A MOVING BED MEMBRANE BIOREACTOR PILOT PLANT FOR CARBON AND NUTRIENT REMOVAL

G. MANNINA*, M. CAPODICI*, A. COSENZA*, D. DI TRAPANI*, G. VIVIANI*, G.A. EKAMA**

* *Dipartimento di Ingegneria Civile, Ambientale, Aerospaziale, dei Materiali (DICAM) Scuola Politecnica, Università di Palermo, Palermo, Italy*

** *Water Research Group, Department of Civil Engineering, University of Cape Town, South Africa*

Keywords: Nutrient removal, WWTP, membrane, MBBR, wastewater

Abstract *The paper reports the main results of an experimental gathering campaign carried out on a moving bed membrane bioreactor pilot plant conceived for carbon and nutrients removal according to a University of Cape Town scheme. Organic carbon, nitrogen and phosphorus removal, biokinetic/stoichiometric constants, membrane fouling tendency and sludge dewaterability have been assessed during experiments. The achieved results showed that pilot plant was able to guarantee very high carbon removal, with average efficiency of 98%. In terms of nitrification, the system showed an excellent performance, with efficiencies higher than 98% for most of the experiments. This result might be related to the presence of biofilm in the aerobic compartment that contributed to sustain the complete nitrification of the influent ammonium. Conversely, the average P removal efficiency was quite moderate, likely due to the increase of the ammonium loading rate that could promote an increased NO_3-N recycled from the anoxic to the anaerobic tank, interfering with PAOs activity inside the anaerobic tank. Referring to membrane fouling, the irreversible resistance due to superficial cake deposition was the mechanism that mostly affected the membrane filtration properties. Moreover, it was noticed the increase of the resistance due to pore blocking and a general worsening of the membrane filtration properties. This result could be due to the increase of the EPS_{Bound} fraction that could be promoted by biofilm detachment phenomena occurred during experiments.*

1. Introduction

It is well known that nutrients (particularly, nitrogen and phosphorus compounds) may have adverse environmental impacts (e.g., eutrophication, toxicity towards the aquatic organisms, etc…) (Wang et al., 2006). Therefore, their removal from wastewater is an imperative requirement, especially when discharging in sensitive areas (Li et al., 2013). In the last years, several biological and physic-chemical methods have been developed to remove nutrients from wastewater. Among these methods, biological treatments are the most cost-effective methods (Chu and Wang, 2011). Biological nutrient removal (BNR) from domestic wastewater has been extensively investigated and developed in the last years and it is usually based on anaerobic, anoxic and aerobic reactors linked in-series (among others, Wanner et al., 1992; Cosenza et al., 2013a; Lu et al., 2015). In BNR

processes, N and P removal is accomplished, respectively, by heterotrophic denitrifying bacteria and polyphosphate-accumulating organisms (PAOs) which require carbon source (Naessens et al., 2012). In particular, the biological phosphorous removal is commonly conducted by exploiting the ability of PAOs to accumulate P and to store it as intracellular polyphosphate (poly-P) under alternating anaerobic/aerobic conditions (Li et al., 2013). However, despite conventional activated sludge (CAS) processes are effective for removal of organic and nutrients compounds, the overall efficiency is strictly related to the performance of the solid-liquid separation into the final settler, which may suffer of separation problems (Wanner, 2002). In this context, membrane bioreactor (MBR) technology may represent a useful solution, since it enables to disconnect the efficiency of the biological processes from the biomass settling properties. Indeed, MBRs have attracted considerable interest due to various advantages compared to conventional process that originate from the use of a membrane for solid–liquid separation (Fu et al., 2009). In particular, MBRs generally feature high quality effluent, small footprint and low sludge production rates compared to CAS systems (Stephenson et al., 2000). Therefore, in the last years the integration of BNR process with MBRs has been proposed for the wastewater treatment to treat the quality of the effluent, including such BNR processes as University of Cape Town (UCT) process, anoxic/oxic (A/O) process and anaerobic/anoxic/oxic (A2O) process (Hu et al., 2014). However, one of the major drawbacks in MBRs is still represented by fouling phenomena that may severely affect the filtration properties of the membrane modules (Judd and Judd, 2010). In particular, the mixed liquor suspended solid (MLSS) concentration has been recognized to play a significant effect on membrane fouling (Poyatos et al., 2008; Di Trapani et al., 2014). An alternative to managing this problem is to couple a MBR system with a moving bed biofilm reactors (MBBR) for the simultaneous growth of suspended biomass and biofilm within the system, realizing a so-called moving bed membrane bioreactor (MBMBR) (among others Leyva-Díaz et al., 2013; Yang et al., 2014). In particular, MBBR technology basically relies on the use of small plastic carrier elements that are kept in constant motion throughout the entire volume of the reactor, for biofilm growth (Ødegaard, 2006). These systems are especially useful when slowly growing organisms as nitrifiers have to be maintained inside a wastewater treatment plant (WWTP) (Kermani et al., 2008). When combined with a MBR system realizing a MBMBR process, there is the potential to utilize best characteristics of both biofilm processes and membrane separation (Ivanovic and Leiknes 2008). Using this technology, the biofilm system may reduce the concentration of suspended solids and improve the extent of membrane fouling. Nevertheless, MBMBRs are relatively new, especially when referring to system performance, biomass biokinetic activity and membrane fouling tendency. Moreover, very few studies have been reported for BNR systems adopting hybrid MBMBR processes (Yang et al., 2010). Therefore, the aim of the study is to gain insight about the behavior of a University Cape Town (UCT) pilot plant, combining both MBR and MBBR technology (UCT-MBMBR), for the treatment of domestic wastewater. In particular, a UCT-MBMBR pilot plant was monitored for almost two months with the aim to investigate the system performance in terms of organic carbon and nutrient removal, biomass biokinetic behavior and membrane fouling tendency and sludge features.

2. Materials and methods

2.1. UCTMBMBR system description

An UCT-MBR pilot plant was built at the Laboratory of Sanitary and Environmental Engineering of Palermo

University (Figure 1). The pilot plant consisted of an anaerobic (volume 62 L), an anoxic (volume 102 L) and an aerobic (volume 211 L) tanks according to the UCT scheme (Ekama et al., 1983). The solid-liquid separation phase was carried out by means of an ultrafiltration hollow fibre membrane (PURON®). The membrane module was located inside an aerated tank (MBR tank) (36 L). An oxygen depletion reactor (ODR) of 40 L allowed the oxygen stripping in the mixed liquor recycled from the MBR tank to the anoxic one (QRAS). The membrane was periodically backwashed (every 9 min for a period of 1 min) by pumping, from the Clean In Place (CIP) tank a volume of permeate back through the membrane module. From the volumes of the reactors and recycle flows the mass fractions of the reactors were calculated from Ramphao et al. (2005) to be; anaerobic 0.071, anoxic 0.232, aerobic 0.481, MBR+ODR 0.216. The anoxic and aerobic compartments were filled with suspended plastic carriers (carriers density = 0.95 g cm^{-3}; carriers specific surface = 500 m^2 m^{-3}), with a 15 and 40% filing ratio, corresponding to a net surface area of 75 and 200 m2 m-3 in the anoxic and aerobic reactor, respectively. Figure 1 reports a schematic view of the UCT-MBMBR pilot plant.

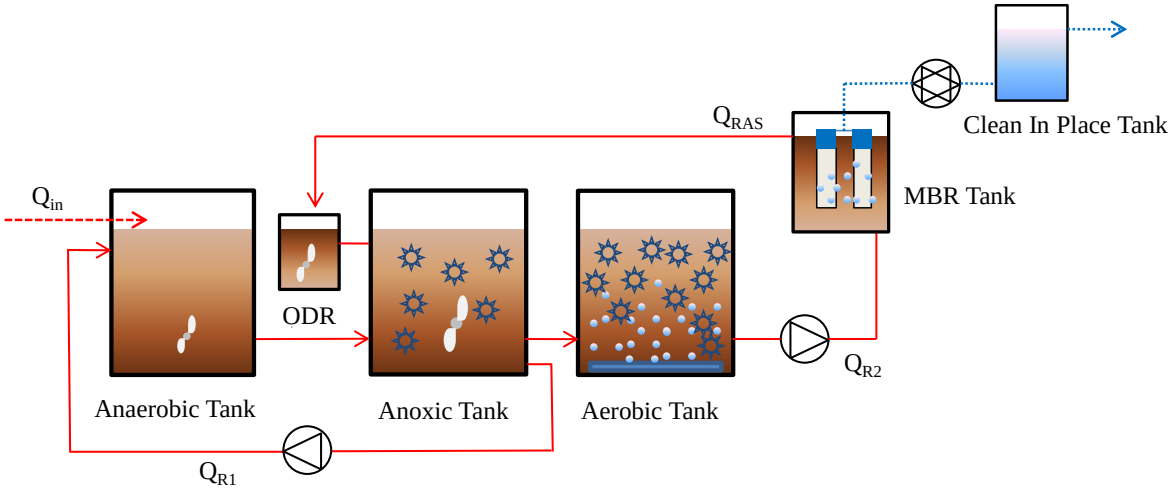

Figure 1. *Schematic lay-out of the UCT-MBMBR pilot plant*

The extraction flow rate was set equal to 20 L h^{-1} (Q_{in}). During the pilot plant operations, a 20 L h^{-1} flow rate (Q_{R1}) was continuously recycled from the anoxic to the anaerobic tank. Furthermore, a 100 L h^{-1} flow rate (Q_{R2}) of mixed liquor was pumped from the aerobic to the MBR tank. A net permeate flow rate of 20 L h^{-1} was extracted (Q_{OUT}) through the membrane module. Therefore, the recycled activated sludge (QRAS) from the MBR to the anoxic tank through the ODR tank was equal to 80 L h^{-1}.

The UCT-MBMBR pilot plant was operated for almost 60 days and was fed with a mixture of real wastewater (deriving from the University buildings and characterized by higher ammonia content compared to typical domestic wastewater) and synthetic wastewater. Briefly, the synthetic wastewater represented almost 50% of the total wastewater in terms of COD, with the 30% constituted by readily biodegradable COD (RBCOD) (dosed as sodium acetate), whilst the remaining 70% was more slowly biodegradable (dosed as glycerol). The synthetic wastewater was added to meet the design organic loading rate to the pilot plant.

The inlet wastewater had the following average features: COD = 607 mg L^{-1}; total nitrogen (TN) = 65 mg L^{-1}; total phosphorus (TP) = 11 mg L^{-1}; COD/TN/TP = 100/10.7/1.8. Permeate flux was maintained equal to 21 L m^{-2} h^{-1},

the hydraulic retention time was equal to 20 h with a permeate flow rate of 20 L h^{-1}.

2.2. Analytical methods

During pilot plant operations, the influent wastewater, the mixed liquor inside the anaerobic, anoxic, aerobic and MBR tank and the effluent permeate have been sampled and analysed for TSS, volatile suspended solids (VSS), total chemical oxygen demand (COD_{TOT}), supernatant COD (COD_{SUP}), ammonium nitrogen (NH_4-N), nitrite nitrogen (NO_2-N), nitrate nitrogen (NO_{3-} N), total nitrogen (TN), phosphate (PO_4-P), total phosphorus (TP). All analyses were carried out according to the Standard Methods (APHA, 2005); pH, dissolved oxygen (DO) and temperature were also monitored in each tank by using a multi-parameter probe. Referring to the COD removal, in order to distinguish the removal due to the biological processes from that one due to the filtration operated by the membrane, two different removal efficiencies have been calculated (Di Trapani et al., 2014): the biological removal efficiency and the total removal efficiency. The biological COD removal efficiency was calculated as the difference between the COD_{TOT} value in the influent and the COD_{SUP} measured in the supernatant of mixed liquor samples (filtered at 0.45 μm) withdrawn from the MBR tank. Conversely, the total COD removal efficiency (including the removal contribution due to membrane filtration) was assessed as the difference between the inlet and the permeate COD_{TOT}, respectively. Periodic test on carrier samples were carried out, in order to establish the biofilm growth on the carriers; briefly, a carriers sample was taken from the anoxic and aerobic reactors (10 and 15 carriers, respectively), dried in an oven for one night at 105◦C and then weighted (W1). After biofilm was removed, the carriers were dried another night at 105◦C and then weighted again (W2); thereafter, the amount of the attached biomass was then calculated as W1− W2. For further details, the reader is addressed to literature (Di Trapani et al., 2013-2014).

Respirometric batch tests were carried out by means of a “flowing gas/static-liquid” respirometer to evaluate the kinetic and stoichiometric parameters for both autotrophic and heterotrophic biomass (Di Trapani et al., 2015). Briefly, the suspended biomass samples were taken from the aerobic reactor and eventually diluted with permeate in order to obtain a mixed liquor volatile suspended solid (MLVSS) concentration in the range of 2.0–3.0 g VSS L^{-1}. The batch tests on biofilm were performed with carriers and permeate, by imposing in the respirometer the same filling fraction of the UCT-MBMBR pilot plant.

In the batch tests aimed to evaluate the heterotrophic biokinetic parameters, the nitrifying biomass was inhibited by adding 10 mg L^{-1} of Allylthiourea (ATU), whilst the exogenous oxygen uptake rate (OUR) was enhanced by the addition of a readily biodegradable organic substrate (sodium acetate in this case). The substrate biodegradation rate was then assumed proportional to the exogenous OUR. On the other hand, the estimation of the kinetic parameters for the autotrophic population was carried out with a very similar procedure. Nevertheless, no inhibiting substance like ATU was added and ammonium chloride (NH_4Cl) was spiked to evaluate the biokinetic parameters. During the batch tests, the pH values were constantly monitored to avoid the process inhibition. Moreover, the evaluation of the nitrification as well as denitrification rate, ammonium utilization rate (AUR) and nitrate utilization rate (NUR) tests were performed by adopting a modified protocol derived by Kristensen et al. (1992).

The soluble EPSs or soluble microbial products (SMPs) were obtained by centrifugation at 5000 rpm for 5 min, while the bound EPSs (EPS_{Bound}) were extracted by means of the thermal extraction method (among others Cosenza et al., 2013b). The extracted EPS_{Bound} and the SMP were then analysed for proteins by using the Folin

method with bovine serum albumin as the standard (Lowry et al., 1951), whereas the carbohydrates were measured according to DuBois et al. (1956), which yields results as glucose equivalent. Moreover, the sum of proteins and carbohydrates was considered as the total EPSs (EPS_T), according to the following expression:

$$EPS_T = \underbrace{EPS_P + EPS_C}_{EPS_{Bound}} + \underbrace{SMP_P + SMP_C}_{SMP} \quad (1)$$

where the subscripts ''P'' and ''C'' indicate the content of proteins and carbohydrates respectively in the EPS_{Bound} and SMP, that typically constitute the main fractions.

Membrane fouling has been analyzed by monitoring the total resistance (R_T) to membrane filtration which is calculated according to Equation 2, derived by the Darcy's law:

$$R_T = \frac{TMP}{\mu J} \quad (2)$$

where TMP is the transmembrane pressure (Pa), μ the permeate viscosity (Pa.s) and J the permeation flux (m s^{-1}).

R_T can be expressed as the sum between the intrinsic resistance of membrane (R_m) and the resistance due to membrane fouling (R_F). This latter can be fractionated according to Equation 3.

$$R_F = R_{PB} + R_{C,irr} + R_{C,rev} = R_T - R_m \quad (3)$$

where: R_{PB} is the irreversible resistance due to colloids and particles deposition into the membrane pore; $R_{C,irr}$ is the fouling resistance related to superficial cake deposition that can be only removed by physical cleanings (hydraulic/sponge scrubbing); $R_{C,rev}$ is the fouling resistance related to superficial cake deposition that can be removed by ordinary backwashing.

In order to analyze the specific fouling mechanisms the resistance-in-series (RIS) resistances method according to Di Trapani et al. (2014) has been applied.

The capillary suction time (CST) and the specific resistance to filtration (SRF) were measured in order to investigate the sludge dewaterability features (Vesilind, 1988; Peng et al., 2011). CST and SFR were measured in accordance with EN 14701-1 (2006) and EN 14701-2 (2006), by analyzing fresh samples collected from the anaerobic, anoxic, aerobic and MBR tanks. For further details on the adopted procedure, the reader is addressed to literature (Mannina et al., 2016).

3. Results and discussion

3.1. Organic carbon and nutrients removal

Figure 2 reports the pattern of influent COD (COD_{IN}), supernatant of MBR ($COD_{SUP,MBR}$) and effluent COD

(COD_{OUT}) throughout experiments (Figure 2a) as well as the COD removal efficiencies, expressed as total (η_{TOT}), biological (η_{BIO}) and physical contribution due to membrane filtration (η_{PHYS}).

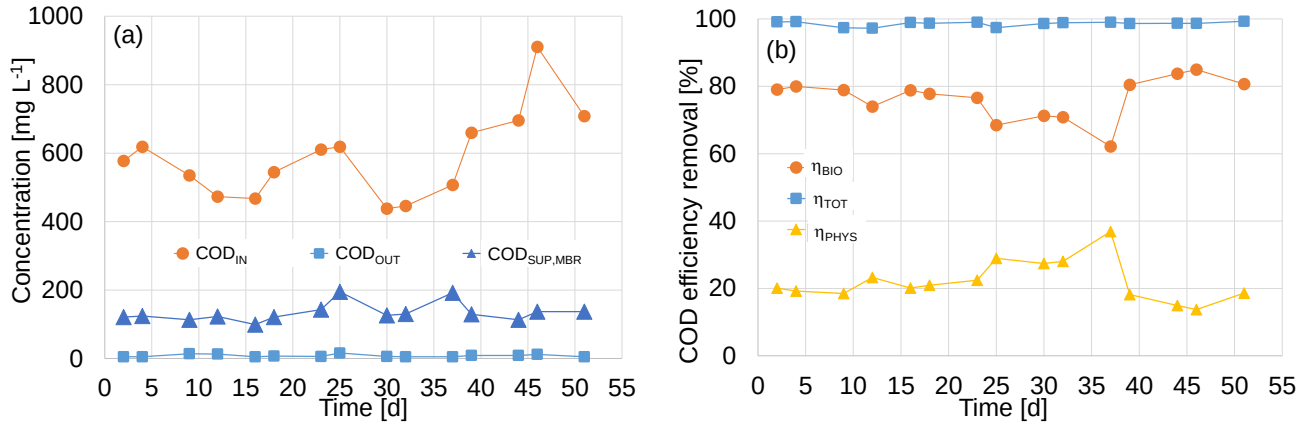

Figure 2. *Profile of COD_{IN}, $COD_{SUP,MBR}$ and COD_{OUT} (a); profiles of COD removal efficiencies expressed as biological (η_{BIO}), physical (η_{PHYS}) and total contribution (η_{TOT}) (b)*

The achieved results highlight that a very high total COD removal was obtained during experiments, with average value higher than 98%. The biological COD removal, evaluated prior to membrane filtration also showed a satisfactory activity of the biological consortium, as confirmed by the respirometric batch tests, reaching an average value during experiments of 77%. Nevertheless, it is worth noting the effect of membrane filtration that contributed to retain inside the bioreactor the particulate COD as well as the portion of the soluble COD characterized by average size higher than membrane porosity (0.04 μm). The achieved results confirmed the robustness of MBR systems towards organic carbon removal.

Referring to nitrogen removal, Figure 3 shows the pattern of influent and effluent ammonia, effluent nitrate (Figure3a) as well as the achieved performance in terms of nitrification (η_{nit}), denitrification (η_{denit}) and total nitrogen removal (ηN_{total}) (Figure 3b).

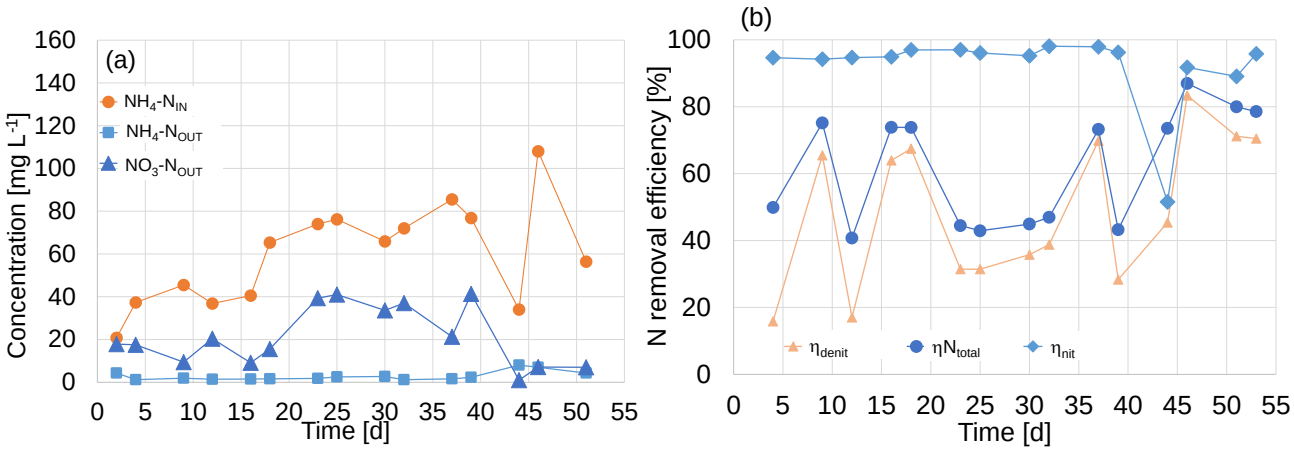

Figure 3. *Profile of NH_4-N_{IN}, NH_4-N_{OUT} and NO_3-N_{OUT} (a); performance of nitrification (μ_{nit}) denitrification (μ_{denit}) and total nitrogen removal (μN_{total}) during experiments (b)*

The obtained results highlight an excellent nitrification performance of the system, with efficiencies higher than 98% for most of the experiments, excepting experimental day 44, when a sudden sharp decrease of the nitrification efficiency occurred. However, this result is likely related to a sudden decrease of the inlet ammonia, due to dilution effect related to a rain event. Indeed, due to the specific composition of the feeding wastewater, we noticed higher ammonia dilutions during rain events.

It is worth noting that the system was able to guarantee high nitrification performances despite the increasing influent ammonia, up to 80-100 mg L^{-1}. This result is likely related to the presence of biofilm (mostly autotrophic) in the aerobic compartment that could sustain an almost complete nitrification throughout experiments.

The TN removal showed significant fluctuations during experiments, reaching an average value of 62%. This result reflected the fluctuations of the denitrification efficiency during experiments. Nevertheless, the denitrification efficiency showed in general an increasing trend in the last experimental days, likely due to the contribution of biofilm growth into the anoxic compartment.

In Figure 4 the profile of the influent and effluent PO_4-P concentrations (Figure 4a) are reported. Moreover, the assimilated or released PO_4-P concentration inside the anaerobic and aerobic tanks is reported in Figure 4b and Figure 4c, respectively. The average P removal efficiency was quite moderate, with average value close to 40.4%. The low performance of the biological phosphorous removal could be due to the increase of the ammonium loading rate during experiments and the consequent decrease of the C/N ratio value. Indeed, the

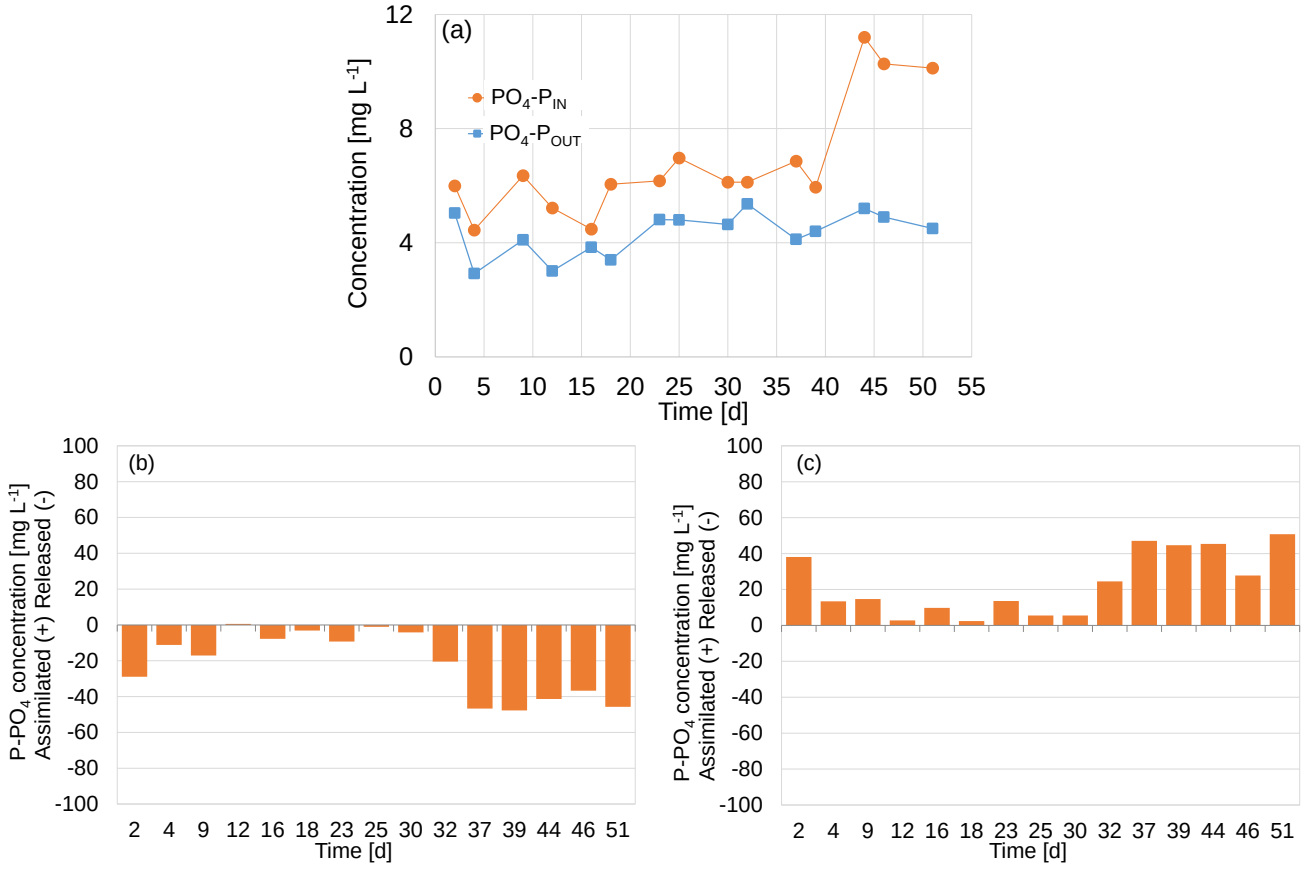

Figure 4. *Profile of the influent and effluent PO_4-P concentration (a); PO_4-P concentration released or assimilated inside the anaerobic (b) and aerobic tank (c).*

higher ammonium loading rate could promote an increased NO_3-N production that were recycled from the anoxic to the anaerobic tank, according to the UCT scheme. This fact could interfere with PAOs activity inside the anaerobic tank, promoting on the other hand the activity of denitrifying PAOs (DPAOs). Indeed, DPAOs have the capacity to grow under anoxic conditions with a very low rate using NO^{3+} and/or NO^{2+} as electron acceptor for P removal instead of oxygen, thus reducing the phosphorus removal efficiency (Parco et al., 2007).

3.2. Biomass respiratory activity and biokinetic/stoichiometric parameters

The respirometric batch tests were run for measuring the biomass activity level throughout experiments by measuring the main kinetic/stoichiometric parameters of both suspended and attached biomass. Figure 5 summarizes the average values achieved for both suspended and attached biomass

Concerning the suspended biomass, the measured parameters were in general consistent with literature results (Hauduc et al., 2011). The specific respiration rates ($SOUR_{max}$) and the maximum growth rates ($\mu_{H,max}$) of heterotrophic species showed a moderate decreasing trend during experiments. This result was mainly related to the fact that the UCT-MBMBR pilot plant was operated without sludge withdrawals, thus promoting the suspended biomass "ageing". Conversely, the increasing sludge age of the suspended consortium favored the

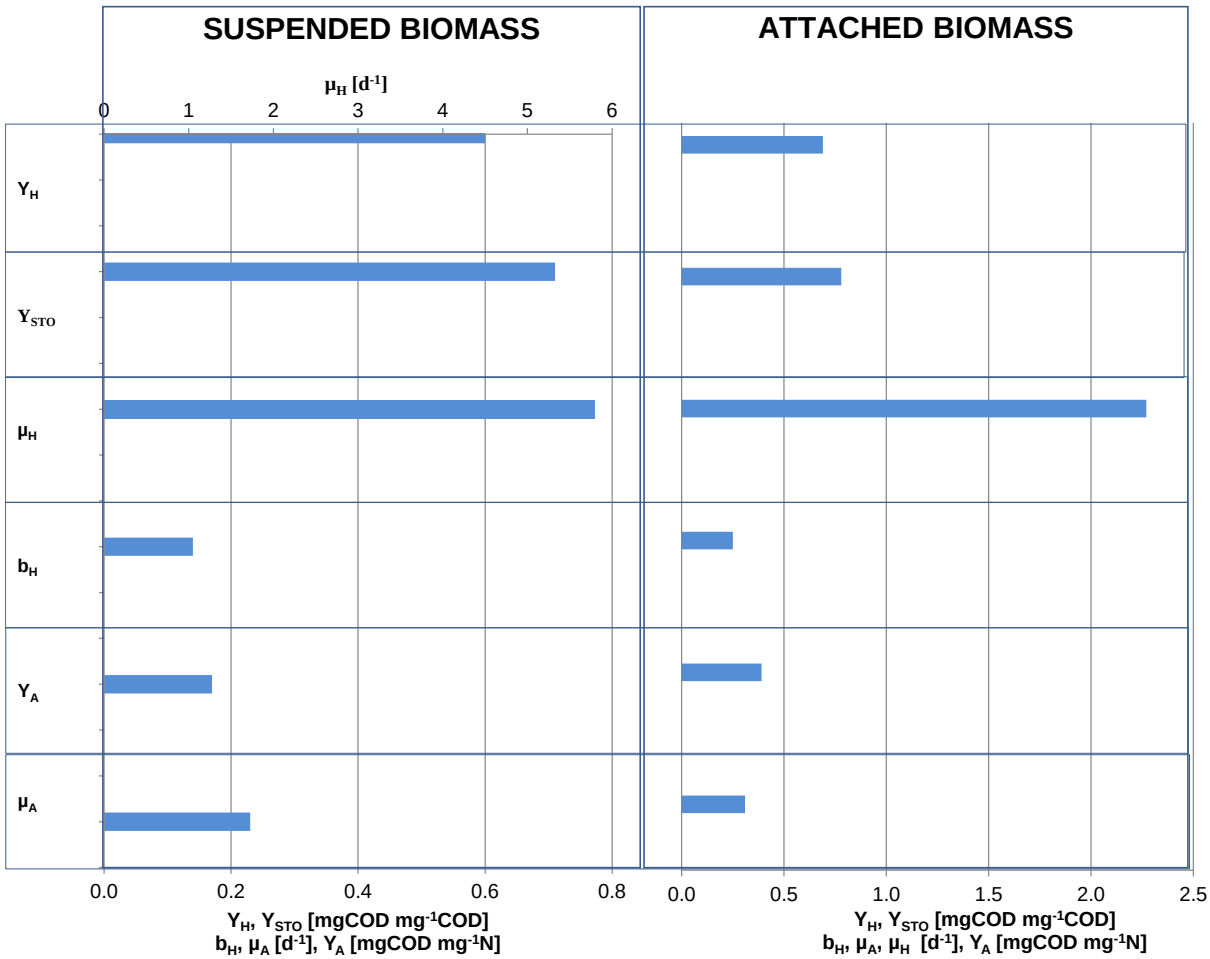

Figure 5. *Average kinetic and stoichiometric parameter values for both suspended and attached biomass.*

development of slow growing microorganisms, like the nitrifying species, that showed an increasing pattern during experiments. Referring to biofilm, it is worth noting that the kinetic/stoichiometric parameters of the autotrophic species were higher compared to the ones of suspended biomass, due to the high residence time, highlighting a sort of “specialization” of biofilm towards the nitrification process. The significant activity of autotrophic species supported the high nitrification level of the system, as previously mentioned.

Furthermore, it was also observed the occurrence of the storage phenomenon in both suspended and attached biomass, related to the ability of specific microorganisms to rapidly convert the external organic substrate into internal storage products under dynamic conditions (Majone et al., 1999; Di Trapani et al., 2014).

3.3. MLSS trend and biofilm growth

Figure 6 reports the pattern of suspended and attached biomass in the different compartments throughout experiments (Figure 6a-d). From the observation of Figure 6, one can notice a general increase of the suspended biomass concentration in the different compartments, related to the absence of sludge withdrawals. The biofilm showed a moderate development, either in the anoxic or aerobic compartment. This result could be related to the completion with the suspended biomass for the availability of the different substrates. In particular, it was also experienced a biofilm detachment into the aerobic compartment after day 25th; with biofilm

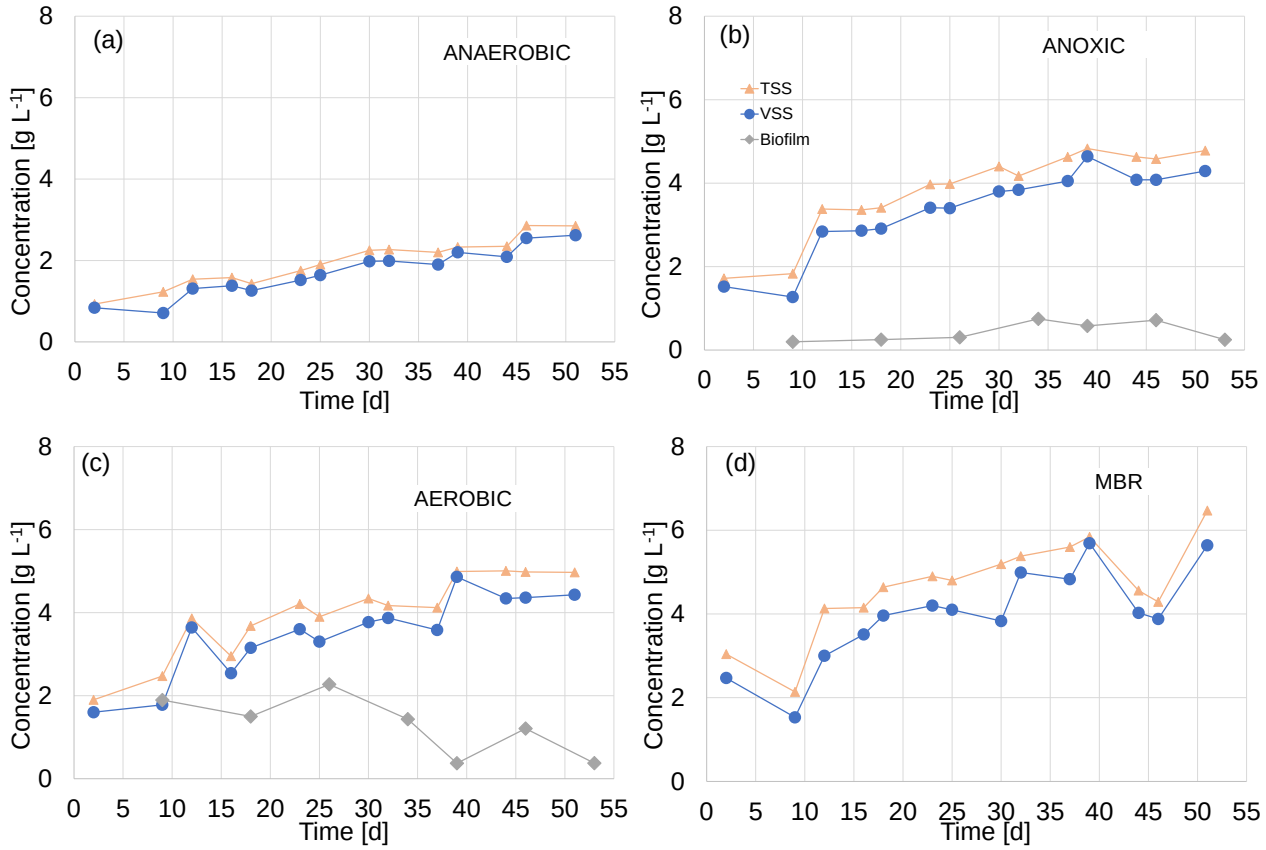

Figure 6. *Biomass trend during experiments, referring respectively to anaerobic (a), anoxic (b), aerobic (c) and MBR (d) compartment.*

concentrations down to 0.4 gTS L^{-1}. This behavior could be related to a stress effect on the biofilm caused by the specific environmental conditions and should contribute to increase the membrane fouling of the system.

3.4. EPS production

Figure 7 reports the pattern of EPS_T concentration during the experimental campaign, expressed as carbohydrates and proteins in microbial flocs (EPS_{Bound}) and dissolved in the bulk liquid (SMP),

From the observation of Figure 7, it is worth noting that the SMP concentration was almost negligible compared to the EPS_{Bound}, excepting some experimental days, especially at the beginning of the experiments. On the other hand, the protein fraction of EPS_{Bound} showed a general increasing trend throughout experiments, reaching values close to 250 mg $gTSS^{-1}$ in the MBR compartment. Such values were higher compared to what achieved in previous experiences with UCT-MBR systems (Cosenza et al., 2013b). This result could be likely due to biofilm detachment that might have contributed to make the mixed liquor more hydrophobic. This variation could likely promote the increase of the mixed liquor hydrophobicity, contributing to worsen the membrane filtration properties compromising the filtration properties of the cake layer, as better outlined in the following section.

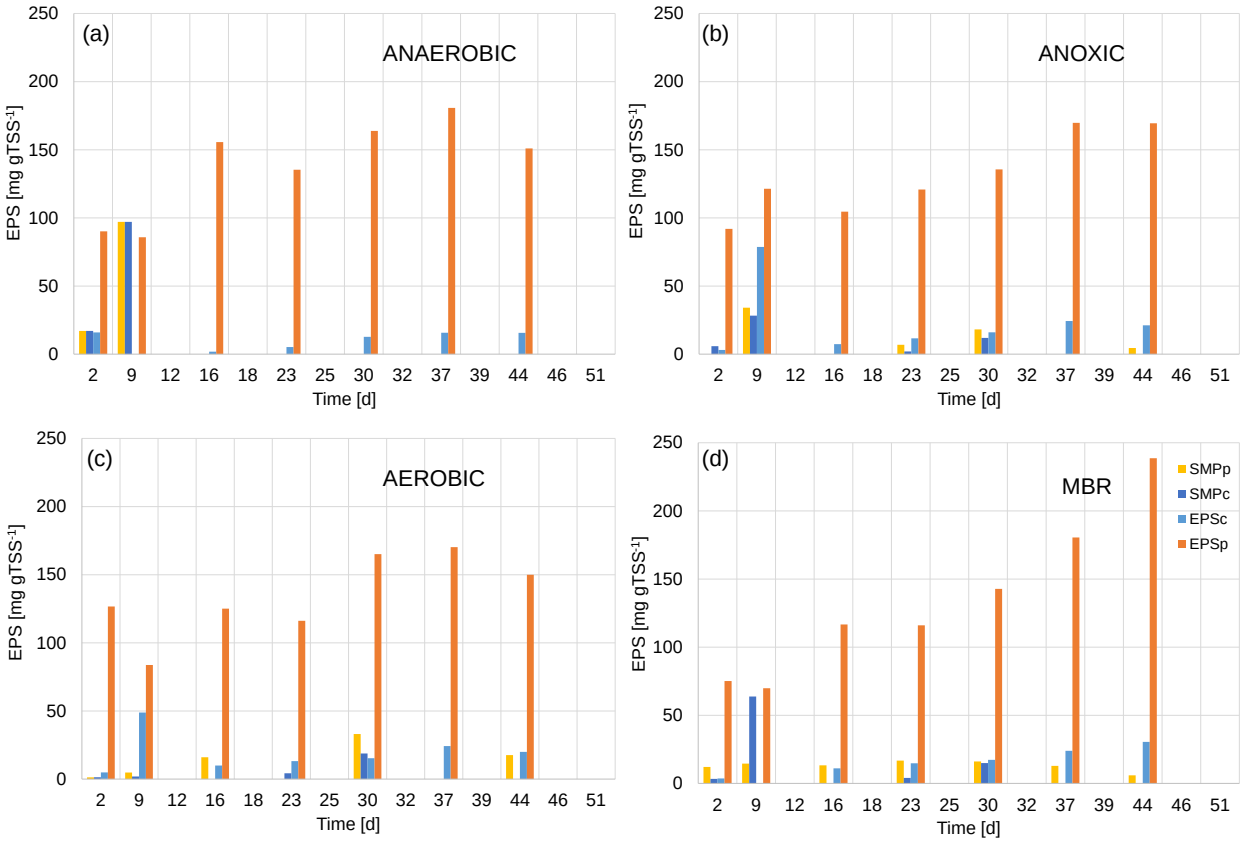

Figure 7. *Pattern of specific EPS_{Bound} and SMP inside the anaerobic (a), anoxic (b), aerobic (c) and MBR (d) reactor, respectively.*

3.5. Sludge dewaterability

The achieved results suggested that UCT-MBMBR pilot plant was characterized by a quite good sludge dewaterability. The CST values were almost constant and slightly affected by the MLSS concentration, with average values of 15.27, 17.27, 15.07 and 18.93 s for the anaerobic, anoxic, aerobic and MBR compartment, respectively.

Furthermore, also the low SRF values confirmed the good sludge filtration properties, with average values for the different compartment close to 4 10^{12} m kg^{-1}, significantly lower compared to what obtained by the same authors in previous experiences, when treating saline wastewater contaminated by hydrocarbons (Mannina et al., 2016). Moreover, the activated sludge filterability was mostly influenced by the specific EPS_{Bound} concentration (i.e., referred to MLSS concentration). Figure 8 shows the relationship between SRF and EPS_{Bound} inside each compartment.

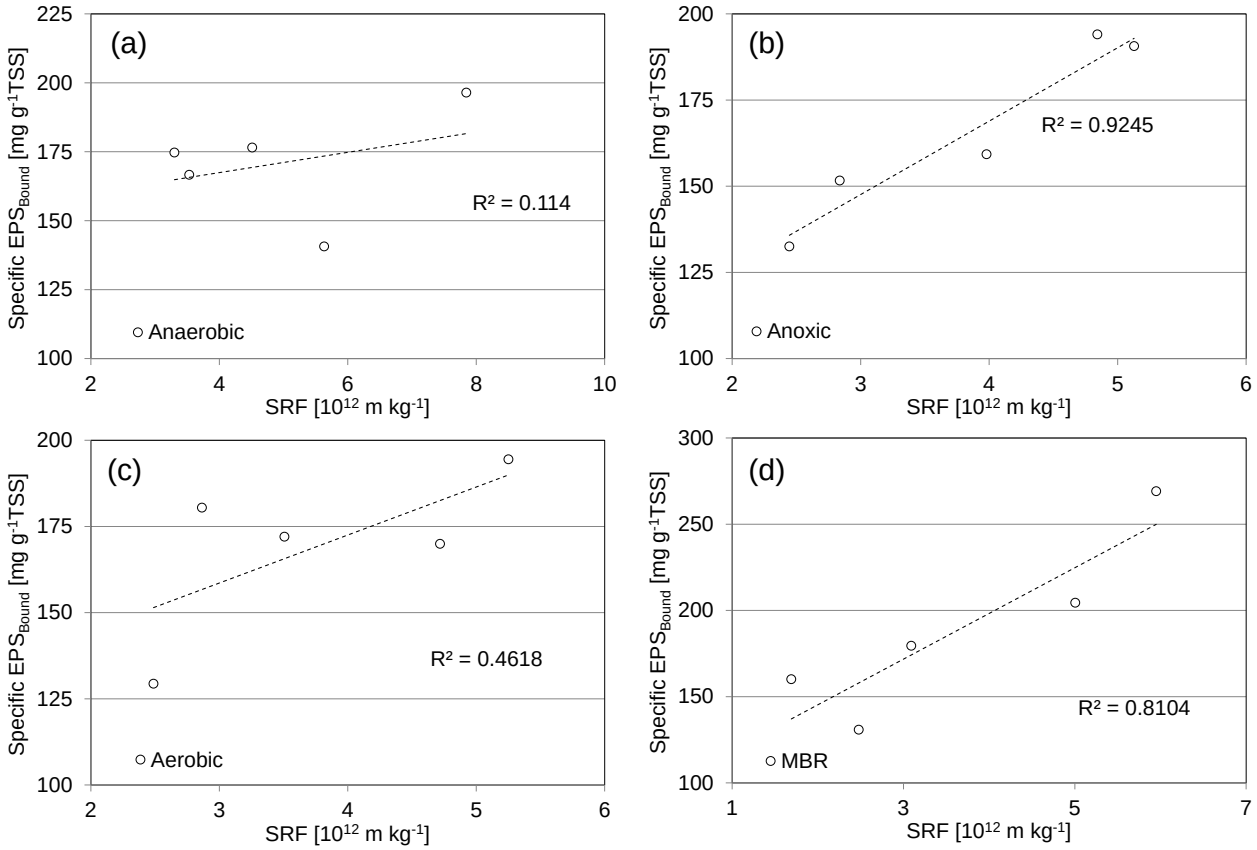

Figure 8. *Correlation between SRF and EPS_{Bound} inside the anaerobic (a), anoxic (b), aerobic (c) and MBR (d) reactor, respectively.*

3.6. Membrane filtration properties

Figure 9 reports the profile of R_T during experiments (Figure 9a) as well as the specific resistance contributions (Figure 9b) evaluated dividing each resistance, derived by applying the aforementioned RIS model, by the R_T. As noticeable from Figure 9a, four extraordinary physical cleanings were carried out during experiments that were necessary in order to prevent the TMP exceeding the critical values defined by the

membrane manufacturer (0.5–0.6 bar). As depicted in Figure 8b, the irreversible resistance due to superficial cake deposition ($R_{C,irr}$) was the mechanism that mostly affected the membrane filtration properties. Moreover, it was noticed the increase of the resistance due to pore blocking (R_{PB}) and a general worsening of the membrane filtration properties. This result could be due to the increase of the EPS_{Bound} fraction that could be enhanced by biofilm detachment phenomena occurred during experiments.

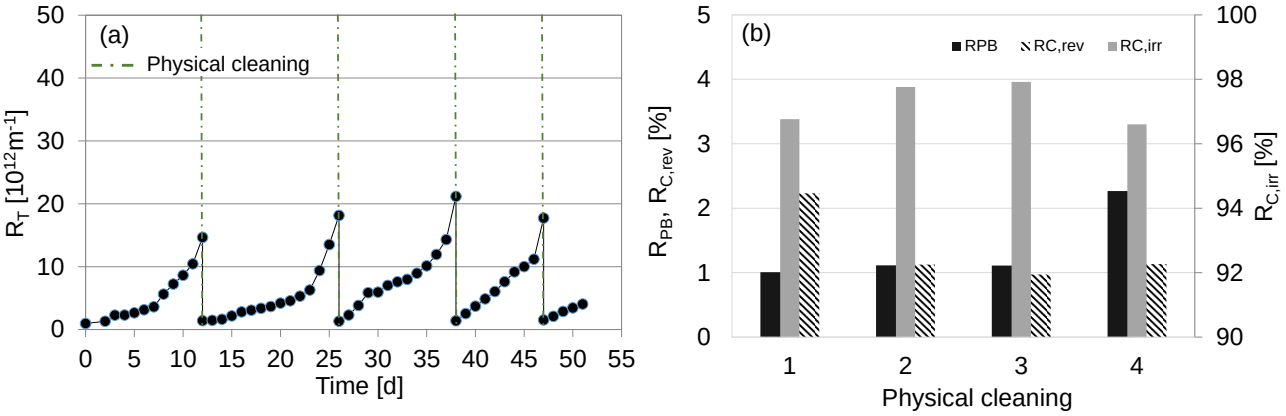

Figure 9. *Profile of total (a) and specific (b) resistances to filtration, respectively.*

The significant relationship that binds EPS with the filtration properties of the system is depicted in the graphs reported in Figure 10, where the main correlations between specific EPS fractions and resistance contributions are shown.

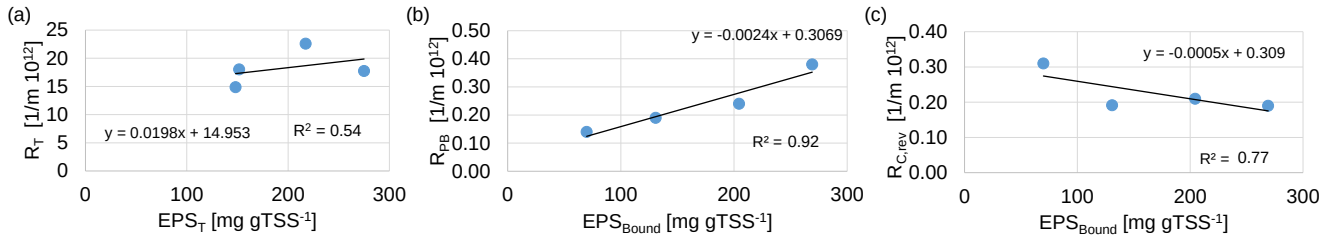

Figure 10. *Correlation between RT and EPST (a), RPB and EPSBound (b), RC,rev and EPSBound (c).*

According to the technical literature, (Judd and Judd, 2010), when the concentration of EPS_T in the mixed liquor increases, the membrane resistance increases as well, highlighting a worsening of the hydraulic performance of the system (see Fig. 10a). Moreover, it is worth noting that an increase of the EPS_{Bound} content promoted an irreversible fouling mechanism, with the increase of the R_{PB} (Figure 10b). On the other hand, the $R_{C,rev}$ significantly decreased with the increase of the EPS_{Bound} (Figure 10c). Such result might seem surprising at first sight. However, it is in good agreement with the following considerations: the increase of protein content in the MBR compartment (as average), which is recognized to be the most hydrophobic fraction, determined a more “bloated” cake layer, similarly to what happens in the bulking phenomenon that occurs in CAS plants

(Jenkins et al., 2003). Therefore, this situation significantly worsened the pre-filter effect of the cake layer (biological membrane). As a consequence, the foulants could reach more easily the internal pores of the membrane, thus promoting the increase of R_{PB} (Figure 10b). Moreover, the biofilm detachment, rich in polymeric substances, could negatively affect the permeability of the cake layer, likely formed on the membrane surface, in agreement with previous findings (Di Trapani et al., 2014).

4. Conclusions

This study has shown that a very high COD and nitrification performance can be achieved in a UCT-MBMBR system. In detail, the nitrification was excellent, with efficiencies higher than 98% for most of the experiments, despite the increasing ammonium loading rates (with influent ammonium concentrations up to 100 mgL^{-1}). The presence of the autotrophic biofilm inside the aerobic reactor of the UCT-MBMBR pilot plant enabled to sustain nitrification throughout experiments. The TN removal showed a lower performance with average efficiency equal to 62%. This result reflected the fluctuations of the denitrification efficiency that could be hindered by the decreasing C/N ratio. Conversely, the average P removal efficiency was quite moderate, likely due to the increase of the ammonium loading rate that could promote an increased NO_3-N recycled from the anoxic to the anaerobic tank, interfering with PAOs activity inside the anaerobic tank. The respirometric batch test enabled to derive the kinetic constant of both suspended biomass and biofilm, highlighting a sort of "specialization" of the two biomasses with the suspended more active in the organic carbon removal, whilst the aerobic biofilm more active towards nitrification. The EPS production highlighted higher protein production in the bound fraction, likely due to the contribution of the detached biofilm. Referring to membrane fouling, the irreversible resistance due to superficial cake deposition mostly affected the membrane filtration properties. As final remark, in order to successfully apply a UCT-MBMBR system for carbon and nutrients removal it is suggested to reduce the sludge age of the suspended biomass, in order to improve the biological phosphorus removal, while the high retention time of the attached biomass would sustain the complete nitrification of the influent ammonia.

Acknowledgements

This work forms part of a research project supported by grant of the Italian Ministry of Education, University and Research (MIUR) through the Research project of national interest PRIN2012 (D.M. 28 dicembre 2012 n. 957/Ric − Prot. 2012PTZAMC) entitled "Energy consumption and GreenHouse Gas (GHG) emissions in the wastewater treatment plants: a decision support system for planning and management − http://ghgfromwwtp.unipa.it" in which the first author of this paper is the Principal Investigator.

References

[1] APHA, 2005. Standard Methods for the Examination of Water and Wastewater. APHA, AWWA and WPCF, Washington DC, USA.

[2] Chu, L., Wang, J., 2011. Comparison of polyurethane foam and biodegradable polymer as carriers in

moving bed biofilm reactor for treating wastewater with a low C/N ratio. Chemosph. 83, 63–68.

[3] Cosenza, A., Di Bella, G., Mannina, G., Torregrossa, M., Viviani, G. 2013a. Biological Nutrient Removal and Fouling Phenomena in a University of Cape Town Membrane Bioreactor Treating High Nitrogen Loads. J. Environ. Eng. 139, 773-780.

[4] Cosenza, A., Di Bella, G., Mannina, G., Torregrossa, 2013b. The role of EPS in fouling and foaming phenomena for a membrane bioreactor. Bioresour. Technol. 147, 184–192.

[5] Di Trapani, D., Christensson, M., Torregrossa, M., Viviani, G., Ødegaard, H., 2013. Performance of a hybrid activated sludge/biofilm process for wastewater treatment in a cold climate region: influence of operating conditions. Biochem. Eng. J. 77, 214–219.

[6] Di Trapani, D., Di Bella, G., Mannina, G., Torregrossa, M., Viviani, G., 2014. Comparison between moving bed-membrane bioreactor (MB-MBR) and membrane bioreactor (MBR) systems: Influence of wastewater salinity variation. Bioresour. Technol. 162, 60–69.

[7] Di Trapani, D., Di Bella, G., Mannina, G., Torregrossa, M., Viviani, G., 2015. Effect of C/N shock variation on the performances of a moving bed membrane bioreactor. Bioresource Technology 189, 250–257

[8] DuBois, M., Gilles, K.A., Hamilton, J.K., Rebers, P.A., Smith, F., 1956. Colorimetric method for determination of sugars and related substances. Anal. Chem. 28, 350–356.

[9] EN 14701-1. European Standard. Characterization of sludges - Filtration properties – Part 1: Capillary Suction Time (CST). European Committee for Standardization. March 2006.

[10] EN 14701-2. European Standard. Characterization of sludges - Filtration properties – Part 2: Determination of the specific resistance to filtration. European Committee for Standardization. March 2006.

[11] Ekama, G.A., Siebritz, I.P., Marais, G.R., 1983. Considerations in the process design of nutrient removal activated sludge processes. Wat. Sci. Tech. 15 (3-4), 283 – 318.

[12] Fu, Z., Yang, F., Zhou, F., Xue, Y., 2009. Control of COD/N ratio for nutrient removal in a modified membrane bioreactor (MBR) treating high strength wastewater. Bioresour. Technol. 100, 136–141.

[13] Hauduc, H., Rieger, L., Ohtsuki, T., Shaw, A., Takács, I., Winkler, S., Héduit, A., Vanrolleghem, P.-A., Gillot, S., 2011. Activated sludge modelling: development and potential use of a practical applications database. Water Sci. Technol. 63, 2164–2182.

[14] Hu, X., Xie, L., Shim, H., Zhang, S. and Yang, D. 2014. Biological Nutrient Removal in a Full Scale Anoxic/Anaerobic/Aerobic/ Pre-anoxic-MBR Plant for Low C/N Ratio Municipal Wastewater Treatment. Chinese Journal of Chemical Engineering, 22 (4) 447—454.

[15] Ivanovic, I., and Leiknes, T. 2008. Impact of aeration rates on particle colloidal fraction in the biofilm membrane bioreactor (BF-MBR). Desalin. 231(1–3), 182–190.

[16] Jenkins, D., Richard, M.G., Daigger, G.T., 2003. Manual on the Causes and Control of Activated Sludge Bulking, Foaming and Other Solids Separation Problems. IWA Publishing, London.

[17] Judd, S.J., Judd, C., 2010. Principles and Applications of Membrane Bioreactors in Water and Wastewater Treatment, second ed. Elsevier, London, UK.

[18] Kermani, M., Bina, B., Movahedian, H., Ami, M.M., Nikaein, M., 2008. Application of moving bed biofilm

process for biological organics and nutrients removal from municipal wastewater. Am J. Environ. Sci. 4(6), 675–682.

[19] Kristensen, G.H., Jørgensen, P.E., Henze, M., 1992. Characterization of functional microorganism groups and substrate in activated sludge and wastewater by AUR, NUR and OUR. Water Sci. Technol. 25, 43–57.

[20] Leyva-Díaz, J.C., Calderón, K., Rodríguez, F.A., González-López, J., Hontoria, E., Poyatos, J.M., 2013. Comparative kinetic study between moving bed biofilm reactor-membrane bioreactor and membrane bioreactor systems and their influence on organic matter and nutrients removal. Biochem. Eng. J. 77, 28–40.

[21] Li, C., Wang, T., Zheng, N., Zhang, J., Ngo, H.H., Guo, W., Liang, S. 2013. Influence of organic shock loads on the production of N2O in denitrifying phosphorus removal process. Bioresour. Technol. 141, 160–166.

[22] Lowry, O.H., Rosebrough, N.J., Farr, A.L., Randall, R.J., 1951. Protein measurement with the Folin phenol reagent. J. Biol. Chem. 193, 265–275.

[23] Lu, Q., Wu, H., Li, H., Yang, D., 2015. Enhanced biological nutrient removal in modified carbon source division anaerobic anoxic oxic process with return activated sludge preconcentration. Chinese Journal of Chemical Engineering 23, 1027–1034.

[24] Majone, M., Dircks, K., Beun, J.J., 1999. Aerobic storage under dynamic conditions in activated sludge processes. The state of the art. Water Sci. Technol. 39, 61–73.

[25] Mannina, G., Cosenza, A. Di Trapani, D., Capodici, M., Viviani, G. 2016. Membrane bioreactors for treatment of saline wastewater contaminated by hydrocarbons (diesel fuel): An experimental pilot plant case study, Chem. Eng. J., 291(1), 269–278.

[26] Naessens, W., Maere, T., Nopens, I., 2012. Critical review of membrane bioreactor models – Part 1: biokinetic and filtration models. Bioresour. Technol. 122, 95–106.

[27] Ødegaard, H., 2006. Innovations in wastewater treatment: the moving bed biofilm process. Water Sci. Technol. 53, 17–33.

[28] Peng, G., Ye, F., Li, Y., 2011. Comparative investigation of parameters for determining the dewaterability of activated sludge. Water Environ. Res. 83(7), 667-671.

[29] Poyatos, J. M., Molina-Muñoz, M. A., Delgado, F., González-López, J. and Hontoria, E. 2008. Flux influence on membrane fouling in a membrane bioreactor system under real conditions with urban wastewater. J. Environ. Sci. Health A., 43(14), 1685–1691.

[30] Stephenson, T., Judd, S.J., Jefferson, B., Brindle, K., 2000. Membrane Bioreactors for Wastewater Treatment. IWA Publishing, London, UK.

[31] Veselind P.A., 1988. Capillary suction time as a fundamental measure of sludge dewaterability. J. Water Pollut. Control Fed. 60, 215-220.

[32] Wang, X.J., Xia, S.Q., Chen, L., Zhao, J.F., Renault, N.J., Chovelon, J.M., 2006. Nutrients removal from municipal wastewater by chemical precipitation in a moving bed biofilm reactor, Process Biochem. 41(4), 824–828.

[33] Wanner, J., Cech, J.S., Kos, M., 1992 New process design for biological nutrient removal, Water Sci.

Technol. 25(4-5), 445–448.

[34] Wanner, J. 2002 Control of filamentous bulking in activated sludge. In: Encyclopedia of Environmental Microbiology, Bitton G Ed., 1306-1315. John Wiley & Sons Inc. New York USA.

[35] Yang, S., Yang, F., Fu, Z., Wang, T., Lei, R. 2010. Simultaneous nitrogen and phosphorus removal by a novel sequencing batch moving bed membrane bioreactor for wastewater treatment. J. Hazard. Mat. 175 551–557.

Yang, W., Syed, W., Zhou, H., 2014. Comparative study on membrane fouling between membrane-coupled moving bed biofilm reactor and conventional membrane bioreactor for municipal wastewater treatment. Water Sci. Technol. 69, 1021–1027.