



Evaluating the impact of water supply strategies on *p*-xylene biodegradation performance in an organic media-based biofilter

G. Gallastegui^a, R. Muñoz^b, A. Barona^a, G. Ibarra-Berastegi^c, N. Rojo^a, A. Elías^{a,*}

^a Department of Chemical and Environmental Engineering, Engineering Faculty, University of the Basque Country, Alda Urquijo s/n, 48013 Bilbao, Spain

^b Department of Chemical Engineering and Environmental Technology, Valladolid University, Dr. Mergelina s/n, 47011 Valladolid, Spain

^c Department of Nuclear Engineering and Fluid Mechanics, Engineering Faculty, University of the Basque Country, Alda Urquijo s/n, 48013 Bilbao, Spain

ARTICLE INFO

Article history:

Received 31 March 2010

Received in revised form 4 October 2010

Accepted 4 October 2010

Available online 12 October 2010

Keywords:

Biofiltration

Dynamic performance

Moisture content

p-Xylene biodegradation

Fluid mechanics

ABSTRACT

The influence of water irrigation on both the long-term and short-term performance of *p*-xylene biodegradation under several organic loading scenarios was investigated using an organic packing material composed of pelletised sawdust and pig manure. Process operation in a modular biofilter, using no external water supply other than the moisture from the saturated inlet air stream, showed poor *p*-xylene abatement efficiencies ($\approx 33 \pm 7\%$), while sustained irrigation every 25 days rendered a high removal efficiency (RE) for a critical loading rate of $120 \text{ g m}^{-3} \text{ h}^{-1}$. Periodic profiles of removal efficiency, temperature and moisture content were recorded throughout the biofilter column subsequent to each biofilter irrigation. Hence, higher *p*-xylene biodegradation rates were always initially recorded in the upper module, which resulted in a subsequent increase in temperature and a decrease in moisture content. This decrease in the moisture content in the upper module resulted in a higher removal rate in the middle module, while the moisture level in the lower module steadily increased as a result of water condensation. Based on these results, mass balance calculations performed using measured bed temperatures and relatively humidity values were successfully used to account for water balances in the biofilter over time. Finally, the absence of bed compaction after 550 days of continuous operation confirmed the suitability of this organic material for biofiltration processes.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Biofiltration is now one of the most cost-effective and sustainable off-gas treatment technologies due to its minimal energy requirements and low environmental impact [1,2]. Based on its simple operation and high removal efficiencies for both hydrophilic and hydrophobic volatile pollutants, biofiltration is the most popular biotechnologies for the treatment of odours and industrial volatile organic compounds (VOCs) [3,4].

Since their introduction in the 1960s, a considerable amount of research has been conducted to elucidate the mechanisms underlying pollutant removal in biofilters and to enhance their robustness in order to increase their acceptance within the industrial community [5,6]. Most studies have focused on the optimization of relevant factors such as the nature of the packing material, moisture content, biomass growth control, pH, nutrient availability, etc. [7–10]. Due to their biological basis, the control of moisture content (water activity) in biofilters is a key operating parameter determining process performance [6]. As a matter of fact, an inefficient control of the

moisture content has been reported as the cause of 75% of biofilter failures [11].

Water activity is responsible for the type (bacteria vs. fungi) and level of activity of the microbial community present, and determines the long-term structural stability of the packed bed (compaction, formation of anaerobic zones or preferential pathways, etc.) [12]. However, the control of moisture content in the packing material is compulsory and complex, as a large number of factors need to be considered: the moisture content of the gas stream entering the biofilter, the frequency and flow of external irrigation, the exothermicity of pollutant mineralization, the organic nature of the packing material, the biomass distribution profile within the biofilter and the water retention capacity of each packing material [3,13]. Water activity in biofiltration systems must therefore be carefully studied on a case-by-case basis in order to ensure consistent VOC and odour treatment efficiencies, especially when using novel organic packing materials. However, despite the importance of this issue, there are few published studies devoted to understanding and further optimizing the temporal and spatial distribution of moisture content in biofilters [11,13,14].

This work focuses the influence of different irrigation and operating strategies on the performance of *p*-xylene abatement using an organic packing material composed of pelletised sawdust and pig manure. Special attention was given to the interdependence

* Corresponding author. Tel.: +34 94 601 4087; fax: +34 94 601 4179.
E-mail address: ana.elias@ehu.es (A. Elías).

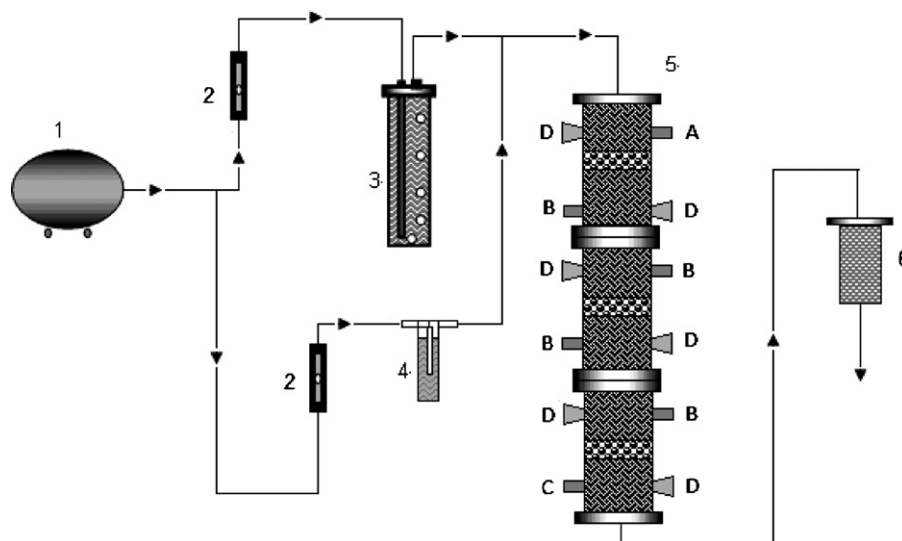


Fig. 1. Diagram of the biofiltration system: 1, air-compressor; 2, flow meter; 3, humidification chamber; 4, p-xylene evaporator; 5, modular biofilter; 6, active carbon chamber; A (inlet gas sampling valve); B (inter-module gas sampling valve); C (outlet gas sampling valve) and D (temperature and air relative humidity measurement ports).

between moisture content and pollutant removal efficiency by carefully monitoring the timeline of the profiles of both parameters throughout the biofilter column. p-Xylene, was used as a model VOC due to its widespread use in the ever-increasing production of polyethylene terephthalate [15].

2. Materials and methods

2.1. Inoculum preparation

Despite the indigenous microflora present in the packing material used in this work had previously shown a good capacity for degrading H_2S [16], it was not capable of mineralizing p-xylene. Thus, an aerobic activated sludge collected at the wastewater treatment plant in Muskiz (Bizkaia, Spain) was used as inoculum for p-xylene biodegradation. Initially, the activated sludge was continuously exposed for 30 days in a stirred tank bioreactor to a p-xylene-laden air at an aeration rate of 0.4 vvm (air volume per unit of liquid volume per minute) and at concentrations ranging from 50 to 100 ppm_v. The measurement of p-xylene degradation rate in batch assays confirmed the acclimatization of the microbial population present in the liquid phase. This procedure has already been explained in a previous work [17].

2.2. Biofilter setup

The biofiltration system consisted of 3 PVC modules (Fig. 1). The packed bed was divided into the three identical sections with a total volume of 4.5 L. The packing material selected in this work was supplied by SLIR S.L (Specialised Engineering in Recycling Agricultural Residues) and its commercial name was ABONLIR. This material was made up of composted pig manure and sawdust, and the pellets were manufactured by mechanical compression without chemical addition. The compost was stored in sealed plastic bags at room temperature to maintain its original moisture content. Table 1 summarizes the main characteristics of the packing material [16]. The biofilter was initially irrigated with an activated sludge acclimated according to the procedure published by Elías et al. [17] and it was operated in a downflow configuration at 23 ± 2 °C. The flow of p-xylene-contaminated air was added from the top of the biofilter at a flow rate of 1–1.5 L min⁻¹ (corresponding to an empty bed residence time ranging from 180 to 270 s) and the contaminated flow was generated by mixing a p-xylene-saturated air stream with a

humidified p-xylene-free air stream in different proportions. The non-humidified fraction of air used for p-xylene saturation was a minor fraction of the whole influent air flow and did not significantly impact the moisture of the contaminated air stream entering the biofilter. Indeed, the relative humidity of the contaminated air at the biofilter inlet remained always higher than 98%. An activated carbon filter was also included in the experimental setup, being fitted to the bioreactor outlet in order to mitigate the environmental impact of the non-degraded contaminant. The biofilter was equipped with several gas sampling valves to monitor the inlet, outlet and inter-module p-xylene and CO₂ concentrations. Additionally, it was also provided with several ports located throughout the three PVC modules for measuring the temperature and relative humidity content of the air.

2.3. Influence of water irrigation on the long-term performance of p-xylene biodegradation

The system was initially operated for 260 days with no further irrigation, as water was supplied solely via the saturated inlet air stream (relative humidity $\geq 98\%$) (dry period). From days 260 to 550 (wet period), each of the 3 modules was individually irrigated every 25 days with 400 mL of mineral salt medium [18], in order to avoid drip of water from the top section to the middle and bottom sections.

Table 1

Physical–chemical properties of the packing material used in the modular biofiltration unit [16].

Organic matter (%) ^a	40.0
Total nitrogen (%)	1.4
P ₂ O ₅ (%)	1.1
K ₂ O (%)	1.5
Total S (%)	3.3
pH	6.5–7.5
Mean length (mm)	10.7
Mean radius (mm)	6.1
Bulk density (g cm ⁻³)	1.3
Real density (g cm ⁻³)	2.3
B.E.T. (N ₂) surface area (m ² g ⁻¹)	12.06 ± 0.09
BJH adsorption average pore diameter (Å)	145
Macropore volume in the pellets (%)	89.42
Micropore area (d < 20 Å) (m ² g ⁻¹)	0.3
Initial moisture content (%)	25.2

^a Value provided by the manufacturer.

During the dry period, the biofilter was continuously fed with the contaminant at an inlet loading rate (IL) of $8 \pm 1 \text{ g m}^{-3} \text{ h}^{-1}$. On the other hand, *p*-xylene was continuously supplied during the wet period, and the different inlet loading rates during this period were 16 ± 4 ; 96 ± 10 ; 30 ± 5 ; 72 ± 10 and $166 \pm 19 \text{ g m}^{-3} \text{ h}^{-1}$, respectively. Apart from monitoring the overall biofilter biodegradation performance, the elimination capacity and moisture content of each individual module were also periodically recorded. Samples of the packing material were collected from the 3 modules during the dry and wet periods in order to monitor the dynamics of microbial population by Scanning Electron Microscopy (SEM).

2.4. Influence of water irrigation on short-term biofilter and module performance

The transient performance of the overall *p*-xylene removal and the temporal and spatial dynamics of moisture content (MC), temperature and partial elimination capacities in each module were evaluated after each periodic irrigation during the wet period.

2.5. Analytical procedure

p-Xylene and CO_2 concentrations were measured using a micro-gas chromatograph CP 4900 (Varian, The Netherlands) equipped with auto-sampling injection, a TCD detector and using He as carrier gas. The Micro GC was equipped with CP-Sil 5 CB ($6 \text{ m} \times 0.15 \text{ mm} \times 2 \mu\text{m}$) and CP PoraPLOTQ ($10 \text{ m} \times 0.25 \text{ mm} \times 8 \mu\text{m}$) columns. The oven, injector and TCD detector were maintained at 80°C , 110°C and 80°C , respectively. External standards prepared from a calibration cylinder (Air Liquide, Spain) containing a *p*-xylene in N_2 enabled the quantitative determination of the target VOC.

The temperature of the packing material was determined by using several thermocouple HI-98804 sensors (Hanna Instruments, Italy) located at six different heights in the biofilter column (Fig. 1). The moisture content within the packing material was determined by a Moisture Analyzer HB43-S (Mettler Toledo) by periodically collecting 2–3 g of support material from each module. Air relative humidity and temperature at the inlet and outlet ports of the biofilter column were measured by using a Testo 625 sensor (Testo, Germany).

Biomass samples extracted from the packing material were fixed in 2% glutaraldehyde in 0.1 M cacodylate buffer (pH 7.4), washed in iso-osmolar cacodylate/sucrose buffer and postfixed in 1% osmium tetroxide in cacodylate buffer. After repeated washing, samples were dehydrated through an ethanol series and washed in hexamethyldisilazane prior to air drying. Finally, samples were mounted onto stubs and gold coated using a JEOL fine coat ion sputter JFC-1100. Samples were visualized and micrographed using a scanning electron microscope (Hitachi S-4800) at 15 kV accelerating voltage.

2.6. Statistical treatment

All results are given as the average value with its corresponding error at 95%. Results were analyzed using a one-way ANOVA with significance at $P \leq 0.05$. The Excel statistical package (Microsoft Corporation, USA) was used for data processing.

3. Results and discussion

3.1. Influence of water irrigation on the long-term performance of *p*-xylene biodegradation

During the “dry period” (the first 260 days of operation), process performance was characterised by an average removal efficiency (RE) of $33 \pm 7\%$ (Fig. 2a). This poor performance was recorded

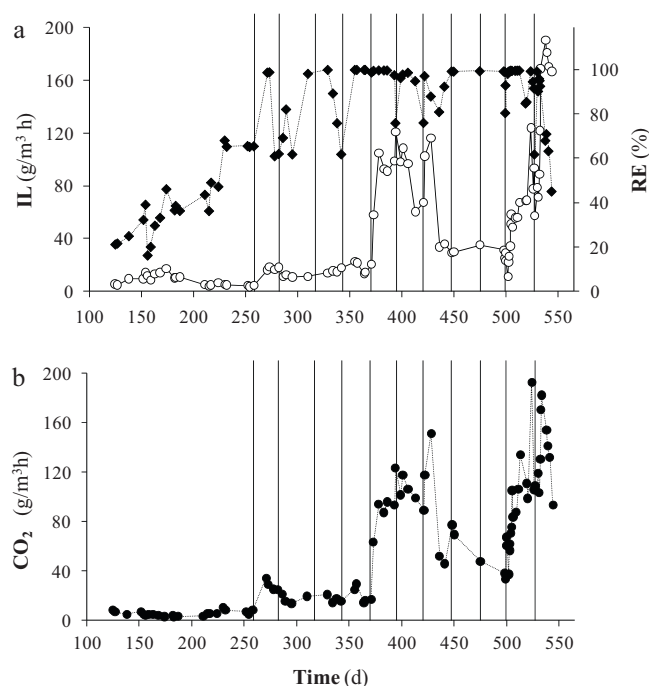


Fig. 2. Evolution of inlet load (○), removal efficiency (◆), and CO_2 production (●) during *p*-xylene biofiltration under dry (from days 0 to 260) and wet conditions (from days 260 to 550). Vertical lines represent biofilter irrigation.

despite the low inlet loading rate of contaminant ($8 \pm 1 \text{ g m}^{-3} \text{ h}^{-1}$) and the long EBRT, which were fairly higher than EBRT used by other authors (Table 2). This long period resulted in a functionally-stable microbial community. The modular biodegradation profile throughout the biofilter column revealed that only the upper module, which received readily available moisture from the inlet stream, was active (data not shown).

Periodic irrigation of the biofilter packing material led to a rapid increase in *p*-xylene removal efficiency, which achieved a sustained RE of $88 \pm 15\%$ from days 259 to 373 (at an inlet loading rate $16 \pm 10 \text{ g m}^{-3} \text{ h}^{-1}$). The irrigated biofilter was also capable of coping with a further increase in IL (days 378 to 428) to $97 \pm 18 \text{ g m}^{-3} \text{ h}^{-1}$, recording a RE of $94 \pm 9\%$. Similar results (an average RE higher than 93%) were achieved during operation from days 436 to 505 at an inlet loading rate of $30 \pm 9 \text{ g m}^{-3} \text{ h}^{-1}$ and from days 505 to 532 at $72 \pm 20 \text{ g m}^{-3} \text{ h}^{-1}$. However, a sudden increase in pollutant loading rate to $166 \pm 24 \text{ g m}^{-3} \text{ h}^{-1}$ resulted in a gradual deterioration of biofilter performance (Fig. 2a), probably due to a partial inhibition of the microbial community present in the biofilm [19,20]. These results therefore confirmed the poor abatement performance reported in uncontrolled biofilters, where the moisture content provided by the humidified air stream might be insufficient to maintain an optimum moisture level for microbial growth [11].

Finally, it must be highlighted that despite the low frequency of irrigation (each 25 days), the good stability of the overall reactor performance within these 25 days without irrigation was in agreement with Son and Striebig [21], who recorded consistent REs greater than 90% in an organic biofilter for 59 days without any nutrient solution or water addition.

The CO_2 production rates recorded also confirmed the higher biofilter performance under periodic irrigation (Fig. 2b). Hence, while CO_2 production and *p*-xylene removal rates had a poor correlation during the dry period ($R^2 \approx 0.13$), a reasonable correlation was obtained during the wet period ($R^2 \approx 0.80$). The reasons underlying this apparent mismatch are still not clear, but they confirmed the dysfunctions of microbial metabolism at low water activities. An overall carbon balance applied to *p*-xylene mineralization

Table 2
Characteristics of conventional biofilters treating p-xylene.

Packing material	EC _{MAX} (g m ³ h ⁻¹)	EBRT (s)	Moisture (%)	Reference
Pig manure–sawdust	130 (>95% RE)	180–270	25–35	This study
Conditioned peat	66 (IL ≥ 200 g m ³ h ⁻¹)	102	50–70	[34]
Peat balls	67 (max)	157	–	[19]
Conditioned peat	236 (66% RE)	68	16–70	[35]
Compost–wood chip	73 ± 14 (max.)	60	60–65	[36]
Peat + Mineral additive with good binding capacity 70:30 (v/v)	61(93% RE)	150	–	[37]
Manure pig compost–forest soil – polyethylene	80 (>96% RE)	132	50	[38]
Biosol (glass and cardboard)	160 (>90% RE)	47	45	[39]
Food waste compost + oyster shell + polyurethane foam	21	60	40–60	[40]
Commercial press mud (sugar industry waste)	67	42	50	[41]
Wood-chip (fungal inoculum)	77 (53% RE)	59	–	[29]
Wood-chip (bacterial inoculum)	58 (49% RE)	59	–	[29]
Scoria	450 (66% RE)	45	–	[42]
Sieved compost + Ceramic balls	101.3 (49% RE)	48.6	55–60	[43]

during the wet period revealed that only 32% of the p-xylene removed was converted into CO₂. This unexpectedly low conversion is however similar to that obtained by García-Peña et al. [22], who also recorded p-xylene conversion to CO₂ of 31%. It is however unlikely that the remaining carbon was totally transformed into biomass, since the production of extracellular metabolites was not analyzed in this work and it might cause a deficit in CO₂ in the gas phase [19]. The low values of CO₂ concentration recorded at the biofilter outlet suggest that the organic packing bed was not actively mineralized during the wet period, which agrees with the high stability of the packing material structure even after 550 days of continuous operation. This experimental result agreed well with the carbon distribution determined during toluene mineralization by *Pseudomonas* sp. [23].

It should be noted that pressure drop over the course of the experimental run was negligible (below 3 cm H₂O), which con-

firmed the stability of this organic packing material. This stability, together with the cost (€130 m⁻³ compared to €475 m⁻³ for activated carbon or €225 m⁻³ for polystyrene foams) makes this packing material an interesting alternative for biofilter scale-up [24].

A comparison between the SEM images of the packing material at day 250 (dry period) (Fig. 3a and b) and day 427 (wet period) (Fig. 3c and d) showed significant differences in the nature and density of the microbial population developed. Hence, while low concentrations of filamentous fungi were present in the filter bed after 8 months of operation in the absence of irrigation, a varied and abundant microbial community composed of both bacteria and fungi was clearly visible after periodic irrigation. These findings are in agreement with previous results on p-xylene biofiltration under xerophilic conditions when using the same pelletised carrier material; an abundant fungal biodiversity was observed by culture-

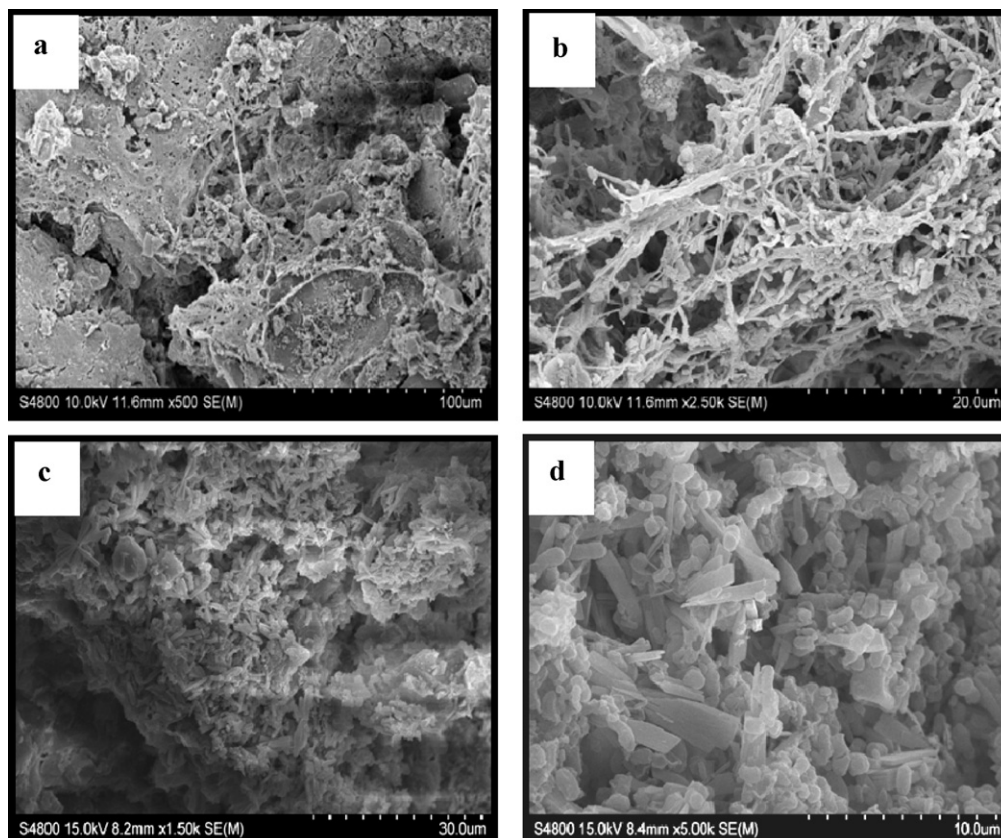


Fig. 3. SEM photographs of packing media under dry (a, b) and wet (c, d) conditions.

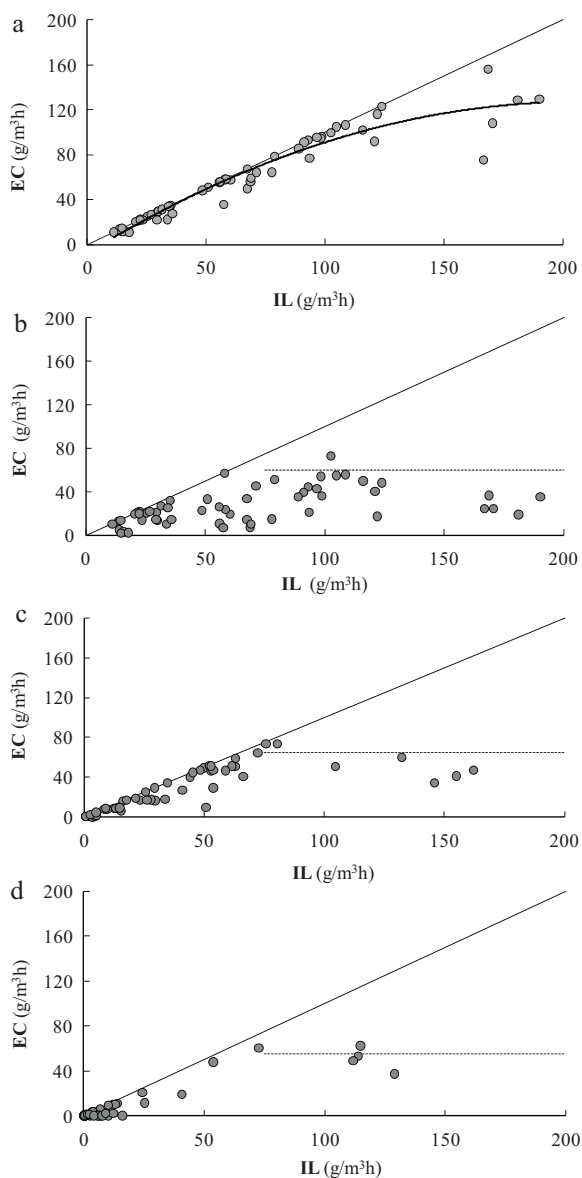


Fig. 4. Elimination capacity versus inlet load for (a) the overall biofilter, (b) upper module, (c) middle module, and (d) lower module during *p*-xylene biofiltration in the wet period.

independent molecular methods [25]. Similarly, Cox et al. [14] and Auria et al. [26] observed a preferential growth of fungi under prolonged periods of low water activity in the biofiltration of styrene and toluene, respectively [14,26].

The overall elimination capacity (EC) of the biofilter as a function of the inlet *p*-xylene loading rate during the wet period is shown in Fig. 4a, while Fig. 4b–d show the EC for the upper, middle and lower modules, respectively, as a function of their corresponding IL. Overall *p*-xylene EC increased at increasing loading rates with RE of approximately 100% up to a critical IL of $120 \text{ g m}^{-3} \text{ h}^{-1}$, where EC remained constant. A detailed analysis of the data confirmed that *p*-xylene diffusion from the gas phase governed biofilter performance under these particular operating conditions. Thus, when loading rates were lower than $120 \text{ g m}^{-3} \text{ h}^{-1}$, the *p*-xylene transferred from the gas phase was rapidly consumed in the outer layers of the biofilm, resulting in an insufficient supply of carbon to the internal layers of the biofilm (adjacent to the packing media). However, when the *p*-xylene IL was higher than $120 \text{ g m}^{-3} \text{ h}^{-1}$, microbial activity, rather than pollutant mass transport, controlled biofil-

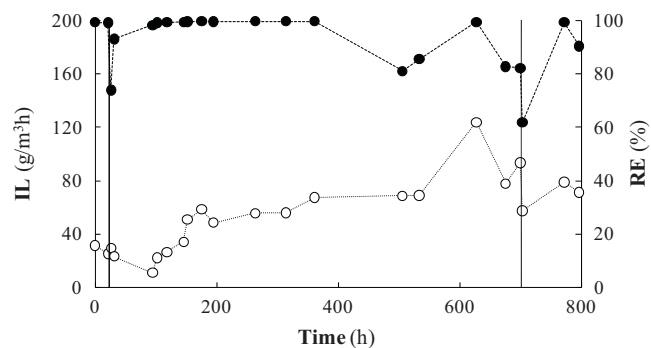


Fig. 5. Short-term transient effect on *p*-xylene removal efficiency (●) after one water irrigation and during the subsequent 25 days. Open circles represent the inlet loading rate.

ter performance. In addition, it should be noted that the highest EC recorded in this study in most cases exceeded the maximum EC obtained by other researchers under comparable experimental conditions, which can be attributed to the high B.E.T. surface area of the packing material (Table 1). Nevertheless, the overall moisture content in our experimental system (25–35%) was slightly lower than those reported in the literature (>40–65%) (Table 2). This high efficiency at lower moisture contents might be explained by the selective pressure placed on microbial enrichment by the initial 8 months of dry period, which rendered a microbial community that was resistant and acclimated to low water activities.

A similar *p*-xylene maximum EC was observed in each of the three modules as shown in the EC vs. IL in Fig. 4b–d, where IL represents the *p*-xylene loading rate entering each individual module during wet period. Despite this similarity, the middle module recorded the most stable performance (with almost complete VOC depletion up to a loading rate of $65 \text{ g m}^{-3} \text{ h}^{-1}$), while the upper module surprisingly showed a rather scattered biodegradation pattern. On the other hand, the third module showed a poor process performance at low IL (Fig. 4d) although comparable performances were recorded above $25 \text{ g m}^{-3} \text{ h}^{-1}$. A detailed analysis of the correlations between *p*-xylene RE and the moisture content in each individual module revealed only a clear correlation in the upper module ($R^2 = 0.82$, in the range 7–32%), while no significant variations in RE were recorded at increasing moisture contents in the middle and lower modules (25–40%) (data not shown, $R^2 < 0.4$). These findings do not rule out a potential correlation between microbial activity and MC, rather than limit the MC range of this correlation. Hence, there was a critical MC (25–30%) below which microbial activity depended on the MC of the packing bed, but above this critical MC, water activity was sufficient to support microbial activity and no correlation between MC and RE was observed.

3.2. Influence of water irrigation on short-term biofilter and module performance

Biofilter performance after one water irrigation and during the subsequent 25 days is shown in Fig. 5. Steady state performance rapidly recovered a few hours after irrigation. This finding was in agreement with that of Auria et al. [26], who explained a similar slight RE decrease based on the rapid desorption of the VOC from the packing material. Likewise, an increase in odour concentration in the area surrounding intensive livestock farming facilities is usually found immediately after rainfall due to odourant VOC desorption (flooding-out effect) [27]. On the other hand, the increase in air velocity (due to pore blocking), together with the increase in the thickness of the water layer surrounding the biofilm, might have

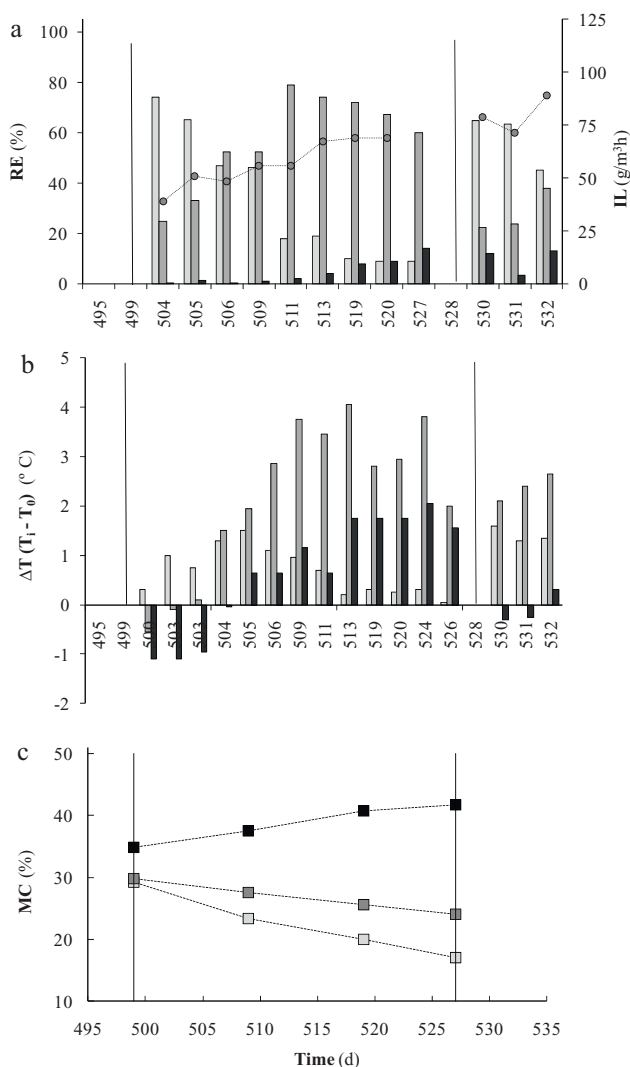


Fig. 6. Evolution of RE (a), temperature variation (b) and moisture content (c) in the upper (□), middle (■) and lower (●) modules over time during p-xylene biofiltration. Vertical lines represent biofilter irrigation and (●) represents the inlet loading rate.

exulted in a higher resistance to the mass transfer of hydrophobic pollutants like p-xylene [28].

Consistent dynamics for RE and MC were recorded in each module during the wet period, although the temperature patterns slightly differed at the beginning of each irrigation period as shown in Fig. 6b. Contaminant biodegradation occurred predominantly in the upper zone during the first week following the irrigation of the three modules (Fig. 6a). Nevertheless, RE in the upper module gradually declined and the RE in the middle module gradually increased within the first 12 days. A steady increase in RE in the lower module from day 12 was observed. The fluctuations in temperature were always correlated with p-xylene biodegradation in each module (Fig. 6b). The temperatures in the middle module increased up to 4 °C by days 13 and 14 after irrigation, while the temperature in the lower zone gradually increased from the first day and obviously decreased with the next irrigation. This close correlation between biofilter's performance and temperature was also observed by Jorio et al. [29]. In accordance with this study, Son et al. [30] showed that the ethylbenzene was not equally degraded over the depth of a biofilter packed with composting material after water irrigation. Hence, the most dominant stage in ethylbenzene degradation gradually shifted over time from

the nearest air-inlet zone to the inner section of the biofiltration units. Finally it must be highlighted that the above-mentioned consistency in RE, temperature variations and MC refers to the trend of these parameters rather than to the quantitative extent of these variations, which itself depends on p-xylene biodegradation rate. It is clear from Fig. 6 than the higher IL on day 528 (compare to day 499) induced higher temperature increases in the modules as a result of a more intense microbial activity. In this context, Son et al. [30] also observed that higher ethylbenzene degradation rates induced a higher temperature increase in biofiltration packed beds [30]. On the other hand, the MC in the upper and middle modules progressively declined from 30% to 17% and 24%, respectively, while it increased in the lower module from 35% to 42% (Fig. 6c). These dynamics in MC can be explained as a result of water evaporation due to a biodegradation-mediated temperature increase. As a result, the unsaturated polluted air was capable of evaporating water from the packing material. The lower temperatures prevailing in the lower module induced later condensation, which explains the increase in moisture content recorded. These MC profiles agreed with those reported by Abumaizar et al. [31], who observed higher MC contents in the lower modules of BTEX treating biofilters packed with activated carbon. An optimum MC range of 20–40% was estimated from the correlation of the dynamic profiles of p-xylene RE and moisture content. This finding confirms the results of Wang and Govind (1997) who reported that naturally bioactive filter beds (like peat or compost) need a range of suitable water content for high-efficiency operation [32].

3.3. Water balance modelling

A simple mathematical model was developed for estimating the dynamics of moisture content within the different modules of the biofilter. This modelling approach was based on the separate contribution of water production during p-xylene mineralization and water evaporation/condensation as a result of the temporal and spatial dynamics of temperature within the biofilter bed.

Thus, the rate of water evaporation from the biofilter (W_{ev}) can be calculated using the theoretical considerations proposed by Morales et al. [33]:

$$W_{ev} = \frac{(y_{IN} \rho_{IN} - y_{OUT} \rho_{OUT})F_{air}}{V_r} \quad (1)$$

where ρ is the air density, F_{air} is the volumetric air flow rate, V_r is the volume of the packing bed and y_i is the mass fraction of water contained in the air circulating through the biofilter calculated as follows:

$$y_i = 0.622 \frac{pv_i}{p - pv_i} \quad (2)$$

The water partial pressure (pv_i) can be estimated as:

$$pv_i = RH_i Pvs_i \quad (3)$$

where Pvs_i is the water saturation pressure expressed as:

$$Pvs_i = e^{(25.775 - 5281.1/T_i)} \quad (4)$$

Moreover, air density can be expressed using Eq. (5):

$$\rho_i = \rho_0 \left(\frac{T_0}{T_i} \right) \left(\frac{p}{p_0} \right) \quad (5)$$

Water production from p-xylene mineralization can be calculated by experimental stoichiometric considerations. Therefore, assuming a biomass composition of $CH_{1.8}N_{0.2}O_{0.5}$ and based on an experimental CO_2 production yield of 1.24 g CO_2 /g p-xylene

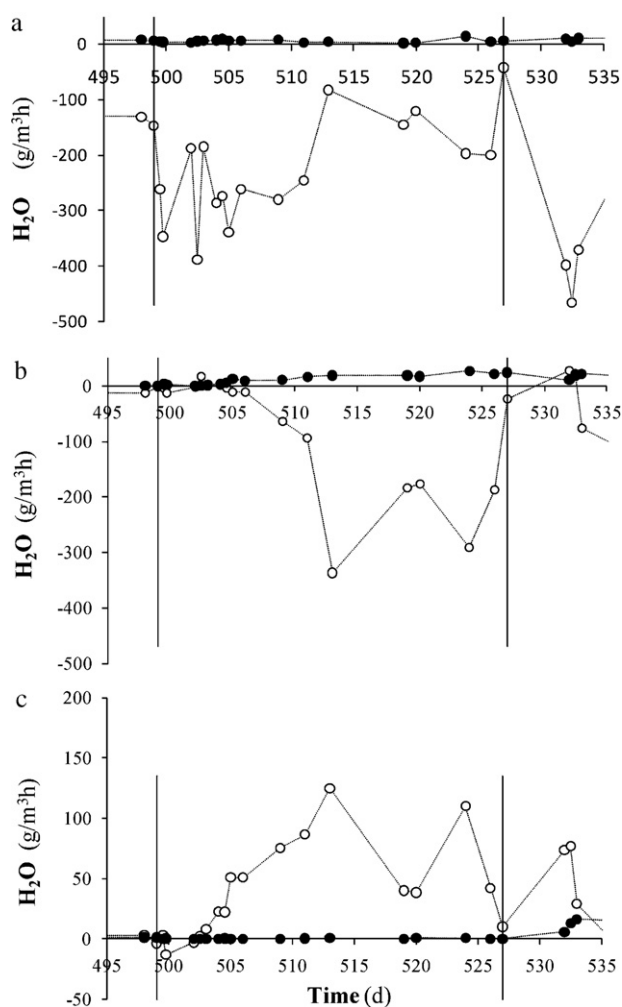
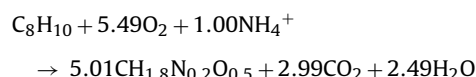


Fig. 7. Evolution of evaporated water (○) and biochemically produced water (●) in the upper (a), middle (b) and lower (c) modules over time.

degraded, the stoichiometry of *p*-xylene mineralization can be represented by the following equation:



The model developed here accurately described the water mass balance within each module. Thus, when applied to the experimental period from days 495 to 435, this model confirmed the very different amounts of evaporated water and biochemically produced water in the upper and middle modules (Fig. 7a and b). In addition, the model was able to explain the increase in MC recorded in the lower module (Fig. 7c). Finally, the goodness of estimation of the mathematical approach proposed here was assessed using the abovementioned experimental period. Accordingly, based on Fig. 6c, the amount of water loss through evaporation in the upper, middle and lower modules accounted for 238, 112, and $-132 \text{ g H}_2\text{O}$, respectively and model predictions based on Fig. 7a–c accounted for 201, 128, and $-60 \text{ g H}_2\text{O}$.

4. Conclusions

Biofiltration operation in the absence of water irrigation resulted in poor *p*-xylene abatement efficiencies ($\approx 33 \pm 7\%$) even in the presence of filamentous fungi, which are known to improve the absorption of hydrophobic compounds (due to their aerial

mycelium) and to maintain their biodegradation activity under low moisturized environment.

On the other hand, a periodic irrigation strategy rendered a considerable increase in the overall RE (about 100%) for an inlet loading rate lower than $120 \text{ g m}^{-3} \text{ h}^{-1}$ which confirmed previous studies on this topic. These results were confirmed by the poor correlation between CO_2 production and *p*-xylene removal at low water activities, and by the absence of a varied and abundant microbial population during the dry period as shown by the SEM images. A temporary decrease in process performance occurred during the first hours after each irrigation, which was attributed to a water-induced desorption of *p*-xylene from the packing material or a higher water-mediated resistance to mass transfer from the gas phase. Consistent dynamic patterns for RE, T and MC were recorded throughout the biofilter column after irrigation (approximately 300 days). Thus, *p*-xylene biodegradation initially occurred in the upper module, resulting in a temperature increase and a consequent decrease in the moisture content. The decline in moisture content in the upper module gradually led to a higher degradation rate in the second module, while the moisture in the lower module steadily increased as a result of water condensation due to the low removal efficiency. Additionally, mass balance calculations performed using bed temperatures and relative humidity values were successfully used to account for water balances in the biofilter over time. The negligible pressure drop ($<3 \text{ cm H}_2\text{O}$) and the absence of bed compaction after 550 days of continuous operation confirmed the suitability of the pelletized pig manure and sawdust material as packing material in biofiltration processes.

Acknowledgements

This research was supported by the Spanish Ministry of Education and Science (project CTM 2006-02460 with FEDER funds, and contract RYC-2007-01667). The authors are also grateful to the University of the Basque Country for financially supporting the research group (GIU08/10) and to SGIker (UPV/EHU) and Dr. Francesc Prenafeta (Giro Centre Tecnològic) for the technical and human support provided.

References

- [1] M.C. Delhoméie, L. Bibeau, M. Heitz, A study of the biofiltration of high-loads of toluene in air: carbon and water balances, temperature changes and nitrogen effect, *Can. J. Chem. Eng.* 83 (2005) 153–160.
- [2] S. Revah, J.M. Morgan-Sagastume, Methods of odor and VOC control, in: Z. Shareefdeen, A. Singh (Eds.), *Biotechnology for Odor and Air Pollution Control*, Springer-Verlag, New York, 2005.
- [3] C. Kennes, Biofiltration of waste gases from a dairy industry, in: C. Kennes, M.C. Veiga (Eds.), *Bioreactors for Waste Gas Treatment*, Kluwer Academic Publishers, Dordrecht, 2001.
- [4] N.J.R. Kraakman, New bioreactor system for treating sulphur- or nitrogen-compounds, in: C. Kennes, M.C. Veiga (Eds.), *Bioreactors for Waste Gas Treatment*, Kluwer Academic Publishers, Dordrecht, 2001.
- [5] A. Elías, A. Barona, A. Arreguy, J. Rios, I. Aranguiz, J. Peñas, Evaluation of a packing material for biodegradation of H_2S and product analysis, *Proc. Biochem.* 37 (2002) 813–820.
- [6] I. Datta, D.G. Allen, Biofilter technology, in: Z. Shareefdeen, A. Singh (Eds.), *Biotechnology for Odor and Air Pollution Control*, Springer-Verlag, New York, 2005.
- [7] F. Gaudin, Y. Andres, P. Le Cloirec, Packing material formulation for odorous emission biofiltration, *Chemosphere* 70 (2008) 958–966.
- [8] I. Ortiz, I. Garcia-Peña, P. Christen, S. Revah, Effects of inoculum type, packing material and operating conditions on pentane biofiltration, *Chem. Biochem. Eng. J.* 22 (2) (2008) 179–184.
- [9] Y. Jin, L. Guo, M.C. Veiga, C. Kennes, Optimization of the treatment of carbon monoxide-polluted air in biofilters, *Chemosphere* 74 (2009) 332–337.
- [10] K. Singh, R.S. Singh, B.N. Rai, S.N. Upadhyay, Biofiltration of toluene using wood charcoal as the biofilter media, *Bioresour. Technol.* 101 (2010) 3947–3951.
- [11] R. Auria, A.C. Aycaguer, J. Devigny, Influence of water content on degradation rates for ethanol in biofiltration, *J. Air Waste Manage. Assoc.* 48 (1998) 65–70.
- [12] R. Corsi, L. Seed, Biofiltration of BTEX: media, substrate, and loading effects, *Environ. Prog.* 14 (3) (1995) 151–158.

- [13] T. Sakuma, T. Hattori, M. Deshusses, The effects of a lower irrigation system on pollutant removal and on the microflora of a biofilter, *Environ. Technol.* 30 (6) (2009) 621–627.
- [14] H.H.J. Cox, F.J. Magielsen, H.J. Dodderma, W. Harder, Influence of the water content and water activity on styrene degradation by *Exophiala jeanselmei* in biofilters, *Appl. Microbiol. Biotechnol.* 45 (1996) 851–856.
- [15] METI, Ministry of Economy, Trade and Industry of Japan, Forecast of Global Supply and Demand Trends for Petrochemical Products, 2005. <http://www.meti.go.jp/english/information/downloadfiles/c0517PetroProe.pdf> (accessed 10.12.09).
- [16] A. Barona, A. Elías, A. Amurrio, I. Cano, R. Arias, Hydrogen sulphide adsorption on a waste material used in bioreactors, *Biochem. Eng. J.* 24 (2005) 79–86.
- [17] A. Elías, A. Barona, G. Gallastegui, N. Rojo, L. Gurtubay, Preliminary acclimation strategies for successful start-up in conventional biofilters, *J. Air Waste Manage. Assoc.* 60 (8) (2010) 959–967.
- [18] A. Barona, A. Elías, I. Cano, A. Uriarte, J. Artetxe, Additional determinations in a potential support material for toluene biofiltration: adsorption and partition in the nutrient solution, *Chem. Biochem. Eng. Q.* 21 (2) (2007) 151–157.
- [19] H. Jorio, L. Bibeau, G. Viel, M. Heitz, Effects of gas flow rate and inlet concentration on xylene vapours biofiltration performance, *Chem. Eng. J.* 76 (2000) 209–221.
- [20] C. Gabaldón, V. Martínez-Soria, M. Martín, P. Marzal, J.M. Penya-Roja, F.J. Alvarez-Hornos, Removal of TEX vapours from air in a peat biofilter: influence of inlet concentration and inlet load, *J. Chem. Technol. Biotechnol.* 81 (2006) 322–328.
- [21] H.K. Son, B.A. Striebig, Ethylbenzene removal in a multiple-stage biofilter, *J. Air Waste Manage. Assoc.* 51 (2001) 1689–1695.
- [22] I. García-Peña, I. Ortiz, S. Hernández, S. Revah, Biofiltration of BTEX by the fungus *Paecilomyces variotii*, *Int. Biodeterior. Biodegrad.* 62 (2008) 442–447.
- [23] R. Muñoz, M. Hernández, A. Segura, J. Gouveia, A. Rojas, S. Villaverde, Continuous cultures of *Pseudomonas putida* mt-2 overcome catabolic function loss under real case operating conditions, *Appl. Microbiol. Biotechnol.* 83 (1) (2009) 189–198.
- [24] O.J. Prado, D. Gabriel, J. Lafuente, Economical assessment of the design, construction and operation of open bed biofilters for waste gas treatment, *J. Environ. Manage.* 90 (8) (2009) 2515–2523, 2009.
- [25] F.X. Prenafeta-Boldú, M. Viñas, M. Guivernau, A. Elías, A. Barona, Molecular characterization of TEX (toluene, ethylbenzene, and xylene) degrading microbial communities from air biofilters, in: I. Santre, Govern de les Illes Balears (Eds.), *The Third International Meeting on Environmental Biotechnology and Engineering*, Palma de Mallorca, 2008, p. 2328.
- [26] R. Auria, G. Frere, M. Morales, M.E. Acuña, S. Revah, Influence of mixing and water addition on the removal rate of toluene vapors in a biofilter, *Biotechnol. Bioeng.* 68 (4) (2000) 448–455.
- [27] D.W. Wright, D.K. Eaton, L.T. Nielsen, F.W. Kuhrt, J.A. Koziel, J.P. Spinhirne, D.B. Parker, Multidimensional gas chromatography-olfactometry for the identification and prioritization of malodours from confined animal feeding operations, *J. Agric. Food Chem.* 53 (22) (2005) 8663–8672.
- [28] M.B. Bagherpour, M. Nikazar, U. Welander, B. Bonakdarpour, M. Sanati, Effects of irrigation and water content of packings on alpha-pinene vapours biofiltration performance, *Biochem. Eng. J.* 24 (2005) 185–193.
- [29] H. Jorio, Y. Jin, H. Elmrini, J. Nikiema, R. Brzezinski, M. Heitz, Treatment of VOCs in biofilters inoculated with fungi and microbial consortium, *Environ. Technol.* 30 (5) (2009) 477–485.
- [30] H.K. Son, B.A. Striebig, Operational condition and temperature study for ethylbenzene treating biofilter, *Kor. J. Environ. Health Soc.* 29 (4) (2003) 4–9.
- [31] R. Abumaizar, W. Kocher, E. Smith, Biofiltration of BTEX contaminated air streams using compost-activated carbon filter media, *J. Hazard. Mater.* 60 (1998) 111–126.
- [32] Z. Wang, R. Govind, Biofiltration of isopentane in peat and compost packed beds, *AIChE J.* 43 (5) (1997) 1348–1356.
- [33] M. Morales, S. Revah, R. Auria, Start-up and the effect of gaseous ammonia additions on a biofilter for the elimination of toluene vapours, *Biotechnol. Bioeng.* 60 (4) (1998) 483–491.
- [34] H. Jorio, K. Kiared, R. Brzezinski, A. Leroux, G. Viel, M. Heitz, Treatment of air polluted with high concentrations of toluene and xylene in a pilot-scale biofilter, *J. Chem. Technol. Biotechnol.* 73 (1998) 183–186.
- [35] H. Jorio, G. Viel, M. Heitz, Biofiltration de l'air pollué par le xylene: observations expérimentales, *Can. J. Civ. Eng.* 29 (2002) 543–553.
- [36] A. Torkian, R. Dehghanzadeh, M. Hakimjavadi, Biodegradation of aromatic hydrocarbons in a compost biofilter, *J. Chem. Technol. Biotechnol.* 78 (2003) 795–801.
- [37] H. Elmrini, N. Bredin, Z. Shareefdeen, M. Heitz, Biofiltration of xylene emissions: bioreactor response to variations in the pollutant inlet concentration and gas flow rate, *Chem. Eng. J.* 100 (2004) 149–158.
- [38] D. Wu, X. Quan, Y. Zhao, S. Chen, Removal of p-xylene from an air stream in a hybrid biofilter, *J. Hazard. Mater.* B136 (2006) 288–295.
- [39] E. Jeong, M. Hirai, M. Shoda, Removal of p-xylene with *Pseudomonas* sp. NBM21 in biofilter, *J. Biosci. Bioeng.* 102 (4) (2006) 281–287.
- [40] J.W. Hwang, S.J. Jang, E.Y. Lee, C.Y. Choi, S. Park, Evaluation of composts as biofilter packing material for treatment of gaseous p-xylene, *Biochem. Eng. J.* 35 (2007) 142–149.
- [41] V. Saravanan, N. Rajamohan, Treatment of xylene polluted air using press mud-based biofilter, *J. Hazard. Mater.* 162 (2009) 981–988.
- [42] J.K. Kim, S.K. Kam, M.G. Lee, Characteristics of benzene, toluene and xylene gas removal by a biofilter using scoria, *Int. J. Environ. Pollut.* 39 (3/4) (2009) 264–278.
- [43] E. Rene, M.E. López, D.V.S. Murthy, T. Swaminathan, Removal of xylene in gas-phase using compost-ceramic ball biofilter, *Int. J. Phys. Sci.* 4 (11) (2009) 638–644.