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A comparative assessment of biofiltration and activated sludge diffusion for odour abatement

Raquel Lebrero, Elisa Rodríguez, Pedro A. García-Encina, Raúl Muñoz*

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

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ABSTRACT

The deodorization performance of a biofilter and an activated sludge diffusion (AS) system was comparatively evaluated in terms of removal efficiency (RE) and process stability at empty bed residence times (EBRT) ranging from 94 to 32 s. Both bioreactors were fed with a synthetic odorous emission containing H_2S, butanone and toluene at 23.6–43.3, 4.3–6.3 and 0.4–0.6 mg m^{-3}, respectively. While the outlet H_2S concentration was always lower than 1.4 mg m^{-3}, the REs for butanone and toluene remained higher than 95% in both bioreactors regardless of the EBRT. The continuous supply of wastewater in the AS unit did not affect removal and appeared to be a requirement for efficient pollutant abatement. Despite the narrow carbon source spectrum treated, the AS system maintained a large bacterial diversity over time. Therefore, the results obtained confirmed the potential of AS systems as a robust and efficient biotechnology for odour treatment in WWTPs.

1. Introduction

Emissions of malodorous gases from Wastewater Treatment Plants (WWTPs) have a negative impact on the nearby local population. The increasing number of malodours-related complaints and the recent enforcement of stricter environmental regulations have ranked minimization and abatement of malodorous emissions among the top priorities in the design and operation of WWTP utilities worldwide [1,2]. The key relevance of the problem, both in terms of compliance to regulations and good public image, initially triggered the implementation of physical/chemical off-gas abatement technologies such as chemical scrubbing, activated carbon filtration and incineration. However, WWTP operators became rapidly aware of the merits of biological treatment processes. Nowadays, biological technologies are the preferred option due to their high efficiency, lower operating costs and absence of hazardous end-products [3,4].

Biofiltration is indisputably the most commonly employed biotechnology for odour treatment in WWTPs [5,6]. In biofilters, the odorous emission is forced through an organic/inorganic packed bed supporting the microbial community responsible for odorant removal. Despite their cost-effectiveness, the widespread implementation of biofilters is often restricted by their large footprint (high empty bed residence times, EBRT, and low packed media heights in order to minimize pressure drops) and by the gradual compaction of packing media. In addition, the technical difficulties to control key parameters such as pH and moisture content within the packed bed, and to avoid the accumulation of inhibitory by-products can also limit biofilter performance [7].

In this context, activated sludge diffusion (AS) system represents a cost-effective alternative to media-based odour treatment bioreactors. In AS systems, the malodorous emission is directly sparged into the aeration tank as the air needed to satisfy the biological oxygen demand of the wastewater [8]. Odorants diffuse into the mixed liquor together with O_2, being subsequently degraded by the AS community [3,9]. AS systems possess all merits of their biological counterparts (environmental friendliness, low operating cost) while overcoming most of their major limitations (packing media compaction, moisture control or accumulation of toxic metabolites in biofiltration, etc.). In addition, the use of the existing aeration tank as odour-abatement unit renders them economically attractive in plants with land limitations. Despite AS systems have been used for over 30 years with high H_2S removal efficiencies (REs), their widespread implementation is still limited by the lack of reliable data concerning its performance during the treatment of odorous volatile organic compounds (VOCs) [9–11].

This work was conducted to systematically compare the performance of a conventional biofilter and an AS system for the treatment of a model WWTP malodorous emission containing four representative odorants with a large range of hydrophobicities. Butanone, toluene, and α-pinene were selected as model VOC odorants representing soluble, moderately soluble and hydrophobic VOCs associated to WWTP emissions [12]. Likewise, H_2S was also selected as model sulphur odorant for being widely present in

* Corresponding author. Tel.: +34 983186424; fax: +34 983423013.
E-mail address: mutora@iq.uva.es (R. Muñoz).

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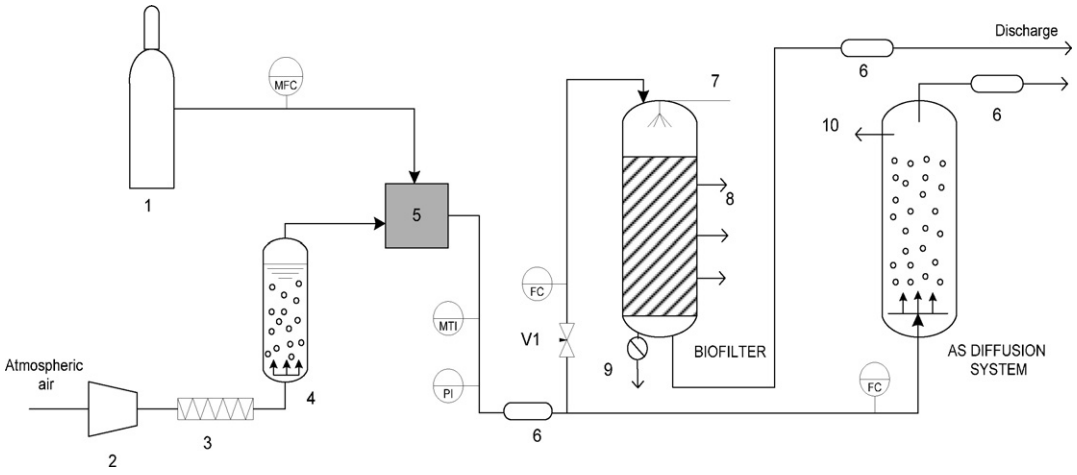

Fig. 1. Schematic representation of the experimental setup. (1) H_2S and VOC reservoir, (2) Compressor, (3) Activated carbon filter bed, (4) Humidifier, (5) Mixing chamber, (6) Gas sampling bulb, (7) Irrigation system, (8) Gas sampling ports, (9) Leachate port, (10) Liquid sampling port, MFC: Mass flow controller FC: Flow controller MTI: Temperature and moisture indicator PI: Pressure indicator VI: Needle valve.

sewage work emissions. The long term performance of both systems along with their detailed characterization at different EBRTs was herein studied. The capacity of these technologies to cope with process fluctuations is reported elsewhere [13].

2. Materials and methods

2.1. Microorganisms and culture conditions

Aerobic bacterial sludge collected at Valladolid WWTP was used here as inoculum. A *Pseudomonas fluorescens* NCIMB 11671, purchased from the National Collection of Industrial and Marine Bacteria (Aberdeen, Scotland), was also added on day 132 to enhance α-pinene biodegradation. A SO_4^{2-}-free mineral salt medium (MSM) was used for biofilter irrigation and as a wastewater matrix to feed the AS unit [14]. MSM was composed of ($g\,l^{-1}$): $Na_2HPO_4{\cdot}12H_2O$, 6.15; KH_2PO_4, 1.52; NH_4Cl, 0.81; $MgCl_2{\cdot}6H_2O$, 0.17; $CaCl_2$, 0.038; and 10 $ml\,l^{-1}$ of a trace element solution containing ($g\,l^{-1}$): EDTA, 0.5; $FeSO_4{\cdot}7H_2O$, 0.2; $ZnSO_4{\cdot}7H_2O$, 0.01; $MnCl_2{\cdot}4H_2O$, 0.003; H_3BO_3, 0.03; $CoCl_2{\cdot}6H_2O$, 0.02; $CuCl_2{\cdot}2H_2O$, 0.001; $NiCl_2{\cdot}6H_2O$, 0.002; $NaMoO_4{\cdot}2H_2O$, 0.003. The final pH of medium was 7.0.

2.2. Chemicals

Butanone, toluene and α-pinene were purchased from Sigma-Aldrich with a purity higher than 99.9%. All other chemicals and reagents were purchased from PANREAC with a purity of +99% (Barcelona, Spain).

2.3. Experimental Set-up

The experiments were carried out in a lab-scale plant consisting of two jacketed column bioreactors operated in parallel: a biofilter and an AS system (Fig. 1). Both bioreactors (120 cm height × 10 cm i.d.) were made of clear PVC with a working volume of 8.5 L and operated at 20 °C. The biofilter was packed with 8.5 L of a mixture of compost and perlite while the AS unit was initially filled with 7.5 L of MSM. The characterization of the packing material was carried out according to standard methods [15] (Table 1).

The odorous stream was prepared by mixing either a concentrated H_2S/butanone/toluene stream or a concentrated H_2S/butanone/toluene/α-pinene stream from calibration cylinders (Abello Linde S.A., Spain) with pre-humidified H_2S and VOCs-free air (ambient air filtered through a 1.6 L activated carbon bed and humidified in a 1 m water column). A mass flow controller (Aalborg, Denmark) was used to accurately dose the concentrated mixture. The final concentrations ranges were 23.6–43.3, 4.3–6.3, 0.40–0.60 and 0.12–0.15 $mg\,m^{-3}$ for H_2S, butanone, toluene and α-pinene, respectively, corresponding to concentrations ranges of 17–31, 1.5–2.2, 0.1–0.2 and 0.02–0.03 ppm, respectively. The concentrations selected were within the typical concentration range of VOC emissions from WWTP according to Zarra et al. [12]. The odorous emission was then equally split and fed to the biofilter from the top of the reactor (downflow configuration) and to the AS system via three ceramic spargers located at the bottom of the bioreactor.

Prior to process start-up, a test was conducted to assess abiotic H_2S and VOC removal. Inlet and outlet H_2S and VOC concentrations were periodically monitored in both bioreactors for 94 h at an EBRT of 94 s in the absence of biofilter packing material and microbial activity to asses for pollutant adsorption and photolysis in the experimental set-up.

Both bioreactors were inoculated with 1–L of concentrated ($17\,g\,L^{-1}$) return AS resuspended in MSM. The systems were first operated for approximately 121 d to evaluate the influence of the EBRT (94, 74, 55, 48, and 32 s) on RE using H_2S, butanone and toluene as model odorants. During this period, the AS system was operated in the absence of glucose addition at an infinite sludge retention time (SRT) (no biomass withdrawal). However, due to biomass aggregation and compaction on day 95 (corresponding to an EBRT of 48 s), the SRT in the AS system was set up at 25 d by daily withdrawal of 340 mL of mixed liquor and replacement with fresh MSM containing 2 g of glucose (corresponding to an organic load of 0.3 kg COD $m^{-3}\,d^{-1}$). SRTs of 10–30 d are typical in WWTP operated in cold climates to guarantee consistent wastewater treatment efficiencies. Once a steady state was reached again in the AS unit, the EBRT was further decreased to 32 s in both bioreactors. At day 121, the ability of both odour abatement biotechnologies to remove hydrophobic odorants was challenged by the supple-

Table 1
Characterization of the biofilter packing material.

Parameter	Value
Composition	75/25 (% compost/% perlite)
Density (as received)	0.22 $g\,mL^{-1}$
Porosity	64.4%
Water holding capacity	41% (volume basis)
pH	5.60

Table 2
Steady state REs and confidence interval ($p = 0.05$) in the biofilter and the AS system at the studied EBRT.

EBRT (s)	Biofilter			AS system		
	Butanone	Toluene	α-Pinene	Butanone	Toluene	α-Pinene
94.5	98.9 ± 1.4	ND		ND	98.4 ± 2.8	
73.5	97.9 ± 0.4	ND		98.4 ± 0.6	97.9 ± 0.3	
55.0	98.9 ± 0.2	ND		99.3 ± 0.1	96.6 ± 1.0	
48.8	99.4 ± 0.1	99.9 ± 0.2		99.3 ± 0.3	95.0 ± 1.4	
32.0	99.3 ± 0.4	99.9 ± 0.2	7.3 ± 1.9	99.7 ± 0.1	96.2 ± 1.2	6.8 ± 1.9

ND: odorant not detected in the outlet stream.

mentation to the above mentioned synthetic odorous stream with α-pinene. Each operational condition was maintained for at least 3 wk in order to ensure stable steady states.

The pH in the AS system was maintained constant at approximately 6.3 ± 0.3 via daily addition of a 50 g L^{-1} NaOH and 15 g L^{-1} Na_2CO_3 solution (in order to supply inorganic carbon for the autotrophic H_2S oxidising bacteria). In addition, 1 L of AS mixed liquor was periodically removed, centrifuged (10 min, 10,000 rpm) and resuspended in MSM in order to maintain sulphate concentration below 3500 mg L^{-1} (corresponding to a conservative salt concentration of 0.9 wt% [5]). Likewise, 250 mL of MSM were periodically irrigated at a frequency inversely proportional to the EBRT via a spray nozzle located at the top of the biofilter. This irrigation avoided media drying and promoted the wash-out of inhibitory sulphate concentrations.

The concentration of CO_2, H_2S, and VOCs was periodically monitored at both inlet and outlet gas sampling ports. H_2S and VOC concentration was also measured at sampling ports located at 0, 40, 80, and 120 cm from the biofilter inlet in order to determine the biodegradation profiles. pH and biomass, SO_4^{2-}, dissolved total organic carbon (DOC), dissolved inorganic carbon (DIC), dissolved total nitrogen (DTN) and ATP concentrations were periodically recorded in the AS system by drawing 20 mL of mixed liquor. Likewise, the pressure drop in the packing media, the pH, SO_4^{2-}, DOC, DIC, and DTN concentrations in the leachate resulting from biofilter irrigation were also periodically measured. In addition, the inlet moisture content of the synthetic odorous emission was also continuously monitored. Finally, the volumetric mass transfer coefficients (k_La) in the AS system were determined according to Quijano et al. [16].

2.4. AS microbial activity monitoring

Batch tests were conducted periodically to monitor microbial acclimation in the AS system. Seven serological bottles of 120 mL were initially filled with 5 mL of MSM and 5 mL of sludge from the AS reactor, closed with butyl septa and sealed with aluminum caps. Butanone and toluene were added to the headspace at 3.0 ± 0.7 and 0.6 ± 0.1 mg m^{-3}, respectively. The microbial assays were incubated at 20 °C in a thermostatic bath under magnetic agitation at 300 rpm and the concentration of butanone and toluene was measured periodically (by removing a test bottle due to the destructive nature of the solid phase microextraction (SPME)–GC–MS analysis) until complete VOC depletion.

2.5. Analytical procedures

H_2S was analysed using an electrochemical sensor (Dräger X-am 5000) calibrated in the 0–40 ppm range. Gas samples for VOC analysis were collected in 250 mL calibrated glass bulbs (SUPELCO) and pre-concentrated by SPME. VOC concentrations were then determined by GC–MS according to Lebrero et al. [13]. External standards prepared in calibrated glass bulbs with humidified air, and sampled under similar conditions as those used during bioreactor sampling, were used for VOC quantification.

Carbon dioxide was analysed in a GC-TCD (Varian CP-3800) according to Hernandez et al. [17].

Biomass concentration in the AS unit was estimated via culture absorbance measurements (optical density at 600 nm) in a Hitachi U-2000 spectrophotometer (Hitachi, Tokyo, Japan) and as total solids concentration according to Standard Methods [18]. ATP was measured using a Microbial ATP kit HS (Biothema, Stockholm, Sweden) and a Microtox 500 luminometer (Azur Environmental, Carlsbad, Germany).

DOC, DIC and DTN concentration was determined in liquid samples from both reactors (mixed liquor in the AS and leachate in the biofilter) according to Hernandez et al. [17]. Sulphate concentration was determined by HPLC-IC using an IC-Pak Anion HC (150 mm × 4.6 mm) column. Liquid samples of 1.5 mL were filtrated through 0.22 μm filters before analysis. The pH was also measured using a pH/mV/°C meter (pH 510 Eutech Instruments, Nijkerk, The Netherlands).

The pressure drop in the biofilter was determined using a homemade differential pressure meter (a clear glass U-tube filled with water and connected directly to the gas inlet and outlet). The moisture content in the influent odorous streams was measured using a Testo 605-H1 thermohygrometer (Testo AG, Germany).

Liquid samples from the AS unit were also drawn and frozen immediately to monitor the population dynamics of the bacterial communities by denaturing gradient gel electrophoresis (DGGE) profiling as described in Lebrero et al. [13]. DGGE was also used to identify the members of the mixed microbial communities detected by DGGE fingerprinting. For this purpose, individual bands were excised from the DGGE gel with a sterile blade, resuspended in 50 μL of ultrapure water and maintained at 60 °C for 1 h to allow DNA extraction from the gel. A volume of 5 μL of the supernatant was used for reamplification with the original primer set. Before sequencing, PCR products were purified with the GenElute PCR DNA Purification Kit (Sigma-Aldrich, St. Louis, MO, USA). The sequences from the excised bands were analysed and compared with sequences in GenBank by BLAST search tool at the NCBI (National Centre for Biotechnology Information) [19]. The sequences were imported into the MEGA program and aligned using the automatic aligner function. The alignment was further corrected manually and phylogenetic trees were constructed by 1000-fold bootstrap analysis using neighbor-joining methods. Trees were edited using MEGA 3.

The sequences were registered in the GenBank Data Library under accession numbers HQ147605-HQ147612.

2.6. Data treatment

Unless otherwise specified, the REs and concentrations recorded during the steady states achieved were presented as the average value with its corresponding error at 95% confidence interval ($p = 0.05$). The Excel statistical package (Microsoft Corporation, USA) was used for data treatment.

3. Results and discussion

No significant differences were recorded between inlet and outlet concentrations of H_2S, butanone or toluene during the 94 h abiotic test, with maximum standard deviations of 1%, 13% and 5% in the biofilter and 0%, 11% and 4% in the AS system, respectively. These results confirmed that neither odorant adsorption nor photolysis occurred in the experimental set-up.

3.1. AS system performance before glucose addition: H_2S and VOC removal

H_2S outlet concentration was below the detection limits of the electrochemical sensor used for H_2S measurement (1 ppm or 1.4 mg m^{-3}) in the AS system regardless of the EBRT employed, even at 32 s. Thus, H_2S REs ranged from 96% to 100% (Fig. 2a). These high REs have been widely reported for AS systems treating varying H_2S concentrations. For example, Barbosa et al. [9] found H_2S REs higher than 98% in the aeration tank of a wastewater treatment pilot plant treating H_2S at 5–25 ppmv (7–35 mg m^{-3}) and up to 99.4% when operating at inlet H_2S concentrations ranging from 30 to 105 ppmv (42–146 mg m^{-3}) [10]. Similarly, Burgess et al. [3] also recorded REs higher than 99% in an AS system treating inlet H_2S concentrations from 77 to 100 ppm (107–140 mg m^{-3}).

A steady increase of sulphate concentration up to 3500 ppm was observed during the first 70 d of operation, remaining stable afterwards (as a result of periodic MSM exchanges). The experimental and theoretical sulphate concentrations exhibited a good correlation ($[SO_4^{2-}]_{theoretical} = 1.011\ [SO_4^{2-}]_{experimental}$; $R^2 = 0.96$), which suggest a complete oxidation of H_2S to sulphate as a result of the high dissolved oxygen concentrations present in the mixed liquor.

Despite H_2S elimination performance has been widely monitored in AS systems due to its ease of measurement, scarce information exists about VOC treatment at trace level concentrations. In our particular study, process start-up was characterized by high initial butanone and toluene REs due to VOC absorption into the mixed liquor followed by a gradual decrease in VOC removal performance. Butanone and toluene REs increased from 45% and 11% at day 2 up to REs > 99% by day 8 and 20, respectively (Fig. 2b and c). Butanone REs in the AS system ranged from 98 to REs > 99% regardless of the EBRT. In the case of toluene, REs of 98.4 ± 2.8% at 94 and 74 s, 95 ± 1.4% at 49 s and 96 ± 1.2% at 32 s were recorded (Fig. 2b and c). These values represent, to the best of our knowledge, the highest REs ever reported for VOCs at such low inlet concentrations and residence times in AS systems. The capacity of AS processes to remove dissolved VOCs was previously studied by Barbosa et al. [10], who recorded high efficiencies for the treatment of a wide range of VOCs and volatile sulphur compounds coming with the wastewater. However, these authors only supplied H_2S to the system with the odorous stream.

The high VOC abatement performance was probably due to the high k_La values driving odorant transfer from the emission and to the efficient microbial pollutant uptake in the mixed liquor (no VOC diffusion limitations). Mass transfer coefficients increased exponentially when decreasing the EBRT from 241 ± 38 h^{-1} at 94 s to 697 ± 40 h^{-1} at 32 s. These k_La values are quite high compared to those reported by Dorado et al. [20] in a biotrickling filter packed with polyurethane foam at 35 s of EBRT ($k_La \approx 43.2\ h^{-1}$). However, the k_La increased up to 700 h^{-1} when the EBRT of the biotrickling filter decreased to 10 s.

The performance of the AS unit for the removal of hydrophobic VOCs was challenged by the addition of α-pinene to the synthetic odorous stream at day 121. While this odorant was initially removed at approximately 21%, its RE decreased to 6.8 ± 1.9% after two days of operation and remained constant for the following 40 d. Despite lower REs compared to toluene and butanone were expected due to mass transfer limitations (low concentration gradient available for mass transfer as a result of its higher partition coefficient), this poor abatement performance was likely due to the lack of a specialized α-pinene degrading community. In this context, the addition of 250 mL of a *Pseudomonas fluorescens* culture (α-pinene degrading species [14]) after 11 days of α-pinene feeding did not result in significant enhancements in the removal of this terpene. The absence of an active microbial community capable of degrading α-pinene at the low concentrations present in the mixed liquor (0.10-0.13 μg l^{-1}) was confirmed by the fact that operation at low pH values triggered the development of an α-pinene degrading community able to degrade up to 50% of this terpene at an EBRT of 50 s (data shown in Lebrero et al. [13]).

Batch VOC biodegradation tests showed a gradual enrichment of bacterial communities specialized in the degradation of trace level concentrations of VOC (2.6 ± 0.4 mg m^{-3} and 0.6 ± 0.1 mg m^{-3} of butanone and toluene, respectively) in the AS. Figure 3 represents the degradation kinetics for butanone and toluene of sludge samples drawn at different operation times. Hence, while microorganisms were initially not able to completely degrade butanone and toluene in 80 min, less than 60 min were required for its complete depletion after 58 and 27 d, respectively.

3.2. AS system performance after glucose addition: biomass growth

After three months of operation with butanone, toluene and H_2S, the AS system collapsed due to an unexpected biomass aggregation followed by compaction and sedimentation at the bottom of the bioreactor. Hence, biomass concentration rapidly decreased from 1 to 0.12 g L^{-1} (Fig. 4) concomitant with a reduction in butanone and toluene REs (63% and 34%, respectively) as a result of the reduction in the specific interfacial area biomass-cultivation broth due to biomass compaction (Fig. 2b and c). The microbiological reasons underlying this phenomenon are however unknown. Glucose was then supplied at a rate of 0.3 kg COD m^{-3} d^{-1} along with a daily biomass withdrawal (sludge retention time of 25 d) in order to maintain the suspended biomass culture at 2.5 g L^{-1} throughout the rest of the experiment. The addition of an easily biodegradable carbon source, simulating wastewater input during real WWTP operation, resulted in a rapid re-suspension of the compacted biomass and in the recovery of the preceding steady state REs. Therefore, the hypothesis of a reduced VOC-degrading activity in the presence of more easily degradable carbon sources (such as organic matter from wastewater) can be ruled-out since the supply of such a source appears to be a requirement for a successful odour removal in AS systems. Moreover, the presence of an easily biodegradable carbon source resulted in a more active biomass as shown by the higher specific ATP contents, which increased from an initial value of $5.0 \pm 0.5 \times 10^{-9}$ mol ATP (g biomass)$^{-1}$ up to constant values of $3.7 \pm 0.2 \times 10^{-8}$ mol ATP (g biomass)$^{-1}$ (Fig. 4). This increase in the energetic level of the cells, herein quantified as specific ATP content, was in agreement with the experimental findings of Bordel et al. [21], who observed an increase in the specific ATP content of *P. putida* F1 at increasing toluene concentrations.

3.3. AS microbial analysis

Despite the acclimation and specialization of the VOC and H_2S degrading community and the addition of glucose (an easily biodegradable carbon source) from day 94, the biodiversity of the microbial community present in the AS unit remained surprisingly constant over time, as shown by the DGGE (Fig. 5). Therefore, the addition of glucose allowed the recovery of the system due to an increase of microbial activity as shown by the increase in RE (Section 3.2), but it did not affect the biodiversity (letter A, Fig. 5).

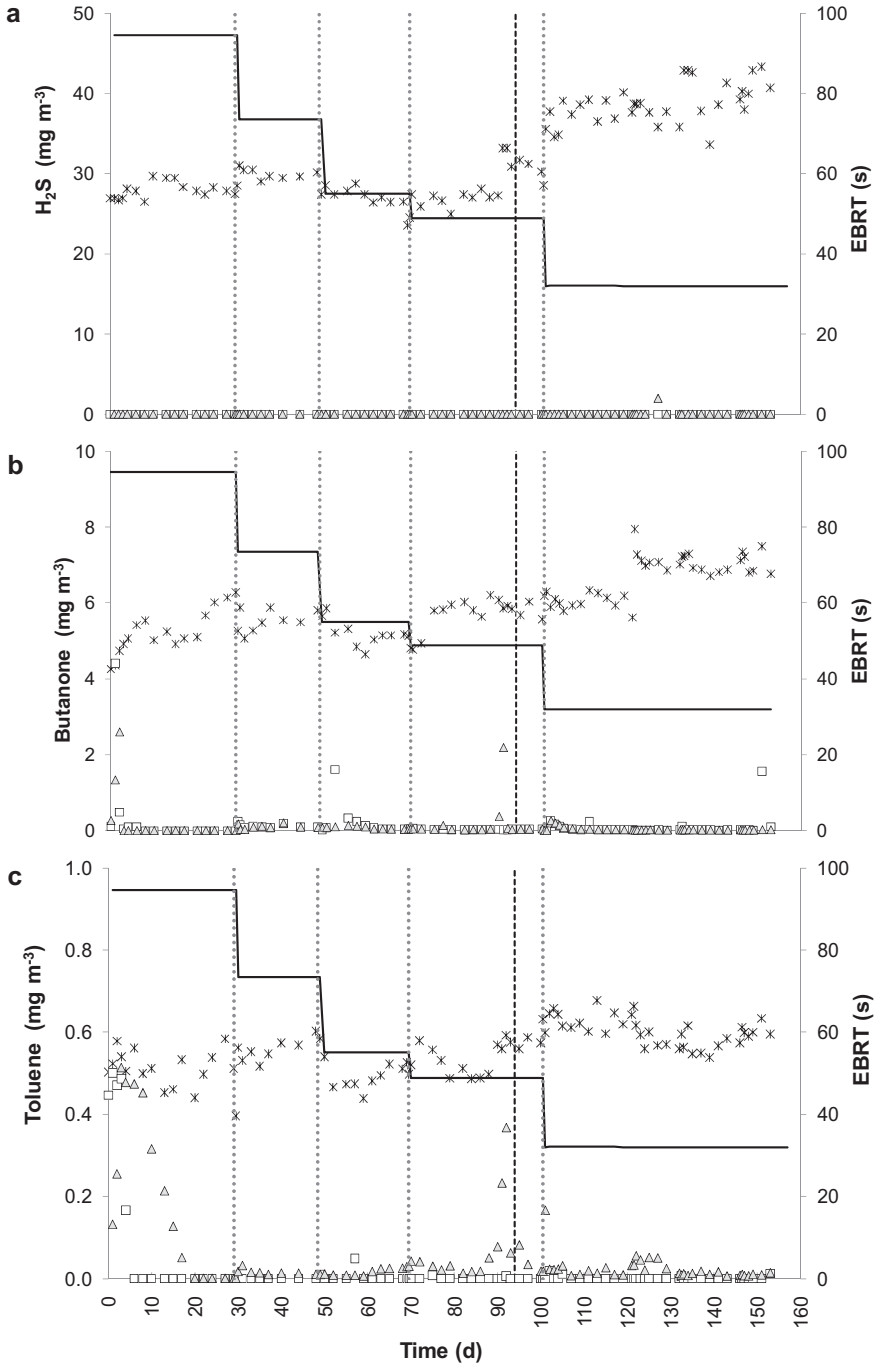

Fig. 2. Time course of H_2S (a), butanone (b) and toluene (c) concentrations in the influent stream (*), biofilter effluent (□) and the AS effluent (△) at different EBRTs (continuous line). Vertical dotted lines represent the operation at different EBRT and the vertical dashed line represents the beginning of AS operation at 25 d of sludge retention time.

Similar results were obtained by Bayle et al. [22] operating two AS reactors supplied with a complex VOC mixture at high and low concentrations. These authors observed a reduction in the bacterial diversity of the bioreactor supplied with the highest VOC loading and the maintenance of a large bacterial diversity in the bioreactor operated at low VOC concentrations. These results support the gradual enrichment of the bacterial communities specialized in the degradation of VOCs and H_2S at trace level concentrations herein obtained and highlight the broad catabolic potential of the AS unit from WWTP. The degree of similarity of DGGE bands to known sequences ranged from 97% to 99%, except for bands 5 and 1, which ranged between 93% and 95%.

DGGE sequencing in our study showed members of four bacterial divisions (Fig. 6). Three out of the eight DGGE bands identified (bands 2, 3 and 6) clustered within the *Proteobacteria* division (γ-*Proteobacteria* and β-*Proteobacteria*). Two DGGE bands (bands 4 and 5) were identified as representatives of *Actinobacteria* while DGGE bands 7 and 8 clustered within *Nitrospirae* and *Chloroflexi,* respectively. The few 16S rRNA-based studies available in literature showed that members of *Proteobacteria*, *Bacteroidetes*, *Chloroflexi* and *Planctomycetes* divisions are abundant in this type of systems [22]. In spite of the addition of a *P. fluorescens* culture on day 132 (letter C in Fig. 5), none of the DGGE bands sequenced were affiliated to the *Pseudomonas* genus, which agreed with the

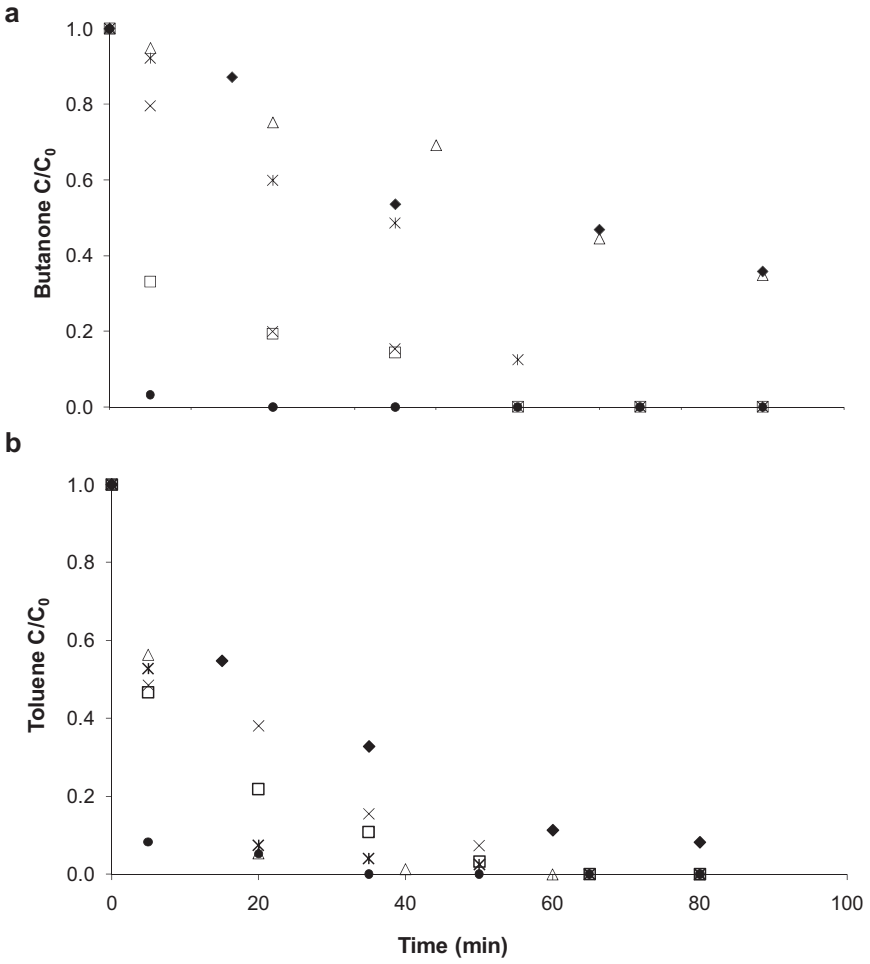

Fig. 3. Time course of butanone (a) and toluene (b) normalized outlet concentrations in the AS mixed liquor at days 9 (♦), 27 (△), 58 (*), 66 (×), 98 (□) and 156 (●).

low REs observed for α-pinene biodegradation. Finally, it must be highlighted that fungi were observed under microscopic analysis, suggesting the presence of a mixed bacterial-fungal population in the AS system. The characterization of these fungal communities is being carried out.

3.4. Biofilter performance: H_2S and VOC removal

H_2S abatement performance in the biofilter was comparable to that recorded in the AS system. Thus, H_2S was also removed at RE > 99% in the biofilter regardless of the EBRT applied (Fig. 2a). High

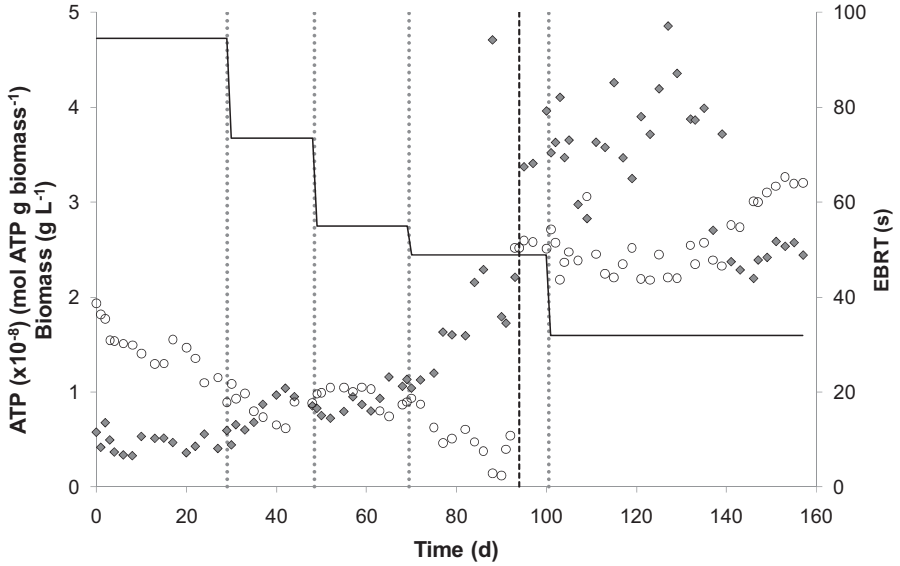

Fig. 4. Time course of biomass concentration (o), specific ATP content (◊) and EBRT in the AS system. The specific ATP values are divided by 10^8. Vertical dotted lines represent the operation at different EBRTs while the vertical dashed line represents the beginning of AS unit operation at 25 d of sludge retention.

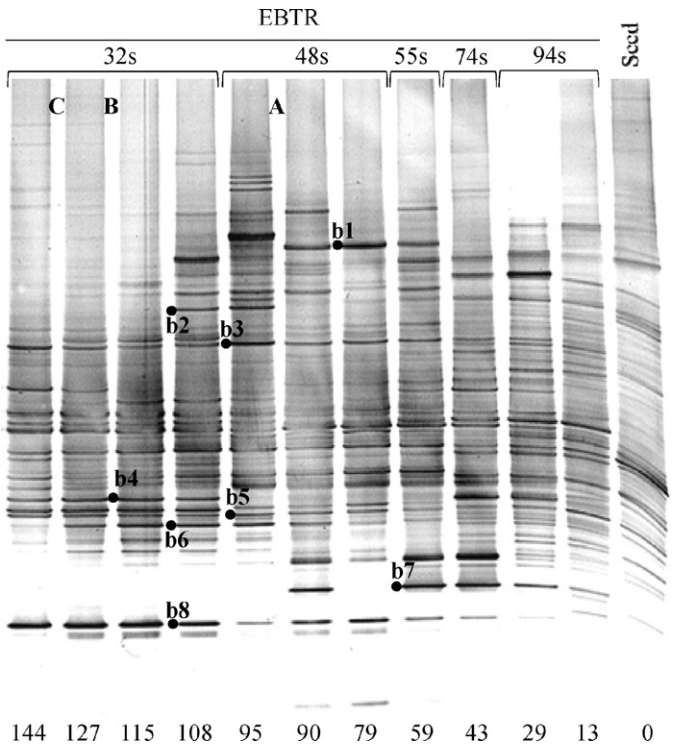

Fig. 5. Bacterial DGGE patterns in the AS system. "DGGE bands are indicated with "B" and the corresponding number of each band. The letters A, B and C indicates the addition of glucose, α-pinene and a *P. fluorescence* culture, respectively. The sampling time and its corresponding EBRT are shown in the lower and upper lane numbers, respectively".

H_2S REs are commonly found in lab and full scale biofilters due to its rapid transfer from the gas phase to the microbial biofilm and the readily biodegradable nature of this volatile inorganic compound (VIC). For example, Morgan-Sagastume and Noyola [23] recorded REs ≈100% in a compost biofilter fed with 100 ppm (140 mg m^{-3}) of H_2S at an EBRT of 50 s. Similarly, Iranpour et al. [24] reported typical H_2S REs ranging from 90% to 100% in full scale biofilters operated in WWTPs in USA. Sulphate, the main byproduct from H_2S oxidation, was periodically washed out from the packed bed as shown by the high concentrations recorded in the leachate after irrigation (maximum value of 18,700 ppm), which prevented from microbial inhibition.

Butanone REs in the biofilter ranged from 98% to 99.5% at the tested EBRTs, which were comparable to those observed in the AS system. Conversely, the biofilter performed better for toluene removal, with REs over 99.9% regardless of the EBRT (Fig. 2b and c). These toluene elimination efficiencies were noticeably higher than those reported in literature. For instance, Liu et al. [25] achieved a maximum RE for toluene in a compost biofilter of 82% at inlet toluene concentration of 0.07–0.73 mg m^{-3} at an EBRT of 65 s. Likewise, Iranpour et al. [24] reported average REs lower than 90% during VOC treatment depending on their hydrophobicity, with hydrophilic VOCs exhibiting highest mass transport gradients due to their lower partition coefficient and slightly soluble compounds presenting poor REs due to a limited mass transfer. Surprisingly, in our particular biofiltration study, toluene REs were slightly higher than those recorded for butanone despite its higher hydrophobicity. This suggests that no mass transfer limitations occurred for these compounds in the biofilter and that other mass transfer mechanism different from the conventional air–water–biofilm (for instance direct air–biofilm transfer) might be present in the biofilm. The biofilter performance for α-pinene treatment was similar to that of the AS system. The REs remained at 7.3 ± 1.9%. Similarly to the AS system, this low terpene removal performance was likely due to the lack of a specialized α-pinene degrading community. The addition of *P. fluorescens* after 11 days of α-pinene feeding did not result in any significant increase in the removal of this terpene. The absence of an active microbial community was further confirmed by the fact that an α-pinene degrading community developed under stress conditions (no irrigation) and α-pinene removal increased up to 65% at EBRT of 50 s (data shown in Lebrero et al. [13]). The low EBRT tested in this study (32 s) might have also mediated the low terpene abatement here recorded. REs of 65% were in the range of those reported for this terpene in conventional biofilters. For example, Jin et al. [26] achieved REs up to 89% in a fungal biofilter treating α-pinene at 2.47 g m^{-3} at EBRTs greater than 1.2 min. A summary of the steady state REs in

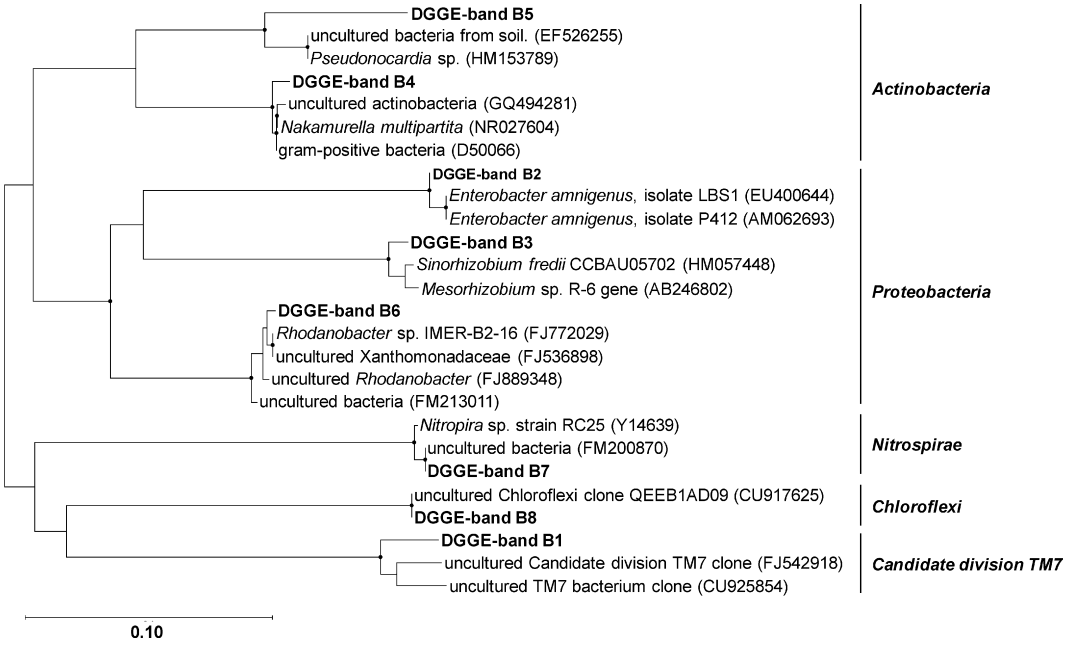

Fig. 6. Phylogenetic tree based on the bacterial 16S rRNA gene sequences obtained from the DGGE bands. Sequences determined in this study are in boldface. Black dots on the nodes indicate bootstrap values of 90% or higher (1000 replicates). The scale bar indicates 10% sequence difference.

the biofilter and the AS system at the studied EBRT is shown in Table 2.

The high REs values recorded in the biofilter were however obtained at the expense of an important pressure drop across the filter bed. The pressure drop in biofilters constitutes the main parameter determining operational costs [7] and 10 cm of water column is often considered as the maximum tolerable pressure drop in biofiltration [27]. This critical value was considerably exceeded in our biofiltration system, where the average pressure drop increased from 6 to 33 cm of water column when decreasing the EBRT from 94 to 32 s, respectively. The gradual deterioration of the packing material used (4.5 cm of bed compaction after 5 months of operation), together with the low diameter of the packing material (1–5 mm for perlite, 0–20 mm for compost) could have contributed to the high pressure drop herein recorded [7].

The packing material lost its buffer capacity at day 80 as shown by the rapid decrease on the pH of the leachate from 4.8 ± 0.2 to 1.3 ± 0.1. Nevertheless, despite the low pH of the biofilter leachate, confirmed by independent measurements of the pH of the packing material (pH of 1.8 at day 157 regardless of the biofilter height), the REs for the tested odorants remained unchanged, which suggests an acclimation of the microbial population to the acidic environment. In this context, a fungal community was observed by microscopy analysis of a packing material sample. Low pH values in biofilters have been previously reported in literature, particularly in fungal biofilters treating sulphur compounds or VOCs and in biofilters inoculated with acidophilic bacterial species [6,28,29].

3.5. Biodegradation profiles of the biofilter

The analysis of the time course of the biodegradation profile along with the biofilter height revealed that most of the H_2S and VOC degradation occurred in the first 40 cm of the bed column, which corresponded to approximately 30% of the bed volume. From the first day of operation, H_2S was totally depleted at the sampling point located 40 cm from the biofilter inlet, whereas butanone and toluene REs at this point increased throughout the experimentation period, being higher than 98.8% for butanone and 95% for toluene from day 29 of operation. Complete removal of VOCs and VICs in the first part of the biofilter column has been already reported in literature. For instance, 40% of a pine bark woodchips biofilter bed was required to completely eliminate oxygenated compounds, while this percentage increased up to 60% when total sulphur and nitrogen compounds removal was evaluated [30].

3.6. CO_2, DOC, DIC and DTN

No significant differences were found between inlet and outlet CO_2 concentrations in the AS system (0.73 ± 0.02 g CO_2 m^{-3} and 0.81 ± 0.03 g CO_2 m^{-3}, respectively) due to the low concentrations of VOCs present in the artificial odour emission treated (data not shown). Slightly higher values (0.87 ± 0.03 g m^{-3}) were recorded in the biofilter during the first three months of operation, probably due to the additional biodegradation of the organic matter present in the compost. However, CO_2 production in the biofilter decreased to 0.76 ± 0.02 g m^{-3} within the last two months of operation, which suggests the complete compost stabilization.

The DOC in the AS system remained stable at 11.1 ± 0.7 mg L^{-1} during the first 72 days of experiment, steadily increasing to a maximum value of 34.8 mg L^{-1} when glucose was added to the system. However, the DOC gradually decreased to the preceding steady value from day 105. In the biofilter, the DOC of the leachate remained approximately constant at 132 ± 10 mg L^{-1} due to the leaching of organic matter from the compost. The concentration of DIC remained constant at 1.6 ± 0.2 and 0.43 ± 0.03 mg L^{-1} in the AS system and the biofilter leachate, respectively. On the other hand, the DTN underwent a sharp decrease in the AS system after glucose addition at day 93 from 162 ± 7 down to 2.3 mg L^{-1} due to the increase in biomass concentration, increasing afterwards to a steady value of 91 ± 5 mg L^{-1}. High DTN values of up to 718 ± 21 mg L^{-1} were initially recorded in the biofilter leachate as a result of N-compounds leaching from the compost. The DTN concentration in the leachate steadily decreased but no limitation was observed throughout the experiment.

3.7. Cost evaluation

AS system is a low cost alternative to biofiltration for odour treatment in WWTP [31]. The investment costs are very limited in AS diffusion since this biotechnology employs the equipment already present in the WWTP. Minimal capital costs are due to ductwork, installation of moisture traps, dust and grease aerosol filters and the replacement of certain equipments with anticorrosion materials (i.e. blowers) [31,32]. In addition, the operating costs are limited to the maintenance of filters and moisture traps, since no packing material is present and the energy cost of air diffusion into the aeration basin is included in the operating cost of the water line. However, if the system needs a dedicated blower or diffuser, or if a long gas pipeline is required, a cost-effectiveness analysis is required [31]. In this context, AS systems are recommended for treating the gaseous emissions of the primary sedimentation tank and the sludge treatment units, the closer unit operations. According to a recent study [32], biofiltration capital costs range from 5 to 28 € $(m^3/h)^{-1}$, while operation costs are about 0.21 € $(1000\ m^3\ treated)^{-1}$.

4. Conclusions

The results here obtained confirmed the potential of AS systems as a robust and efficient biotechnology for odour treatment in WWTPs, with comparable steady state REs (>95%) for H_2S, butanone and toluene to those recorded in the biofilter regardless of the EBRTs (94–32 s). High k_La values were recorded in the AS, which increased exponentially with decreasing EBRT. The supply of wastewater to the AS unit contributed to an enhanced process stability by preventing biomass compaction. Therefore, a complete VOC removal can be expected in aerated tanks with fine bubble diffusers in WWTPs under the typical operational conditions.

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