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A study of clogging in a biofilter treating toluene vapors

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Abstract

In ideal biofilters, organic pollutants are essentially converted into mineral end products, while biomass is produced at low rates. Nevertheless, in real cases, excess biofilm develops within the porous medium, and thus causes clogging of fixed-bed bioreactors. This paper presents a characterization of biofilters clogging. Two identical, laboratory-scale biofilters have been operated in parallel, for a 2-month period, for the removal of toluene vapors. The total air flow rate was set at 1 $m^3\,h^{-1}$ and the toluene concentration at 1.9 $g\,m^{-3}$. The periodic supply of incremental nitrogen concentrations led to the progressive and cumulative formation of biomass, and the consequential increasing pressure drop. This nutrient addition allowed to control biofilter performance, and to achieve elimination capacity values around 90–95 $g\,m^{-3}\,h^{-1}$, in both units, for nitrogen flow rates near 0.4 g N day^{-1}. Relationships were developed to evaluate the biofilm thickness (∼510 μm) and to correlate the pressure drop with the bed porosity, according to an equation inspired by Ergun. Further, four methods of clogging control have been tested. In the first time, two mechanical methods have been employed: bed stirring and bed washing, which permit removal of part of the excess biofilm, without affecting the performance (removal efficiency ≥80%). Secondly, two chemical methods, based on the nutrient (nitrogen) control, have also been evaluated. However, they did not lead to significant control of pressure drop and their corresponding performance was not satisfactory (removal efficiency ≤60%).

Keywords: Biofiltration; Air treatment; Clogging; Toluene; Biofilm; Nutrient control; Pressure drop

1. Introduction

Biological air pollution control technologies constitute recent alternative process for pollution control and are used for an increasing variety of industrial processes [3,4,30]. There are several reasons for such a development: firstly, the relatively low investment and operation costs; secondly, the increasing competitiveness of these biotechnologies in comparison to the more traditional and well-established processes, such as the adsorption, absorption, incineration or condensation process [31]. The basic principle of volatile organic compound (VOC) aerobic biofiltration is that of catalyzing the oxidation of pollutants diluted in atmospheric effluents, in a biological way. Biocatalysts consist of consortia of various microbial species immobilized on packing materials. The microorganisms utilize these pollutants in two ways: (a) they contribute to the growth of new cellular material (anabolic pathway) and (b) they are sources of energy, which is synthesized through the respiratory system (catabolic pathway).

The ideal biofiltration process mineralizes toxic VOCs to inorganic products (CO_2, H_2O, etc.) [13,19] and minimizes the formation and accumulation of biological matter, phenomena, favored by increasing pollutant loading rates. Thus, for those fixed-bed bioreactors, operated with significant inlet loads of VOC, optimum sustainable removal efficiency, and control of bed clogging processes remain major challenges [13,21]. Few published works actually deal with the clogging as a limitation on air biofiltration, and with methods designed for blockage control. Processes for excess biomass removal are mostly encountered in water treatment bioreactors or in air biotrickling filters and are mainly derived from three major types: physical/mechanical, chemical and biological. Physical treatments consist mainly of bed stirring or in draining the accumulated biomass via water backwashings [2,10,23,35]. Chemical treatments use reagents that dissociate the chemical bonds between the biofilm and the supporting solid surfaces or that directly degrade the biomass to a removable condition (NaCl, H_2O_2, HClO, etc.) [1,5,7,14,33]. Biological treatments utilize microorganisms that are able

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Nomenclature

a_s	specific surface area ($m^2\,m^{-3}$)
A	empirical correction factor in Ergun's correlation (–)
C_I	inlet concentration of toluene ($g\,m^{-3}$)
C_o	outlet concentration of toluene ($g\,m^{-3}$)
d	diameter of the pellets (m)
EC	elimination capacity ($g\,m^{-3}\,h^{-1}$)
H	bed height (m)
IL	inlet load of toluene ($g\,m^{-3}\,h^{-1}$)
n_1, n_2	empirical correction exponent in Ergun's correlation (–)
ΔP	pressure drop (cm $H_2O\,m^{-1}$)
Q	volumetric air flow rate ($m^3\,h^{-1}$)
v_0	empty tower gas velocity ($m\,s^{-1}$)
V	bed volume (m^3)
X	removal efficiency (%)

Greek letters

δ	biofilm thickness (μm)
ε	bed porosity (–)
μ	air dynamic viscosity ($kg\,m^{-1}\,s^{-1}$)
ρ	air density ($kg\,m^{-3}$)

to degrade the biomass accumulated throughout the bed [8]. The major drawback of all of these methods is the reduction in biofilter performance due to the loss of biomass, including the active degrading microorganisms. So, the balance between the control of excess biomass generation and the maintenance of optimum sustainable pollutant removal efficiency remains an important challenge in biofiltration.

The objective of the study presented here was the examination of various processes involved in clogging; in particular, the evolution of the pressure drop through the bed, the changes in filtering material characteristics, such as the density and porosity, with time. To this end, two biofilters have been operated for the removal of toluene vapors, specially until the biofilm produced was sufficient to obstruct flow in the bed. Then, four methods of clogging prevention and control have been evaluated. Two mechanical methods (stirring and washing) were tested to remove excess biological matter accumulated, and two chemical methods, based on nutrients supply control, were also tested to avoid excess biomass formation.

2. Materials and procedures

2.1. Equipment

Two identical biofilters, as detailed in Fig. 1, have been operated: units U1 and U2. The main component of this system was the biofiltration column, composed of three identical modular sections, made from Plexiglas®. Each section was provided with a stainless steel screen that supported the weight of the filtering material and ensured a radial distribution of gas between adjacent sections. The gas inlet was located at the biofilter base, so that the airflow through the beds was upwards. Before being introduced into the column, the air was separated into two flows. The main fraction was directed to a humidification unit, which guaranteed a minimum 95% water saturation level. The other flow was conducted through a thermostated (20 °C) toluene-saturation chamber. Both airflows were then mixed in a homogeniza-

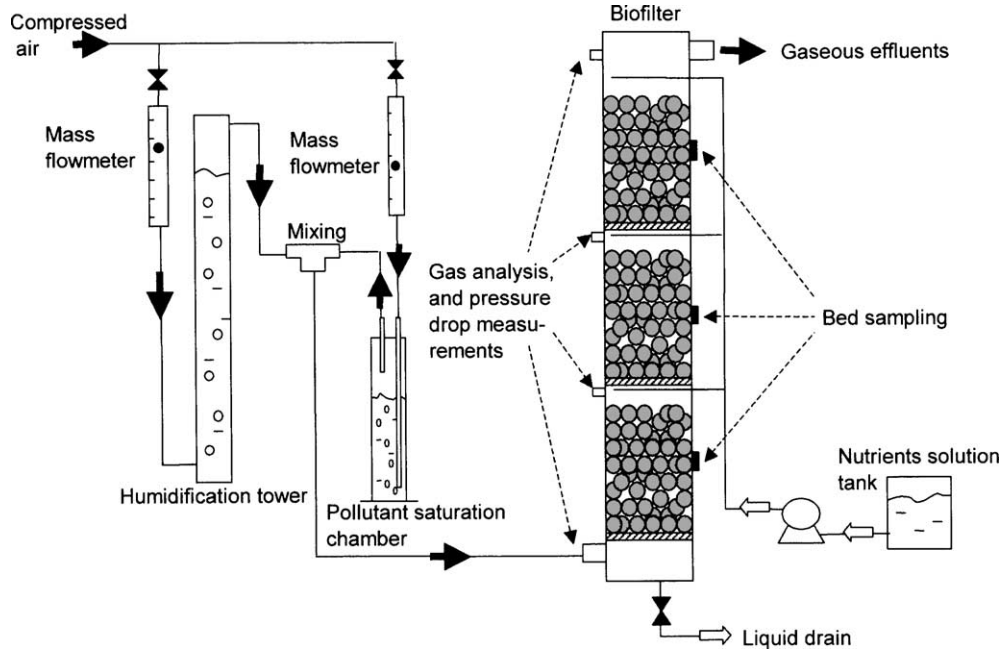

Fig. 1. Biofiltration unit.

Table 1
Chemical composition of raw compost (main components)

	Raw compost
pH	6.8
Organic matter (%)	31
Total N (%)	1.0
Total P (%)	1.0
Available P (% of P_2O_5)	1.3
Total K (%)	0.37
Available K (% of K_2O)	<0.1
Na (%)	0.18
Fe, Al (%)	2.0–2.4

Note: Percentages are given by weight.

tion vessel and conveyed through the biofilter, via Teflon® tubing.

2.2. Filtering material and irrigation solutions

The filtering material was composed of a mixture of matured compost provided by Condor Gestion Ltd., Montréal, Que., Canada) and an organic binder (details not available here because of existing confidentiality agreements), in 90:10 (w:w) proportions. The chemical composition of the raw compost is provided in Table 1. Each bed section, of 15.3 cm in diameter, was filled with the bed material to a height of 33 cm. As the compost naturally contained a wide variety of microbial species, among them the toluene degrading microflora [10], the bed was not inoculated. The particles that composed the bed were quite spherical, and after sieving only the fraction having an average diameter of 8.0 mm was kept.

In order to maintain constant the bed moisture content and to provide the microorganisms with the required nutrients, an irrigation solution was delivered to each biofilter section, individually, twice a day (total 1.3 l per section, per day). One liter of this solution contained the following components: urea: variable quantity; $MgSO_4{\cdot}7H_2O$: 0.20 g; $FeCl_3{\cdot}6H_2O$: 0.05 g; $CaCl_2$ anhydrous: 0.02 g; NaCl: 10.0 g; H_3PO_4 (85%): 0.96 ml; K_2SO_4 anhydrous: 0.36 g; distilled water: 1 l. To study the effect of nitrogen, the quantity of urea was varied to provide the scheduled nitrogen concentrations listed in Table 2. The pH of this solution was adjusted at 7.8 by using a NaOH solution (5N).

2.3. Analytical methods

The determination of toluene concentration at each gas sampling port was effected daily, with a gas chromatograph coupled to a mass spectrometer (Hewlett-Packard, 5890 Series II, column HP5: 30 m × 2.5 mm × 2.5 μm, crosslinked 5% Ph Me Silicone, oven temperature: 80 °C, sampling loop, carrier gas: He). The GC/MS was calibrated every week with appropriate toluene standards. Bed pressure drop was measured with a differential manometer (Air Flow Developments Ltd., UK, Type 4). The filter bed porosity (ε) was also measured, according to the method proposed by Hodge and Devinny [17]. It was estimated from 1 l of particles, sampled from the three biofilter sections, mixed together, and then placed in a 1 l graduated cylinder. The volume of water added to fill the void spaces yields the value of ε, according to $\varepsilon = V_{\text{added water}}/V_{\text{total}}$. The bed density ($\rho$, kg m^{-3}) was determined experimentally by measuring the mass of 1 l of the same bed pellets as for the porosity measurements. After utilization, pellets were reintroduced into the three biofilter sections. It is to be noted that only 1 l was sampled each time. Thus, performance and pressure drop were not perturbed by these operations, which was confirmed by performance continuity on the following days.

The total and toluene-specific bacterial counts were performed according to the Most Probable Number method, adapted from Wrenn and Venosa [34]. Five grams of pellets were sampled from the three biofilter sections and mechanically vortexed in a buffer containing 2% sodium chloride and 0.1% sodium pyrophosphate. Microbial extracts were obtained after a low speed centrifugation (1000 × g, 5 min) of the mixture. The total counts were obtained by incubation of extracts in a medium containing a commercial saline solution

Table 2
Nitrogen concentrations in the nutrient solutions delivered to biofilters U1 and U2, and their corresponding operating periods

Biofilter U1			Biofilter U2		
Days	Operating period	N flow rate (g of N day^{-1})	Days	Operating period	N flow rate (g of N day^{-1})
0–3	A1	0.0	0–3	A2	0.0
4–14	B1	0.2	4–14	B2	0.2
15–18	C1	0.3	15–18	C2	0.3
19–35	D1	0.4	19–35	D2	0.4
Bed stirring					
36–42	E1	0.5	36–42	E2	0.5
Bed washing					
41–42	F1	0.5	41–42	F2	0.5
43–50	G1	0.3	45–58	G2	Alternating 0.5–0.0 per 24 h
51–57	H1	0.2	59–67	H2	Alternating 0.5–0.04 per 24 h
58–60	I1	0.04		–	–

(BH, Büshnell-Haas, Difco) and completed with tryptone (2.5 g l^{-1}), yeast extract (1.25 g l^{-1}) and glucose (0.5 g l^{-1}). The toluene-specific counts were obtained after incubation in a BH medium, amended with toluene vapors as sole carbon source. Incubations were carried out at 250 °C, for 1 week (total counts) or 2 weeks (toluene-specific). Finally, positive wells were detected by the reduction of iodonitrotetrazolium violet [34].

2.4. Operating protocol

Biofilters, U1 and U2, were operated over a 60-day period at the ambient laboratory temperature (22 °C). The input air flow rate was maintained at 1 m^3 h^{-1} (empty bed residence time = 65 s), and the toluene inlet concentration was fixed at 1.9 g m^{-3}. Initially, both biofilters were exposed to the same incremental nitrogen flow rates, from 0 to 0.5 g of N day^{-1}, as indicated in Table 2. The aim of this incremental nitrogen distribution was firstly, to evaluate the performance and the reproducibility of the biofilters, and secondly, to favor the formation of biomass and increase the pressure drop. Then, four methods for control of clogging were evaluated: (1) bed stirring, which consists in emptying the column, stirring media in a separate tank and reintroducing the pellets in the biofilter, (2) bed washing with a co-current flow of fresh water fed by a centrifugal pump, (3) decreasing nitrogen input concentrations for unit (U1), and (4) alternate distribution of nitrogen concentration for (U2), as detailed in Table 2. The various performance parameters and their definitions are listed in Table 3.

Table 3
Performance parameters and their definitions

Parameter	Definition
Inlet load (g m^{-3} h)	$IL = \frac{C_I Q}{V}$
Removal efficiency (%)	$X = 100\left(\frac{C_I - C_O}{C_I}\right)$
Elimination capacity (g m^{-3} h^{-1})	$EC = X \times IL$

3. Results and discussion

3.1. Biofilters' performance and pressure drop

Figs. 2 and 3 present the performance and the pressure drops versus operation time for U1 and U2, respectively. For both reactors, a major decline in the outlet toluene concentrations was observed on the second day of operation; from 1.6 to 0.05 g m^{-3} for U1, and from 1.9 to 0.4 g m^{-3} in U2. Delhoménie et al. [11] have demonstrated that this early

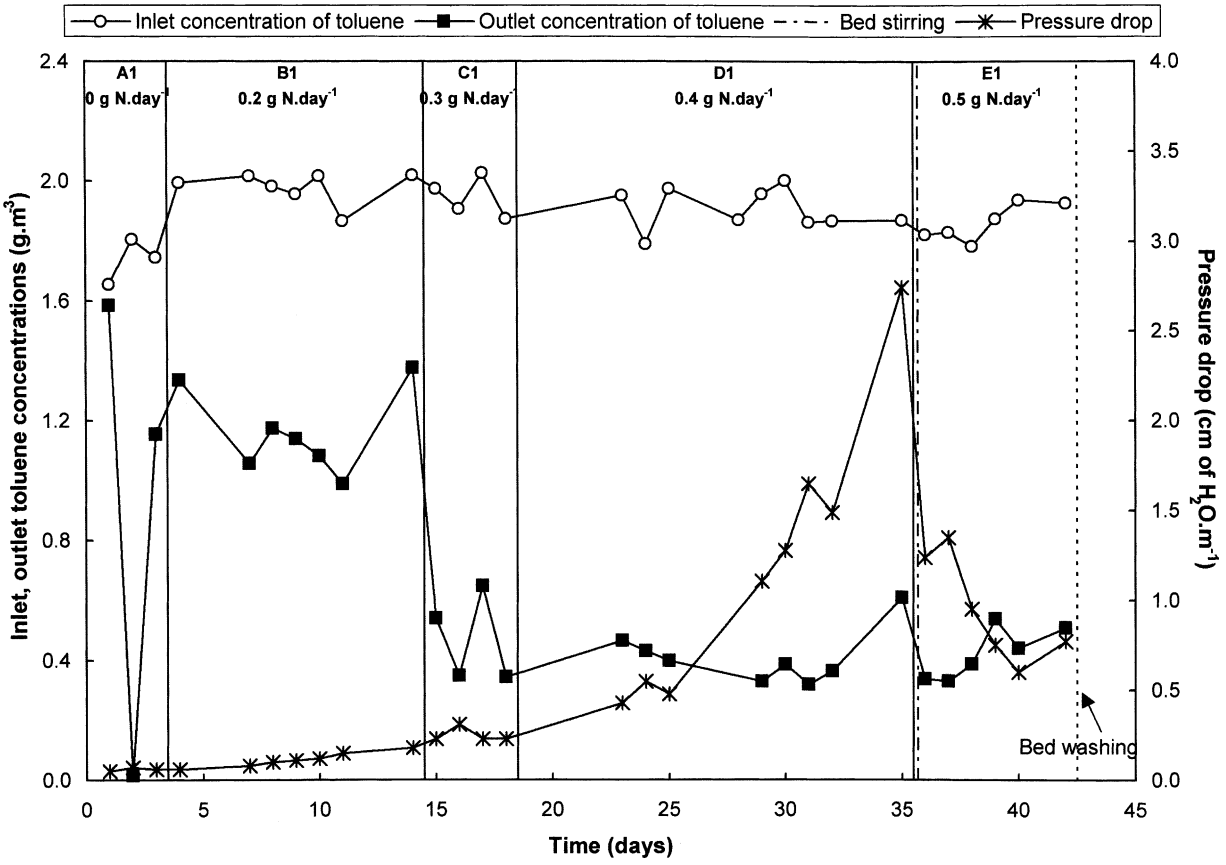

Fig. 2. Evolution of inlet and outlet concentrations of toluene, and pressure drop vs. time in biofilter U1.

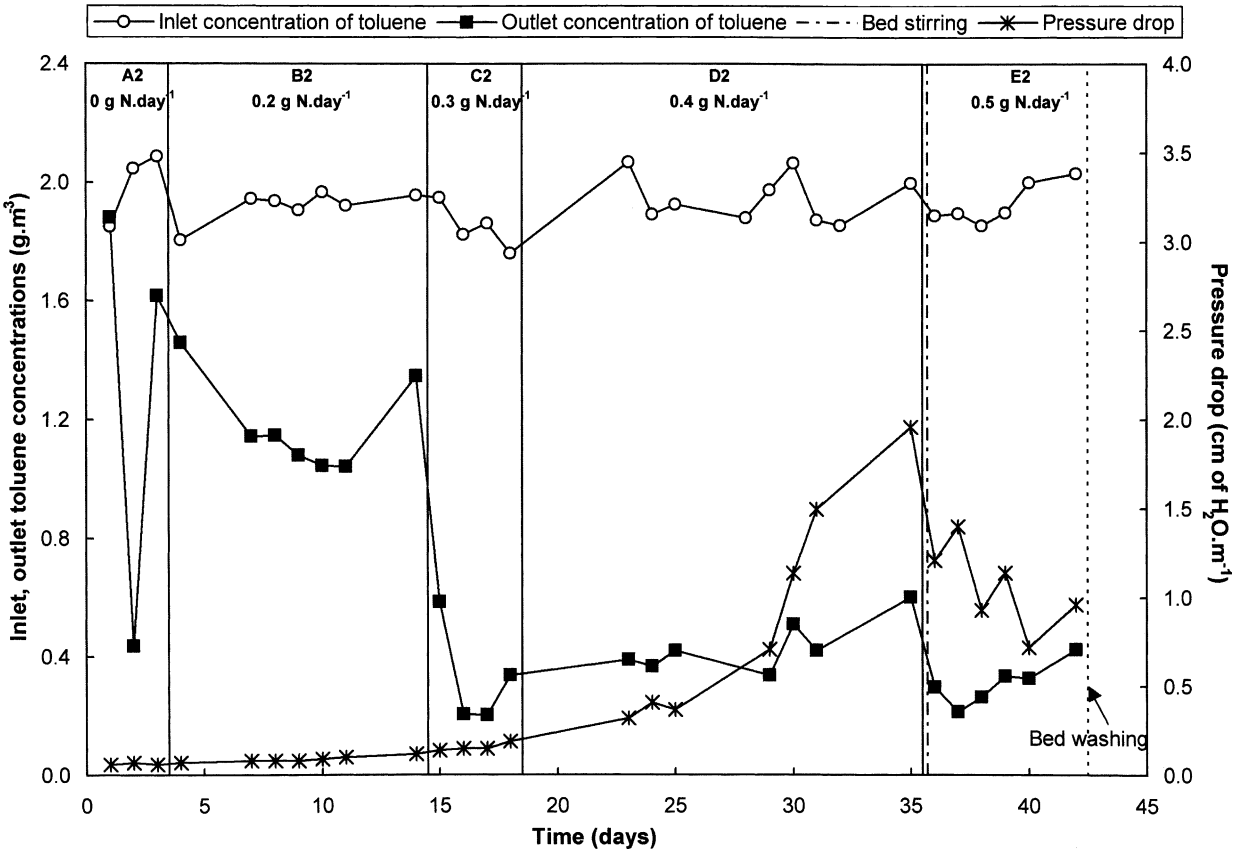

Fig. 3. Evolution of inlet and outlet concentrations of toluene, and pressure drop vs. time in biofilter U2.

peak results from a pulse of degradation activity that occurs within the start-up period. During the subsequent periods, A to C, the quantity of toluene degraded seems to be related to the flow rate of nitrogen introduced to the biofilters. Indeed, from A to C, the increase in nitrogen input (from 0 to 0.3 g of N day^{-1}) leads to corresponding incremental decreases in the toluene outlet concentrations (from 1.2 (U1) and 1.6 (U2) to 0.3 g m^{-3}), for constant inlet concentrations of about 1.9 g m^{-3}. This shows that over these periods, nitrogen input was in deficit in comparison to the carbon input, and limited the degradation level. Then, over the last two periods, D (0.4 g of N day^{-1}) and E (0.5 g of N day^{-1}), the outlet concentrations of toluene remained quite stable at around 0.3–0.4 g m^{-3}. This stability in degradation level indicates that introducing more than 0.4 g of N day^{-1} was effectively in excess in comparison to the biological needs and had no influence on the performance. Moreover, it is to be noted that the toluene elimination during periods D and E was not complete (~80%), even with excess nitrogen. This point can be explained by the fact that limitations (mass transfer, physical, biological, etc.), other than nitrogen-limitation, could have taken place in the filtering material.

The nutrients dependency of biofilters has previously been noted by Corsi and Seed [6] and Morgenroth et al. [26] who studied the impact of nutrients exhaustion intrinsic to the filtering media. Gribbins and Loehr [16] noted the significant benefits of nitrogen-compounds additions to compost beds on the overall performance. The impact of the irrigation solution nitrogen content has been reported on by Delhoménie et al. [9] who also demonstrated the existence of a N-limitation, and a N-excess regimes.

Figs. 2 and 3 also present the evolution of pressure drop for both units U1 and U2. The pressure drops remained stable at very low values (<0.3 cm H_2O m^{-1}) over periods A–C, and then rapidly increased over period D: the pressure drop rose to 2.7 and 2.0 cm of H_2O m^{-1}, in U1 and U2, respectively. The evolution of pressure drop was similar in both bioreactors, which suggests that the hydrodynamic behavior was equivalent in U1 and U2.

3.2. Bacterial numeration, bed specific gravity and porosity

A comparison of data in Figs. 2 and 3 illustrates the performance similarity between biofilters U1 and U2. For this reason, the following hydrodynamic considerations and bacterial numerations have been effected on the reactor U1 only. Fig. 4 presents the changes in bacterial counts (total and toluene-specific), with time, before the bed washing. Fig. 4 shows the existence of three bioactivity domains: (1)

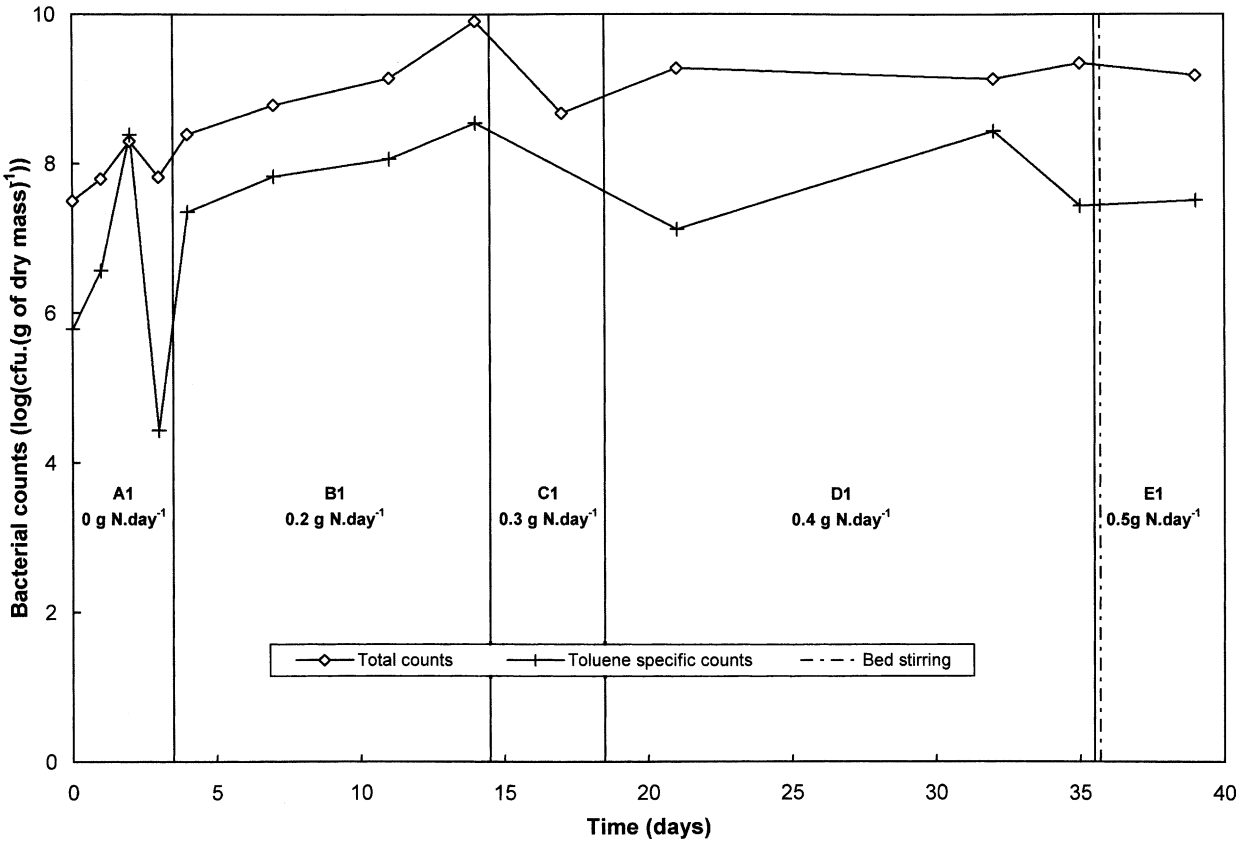

Fig. 4. Evolution of the total and toluene-specific bacterial counts in biofilter U1 vs. time.

a start-up peak, on the 2nd day, (2) a rapid activity increase region on the 3rd and 4th days, and (3) a stabilization zone until the 40th day. During the start-up peak, the total bacterial counts rose from 7.5 to 8.4 log(cfu(g of dry mass)$^{-1}$) and the toluene-specific counts from 5.8 to 8.4 log(cfu(g of dry mass)$^{-1}$). It is to be noted that on the peak summit, the bacterial populations were mostly composed of toluene degrading microorganisms. This peak is certainly related to the consumption of nutrients that were readily available in the filter material. On the third day, both counts steeply declined, down to 7.8 and 4.4 log(cfu(g of dry mass)$^{-1}$). This decrease could be explained by the exhaustion of the nutrients initially present in the filtering material. During the subsequent growth phase, the total and toluene-specific counts increased and then remained stable at around 9.2 and 8.0 log(cfu(g of dry mass)$^{-1}$), respectively, even after the filter bed stirring (35 days).

The microbial count values obtained during these experiments are of same magnitude as bacterial counts proposed by Ottengraf and van den Oever [27], Medina et al. [24], and more recently by Krailas et al. [22] (from 6 to 10 log(cfu(g of sample)$^{-1}$). Moreover, the stabilization phase shows that a microbial equilibrium (no apparent cell growth) has established in the bed and the toluene degrading species represented 6–7% of the total populations. Such ratios of degrading to total bacterial populations have also been reported by Pedersen et al. [28] who determined that it takes only 1–10% of the total population to be active in the biodegradation process.

Fig. 5 represents the evolution of the bed porosity and specific gravity (ratio to the water density) over the first 35 days, before bed stirring. Over these 35 days, the specific gravity continuously increased from 0.55 to 0.81, indicating that biological matter accumulated in the bed structure. Over the same period (apart from the start-up phase), the bacterial counts were relatively constant (no cell growth), indicating that the additional biomass was mostly composed of microbial secretions, such as polysaccharides or various proteins [20], and dead cells. The accumulated biomass occupied the available void spaces of the bed, physically measured by the porosity. Therefore, the bed porosity is expected to decrease in the same way as the specific gravity increases. As presented in Fig. 5, the porosity and specific gravity evolve in antisymmetric ways and the bed porosity effectively decreased, from 0.53 to 0.31 before bed stirring.

Thus, the data displayed in Figs. 2–5 permit us to directly relate the continuous pressure drop increase to the accumulation of exocellular compounds in the bed, which subsequently translates to a specific gravity increase in the filter bed, or a bed porosity decrease with time.

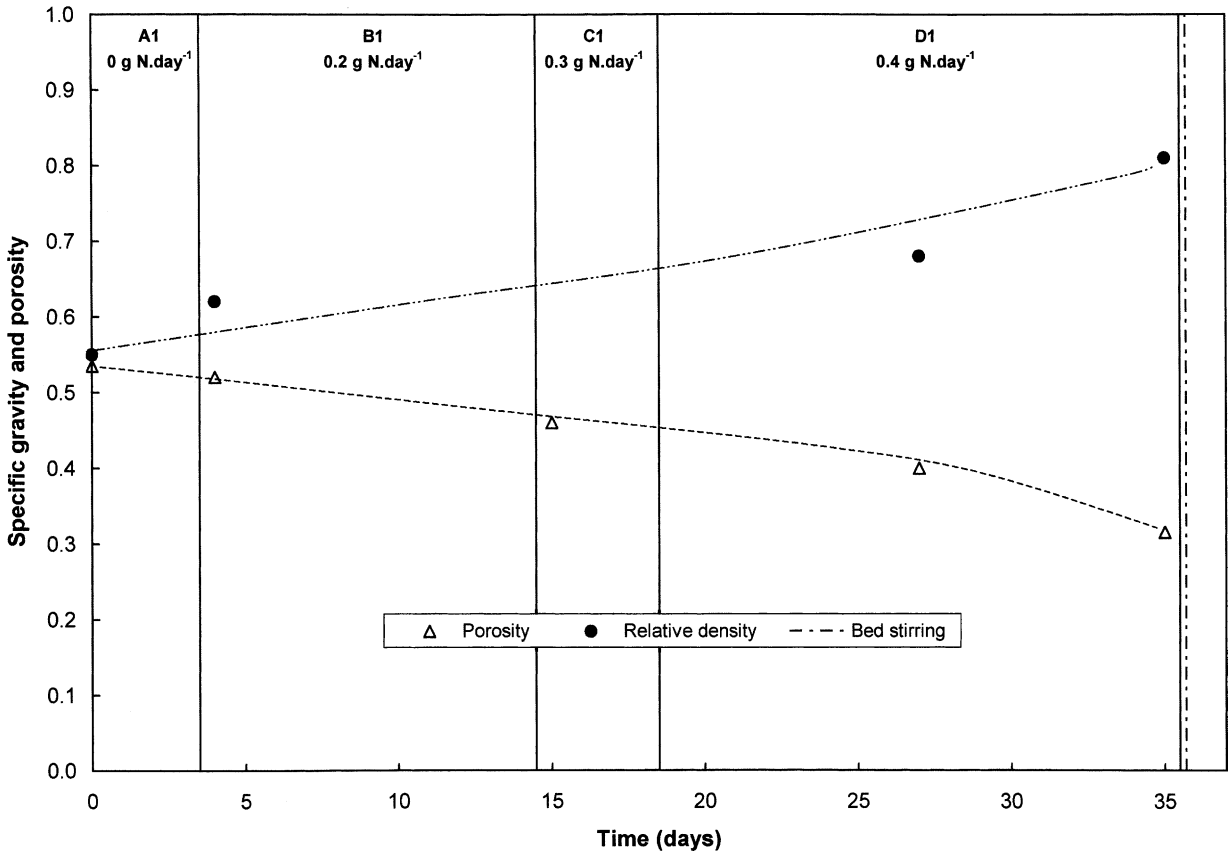

Fig. 5. Evolution of the bed specific gravity and porosity of in biofilter U1 vs. time.

3.3. Correlation between bed porosity and pressure drop

As previously demonstrated, the biomass growth, which is the main cause of biofilter clogging, takes place on the bed pellets surface, i.e. in the interstitial bed porosities [32]. In a simple geometrical case, a surface element of a particle can be considered as a plane. Under these conditions, porosity (ε) and biofilm thickness (δ) can be easily correlated, via Eq. (1) [12]:

$$\varepsilon = \varepsilon_0 - a_s\delta \tag{1}$$

where ε is the initial bed porosity (measured as 0.53), a_s is the specific surface area ($m^2\,m^{-3}$). By assuming that the particles are spherical, the specific area can be estimated through the following equation:

$$a_s = \frac{6(1-\varepsilon)}{d} \tag{2}$$

where d is the mean diameter of the bed pellets and evolves with biofilm thickness on the pellets according to

$$d = d_0 + \delta \tag{3}$$

where d_0 is the initial mean diameter (measured as 8.0 mm).

Fig. 6 presents the evolution of the biofilm thickness, estimated via an iterative solution for the system of Eqs. (1)–(3), with respect to time. The biofilm thickness is an increasing function of time, which is in accordance with the fact that porosity is a decreasing parameter (Fig. 5). Moreover, the biofilm formation on the pellets surface was a visible phenomenon during the experiment. The thickness values predicted by this simplified model rise to 510 μm, as pressure drop in biofilter U1 reached its highest value ($2.7\,cm\,H_2O\,m^{-1}$). The values for biofilm thickness, obtained in this work, are of the order of those reported elsewhere, ranging between 30 μm and 2.5 mm [29].

The biofilm growth creates increasing resistance to gas flow through the porous bed, as the void volume (ε) available for convection is reduced. In considering a monophasic fluid system (i.e. homogeneous mixture between air, toluene and water), Ergun [15] established that the pressure drop, ΔP, through an ideal porous fixed-bed plug-flow reactor could be correlated with the porosity of the bed through the following relationship:

$$\frac{\Delta P}{H} = \frac{150\mu v_0}{d_p^2}\frac{(1-\varepsilon)^2}{\varepsilon^3} + \frac{1.75\rho v_0^2}{d_p}\frac{(1-\varepsilon)}{\varepsilon^3} \tag{4}$$

where μ is the air dynamic viscosity ($1.8\times10^{-5}\,kg\,m^{-1}\,s^{-1}$), ρ the density of air ($1.2\,kg\,m^{-3}$), v_0 the empty column air velocity ($1.5\times10^{-2}\,m\,s^{-1}$), d_p the mean diameter of the pellets (8.0 mm) and H is the bed height (1 m).

Fig. 7 presents the variations of the pressure drop versus the porosity in the biofilter U1. The pressure drop values

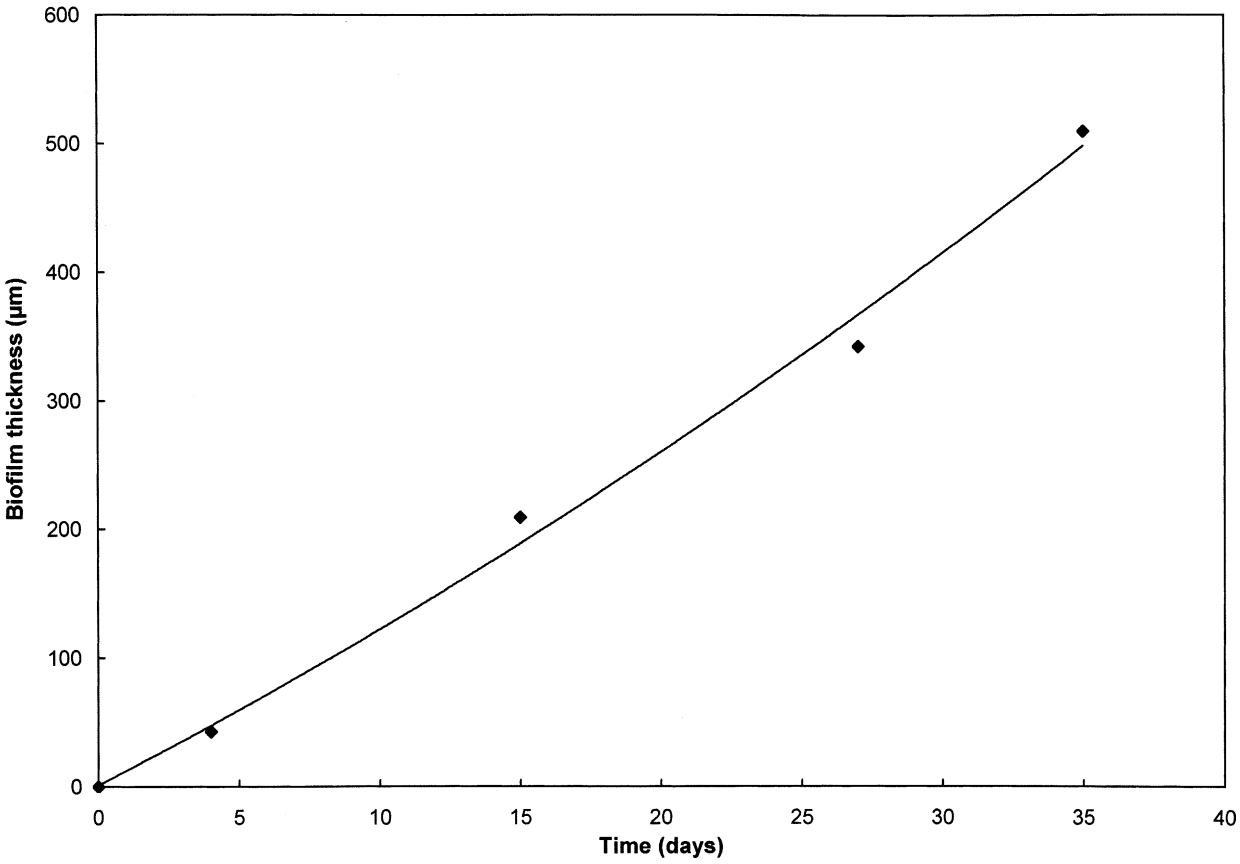

Fig. 6. Simple modeling of the evolution of the biofilm thickness with time, based on a planar geometry.

correlated according to Eq. (4) are also reported in Fig. 7. Both experimental and correlated data follow the same trend: from high to low porosity values, the pressure drop increases slightly, down to $\varepsilon = 0.6$, then exponentially under $\varepsilon = 0.6$. These observations are in qualitative agreement with the fact that lower porosities induce higher resistance to gas flow, whereas high interparticular volumes facilitate gas convection. Nevertheless, the gap between the experimental points and the Ergun's correlation is quite significant. The difference is certainly due to the following points: the fixed-bed bioreactor is not an ideal plug-flow reactor, as assumed in Eq. (4), and the uncertainty of the ε evaluation method provides an overestimated value of ε in comparison with the real value (biofilm junctions partially damaged during the sampling).

To provide a better fit of the experimental data, an empirical correlation is proposed as follows:

$$\frac{\Delta P}{H} = A\left(\frac{150\mu v_0}{d_p^2}\frac{(1-\varepsilon)^2}{\varepsilon^{n_1}} + \frac{1.75\rho v_0^2}{d_p}\frac{(1-\varepsilon)}{\varepsilon^{n_2}}\right) \quad (5)$$

where A is a correction factor, and n_1 and n_2 are porosity exponents, which take into account the uncertainties during the experimental determination of ε. The empirical correlation, calculated through Eq. (5), fits to the experimental points with $A = 0.9$, and $n_1 = n_2 = 6$. The empirical set of data is reported in Fig. 7 (dotted line). As the empirical porosity exponents ($n_1 = n_2 = 6$) are far from the Ergun's proposition ($n = 3$), it is confirmed that the major uncertainty came from the experimental determination of ε. A similar prediction has been proposed by Morgan-Sagastume et al. [25].

3.4. Clogging prevention

According to previous discussions, the biofilters long-term efficiency relies on the prevention and/or remediation of clogging. During these experiments, four remedial methods have been tested on biofilters U1 and U2.

3.4.1. Bed stirring

At the end of period D, pressure drop values became sufficiently high (2.0 and 2.7 cm $H_2O\,m^{-1}$, in U1 and U2, respectively), and a bed stirring was operated on the 35th day. As pointed out by data plotted in Figs. 2 and 3, this operation effectively led to a substantial decline in pressure drop the following day (1.2 cm of $H_2O\,m^{-1}$ in U1 and U2). Thus, the breakdown of the biofilm bonds between bed particles, permitted to restore bed aeration and to favor gas/microorganisms exchanges. Consequently, the outlet concentrations of toluene observed over period E remained stable at their pre-agitation levels, around 0.3–0.4 g m^{-3},

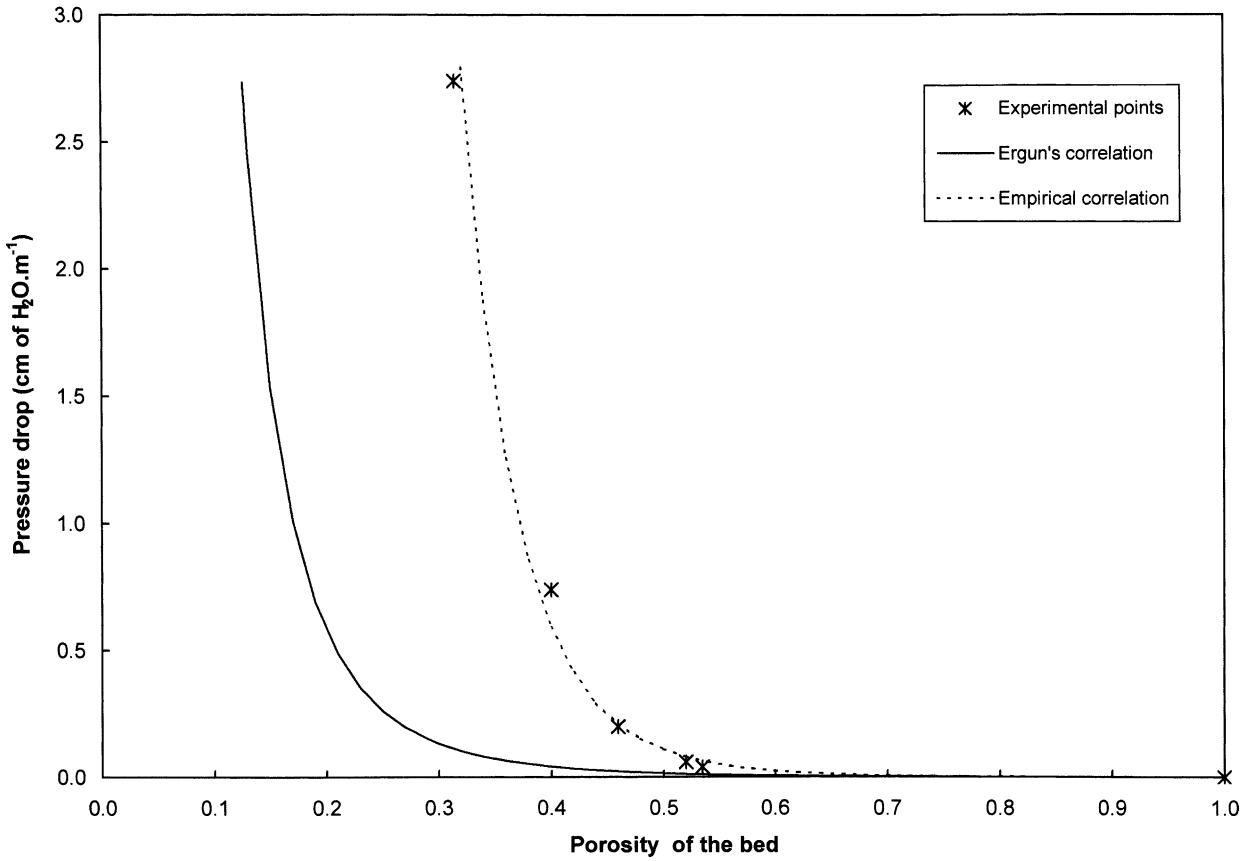

Fig. 7. Influence of the bed porosity on the pressure drop through the filtering medium of biofilter U1.

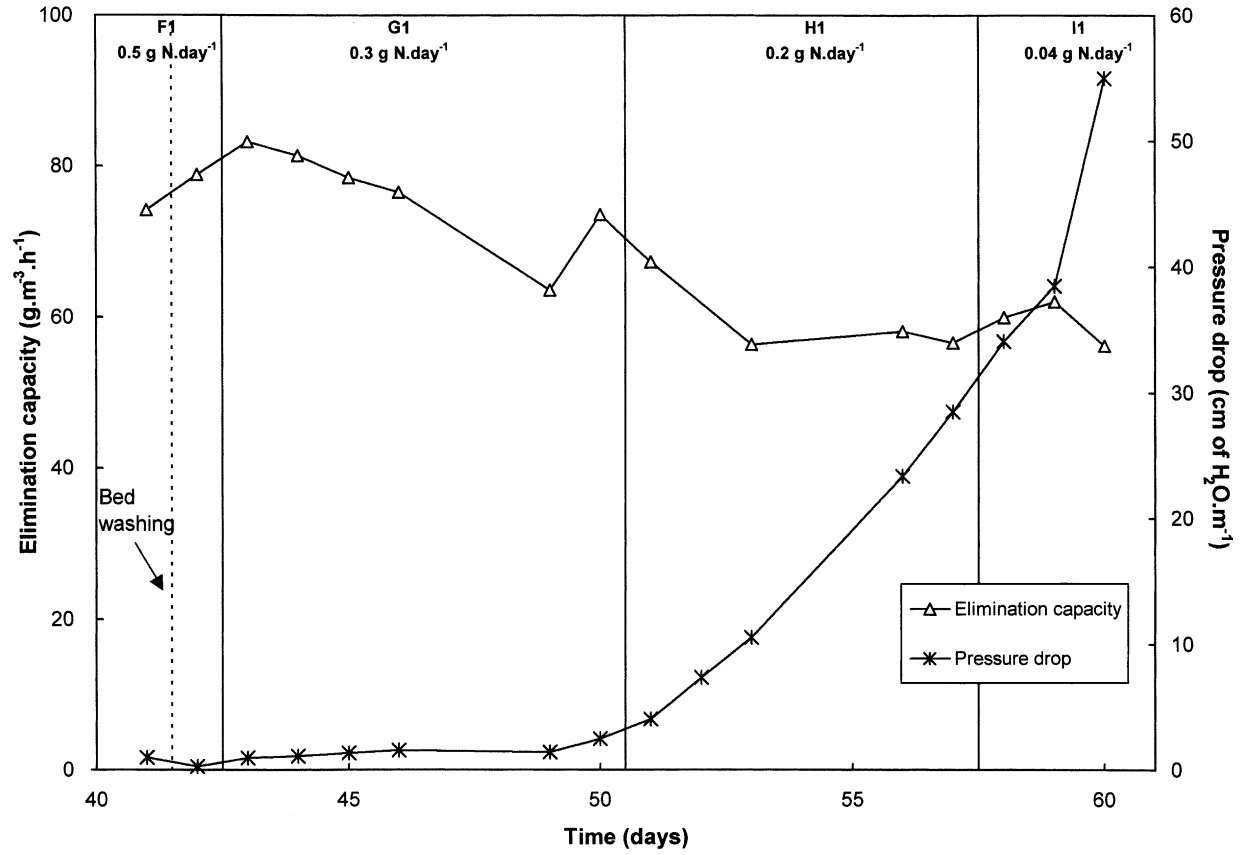

Fig. 8. Evolution of the elimination capacity and the pressure drop in the biofilter U1 after a bed washing. The inlet concentration was constant at 1.9 g m^{-3}.

suggesting that bed stirring could provide a satisfactory treatment for clogging.

3.4.2. Bed washing

A bed washing was operated on day 42 in both biofilters. Figs. 8 and 9 present the variations of elimination capacity and pressure drop with time, in biofilters U1 and U2, respectively. After the bed washing, pressure drop significantly decreased down to negligible values, while elimination capacities rose up to 85 and 105 g m^{-3} h^{-1} in U1 and U2, respectively. These results show that a bed washing constitutes an efficient method for blockage remediation as it permits to remove and drain excess biological matter from the pellets, but still, it remains energetically unfavorable.

3.4.3. Nutrients control

The methods that consist in applying a nutrients (nitrogen) control on the biofiltration system were based on the assumption that the microbial degrading activity is closely linked to the amounts of nutrients (nitrogen in particular) consumed [18,33,35].

3.4.4. Incremental decrease

As illustrated in Fig. 8, after the bed washing, biofilter U1 was submitted to decreasing increments of nitrogen input, from 0.5 to 0.04 g of N day^{-1}, over periods F1 to I1. The objective was to inhibit the biofilm formation, by progressively diminishing the nutrients supply. However, the elimination capacity also seemed to be affected by the lack of nutrients and declined from 85 (F1) to 70 (G1), then to 60 g m^{-3} h^{-1} (H1 and I1). It is to be noted that the elimination capacities observed during the last two periods were similar (60 g m^{-3} h^{-1}), whereas the flow rate of nitrogen during H1 (0.2 g of N day^{-1}) was five times greater than during I1 (0.04 g of N day^{-1}). The unexpected performance achieved in period I1 was certainly due to the fact that part of the nutrients distributed over the previous periods (F1–H1) may have accumulated in the bed and was subsequently utilized by the microorganisms. Moreover, the pressure drop continuously increased during the whole test (from 0.8 to more than 55 cm of H_2O m^{-1}—the measurement limit of the manometer), whereas the nitrogen concentration decreased.

This test shows that a progressive diminution of the nitrogen supply does not constitute a suitable method for preventing biofilter blockage, particularly as this method gives rise to a drastic reduction of the performance.

3.4.5. Alternative nitrogen distribution

After the bed washing, the biofilter U2 was submitted to alternate distribution of nitrogen, as indicated in Table 2.

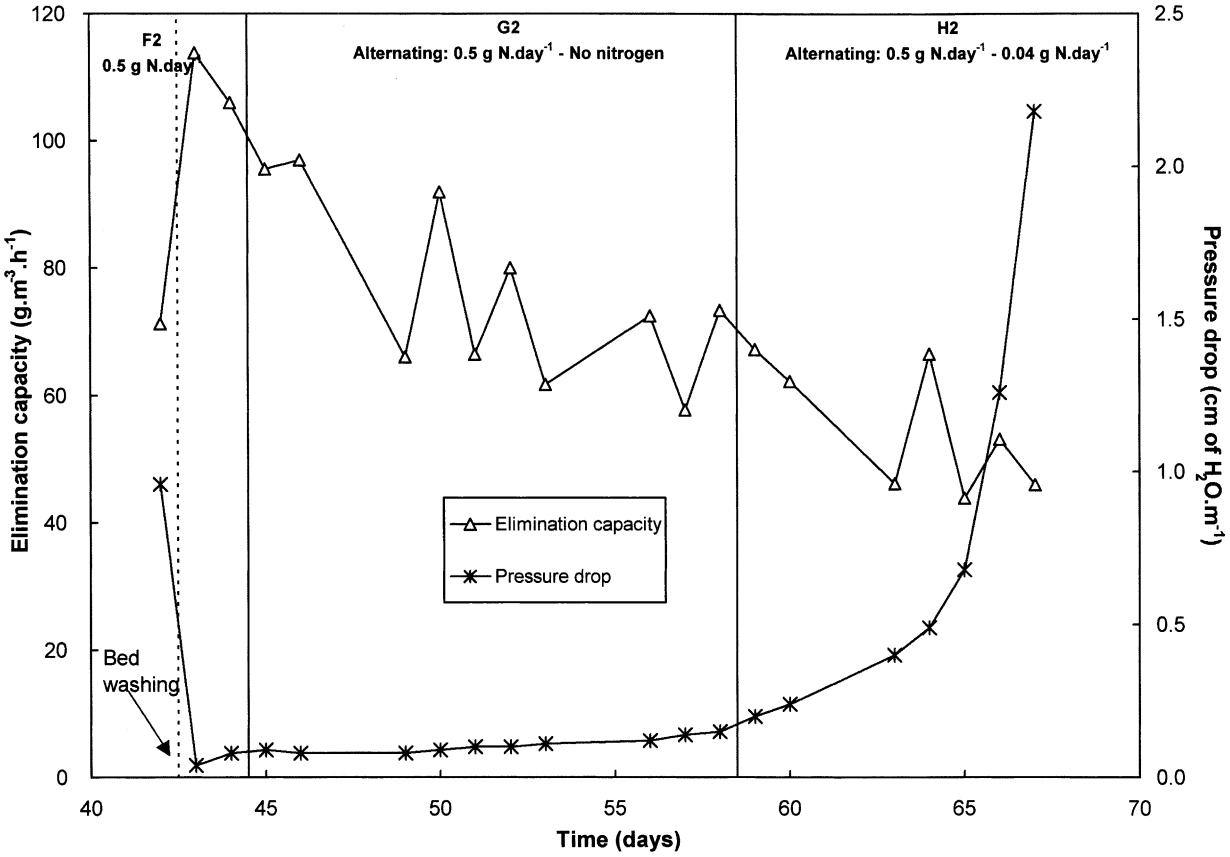

Fig. 9. Evolution of the elimination capacity and the pressure drop for biofilter U2 after bed washing. The inlet concentration was constant at 1.9 g m^{-3}.

The objective of these alternating periods was to promote the degrading activity with the N-enriched solutions and to slow the biomass growth with nitrogen deficiency periods. As presented in Fig. 9, the elimination capacities obtained at the end of period F2 and at the beginning of period G2 were close to 100 g m^{-3} h^{-1}. These good performances resulted from the enriched input of nitrogen (0.5 g of N day^{-1}) and the favorable bed aeration conditions following the bed washing. From period G2 to period H2, the elimination capacity evolved in a cyclic way, according to the 24 h imposed nutrient cycle: decrease the day following low or absence of nitrogen input, and increase after irrigations with N-enriched solutions. Moreover, the maximum values of the peaks, obtained after 0.5 g of N day^{-1}, continuously decreased with time, from 100 down to 75 g m^{-3} h^{-1}. This behavior is in agreement with the fact that nitrogen is essential for microbial activity and that the nutrient resources, either intrinsic to compost or accumulated in the media within 24 h, are not sufficient to maintain a satisfactory degrading activity level. Fig. 9 also shows that pressure drop continuously increased between G2 and H2, from negligible values to 2.2 cm of H_2O m^{-1}. This proves that biofilm formation during a normal biofilter operation cannot be significantly cut-back through periods of reduced nutrients. However, the values for pressure drops obtained in biofilter U2 (<2.2 cm of H_2O m^{-1}) are far lower than those reached in biofilter U1 (>50 cm of H_2O m^{-1}). This indicates that N-starvation periods could constitute a preliminary to prevent biomass formation.

4. Conclusion

In this work, two upflow laboratory-scale biofilters were operated in parallel at constant inlet concentration of toluene (1.9 g m^{-3}) and air flow rate (1.0 m^3 h^{-1}). Variable nitrogen flow rates have been delivered to the biofilters (from 0.0 to 0.5 g N day^{-1}). The optimum elimination capacities (90–95 g m^{-3} h^{-1}) were achieved for N-concentration at near 0.4 g N day^{-1}. Analyses of the results showed that conditions most favorable for toluene biodegradation are equally those that contribute strongly to excess biomass formation and biofilter clogging. It was also shown that pressure drop could be correlated to physical properties such as bed porosity via a modified Ergun's relationship. Moreover, in the course of these experiments, four methods for clogging prevention have been evaluated. Two mechanical methods, bed stirring and bed washing, have been operated, and effectively gave rise to improved performance, through removal of excess biomass. Two chemical methods, based upon nutrients control, have also been tested. In order to create a nutrient deficiency, nitrogen was delivered, either by following a schedule of decreasing flow rates, or by following 24 h cycles: alternating solution enriched with N/solution without or poor in N. However, the resulting performance of the chemical methods reveals that the overall microbial degrading activity was significantly reduced and that the biofilm accumulation could not be avoided through such nutrient controls.

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