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Electrifying biotrickling filters for the treatment of aquaponics wastewater

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Abstract

This work aimed to study the electrification of biotrickling filters by means of Microbial electrochemical technologies (MET) to develop an easy-to-assemble and easy-to-use MET for nitrogen removal without external aeration nor addition of chemicals. Four different designs were tested. The highest ammonium and nitrate removal rates (94 $gN \cdot m^{-3} \cdot d^{-1}$ and 43 $gN \cdot m^{-3} \cdot d^{-1}$, respectively) were reached by combining an aerobic zone with an electrified anoxic zone. The standards of effluent quality suitable for hydroponics were met at low energy cost (8.3×10^{-2} $kWh \cdot gN^{-1}$). Electrified biotrickling filters are a promising alternative for aquaponics and a potential treatment for organic carbon-deficient ammonium-contaminated waters.

Keywords: Circular bioeconomy, Bioelectrochemical systems, Biologic nitrogen removal, Microbial electrochemical technologies, Power-to-food.

1. INTRODUCTION

The development of innovative and environmental-friendly food cultivation methods is required to face the near future (Godfray et al., 2010). One of the fastest growing food-producing sectors is aquaculture (FAO, 2018). World production has increased from 3 to 80 million tonnes of fish from 1970s to 2017. Thus, it accounts for about 50 % of the world's fish consumption. Aquaculture could decrease the pressure on the endangered aquatic wildlife, but its development needs a revision. Aquaculture impacts the environment by producing fish feed (usually produced from fish oils/flours) and nitrogen/antibiotics discharges (Read and Fernandes, 2003). At the same time, industrial agriculture is also being scrutinized. The expansion of agriculture causes increasing land use, higher fresh water consumption as well as nitrogen, phosphorus, and pesticides overloads (Tilman et al., 2001). In this perspective, hydroponics (soilless plant cultivation) is considered as an alternative to conventional agriculture as it decreases the demand for land, water, nutrients, and pesticide dosing (Gwynn-Jones et al., 2018). If nutrient-rich effluents coming from aquaculture are used in hydroponics and vice versa, a virtuous loop is generated, i.e., this is aquaponics. Aquaponics allows the production of both fish and edible plants while minimizing the environmental impact compared to conventional fishing and agriculture (FAO, 2014; Tyson et al., 2011), closing urban biocycles (Venkata Mohan et al., 2020).

From the conceptual point of view, aquaponics is a win-win situation, but its real-world implementation requires the correct management of the nitrogen cycle inside the system (Wongkiew et al., 2017). On the one hand, aquaculture effluents are usually characterized by high ammonium content, since about 60 - 70 % of the feed is excreted as ammonia (Kissil and Lupatsch, 2004). On the other hand, hydroponics requires almost ammonium-free water (< 0.8 mgN-$NH_4^+ \cdot L^{-1}$) but with a certain amount of nitrate

(1 - 34 mgN-NO_3^-·L^{-1}) as nitrogen source of cultured plants (FAO, 2014). In consequence, conventional nitrification-denitrification processes, usually focusing on full nitrogen removal, need to be adapted to the specific requirements of aquaponics. Firstly, ammonium generated in the aquaculture pond should be converted into nitrate followed by a controlled denitrification in order to avoid high nitrate accumulation (< 90 mgN-NO_3^-·L^{-1}) that could affect fish and plants growth (FAO, 2014; van Rijn et al., 2006), and to ensure no nitrite presence (< 0.3 mgN-NO_2^-·L^{-1}) due to its toxicity for plants and fish (Colt, 2006; FAO, 2014). Aquaculture recirculating systems can be easily adapted to aquaponics as they are already equipped with, e.g., biotrickling filters being characterized by a good nitrification performance. However, the denitrification performance of such systems is poor due to the lack of organic matter (C/N < 3) (Mook et al., 2012; van Rijn et al., 2006). Thus, an externally added electron donor is needed to control and adjust the nitrate content. The most common external electron donor is organic matter, but it introduces additional cost factors (i.e. chemical dosage and sludge disposal). By finding a solution for the treatment of aquaponics, a solution for the treatment of other wastewaters with low C/N ratio wastewaters (e.g. some urban wastewater) could be also found (Mook et al., 2012).

Primary microbial electrochemical technologies (MET) have emerged as a biotechnological alternative for directly supplying an electron donor/acceptor to electroactive microorganisms by means of an electron conductor termed electrode (Schröder et al., 2015). Integrating primary MET in aquaponics could result in a considerable improvement thereof, as they were demonstrated to drive both nitrification (Vilajeliu-Pons et al., 2018) and denitrification (Gregory et al., 2004). Still little is known about the recently discovered electricity-linked ammonium removal (Shaw et al., 2020), thus ammonium is usually oxidized into nitrate aerobically (He et al., 2016,

Virdis et al., 2008). Microbial electrochemical denitrification has been widely tested in different waters such as wastewater (Virdis et al., 2008), groundwater (Pous et al., 2015a), or aquaculture effluents (Marx Sander et al., 2018). The microbial structure and activity of denitrifying MET rapidly changes with the mode of operation (Pous et al., 2015b) allowing better control of denitrification by fine-tuning different operational parameters, e.g., cathode potential (Virdis et al., 2009), current density (Park et al., 2005), pH (Clauwaert et al., 2009), or the hydraulic retention time (HRT) (Pous et al., 2017).

Besides MET implementation in aquaponics could be effective at low operational expenditures, the complexity and capital expenditures associated to its conventional configuration represents a matter of concern (for instance, usage of electrodes, membranes, potentiostats, etc.) (Sleutels et al., 2012). However, the development of MET-based treatment concepts such as snorkels (Hoareau et al., 2019; Viggi et al., 2015) or METlands (Aguirre-Sierra et al., 2020; Prado et al., 2020) highlights the importance of the microbial ecology function over reactor materials and engineering (Koch et al., 2018). In consequence, only two components might be needed to reach an improvement of bioremediation activities: the appropriate microbiome inhering electroactive microorganisms and a conductive support serving as electrode. Conventional technologies currently used in aquaculture and aquaponics (e.g., biofilters) are based on microbial degradation at non-conductive supports (Crab et al., 2007). Yet, it can be hypothesized that a conductive support integrated in the effluent treatment site will enhance nitrification and denitrification due to the activity of electroactive microorganisms. For this reason, this work explored the potential of biotrickling filters to be electrified for improving nitrification/denitrification rates and the efficient control of the nitrate content in the effluent. Consequently, a sustainable

system was developed to improve aquaponics water recirculation by a controlled optimization of the nitrogen content in the aquaculture effluent according to actual requirements of hydroponics. This technology could be used for the treatment of other wastewaters containing ammonium at low C/N ratio.

2. MATERIALS AND METHODS

2.1. Influent characteristics

All reactors were fed with synthetic aquaculture effluent containing a representative amount of ammonium (0.2 $g \cdot L^{-1}$ NH_4Cl ; 50 $mgN\text{-}NH_4^+ \cdot L^{-1}$) (Yin et al., 2018) and 0.1 $g \cdot L^{-1}$ $MgSO_4$, 0.015 $g \cdot L^{-1}$ $CaCl_2$, 0.162 $g \cdot L^{-1}$ Na_2HPO_4, 1.072 $g \cdot L^{-1}$ KH_2PO_4, 0.25 $g \cdot L^{-1}$ NaCl, 1.05 $g \cdot L^{-1}$ $NaHCO_3$, 0.1 $mL \cdot L^{-1}$ micronutrients (Rabaey et al., 2005). All chemicals were of analytical or biochemical grade.

2.2. Study of the effect of material filling and electricity input (Reactor designs A, B, and C)

2.2.1. Reactor set-up and inoculation of reactor designs A, B, and C

Experiments were performed in tubular polyvinyl chloride (PVC) reactors of 100 cm height and 4.2 cm of internal diameter (5.0 cm external diameter – PVC 50 - 10 Atm), implying a total volume of 1385 mL (see supplementary files). In all reactors, the inlet was located at the upper side of the reactor. Influent water was dropped spread on a stainless steel mesh (mesh path light 5 x 5 mm) to get a better distribution along the whole reactor diameter and promote aeration. Water circulated downwards and the reactor water level (WL) was controlled by moving the outlet discharge point as shown in Figure 1. In a first round of tests, 3 different designs (A, B, and C) with different filling materials were used leading to different reactor net liquid volumes, A: One reactor filled with PVC granules (diameter 2 - 8 mm) representing a conventional biofilter (534 mL net liquid volume), B: Two non-polarized reactor replicates filled with

granular graphite (model 00514, diameter 1.5-5 mm, Enviro-cell, Germany) (633 ± 38 mL net liquid volume), and C: Two polarized reactor replicates filled with granular graphite (model 00514, diameter 1.5-5 mm, Enviro-cell, Germany) (655 ± 21 mL net liquid volume). In reactor C, nine graphite rods (6 mm diameter, Mersen Iberica, Spain) located every 10 cm height and inserted ca. 3 cm in the tube serving as current collectors (CCs). CCs were connected to a power source (IMHY3, Lendher, Spain). All graphite electrodes (rods and granules) were washed with 1 M NaOH and 1 M HCl prior to use. An Ag/AgCl reference electrode (+ 0.197 V vs. SHE, SE 11, Xylem Analytics Germany Sales GmbH & Co. KG Sensortechnik Meinsberg, Germany) was introduced at height of 20 cm. If not stated otherwise, all potentials provided refer to Ag/AgCl sat. KCl reference electrodes (+0.197 V vs. standard hydrogen electrode (SHE)).

All reactors were inoculated in batch mode for 23 days. Each reactor was connected to a 10 L buffer tank containing a solution with synthetic aquaculture medium (section 2.1.) and a mixed inoculum. The inoculum contained effluent from different reactors performing nitrification (Vilajeliu-Pons et al., 2018), denitrification (Pous et al., 2017), anammox (Akaboci et al., 2018) and activated sludge from the urban wastewater treatment plant of Quart (N.E. Catalonia, Spain).

2.2.2. Operation and testing of reactor designs A, B and C

After 23 days of inoculation in batch mode, reactors were switched to continuous flow mode at 0.6 ± 0.1 $L \cdot d^{-1}$ (around 1.0 d hydraulic retention time, HRT, depending on the reactor design). The reactors were operated for 118 days with these flow conditions while testing the influence of electrically connecting different current collectors located at different heights of design C (see supplementary files). The configuration of 8 connected CCs (4 anodes and 4 cathodes) was finally used for the main experiments as

it provided a better potential distribution. With this configuration, the CCs located at 50, 60, 70, and 80 cm height were used as anodes, while those at 10, 20, 30, and 40 cm height were used as cathodes. A cathode potential of – 0.3 V (Pous et al., 2015a) ought to be applied by manual tuning of the power source.

When fairly stable performance in terms of nitrogen concentrations, current density and cathode potential under the applied condition (0.6 ± 0.1 $L \cdot d^{-1}$) was reached (see supplementary files), further operational reactor parameters were tested, each for two weeks, including: i) volumetric flow rate (Q) between 0.6 ± 0.1 and 2.6 ± 0.2 $L \cdot d^{-1}$ corresponding to HRTs between 0.3 and 1.1 d, respectively, and ii) presence of oxygen at the influent (Influent reservoir stored in a 10 L self-collapsible bags and flushed, or not, with N_2 gas for 15 min). In total, the reactors were operated for 189 d.

2.3. Reactor for performance enhancement (Reactor design D)

2.3.1 Reactor set-up and inoculation of reactor design D

After taking into consideration the obtained results from the first reactor designs, a second set of experiments was performed using reactor design D (Figure 1 and supplementary files). Two reactor replicates were constructed with the lower half of the reactors filled with granular graphite. Two titanium rods (Grade 1, 8 mm diameter, Polymet Reine Metalle, Germany) were inserted ca. 3 cm in the tube serving as CCs for the anode and the cathode zone. Thereby, the cathodic and anodic CCs were located at 12 and 45 cm height, respectively. A stainless steel mesh (30 cm length, mesh path light 5 x 5 mm) was introduced at the inner wall of the PVC tube for improving potential distribution in the cathode zone. According to this set-up, the cathode zone had a height of 30 cm with 280 ± 6 mL of net cathode volume (NCC). The upper half of the reactors was filled with PVC granules. In consequence, the net liquid volume of the whole reactors was 777 ± 10 mL. An Ag/AgCl reference electrode (+ 0.197 V vs. SHE, SE 11,

Xylem Analytics Germany Sales GmbH & Co. KG Sensortechnik Meinsberg, Germany) was introduced next to the cathode collector (height 12 cm) to measure the electrode potentials.

Reactors were inoculated following the same protocol and using effluent taken from the same reactors as it was performed for reactors A, B, and C (described in section 2.2.1.).

2.3.2. Operation and testing of reactor design D

After 21 days of inoculation in batch mode, reactors were switched to continuous flow mode with 0.7 ± 0.1 $L \cdot d^{-1}$ (1.2 ± 0.2 d HRT). This operation was followed for 44 d (days 30-44 constitute representative reactor operation reactors under this condition). Subsequently, different operational parameters were tested, each for 2 - 3 weeks: i) volumetric flow rates between 0.7 ± 0.1 and 2.3 ± 0.2 $L \cdot d^{-1}$ corresponding to HRTs between 0.3 and 1.2 d, respectively, ii) Water level (WL) of 50 % and 75% of reactor height, and iii) not polarized graphite granule bed (i.e., open cell potential, OCP) for one week. In total, the reactors were operated for 142 d. All tests were performed with an influent flushed with N_2 for 15 min.

2.4. Chemical analyses and calculations

Influent and effluent samples were taken twice a week to measure pH, conductivity, nitrite ($N\text{-}NO_2^-$), nitrate ($N\text{-}NO_3^-$), and ammonium ($N\text{-}NH_4^+$) in accordance with the American Public Health Association (APHA) standards (APHA, 2005). Nitrous oxide and dissolved oxygen (DO) were measured at the effluent of the reactor D using a N_2O liquid-phase microsensor (Unisense, Denmark) and an oxygen sensor (O.D. 6050, Crison - Hach Lange GmbH, Germany).

Ammonium removal was calculated as the difference between influent and effluent ammonium content. Total nitrogen (N-TN) removal was calculated as the total nitrogen

($N\text{-}NH_4^+$ + $N\text{-}NO_2^-$ + $N\text{-}NO_3^-$) removal difference between influent and effluent. Ammonium and total nitrogen removal rates ($N\text{-}NH_4^+{}_{RR}$ and $N\text{-}TN_{RR}$) were calculated taking into account the HRTs of reactors. HRT was calculated using the total net reactor volume of each reactor and the respective flow rates.

The electricity consumption of the systems C and D ($kWh \cdot gN_{rem}^{-1}$) were calculated using the voltage and current applied with the power source together the mass of nitrogen removed.

3. RESULTS AND DISCUSSION

3.1. Towards the electrification of biotrickling filter: Understanding the effect of material filling and electricity input over nitrogen removal (Reactor designs A, B, and C)

3.1.1. Influence of volumetric flow rates

Reactor designs A, B and C were designed with different configurations (Figure 1) to evaluate how the material filling and electricity input can influence biologic ammonium removal in a biotrickling filter. After an start-up process, the systems reached an steady-state (see supplementary files). As the last period of this first test phase (ca. 14 d) was fairly stable in terms of nitrogen concentrations, current density, and cathode potential at the applied condition (1.0 ± 0.1 d HRT), experimental test series started. In order to study the effect of different hydraulic retention times (HRT) on the performance, tests with different volumetric flow rates (Q) were performed with an oxygen-deficient influent. Under these conditions, all oxygen available for aerobic nitrification would come from air dissolution at the upper layers of the reactors. Figure 2 shows the ammonium and total nitrogen removal performance at different HRTs.

As a general trend, higher nitrification rates were observed at lower HRTs. Design A (PVC as filling material) exhibited the highest ammonium removal rates ($N\text{-}NH_4^+{}_{RR}$),

showing a maximum of 56 ± 15 $gN \cdot m^{-3} \cdot d^{-1}$ at an HRT of 0.4 d (49 ± 12 % $N\text{-}NH_4^+$ removal). Assuming that aerobic nitrification was the major microbial process for ammonium removal, it can be deduced that the usage of PVC granules allowed a better oxygen distribution from the upper reactor layers to the inner parts of the reactor compared to graphite granules, which could be related to the bigger size of PVC granules (diameter 2 - 8 mm) compared to granular graphite. For the reactor designs with granular graphite, higher nitrification performance was observed for higher Q (low HRTs) and for a polarized granule bed (i.e., reactor design C). For example, the maximum ammonium removal rate of reactor design C (39 ± 8 $gN \cdot m^{-3} \cdot d^{-1}$) was ca. 1.4 times higher than of reactor design B (28 ± 7 $gN \cdot m^{-3} \cdot d^{-1}$) at an HRT of 0.3 d. This increase of performance could be related to the usage of a power supply in design C, which was daily adjusted for keeping the cathode CCs at – 0.3 V for providing suitable conditions for bioelectrochemical denitrification (Pous et al., 2015a), resulting in varying cell potential (1.3 ± 0.4 V) and anode potential (+ 0.5 ± 0.3 V). Consequently, a stratification of redox potential was observed along the different CCs between + 0.7 ± 0.2 V (height 70 cm) and -0.3 ± 0.2 V (height 20 cm) (see supplementary files). Within this potential gradient, the higher ammonium removal rates observed in design C in comparison to design B can be explained by both current-driven ammonium oxidation (Shaw et al., 2020; Vilajeliu-Pons et al., 2018) as well as oxygen supply by electrochemical water splitting (> + 0.6 V). However, the letter electrochemical reaction is not sustainable when using graphite due to exfoliation (Lai et al., 2017).

In terms of total nitrogen removal (Figure 2B), a similar rate was observed for all reactor designs. Operation at the lowest HRT (0.3 d) yielded the highest but still similar low total nitrogen removal rates ($N\text{-}TN_{RR}$) (mean values < 20 $gN \cdot m^{-3} \cdot d^{-1}$). Polarization of graphite granules in case of reactor design C did not provide additional denitrification

activity. Due to the potential distribution, only a limited zone was at the desired potential for denitrification (– 0.3 V) (Pous et al., 2015a). Nevertheless, a poor potential distribution could not have been the sole reason for the low denitrifying activity. The effluent nitrate concentrations were low (5 - 6 mg $N\text{-}NO_3^- \cdot L^{-1}$), suggesting that substrate was scarcely available for denitrifiers and identifying nitrification as the limiting step

3.1.2. Influence of dissolved oxygen in the influent

For analysing the effect of real-world aquaponics conditions on the performance of the developed reactor designs, reactors were fed with an influent not flushed with N_2 (aerobic). Reactor's influent in a real aquaponics application will have a DO concentration of around 4 – 8 $mgO_2 \cdot L^{-1}$, constituting a requirement for efficient fish respiration and growth (Wongkiew et al., 2017).

Although a general increase of the $N\text{-}NH_4^+{}_{RR}$ was expected for all reactor designs with this influent because of an increased oxygen availability for ammonium-oxidizing bacteria, this was only observed for high HRTs (> 0.8 d). In reactor design B (i.e. unpolarised graphite granule bed), the $N\text{-}NH_4^+{}_{RR}$ increased 187 % (34 ± 9 $gN \cdot m^{-3} \cdot d^{-1}$) at 1.0 d HRT, but decreased to only 21 % (34 ± 5 $gN \cdot m^{-3} \cdot d^{-1}$) at 0.2 d HRT. This improvement of $N\text{-}NH_4^+{}_{RR}$ for lower Q was higher in design B than in designs A and C either because of the better oxygen diffusion in PVC compared to graphite granules (in case of reactor design A) or because the presence of electricity-driven ammonium removal (in case of reactor design C) already brought these reactor designs close to their upper performance limits in terms of $N\text{-}NH_4^+{}_{RR}$. Still, the highest $N\text{-}NH_4^+{}_{RR}$ was exhibited by reactor design A, showing a maximum value of 68 ± 9 $gN \cdot m^{-3} \cdot d^{-1}$ at 0.4 d HRT. This value represented an increase of 21 % in respect to the maximum observed $N\text{-}NH_4^+{}_{RR}$ with a N_2-flushed influent (Fig. 2). In case of the N_2-flushed influent, the DO needed for nitrification was obtained only from air dissolving in the upper reactor

layers. Influent flow rate affects the oxygen mass transfer from gas to liquid phase, and thus, at high Q (low HRT), the oxygen mass transfer increased, and aerobic nitrification improved. However, when oxygen was already available in the influent, the increase of nitrification activity related to additional oxygen was negligible, as DO was already saturated in the upper reactor layers.

As nitrification rates ought to increase, more nitrate was available for bioelectrochemical denitrification. As a consequence, a sharp increase of N-TN_{RR} performance could be observed at high HRTs, but few differences at low HRTs (Fig. 3).

This initial testing phase revealed that designs A, B, and C lacked a sufficient oxygen distribution in the reactor for nitrification. Clearly, PVC was the filling material that provided better ammonium removal performances (Figures 2 and 3). The characteristics of PVC compared to granular graphite (lower porosity, larger size of granules) probably allowed a better oxygen penetration and distribution inside the reactor. In terms of total nitrogen removal using PVC provided some, but limited denitrification activity. This limitation is due to the fact that no electron donor is present, therefore this process is enhanced and better controlled by polarizing the granular graphite bed and providing cathodic electrons (i.e. reactor design C). Granule polarization was required for total nitrogen removal, as the usage of non-polarized graphite granules (i.e. reactor design B) did not improve the results obtained with PVC (Figures 2 and 3). Nevertheless, in addition to the poor oxygen distribution, reactor design C was probably also affected by a poor potential distribution within the graphite granule bed. The redox potential control of the whole cathode zone at the desired potential was not achieved in the present reactor architecture and thus bioelectrochemical denitrification was limited as the cathode could not deliver sufficient electrons at a sufficient redox potential. Therefore,

the results suggested that a proper coupling of the different materials and conditions in a single reactor design could enhance the reactor performance even further.

3.2. Performance enhancement

A fourth reactor design was developed according to the knowledge obtained during the testing phase of the first reactor designs (see section 3.1). For this reactor configuration (i.e. reactor design D in Fig. 1), an aerobic zone filled with PVC granules for promoting aerobic nitrification was coupled with an anoxic zone in the lower half of the reactors filled with a polarized granular graphite bed. Furthermore, a conductive stainless steel mesh was incorporated as CC in the granular graphite bed for setting a homogeneous potential distribution, and thus for improving and better controlling of denitrification rates (section 2.3).

As Figure 4 shows, the coupling of a polarized graphite granule bed with a PVC granule bed resulted in increased nitrification and denitrification rates. Initial testing was performed with the water level at 50 %, thus leaving the PVC granule bed fully exposed to air, while the graphite granule bed was completely covered with medium. At the initial HRT (1.2 ± 0.0 $L \cdot d^{-1}$, i.e. same initial flow rate than the other reactor designs but different HRT due to the different net volume), an $N\text{-}NH_4^+{}_{RR}$ of 39 ± 3 $gN \cdot m^{-3} \cdot d^{-1}$ was achieved corresponding to 89 ± 9 % $N\text{-}NH_4^+$ removal and representing a higher removal rate than the previous reactor designs (Figures 2 and 3). However, the observed $N\text{-}TN_{RR}$ was still similar to reactor designs A, B, and C (11 ± 8 $gN \cdot m^{-3} \cdot d^{-1}$). The performance of reactor design D was further enhanced by lowering the HRT. The ammonium and total nitrogen removal rates increased to 94 ± 44 $gN \cdot m^{-3} \cdot d^{-1}$ (72 ± 29 % $N\text{-}NH_4^+$ removal, Fig. 4A) and 39 ± 6 $gN \cdot m^{-3} \cdot d^{-1}$ (39 ± 13 % N-TN removal, Fig. 4B), respectively, when a HRT of 0.3 d was applied. These values represented an increase of

39 % and 13 % of the $N\text{-}NH_4^+{}_{RR}$ and $N\text{-}TN_{RR}$, respectively, compared to the maximum values achieved with the first reactor designs (Figures 2 and 3).

It is of note that the cathode potential was high (+ 0.1 ± 0.2 V ; see supplementary files) even though higher cell potentials were applied to the reactors (3.3 ± 1.3 V) suggesting that a high concentration of DO occurred due to the low water level (WL, 50 %) limiting nitrate removal. Once the WL was increased to 75%, the oxygen diffusion to the denitrifying zone was hindered leading to a depletion of oxygen already in the upper parts of the water column (i.e., PVC granule bed and anode zone of granular bed).

First, the increase of the WL to 75 % also resulted in an improved cathode potential of – 0.2 ± 0.1 V (by applying a cell potential of 3.8 ± 0.5 V) indicating suitable conditions for bioelectrochemical denitrification (Pous et al., 2015a). Furthermore, a WL of 75 % ought to decrease the aerobic zone further suggesting a negative effect on ammonium removal performed by ammonium-oxidizing bacteria. However, this was not the case for high HRTs. The ammonium removal varied only slightly compared to a WL of 50 % and reached removal rates of 66 ± 10 and 38 ± 2 $gN \cdot m^{-3} \cdot d^{-1}$ (88 ± 7 and 99 ± 1 % $N\text{-}NH_4^+$ removal) for HRT of 0.7 and 1.2 d, respectively (Figure 4). However, the $N\text{-}NH_4^+{}_{RR}$ stabilized to 70 ± 17 $gN \cdot m^{-3} \cdot d^{-1}$ at HRT of 0.3 d, corresponding to 51 ± 15 % $N\text{-}NH_4^+$ removal.

The maximum $N\text{-}TN_{RR}$ obtained in reactor design D operated at WL 75 % was 43 ± 2 $gN \cdot m^{-3} \cdot d^{-1}$ (HRT of 0.7 d), representing the highest removal rates being observed during the experiments. Hence, the increase of the WL from 50% to 75% also improved the reactor performance in terms of total nitrogen removal. Total nitrogen removal performance was particularly enhanced at 1.2 d, HRT where a change of WL from 50 % of 75 % increased the total nitrogen removal from 25 ± 18 % to 78 ± 14 %.

The effect of the polarization of the graphite granule bed on nitrogen removal was tested by operating the system under open circuit potential (OCP) while the water level and the HRT were 75 % and 1.2 d, respectively (Figure 5). By switching the reactors to OCP, a collapse on denitrification performance was observed. $N\text{-}TN_{RR}$ declined from 31 ± 6 to 8 ± 6 $gN \cdot m^{-3} \cdot d^{-1}$, indicating that denitrification in the reactors was mainly based on activity of electroactive bacteria. The application of OCP conditions not only affected the $N\text{-}TN_{RR}$ but also the $N\text{-}NH_4^+{}_{RR}$, which decreased from 39 ± 8 to 23 ± 12 $gN \cdot m^{-3} \cdot d^{-1}$. The decrease of $N\text{-}NH_4^+{}_{RR}$ by 25% indicates that aerobic nitrification in the PVC bed was the dominating but not the sole process for ammonium removal. Apparently, the electricity-linked ammonium removal had a certain relevance in the reactor design D (Shaw et al., 2020; Vilajeliu-Pons et al., 2018). This was further supported by recovery of both $N\text{-}NH_4^+{}_{RR}$ and $N\text{-}TN_{RR}$ to 36 ± 2 and 31 ± 4 $gN \cdot m^{-3} \cdot d^{-1}$, respectively, when reactors were polarized again (Figure 5).

3.3. Moving towards a sustainable aquaponics treatment: Effluent qualities and energetic requirements of the developed reactor designs

In this section, the performances of the developed reactor designs are discussed regarding their capabilities to treat a reasonable ammonium-rich influent (i.e., 50 $mgN\text{-}NH_4^+ \cdot L^{-1}$ (Yin et al., 2018)) and to achieve nitrogen concentrations that FAO considers as ideal for aquaponics loop: < 0.8 $mgN\text{-}NH_4^+ \cdot L^{-1}$, < 0.3 $mgN\text{-}NO_2^- \cdot L^{-1}$ and 1 - 34 $mgN\text{-}NO_3^- \cdot L^{-1}$ (FAO, 2014). If these conditions are reached, aquaculture effluent could be used as hydroponics influent, closing the aquaponics loop. Table 1 summarizes the highest effluent qualities obtained with the different reactor configurations.

Notably, the best effluent qualities for the different reactor designs were obtained with a similar HRT (around 1.0 d), representing the highest tested. All reactor designs achieved the required nitrate concentrations being sufficient for cultivating plants in

hydroponics, allowing the establishment of the aquaponics loop. Thus, the difference between the reactor designs is mainly based on their capacity to degrade the most harmful compounds for fish and plants growth (i.e. ammonium and nitrite). Considering all different experimental conditions and reactor designs, only reactor design D (WL 75 %) provided a sufficient effluent quality to be used in aquaponics applications (0.5 ± 0.4 mgN-$NH_4^+ \cdot L^{-1}$ and 0.2 ± 0.2 mgN-$NO_2^- \cdot L^{-1}$). With this configuration, a proper nitrification-denitrification process without intermediates accumulation was obtained (N_2O was rarely detected in the effluent when testing the different HRTs) and no main changes on pH was observed between influent (7.5 ± 0.3 pH) and effluent (7.4 ± 0.2 pH).

Nevertheless, for a real-world application, it is also needed to take into account the fish feed, the dynamic fish output rates, dynamic plant uptake rates, and the actual flow regime to understand how the here described system can be further adapted to different cultivated plants and fishes (Buzby and Lin, 2014; Endut et al., 2010; Hu et al., 2015). Therefore, a discussion of obtained results should also consider the maximum N-$NH_4^+{}_{RR}$ and N-TN_{RR} because they normalize the reactor activity according to its reactor volume and HRT of operation. In this sense, the highest performances were also provided by reactor design D (94 $gN \cdot m^{-3} \cdot d^{-1}$ and 43 $gN \cdot m^{-3} \cdot d^{-1}$, respectively).

The $NH_4^+{}_{RR}$ values obtained here are in the range of conventional biotrickiling filters reported for the treatment of aquaculture in literature (about 90 $gN \cdot m^{-3} \cdot d^{-1}$ – Losordo et al., 1999; Tyson et al., 2008) while being lower than those expressed for a commercial reactor (e.g. MAT-RAS company biofilters removes NH_4^+ at around 500 $gN \cdot m^{-3} \cdot d^{-1}$ (MAT-RAS, 2020)). However, it should be noted that the aerobic zone in reactor design D (i.e., main nitrification zone) is comparable small as it occupies only 25 - 50 % of the total reactor volume. Further potential improvements of the reactor design D for

increasing nitrification rate include optimization of the used filling material (e.g. higher surface area for bacterial growth, good drainage properties), increasing air flow distribution (e.g., by incorporating venting tubes), and obtaining a better knowledge on electricity-linked ammonium removal (Shaw et al., 2020; Vilajeliu-Pons et al., 2018). Remarkably, the electrified biotrickling filter provides not only ammonium but also nitrate removal without the need for adding chemicals, being current standard procedure in aquaculture recirculating systems (van Rijn et al., 2006), but by polarizing the graphite granule bed. The usage of autotrophic denitrifiers also decreases the risk filter blocking by heterotrophic denitrifiers growth. Although the anaerobic requirements for bioelectrochemical denitrification decreased the DO in the effluent of the reactor (< 0.2 $mgO_2 \cdot L^{-1}$), DO values required for the aquaponics loop (FAO, 2014) could be easily recovered by integrating cascade systems between electrified biotrickling filter and hydroponics tank.

The introduced reactor configuration moves METs one step closer to its application for the removal of nitrogen in wastewaters deficient in organic matter (C/N < 3), as it has no need of mechanical aeration, membranes or tailor-made structures as well as representing an approach with a low complexity. For comparing the here obtained total nitrogen removal rates with MET-based denitrification processes, the bioelectrochemical denitrification rates should be normalized to the net cathode volume (NCC) (Clauwaert et al., 2007). Considering that denitrification in reactor design D occurs only in the cathode zone (280 ± 6 mL), the maximum observed $N\text{-}TN_{RR}$ (43 ± 2 $gN \cdot m^{-3} \cdot d^{-1}$ at 0.7 d HRT and 75 % WL) corresponds to 120 ± 6 $gN \cdot m^{-3}_{NCC} \cdot d^{-1}$ being comparable to values commonly found in literature (Marx Sander et al., 2018; Pous et al., 2015a; Virdis et al., 2009). However, the denitrification potential of reactor design D is underachieved, since reactors specifically optimized for bioelectrochemical

denitrification have achieved rates higher than 500 $gN \cdot m^{-3}_{NCC} \cdot d^{-1}$ (Clauwaert et al., 2009; Pous et al., 2017). Nevertheless, rates on total nitrogen removal obtained in the present study (43 ± 2 $gN \cdot m^{-3} \cdot d^{-1}$) are competitive when compared to already reported low-complex MET for the treatment of urban wastewater. Noteworthy, these wastewaters contained organic matter, while the system tested here was fully autotrophic. For instance, microbial electrochemical wetlands reached values below 15 $gN \cdot m^{-3} \cdot d^{-1}$ (Aguirre-Sierra et al., 2020; Xu et al., 2017). When a constructed wetland was coupled with a denitrifying MET, a maximum total nitrogen removal rate of 76 $gN \cdot m^{-3} \cdot d^{-1}$ was observed (He et al., 2016). It is worth noting that this process required two steps (Wetland + MET) and the influent wastewater was composed of a mixture of nitrate and ammonium (40 $mgN\text{-}NO_3^- \cdot L^{-1}$ and 20 $mgN\text{-}NH_4^+ \cdot L^{-1}$) with a C/N ratio of 0.75. Thereby denitrification was not limited by nitrification, and the C/N ratio could explain an equal contribution of heterotrophic and autotrophic denitrification. Yet, total nitrogen removal rates achieved here with reactor design D need to be increased, if they should reach the performance of conventional alternatives pursuing full nitrogen removal in organic-carbon deficient waters. For example, partial-nitritation anammox processes can provide removal rates of around 200 $gN \cdot m^{-3} \cdot day^{-1}$ when treating wastewater containing 50 $mgN\text{-}NH_4^+ \cdot L^{-1}$ (Chatterjee et al., 2016).

However, the low complexity of the presented approach should be considered when interpreting results. In order to improve the applicability of this technology into the aquaponics sector, the complexity and hence the capital expeditures of the reactors can be expected to be significantly lower than a conventional MET used for nitrogen removal (i.e. 2-chamber BES connected to a potentiostat (Virdis et al., 2009)). The electrified biotrickling has no need of a membrane, decreases the number of pumps (no recirculation is applied and influent water flows from the anode to the cathode by

gravity), uses cheap materials (i.e. PVC tubes as reactor body and granular graphite bed as electrode), and only needs a DC power supply instead of a potentiostat. Furthermore, in this work and based on aquaponics characteristics, a redox-stratified food web was established. The influent water contained only ammonium that was aerobically and anodically degraded in the upper parts of the reactors. The therefrom produced nitrate was bioelectrochemically treated in the lower reactor parts by cathodic electron supply. Thus denitrifying activity was dependent on nitrification performance. A poor ammonium oxidation rate implied low nitrate availability for denitrifiers, being its activity restricted by substrate limitation. By improving, e.g. flow conditions and potential distribution, the capability of redox-stratified food web is likely to be enhanced.

The electrification of the biotrickling filter implies extra costs related to the usage of a DC power supply, electrodes and few electrical components. However, as the cost of the conductive filling material is low (about 0.2 €·L^{-1}) and as the components required for electrification were kept deliberately simple compared to previous researches, the envisaged reactor system would be comparable cheap. Moreover, an electrified biotrickling filter allows the suppression of organic matter dosing for denitrification and chemical dosing for maintaining appropriate aquaponics conditions representing two main operational costs. Due to the lower biomass accumulation, the sludge management of the electroactive microbiome presumably also represents an economic advantage compared to conventional aquaponics systems (Brown et al., 2015; Delaide et al., 2019). Although the energy consumption for nitrogen removal is relatively high comparing MET literature, it should be considered that this work represents a proof-of-concept offering several opportunities for improvements (e.g., flow conditions, potential distribution, oxygen leakage). For instance, the electricity consumption related to the

DC power supply of reactor design D (between 2.7×10^{-1} and 8.3×10^{-2} kWh·gN^{-1}) was higher than those related to the usage of a potentiostat for bioelectrochemical anoxic ammonium removal (1.16×10^{-3} kWh·gN^{-1} (Vilajeliu-Pons et al., 2018)) or denitrification (1.3×10^{-2} kWh·gN^{-1} (Pous et al., 2015a)) but similar comparing literature also using conventional power supply for bioelectrochemical nitrate removal (7.0×10^{-2} kWh·gN^{-1} (Sakakibara and Nakayama, 2001)). This electricity cost would not be the total operational cost related to the system, since pumps would probably be the most important contributor to electricity consumption. Actually, pumps are already present in aquaponics, and they could be used as well to feed the system. Thus, the savings provided by the electrified biotrickling filter in terms of less sludge production and less chemical demand (pH adjustment and organic matter) could overcome the costs related to the power source.

Finally, the recent Covid-19 lockdown allowed observing that reactor design D also inheres a certain resilience and robustness (see supplementary files). The results presented in this work were obtained before the lockdown and during this period, the volumetric flow rate was decreased to 0.3 L·d^{-1} (4.6 d HRT) for 2 months. After the lockdown, HRTs of 1.2, 0.7, and 0.4 days were tested again (2 weeks each) and similar maximum N-$NH_4^+{}_{RR}$ and N-TN_{RR} rates were observed (97 ± 18 gN·m^{-3}·d^{-1} and 55 ± 15 gN·m^{-3}·d^{-1}, respectively).

4. CONCLUSIONS

Sustainable electrification of biotrickling filters was achieved by combining an aerobic zone (filled with a non-conductive material) with an anoxic electrified zone (filled with a conductive material). Relevant ammonium and nitrate removal rates were obtained (94 gN·m^{-3}·d^{-1} and 43 gN·m^{-3}·d^{-1}, respectively) and the effluent quality criteria for an aquaponics application was reached. The reactor design developed in this study is a

promising alternative for aquaponics but also for the treatment of organic carbon-deficient ammonium-contaminated waters.

E-SUPPLEMENTARY MATERIAL

E-supplementary material data of this work can be found in online version of the paper.

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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:

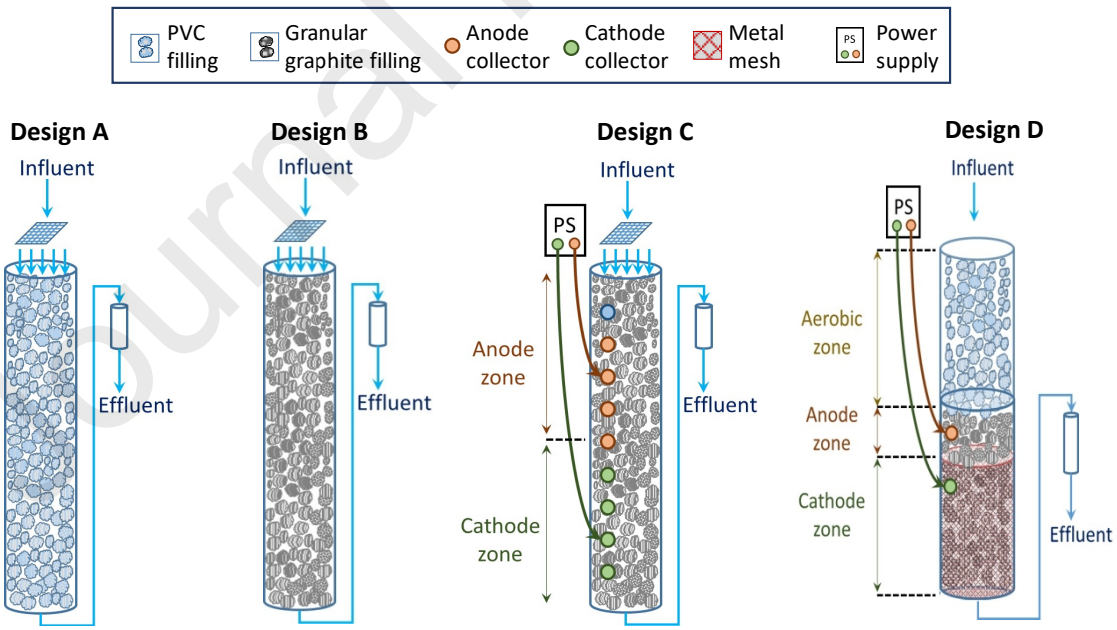

Figure 1. Reactor designs used in this study (details see 2.2.1. Reactors set-up and inoculation).

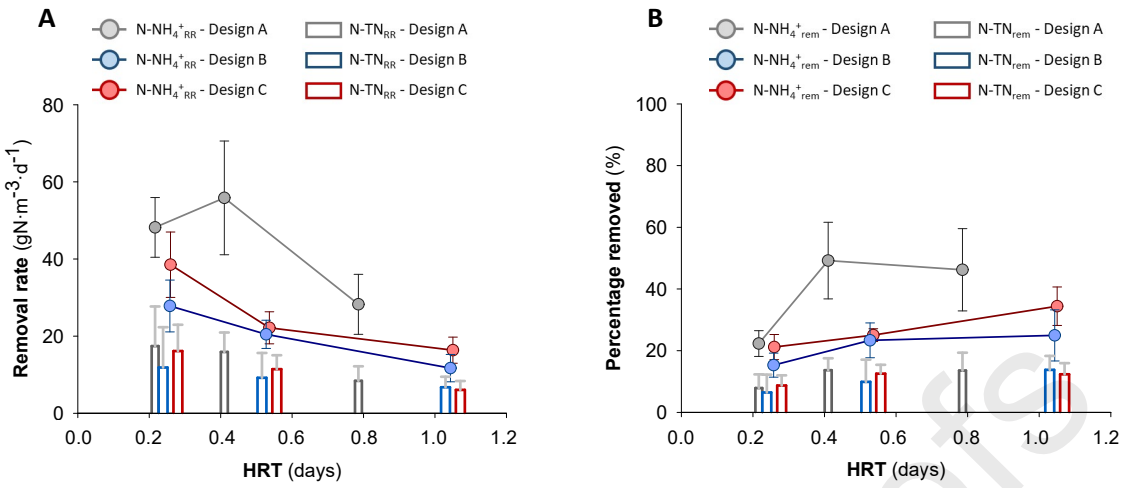

Figure 2. Removal rates of reactor designs A, B, and C operated at different HRTs treating an influent flushed with N_2. A) Evolution of N-NH_4^+ removal rates (Solid circles) and N-TN removal rates (Bar charts). B) Evolution of percentages of N-NH_4^+ removal (Solid circles) and percentages of N-TN removal (Bar charts). Error bars indicate standard deviation.

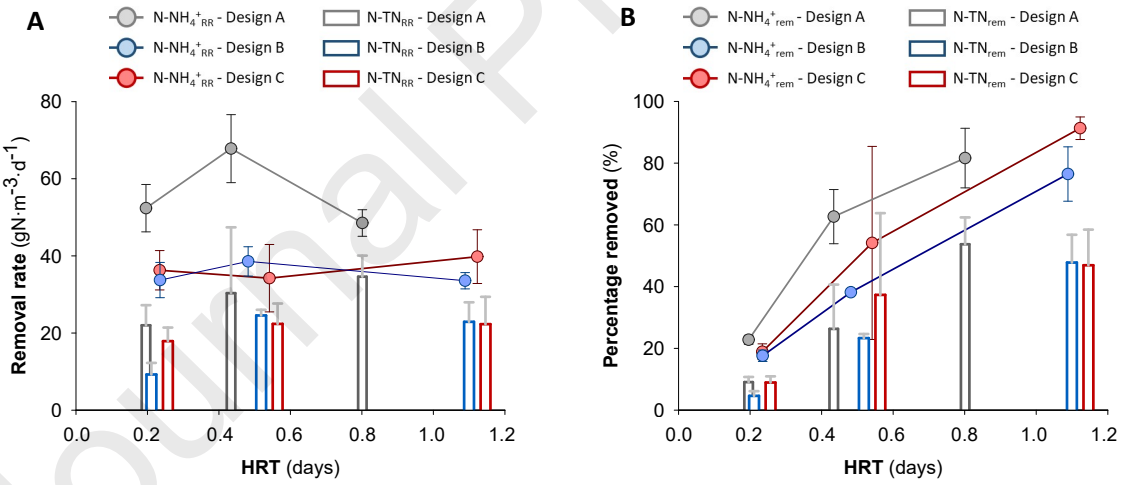

Figure 3. Removal rates of reactor designs A, B, and C at different HRTs treating an influent not flushed with N_2. A) Evolution of N-NH_4^+ removal rates (Solid circles) and N-TN removal rates (Bar charts). B) Evolution of percentages of N-NH_4^+ removal (Solid circles) and percentages of N-TN removal (Bar charts). Error bars indicate standard deviation.

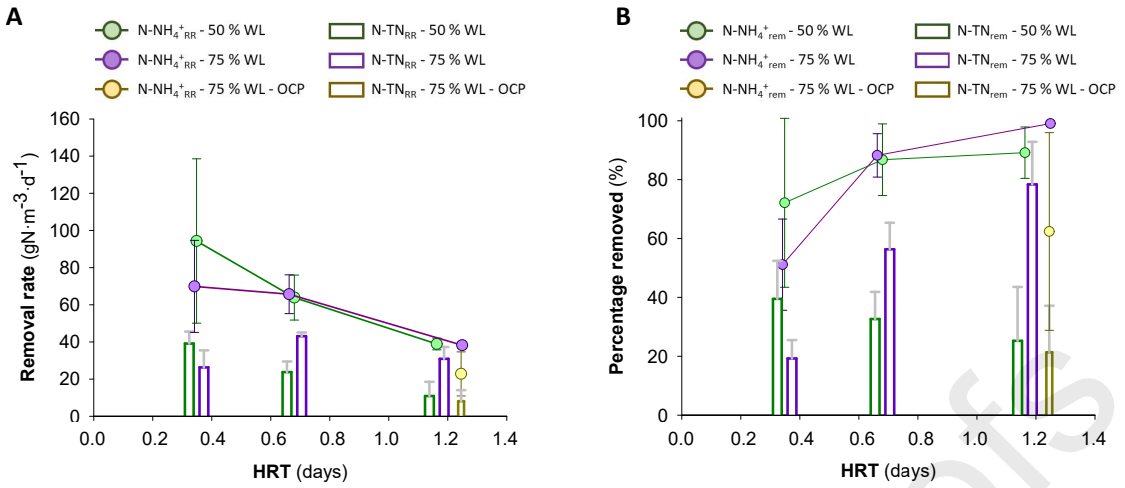

Figure 4. Removal rates of reactor design D at different HRTs treating an N_2-flushed influent at different water levels (WL) and without polarization of graphite granules (open circuit potential, OCP). A) Evolution of N-NH_4^+ removal rates (Solid circles) and N-TN removal rates (Bar charts). B) Evolution of percentages of N-NH_4^+ removal (Solid circles) and percentages of N-TN removal (Bar charts). Error bars indicate standard deviation.

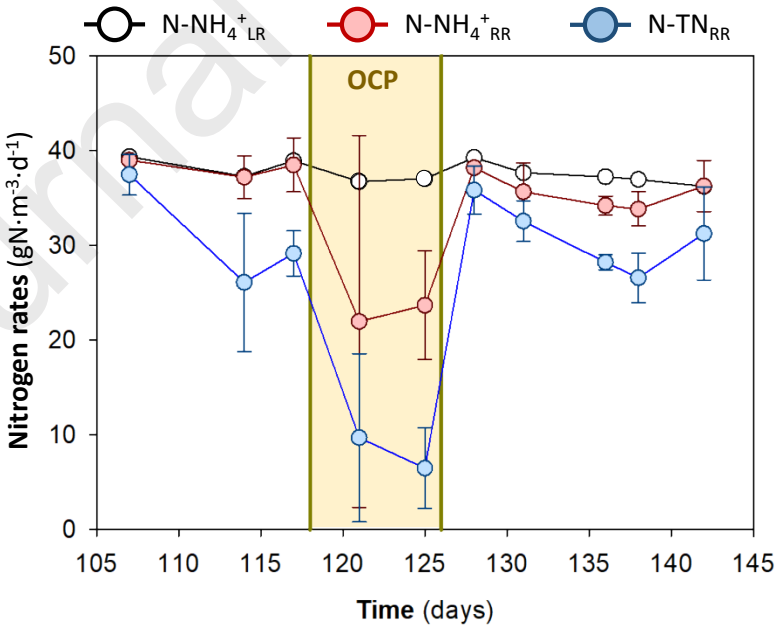

Figure 5. Removal rates of reactor design D during open circuit potential (OCP) conditions. Evolution of N-NH_4^+ loading rate (N-$NH_4^+{}_{LR}$), NH_4^+ removal rate (N-$NH_4^+{}_{RR}$), and N-TN removal rate (N-TN_{RR}).

Table 1. Best effluent conditions reached with the different reactor designs.

Reactor design	Condition	HRT (days)	N-NH_4^+ effluent (mgN-NH_4^+·L^{-1})	N-NO_2^- effluent (mgN-NO_2^-·L^{-1})	N-NO_3^- effluent (mgN-NO_3^-·L^{-1})	Electricity consumption (kWh·gN_{rem}^{-1})
A	N_2-flushed influent	1.0	26.1 ± 7.2	2.4 ± 1.2	13.5 ± 4.9	-
	Aerobic influent	1.0	8.9 ± 4.9	3.0 ± 1.7	12.1 ± 5.1	-
B	N_2-flushed influent	1.0	37.0 ± 5.1	0.4 ± 0.2	5.4 ±3.7	-
	Aerobic influent	1.0	11.3 ± 4.4	0.5 ± 0.2	15.1 ± 4.0	-
C	N_2-flushed influent	1.0	32.4 ± 4.3	1.0 ± 1.0	10.1 ± 2.7	$1.1x10^{-1}$ ± $1.1x10^{-1}$
	Aerobic influent	1.0	4.2 ± 1.8	0.6 ± 0.5	22.6 ± 5.5	$2.8x10^{-2}$ ± $2.7x10^{-2}$
D	WL 50 % N_2-flushed influent	1.0	5.7 ± 4.7	1.0 ± 1.1	31.5 ± 8.5	$9.3x10^{-2}$ ± $10.2x10^{-2}$
	WL 75 % N_2-flushed influent	1.0	0.5 ± 0.4	0.2 ± 0.2	9.8 ± 6.8	$8.3x10^{-2}$ ± $4.6x10^{-2}$

Highlights

- Non-conductive filling material brought insufficient and uncontrollable N removal.
- Polarized conductive filling material provided limited but controllable N removal.
- Mix of non-conductive and polarized conductive material were the most active.
- The electrified biotrickling filter met the N standards, closing aquaponics loop.