Int. J. Environ. Sci. Technol. (2012) 9:395–408
DOI 10.1007/s13762-012-0073-3

ORIGINAL PAPER

Effect of aeration on steady-state conditions in non- and partially aerated low-loaded biofilter

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Received: 12 April 2011 / Revised: 26 October 2011 / Accepted: 8 December 2011 / Published online: 12 April 2012

Abstract Excessive growth of biomass and retention of solids associated with air bubbles lead to bed clogging, which affects the biofilters' performance. Two experiments were carried out in a submerged biofilter at the flow velocity of 0.5 m h^{-1}, for an organic loading rate of 51 g C m^{-3} h^{-1} and a nitrogen loading rate of 13 g NH_4-N m^{-3} h^{-1}, one with the biofilter not aerated, the other with the biofilter partially aerated. The results showed that the higher head losses occurred in the upper section of the biofilter, where there was a greater biomass development and a higher removal of organic carbon, ammonia and solids, with the maximum allowed head loss being reached in 16 and 8 days. In any case, the steady-state conditions were achieved after 2 days and were interrupted on the tenth day of experiment E1 and on the fifth day of experiment E2. This allowed defining different operating cycles that enabled an average organic removal rate of 12.7 g C m^{-3} h^{-1} (27 %) and an average ammonia removal rate of 1.1 g NH4-N m^{-3} h^{-1} (9 %) without aeration, and of 35.8 g C m^{-3} h^{-1} (76 %) and 6.3 g NH4-N m^{-3} h^{-1} (51 %) with aeration. Regardless of the aeration conditions, more than 90 % of TOC and NH4-N removal occurred in the upper section. After the backwashing cycle, the biofilter returned to steady-state conditions in 6 h (without aeration) and 7 h (with aeration).

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Keywords Aerated filter · Backwashing · Head losses · Nitrogen removal · Organic carbon removal

Introduction

Submerged biofilters have been used for on-site treatment (Coetzee et al. 2011), secondary treatment of wastewater (domestic, industrial and stormwater effluents) (Garzon-Zuniga et al. 2005; Osorio et al. 2006; Farabegoli et al. 2009; Liu et al. 2010; Rajakumar et al. 2011) or as a tertiary step to upgrade other treatment systems (Hidaka and Tsuno 2004; Liu et al. 2008; Ha et al. 2010b) and can be a promising alternative to retrofitting (Schulz and Menningmann 2008; Farabegoli et al. 2009). The construction of biofilters as a tertiary/polishing step does not interfere with the operation of existing reactors and its maintenance is considered to be simple, as there is no need for sludge recycling and a final clarifier (Hidaka and Tsuno 2004).

The interest of this bioreactor is the maintenance of high concentration of active biomass and the good control of its excess, need for lower volumes for reaction, hydraulic retention time and area for construction and similar cell residence times compared with completely mixed reactors, good efficiency of pollutant removal (organic carbon, nitrogen, phosphorous, heavy metals and pathogen) and high filtering capacity (Mendoza-Espinosa and Stephenson 1999; Grady Jr et al. 1999; Osorio et al. 2006; Hidaka and Tsuno 2004; Schulz and Menningmann 2008). These benefits are associated with the use of bed media with high specific surface area (500–2,000 m^2 m^{-3}) to allow a good biofilm development, and a particle diameter ranging from 1 to 4 mm to obtain a proper void ratio (over 0.4) for a suitable hydraulic flow rate and to delay clogging (Mendoza-Espinosa and Stephenson 1999; Schulz and Menningmann

2008; Farabegoli et al. 2009). In order to minimize the negative effects of clogging, the filter must be washed from time to time. However, the type of wash (water, air/water), its duration and velocity must be carefully chosen in order to avoid biomass loss and to allow a quick recovery of the steady-state conditions in the filter. The washing energy requirements constitute an important percentage of the total cost of treatment plants (Robinson et al. 1994).

The costs associated with aeration can be reduced by optimizing washing cycles (i.e., the washing procedure is only used when the head losses increase to a level that can interrupt the steady-state conditions and the treatment performance) and using low air-flow rate or intermittent aeration (Grady Jr et al. 1999; Tchobanoglous et al. 2002). This last procedure may also be useful to change the biochemical environment inside the biofilter from aerobic to anoxic or anaerobic, in order to promote nitrification/denitrification mechanisms, either simultaneously or alternatively.

The steady-state conditions are normally achieved when the removal of organic matter [such as total organic carbon (TOC) or chemical oxygen demand (COD)], ammonia nitrogen (NH_4-N) or other compounds remains approximately constant in each point along the biofilters over time (Tchobanoglous et al. 2002; Albuquerque et al. 2011). These conditions can be achieved in 1–3 weeks (Grady Jr et al. 1999) and may deteriorate when the biofilm grows excessively, since the solid material and excess of biomass accumulate in the void spaces and fine material is released from the media, leading to the development of stagnated and dead volume areas that clog the filter. One way to follow the evolution of clogging in biofilters is by measuring head losses in different sections of the media by measuring the difference between piezometric heads (Gonçalves and Oliveira 1996; Ozis et al. 2007). As the solids are retained by filtration and the biomass increases, the flow loses energy, which will raise the hydrostatic pressure inside the filter and, therefore, will increase the level of a piezometer.

The effect of aeration on the maintenance of the steady-state conditions in tertiary/polishing partially aerated biofilters was not well studied and needs to be optimized. According to Soewondo and Yulianto (2008), intermittent aeration in biofilters could reduce the use of energy when the aeration system is turned down, especially at night. The same energy savings can be achieved with partially aerated biofilters, since the air-flow rate is reduced in order to maintain a portion of the filter in an aerobic condition and another portion anaerobic. Ha and Ong (2007) have used a partially aerated biofilter with recirculation for the secondary treatment of a feedwater with 250 mg COD L^{-1} and 35 mg N L^{-1} and an hydraulic retention time (HRT) of 4 h. Later on, these authors evaluated the effects of temperature on nitrification in a polishing biological aerated filter (Ha et al. 2010a).

Therefore, this work aims to identify the influence of aeration conditions on the increase of head losses and the interruption of steady-state conditions in a partially aerated biofilter for the removal of residual loads of carbon (approximately 40 mg C L^{-1}) and nitrogen (approximately 10 mg NH_4-N L^{-1}), as well as to define the optimal washing cycle in order to maintain a good treatment performance. Most of the experiments developed with aerated biofilters used full aeration and the reactors were operated as secondary treatment, with air/water ratios between 4 and 10 to remove higher concentrations of carbon and ammonia, usually from 100 to 2,000 mg C L^{-1} and from 25 to 2,000 mg N L^{-1}, respectively (Mendoza-Espinosa and Stephenson 1999; Villaverde et al. 2000; Osorio and Hontoria 2001; Stephenson et al. 2003; Garzon-Zuniga et al. 2005; Lei et al. 2009; Ha et al. 2010b; He et al. 2007; Coetzee et al. 2011; Rajakumar et al. 2011).

This research work was carried out at the Laboratory of Environmental Sanitation of the Department of Civil Engineering and Architecture of the University of Beira Interior (Portugal), between September 2010 and May of 2011, with the technical support of the Department of Civil Engineering of the University of Granada (Spain).

Materials and methods

Experimental setup

A bench-scale cylindrical vertical biofilter was used for this study, made in acrylic glass with 7.0 cm × 40.5 cm size [internal diameter × packing height (he)] and filled with natural porous volcanic rock material (puzzolane) with an effective diameter of 4 mm, specific surface area of 1,740 m^2 m^{-3} and void ratio of 0.52 [a similar device and the same bed media were used in the works of Albuquerque et al. (2009a) and (2011) to study of nitrogen removal, in a downward flow configuration (Fig. 1)].

According to Buitrón et al. (2004), puzzolane presents suitable properties for application in bioreactors and has already been tested in a sequencing batch biofilter for the removal of azo dye. Villaverde et al. (2000) have also tested this material in a fully aerated biofilter operating at an air/water ratio of 4, but for the removal of ammonia concentrations above 100 mg N L^{-1}.

The media bed was submerged 3 cm below the water level (h_{WL}) and 5 sampling ports (P1–P5, 5 mm in diameter) were provided along the height of the reactor to collect water samples for analytical measurements. Another five ports (in the opposite side of the bioreactor) were connected to five piezometers to evaluate variations in the hydrostatic pressure between five sections (TM–P1, P1–P2, P2–P3, P3–P4 and P4–P5).

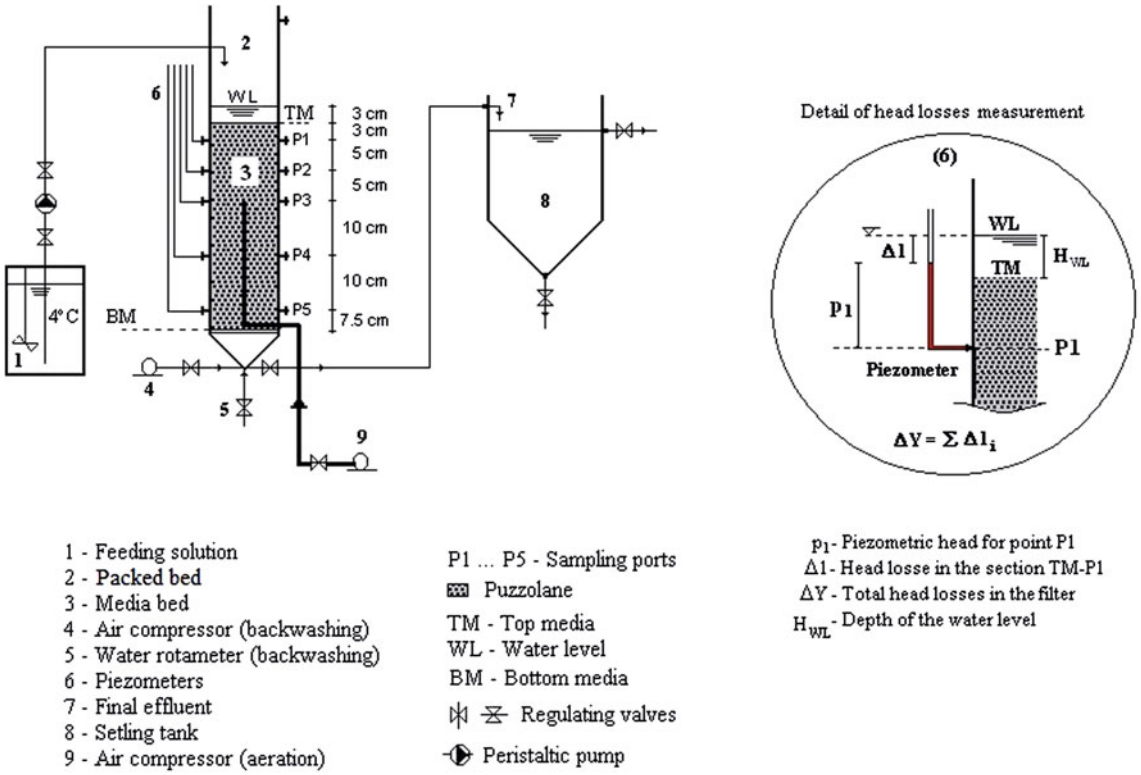

Fig. 1 Schematic representation of the biofilter

An external aeration device (TetraTec AP150 pump) with a maximum capacity of 150 L h^{-1} was connected in port P3 (i.e., 13 cm from the media top) in order to allow the aeration of the upper part. The air was injected into the reactor through a 4-mm tube with a fine bubble micro-diffuser. The aeration device introduced the air in an upward direction in order to keep dissolved oxygen (DO) concentrations above 2 mg O_2 L^{-1} in section TM-P3 (i.e., the reactor was partially aerated).

This is one of the innovations of this work, since most of the previous studies with BAF (Villaverde et al. 2000; Stephenson et al. 2003; Lei et al. 2009; Ha et al. 2010b; He et al. 2007) used fully aerated filters with the aeration device located at the bottom of the reactors. A backwashing system was included in order to remove the excess of both filtered material and biomass (sludge) produced during the biofilter operation. The backwashing water-flow rate was controlled by a rotameter (GARDENA T120, Italy), whilst the backwashing air-flow rate was controlled by a pressure pump (VACUUBRAND ME 4R, Germany).

Feeding solution

The feeding solution used for the experiments included a mineral medium (buffer, magnesium sulphate, calcium chloride and iron chloride solutions), prepared as proposed by Dang et al. (1989), an oligoelements solution (according to the standard ISO 9408 1999), a source of organic carbon (sodium acetate) and a source of ammonia nitrogen (ammonia chloride). The concentrated mineral medium had the following composition: buffer solution (8.50 g KH_2PO_4 + 21.75 g K_2HPO_4 + 33.40 g $Na_2HPO_4{\cdot}7H_2O$ + 1.70 g NH_4Cl per L), magnesium sulphate solution (22.50 g $MgSO_4{\cdot}7H_2O$ per L), calcium chloride solution (36.43 g $CaCl_2{\cdot}2H_2O$ per L), iron chloride solution (0.25 g $FeCl_3{\cdot}6H_2O$ per 1 L) and oligoelements solution (0.04 g $MnSO_4{\cdot}4H_2O$ + 0.06 g H_3BO_3 + 0.04 g $ZnSO_2{\cdot}7H_2O$ + 0.032 g $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$ + 0.10 g $C_{10}H_{12}FeN_2NaO_8{\cdot}3H_2O$ + 0.0555 g EDTA ($C_{10}H_{14}N_2Na_2O_8{\cdot}3H_2O$) + 0.0445 g $FeCl_3{\cdot}6H$ per L). The concentrated sodium acetate solution (113.4 g $C_2H_3O_2Na{\cdot}3H_2O$ L^{-1}) had a TOC of 20 g C L^{-1}. The concentrated ammonia chloride solution (76.41 g NH_4Cl L^{-1}) had a NH_4-N of 20 g N L^{-1}.

Operating conditions

The biofilter was first inoculated with biomass from an activated sludge system treating domestic wastewater. The bed's colonization took approximately 15 days in a closed circuit at the flow rate of 1 L h^{-1} and with discontinuous feeding. Approximately 38.9 and 9.7 mL of acetate and ammonia solutions, respectively, and a proportional

volume of the mineral solutions were added to the reactor every 24 h, in order to ensure concentrations of approximately 40 mg C L^{-1} and 10 mg NH_4-N L^{-1} ($C/N = 4$).

The biofilter was therefore continuously fed with the synthetic wastewater, which was prepared with tap water by diluting the concentrated solutions in the following proportions: 2 mL L^{-1} of buffer solution, 0.2 mL L^{-1} of magnesium sulphate solution, 0.2 mL L^{-1} of calcium chloride solution, 0.2 mL L^{-1} of iron chloride solution, 0.2 mL L^{-1} of trace element solution, 2 mL L^{-1} of sodium acetate solution, and 0.5 mL L^{-1} of ammonia chloride solution. The concentrations of TOC and NH_4-N were obtained by diluting the concentrated solutions of sodium acetate and ammonia chloride and were chosen in order to obtain influent values of 40 mg C L^{-1}, 10 mg NH_4-N L^{-1} and a C/N ratio of 4 (characteristic ratio of secondary effluents, which was also used in previous studies (Albuquerque et al. 2009a, b, 2011). The feeding solution was kept in a storage tank (ISCO FTD 220, Italy) at a constant temperature of approximately 4 °C and pumped to the biofilter through a peristaltic pump (ISMATEC MCP CA4, Switzerland). The experiments were carried out in an acclimated laboratory at the temperature 20 ± 1 °C.

The biofilter was first operated without aeration until achieving steady-state conditions (experiment E1) in terms of TOC removal (ΔC) in all the five ports (P1–P5). Then, the operation continued until reaching the maximum total head loss that was possible to measure in the biofilter ($\Delta Y = 50$ cm of water column), which corresponded to the complete clogging of the biofilter. The TOC, NH_4-N, nitrite nitrogen (NO_2-N), nitrate nitrogen (NO_3-N), total suspended solids (TSS), total volatile suspended solids (VSS), temperature, pH and DO were measured daily at the entrance and in each of the five ports. The water levels (piezometric heads, p1) in the five piezometers and the depth of the water table (h_{WL}) above the biofilter were measured daily in order to calculate the head losses (Δl) in the sections TM–P1, P1–P2, P2–P3, P3–P4 and P4–P5. The experiment was run at a hydraulic loading rate (HLR) of 0.5 m h^{-1} (flow rate of 1 L h^{-1}) and for the mean loads presented in Table 1 (fist column).

Recent studies showed good performance for ammonia removal at HLR up to 3 m h^{-1} and air-flow rates from 18 to 200 L h^{-1} (Taghizadeh et al. 2007; Schulz and Menningmann 2008; Lei et al. 2009; Ha et al. 2010b; He et al. 2007; Rajakumar et al. 2011) in fully aerated biofilters fed with higher ammonia concentrations (>100 mg NH_4-N L^{-1}). The HRT was approximately 50 min, which is in the range (20 min to 2 h) reported by Mendoza-Espinosa and Stephenson (1999) and Ha et al. (2010b) for downflow aerated biofilters treating real and synthetic wastewater. Mendoza-Espinosa and Stephenson (1999) point out organic and nitrogen removal efficiencies above 80 % in fully aerated biofilters running at HLR from 1 to 10 m h^{-1}. Coetzee et al. (2011) used an HLR of 0.001 m h^{-1} in small fully aerated biofilters to remove loads of 2 000 to 12000 mg N L^{-1} in pit latrines.

After the backwashing cycle has been defined, the recovery of the steady-state conditions after washing was assessed by observing the ΔC variation in the sampling points P1, P3 and P5 over time (hourly samples were taken at each port until steady-state conditions were observed again). In addition, before and after backwashing, four grains of the media were collected at ports P1 and P3 in order to evaluate biofilm thickness variations after washing.

Then, the aeration device was introduced at the level of port P3 (13 cm from the TM), the air-flow rate was regulated and the second experiment (E2) started, following the same procedure as defined for experiment E1. The aeration rate was kept at 4 L h^{-1}, which resulted in an air/water ratio of 8. According to Schulz and Menningmann (2008), Chang et al. (2008) and Ha et al. (2010b), continuous or intermittent air-flow rates between 3.5 and 18 L h^{-1} are considered enough for the simultaneous removal of organic carbon and nitrogen.

Backwashing was performed by simultaneously injecting air at a low rate of 5 L m^{-2} s^{-1} (≈68 L h^{-1}, ≈0.4 m^3 air m^{-3} media min^{-1}) and water at a flow rate of 1 L m^{-2} s^{-1} (≈14 L h^{-1}, ≈0.3 m^3 water m^{-3} media min^{-1}), in an upward direction during 10 min (air/water ratio of 5). These conditions were set based on the recommendations of Mendoza-Espinosa and Stephenson (1999), i.e., air-flow rates and water-flow rates in the ranges of 0.4–0.5 m^3 air m^{-3} media min^{-1} and 0.33–0.35 m^3 water m^{-3} media min^{-1}, respectively, and are within the range of flow rates observed in the studies of Yang et al. (2010) and Liu et al. (2010), 5.3–15 L m^{-2} s^{-1} (backwash air-flow rate) and 0.18–5 L m^{-2} s^{-1} (backwash water-flow rate).

Analytical methods

The measurements of DO, pH and temperature were carried out with two D201 flow through vessel using probes SenTix 41 and CellOx 325 connected to the Multi 340i meter (WTW, Germany). The TOC was measured using the TOC-5000 analyzer (Shimadzu, Japan). Concentrations of NH_4-N, NO_2-N and NO_3-N were obtained using the cuvette-tests LCK 303 (2–47 mg NH_4-N L^{-1}), LCK 342 (0.6–6 mg NO_2-N L^{-1}) and LCK 339 (0.23-13.5 mg NO_3-N L^{-1}), following the standards DIN 38406-E 5-1 (ammonia), DIN 38405 D10 (nitrite) and DIN 38405-9 (nitrate), and the CADAS 50 spectrophotometer UV–Vis (HACH LANGE, Germany). Total and volatile suspended solids (TSS and VSS) concentrations were determined

Table 1 Results for experiments E1 and E2

Parameters	Entrance[a]	P1 3 cm	P2 8 cm	P3 13 cm	P4 23 cm	P5 33 cm
Experiment E1: no aeration, 16 days						
pH	7.20 ± 0.12	7.33 ± 0.08	7.34 ± 0.05	7.35 ± 0.06	7.37 ± 0.08	7.38 ± 0.05
Temperature (°C)	20.15 ± 0.05	20.33 ± 0.06	20.51 ± 0.05	20.84 ± 0.03	20.78 ± 0.05	20.82 ± 0.03
OD (mg O_2 L^{-1})	7.73 ± 0.16	0.62 ± 0.11	0.18 ± 0.02	0.12 ± 0.01	0.09 ± 0.00	0.05 ± 0.00
TOC (mg C L^{-1})	41.21 ± 0.26	34.71 ± 0.61	34.49 ± 0.90	31.94 ± 0.87	31.64 ± 0.80	31.45 ± 0.81
NH_4-N (mg N L^{-1})	10.56 ± 0.17	9.96 ± 0.26	9.98 ± 0.25	10.04 ± 0.17	10.06 ± 0.22	10.08 ± 0.26
NO_2-N (mg N L^{-1})	–	<0.6	<0.6	<0.6	<0.6	<0.6
NO_3-N (mg N L^{-1})	–	0.33 ± 0.04	<0.23	<0.23	<0.23	<0.23
TSS (mg L^{-1})	–	50.2 ± 6.1	44.8 ± 6.2	41.6 ± 5.5	36.9 ± 5.5	32.7 ± 4.1
VSS (mg L^{-1})	–	42.3 ± 5.1	36.9 ± 4.9	33.8 ± 4.5	29.2 ± 4.1	25.5 ± 2.9
Experiment E2: with partial aeration, 8 days						
pH	7.21 ± 0.11	7.43 ± 0.12	7.51 ± 0.10	7.55 ± 0.11	7.58 ± 0.05	7.56 ± 0.07
Temperature (°C)	20.21 ± 0.04	20.52 ± 0.08	20.61 ± 0.05	20.77 ± 0.05	20.75 ± 0.05	20.72 ± 0.04
OD (mg O_2 L^{-1})	7.75 ± 0.20	2.57 ± 0.31	2.52 ± 0.28	2.78 ± 0.23	0.15 ± 0.03	0.08 ± 0.01
TOC (mg C L^{-1})	40.9 ± 0.40	18.8 ± 2.3	16.8 ± 2.5	15.8 ± 2.2	15.1 ± 2.4	14.6 ± 2.6
NH_4-N (mg N L^{-1})	10.51 ± 0.10	8.87 ± 0.41	7.95 ± 0.65	7.06 ± 0.95	7.04 ± 0.97	7.09 ± 0.91
NO_2-N (mg N L^{-1})	–	<0.6	<0.6	<0.6	<0.6	<0.6
NO_3-N (mg N L^{-1})	–	1.25 ± 0.17	1.95 ± 0.26	2.32 ± 0.41	<0.23	<0.23
TSS (mg L^{-1})	–	57.5 ± 5.6	51.9 ± 5.3	45.0 ± 6.3	40.3 ± 5.1	35.3 ± 4.2
VSS (mg L^{-1})	–	48.8 ± 4.4	43.8 ± 3.1	35.9 ± 3.8	32.5 ± 4.1	25.6 ± 1.8

Average and confidence interval [calculated for a confidence level of 95 % and the following number of samples: 16 (E1) and 8 (E2)]

[a] Measured in the entrance tube, before the water level

using 0.45 μm pore size filters through the gravimetric method (APHA-AWWA-WEF 1999). The biofilm thickness was evaluated through an electronic microscope (Hitachi S 2700, Japan) after the sample (media/biofilm) dehydration with acetone.

Results and discussion

Evolution of head losses

The head losses (Δl) in each section of the biofilter (TM–P1, P1–P2, P2–P3, P3–P4 and P4–P5) were calculated by measuring the difference between the piezometric heads of two consecutive piezometers, as illustrated in Fig. 1 for the initial section (TM–P1). The total head loss in the filter (ΔY) is the sum of the Δl measured in each section. The daily values were computed for each experiment (E1 and E2) until reaching the maximum head loss allowed in the biofilter ($\Delta Y = 50$ cm, which corresponds to the complete clogging of the biofilter) and are presented in Figs. 2 and 3.

In both experiments, the results show that the head losses occurred mainly in section TM–P2 (first 8 cm), but occurred more quickly in the experiment with aeration, reaching the maximum allowed head loss (50 cm) in half the time, as it can also be seen in Figs. 4 and 5.

Table 1 presents the results of the physicochemical analysis for each experiment over the experimental period.

The pH and temperature presented no significant variation throughout the biofilter and over time in experiment E1, but increased slightly in experiment E2. This increase is associated with the removal of acetate and ammonia, especially in section TM–P3, that according to Grady Jr et al. (1999) and Albuquerque et al. (2009a, b) generate alkalinity. DO concentrations show that oxygen was a limiting factor in the biofilter in experiment E1 (the average value in section TM–P3 was 0.31 mg O_2 L^{-1}). In experiment E2, the biofilter was aerobic in section TM–P3 (average DO of 2.62 mg O_2 L^{-1}) and anoxic in section P3–P5 (average DO of 0.12 mg O_2 L^{-1}).

In the experiment E1, the average organic loading rate (OLR) and the average nitrogen loading rate (NLR) were 50.9 g C m^{-3} h^{-1} and 13 g NH_4-N m^{-3} h^{-1}, respectively, whilst for experiment E2 were 50.5 g C m^{-3} h^{-1} and 13 g NH_4-N m^{-3} h^{-1}, respectively. These values were computed by multiplying the average TOC or NH_4-N concentrations given in Table 1 by the flow rate (0.001 m^{-3} h^{-3}) and then dividing it by the effective volume of the biofilter (0.00081 m^3). The availability of oxygen has allowed an improvement of TOC and NH_4-N removal in the aerated biofilter, in the later case with the production of some nitrate (ports P1–P3). The overall average removal efficiency of NH_4-N for the 8 days was 37.5 %, which corresponds to an average ammonia removal rate (r_{NH_4N}) of 4.9 g NH_4-N m^{-3} h^{-1}. Approximately 99 % of the ammonia was removed in the aerated section TM-P3 (approximately 6.7 g NH_4-N m^{-3} h^{-1}) where the average DO was 2.62 mg O_2 L^{-1}. The r_{NH_4N} was computed by multiplying the mean NH_4-N mass removal (ΔNH_4-N = influent concentration − effluent concentration given in Table 1) by the flow rate (0.001 m^{-3} h^{-1}) and then dividing it by the effective volume of the biofilter (0.00081 m^3).

Analogous conclusions were observed in Albuquerque et al. (2011) using a similar biofilter, but with both higher NLR and air/water ratio (20). However, the aeration device was located in port P2 (8 cm from the TM), which may have allowed more time for the oxygen to be transferred to

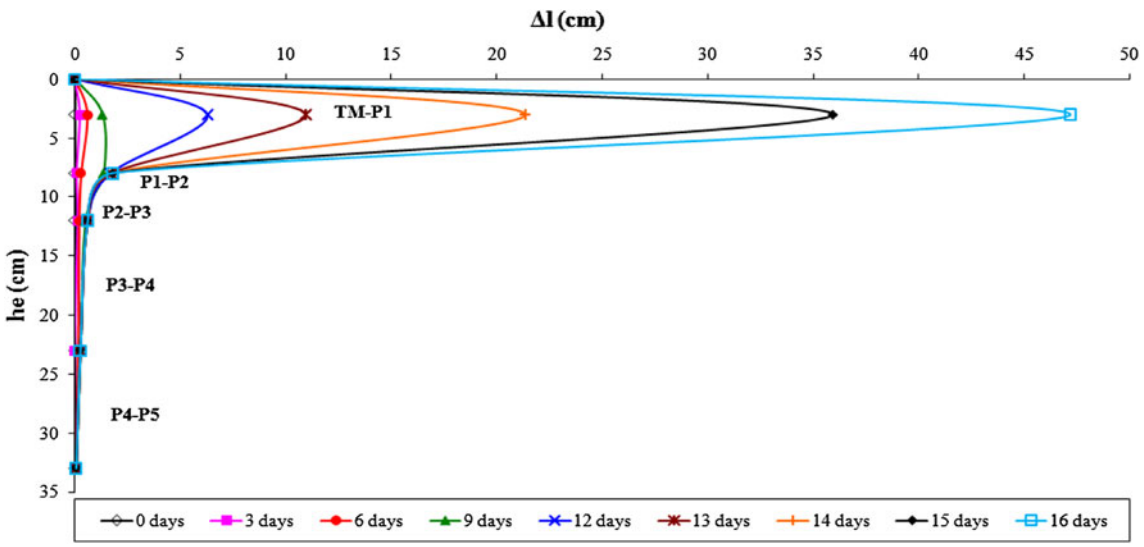

Fig. 2 Evolution of head losses in each section of the biofilter (E1)

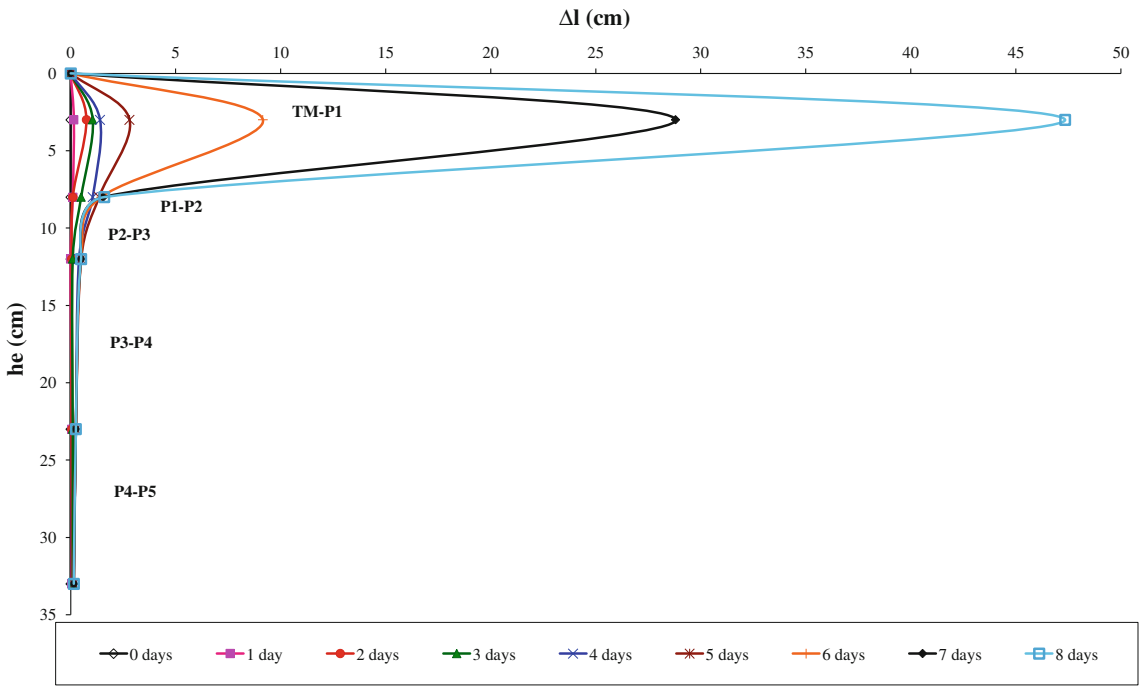

Fig. 3 Evolution of head losses in each section of the biofilter (E2)

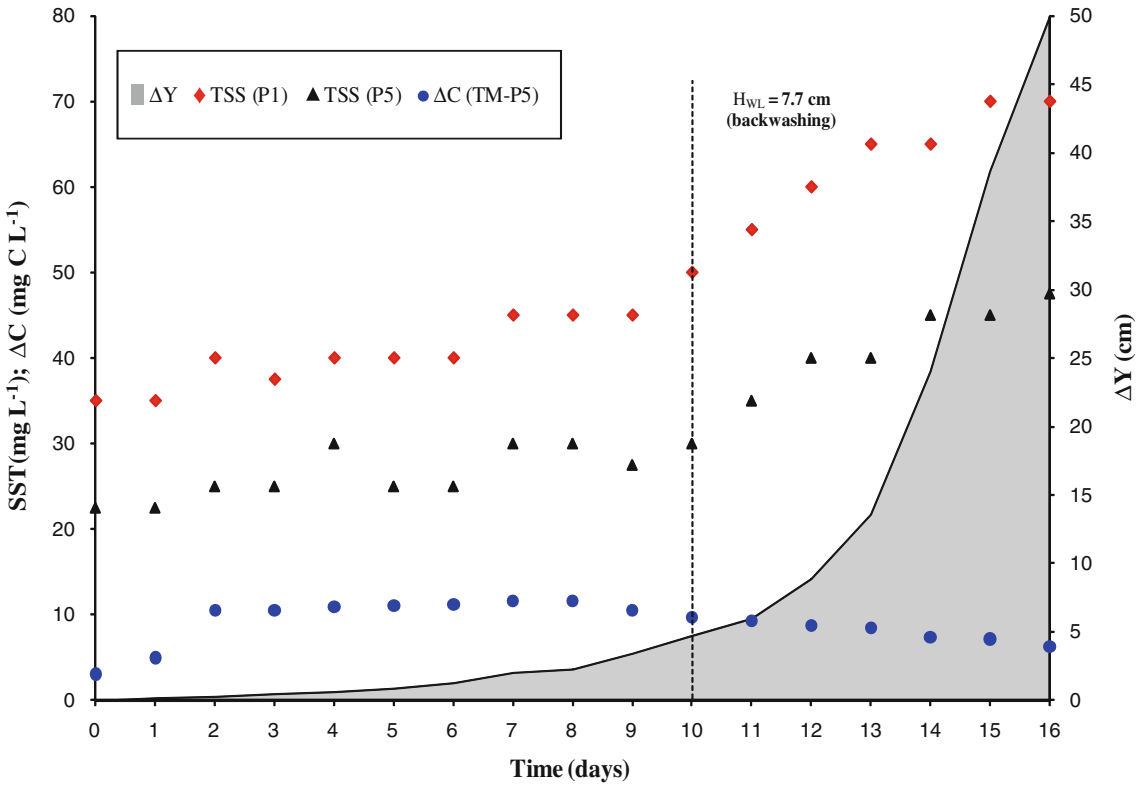

Fig. 4 Evolution of the total head losses, TSS and Δ*C* in the biofilter (E1)

the cells and less oxygen lost to the atmosphere and, therefore, the observation of a higher average DO concentration in the aerated sections (4.3 mg O_2 L^{-1}). The overall average removal of ammonia was 41.6 %, 90 % of which occurred in section TM–P2 where the average r_{NH_4N} was 6.8 g NH_4-N m^{-3} h^{-1}. Liu et al. (2009) observed a

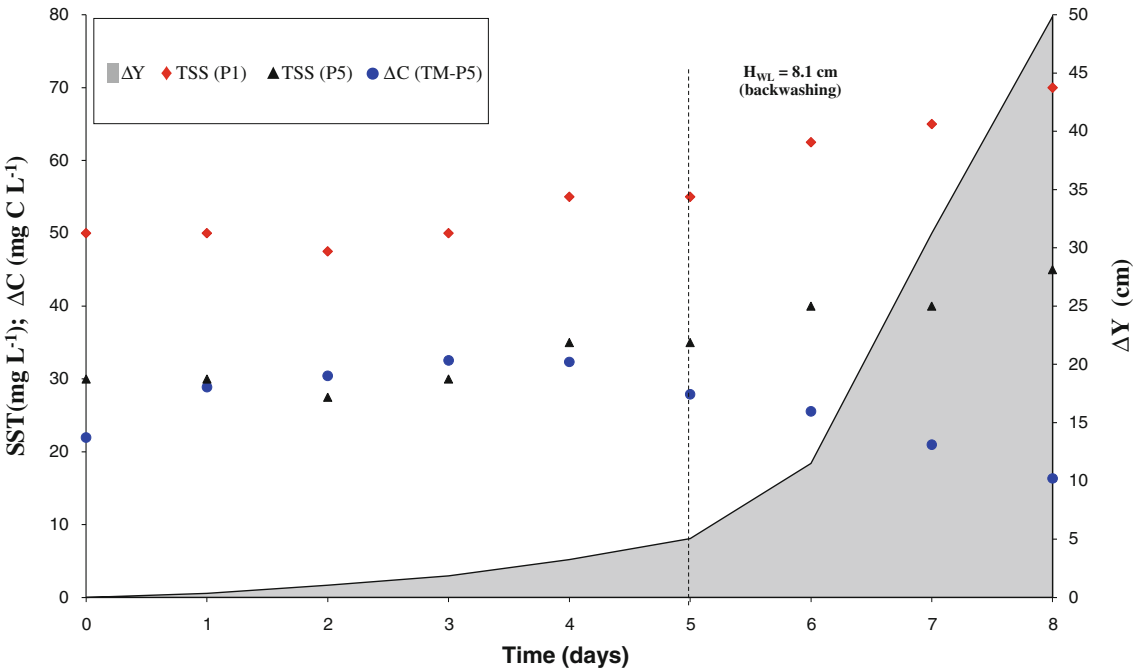

Fig. 5 Evolution of the total head losses, TSS and ΔC in the biofilter (E2)

higher average removal efficiency (55.5 %) in an aerated biofilter operating at an average NH_4-N influent concentration of 17.1 mg L^{-1} and an air/water ratio of 4. Therefore, high air/water ratios do not seem to benefit nitrification. According to Liu et al. (2008), if the air/water ratio is above 6 and the DO concentration exceeds 5 mg L^{-1}, nitrification becomes weakened as a result of the extremely high DO concentration. In addition, high airflow rates damage the biofilm, resulting in decreased NH_4-N removal. (Tchobanoglous et al. 2002; Liu et al. 2009).

There was no significant removal of ammonia in experiment E1, nor detection of nitrite or nitrate in any of the five samplings ports. The concentrations of oxidized nitrogen forms were below 0.6 mg NO_2-N L^{-1} and 0.23 mg NO_3-N L^{-1} (detection limits of the analytical methods). The overall average removal efficiency of NH_4-N for the 16 days was 7.6 %, which corresponds to an average r_{NH_4N} of 0.8 g NH_4-N m^{-3} h^{-1}. Approximately 99 % of the ammonia was removed in section TM–P2 where the average DO was 0.4 mg O_2 L^{-1} (P1) and 0.18 mg O_2 L^{-1} (P2). However, these values were measured at the ports and the nitrifiers benefited from the higher DO concentrations coming from the feeding solution (average value of 7.73 mg O_2 L^{-1}). These results are worser than the ones obtained in another study (Albuquerque et al. 2009a), using a similar not aerated biofilter fed with NH_4-N concentrations up to 31.2 mg L^{-1}, where an overall average NH_4-N removal of 28 % (74 % of which was observed in section TM–P2) was obtained. This higher removal rate is associated with the higher NLR used in that study, since it was also proved that the increase of the nitrogen loads influenced the respective removal rates (for influent concentrations from 5 to 31 mg NH_4-N L^{-1}).

In experiment E2, there was only production of nitrate until port P3, which can be explained by the presence of an average DO concentration of 2.62 mg O_2 L^{-1}. However, all the NO_3-N was removed in the section P3–P5, where the average DO concentration was 0.12 mg O_2 L^{-1} presumably through denitrification. According to Grady Jr et al. (1999), denitrification under anoxic environments occurs preferably at DO concentrations below 0.2 mg O_2 L^{-1} (critical value). Behera et al. (2007) observed denitrification efficiencies between 78 and 98 % in an anoxic non-aerated biofilter operating at similar HLR, but with 12 h of HRT.

The complete clogging of the biofilter was reached in 16 days without aeration and in 8 days with aeration. When observing the organic carbon removal (ΔC) and TSS evolution for experiment E1, a decrease in ΔC and an increase in TSS after the tenth day of operation (Fig. 4), which corresponds to a ΔY of 4.8 cm (3.9 cm in section TM-P2) can be seen. With aeration (Fig. 5), a similar behaviour occurred after the fifth day of operation with a ΔY of 5.1 cm (4.2 cm in section TM–P2).

Evolution of the steady-state conditions

The analysis of the steady-state conditions was focused on the variation of TOC and ΔC over time in the different ports, as also observed in Colt et al. (2005), since there was

no significant removal of ammonia in experiment E1. The measure of uncertainty used was the highest absolute deviation between the average TOC concentration and the measured TOC concentrations for each point over time (adapted from the procedure suggested in Rabinovich 2005). The results for sections TM–P2 (8 cm), TM–P3 (13 cm) and TM–P5 (33 cm) are presented in Figs. 6 and 7.

When observing the Figs. 4, 5, 6, 7, it can be seen that the biofilter reached steady-state conditions after approximately 2 days (experiment E1) and 1 day (experiment E2) of the operating period. For non-aeration conditions, it seems that the optimal operating period occurred between the second and the ninth day with an overall average TOC removal efficiency of 27 % (9 % for ammonia), which corresponded to an overall average organic mass removal rate (r_C) of 13.6 g C m^{-3} h^{-1} (Fig. 6), 90 % of which occurred in section TM-P2. In the presence of aeration, the optimal operating period occurred between the first and the fourth day with an overall average TOC removal efficiency of 76 % (51 % for ammonia), which corresponded to a r_C of 38.3 g C m^{-3} h^{-1} (Fig. 7), 96 % of which occurred in section TM–P3. Liu et al. (2009) observed steady-state conditions between the 8th and the 14th day, but using a higher HLR (1.2 m h^{-1}) and a lower air/water ratio (4) during 22 days of operation. However, the overall average organic matter removal was 70 %. Therefore, an air/water ratio of 8 seems to reduce the time required to achieve steady-state conditions, but may reduce nitrification as discussed before.

The removal of organic carbon was higher in the aerated biofilter (E2). The overall mean mass removal observed in the final of the experiment E2 was 26.3 mg C L^{-1} (i.e., an overall mean removal efficiency of 64 %), which is 2.4 times higher than the maximum average organic mass removal observed in experiment E1 (11 mg C L^{-1}, between the second and the ninth day). These results are associated with a better contact between biomass and both organic matter and nitrogen in the experiment E2, as a consequence of the turbulence caused by aeration, as well as due to the presence of a mean DO concentration of 2.62 mg L^{-1} between TM and P3. In the experiment E1, oxygen was a limiting factor (the average DO concentration between TM and P3 was only 0.31 mg L^{-1}), and the overall average organic mass removal at the end of the experiment was 9.8 mg C L^{-1} (i.e., an overall average removal efficiency of 24 %).

In experiment E1, the average TOC mass removal in sections TM–P1 and P1–P2 was 7.2 and 2.8 mg C L^{-1}, respectively (i.e., 90 % of the total TOC removal occurred in section TM–P2). In experiment E2, the TOC removal occurred mainly in the sections TM–P1, P1–P2 and P2–P3 with mass removals (ΔC = influent concentration − effluent concentration) of 26, 2.5 and 1.1 mg C L^{-1} (i.e., 96 % of the total TOC removal occurred in section TM–P3).

The deterioration of the effluent quality in terms of TSS was faster when the biofilter was aerated with a significant increase of TSS in all the ports after the fourth day of operation (Fig. 5). This occurrence is associated with a higher production of biomass (measured as VSS), the break-up of the attached biofilm with release of biofilm particles (due to air bubble friction) and the release of other reaction by-products in the aerated sections. The presence of DO concentrations above 2 mg L^{-1} stimulated the activity and fast growth of aerobic heterotrophic microorganisms, for the oxidation of organic carbon, and of nitrifiers, for the oxidation of ammonia. However, as referred by Di Iaconi et al. (2005), the continuous increase of biofilm thickness produces a corresponding increase of the shear forces with negative effects on biomass stability, causing the detachment of biofilm particles. As a consequence, the media became progressively clogged especially after the fifth day, the filter capacity decreased, short-circuiting increased and more solid particles were released through the section ports.

The results seem to indicate that the filtration capacity of the biofilter began to become saturated after the tenth day (non-aerated biofilter) with a ΔY of 4.8 cm and after the fifth day (aerated biofilter) with a ΔY of 5.1 cm, and a significant increase of head losses and a decrease in TOC and TSS removal was observed. Therefore, in order to ensure a good performance treatment, the biofilter should be operated in cycles of 10 days (without aeration) and 5 days (with aeration). After these periods, or whenever the head losses exceed 5 cm, backwashing should be performed. Standard fully aerated tertiary biofilters are usually backwashed every week (Mendoza-Espinosa and Stephenson 1999; Xie et al. 2004). Liu et al. (2009) also established 10 days for biofilter operation and used an air/water ratio of 4. Gonçalves and Oliveira (1996) observed an average TSS removal of 63 % in a submerged aerated biofilter operating at a HLR of 2 m h^{-1}, but for 3-day operating cycles. After this time, the head losses increased up to 50 cm and TSS removal decreased to less than half.

The assessment of the biofilter recovery after washing is of interest in order to know how long it takes to return to the steady-state conditions (i.e., the time that the biofilm needs to reach again its maximum capacity to remove organic matter and nitrogen). Figure 8 shows the variation of ΔC over time after backwashing in ports P1, P3 and P5 and Table 2 the biofilm thickness in ports P1 and P3, before and after backwashing, for the two experiments. It can be seen that the biofilter reached steady-state conditions again after 6 h (E1) and 7 h (E2) of washing.

Although after washing the biofilm thickness was similar in both experiments, a thinner biofilm was observed in

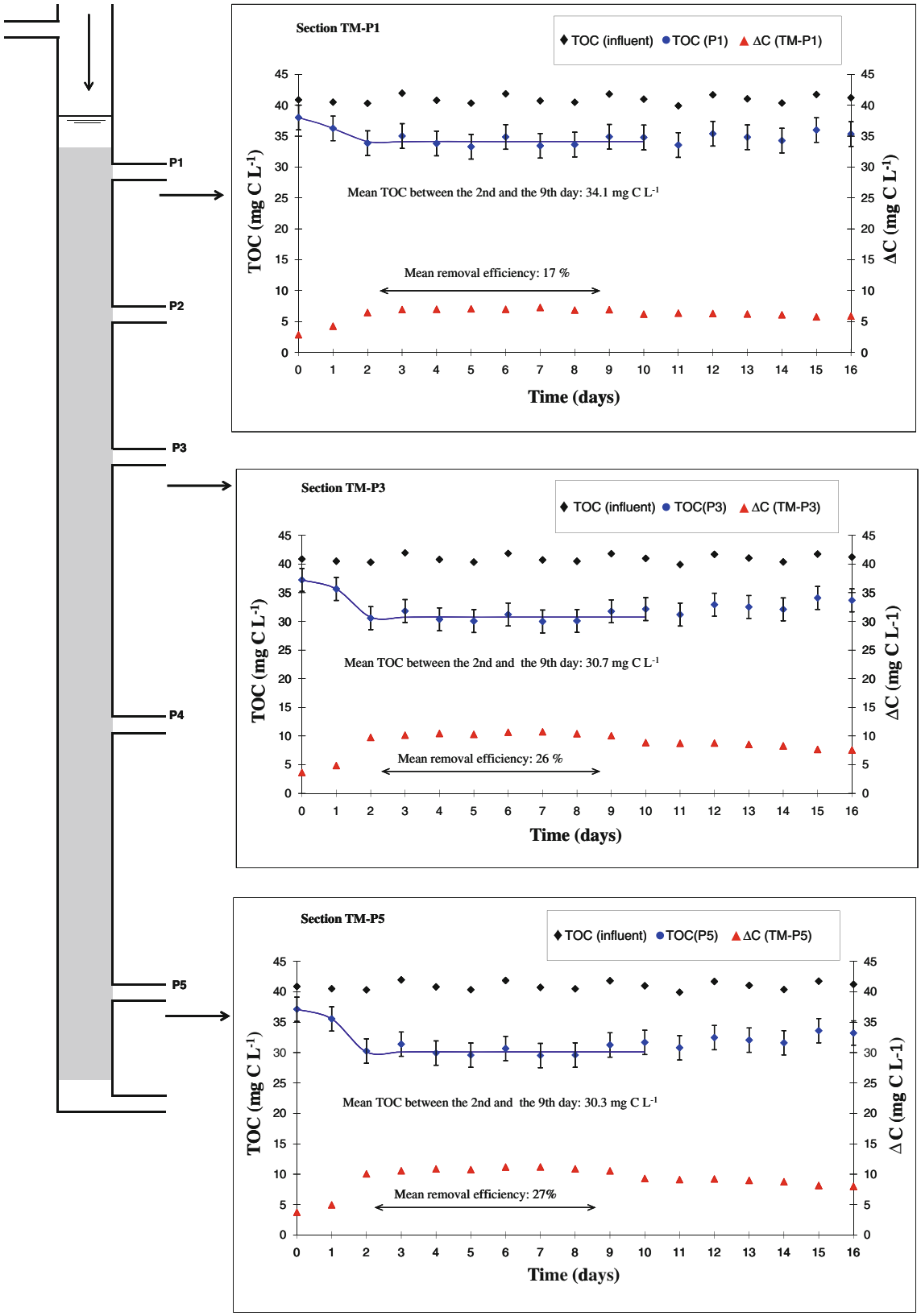

Fig. 6 Variation of the steady-state conditions in points P1, P3 and P5 (E1)

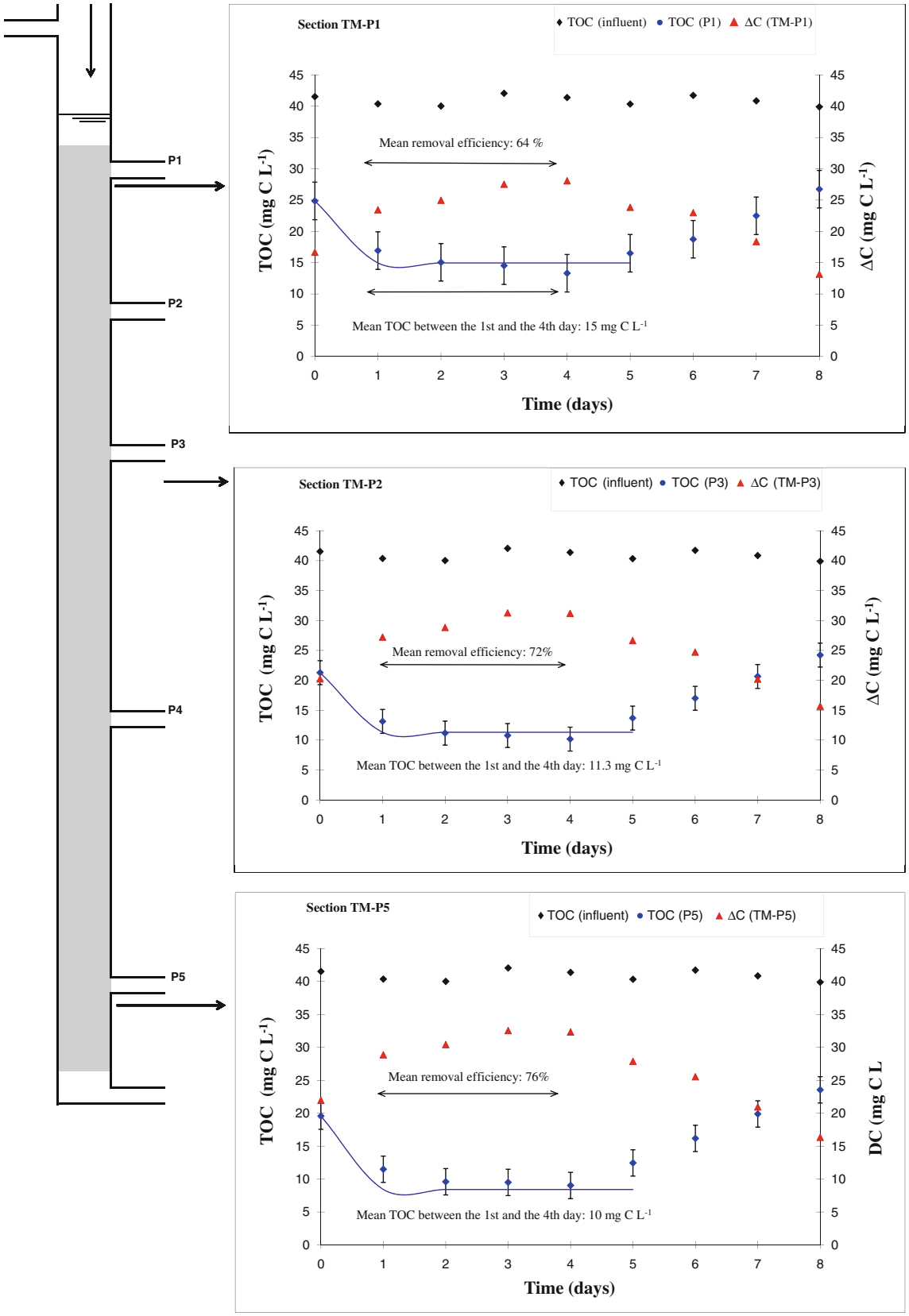

Fig. 7 Variation of the steady-state conditions in points P1, P3 and P5 (E2)

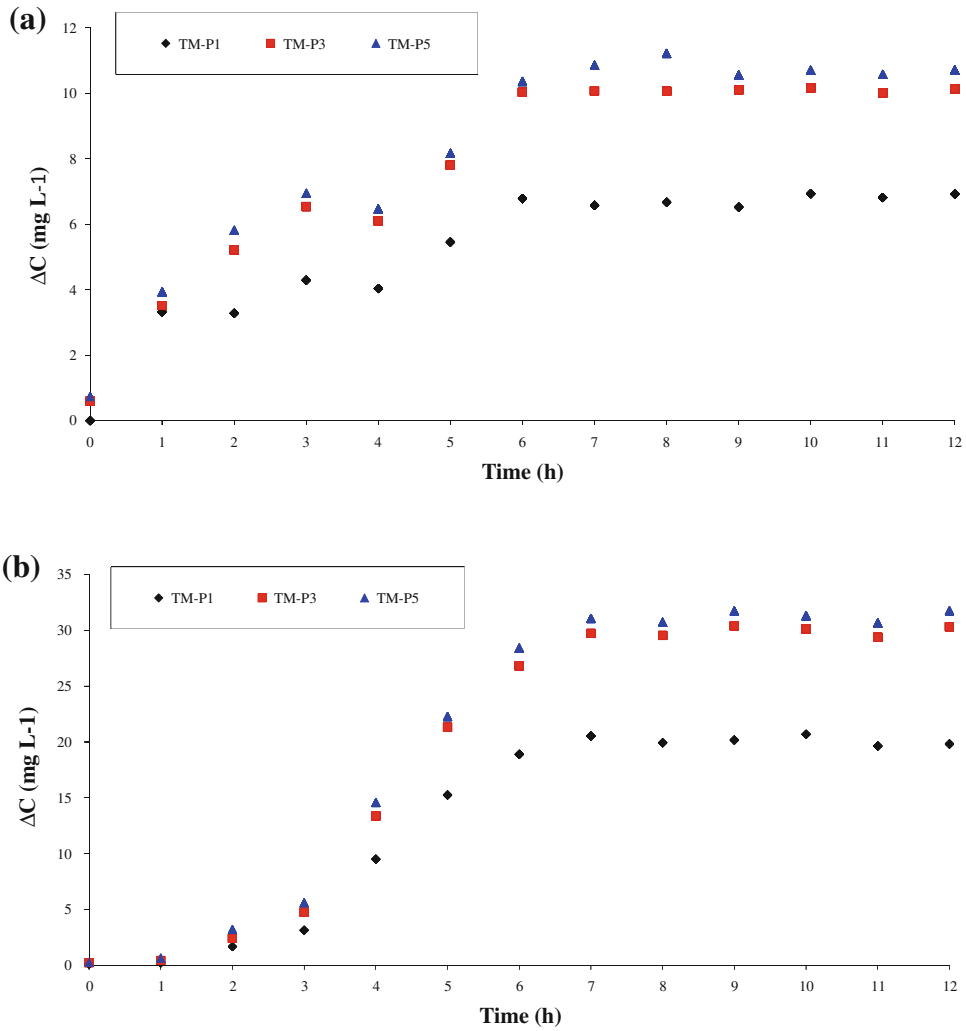

Fig. 8 Recovery of the steady-state conditions after backwashing: **a** experiment E1, **b** experiment E2

Table 2 Variation of the biofilm thickness before and after washing

Sampling point	Biofilm thickness (mm)			
	Experiment E1		Experiment E2	
	Before	After	Before	After
P1	0.76 ± 0.13	0.52 ± 0.08	0.65 ± 0.06	0.51 ± 0.05
P3	0.13 ± 0.05	0.10 ± 0.02	0.12 ± 0.04	0.10 ± 0.01

Average and confidence interval (calculated for a confidence level of 95 % and 4 samples)

the experiment with aeration before washing. This circumstance, as observed by Visvanathan and Nhien (1995), is associated with the friction caused by the release of air bubbles during the aeration, which led to the release of old biofilm in decay process. Liu et al. (2009) observed recovery times between 3 and 6 h in aerated biofilters with equal backwash air/water ratio (equal to 5), but using lower washing times (5 against 10 min used in this study). Therefore, lower washing times may allow a quick recovery of the steady-state conditions, provided that there is an efficient removal of sludge and old biofilm.

Conclusion

Tertiary biofilters may be suitable bioreactors for the removal of residual loads of organic matter, nitrogen and solids if the steady-state conditions and head losses can be properly controlled. The biofilter used in this study allowed obtaining, during the periods of steady-state conditions, overall average removal efficiencies of TOC and NH_4-N of 27 and 9 % (without aeration) and of 76 and 51 % (with aeration), respectively. The introduction of aeration did not influence too much the time necessary to achieve steady-state conditions, since such conditions were observed 2 days (E1) and 1 day (E2) after the start-up and 6 h (E1) and 7 h (E2) after backwashing. However, when the biofilter was aerated, steady-state conditions were interrupted in half the operating time, besides resulting in higher

average removal rates of organic carbon and ammonia (35.8 g C m^{-3} h^{-1} and 6.3 g NH_4-N m^{-3} h^{-1} against 12.7 g C m^{-3} h^{-1} and 1.1 g NH_4-N m^{-3} h^{-1} when the biofilter was not aerated). The biofilters should be operated at air/water ratios below 8 in order to ensure a good treatment performance. Backwashing should be performed every 10 days (E1) or every 5 days (E2), using an air/water ratio of 5 and a time period below 10 min.

Acknowledgments The authors also wish to extend their sincere gratitude to all who assisted in promoting the present work.

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