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Assessment of dynamic membrane filtration for biological treatment of old landfill leachate

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Abstract

This study investigated the behaviour of dynamic membrane (DM) filtration for the treatment of stabilised landfill leachate in a bench-scale pre-anoxic and aerobic submerged dynamic membrane bioreactor (DMBR). Four meshes with different openings (10, 52, 85 and 200 µm) were tested to support the development of DM. Differences were observed among the meshes in supporting the development of the cake layer constituting the DM. The treatment of landfill leachate had an impact on sludge characteristics resulting in deteriorated filtration performance of the DM. Effluent turbidity was often higher than 100 NTU for larger mesh pore size (85 and 200 µm). Low effluent turbidity was achieved with meshes with 10 and 52 µm (13±2 and 26±4 NTU, respectively) although at membrane fluxes lower than 10 L m^{-2} h^{-1}. The bioreactor exhibited a moderate organics removal of 50-60% and an ammonia oxidation between 80 and 90%. Incomplete nitrification was observed due to increased concentrations of free ammonia and free nitrous acid, with nitrite effluent concentrations up to 1062 mgNO_2^--N L^{-1}. Due to the large presence of refractory organic matter in landfill leachate, denitrification was limited resulting in a total nitrogen removal of approximately 20%.

Keywords: Dynamic membrane; fouling; landfill leachate; mesh filtration; nitrification; denitrification.

1. Introduction

Sanitary landfill has been acknowledged as the most economically viable ultimate disposal option for municipal solid waste in most parts of the World, despite being placed at the bottom of waste management hierarchy (Fudala-Ksiazek et al., 2016). A major concern arising during landfill operation is the production of leachate resulting from the infiltration of water through the landfill body and the decomposing of waste. If not properly managed, leachate could severely contaminate groundwater sources, raising concerns regarding the protection of natural environment and public health (Renou et al., 2008).

Landfill leachate (LFL) treatment is challenging due to the high levels of contaminants including organics, ammonia, inorganic substances, heavy metals and toxic hydrocarbons (aromatic and phenolic compounds) together with the variability in its quantity and quality in both space and time (Kulikowska and Klimiuk, 2008; Renou et al., 2008). Moreover, the worldwide application of recent environmental legislation is changing the waste management chain reducing the disposal to landfills and, as a result, changing the leachate production and composition (Fudala-Ksiazek et al., 2016).

Biological processes have been proved to be effective in treating young leachates whereas their efficacy reduces with the increase of leachate age due to a shortage of biodegradable matter and an increase of refractory organics (Brennan et al., 2017; Mohammad-pajooh et al., 2017; Renou et al., 2008; Oloibi et al., 2017).

Membrane bioreactor (MBR), which consists in the integration of microfiltration or ultrafiltration (MF/UF) membranes with biological reactors, has gained much appreciation over the last decade and has been perceived as an advanced treatment process considering its excellent effluent quality and flexible operation (Judd, 2011). Studies on leachate treatment have demonstrated that MBRs are very effective under a wide range of loading conditions as compared to conventional biological treatment systems, particularly in treating LFL from old

landfills (Alvarez-Vazquez et al., 2004; Hashisho and El-Fadel, 2016). However, the application of high loading conditions, long hydraulic retention time (HRT) and solids retention time (SRT) and the high concentrations of contaminants can increase membrane fouling (Ahmed and Lan, 2012). In addition, excessive amount of humic and fulvic acids usually present in LFL have shown to speed up membrane fouling (Sutzkover-Gutman et al., 2010). In a recent review on MBR application treating LFL, Hashisho and El-Fadel (2016) concluded that membrane fouling was the main bottleneck in the widespread application of MBR in leachate treatment due to its high fouling potential especially while treating stabilised LFL.

In this regard, dynamic membranes (DMs) could represent an innovative approach by purposefully exploiting fouling as a mean for solid liquid separation (Alibardi et al., 2014, 2016; Saleem et al., 2016; Xiong et al., 2016; Zhang et al., 2010). DM is defined as a self-forming and regenerative fouling surface that is formed by the deposition of suspended solids, colloids and microbial cell particles over a coarse underlying support material (Ersahin et al., 2012; Li et al., 2011; Liu et al., 2009).

Most of the studies on DM have been carried out on synthetic or real municipal wastewater under aerobic or anaerobic conditions and for anaerobic sludge digestion (Alibardi et al., 2014, 2016; Saleem et al., 2016; Ersahin et al., 2016; Jeison et al., 2008; Li et al., 2011; Liu et al., 2009; Kiso et al., 2000; Hu et al., 2016; Xiong et al., 2016; Zhang et al., 2010). Xie et al. (2014) studied the performances of an anaerobic dynamic MBR for the treatment of leachate by using a 40 µm mesh as support material. Although these authors achieved solids retentions of the DM that were not comparable to those from MF/UF membranes, they reported a better effluent quality than conventional anaerobic treatment systems. To the best knowledge of the authors, no studies have yet evaluated the optimisation of organic matter and nitrogen removal for biological LFL treatment by using DMs. Similarly, the effect of the use of

meshes with different pore sizes on the filtration performances of DMs treating LFL is also lacking.

This study aimed at evaluating the application of DMs in anoxic-aerobic process for the treatment of LFL from an old landfill. In particular, the effect of the use of different mesh sizes on the development of the DM was evaluated. The behaviour of developed DM was studied in conjunction with the effect of change in feed characteristics and operating conditions.

2. Materials and methods

2.1. Experimental setup

The study was conducted using a laboratory-scale, continuously mixed, anoxic-aerobic system (Fig. 1a). The experimental setup consisted of a pre-anoxic tank with a working volume of 2.8 L connected to an aerobic tank with a working volume of 7.5 L. The tanks were made up of 5 mm thick Plexiglas cylinders. The internal diameter was 24 cm and 18 cm for aerobic and anoxic tanks, respectively, while depth was 30 cm for both tanks.

The filtration modules were constituted by a nylon mesh wounded over a cylindrical frame. The frame was a plastic body having an external diameter of 15 mm and a length of 70 mm with uniformly distributed openings of 5 mm X 3 mm. The total surface area of the filtration module was 33 cm^2 and approximately 61% (ca. 20 cm^2) was the effective filtration area of each mesh. Three filtration modules were continuously immersed in the aerobic vessel and operated in parallel, resulting in a total effective filtration area of 60 cm^2. Filtration flux were controlled through a three-line peristaltic pump (Watson Marlow SCI 400) which was connected to the three modules.

Four different meshes with pore sizes of 10, 52, 85 and 200 µm were tested (Table 1). Meshes with porosities of 10, 85 and 200 µm were initially evaluated; however, due to changes in

filtration behaviour of the sludge of the bioreactor, after 105 days of continuous operation the mesh with openings of 200 µm was replaced with a new one of 52 µm pore size.

The study was performed at ambient temperature (21±1 °C). Aeration of the aerobic tank was provided by a small air pump and diffusers. The air flow was controlled by using an air flowmeter (ColeParmer 1-800-323-4340). Leachate was fed to the anoxic tank through a peristaltic pump (Watson Marlow SCI 400) connected to a level sensor. Sludge recirculation flow was approximately four to five times the influent flow and was provided by means of a peristaltic pump (Watson Marlow SCI 400). The two bioreactors were kept completely mixed by using two overhead stirrers (LS F201A0151, VELP Scientifica). Solids retention time was maintained at 30-40 days.

2.2. Inoculum and Feed

Sludge collected from a full-scale municipal wastewater treatment plant (Padova, Italy) was used as inoculum. The sludge had a total suspended solids (TSS) concentration of 8.7 g L^{-1} and volatile suspended solids (VSS) of 5.4 g L^{-1}.

The feed to the reactor consisted of raw LFL collected from an old (> 25 years) landfill site located in Veneto Region, Italy. The characteristics of the LFL samples are reported in Table 2. The LFL sample used for this study can be considered as stabilised and typical of old landfills (Kjeldsen et al., 2002). The leachate was collected approximately every month and stored at 4 °C before use. To ensure the availability of essential micronutrients to support biomass activity following micronutrients were added in the feed wastewater: $Na_2MoO_4*2H_2O$ (0.22 mg Mo L^{-1}), $ZnSO_4*7H_2O$ (0.23 mg Zn L^{-1}), $CuSO_4*5H_2O$ (0.128 mg Cu L^{-1}), $NiCl_2*6H_2O$ (0.1 mg Ni L^{-1}), H_3BO_4 (0.007 mg B L^{-1}), Ne_2SeO_3 (0.06 mg Se L^{-1}), $MnCl_2*4H_2O$ (0.56 mg Mn L^{-1}) and $CoCl_2*6H_2O$ (0.124 mg Co L^{-1}).

Since the leachate used in this study has low BOD_5/N ratio, after approximately two months of operation, sodium acetate was also added with the leachate to the anoxic vessel in order to support the denitrification process. The amount of sodium acetate was provided to sustain the denitrification process.

2.3. Short-term filtration experiments

Short-term filtration experiment is a simple way to evaluate the performance of the coarse meshes used to develop DM (Li et al., 2012). These experiments were performed in a separate filtration system according to the procedure previously described in Saleem et al. (2017). Briefly, filtration was performed under a constant transmembrane pressure (TMP) of 3.43 kPa provided by the hydrostatic water head maintained above the filtration module connected to a 5 L stirring tank by a peristaltic pump (Fig. 1b). Filtration fluxes were estimated by measuring the time required to collect a known volume of permeate.

Short-term gravity driven filtration experiments were carried out with 200, 85 and 10 μm meshes. New meshes were used in the filtration module. The experiments were carried out with the initial inoculum and with the sludge sampled from the aeration tank after 67 days of continuous bioreactor operation (bulk sludge). TSS and VSS concentration inside the bioreactor on 67^{th} day of the continuous bioreactor operation was 7.4 and 4.3 g L^{-1} respectively.

Only for the experiments with bulk sludge, the filtration fluxes were increased to approximately 100 L m^{-2} h^{-1} by means of a peristaltic pump (Watson Marlow 505U) when the fluxes reduced to less than 5 % of the initial values.

2.4. Dynamic membrane operation and cleaning

Periodical cleaning of the excessively fouled DM layer was performed at the same time for all meshes when the TMP values were higher than 20 kPa or the fluxes were lower than 2 L m^{-2} h^{-1} for any of the mesh under investigation (set as the lower limit for this study). The meshes were cleaned in situ (i.e. inside the aerobic bioreactor) with the help of a brush.

Since the formation of DM layer after every cleaning operation could greatly compromised the effluent quality in terms of suspended solids removal (Alibardi et al., 2014; 2016), after every cleaning operation, a constant hydrostatic water head of 1.7 kPa was applied to the filtration module to establish high initial filtration fluxes in order to expedite the process of DM formation (Saleem et al., 2016). After the development of DM layer, characterised by the production of a "clear" permeate (visual inspection), constant flux filtration operation (to maintain the design HRT) was resumed. The permeates collected during this interval were returned to the bioreactor. A similar recirculation strategy was also proposed by Ersahin et al. (2012), Alavi Moghaddam et al. (2002) and Fan and Huang (2002) for the start-up of DMs systems.

2.5. Analytical Method and Measurements

Total suspended solids (TSS), volatile suspended solids (VSS), ammonium, nitrates, nitrites nitrogen, total phosphorous, 5-day biochemical oxygen demand (BOD_5), were measured according to standard methods (APHA, AWWA, WEF 2012). Organics matter and alkalinity were estimated measuring the total carbon (TC) and total organic carbon (TOC) by using Shimadzu TOC-V_{CSN} analyser. Inorganic carbon (IC) was calculated as difference between TC and TOC.

The concentrations of free ammonia (FA) and free nitrous acid (FNA) were estimated according to Anthonisen et al. (1976).

Transmembrane pressure (TMP) was measured separately for each DM by using an electronic pressure gauge (COMARK C9505/IS, Pressure Meter, 0–30 PSI). Darcy's equation was used to estimate total DM resistance as follows (Li et al., 2012):

$$R = \frac{\Delta P}{\mu \cdot J} \quad (1)$$

Where J is the permeate flux, ΔP is TMP across the membrane, μ is the viscosity of the permeate (assumed of clean water), and R is total membrane resistance.

Dissolved oxygen (DO) concentration inside the bioreactors was monitored by using a DO meter (HANNA HI 9147). Effluent turbidity and pH were measured using a turbidimeter (HACH 2100 P ISO TURBIDIMETER) and a pH-meter (Crison GLP 22), respectively. Average daily fluxes from the three DM modules were estimated by dividing the volume of the filtrate collected from each filtration module by the filtration area of each module.

3. Results and Discussion

3.1. Dynamic membrane behaviour

Filtration was started up applying high fluxes in order to speed up the formation of the cake layer on the mesh supports (Saleem et al., 2016). Effluent quality improved rapidly, indicating a quick formation of DM for all meshes and confirming previous results obtained under batch and continuous conditions (Alibardi et al., 2016; Saleem et al., 2017).

The TMP of the different meshes showed similar trends characterised by a typical progressive increase in value during operational filtration (Fig. 2). However, the behaviour of these trends showed a significant change during the study (Fig. 2). For the first 25-30 d, a slow and gradual rise in TMP was observed, according to the local flux theory proposed for conventional membranes (Cho and Fane, 2002). The first week of operation was characterised

by very low and stable TMP of approximately 1-2 kPa for all meshes, irrespective of the considerable difference in porosities. Afterwards, TMP values gradually and almost steadily increased up to approximately 60 kPa in about 20 days. Owing to the high TMP, the meshes were cleaned following the procedures described in Section 2.4. Thereafter, contrariwise to the first 30 days of operation, the TMP trends were characterised by sharp and fast increases after every cleaning procedure with different maximum values for each mesh pore size.

The result obtained over the first 30 days of operation confirms a previous study (Saleem et al., 2017) which demonstrated that the mesh pore size does not significantly affect the filtration flux (Fig. 2). However, after the first 30 days of operation (corresponding to the first filtration period ending with mesh cleaning), the mesh with larger pore-size showed the lowest maximum TMP values when reaching the minimum flux value of 2 L m^{-2} h^{-1} (Fig. 2). This suggests that a lower resistance (Fig. 3) was obtained when DM developed on larger pore size. It is of note that the maximum TMP achieved during each filtration cycle (where a filtration cycle can be identified between two cleaning procedures) decreased during the entire duration of the study (Fig. 2). This is particularly evident for the mesh with large pore size (i.e. 200 and 85 µm) where maximum TMP decreased from approximately 60 kPa to less than 20 kPa (Fig. 2a and 2b).

The variation of TMP also affected the filtration fluxes (Fig. 2). Despite the use of a peristaltic pump to control effluent flow, fluxes through each filtration module resulted variable and the observed fluxes were in general higher for larger mesh pore sizes. Due to the very variable *J*, HRT of the system also showed a fluctuating profile with an average value of 20±9 days.

A variation of the characteristics of the DM can be highlighted also by the trends of the membrane resistance (*R*) as calculated by equation 1 (Fig. 3). During the first filtration cycle of approximately 30 days of operation, DM resistance increased gradually from

approximately 1.0×10^{12} to 1.0×10^{14} m^{-1} for all the meshes under investigation, without large differences among them (Fig. 3). Results suggest that the resistance of the DM was the main contributor of the total resistance of the filtration module, as the intrinsic *R* of all the meshes measured using tap water was of the order of 1×10^{9} m^{-1} (Table 1).

The behaviour of DM *R* changed after the first 30 days of operation, showing a much faster build-up at any filtration cycle (Fig. 3). This observation confirms what observed also for TMP, despite *J* resulted variable. The *R* measured immediately after every cleaning procedure was always higher than the values measured during the first filtration cycle. This result indicates either that the cleaning process was not able to completely remove the cake layer from the mesh or that the sludge characteristics changed over the experimental study with a measurable impact on DM formation. Moreover, the initial resistance increased with decreasing mesh pore size (Fig. 3).

The change of the characteristics of the sludge and the impact on DM formation is also evident from the measured effluent turbidity (Fig. 3). Very low effluent turbidity values were measured over the first filtration cycle (with values less than 5 NTU), for all the three meshes, demonstrating an excellent solid rejection of the DM, regardless of the difference in mesh size. Despite TMP greatly increased during the first filtration cycle (Fig. 2), effluent turbidity remained almost stable (Fig. 3) confirming the formation of a stable cake layer over the mesh independently, once more, of the pore size. After the first cleaning procedure, the effluent turbidity increased and remained above the low values measured during the first 30 days for the rest of the experimental period (Fig. 3). The related mean turbidity values for 200, 85 and 10 μm meshes were 2126±253, 615±81 and 37±20 NTU, respectively. As a result, the different DM showed different suspended solid rejection. The DM developed over the mesh of 10 μm exhibited very high suspended solids rejection which was always above 95%, while

the meshes with openings of 85 and 200 μm achieved mean solid rejection of approximately 85 and 55 %, respectively (data not shown).

In order to find a trade-off between high filtration fluxes observed for 200 and 85 μm meshes and high effluent quality observed for 10 μm mesh, it was decided to evaluate the performance of 52 μm mesh by starting on day 104 of the continuous bioreactor operation. The results showed that the solids rejection performance of 52 μm mesh was comparable to that of 10 μm in terms of effluent turbidity (Fig. 3c and 3d). Furthermore, the average filtration fluxes of the DM developed on the mesh with openings of 52 μm was higher than those obtained for 10 μm mesh and was rather comparable to 200 and 85 μm meshes (Fig. 2). The difference in behaviour of the DMs developed over the different meshes between the first filtration cycle (first 30 days) and the cycles during the following three months of operation could be due to two reasons. On the one hand, the cleaning procedures based on brushing, could have been not effective in completely removing the material deposited over the mesh. On the other hand, the applied operating conditions could have changed the sludge characteristic, increasing its fouling propensity. Li et al. (2016) observed by scanning electron microscopy that a significant amount of fouling material remained deeply entrapped inside the mesh of an anaerobic bioreactor, even after intense water flushing and scraping. It is also well documented that the operating conditions of MBR or DMBR affects sludge filtration performance (e.g. Ersahin et al., 2017; Sabia et al., 2013). In addition, the high fouling propensity of landfill leachate, as observed in conventional membrane bioreactor (Ahmed and Lan, 2012), could have changed filtration characteristics of the aerobic/anoxic sludge developed in the experimental bench-scale plant.

A previous study on the evaluation of DM development (Saleem et al., 2017) indicated that mesh pore size does not affect DM development, similarly to the results of the first filtration cycle in this research study. It is of note that the results in Saleem et at., 2017 were obtained

in short term experiments with an anaerobic sludge. On the contrary, other authors (Wu et al., 2003) reported that large mesh pore size favours high filtration fluxes under similar conditions of applied TMP and this observation is in agreement with the second phase of the present research study. The diversity of results between the two phases of the dynamic MBR treating landfill leachate suggests that the operating condition and/or the feed and sludge characteristics influence the development of the cake layer composing the DM and thus its filtration characteristics. As a consequence of this, a proper selection the mesh pore size could facilitate the filtration performance of the DM in particular for those cases where excessive amount of organic foulants (humic and fulvic substances, largely present in stabilized LFL) would have contributed towards much faster DM fouling in the later stage of bioreactor operation (Ahmed and Lan, 2012).

3.2. Short-term filtration experiments

During the short-term filtration tests under gravity driven filtration, the inoculum showed flux reduction to less than 10% of the initial values in 30 minutes (Fig. 4). On the contrary, when the sludge collected from the aerobic tank after more than two months of leachate treatment was used, fluxes reduced to less than 5% of their initial values within 10 to 15 min demonstrating a much higher fouling propensity of the bulk sludge than the inoculum (Fig. 4). As a result, the fluxes measured filtering the inoculum were of approximately 10 time higher than those obtained by using the bulk sludge from the reactor if compared after approximately the same filtration time. The much higher fouling propensity of the bulk sludge if compared with the inoculum, can also be evinced in the filtration resistance (Fig. 4).

Since the fluxes reduced very quickly to values well below the 5% of the initial flux when using the bulk sludge, the fluxes were increased (using a peristaltic pump) to constant values of approximately 100 L m^{-2} h^{-1}. This operation was carried out in order to assess the

behaviour of the DM filtration under constant flux as typical condition for MBRs operation. The constant flux filtration caused a slight decrease of filtration resistance for 200 and 85 meshes which was however quickly followed by an increase (Fig. 4). The initial decrease of the filtration resistance for 200 and 85 μm meshes suggests the initial formation of a weak DM structure under gravity driven filtration, that was not resistant to the increased flux and the resulting TMP (Fig. 4). Alibardi et al. (2014) have reported a similar observation, during flux-step experiment performed to assess the strength of DM formed under anaerobic conditions treating synthetic wastewater. These results suggest that larger mesh pore sizes form unstable DM which can be easily destabilised with sudden increases of flux.

The different behaviour of the bulk sludge if compared with inoculum is also well evident in the turbidity measured in the short-term experiments. During the filtration tests of the inoculum, effluent turbidity reduced to values lower than 10 NTU in 5-10 min, with no particular differences among the three meshes (Fig. 4). When the bulk sludge was used, turbidity remained above 400 NTU although the much lower fluxes and higher resistance. Under constant flux condition, turbidity values increased markedly for 85 and 200 μm mesh (Fig. 4) and effluent quality deteriorated due to the loss of loosely bounded particles at high TMP (Fig. 4). In contrast, the continuous deposition of materials on the 10 μm mesh formed a DM which was more resistant to much higher TMP values, averaging around 50 kPa (Fig. 4), resulting in an improved effluent quality (Fig. 4).

These results suggest that the cake layer formed by the bulk sludge has a higher propensity to fouling, a more unstable structure and cannot effectively reject solids, if compared to performance obtained from inoculum sludge. The results also indicate that the operation of a DM treating landfill leachate causes a significant deterioration of biofilm on the mesh, at least for its filtration characteristics. The use of large mesh openings seems beneficial on the basis of higher filtration flux and lower operating TMP values for 200 and 85 μm meshes than

those measured for 10 μm mesh (Fig. 4); however, such advantages were associated with highly deteriorated effluent quality due to loss of biomass in the effluent. Therefore, these results demonstrate that the behaviour of the DM is affected by the characteristics of the filtered sludge and by the operating conditions applied (i.e. flux and TMP).

3.3. Landfill leachate treatment

Dissolved oxygen concentration of the aerobic vessel was always maintained above 1.0 mg L^{-1} (data not shown) during the entire bioreactor operation to sustain nitrification. The pH value of aerobic bioreactor varied between 6.4 and 8.9 (Figure S1 supplementary material).

The bioreactor exhibited moderate TOC removal performance due to the recalcitrant nature of the organics in the leachate. The average TOC removal recorded after 20 days of continuous bioreactor operation was 58±1.4% (Figure 6). Moderate organic removals from leachate collected in old landfill are usually observed applying conventional biological processes (e.g. Spagni et al., 2007). In addition, Ahmd and Lan, (2012) reported that conventional MBRs treating stabilised LFL achieve COD removal efficiencies ranging from 54-78%, similarly to the results of this study. Galleguillos et al. (2011) evaluated the performance of a pilot MBR with a microfiltration membrane in treating stabilised LFL. The system exhibited high BOD and ammonia removal of 94% and 98% respectively; however, COD removal was rather low (approx. 40%) due to the high concentration of recalcitrant organics, confirming the results obtained in this study using a DM bioreactor.

NH_4^+-N oxidation showed a fluctuating trend throughout the study, ranging from 70 to 99%, and despite the high influent NH_4^+-N concentration (1073-1767 mgN L^{-1}), the average NH_4^+-N oxidation was of 84±1.4% (Figure 6b). Although the system exhibited high NH_4^+-N oxidation, the biological nitrification process was incomplete and NO_2^--N was the main product of ammonia oxidation (Figure 6b). Along with the increase in influent NH_4^+-N

concentration towards the end of the experimental phase, a progressive increase in effluent NO_2^--N concentration was observed, reaching NO_2^--N values as high as 1062 mg L^{-1} (Figure 7a). As a consequence of incomplete nitrification, the effluent NO_3^--N concentration always remained below 160 mgN L^{-1} (Fig. 6) with average concentration of 86±6 mgN L^{-1}, showing a limited activity of nitrite oxidising bacteria (NOB).

The severe inhibition of NOB activity can be explained by considering the free ammonia (FA) and free nitrous acid (FNA) concentrations inside the bioreactor (Fig. 7). Their concentrations were above the minimum threshold proved to be toxic for NOB. Anthonisen et al. (1976) reported FA and FNA inhibitory concentrations for NOB bacteria ranging from 0.1 to 150 mgN L^{-1} and 0.2 to 2.8 mgN L^{-1}, respectively. Similar inhibitory concentrations have also been confirmed by other authors (Kim et al., 2006; Zhou et al., 2011). Figure 7 shows that inhibition of NOB was triggered by FA concentrations since this compound was mostly above the highest toxicity limit (according to Anthonisen et al., 1976) while FNA was only occasionally higher than the toxicity concentration.

It should also be highlighted that the addition of external carbon to support denitrification could have affected the nitrification activity: Remmas et al. (2016) have recently observed a significant nitrification inhibition when glycerol was added to MBR to sustain denitrification. Although acetate was used in this study instead of glycerol to support nitrogen removal, the addition of external organic material could have affected the microbial community including the specific abundance of nitrifying populations (Remmas et al., 2016).

Even though denitrification via the nitrite route could have offered considerable cost savings in terms of organics and aeration requirements (Spagni and Marsili-Libelli, 2010), the denitrification performance and consequently the total nitrogen removal were rather poor (Fig. 6). Average total nitrogen removal after 20 days of continuous bioreactor operation and before the addition of supplemental organics was only 25±3%. Moreover, the gradual addition

of external organics did not bring significant improvement in the denitrification performance (Fig. 6). Furthermore, it can also be inferred that the contribution of heterotrophic denitrification in total TOC removal was very limited and a large fraction of TOC was removed in the aerobic tank instead of anoxic one (data not shown).

Zhou et al, (2011) summarised the results of several studies done on determining the toxicity threshold of FNA concentration on denitrification activity. Depending upon the microbial community structure and operating conditions (pH, temperature, etc.), FNA concentration as low as 0.01-0.025 mgHNO_2-N L^{-1} can initiate inhibition (up to 40%) while concentration up to 0.2 mgHNO_2-N L^{-1} was proved to be extremely toxic on denitrification activity. In this study, the observed FNA concentration in the anoxic tank ranged from 0.001 to 0.079 mgHNO_2-N L^{-1} and averaging around 0.011 mgHNO_2-N L^{-1} that might have contributed to the poor denitrification activity of the system (Fig. 6).

4. Conclusions

This study showed the possibility of using DM developed over nylon meshes (10, 52, 85 and 200 µm) in a two-stage anoxic/aerobic bioreactor for the treatment of stabilised LFL.

The results demonstrated the change of the filterability characteristics of the bulk sludge due to the applied operating conditions and to the use of stabilised LFL. As a consequence, severe DM fouling was observed, which was characterised by very sharp increase in TMP. DM solids rejection was also deteriorated and the effect of mesh porosity on solid-liquid separation was heightened. Effective solids rejection was achieved with the mesh with the smallest openings tested in this study of 10 µm, though at low permeate fluxes (approximately of 5 L m^{-2} h^{-1}). In this regard, among the four meshes tested in this study, 52 µm mesh showed to be a reasonable compromise in terms effluent turbidity and achievable operating fluxes.

The bioreactor achieved organics removal similar to values reported in literature for conventional MBR systems. Even though bioreactor exhibited high ammonia oxidation, the increased concentrations of free ammonia (FA) and free nitrous acid (FNA) inside the system severely affected the nitrification and denitrification performance that resulted in high nitrite accumulation.

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Table captions

Table 1. Properties of the meshes used in this study.

Table 2. Average characteristics of the leachate samples.

Table 1. Properties of the meshes used in this study.

Product information	Mesh opening (µm)	Open area (%)	Mesh count (cm^{-1})	Thread diameter (µm)	Resistance (Clean mesh) (m^{-1}) [2]	Tap water permeability ($L\ m^{-2}\ h^{-1}\ kPa^{-1}$) [3]
SaatiMil PA[1] 7	200	39	31	120	5.5×10^9	1570
SaatiMil PA 15	85	49	81	37	5.4×10^9	1580
Saatifil PA 52/32	52	32	110	38	5.6×10^9	1530
Saatifil PA 10/4	10	4	200 x 220	30 x 38	6.5×10^9	1330

(1) PA is an acronym for polyamide

(2) Resistance of the mesh measured at TMP of 5 kPa

(3) 20 ^{o}C normalised permeability measured at TMP of 5 kPa

Table 2. Average characteristics of the leachate samples.

Parameter	**Value**
BOD_5 (mgO_2 L^{-1})	400
TOC (mgC L^{-1})	1300
TKN (mgN L^{-1})	2060
NH_4^+-N (mgN L^{-1})	1700
NO_3^--N (mgN L^{-1})	< 1
NO_2^--N (mgN L^{-1})	7.7
Total phosphorus (mgP L^{-1})	9.6
pH	8.56
Alkalinity (mg $CaCO_3$ L^{-1})	14600
Cd (µg L^{-1})	< 10
Cr (µg L^{-1})	753
Cu (µg L^{-1})	52
Fe (µg L^{-1})	3860
Mn (µg L^{-1})	172
Ni (µg L^{-1})	148
Pb (µg L^{-1})	< 10
Zn (µg L^{-1})	112

Figure captions

Figure 1. Schematic diagrams (a) experimental setup and (b) short-term filtration test set-up

Figure 2. Observed filtration flux and TMP profiles for (a) 200 µm, (b) 85 µm, (c) 52 µm and (d) 10 µ.

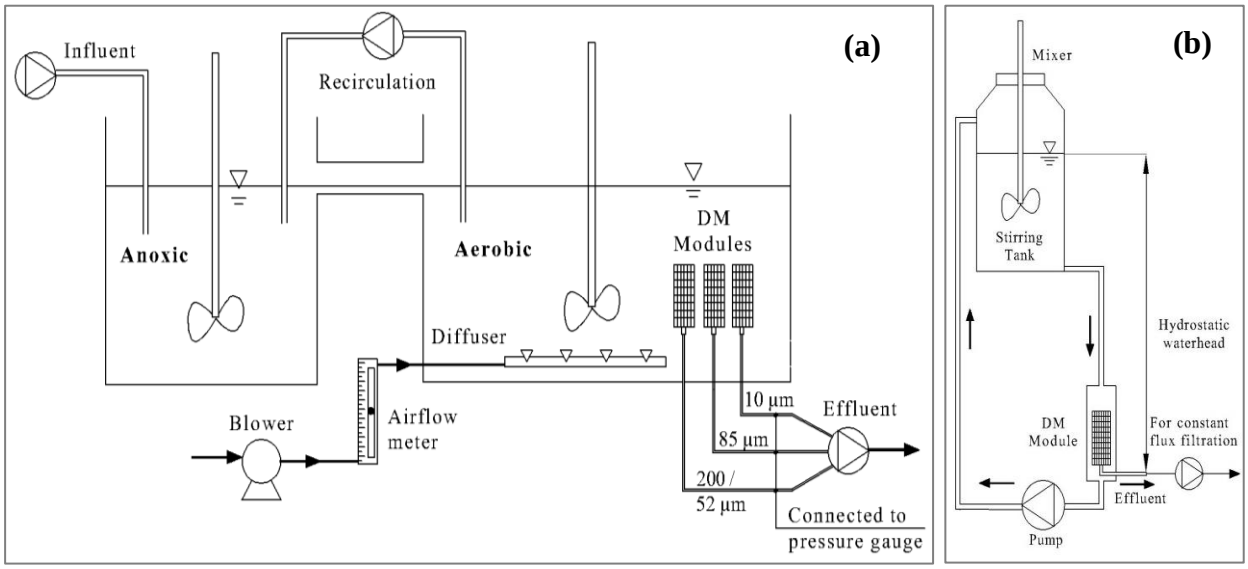
Figure 3. Dynamic membrane resistance profiles along with effluent turbidity values (a) 200 µm, (b) 85 µm, (c) 52 µm and (d) 10 µm

Figure 4. Results of short-term gravity driven filtration tests: flux (a,d) resistance (b, e) and turbidity (c, f) profiles for initial inoculum and for bulk sludge, respectively. Arrows indicate when the peristaltic pump was switched on (bulk sludge only) to increase membrane flux; square, triangle and circle below the arrows are for mesh of 200, 85 and 10 µm, respectively. TMP profile for bulk sludge experiment is inserted in graph (d).

Figure 5. Influent and effluent TOC profiles and TOC removal performance.

Figure 6. (a) influent (INF) and effluent (EFF) ammonia and effluent nitrite and nitrate concentration; (b) ammonia oxidation and nitrogen removal performance

Figure 7. Free ammonia (a) and free nitrous acid (b) concentration and values of toxicity for nitrifying microorganism according to Anthonisen et al. (1976).



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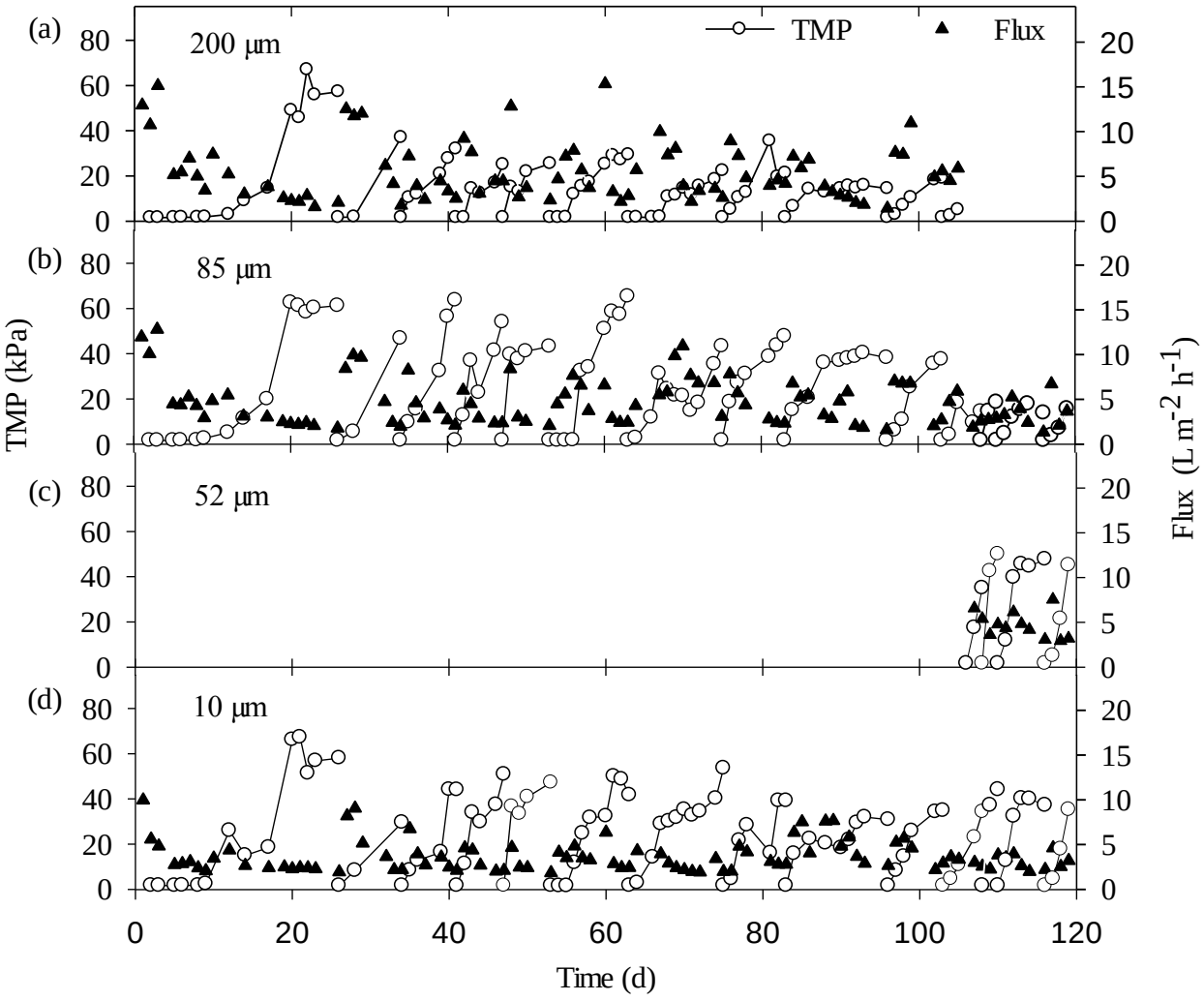


Figure 2. Observed filtration flux and TMP profiles for (a) 200 μm, (b) 85 μm, (c) 52 μm and (d) 10 μ.

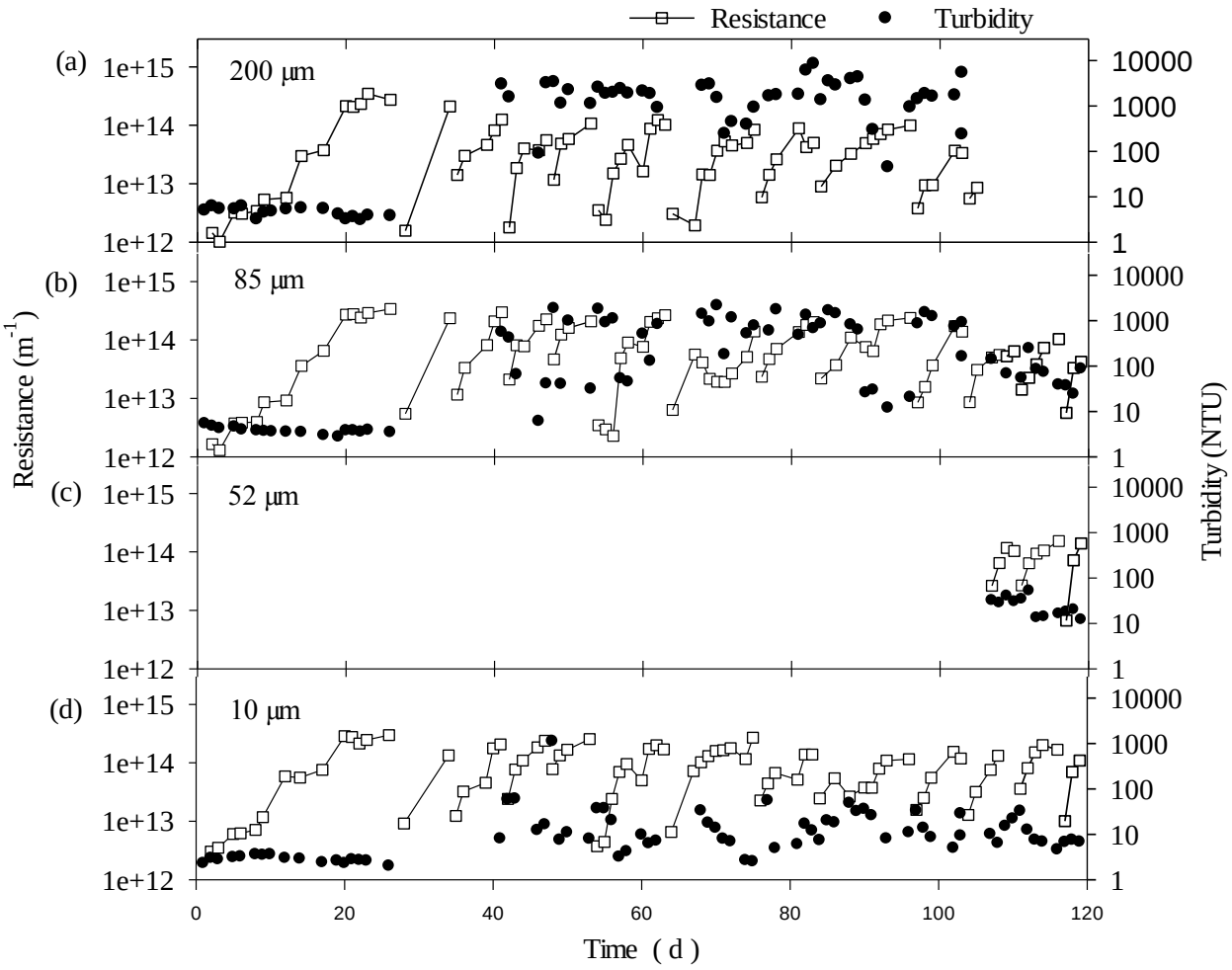
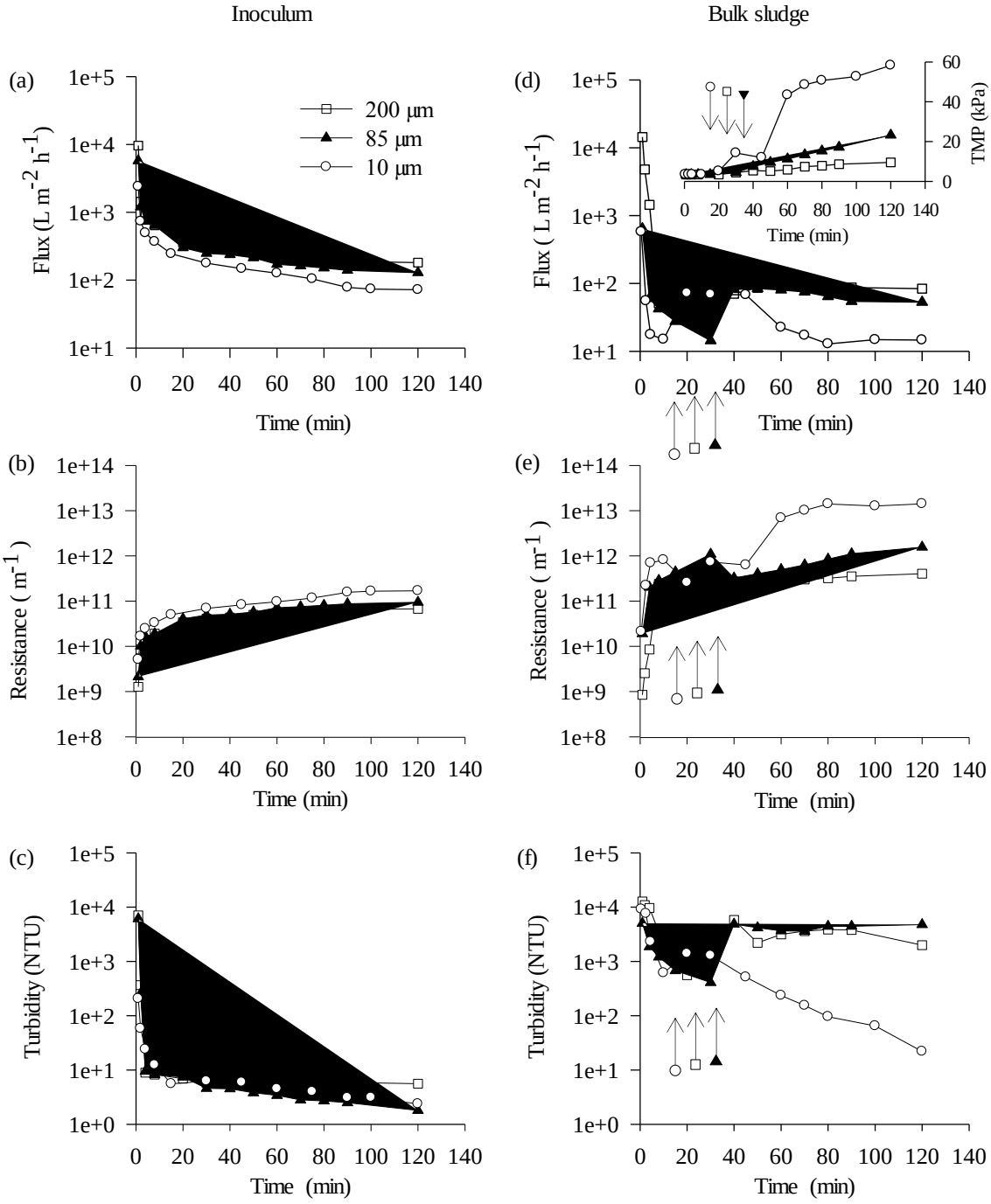


Figure 3. Dynamic membrane resistance profiles along with effluent turbidity values (a) 200 μm, (b) 85 μm, (c) 52 μm and (d) 10 μm



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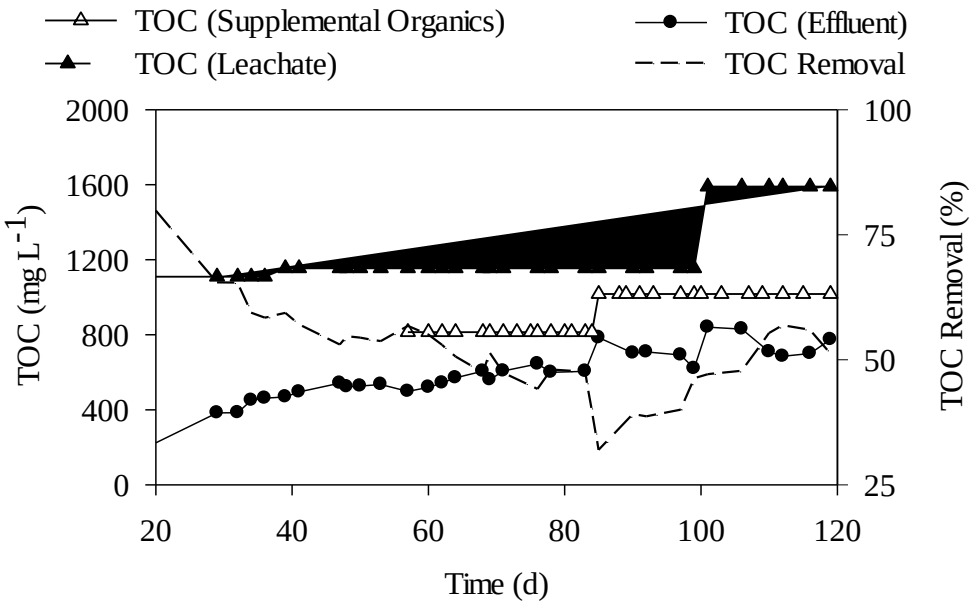
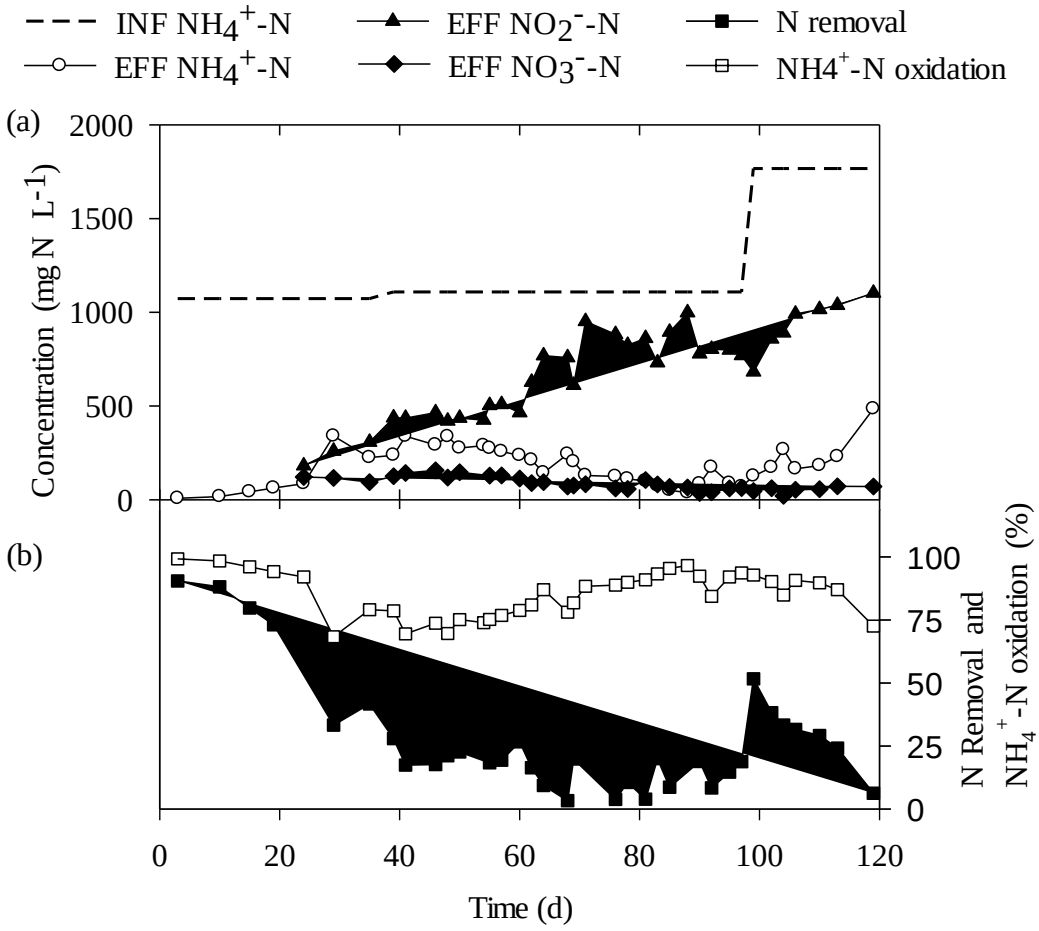


Figure 5. Influent and effluent TOC profiles and TOC removal performance.



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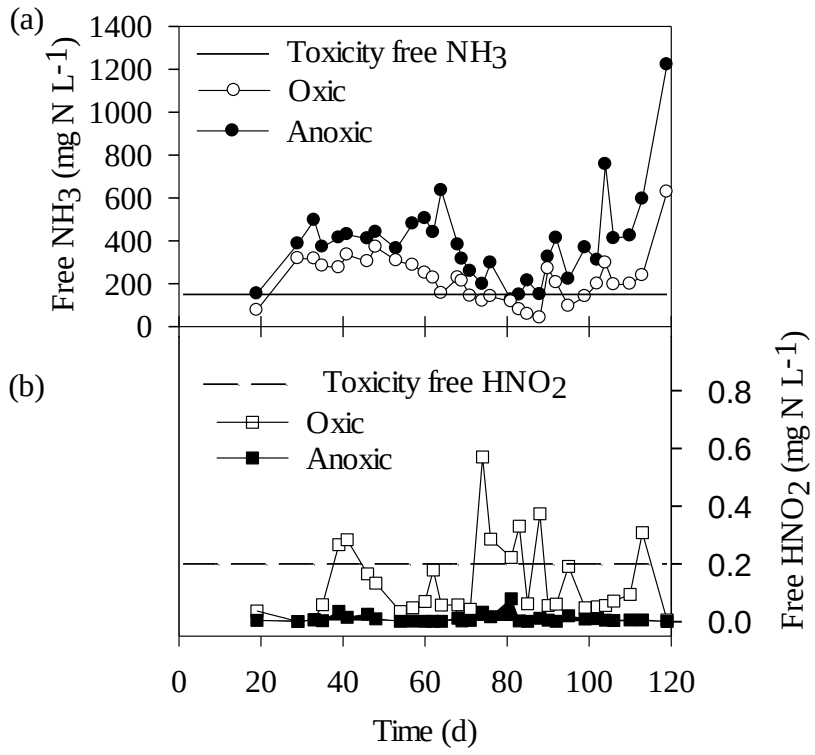

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Supplementary material

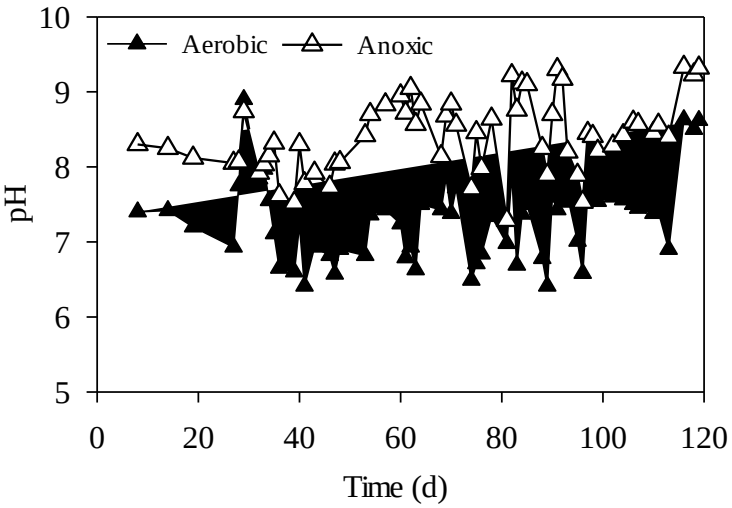

Figure S1. Observed pH profile inside the aerobic and anoxic tank.