-sparging method used Nitrogen gas (N_2) by alternating 10 seconds of gas off and 10 seconds of gas on with a specific flowrate value of 2m^3/m^2/h. Maximum solids concentration of about 300 mg/L for UASB effluent was identified as limiting for the critical flux (14 L/m^2/h), therefore operating flux was maintained at 8.8±1.9 L/h/m^2 and the average Transmembrane Pressure (TMP) was detected equal to 44 mbar. Membrane cleaning was carried out using sodium hypochlorite solution (400 ppm) each 45 days to remove organic fouling and to recover the initial permeability of the membrane. Increment of influent Organic Loading Rate (OLR) was studied from 1 to 2 kgCOD/m^3/d by feeding raw wastewater (Period 1), methanol (Period 2) and fermented supernatant from cellulosic sludge (Period 3). The performance in terms of macropollutants and bacteria removal efficiencies and biogas production were investigated in the different scenarios.
Other tertiary treatments using UV lamp (VIQUA D4+ with a volume of 2.85 L and 40 Watts of power) at dose of 8-70 mJ/$cm^2$and Hydrogen Peroxide (H_2O_2) at concentrations of 5, 20, 50 and 100 mg/L with a contact time of 30 min, have been carried out to determine the effect on E. Coli log reduction on the effluent from UASB process.
Moreover, Active Granular Carbon (GAC) adsorption (contact times from 10 to 180 minutes) have been experimented to define the potential removal of the metals and of the recalcitrant organic pollutants. GAC dosages ranged from 2 to 20 mg/L (Kårelid et Al., 2017). Three type of GAC were tested and Table 2 reported their specific characteristics. The tests were realized by using effluent from UASB processes both in batch and in column configurations. The adsorption capacity of the activated carbon was investigated at constant temperature of 20 °C by the adsorption isotherm.

Table 2: Characteristics of Activated Granular Carbon

Name	Granulometry	Density	Iodine Index	Blue methylene index	Specific Surface
-	U.S. mesh	g/l	mg/g	mL/g	m^2/g
ST100	8x30	520±20	>750	>150	>800
ST300	8x30	500±20	>950	>180	>950
STW400	8x30	500±20	>1000	>190	>1000

Finally, Ozone and NaOCl dosages and their effect on MPs were studied in batch tests. Microplastics were characterized before and after the treatments by µFT-IR technology. Ozone dosages were 50, 200, 400, 500 and 1000 mgO_3/L and NaOCl (at 14% w/v) dosages were from 40 to 114 mlNaOCl/m^3. Microplastics were quantified in the liquid fraction collected at different points of the system, and characterized according to shape, size and polymer typology by microscopy and µFT-IR technology.

3. Results and discussion

3.1 Performances of UASB

During Period 1 the biogas production was assessed equal to 0.4±0.17 L/d. Regards Periods 2 and 3, the increase of soluble organic content has significantly affected the amount of biogas production, reaching an average value 10 times higher than the previous period (3±1.1 L/d in period 2 and 4.11+3.2 L/d in period 3). Biogas samples were collected and methane content was analyzed with gas chromatography technique. The CH_4 amount was quite constant over the three OLR periods with percentages from 30 to 40% up to 50%. Specific Gas Production are in the same range reported in literature, from 0.2 to 0.5 m^3biogas/kg COD_{added} (Song et Al., 2018), the highest value is reached with methanol addition to urban wastewater.

Table 3: Biogas production and CH_4 content at different OLRs

Configuration	Period	Feed	OLR	Biogas production	CH_4	SGP
			kgCOD/m^3/d	L/d	%	m^{3}Biogas/kgCOD added
UASB+AnMBR	1	Raw wastewater	1.05±0.4	0.40±0.17	33.0±6.0	0.13±0.02
UASB+AnMBR	2	Methanol	2.01±0.4	3.00±1.10	33.6±10.0	0.57±0.30
UASB+AnMBR	3	Centrate	1.85±0.6	4.11±3.2	32.7±10.3	0.24 ±0.15

3.2 Performances of UASB coupled with AnMBR

Considering UASB process coupled with membrane filtration, removals of Chemical Oxygen Demand (COD) and Total Suspended Solids (TSS) were similar during overall experimental phases and respectively equal to 86±1% and 100% (Period 1), to 85±1% and 100% (period 2) and to 83±1% and 100% (Period 3). Total Nitrogen (TN) and Total Phosphorus (TP) were released with the effluent reaching up to 83% of TN release and 76% of TP release during Period 2 and 76% of N release and 85% of P release during Period 3. An increase in the concentration of soluble forms of the nutrients occurred in the effluent because of the degradation of the organic matter, which entails the solubilisation of the organic nitrogen and phosphorous to ammonium and phosphate (Moñino et al. 2017).
The removal of E.Coli was investigated in the permeate of the AnMBR process. At clean membrane conditions, complete removal of bacteria (99±1%) was found.
Moreover, the evaluation of microplastics distribution was studied both in the demonstrative anaerobic and the conventional full-scale aerobic flow schemes identifying the roles of the different operative units in the microplastics removal. About 97% of microplastics were removed, providing 1.7 MPs/L after UASB reactor, further reduced to 0.1 MPs/L (100 MPs/m^3) after ultrafiltration. MPs larger than 1mm were retained by intermediate steps. After the ultrafiltration unit only fibers, mainly of polyesters, were found.

Table 4: Removals efficiency and nutrients release UASB+AnMBR

Period	COD removal	TSS removal	E.Coli removal	MPs removal	TN release	TP release
	%	%	%	%	%	%
1	86±1	100	99±1	-	-	-
2	86±1	100	99±1	97	83	76
3	83±1	100	99±1	-	76	85

The final effluents coupling UASB and AnMBR during Periods 2 and 3, are reported in Table 5, to make a comparison with the new proposal for the "*Regulation of the European Parliament and of the Council*" (28 May 2018) on minimum requirements for water reuse. About uses and minimum requirements, the new proposal allows to use reclaimed water for agricultural irrigation both for food crops consumed raw, for processed food crops and for non-food crops. There are different classes of reclaimed water quality (A, B, C, D) and the minimum requirements are set out in the proposal for each allowed use, class and irrigation method.

Table 5: Effluent quality of UASB+AnMBR in different periods

Period	COD	TSS	TN	TP	E.Coli
	mg/L	mg/L	mg/L	mg/L	UFC/100mL
1	55±15	0	-	-	-
2	43±13	0	15±10	1.9±1.0	1
3	77±87	0	60±55	6.0±4.3	1

In this study, the effluent from AnMBR complies with the minimum requirements set out in the EU proposal in terms of E.Coli, other national and regional laws actually in force in Europe present more restrictive limits on microbiological parameters and micropollutants that make wastewater reuse not already possible (i.e. Italy and Greece).

3.3 Performances of UASB and others post-treatments

Tertiary treatments have been applied to UASB effluent. The UV post-treatment was studied to achieve the complete removal of the microbiological population. Moreover, an experiment was carried out using only H_2O_2

to remove E.Coli. Results demonstrate that only the highest dosage (100 mgH_2O_2/L) with 30 min contact time was able to remove E.Coli. Higher contact time (example 60 min) did not increase bacteria reductions.
Using only the UV lamp, removal of 99.5% and log reduction of 2.3 were achieved by testing the effluent of the UASB reactor with UV dose of 80 mJ/cm^2. UASB effluent is characterized by very low transmittance (about 30%), therefore, to reach literature UV doses (70mJ/cm^2) longer contact times are needed. No specific improvement of the log reduction was found applying pre-filtration at 11 micrometers before the UV disinfection. Finally, granular active carbon adsorption was tested to define the potential removal of soluble metals and of recalcitrant organic pollutants. Affinity of activated carbon to the removal of some heavy metals was established for UASB anaerobic effluent and was found equal to 36% for Zinc, 46% for Nickel and only 8% for Copper. Moreover, the reduction of the soluble organic COD was 55% using GAC ST100 and 92% using GAC ST300.
Other advanced post-treatments like Ozone and NaOCl disinfection, tested in batch tests with different contact times, have highlighted changes in the specific characteristics of original MPs polymer (polyethylene) with elevated dosages above 1000 mgO_3/L and 95 lNaOCl/m^3.

4. Conclusions

In this work it has been studied a pilot scale UASB treating urban wastewater. This system can allow to save energy, producing biogas from 0.2 to 0.5 m^3biogas/kg COD_{added} depending on the Organic Loading Rate. UASB effluent does not show effluent quality to comply limit values for reuse, especially for microbiological parameters. Further treatments could be necessary to achieve bacteria, recalcitrant organic traces and metals removals. Therefore, some advanced post treatments have been studied after UASB treatments, like UV disinfection, UV coupled with H2O2 dose and GAC adsorption. Removal of 99.5% of E.Coli and 2.3 of log reduction were achieved by testing the effluent of the UASB reactor with UV dose of 80 mJ/cm^2. Heavy metals removals were established for UASB effluent with GAC adsorption equal to 36% for Zinc, to 46% for Nickel and to 8% for Copper. Reduction of recalcitrant organic compounds were evaluated keeping into account the soluble organic COD reduction that ranged from 55% using GAC ST100 to 92% using GAC ST300.
Coupling UASB and AnMBR, the almost total COD and TSS removals can be reached (85% COD% and 100% TSS%). Thanks to TN and TP releases (from 75% to 85%) the permeate is suitable for fertigation and agriculture application. The effluent fits the limit values for reuse according the new EU proposal on water reuse, also in terms of E.Coli. In addition, the innovative UASB and AnMBR system removed 97% of influent microplastics, providing 1.7 MPs/L after UASB reactor up to 0.1 MPs/L (100 MPs/m^3) after ultrafiltration. After the ultrafiltration unit only fibers, mainly of polyesters, were found. Others tertiary treatments, like Ozone and Sodium Hypochlorite, were studied to verify the effects on MPs. For both the technologies, changes of specific characteristics of MPs polymers at high dosages of oxidants (1000 mgO_3/L and 95 lNaOCl/m^3) were highlighted.

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