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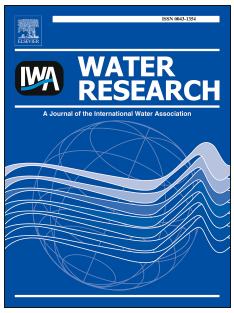

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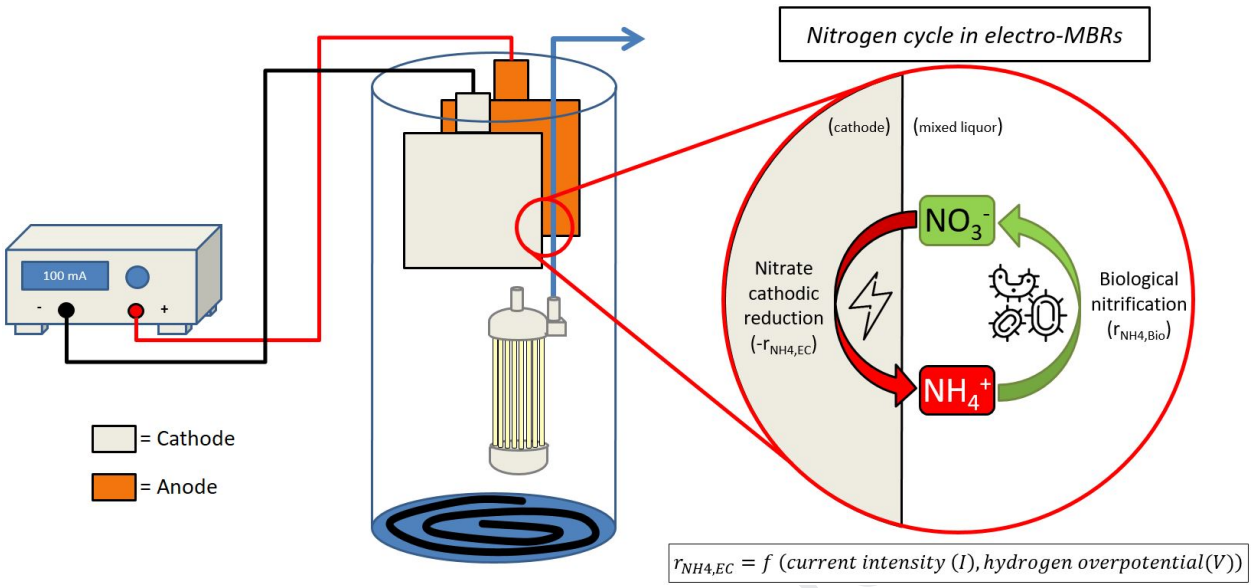

Effect of cathode material and charge loading on the nitrification performance and bacterial community in leachate treating Electro-MBRs

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Abstract:

Electro-MBR technology, which combines an electrocoagulation process inside the mixed liquor of a membrane bioreactor, was studied for the treatment of a high-strength ammonia leachate (124 ± 4 mg NH_4-N L^{-1}). A lab-scale aerobic Electro-MBR was operated with a solid retention time of 45 days, hydraulic retention times of 24h and 12h, and charge loading ranging from 100 to 400 mAh L^{-1}. At 400 mAh L^{-1}, with a combination of a Ti/Pt cathode and a sacrificial iron anode, removal percentages for ammonia nitrogen, total organic carbon, and total phosphorus were 99.8%, 38%, and 99.0%, respectively. At 400 mAh L^{-1}, the estimated ferric ion dosage was 325 mg Fe^{3+} L^{-1}. Experiments conducted with different cathode materials showed that previously reported inhibition phenomena may

result from a cathodic nitrate reduction into ammonia nitrogen. Conventional cathode materials, such as graphite, have electrochemical nitrate reduction rates of -0.03 mg NO_3-N mAh^{-1}. By comparison, when using Ti/Pt, the rate was -0.0045 mg NO_3-N mAh^{-1}(85% lower than graphite due to its low hydrogen overpotential). Charge loading tested in this study had no significant impact on both nitrification performance and microbial population diversity. However, the relative abundance of the mixed liquor's Nitrosomonas increased from 4.8% to 8.2% when the charge loading increased from 0 to 400 mAh L^{-1}. Results from this study are promising for future applications of the Ti/Pt - Iron Electro-MBR in various high-strength ammonia wastewater treatment applications.

Keywords: electro membrane bioreactor, electro-coagulation, nitrification, ammonia nitrogen, leachate, microbial community

List of abbreviation

AOB	Ammonia oxidizing bacteria
BOD_5	Biological oxygen demand over a period of 5 days
COD	Chemical oxygen demand
DC	Direct current
DO	Dissolved oxygen
EC	Electro-coagulation
HRT	Hydraulic retention time
HOP	Hydrogen overpotential
MBR	Membrane bioreactor
MLVSS	Mixed liquor volatile suspended solids
NLR	Nitrogen loading rate
OLR	Organic loading rate
OTU	Operating taxonomic units
PCoA	Principal component analysis
PLR	Phosphorus loading rate
SRT	Solid retention time
SS	Stainless steel
TSS	Total suspended solids

1. Introduction

Waste originating leachates are known for their composition complexity and for the threat they pose to the preservation of water resources adjacent to waste management facilities. Amongst the contaminants found in these leachates, ammonia nitrogen is of primary concern due to its numerous adverse effects, such as its promotion of eutrophication, its toxicity to aquatic organisms, and the depletion of dissolved oxygen caused by the oxidation of ammonia nitrogen to nitrate (He, Xue et al. 2009). Previous studies reported ammonia nitrogen concentrations in composting and landfill leachates ranging from 5 to more than 21 000 mg NH_4-N L^{-1} (Roy, Azaïs et al. 2018).

The treatment of these high-strength ammonia leachates has become a major concern in recent years due to their negative effect on conventional municipal wastewater biological treatment processes. Gagnaire et al. (2011) reported that even when co-treating leachate with municipal wastewater, the dilution factor is often insufficient to dilute the ammonia nitrogen concentration peaks that inhibit the microbial activity in the mixed liquor (Gagnaire, Wang et al. 2011). Consequently, treatment systems have been engineered specifically for waste originating leachates. Amongst these technologies, the membrane bioreactor (MBR) is one of the most efficient in terms of removing ammonia nitrogen from leachates. In a previous study conducted on co-composting leachates, MBRs were shown to reach ammonia nitrogen removal rates of >740 mg NH_4-N L^{-1} d^{-1}, which corresponds to 99.8% removal (Roy, Drogui et al. 2020). The main advantage of MBRs is their excellent control over the SRT, due to the membranes that prevent the passage of microorganisms in the permeate. Such a control over the SRT allows for the proliferation of low growth rate ammonia-oxidizing bacteria (AOB) (Canziani, Emondi et al. 2006) and their acclimation to the inhibitive compounds potentially found in leachates (Vuono, Regnery et al. 2016). However, membranes are also at the origin of the main drawback of MBRs: membrane fouling. Membrane fouling is mainly caused by the small size of microbial floc (Bani-Melhem and

Elektorowicz 2011), high concentrations of extracellular polymeric substances in the mixed liquor (Wei, Elektorowicz et al. 2012), and soluble organic substances (Borea, Naddeo et al. 2017). Furthermore, aerobic MBRs have low phosphorus removal efficacy, which can be prohibitive for leachate treatment applications. For example, Roy et al. (2020) obtained assimilation/precipitation rates ranging from 0.026 to 0.041 g P/g total suspended solids (TSS) when treating co-composting leachates, which corresponds to 37% to 71% removal (Roy, Drogui et al. 2020).

Lee et al. (2001) made a first attempt at solving both of the aforementioned drawbacks by feeding an inorganic coagulant directly in the mixed liquor. Using alum at a molar ratio of 1.5Al:1 P, they found that alum can efficiently coagulate colloidal particles in the range of 0.1 to 2 µm, as well as increase microbial particles' zeta potential from -14.6 to -6.8 mV, both of which lead to a significant improvement in membrane permeability. Adding alum also increased phosphorus removal by 90% through chemical precipitation. However, the formation of $AlOH_2$ (precipitate) and its conjugated acid, H_2SO_4, led to a reduction in pH, which inhibited biological nitrification (Lee, Kim et al. 2001). Similarly, Wu et al. (2006) tested the addition of monomeric coagulants ($Al_2(SO_4)$, FeCl3) and polymeric coagulants (polymeric aluminum chloride, and polymeric ferric sulfate) to control membrane fouling. Polymeric ferric sulfate was found to be the most effective in providing the highest removal percentage of supernatant organic matter as well as the greatest enlargement of sludge floc size. However, the required high coagulant dosage in mixed liquor (> 0.5 mM Al or Fe) quickly led to a pH drop below 7 and inhibited biological nitrification (Wu, Chen et al. 2006).

To overcome the acidification problems related to the addition of inorganic coagulants, researchers proposed the combined use of electrocoagulation (EC) and MBRs. Patented in 1976 by Ramirez (Ramirez 1976), EC consists of the in-situ generation of multivalent metal ions (coagulants) from the electrochemical dissolution of immersed, sacrificial anodes (Ensano, Borea et al. 2016). The main advantages of EC when compared to inorganic coagulant is: 1) the reduction in materials and

transportation costs compared to chemical reagents, 2) the reduced sludge production due to a lower content in bound water, and 3) the simplified dosage automation (Wei, Oleszkiewicz et al. 2009). There are two main categories of electrocoagulation-MBRs (Electro-MBR): 1) external EC unit upstream of the MBR and 2) EC directly submerged in the mixed liquor with the membrane (Ensano, Borea et al. 2016). The first configuration was initially proposed by Kim et al. (2010) for the treatment of municipal wastewater in South Korea. With a 50 m^3/h electro-MBR pilot unit using aluminum cathodes and anodes, they obtained total phosphorus removal efficiency of 79.9%, as opposed to 41% for a MBR without EC (Kim, Jang et al. 2010). In the same period of time, Wei et al. (2009) studied the second configuration using synthetic municipal wastewater. Also using aluminum electrodes, they found that the Electro-MBR can significantly reduce membrane fouling and improve phosphorus removal (Wei, Shi et al. 2009). Using the same Electro-MBR as Wei et al. (2009), Bani-Melhem et Elektorowicz (2011) tested the effect on treatment performance of an intermittent DC with an operational mode of 15 min ON – 45 min OFF using iron electrodes. They found that the addition of an EC system enhanced the removal of COD and PO_4-P up to 96% and 98%, respectively, but reduced ammonia nitrogen removal from 97% to 70%. These results raised concerns about the application of this technology with high strength ammonia wastewaters such as leachates. According to Bani-Melhem et Elektorowicz (2011), lower nitrification activity may be caused by two factors: 1) the greater sensitivity to the applied DC field of the nitrifying bacteria; and/or 2) the accumulation of iron in the Electro-MBR, which has some inhibitory effects on the activity of nitrifying bacteria (Bani-Melhem and Elektorowicz 2011). Similarly, Li et al. (2001) previously shown that an electricity quantity of 50 mAh L^{-1} in an activated sludge system with stainless steel (SS) electrodes reduced the nitrification rate by 50% (Li, Cao et al. 2001). Other studies reported similar inhibitive effects on mixed liquor microbial populations and nitrification performance. Wei et al. (2011) exposed mixed liquor samples to various electricity quantities for 4 hours in a batch system and found that the percentage of live cells dropped by 15% and 29% when exposed to

440 mAh L^{-1} and 890 mAh L^{-1}, respectively (Wei, Elektorowicz et al. 2011). Bani-Melhem et Smith (2012) reported a drop in ammonia nitrogen removal from 97.4% to 77.8% when adding an EC system operated with a charge loading of 680 mAh L^{-1} and aluminum electrodes to their MBR (Bani-Melhem and Smith 2012).

On the other hand, other studies reported contradictory effects of the EC on nitrification performance. Tafti et al. (2015) reported that nitrogen removal efficiency improved with an electricity quantity below 144 mAh L^{-1} in an Electro-MBR (iron electrodes) when compared to an unmodified MBR (Tafti, Mirzaii et al. 2015). Similarly, Borea et al. (2016) reported that the EC increased ammonia nitrogen removal by 10.5% to 27% when charge loading of 15 and 40 mAh L^{-1} were applied with an aluminum anode and a SS cathode (Borea, Naddeo et al. 2017). Li et al. (2018) and Battistelli, da Costa et al. (2018) each compared the microbial population of an Electro-MBR to that of a MBR treating the same wastewater. Li et al. (2018) found that the relative abundance of Nitrosomonas in the MBR's mixed liquor increased from 1.45 to 3.12% when a 14 mAh L^{-1} charge load was applied (Li, Dong et al. 2018). Similarly, Battistelli, da Costa et al. (2018) reported an increase in Nitrospira relative abundance from 10% to 38% as well as higher ammonia oxidizing bacteria activity (OUR-NH_4^+) when an electric density of 10 A m^{-2} was applied (Battistelli, Belli et al. 2019).

This inconsistency in the reported effects of EC systems on MBRs nitrification performances might find its origin in the selection of the cathode material and the charge loading. Dia et al. (2017) previously studied landfill leachate treatment using biofiltration followed by EC (Dia, Drogui et al. 2017). Interestingly, they reported that nitrates reacted at the cathode and formed ammonia nitrogen according to the electrochemical reduction reactions presented in Figure 1.

In addition, the nitrate electrochemical reduction rate was found to be correlated with the hydrogen overpotential (HOP). In an EC process, gaseous hydrogen generation is the main chemical reaction

occurring on the cathode's surface. However, when the cathode material HOP value is high, nitrate reduction can compete with hydrogen generation, leading to an increase in ammonia nitrogen concentration. In order to avoid ammonia nitrogen formation, Dia et al. (2017) suggested the use of Ti/Pt as the cathode material, which has a HOP of 0.95 V, compared to 1.3, 1.75, and 1.37 V for Fe, Al, and SS, respectively. When applying 500 mAh L^{-1} on a leachate containing 300 mg NO_3-N L^{-1}, they found that the Ti/Pt cathode reduced ammonia nitrogen formation by more than 90% compared to Fe, Al, and SS electrodes. The application of these concepts is promising in terms of Electro-MBRs' applications in the treatment of high-strength ammonia nitrogen wastewaters.

Another challenge with leachate is the presence of high concentrations of recalcitrant organic carbon (Roy, Benkaraache et al. 2019). MBRs are efficient technologies for the removal of easily to moderately biodegradable organic carbon. However, they are limited in terms of removing leachates' recalcitrant organic carbon at hydraulic retention times below 48h when operated at SRTs of 30 to 45 days (Roy, Drogui et al. 2020). The addition of an EC process to the MBR process has been reported to increase the organic carbon removal rates in municipal wastewaters and synthetic wastewater. However, no studies have reported its effect on recalcitrant TOC from leachate, which is mostly comprised of fulvic- and humic-like substances (high aromaticity, and presence of polar functions such as carboxyl and hydroxyl groups) (Roy, Benkaraache et al. 2019).

The main objective of this study is to assess the potential application of Ti/Pt Electro-MBRs in the treatment of high-strength ammonia nitrogen leachates, as well as to define the effect of the EC process on nitrification and leachate contaminant removal performances (COT and total phosphorus). Since membrane fouling is largely covered in the scientific literature, the subject will not be discussed in this study. In order to reach these objectives, the nitrate electrochemical reduction hypothesis was validated by testing a graphite and a Ti/Pt cathode in a batch EC system. Then, high-strength ammonia co-composing leachate was treated using an Electro-MBR operated at hydraulic retention times (HRT) of

172 24h and 12h, and charge loading of 0, 100, 200, and 400 mAh L^{-1}. During this study, the following
173 contaminants were studied: ammonia nitrogen, total organic carbon (TOC), and total phosphorus (P_{tot}).
174 Finally, metagenomic analyses, with a focus on amoA gene expressing bacteria, were conducted on
175 mixed liquor samples taken at each charge loading tested in order to assess the effect of the EC on the
176 ammonia oxidizing bacteria population activity.

177 2. Material and methods

178 2.1. Electrocoagulation batch reactor

179 The experiments on the effect of the cathode material on the cathodic nitrate reduction rate were
180 carried out in a 0.5 L parallelepiped batch reactor made of clear PVC, as presented in Figure 2a. A
181 magnetic stirrer was used in order to create a homogeneous internal mixing. The rectangular electrodes
182 (10 cm x 11 cm) were immersed in the reactor at 1 cm distance from one another (Figure 2b). The
183 sacrificial anode was made of iron. Before each experiment, the anode was first polished using an
184 abrasive paper, and then rinsed in a 5% (v:v) diluted HCl solution for 10 minutes. Two cathodes made of
185 different materials were tested: 1) graphite, and 2) platinized titanium (Ti/Pt). Graphite was selected for
186 its low cost and because it had not previously been tested by Dia et al. (2017) on leachates in EC systems
187 (Dia, Drogui et al. 2017). Ti/Pt was selected for its low HOP. The electrodes were connected to a digital
188 DC power supply (BK Precision 9184 DC power supply, 0-100V, 2A). Co-composting leachate containing
189 128±1 mg NO_3-N L^{-1}, treated with the MBR before the addition of electrodes (Table 1, Condition #1), was
190 used during this experiment.

191 2.2. Lab-scale submerged electrocoagulation-MBR

192 The submerged Electro-MBR lab-scale experimental set-up was designed according to figure 3a and
193 Figure 3b.

194

195 The leachate was kept in a 50 L polyethylene tank placed in a refrigerator at 4°C in order to avoid any
196 biological activity that could alter the leachate's composition. The 10 L reactor was made of a 146.33
197 mm I.D. clear PVC tube (schedule 80). The leachate was fed into the reactor through a side port using a
198 peristaltic pump (Masterflex, model #7528-10). An ultrafiltration hollow-fiber membrane module (Zee-
199 Weed, ZW-1) was used. The total surface area of the module's membrane is 47 cm^2, while the
200 membrane's nominal pore diameter is 0.04 μm.

201 The permeate flow rate (filtration: 300 s, flux 7.4 L m^{-2} h^{-1}; backwash: 20 s, flux 46.4 L m^{-2} h^{-1}) was
202 maintained constant during each experiment by controlling the vacuum applied to the membrane (5 to
203 50 kPa) with a peristaltic pump (Masterflex, model #7528-10). To prevent the accumulation of a cake
204 layer on the membrane's surface, filtration/backwash cycles were applied, and air was introduced
205 between the membrane fibers (2.5 L air min^{-1}) in order to create enough shear stress to dislodge most of
206 the biofilm. Membrane module cleaning was conducted when the transmembrane pressure reached -50
207 kPa.

208 The electrodes (sacrificial iron anode and Ti/Pt cathode) were immersed around the membrane module
209 at 5 cm distance from one another, similar to previous studies (Bani-Melhem and Elektorowicz 2011,
210 Borea, Naddeo et al. 2017). This distance, combined with the high conductivity of the leachate, reduced
211 the power requirements needed to obtain the desired charge loading, while minimizing the potential
212 inhibitive effect of an acidic/oxidation zone on the biological nitrification (Bani-Melhem and
213 Elektorowicz 2010). The electrodes were connected to a digital DC power supply (BK Precision 9184 DC

power supply, 0-100V, 2A). When the voltage reached 3V, the iron anode was changed and the Ti/Pt cathode was cleaned in a 0.1M HCl solution for 1h.

The Electro-MBR was operated in oxic conditions (D.O. = 6 - 7 mg O_2 L^{-1}) at room temperature (20±1°C). Compressed air was introduced through a perforated tube placed at the bottom in the reactor (2.5 L min^{-1}) and arranged in such way as to create a homogeneous internal mixing of the reactor from the rising bubbles. SRT was controlled by purging mixed liquor from the reactor on a daily basis. HRT was controlled by adjusting the mixed liquor volume; it was calculated by measuring the collected permeate and mixed liquor volume on a daily basis.

2.3. Experimental procedure

Over the course of the experiment's 70-day duration, three different charge loading (100, 200, and 400 mAh L^{-1}) were tested at a constant SRT of 45 days. The electric current was applied with a continuous operation mode. The details of the experimental conditions are presented in Table 1.

The charge loading ($\dot{q}$) is defined by the amount of electricity transferred through electrochemical reaction for a given amount of water treated, and it is calculated using the following equation (Eq. 1):

$$\dot{q} = \frac{I}{V} * HRT \qquad \text{Eq. 1}$$

Where I is the applied current (mA), V is the reactor's volume (L), and the HRT is expressed in hours. The mixed liquor used in this study originated from a previous experiment in which the MBR (without electrodes) was operated at a HRT of 45 days during 60 days with composting leachate collected in the same composting facility as were collected this study's leachate (St-Henri-de-Lévis's composting facility, Canada) (Roy, Drogui et al. 2020). In order to adapt the mixed liquor's microbial population to this study's leachate contaminant concentrations, the MBR was initially operated without applied current for 28 days (Condition #1). Then, the three different applied charge loading were maintained for 14 days

235 each (Conditions #2, #3 and #4) in order to reach steady-state conditions and to provide an adequate
236 duration for sample collection. After 10 days of operation under the same operating conditions, samples
237 were taken every day for 4 days. If the difference in TOC and $NH4^+$ removal rates, as well as MLVSS
238 between two samples taken within a 4-day interval was less than 5%, then the MBR was considered to
239 be operating at steady-state. The characterization of the co-composting leachate treated in this study is
240 presented in Table 2.

241 *2.4. Analytical methods*

242 *2.4.1. Microbial community analysis*

243 **Genomic DNA extraction**

244 Genomic DNA was extracted from 10 ml of each sample using a DNeasy Powersoil kit following the
245 manufacturer's protocol (Quiagen). The final elution volume was 100 µl, and the concentration was
246 determined with the Quant-it Picogreen kit (Thermofisher) and the NanoDrop 330 Fluorospectrometer
247 (thermofisher).

248 **Illumina MiSeq**

249 The first PCR amplifications were performed with the primer Bakt_341F (5'-CCTACGGGNGGCWGCAG-3')
250 and the primer Bakt_805R (5'-GACTACHVGGGTATCTAATCC-3'), designed by Herlemann et al. (2011)
251 (Herlemann, Labrenz et al. 2011). Common sequence 1 (CS1) (5'-ACACTGACGACATGGTTCTACA-3') and
252 common sequence 2 (CS2) (5'-TACGGTAGCAGAGACTTGGTCT-3') universal primer sequences, required
253 for Illumina MiSeq amplicon tagging and indexing, were added to the 5' ends of forward and reverse
254 primers, respectively. Reaction conditions were 98°C for three minutes, followed by 30 cycles of 98°C for
255 10 seconds, 62°C for 20 seconds and 72°C for 30 seconds. Then a final elongation step of 72°C for 2
256 minutes was conducted. The first PCR products were used as templates for the second PCR. The second

PCR conducted in order to add adapters and tags, as well as the amplified sequences purification were made by Génome Québec inc.

Statistical analysis

Bacterial population analysis results were interpreted using MicrobiomeAnalyst software (Dhariwal, Chong et al. 2017). The Marker Data Profiling module was used to compare the bacterial population in the samples taken at different charge loading.

2.4.2. Physico-chemical parameters analysis

Water samples (feed and permeate) were analyzed for pH (Mettler Toledo SevenEasy), electrical conductivity (Mettler Toledo SevenCompact Conductivity), alkalinity (bromocresol green titration), solid content (total, dissolved, and volatile) (EPA Method 160.2), chemical oxygen demand (CEAEQ MA. 315 – DCO 1.1, Potassium dichromate), biological oxygen demand (5 days) (CEAEQ MA. 315 – DBO 1.1,), dissolved ammoniacal nitrogen (QuickChem Method 10-107-06-2-O, salicylate – nitroprusside colorimetric method), total organic carbon and total nitrogen (Shimadzu VCPH), and total phosphorus (Varian Vista AX ICP-AES). Total phosphorus content was determined after preliminary sample digestion (15% trace metals grade HNO_3 and 5% H_2O_2 at 95°C for 2 hours). Mixed liquor samples were analyzed for solid content (total, dissolved, and volatile) (EPA Method 160.2).

3. Results and discussion

3.1. Nitrate reduction hypothesis validation

In order to confirm the hypothesis of an electrochemical cathodic reduction of nitrates into ammonia nitrogen suggested by Dia et al. (2017), treated leachates from Condition #1 were exposed to charge

loading ranging from 200 to 1 000 mAh L^{-1} in an electrocoagulation batch reactor (Dia, Drogui et al. 2017). The leachate's nitrate and ammonia nitrogen concentrations after a 30-minute exposure to different current intensities in Figure 4a and Figure 4b, respectively. Figure 4a and Figure 4b show the concentrations using both a Ti/Pt cathode and iron anode, and graphite cathode and iron anode.

Initial concentrations of nitrate and ammonia nitrogen in the aerobically treated leachates were 128 mg NO_3-N L^{-1} and 1.1 mg NH_4-N L^{-1}, respectively. In Figure 4a, results show that nitrate concentrations linearly decreased at a rate of -0.03 mg NO_3-N mA^{-1} h^{-1} when a graphite cathode was used. At this rate, the leachate's nitrate concentration was reduced by 32 mg NO_3-N L^{-1} after 30 minutes of exposure to 1 000 mAh L^{-1}. With the Ti/Pt cathode, no significant difference in nitrate concentration was observed at charge loading below 750 mAh L^{-1}. Leachate exposed to 1 000 mAh L^{-1} for 30 minutes using a Ti/PT cathode saw its concentration reduced by approximately 5 mg NO_3-N L^{-1}, which is 85% less than with the graphite electrode at the same charge loading. In Figure 4b, results show that the leachate's ammonia nitrogen concentration increased proportionally to the applied charge loading when using the graphite cathode. More precisely, the ammonia nitrogen increased from 1.1 to 25 mg NH_4-N L^{-1} after 30 minutes with a charge loading of 1 000 mAh L^{-1}. This ammonia nitrogen concentration is in accordance with its associated nitrate concentration drop (32 mg NO_3-N L^{-1}). With the use of a Ti/Pt cathode, the ammonia nitrogen only increased from 1.1 to 1.3 mg NH_4-N L^{-1} after 30 minutes of exposure to a charge loading of 1 000 mAh L^{-1}.

According to the results presented in Figure 4a, the rate at which the nitrates are reduced into ammonia during the electrocoagulation process depends on the cathode material properties and also is directly correlated to the charge loading at high nitrate concentrations. This rate is expressed by Eq. 2.

$$r_{NO3,EC} = -k_i \frac{I}{V} = -r_{NH4,EC} \qquad \text{Eq. 2}$$

Where k_i is the cathode material's (*i*) nitrate mass reduction rate (mg NO_3-N mA^{-1} h^{-1}), *I* is the current intensity (A), and *V* is the reactor's volume (L). These results confirm the Dia et al. (2017) hypothesis that a Ti/Pt cathode is a well-suited cathode material for an EC process combined with a bioprocess treating ammonia nitrogen, due to its low electrochemical nitrate reduction rate (Dia, Drogui et al. 2017). In an Electro-MBR, the steady-state ammonia nitrogen removal is expressed by Eq. 3 (adapted from Roy et al. (2020) to include the effect of the EC process (Roy, Drogui et al. 2020)).

$$\frac{(S_{NH4,f} - S_{NH4,p})}{HRT} - r_{NH4,EC} = r_{NH4,Bio} * \left(1 - \frac{S^*_{Alk,p}}{S_{Alk,p}}\right)^n \qquad \text{Eq. 3}$$

According to Eq. 2, ammonia nitrogen removal objectives are obtained when the biological nitrification rate ($r_{NH4,Bio}$) is higher than the ammonia load rate combined with the nitrate reduction rate minus the maximum allowed ammonia exit rate. Thus, Electro-MBRs' reduced nitrification rates reported in the literature could be the result of the use of high HOP cathode materials and high charge loading, both of which increase the nitrate reduction rate. To further support this hypothesis, reported effects of electro-coagulation systems on Electro-MBR performances in treating ammonia nitrogen from wastewater are summarized in Table 3.

In both cases where the electrocoagulation was reported to have a negative impact on nitrification performances, ammonia loading rates were low (36 mg NH_4-N L^{-1} d^{-1} in the synthetic municipal wastewater and 13.6 mg NH_4-N L^{-1} d^{-1} in the grey water) and the charge loading were above 100 mAh L^{-1}. Considering that the maximum biological nitrification rate ($r_{NH4,Bio}$) value is equal to the ammonia load rate when no EC process is used, ammonia obtained from cathodic reduction at the aforementioned charge loading after the addition of an EC process will most probably not be removed. This sudden excess in ammonia nitrogen can thus significantly impact the reported ammonia removal percentage, especially when using cathode materials with high HOP values (Al and Fe).

319 Positive effects were reported at charge loading lower than 144 mAh L^{-1}, indicating that the ammonia
320 oxidizing bacteria can adapt their rate of biological nitrification to the small excess of ammonia nitrogen
321 generated on the cathode in the EC system. Based on a reaction engineering approach, the increased
322 Nitrosomonas relative abundance reported by Li, Dong et al. (2018) is likely to be the result of an
323 increased substrate concentration (ammonia nitrogen) resulting from the cathodic reduction of their
324 product (nitrates). Furthermore, as suggested by Battistelli, da Costa et al. (2018) and Li, Dong et al.
325 (2018), electrostimulation of both the proton transfer and enzyme activity can improve microbial
326 metabolism and increase cell growth (Battistelli, da Costa et al. 2018, Li, Dong et al. 2018).

327 *3.2. Anodic dissolution rate and coagulant dosage*

328 The anodic dissolution rate governs the charge loading to be applied in order to reach the desired
329 coagulant concentrations in the mixed liquor. The anodic dissolution rate can be theoretically estimated
330 using Faraday's law (Eq. 4) (Li, Dong et al. 2018).

$$\dot{m} = \frac{M}{Fn} I \qquad \text{Eq. 4}$$

331 Where $\dot{m}$ is the anodic dissolution rate (g s^{-1}), M is the molar mass of the anode material (g mol^{-1}), I is
332 the applied current (A), F is Faraday's constant (96.5 C mol^{-1}), and n is the valence of the anode material.
333 In order to estimate the anodic dissolution rate of iron anodes in a leachate treating Electro-MBR,
334 experiments were conducted in the batch reactor at 0.2A with both a NaCl solution with a similar
335 conductivity as composting leachate (0.2A – Synthetic), as well as real composting leachates (0.2A –
336 Leachate). Figure 5 compares the experimental results to the theoretical dissolution rate calculated with
337 Faraday's law.

338 According to Faraday's law, an iron anode should dissolve in the form of ionic three-valent iron at a rate
339 of 0.7 mg Fe^{3+} mAh^{-1}. When conducting the anodic dissolution experiment with real leachates, the

measured rate was 0.82 ± 0.05 mg Fe^{3+} mAh^{-1} (obtained from a duplicate), which is consistent with the expected theoretical rate. Iron dosages for Condition #2 (100 mAh L^{-1}), #3 (200 mAh L^{-1}), and #4 (400 mAh L^{-1}) were estimated to be 82, 165, and 325 mg Fe^{3+} L^{-1}, respectively, using the following equation (Eq. 5):

$$\dot{Fe} = \dot{m} * \frac{I}{V} * HRT \qquad \text{Eq. 5}$$

3.3. Where $\dot{Fe}$ is the iron dosage from anodic dissolution (mg Fe^{3+} L^{-1})Effect of the charge loading on leachate contaminant removal

3.3.1. Nitrification process performance

MBRs' nitrification performances were reported to be either positively or negatively affected by the addition of a submerged EC process inside its mixed liquor. The discussion in Section 3.1. showed that negative impacts of the EC process were potentially attributed to the selection of the cathode material. With high HOP material, the cathodic nitrate reduction rate pushes the equilibrium toward higher concentrations of ammonia nitrogen in the reactor, leading to lower nitrification performances. Ti/Pt cathodes were found to have an electrochemical nitrate reduction rate of more than 85% lower than conventional cathode materials used in EC processes (Al, Fe and stainless steel) (Dia, Drogui et al. 2017). Results in Figure 4b showed that no significant quantities of ammonia nitrogen are produced from nitrate cathodic reduction when exposed to charge loading ranging from 200 to 1 000 mAh L^{-1} when a Ti/Pt cathode is used. Thus, by using a Ti/Pt cathode in an Electro-MBR, it was possible to measure the effect of different DC charge loading, as well as the iron concentration on the Electro-MBR's nitrification performances, by avoiding nitrate reduction. Table 4 presents the nitrogen load rates (NLR) and removal percentage for the Electro-MBR operated under charge loading ranging from 0 to 400 mAh L^{-1}

360 (Coagulant dosage ranging from 82 to 325 mg Fe^{3+} L^{-1}). The high NLR measured at Condition #3 (200
361 mAh L^{-1}) is the result of a HRT change from 24h to 12h, which was done in order to assess the effect of
362 doubling the NLR on the Electro-MBR's capacity to remove ammonia nitrogen.

363 Interestingly, the ammonia nitrogen removal percentage remained at >99.7% in all conditions with
364 permeate concentrations ranging from 0.18 to 0.43 mg NH_4-N L^{-1}, which indicate no inhibitory effect of
365 the charge loading or the iron concentration on the nitrification performance. Removal rates of 119 to
366 125 mg NH_4-N L^{-1} d^{-1} and 243 NH_4-N L^{-1} d^{-1} were measured at HRTs of 24h and 12h, respectively. A
367 previous study on the performance of a MBR treating composting leachate reported similar removal
368 percentages and rates reaching up to 740 mg NH_4-N L^{-1} d^{-1}, indicating no negative impact of the
369 electrocoagulation on the MBR process (Roy, Drogui et al. 2020). Furthermore, the change in rates
370 when the HRT was decreased from 24h to 12h indicates that the amoA gene expressing bacteria from
371 the mixed liquor can quickly adapt to variable ammonia load rates and therefore maintain an almost
372 complete ammonia removal without inhibition from the application of a charge loading up to 400 mAh L^{-}
373 ^{1}.

374 The nitrification performances of a biological system also depend on the buffering capacity of the
375 wastewater to be treated. According to the stoichiometry of the nitrification reaction, 7.14 mg of
376 alkalinity (as $CaCO_3$) is consumed for every milligram of ammonia nitrogen oxidized to nitrate (Roy,
377 Benkaraache et al. 2019). If this ratio is not maintained, a pH drop will occur and nitrification will be
378 inhibited (Lee, Kim et al. 2001). Roy et al. (2020) reported that a residual concentration of 200 mg
379 $CaCO_3$ L^{-1} is required to maintain high nitrification performances (Roy, Drogui et al. 2020). Therefore,
380 alkalinity consumption was monitored at each tested condition. Both feed and permeate concentrations
381 are presented in Table 4. Without applied current, the measured alkalinity consumption was 7.51 mg
382 $CaCO_3$ per 1 mg NH_4-N L^{-1} and was entirely attributed to biological nitrification. This ratio was assumed
383 to be constant for all other conditions, and any consumed excess alkalinity was attributed to the EC

process. The EC process's alkalinity consumption increased from 8 mg $CaCO_3$ L^{-1} to 269 mg $CaCO_3$ L^{-1} when the applied charge loading increased from 100 to 400 mAh L^{-1}. At 400 mAh L^{-1}, permeate's Ca^{2+} concentration decreased from 178.1 to 78.6 mg Ca^{2+} L^{-1}, which is almost equimolar to the loss in alkalinity (0.93 mol Ca^{2+}:mol $CaCO_3$). This alkalinity consumption thus results from the formation of a $CaCO_3$ precipitate on the cathode surface where highly alkaline conditions are created by the release of OH^-. Therefore, EC can consume alkalinity if no current inversion is done in order to dissolved the $CaCO_3$ precipitate on the cathode surface (Zhu, Clifford et al. 2005). Therefore, if the alkalinity in the wastewater is not sufficient to account for both the biological and the EC consumption, this parameter can also lead to lower nitrification performances in Electro-MBRs.

3.3.2. Total organic carbon and total phosphorus removal performances

Table 5 presents the feed and permeate TOC and dissolved iron concentrations from the Electro-MBR when treating co-composting leachates operated with charge loading ranging from 0 to 400 mAh L^{-1}.

When operated without applied current, the MBR achieved a mere 13% removal in TOC, indicating that the recalcitrant fraction of TOC represents 87% of the organic contamination. The electrochemical dosage of ferric ions at concentrations of 82 to 325 mg Fe^{3+} L^{-1} (corresponding to charge loading of 100 to 400 mAh L^{-1}) then increased the TOC removal from 25 to 38%, respectively. These results are in accordance with those of Maleki and al. (2009) which reported COD removal percentages of 28% from composting leachate at an iron dosage of 680 mg Fe L^{-1} using iron salts as coagulating agents (Maleki, Zazouli et al. 2009, Mahvi, feizabadi et al. 2015). Lower dissolved iron concentrations measured in the permeate when compared to the feed indicate that all the iron released from the sacrificial anode was precipitated with the biomass and remained in the mixed liquor.

In order to assess the efficiency of the ferric ions in coagulating the leachate's TOC in an Electro-MBR, a mass balance between the amount of ferric ions generated and the TOC removed was conducted. Figure

6a distinguishes the biologically oxidized TOC from the chemically precipitated TOC, while Figure 6b presents the ratios of TOC coagulated per mass of ferric iron generated in the mixed liquor at charge loading from 100 to 400 mAh L^{-1}.

According to the results shown in Figure 6a, the MBR removed 35 mg TOC L^{-1} when operated without applied current at a HRT of 24h and a SRT of 45 days. Then, at Conditions #2, #3, and #4, all the TOC that was removed beyond the aforementioned concentration was assumed to be physico-chemically precipitated by the EC process. The most TOC removal was obtained at 400 mAh L^{-1}, with a total of 67 mg TOC L^{-1}. However, in terms of efficiency, the highest ratio of TOC:Fe^{3+} was obtained at 100 mAh L^{-1}, with a value of 0.44 ± 0.5 mg TOC mg Fe^{-1}. For charge loading of 200 to 400 mAh L-1, the TOC:Fe^{3+} efficiency reached a plateau at 0.19 ± 0.2 mg TOC mg Fe^{-1}. These results are in accordance with those from Amokrane et al. (1995) (Amokrane, Comel et al. 1997). At low iron concentrations (<200 mg L^{-1}), highly charged organic molecules react quickly with ferric ions and then precipitate. The TOC:Fe^{3+} ratio then remains constant (removal percentage is proportional to the ferric iron dosage) until most organic molecules with polar moieties have precipitated.

The Electro-MBR efficiency in removing phosphorus from leachate was also assessed by conducting a mass balance between the quantity of generated ferric ions and the precipitated total phosphorus (P_{tot}). Table 6 presents the feed and permeate P_{tot} concentrations, as well as the P_{tot}:Fe^{3+} ratio, for the Electro-MBR treating co-composting leachates and operated under charge loading ranging from 0 to 400 mAh L^{-1}.

P_{tot} removal percentages are positively correlated with the iron dosage, which is in accordance with previous studies on Electro-MBRs (Bani-Melhem and Smith 2012, Tian, He et al. 2016). As opposed to the TOC, P_{tot} removal efficiency was at its highest at 400 mAh L^{-1} with a P_{tot}:Fe^{3+} ratio of 60 mg P_{tot} g Fe^{-1}. The increased P_{tot} removal efficiency at higher ferric ion concentrations is linked to the TOC removal. At

low Fe^{3+} concentrations, all the coagulant is scavenged by the highly charged TOC. Then, at higher Fe^{3+} concentrations, more coagulant remains available for the P_{tot} to react with, leading to higher P_{tot}:Fe^{3+} ratios.

3.4. Effect of the charge loading on the mixed liquor microbial population diversity and structure

In the previous sections, it was demonstrated that lower nitrification performances, rather than being related to inhibition phenomena, were instead potentially related to the magnitude of the cathode material's nitrate electrochemical reduction rate and to a possible lack of buffering capacity (alkalinity) in the wastewater. To further support this statement, changes in microbial population diversity and structure were monitored at each tested condition. The α-diversity (Chao1), as well as the rarefaction curves, are presented in Figure 7a and Figure 7b, respectively. The Chao1 method was selected in order to estimate the number of unique species while accounting for low-abundance operational taxonomic units (OTUs) (Dhariwal, Chong et al. 2017).

The Chao1 α-diversity analysis conducted on both the MBR's and the Electro-MBR's mixed liquors showed a stable diversity in the microbial community (α-diversity = 169 ± 3), except at Condition #3 (α-diversity = 140), where the Electro-MBR was operated at a HRT of 12h instead of 24h. In terms of species richness analysis, Condition #3 also resulted in the lowest number of OTUs, with 1 980 OTUs. The number of OTUs in the other samples ranged from 2 120 to 2 425, with the sample that was exposed to a charge loading of 400 mAh L^{-1} having the highest count. The similarities in α-diversity and species richness between the mixed liquor samples collected from both the MBR and the Electro-MBR when operated at a HRT of 24h, indicate that both charge loading that range from 100 to 400 mAh L^{-1}, as well as the associated ferric ions concentrations, both have no significant impact on the diversity of microbial genus in mixed liquor. Furthermore, the significantly lower α-diversity measured at a HRT of

453 12h indicates that the contaminant load rate has more impact on the community diversity than the
454 charge loading applied would.

455 Both the MBR's and the Electro-MBR's microbial population structure at the genus level is presented in
456 Figure 8. No dominant OTU genus (relative abundance >10%) was detected in the MBR's mixed liquor
457 without applied current. Instead, the population contained three sub-dominant OTU genera (relative
458 abundance >3%) that were: Reyranella (7.8%), Trichococcus (5.1%), and Nitrosomonas (4.8%). Both
459 Reyranella and Trichococcus are heterotrophic bacteria ubiquitous to biological wastewater treatment
460 processes. Trichococcus are filamentous bacteria commonly found in bulking sludge (Scheff, Salcher et
461 al. 1984). Vandewalle et al. (2012) also reported this genus to be dominant in urban sewer infrastructure
462 (Vandewalle, Goetz et al. 2012). After the addition of the EC system with a HRT of 24h (Condition #2 and
463 #4), Reyranella and Nitrosomonas remained subdominant genera, while Trichococcus became a
464 dominant genus. At charge loading of 100 and 400 mAh L^{-1}, the dominant and sub-dominant genera's
465 relative abundance were: 10.3% and 12.7% for Trichococcus, 8.7% and 4.3% for Reyranella, and 5.6%
466 and 8.2% for Nitrosomonas. These results indicate that the presence of coagulant might have favoured
467 the development of floc-forming bacteria, which could be related to the lower membrane fouling
468 propensity in Electro-MBRs (Borea, Naddeo et al. 2017). Another interesting observation is the positive
469 correlation between the Nitrosomonas, the main ammonia nitrogen oxidizing bacteria identified in the
470 mixed liquor, and the charge loading. Compared to the MBR without applied current, Nitrosomonas
471 relative abundance almost doubled when exposed to a charge loading of 400 mAh L^{-1}, increasing from
472 4.8% to 8.2%. This result supports the findings of Li et al. (2018), which reported an increase in
473 Nitrosomonas relative abundance of 1.45% to 3.12% when exposed to a charge loading of 14 mAh L^{-1} in
474 the study's Electro-MBR treating a synthetic wastewater (40 mg NH_4-N L^{-1}) (Li, Dong et al. 2018). The
475 effect of the electrocoagulation system is explained by two elements; first, since a microbial population
476 size is generally proportional to the amount of substrate available, the increased amount of substrate

477 (ammonia nitrogen) from nitrate cathodic reduction is likely to result in a larger population of
478 Nitrosomonas. Second, as previously reported by Qian et al. (2017), the use of EC integrated in
479 bioreactors can improve the biomass enzymatic activity of nitrifying bacteria, mainly the ammonia
480 monooxygenase (AMO), hydroxylamine oxidoreductase (HAO), and nitrite oxidoreductase (NOR), which
481 would result in increased nitrifying bacteria growth (Qian, Hu et al. 2017). In terms of contaminant load
482 rate, the change in HRT had a significant impact on the microbial population structure. At a HRT of 12h
483 (Condition #3), Trichococcus genus increased its relative abundance to 20.2%, and several subdominant
484 genera also appeared, including: Nitrobacter (5.4%), Chryseobacterium (4.1%), Pirellula (4.1%),
485 Proteiniclasticum (3.3%), and Paludibaculum (3.0%). In a previous study on MBR treating composting
486 leachates, all these subdominant genera, with the exception of Chryseobacterium, were identified at a
487 SRT of 45 days under similar contaminant load rates. Nitrobacter was also reported in higher
488 abundances at low HRT (Roy et al. 2020). Interestingly, at a HRT of 12h, Nitrosomonas relative
489 abundance decreased to only 0.8% while the NLR increased from 124 to 244 mg NH_4-N L^{-1} d^{-1}. Despite a
490 lower Nitrosomonas relative abundance when compared to the other conditions, the ammonia nitrogen
491 removal percentage at Condition #3 remained above 99.7%. These results are in accordance with those
492 of Roy et al. (2020), which reported that Nitrosomonas growth is inhibited in a MBR by increasing the
493 organic load rate (OLR), yet nitrification performances are unaffected by the OLR and the Nitrosomonas
494 relative abundance.

495 Results from the microbial community structure and diversity analyses indicate that neither charge
496 loading ranging from 100 to 400 mAh L^{-1} nor their associated ferric iron concentrations have any
497 inhibitive impact on Nitrosomonas growth. Rather, Nitrosomonas growth is positively correlated to the
498 applied charge loading under these conditions. The HRT was found to have significantly more impact on
499 the microbial population structure than the DC current. Thus, lower nitrification performances are
500 expected in studies where membrane fouling is compensated by constantly varying the HRT (ex.: Bani-

Melhem and Smith (2012) (Bani-Melhem and Smith 2012)), whereas higher performances are expected when the membrane is instead changed or frequently cleaned. As opposed to the hypothesis made from the authors of these studies, these lower performances are the result of a constant adaptation of the mixed liquor to the contaminant load rates, as well as a loss in Nitrosomonas population, and are not related to the applied current.

4. Conclusion

In this study, Electro-MBR technology was evaluated for the treatment of high-strength ammonia wastewater with an initial ammonia nitrogen concentration of 124 ± 4 mg NH_4-N L^{-1}. Using a combination of a sacrificial iron anode and a Ti/Pt cathode, as well as a charge loading of 400 mAh L^{-1}, removal percentages of 99.8%, 38%, and 99.0% of ammonia nitrogen, TOC, and P_{tot}, respectively, were obtained. This configuration of Electro-MBR is thus promising in solving the challenges associated with the treatment of waste originating leachates.

This study's second objective was to shed light on reported divergent impacts on nitrification performances when an EC process was added to an MBR. First, the effect of the cathode material on the cathodic nitrate reduction rate was evaluated. Results showed that high HOP cathode materials, such as graphite, reduce nitrate to ammonia nitrogen at a rate that outpaces the biological nitrification rate. The solution to overcome this issue is to use low HOP cathode material such as Ti/Pt, which favours water reduction to hydrogen. Then, the effect of the charge loading the nitrification performances was assessed. No significant impact was observed on the nitrification performances when using a Ti/Pt cathode with charge loading ranging from 100 to 400 mAh L^{-1}. These charge loading had also no significant impact on the microbial community diversity, while a lower HRT resulted in a lower OTUs count. In terms of community structure, Nitrosomonas relative abundance increased proportionally with

the charge loading, showing no inhibitive phenomena due to the iron concentration or the applied DC current. In general, high nitrification rates in Electro-MBR can be obtained when using a Ti/Pt cathode.

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Table 1 Operating parameters of the Electro-MBR

Parameters		Condition #1	Condition #2	Condition #3	Condition #4
Period length	(d)	28	14	14	14
Charge loading	(mAh L^{-1})	0	100	200	400
Current density	A m^{-2}	0	2.3	4.5	9.1
Temperature	(°C)	20 ± 1	20 ± 1	20 ± 1	20 ± 1
HRT	(hr)	24 ± 2	24 ± 2	12 ± 1	24 ± 2
TSS	(mg L^{-1})	7 800 ± 50	7 870 ± 250	14 340 ± 620	12 660 ± 710
MLVSS	(mg L^{-1})	5 100 ± 50	4 995 ± 170	6 840 ± 260	6 480 ± 570
OLR	(mg COD L^{-1} d^{-1})	1100 ± 110	1100 ± 110	1790 ± 180	1100 ± 110
NLR	(mg NH_4-N L^{-1} d^{-1})	125 ± 1	124 ± 2	244 ± 2	119 ± 4
PLR	(mg P L^{-1} d^{-1})	4.5 ± 0.2	4.9 ± 0.3	17.4 ± 0.8	9.7 ± 1.2

Table 2 Co-composting leachate characterization

Parameter	Units	Min. - Max. value
Alkalinity	(mg $CaCO_3$ L^{-1})	1 493 - 1 593
Ammonia nitrogen	(mg NH_4-N L^{-1})	114 - 129
BOD_5	(mg L^{-1})	70 - 210
COD	(mg L^{-1})	860 - 1 200
TOC	(mg L^{-1})	244 - 297
Conductivity	(mS cm^{-1})	3.65 - 3.78
Nitrate	(mg NO_3-N L^{-1})	0.1 - 0.6
pH	-	7.9 - 8.3
Total dissolved solids	(mg L^{-1})	2 190 - 2 820
Total nitrogen	(mg N L^{-1})	129 - 145
Total phosphorus	(mg P L^{-1})	4.3 - 10.7
Total solids	(mg L^{-1})	2 440 - 2 830

Table 3 Reported effects of electro-coagulation systems on Electro-MBR performances in treating ammonia nitrogen

Electrodes material		Charge loading	Wastewater	Effect of Electro-coagulation on nitrification	Authors
Anode	**Cathode**	**(mAh L^{-1})**			
Aluminum	Iron	110	Synthetic municipal wastewater	**Negative** 27% loss in ammonia nitrogen removal	(Bani-Melhem and Elektorowicz 2011)
Aluminum	Aluminum	680	Grey water	**Negative** 20% loss in ammonia nitrogen removal	(Bani-Melhem and Smith 2012)
Iron	Stainless steel	36 - 165	Synthetic municipal wastewater	**Positive** Improved nitrification at charge loading <144 mAh L^{-1}	(Tafti, Mirzaii et al. 2015)
Aluminum	Stainless steel	15 - 40	Synthetic municipal wastewater	**Positive** 10.5 to 27% increased in ammonia nitrogen removal	(Borea, Naddeo et al. 2017)
Graphite	Iron	14	Synthetic municipal wastewater	**Positive** Nitrosomonas relative abundance increase from 1.45 to 3.12%	(Li, Dong et al. 2018)

Table 4 Ti/Pt - Fe Electro-MBR's ammonia nitrogen removal performances and associated alkalinity consumption

Charge loading	Ammonia nitrogen				Alkalinity		
	NLR	Feed	Permeate	Removal	Feed	Permeate	EC consumption
(mAh L^{-1})	(mg NH_4-N L^{-1} d^{-1})	(mg NH_4-N L^{-1})	(mg NH_4-N L^{-1})	(%)	(mg $CaCO_3$ L^{-1})	(mg $CaCO_3$ L^{-1})	(mg $CaCO_3$ L^{-1})
0	125	125	0.43	99.7%	1541	598	0
100	124	124	0.34	99.7%	1528	590	8
200	244	122	0.38	99.7%	1563	426	221
400	119	119	0.18	99.8%	1548	383	269

Table 5 Ti/Pt - Fe Electro-MBR's TOC removal performances

Charge loading	**Feed**		**Permeate**		**TOC Removal**
(mAh L-1)	**(mg TOC L^{-1})**	**(mg Fe L^{-1})**	**(mg TOC L^{-1})**	**(mg Fe L^{-1})**	**(% TOC)**
0	260	3.2	225	0.7	13%
100	280	3.7	211	0.5	25%
200	263	3.2	187	0.3	29%
400	268	4.0	166	0.2	38%

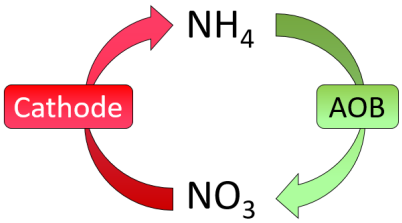
Table 6 Ti/Pt - Fe Electro-MBR's Total phosphorus removal performances

Charge loading	PLR	Feed	Permeate	Removal	$Y_{(P/Fe)}$
(mAh L^{-1})	(mg P L^{-1} d^{-1})	(mg P L^{-1})	(mg P L^{-1})	(%)	(mg P:g Fe)
0	4.5	4.5	1.3	71.1%	0
100	4.9	4.9	0.5	89.8%	45.7
200	17.4	8.7	0.3	96.6%	43.4
400	9.7	9.7	0.1	99.0%	60.3

Electrochemical reduction:

$NO_3^- + H_2O + 2e^- \leftrightarrow NO_2^- + 2\,OH^-$

$NO_2^- + 5H_2O + 6e^- \leftrightarrow NH_3 + 7\,OH^-$

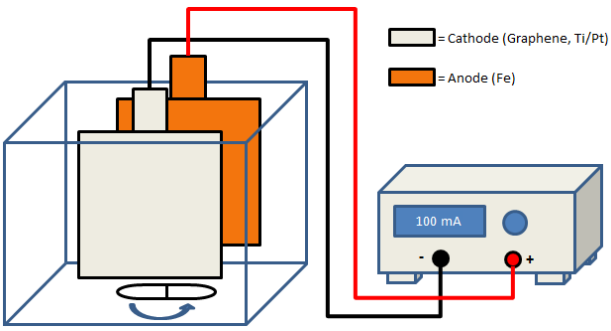


Biological nitrification:

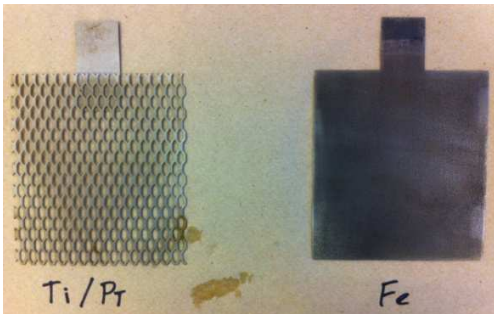
$NH_4^+ + 1.5O_2 \xrightarrow{Nitrosomonas} NO_2^- + H_2O + 2H^+$

$NO_2^- + 0.5O_2 \xrightarrow{Nitrobacter} NO_3^-$

Figure 1 Nitrogen cycle in an aerobic biological / electrochemical combined process

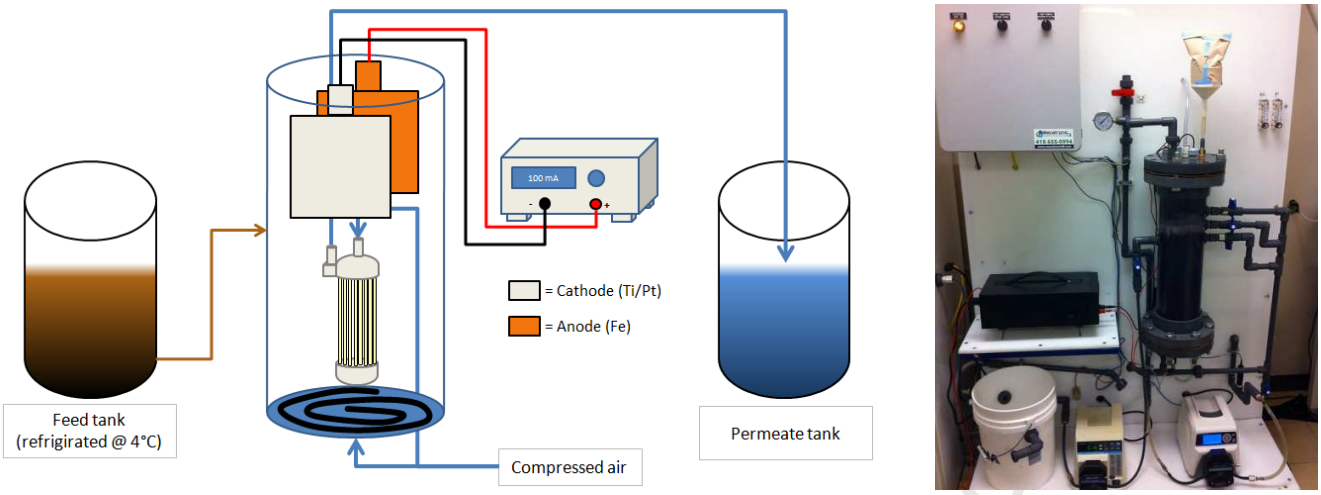


(a)



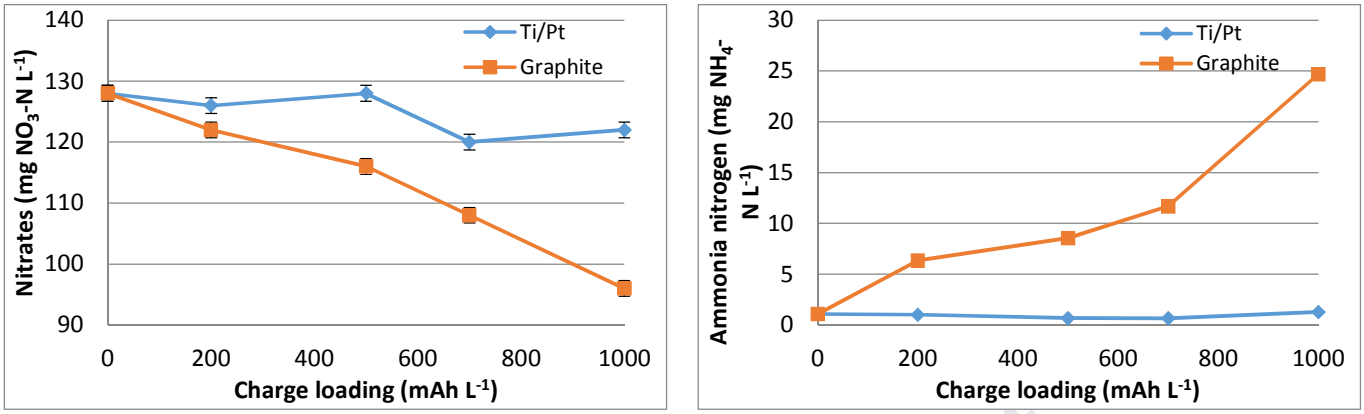
(b)

Figure 2 (a) Diagram of the lab-scale electrocoagulation batch reactor, and (b) set of electrodes used in this study



(a) (b)

Figure 3 (a) Diagram of the lab-scale submerged membrane bioreactor and (b) Picture of experimental set-up



(a) **(b)**

Figure 4 Effect of the charge loading on leachate's a) nitrate concentrations and b) ammonia nitrogen concentration

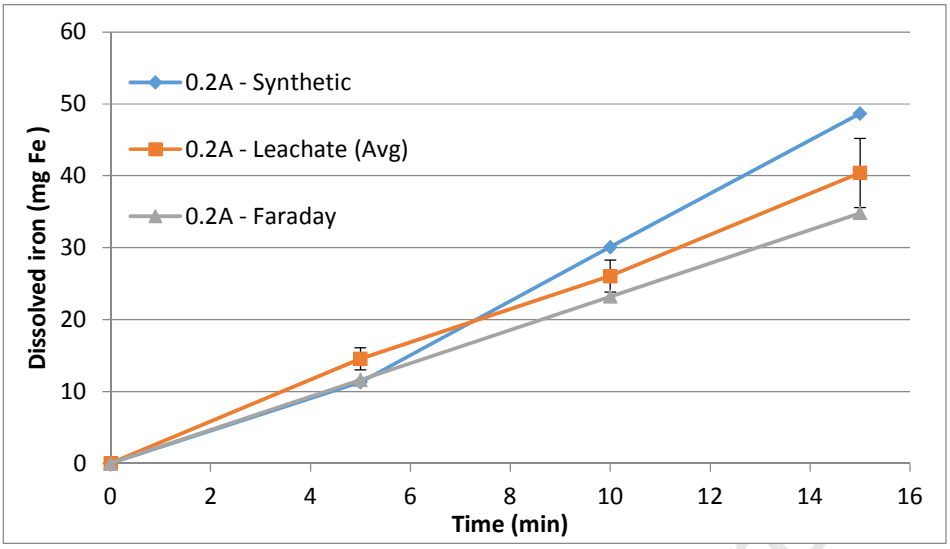
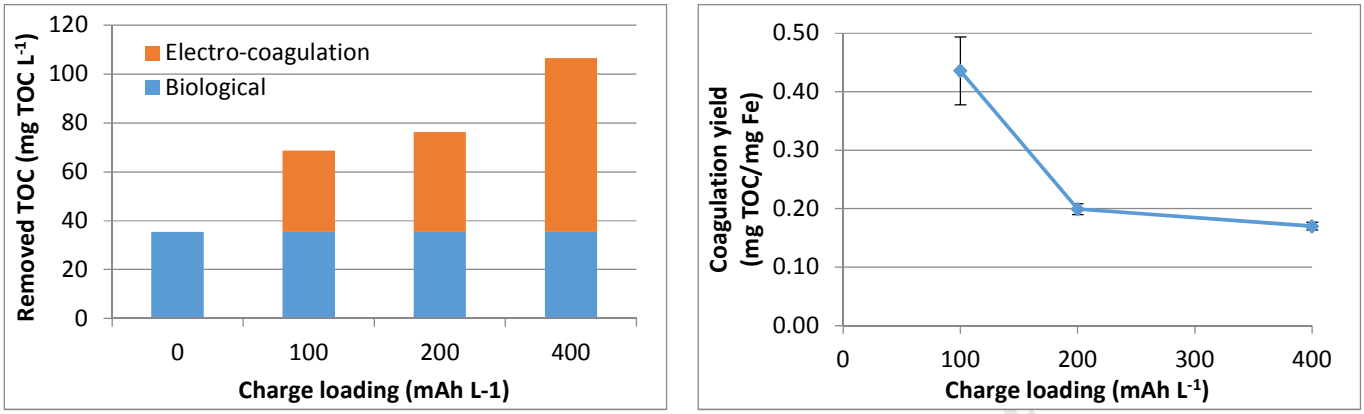
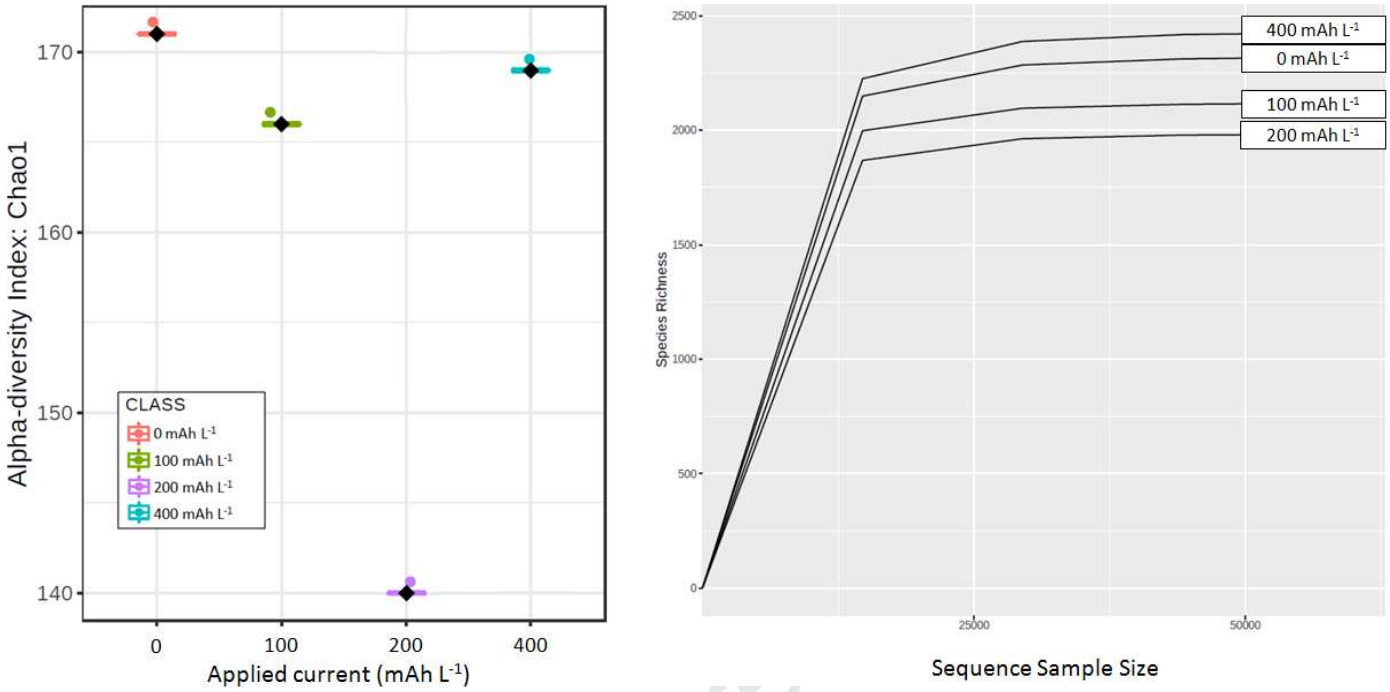


Figure 5 Anodic dissolution rate measurements at a current intensity of 200 mA



(a) (b)

Figure 6 (a) Biological and physico-chemical fraction of TOC removed by the Electro-MBR and (b) TOC coagulation efficiency at different charge loading



(a) (b)

Figure 7 (a) Alpha-diversity (Chao1) and (b) rarefaction curves of mixed liquor samples collected from the different Electro-MBR's operating conditions

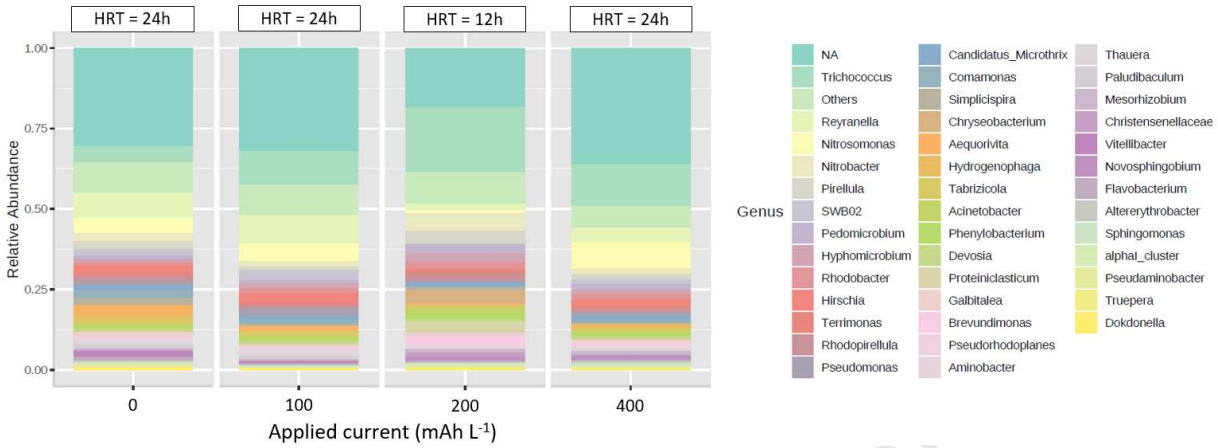


Figure 8 MBR's and Electro-MBR's mixed liquor microbial population structure at the genus level

Highlights:

- Nitrate cathodic reduction rate depends on the cathode material's hydrogen overpotential
- Ferric ion dosage of 82 to 325 mg Fe L^{-1} has shown no nitrification inhibition phenomena
- Nitrification inhibition is avoided by the use of a platinized titanium cathode
- Nitrosomonas relative abundance is positively correlated to the charge loading
- A Ti/Pt-Fe Electro-MBR can simultaneously remove >99% of NH_4 and P_{tot} at 400 mAh L^{-1}

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: