Sustainable Chemistry and Pharmacy 31 (2023) 100960

Contents lists available at ScienceDirect

Sustainable Chemistry and Pharmacy

journal homepage: www.elsevier.com/locate/scp

Biofiltration based on bioactive coatings for the abatement of indoor air VOCs

Javier González-Martín [a, b], Sara Cantera [a, c], Raquel Lebrero [a, b], Raúl Muñoz [a, b, *]

[a] Institute of Sustainable Processes, University of Valladolid, Dr. Mergelina s/n., Valladolid, 47011, Spain
[b] Department of Chemical Engineering and Environmental Technology, University of Valladolid, Dr. Mergelina s/n., Valladolid, 47011, Spain
[c] Laboratory of Microbiology, Wageningen University and Research Center, the Netherlands

ARTICLE INFO

Keywords:
Bioactive coating
Biofiltration
Indoor air quality
VOCs

ABSTRACT

Biotechnologies are a feasible alternative for indoor air pollutant abatement. Among biotechnologies, bioactive coatings consist of microorganisms embedded in polymeric matrices, allowing a direct contact between microorganisms and gas pollutants, thus enhancing their abatement. Three bioreactors (BR1, BR2 and BR3) were inoculated with a VOC-degrading enriched culture, a latex bioactive coating with the VOC-degrading enriched culture, and a latex bioactive coating with fresh activated sludge. The influence of empty bed residence time (EBRT) and inlet concentrations on the removal of toluene, α-pinene and n-hexane was assessed. BR1 and BR2 achieved steady-state toluene and pinene removals >90% down to 30 s. BR3 lower removals could be attributed to the lack of acclimation of activated sludge. When inlet concentrations were progressively reduced to <2 mg m^{-3} at 15 s of EBRT, toluene removals increased to >80% in BR1 and BR2, but only to 64.2% in BR3. Pinene removals reached 90.9% in BR1, and >70% in BR2 and BR3. The bacterial population was dominated by *Rhodococcus*, *Mycobacterium*, *Devosia* and *Rhodobacteraceae* members in BR1 and BR2. No significant and robust hexane removal was observed regardless of the inoculum or operational conditions, probably due to mass transfer limitations, which entailed a low dominance of organisms with this metabolic capability.

1. Introduction

The quality of indoor air is a health issue worldwide. Nowadays, citizens in developed countries spend up to 90% of their lives indoors, and paradoxically indoor air often exhibits higher concentrations of gas pollutants than outdoors air (European Environment Agency, 2020). Indeed, approximately 3.8 million deaths are attributed to indoor air pollution every year worldwide. These deaths are associated with chronic obstructive pulmonary disease, lung cancer, ischemic heart disease, and strokes, among others (Lee et al., 2020; Van Tran et al., 2020; World Health Organization, 2018). Poor indoor air quality is also responsible for the sick building syndrome, which is characterized by symptoms such as mucous membrane and skin irritations, headache, asthma and mental fatigue (Ghaffarianhoseini et al., 2018; Nakaoka et al., 2014; Van Tran et al., 2020). This problem is expected to aggravate in the coming years, as new building regulations, aligned with the Sustainability Development Goals, tend to increase the tightness of constructions to focus on energy efficiency, then reducing ventilation rates and contributing to the rise of indoor pollution levels.

One of the most well-known contributors to indoor air pollution are volatile organic compounds (VOCs), which have been identified in indoor environments such as homes (Pei et al., 2020; Wickliffe et al., 2020), offices (Mandin et al., 2017), stores (Robert et al.,

* Corresponding author. Department of Chemical Engineering and Environmental Technology, University of Valladolid, Dr. Mergelina s/n., Valladolid, 47011, Spain.
E-mail addresses: javier.gonzalezm@uva.es (J. González-Martín), sara.canteraruizdepellon@wur.nl (S. Cantera), raquel.lebrero@iq.uva.es (R. Lebrero), mutora@iq.uva.es (R. Muñoz).

https://doi.org/10.1016/j.scp.2022.100960
Received 4 October 2022; Received in revised form 7 December 2022; Accepted 26 December 2022
Available online 4 January 2023

2021; Singh et al., 2016), schools (Zhong et al., 2017), libraries (Cincinelli et al., 2016), sports centers (Bralewska et al., 2022), and even in public transport (Gastelum-Arellanez et al., 2021). This group of pollutants include a wide variety of compounds found in indoor spaces at very low and variable concentrations (Tsai, 2019; World Health Organization Regional Office for Europe, 2010). In this context, indoor air purification methods are needed to solve an important Sustainability Development Goal such as the Good Health and Well Being, which includes the increased death rate by indoor air pollution and other illnesses attributable to this problem. The different hydrophobicity, reactivity, and trace level concentrations of indoor air pollutants hinder the effectiveness of conventional physical-chemical technologies for indoor air treatment and represents an opportunity for biotechnologies. As an example, very common indoor VOCs such as toluene or α-pinene have been found in indoor air at average concentrations in the range of 1.8–164 μg m^{-3} and 1.5–32 μg m^{-3}, respectively (González-Martín et al., 2021). Furthermore, sharp concentration peaks are often recorded due to regular indoor activities such as cooking or cleaning.

Biofiltration has emerged as a cost-effective technology for the removal of organic pollutants in off-gases. To date, biofiltration has been successfully applied to the treatment of industrial waste gas streams, which are typically composed of few pollutants at relatively high concentrations (Kennes and Veiga, 2013; Soreanu and Dumont, 2020). The mechanism of VOC biodegradation in biofiltration is based on a first step of mass transfer of the pollutant from the gas phase to the microbial community present in the bioreactor. Then, the pollutant is biodegraded by microorganisms. The gas-microorganism mass transfer is limited for VOCs with poor aqueous solubility or high Henry's law coefficient, which ultimately limits their bioavailability (Ferdowsi et al., 2017; Kraakman et al., 2021; Lamprea Pineda et al., 2021). Innovative bioreactor configurations such as membrane bioreactors or capillary bioreactors have been investigated in order to overcome mass transfer limitations during indoor air treatment. However, these alternative biotechnological platforms require complex and costly systems and have not yet been upscaled (Kraakman et al., 2011). In this context, bioactive coatings can be engineered in conventional packed bed reactors. A bioactive coating consists of a metabolically active microbial community entrapped in a nanoporous polymer matrix (Lyngberg et al., 2001). Therefore, VOC mass transfer is increased since microorganisms are in direct contact with the gas pollutants and pollutant diffusion through an aqueous phase is avoided. Additionally, bioactive coatings allow for the confinement of remarkably high cell densities, which improves the volumetric elimination capacity of VOCs. Other advantages of bioactive coatings are the prevention of an undesired release of microorganisms and their easy implementation onto different support materials (Cortez et al., 2017; Estrada and Quijano, 2020).

Bioactive coatings for air pollution control are still in an embryonic phase. Thus, even though some methods for bioactive coating preparation have been described (Flickinger et al., 2007; Gosse and Flickinger, 2011), only a limited number of successful applications can be found in literature. For instance, Schulte et al. (2016) reported a syngas to acetate bioconversion in a bioactive coating based on *Clostridium ljungdahlii*. In the field of VOC abatement, Estrada et al. (2015) reported the superior performance of latex-based biofilms compared to water-based biofilms during toluene biodegradation by *Pseudomonas putida* F1. Likewise, Gonzalez et al. (González-Martín et al., 2022) recently pointed out the potential of bioactive coatings on polyurethane foam for the simultaneous removal of a mixture of VOCs using a mixed microbial culture enriched from activated sludge in short-term experiments conducted in flat chambers. To date, the continuous abatement of VOCs in bioactive coating based packed bed bioreactors has never been investigated.

This work assessed the potential of bioactive coatings engineered in biofilters and biotrickling filters for the continuous abatement of n-hexane, toluene and α-pinene. These gas pollutants were selected based on their ubiquity in indoor air and different hydrophobicity. The influence of EBRT and inlet VOC concentrations on pollutant biodegradation was investigated.

2. Materials and methods

2.1. Chemicals

Three VOCs were used as model air pollutants: α-pinene (CAS 80-56-8), toluene (CAS 108-88-3) and n-hexane (CAS 110-54-3). The VOCs liquid mixture used throughout the experiment was composed of 22% of α-pinene, 34% of toluene and 44% of n-hexane (%v/v). α-pinene was supplied by Sigma-Aldrich (Madrid, Spain), while n-hexane and toluene were supplied by Panreac® (Barcelona, Spain). The composition of macro and micronutrients in the mineral salt medium (MSM) was described elsewhere (González-Martín et al., 2022). The salts used for MSM preparation were purchased from Panreac® (Barcelona, Spain). The polymer PRIMAL™ SF-208 ER (alkylphenol ethoxylates and biocide free, acrylic-styrene copolymer; solids content 48.05%; pH 8.0–9.5; Dow Chemical, Germany), kindly supplied by Brenntag Química (Barcelona, Spain), was used for the preparation of the bioactive coatings. Two additives were used as osmoprotectant as described in Gosse et al. (2007): D(+)-saccharose purchased from Labkem (Barcelona, Spain), and glycerol purchased from Panreac® (Barcelona, Spain).

2.2. Microorganisms

Two types of microbial communities were used for the preparation of the bioactive coatings: a) an enriched culture obtained after an acclimation period of 3 months of activated sludge in a 3 L stirred tank bioreactor fed with air containing the three VOCs; b) fresh activated sludge obtained from Valladolid wastewater treatment plant.

For the preparation of the bioactive coating, an aliquot of enriched culture or fresh activated sludge was centrifuged at 10,000 rpm for 10 min (4 °C). Then, the biomass pellet (≈1.4 g) was resuspended in 115 mL of PRIMAL™ SF-208 ER, 30 mL of a saccharose solution (0.72 g L^{-1}) and 16 mL of glycerol (100 %V/V). The latex in the mixture of bioreactor 1 was replaced by 35 mL of distilled water to guarantee a uniform distribution of the microorganisms.

2.3. Experimental set-up

The experimental set-up consisted of three PVC column bioreactors (BR1, BR2 and BR3) of 10 cm of internal diameter packed with 32 cm of expanded clay (Arlite Light Plus, LECA Portugal S.A., Portugal), resulting in a working volume of 2.5 L (Fig. 1). The characteristics of this porous packing material were diameter 10–20 mm, bulk density 275 kg m^{-3}, water absorption <25% w/w. The bioreactors were filled with the coated packing material and a 5 cm top layer of plastic Kaldness K1 Micro rings (Evolution Aqua, UK) (ring diameter 1 cm, density as received 0.17 g mL^{-1}, void fraction 83%, water-holding capacity on a volume basis 11%) to ensure a homogeneous supply of the recirculating MSM. Expanded clay beads were first cleaned with water, dried for 24 h and then hydrated in fresh MSM for 8 h before usage. For each bioreactor, 2.5 L of expanded clay beads were homogeneously coated with the corresponding microbial mixture, and then left to dry overnight before the start of the experiment. BR1 was set as a control by replacing the latex by water, while bioactive coatings were used in BR2 and BR3. The enriched microorganisms were used in BR1 and BR2, while BR3 was inoculated with fresh activated sludge. The air flow, which was supplied by an air compressor (ABAC B2500-50 2, Italy), was humidified in a 1 m bubble column. Air rotameters (Aalborg, New York, USA) were used to control the air flow of each bioreactor. The mixture of liquid VOCs was supplied using a syringe pump (Fusion 100, Chemyx Inc., USA) and glass liquid syringes of different volumes depending on the EBRT and VOC concentration (Hamilton, USA). The VOCs injection flowrate varied between 12 and 100 μL h^{-1}, depending on the operational stage. The operational parameters are summarized in Table 1.

The bioreactors were operated in a biofilter configuration with daily irrigation of fresh MSM (100 mL day^{-1}, divided in 16 additions) from stage 1–4. Then, the bioreactors were operated in a biotrickling filter configuration from stages 5–8 under continuous MSM recirculation at 8 L day^{-1}. A multichannel peristaltic pump (model 205S; Watson-Marlow Limited, Falmouth, UK) was used for MSM irrigation and recirculation. The EBRT in the biofilters was reduced from 120 to 60, 30 and 15 s in stages 1, 2, 3 and 4, respectively, at a constant inlet concentration of hexane, pinene and toluene of 9.1 ± 3.4, 15.2 ± 2.3 and 15.7 ± 3.2 mg m^{-3}. In stage 5, VOC concentration and EBRT were maintained and the liquid recirculation was set at 8 L day^{-1}. Then, the concentrations of hexane, pinene and toluene were progressively reduced to 5.8 ± 0.7, 7.7 ± 0.9 and 8.4 ± 1.1 mg m^{-3} in stage 6, 2.9 ± 0.4, 4.2 ± 0.6 and 4.0 ± 0.8 mg m^{-3} in stage 7 and 1.5 ± 0.2, 2.0 ± 0.2 and 1.9 ± 0.1 mg m^{-3} in stage 8, at a constant EBRT of 15 s. Although indoor air VOC concentrations are usually reported in the range of μg m^{-3}, concentrations in the range of 1–20 mg m^{-3} have been used in this work. These higher concentrations have been preferred in order to enhance the mass transfer and also facilitate the analysis by GC-FID. Gas sampling bulbs (Sigma-Aldrich, Madrid, Spain) of 250 mL were placed at the inlet and outlet air stream of each bioreactor to monitor VOC concentration by solid phase microextraction (SPME) coupled with GC-FID. The bioreactors were maintained at 22 ± 2 °C throughout the 218 days of experiment.

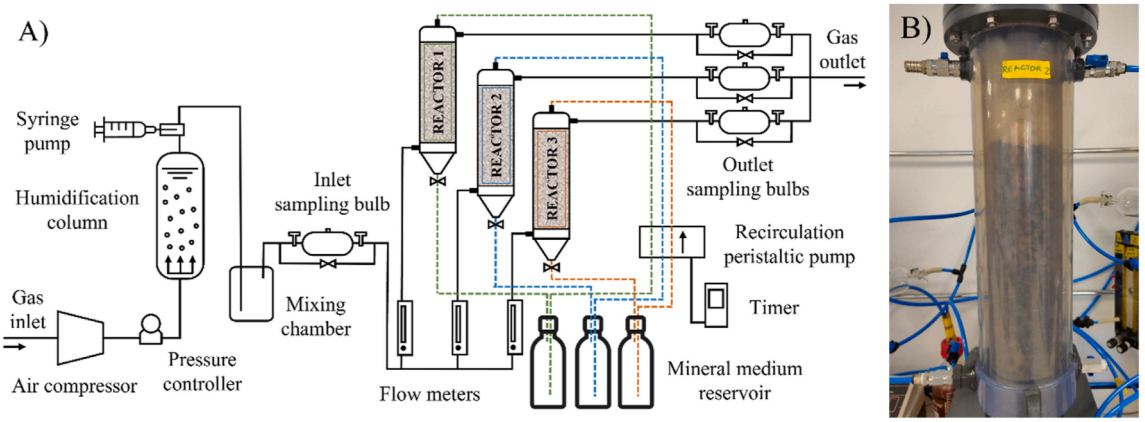

Fig. 1. A) Schematic overview of the experimental set-up and B) picture of a bioreactor.

Table 1
Operational parameters evaluated in the three bioreactors. Inlet concentrations (Cin) are provided with their standard deviation in each stage.

Stage	Q_{gas} (L min^{-1})	EBRT (s)	Cin_{Hex} (mg m^{-3})	Cin_{Tol} (mg m^{-3})	Cin_{Pin} (mg m^{-3})	Irrigation	Elapsed time (days)
1	1.25	120	7.8 (1.3)	14.6 (2.2)	13.4 (3.2)	100 mL MSM day^{-1} at 6.25 mL intervals	41
2	2.5	60	8.2 (0.8)	14.9 (1.1)	18.3 (1.5)		24
3	5	30	10.8 (2.5)	16.7 (2.7)	17.5 (2.8)		36
4	10	15	14.3 (3.1)	14.6 (1.6)	15.1 (1.7)		38
5	10	15	14.2 (1.9)	14.4 (1.5)	15.9 (2.6)	MSM recirculation at 8 L day^{-1}	31
6	10	15	5.8 (0.7)	7.7 (0.9)	8.4 (1.1)		19
7	10	15	2.9 (0.4)	4.2 (0.6)	4.0 (0.8)		14
8	10	15	1.5 (0.2)	2.0 (0.2)	1.9 (0.1)		15

2.4. Analytical procedures

The inlet and outlet concentrations of toluene, pinene and hexane were measured daily by SPME-GC-FID. First, a 10 min preconcentration step was performed by exposing the SPME fiber (85 μm CAR/PDMS; Supelco, Bellefonte, USA) to the VOC laden emission in 250 mL glass bulbs. Then, VOC analysis was carried out in a GC-FID (Varian 3900) equipped with an Agilent HP-5MSI capillary column (30 m × 0.25mm × 0.25 μm) according to a previous study (González-Martín et al., 2022). A blank injection was performed before sampling to clean the SPME fiber. New fibers were conditioned prior to first usage at 300 °C for 1 h at the GC-FID injector. The fibers were then calibrated with external standards of the VOCs, prepared in 250 mL glass bulbs, following the SPME-GC-FID method above mentioned. The concentration of biomass in the aliquots was determined according to Standard Method 2540 D (Standard Methods Committee of the American Public Health Association et al., 2018).

2.5. Bacterial community analysis

Samples of 20 mL of each bioreactor were withdrawn in duplicate for microbial analysis. Cells were detached from the packing material with distilled water using a gentle shaking (day 218). The biomass was then centrifuged at 13,000 × *g* for 10 min and preserved at −20 °C for bacterial population analysis. DNA extraction and Illumina Miseq amplicon sequencing were carried out in the Foundation for the Promotion of Health and Biomedical Research of the Valencia Region (FISABIO, Spain). Amplicon sequencing to analyze the bacterial population was developed targeting the 16S V3 and V4 regions (464bp, *Escherichia coli*-based coordinates) with the bacterial primers S-D-Bact-0341-b-S-17 and S-D-Bact-0785-a- A-21. Libraries were prepared and sequenced on a Miseq-Illumina platform using nucleotide paired-end reads at FISABIO (Spain). Mothur v.1.47.0 was used to process and quality filtered the sequences (Kozich et al., 2013). Afterwards, bacterial sequences were clustered into Operational Taxonomic Units (OTUs) using the SILVA 16S rRNA gene reference database (Version: 138.1) at 97% identity threshold and the ribosomal data base project (version 18) using Mothur v.1.47.0 (Cole et al., 2014; Quast et al., 2012). The data have been deposited with links to BioProject accession number PRJNA841443 in the NCBI BioProject database (https://www.ncbi.nlm.nih.gov/bioproject/). Community structure and hierarchical relations were plotted with R version 3.6.1 (R Core Team, 2019). The main genera are shown in heatmaps plotted using the package *pheatmap* (Kolde, 2019).

2.6. Data treatment

The removal efficiency (RE) of the bioreactors for each VOC was calculated using Eq. (1):

$$RE = 100 \times \left(1 - \left(Cout_i / Cin_i\right)\right) \quad \text{Eq. 1}$$

where Cin_i and $Cout_i$ are the inlet and outlet concentrations (in mg m^{-3}) of the target VOCs, respectively.

3. Results and discussion

3.1. Influence of EBRT on VOC removal

The REs achieved in BR1, BR2 and BR3 in stages 1–5 are presented in Fig. 2. Average RE values at steady state of each stage are summarized in Table S1. Daily inlet and outlet concentrations are represented in Fig. S1 (stages 1–5) and S2 (stages 5–8). Stage 1 exhibited an acclimation period for the microbial communities inoculated in the bioreactors. Toluene and pinene adsorption onto the packing material resulted in high initial REs in BR1 (91.1% and 77.7%) and BR2 (77.0% and 57.0%), with lower values recorded in BR3 (47.0% and 25.5%). Then, the REs of toluene and pinene rapidly decreased in the three bioreactors to minimum values of 40.3% and 27.7% in BR1, 17.7% and 1.7% in BR2, and 7.4% and 0.0% in BR3, respectively. From day 10 onwards, the REs of both pollutants increased again to reach steady state values of ≈90% in the three bioreactors within the fourth week of operation. The low removal performance recorded could be attributed to a long initial growth lag phase of the microorganisms as a result of their adaptation to the new environmental conditions prevailing in the reactor (e.g. growth as biofilm, partial desiccation, etc.). In the case of BR3, a first period of enrichment driven to select those organisms able to use VOCs as the carbon/energy source was encountered. Thus, the lag phase of VOC degradation was longer. Similarly, startup periods of up to several weeks have been commonly reported for gas-phase bioreactors treating VOCs (Yang et al., 2011; Zhao et al., 2011). Interestingly, a high hexane removal was only initially observed in BR1 (86.2%), which rapidly decreased to remain < 5% during stage 1. BR2 and BR3 only supported average REs lower than 5%. Previous batch experiments carried out during the enrichment of the inoculum of BR1 and BR2 (Fig. S3) showed that the microbial population had the capacity to remove hexane at high concentrations, which suggested that the high hydrophobicity and the low mass transfer gradient of hexane hindered the growth and dominance of hexane degraders in the bioreactors in comparison with other microbial members.

The EBRT was reduced to 60 s in stage 2. BR1 and BR2 supported REs of toluene and pinene over 90%, but in BR3, toluene and pinene REs slightly decreased to average values of 89.9 ± 1.6% and 87.3 ± 4.1%, respectively. Similar to stage 1, no significant hexane removal was observed in any of the bioreactors.

The reduction in EBRT from 60 to 30 s in stage 3 resulted in a temporary decrease in the REs in BR1 and BR2 down to 85.1% and 84.7% for toluene, and 66.1% and 68.7% for pinene, respectively. Then, the REs gradually recovered within a week to finally reach steady state values of 91.3 ± 3.0% for toluene and 97.9 ± 2.3% for pinene in BR1, and 90.3 ± 2.8% for toluene and 92.5 ± 6.4% for pinene in BR2. This transitory deterioration in the VOC abatement efficiency when decreasing the EBRT has been previously observed in other gas-phase bioreactors aerobically treating VOCs (Lebrero et al., 2012, 2013, 2014). However, BR3, containing the bioactive coating with activated sludge, only reached steady state values of 67.0 ± 7.8% for toluene and 49.8 ± 12.0% for pinene.

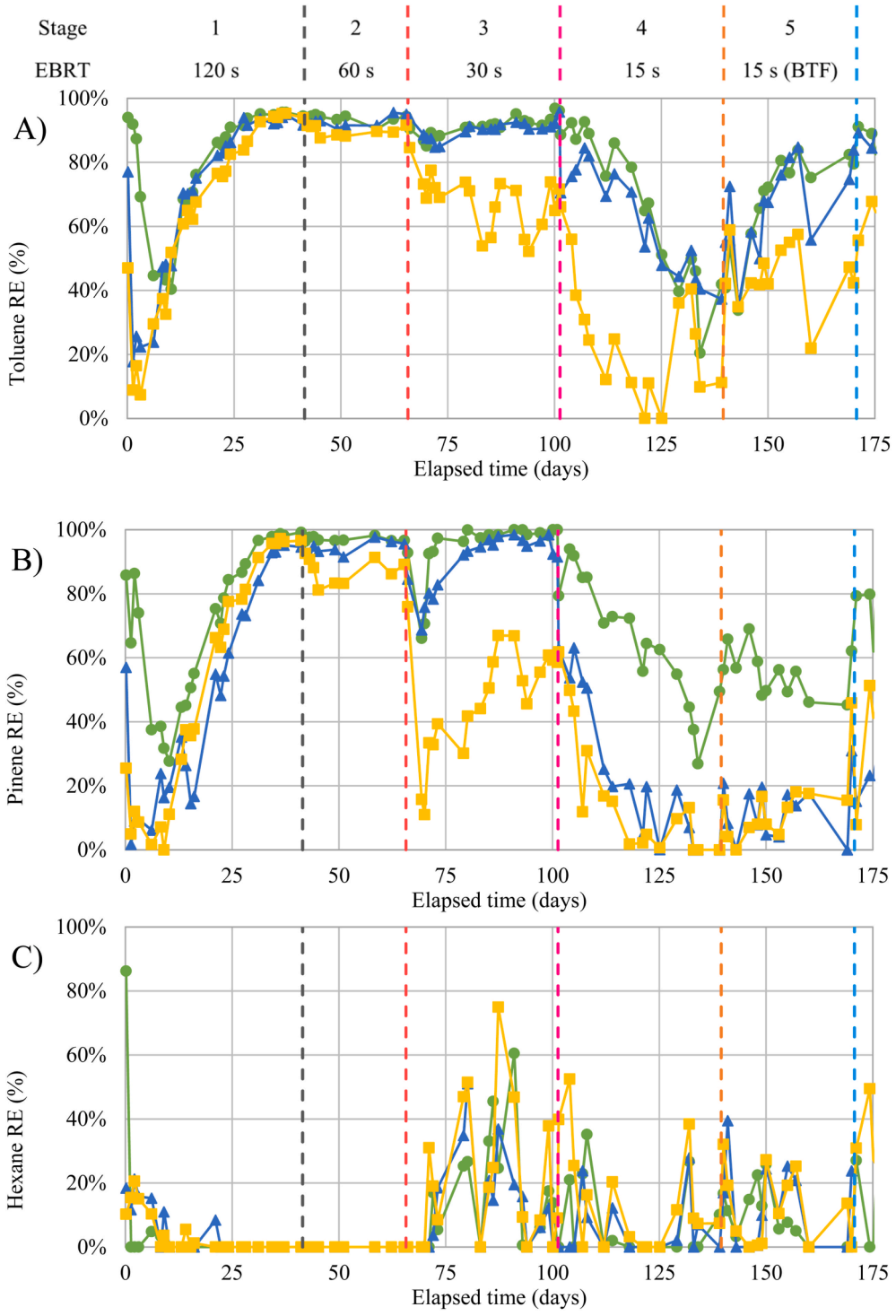

Fig. 2. Time course of the removal efficiency of A) toluene, B) pinene and C) hexane in stages 1 (EBRT = 120 s), stage 2 (EBRT = 60 s), stage 3 (EBRT = 30 s), stage 4 (EBRT = 15 s) and stage 5 in BR1 (—●—), BR2 (—▲—) and BR3 (—■—).

This phenomenon was likely caused by the lack of specialization of the activated sludge used for VOC biodegradation compared to BR1 and BR2, where VOC degraders were previously enriched and selected. A slight increase in hexane removal was observed in stage 3, which might be due to the gradual establishment of a hexane degrading community.

Process operation at an EBRT of 15 s in stage 4 entailed a gradual deterioration in VOC REs. Thus, toluene and pinene REs decreased to 41.5 ± 11.2% and 55.7 ± 15.0% in BR1, to 44.3 ± 5.4% and 10.5 ± 10.2% in BR2, and to 22.2 ± 16.1% and

8.2 ± 9.3% in BR3, while hexane removal followed the same trend than in stage 3. This deterioration likely resulted from the progressive loss of moisture in the bioactive coating due to the higher air flowrate, which could have deteriorated the metabolic activity of the VOC degrading microorganisms. The shorter gas-microorganism contact time might be also responsible of the decrease in VOC elimination. Despite the different nature of natural biofilms and bioactive coatings, toluene REs were similar in BR1 and BR2. However, lower pinene REs were recorded in BR2. This finding was not in agreement with previous experiments where the bioactive coating provided an enhanced resistance to the desiccation caused by increases in air flowrate over a conventional biofilm (González-Martín et al., 2022), thus further research is needed to elucidate this effect.

Adapting the operation of the bioreactors from a biofilter (discontinuous irrigation of 100 mL day^{-1}) to a biotrickling filter configuration (continuous liquid recirculation at 8 L day^{-1}) in stage 5 resulted in an increase in toluene REs from 41.5 ± 11.2% to 76.3 ± 5.9% in BR1, from 44.3 ± 5.4% to 74.0 ± 9.9% in BR2, and from 22.2 ± 16.1% to 45.1 ± 10.0% in BR3. However, the REs of pinene remained similar in all bioreactors. As observed in stage 4, the variability in hexane REs was very high. The improvement in toluene removal could be attributed to the increase in the irrigation rate, which enhanced the catabolic activity of the microbial population. On the contrary, the higher hydrophobicity of pinene ($H = 2.1 \times 10^{-4}$ mol m^{-3} Pa^{-1}) and hexane ($H = 6.0 \times 10^{-4}$ mol m^{-3} Pa^{-1}) compared to that of toluene ($H = 1.5 \times 10^{-3}$ mol m^{-3} Pa^{-1}) hindered a further improvement of REs (Sander, 2015).

Overall, the biofilter containing an aqueous biofilm achieved comparable VOC removals and shorter acclimation periods than the bioactive coatings. Similar abatement performances between natural biofilms and bioactive coatings were reported in previous work (González-Martín et al., 2022). Moreover, toluene and pinene removals in the biofilter with activated sludge bioactive coating (BR3) were always lower than those recorded in BR1 and BR2, and longer acclimation periods to EBRT reductions were obtained. This fact suggested that the microbial inoculum in bioactive coatings is a key factor determining the bioreactor performance and confirmed the relevance of the acclimation period to the selected VOCs.

3.2. Influence of inlet concentrations on VOC removal

The REs achieved by BR1, BR2 and BR3 in stages 5–8 are represented in Fig. 3. In stage 5, BR1 and BR2 achieved similar steady state REs for toluene (76.3 ± 5.9% and 74.0 ± 9.9%, respectively), while BR3 only supported a moderate toluene removal (45.1 ± 10.0%). A significantly higher removal of pinene was observed in BR1 (55.4 ± 7.4%) compared to BR2 (12.5 ± 9.2%) and BR3 (13.4 ± 11.4%), while the low removal of hexane was similar to previous stages.

The first decrease in hexane, toluene and pinene inlet concentrations to 5.8 ± 0.7, 7.7 ± 0.9, 8.4 ± 1.1 mg m^{-3} in stage 6 mediated an improvement of ≈10% in toluene and of ≈30% in pinene REs in the three bioreactors. Indeed, toluene removal increased to 87.1 ± 2.7% in BR1, 86.5 ± 2.7 in BR2 and 58.8 ± 7.0% in BR3. Interestingly, pinene removal increased much faster up to steady state REs of 83.1 ± 8.4% in BR1, 44.2 ± 10.3% in BR2 and 43.4 ± 5.2% in BR3. Hexane REs remained at ≈10%, with a high variability. Although the mass transfer gradient decreased, the reduction in VOC inlet concentrations supported the higher REs observed.

In stage 7, the inlet concentrations of hexane, toluene and pinene were set at 2.9 ± 0.4, 4.2 ± 0.6 and 4.0 ± 0.8 mg m^{-3}, respectively. In this context, toluene average REs slightly decreased in BR1 and BR2 (85.4 ± 2.1% and 79.0 ± 4.0%), while BR3 supported a higher removal than in stage 6 (61.9 ± 9.0%). On the contrary, pinene REs improved from 83.1 ± 8.4% to 90.9 ± 2.3% in BR1, exhibiting a considerably higher enhancement in BR2 and BR3 (from 44.2 ± 10.3% to 76.5 ± 3.2% and from 43.4 ± 5.2% to 70.6 ± 8.7%, respectively). Highly variable hexane REs were again observed in this stage.

Finally, the inlet concentrations were reduced to 1.5 ± 0.2 mg m^{-3} for hexane, 2.0 ± 0.2 mg m^{-3} for toluene and 1.9 ± 0.1 mg m^{-3} for pinene in stage 8. Toluene REs continued decreasing to 70.8 ± 6.4% and 62.0 ± 12.7% in BR1 and BR2, respectively and remained similar in BR3 (64.2 ± 6.5%). Pinene REs slightly decreased in BR1 to 83.5 ± 6.2% and remained constant in BR2 (76.6 ± 8.1%) and BR3 (70.5 ± 4.6%), compared to stage 7. In this context, toluene and pinene REs over 90% have been reported at EBRTs shorter than 15 s and concentrations of 0.82 ± 0.07 and 0.91 ± 0.10 mg m^{-3} (Lebrero et al., 2014) and 1.32 ± 0.10 and 1.31 ± 0.14 mg m^{-3} (Lebrero et al., 2013). The lower removals during stages 5–8 were likely caused by the deterioration and/or lack of adaption of the microbial community to decreasing EBRTs at stages 1–4, which may have caused partial desiccation of the system.

Overall, no consistent hexane removal was observed in any of the bioreactors regardless of the decreasing VOC concentrations. Hexane treatment in gas-phase bioreactors inoculated with activated sludge has been previously reported at higher inlet concentrations of up to 1700 mg m^{-3} (Amin et al., 2017; Cheng et al., 2020; Zhao et al., 2011). At low concentrations, robust hexane removals were achieved in a biofilter (93.7 ± 0.7%) at an EBRT of 8 s, in a biotrickling filter (88.4 ± 1.1%) at an EBRT of 4 s, and in a membrane bioreactor (38.3 ± 6.2%) at an EBRT of 18 s (Lebrero et al., 2014) at inlet concentration of 0.75 ± 0.08 mg m^{-3}. Hexane treatment was also demonstrated in a biotrickling filter, achieving REs of 84 ± 2% and 60 ± 5% at average inlet concentrations of 1.27 ± 0.11 mg m^{-3} and EBRTs of 12 and 6 s, respectively (Lebrero et al., 2013). The concept of bioactive coatings in continuous biofiltration was confirmed as valid, as the removal efficiencies of toluene and pinene achieved by the bioactive coating system were close to the removals achieved by the traditional biofilm-based bioreactor. However, no clear improvement was observed in the treatment of hexane, the most hydrophobic VOC in this work, which requires further research to benefit from the potential of bioactive coatings. Further research is therefore needed to clarify the cause underlying the poor removal obtained in our bioreactor configurations.

3.3. Bacterial community structure

The analysis of the bacterial community in the sample set showed that the most diverse community belonged to BR3 and the population found in this reactor was the most dissimilar to the rest (Fig. 4). This fact can be explained by the 3-month acclimation period of the inoculum prior inoculation of BR1 and BR2.

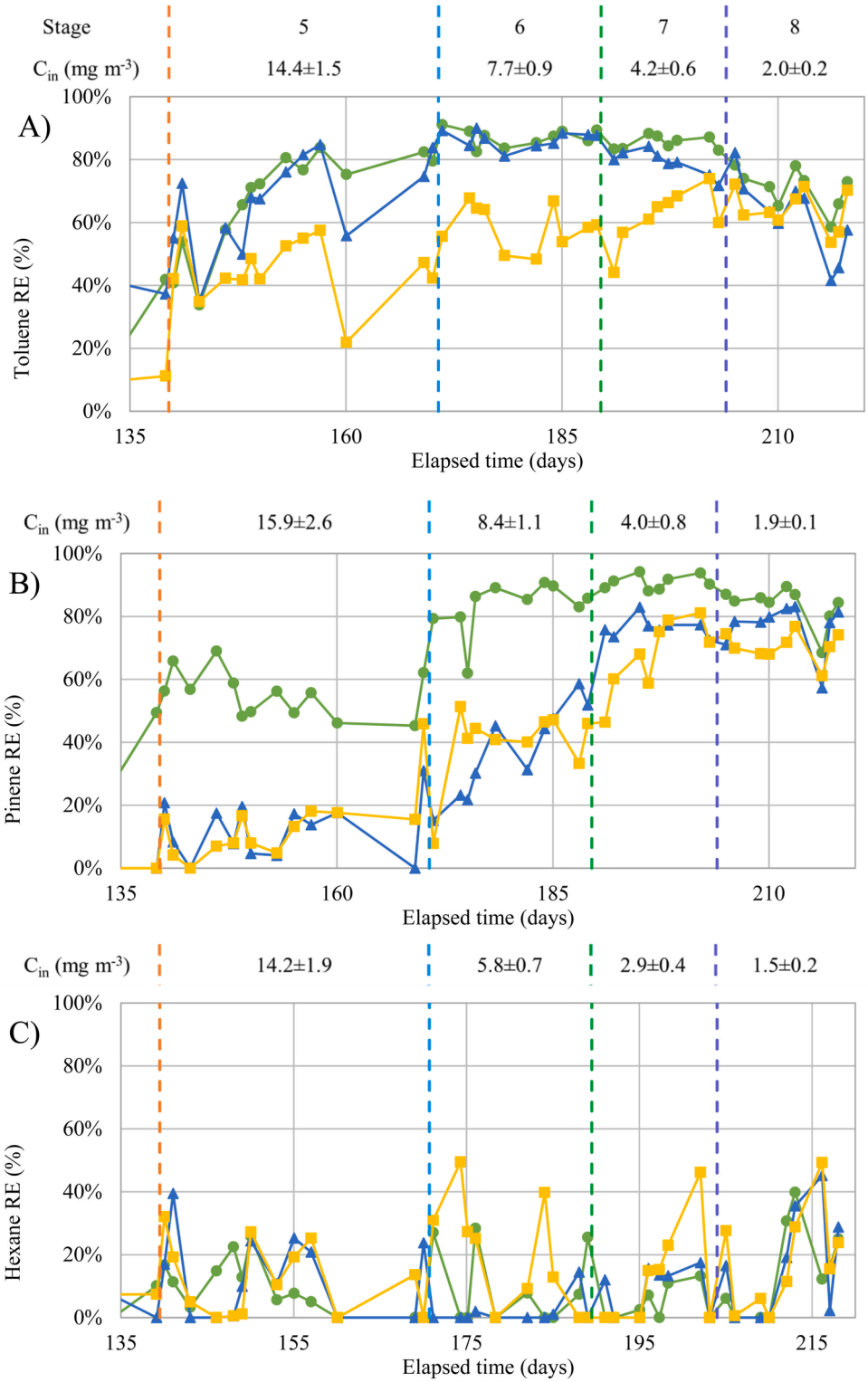

Fig. 3. Time course of the removal efficiency of A) toluene, B) pinene and C) hexane during stages 5–8 (dotted lines) in BR1 (—●—), BR2 (—▲—) and BR3 (—■—).

The prior adaptation and enrichment of the inoculum used in BR1 and BR2 towards the metabolism of aromatic VOCs and terpenes resulted in similar populations by the end of operation. However, the presence of latex in BR2 promoted the growth of some populations over others. The most representative bacteria found in BR1 belonged to the genus *Devosia* (9.0%) and uncultured members of *Rhodobacteraceae* (8.7%). The genera *Sphingopyxis* (5.7%), *Rhodococcus* (5.5%), *Lacipirellula* (4.8%), *Mycobacterium* (4.6%) and

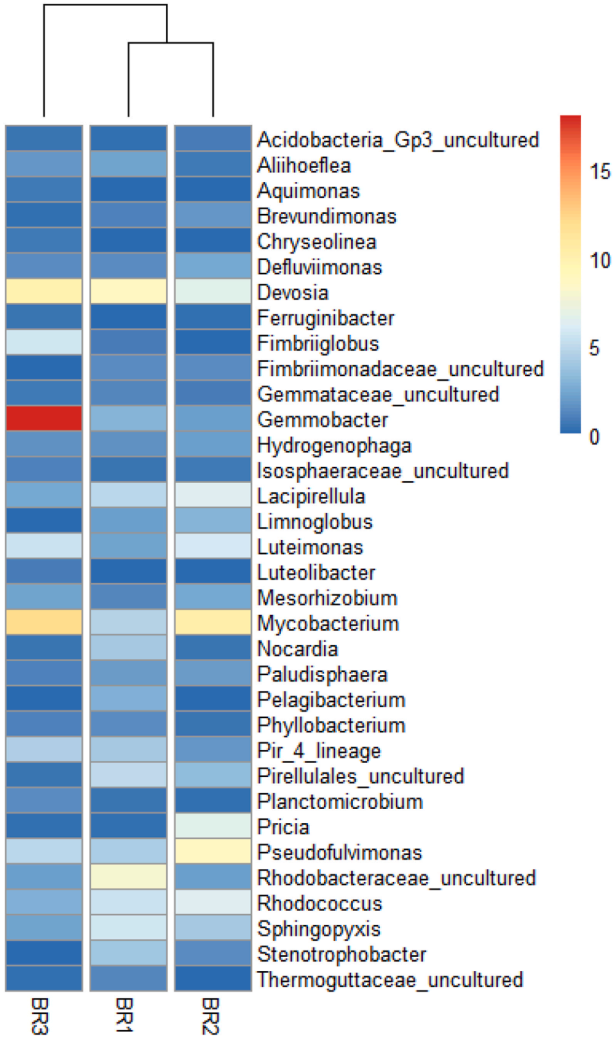

Fig. 4. Heat map of the most representative bacterial genera (99% of the total genera) in the three bioreactors. The dendrogram on top represents hierarchical clustering of the samples. Data is presented as the relative abundance (%).

Pseudofulvimonas (4.3%) were also present in BR1. In BR2, the genera *Mycobacterium* (10.2%) and *Pseudofulvimonas* (8.6%) were more abundant than in BR1, while the representation of members of *Devosia* (6.7%) and uncultured members of *Rhodobacteraceae* (2.0%) decreased. *Rhodococcus* and *Lacipirellula* (6.2 and 6.1%, respectively) had a similar relative abundance than in BR1. Moreover, the abundance of the genus *Pricia* (6.7%) was significant in BR2, although this genus was not representative in the other reactors (<0.2%). On the other hand, the most representative bacterial genus in BR3 was *Gemmobacter* (17.9%), likely as a result of the inoculation with not acclimatized fresh activated sludge. Members of the genera *Mycobacterium* and *Devosia* were also abundant in this reactor (12% and 9.9%, respectively). Other representative genera in BR3 were *Fimbriiglobus* (5.7%), *Luteimonas* (5.5%) and *Pseudofulvimonas* (4.9%).

Broadly, typical bacterial genera related to VOCs degradation were abundant in BR1 and BR2. Several bacteria from the family *Rhodobacteraceae* and *Sphingomonadaceae*, such as *Rhodococcus*, *Paracoccus* and *Sphingopyxis*, are well known degraders of terpenes and toluene (Lee and Cho, 2009; Zhao et al., 2013). Some *Rhodococcus* species have been also demonstrated to remove n-hexane (Lee and Cho, 2009). *Mycobacterium* is able to efficiently grow with toluene and different aromatic compounds as carbon/energy source (Kastner et al., 1999; Tay et al., 1998) and *Devosia* has the genes for the utilization of toluene and some terpenes (Talwar et al., 2020). BR3 was dominated by members of the genus *Gemmobacter*. This genus, although is usually found in bioreactors for waste treatment, to the best of our knowledge, does not possess the genes for the degradation of toluene and terpenes (Chen et al., 2013). Most likely, the population of *Gemmobacter* was abundant in the aerobic sludge used as inoculum, but it was not directly involved in VOC degradation. Nevertheless, known toluene and pinene degraders were also abundant in BR3, such as *Mycobacterium* and *Devosia.* This fact promoted the degradation of pinene and toluene in BR3. The lack of hexane degradation in all the reactors, though, could be related to the absence of metabolic functionality involved in the removal of this hydrophobic gas in the inoculum. Although hexane degradation was observed in the inoculum of BR1 and BR2, probably the low concentrations of hexane available for the microorganisms during

operation hindered the growth of hexane degraders. These ones most likely could not compete for the niche resources with those bacteria that had more carbon and energy available.

4. Conclusions

To the best of our knowledge, this study represents the first long-term evaluation of bioactive coating-based biofiltration for indoor air pollution treatment. Bioactive coatings implemented in biofilters supported a similar VOC abatement performance than conventional biofilm-based bioreactors at decreasing EBRTs and inlet concentrations. Steady state toluene and pinene REs over 90% were achieved by conventional and bioactive coating biofilters at EBRTs of 60, 45 and 30 s. Toluene removals decreased to ≈40% at a EBRT of 15 s, while pinene REs decreased to 55.7% and 10.5% in conventional and bioactive coating biofilters, respectively. The stepwise decrease in VOC inlet concentration resulted in a progressive improvement of pinene degradation performance in the bioactive coating biofilter, while toluene REs gradually increased over 80% in both bioreactors. Overall, the use of biomass not previously acclimated to VOC removal during bioactive coating preparation mediated lower VOC REs and longer stabilization periods compared to the bioreactors inoculated with pre-acclimated cultures. Poor hexane removals were recorded regardless of the type of inoculum and operational conditions, which were attributed to the extremely high hydrophobicity of this VOC and the low inlet concentrations used in the experiment (thus hindering the growth of hexane degraders). New bioactive coatings formulations need to be investigated to further overcome mass transfer limitations of highly hydrophobic VOCs at low concentrations. Future work should focus as well on the elimination of VOCs at even lower concentrations, more representative of indoor air. In addition, special attention should be brought to the identification of microbial VOCs (Veselova et al., 2019). These secondary pollutants are originated during the metabolism of the microorganisms used in the biofiltration devices and could be more harmful than the original pollutants.

Author contributions statement

Javier González-Martín: Conceptualization; Formal analysis; Investigation; Writing - Original Draft; Writing - Review & Editing; Visualization, Sara Cantera: Formal analysis; Writing - Original Draft; Writing - Review & Editing, Raquel Lebrero: Conceptualization; Writing - Review & Editing; Supervision; Funding acquisition; Project administration, Raúl Muñoz: Conceptualization; Writing - Review & Editing; Supervision; Funding acquisition; Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This work was supported by the Ministry of Science, Innovation and Universities [project RTI2018-0-096441-B-I00]. The Regional Government of Castilla y León and the EU-FEDER program [grant number CLU 2017–09, CL-EI-2021-07 and UIC 315] are also gratefully acknowledged. The financial support of the Regional Government of Castilla y León (Consejería de Educación) and the European Social Fund is also acknowledged for the PhD grant of Javier González-Martín (BDNS 487971). This work was also supported by the European Commission-H2020-MSCA–IF–2019 grant (ENHANCEMENT, ref. 897284) attributed to S. Cantera.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scp.2022.100960.

References

Amin, M.M., Rahimi, A., Bina, B., Nourmoradi, H., Hassanvand, M.S., Mohammadi-Moghadam, F., Norouzi, S., Heidari, M., 2017. Biodegradation of n -hexane as single pollutant and in a mixture with BTEX in a scoria/compost-based biofilter. Process Saf. Environ. Protect. 107, 508–517. https://doi.org/10.1016/j.psep.2017.03.019.

Bralewska, K., Rogula-Kozłowska, W., Bralewski, A., 2022. Indoor air quality in sports center: assessment of gaseous pollutants. Build. Environ. 208, 108589. https://doi.org/10.1016/j.buildenv.2021.108589.

Chen, W.-M., Cho, N.-T., Huang, W.-C., Young, C.-C., Sheu, S.-Y., 2013. Description of Gemmobacter fontiphilus sp. nov., isolated from a freshwater spring, reclassification of Catellibacterium nectariphilum as Gemmobacter nectariphilus comb. nov., Catellibacterium changlense as Gemmobacter changlensis comb. nov. Catellibacte. Int. J. Syst. Evol. Microbiol. 63, 470–478. https://doi.org/10.1099/ijs.0.042051-0.

Cheng, Y., Li, X., Liu, H., Yang, C., Wu, S., Du, C., Nie, L., Zhong, Y., 2020. Effect of presence of hydrophilic volatile organic compounds on removal of hydrophobic n-hexane in biotrickling filters. Chemosphere 252, 126490. https://doi.org/10.1016/j.chemosphere.2020.126490.

Cincinelli, A., Martellini, T., Amore, A., Dei, L., Marrazza, G., Carretti, E., Belosi, F., Ravegnani, F., Leva, P., 2016. Measurement of volatile organic compounds (VOCs) in libraries and archives in Florence (Italy). Sci. Total Environ. 572, 333–339. https://doi.org/10.1016/j.scitotenv.2016.07.201.

Cole, J.R., Wang, Q., Fish, J.A., Chai, B., McGarrell, D.M., Sun, Y., Brown, C.T., Porras-Alfaro, A., Kuske, C.R., Tiedje, J.M., 2014. Ribosomal Database Project: data and tools for high throughput rRNA analysis. Nucleic Acids Res. 42, D633–D642. https://doi.org/10.1093/nar/gkt1244.

Cortez, S., Nicolau, A., Flickinger, M.C., Mota, M., 2017. Biocoatings: a new challenge for environmental biotechnology. Biochem. Eng. J. 121, 25–37. https://doi.org/10.1016/j.bej.2017.01.004.

Estrada, J.M., Bernal, O.I., Flickinger, M.C., Muñoz, R., Deshusses, M.A., 2015. Biocatalytic coatings for air pollution control: a proof of concept study on VOC

biodegradation. Biotechnol. Bioeng. 112, 263–271. https://doi.org/10.1002/bit.25353.
Estrada, J.M., Quijano, G., 2020. Bioremediation of air using microorganisms immobilized in bedding nanomaterials. In: Nanomaterials for Air Remediation. Elsevier, pp. 211–225. https://doi.org/10.1016/B978-0-12-818821-7.00011-7.
European Environment Agency, 2020. Air Quality in Europe: 2020 Report. EEA Report. Publications Office, Luxembourg. https://doi.org/10.2800/786656.
Ferdowsi, M., Avalos Ramirez, A., Jones, J.P., Heitz, M., 2017. Elimination of mass transfer and kinetic limited organic pollutants in biofilters: a review. Int. Biodeterior. Biodegrad. 119, 336–348. https://doi.org/10.1016/j.ibiod.2016.10.015.
Flickinger, M.C., Schottel, J.L., Bond, D.R., Aksan, A., Scriven, L.E., 2007. Painting and printing living bacteria: engineering nanoporous biocatalytic coatings to preserve microbial viability and intensify reactivity. Biotechnol. Prog. 23, 2–17. https://doi.org/10.1021/bp060347r.
Gastelum-Arellanez, A., Esquivel-Días, J., Lopez-Padilla, R., Robledo, V.H., Paulina, R., Beltrán, M.F., Saucedo-Lucero, J.O., 2021. Assessment of persistent indoor VOCs inside public transport during winter season. Chemosphere 263, 128127. https://doi.org/10.1016/j.chemosphere.2020.128127.
Ghaffarianhoseini, Amirhosein, AlWaer, H., Omrany, H., Ghaffarianhoseini, Ali, Alalouch, C., Clements-Croome, D., Tookey, J., 2018. Sick building syndrome: are we doing enough? Architect. Sci. Rev. 61, 99–121. https://doi.org/10.1080/00038628.2018.1461060.
González-Martín, J., Cantera, S., Lebrero, R., Muñoz, R., 2022. Optimization of acrylic-styrene latex-based biofilms as a platform for biological indoor air treatment. Chemosphere 287, 132182. https://doi.org/10.1016/j.chemosphere.2021.132182.
González-Martín, J., Kraakman, N.J.R., Pérez, C., Lebrero, R., Muñoz, R., 2021. A state–of–the-art review on indoor air pollution and strategies for indoor air pollution control. Chemosphere 262, 128376. https://doi.org/10.1016/j.chemosphere.2020.128376.
Gosse, J.L., Engel, B.J., Rey, F.E., Harwood, C.S., Scriven, L.E., Flickinger, M.C., 2007. Hydrogen production by photoreactive nanoporous latex coatings of nongrowing rhodopseudomonas palustris CGA009. Biotechnol. Prog. 23, 124–130. https://doi.org/10.1021/bp060254.
Gosse, J.L., Flickinger, M.C., 2011. Uniform lab-scale biocatalytic nanoporous latex coatings for reactive microorganisms. In: Wang, P. (Ed.), Nanoscale Biocatalysis (Methods and Protocols). Humana Press, pp. 213–222. https://doi.org/10.1007/978-1-61779-132-1_17.
Kastner, J.R., Thompson, D.N., Cherry, R.S., 1999. Water-soluble polymer for increasing the biodegradation of sparingly soluble vapors. Enzym. Microb. Technol. 24, 104–110. https://doi.org/10.1016/S0141-0229(98)00108-2.
Kennes, C., Veiga, M.C., 2013. Air Pollution Prevention and Control: Bioreactors and Bioenergy, Sustainability. Wiley. https://doi.org/10.3390/su5052272.
Kolde, R., 2019. Pheatmap: Pretty Heatmaps. R Package Version 1.0.12. URL. https://cran.r-project.org/package=pheatmap.
Kozich, J.J., Westcott, S.L., Baxter, N.T., Highlander, S.K., Schloss, P.D., 2013. Development of a dual-index sequencing strategy and curation pipeline for analyzing amplicon sequence data on the MiSeq Illumina sequencing platform. Appl. Environ. Microbiol. 79, 5112–5120. https://doi.org/10.1128/AEM.01043-13.
Kraakman, N.J.R., González-Martín, J., Pérez, C., Lebrero, R., Muñoz, R., 2021. Recent advances in biological systems for improving indoor air quality. Rev. Environ. Sci. Bio/Technology 20, 363–387. https://doi.org/10.1007/s11157-021-09569-x.
Kraakman, N.J.R., Rocha-Rios, J., van Loosdrecht, M.C.M., 2011. Review of mass transfer aspects for biological gas treatment. Appl. Microbiol. Biotechnol. 91, 873–886. https://doi.org/10.1007/s00253-011-3365-5.
Lamprea Pineda, P.A., Demeestere, K., Toledo, M., Van Langenhove, H., Walgraeve, C., 2021. Enhanced removal of hydrophobic volatile organic compounds in biofilters and biotrickling filters: a review on the use of surfactants and the addition of hydrophilic compounds. Chemosphere 279, 130757. https://doi.org/10.1016/j.chemosphere.2021.130757.
Lebrero, R., Gondim, A.C., Pérez, R., García-Encina, P.A., Muñoz, R., 2014. Comparative assessment of a biofilter, a biotrickling filter and a hollow fiber membrane bioreactor for odor treatment in wastewater treatment plants. Water Res. 49, 339–350. https://doi.org/10.1016/j.watres.2013.09.055.
Lebrero, R., Rodríguez, E., Estrada, J.M., García-Encina, P.A., Muñoz, R., 2012. Odor abatement in biotrickling filters: effect of the EBRT on methyl mercaptan and hydrophobic VOCs removal. Bioresour. Technol. 109, 38–45. https://doi.org/10.1016/j.biortech.2012.01.052.
Lebrero, R., Rodríguez, E., Pérez, R., García-Encina, P.A., Muñoz, R., 2013. Abatement of odorant compounds in one- and two-phase biotrickling filters under steady and transient conditions. Appl. Microbiol. Biotechnol. 97, 4627–4638. https://doi.org/10.1007/s00253-012-4247-1.
Lee, E.-H., Cho, K.-S., 2009. Effect of substrate interaction on the degradation of methyl tert-butyl ether, benzene, toluene, ethylbenzene, and xylene by Rhodococcus sp. J. Hazard Mater. 167, 669–674. https://doi.org/10.1016/j.jhazmat.2009.01.035.
Lee, K.K., Bing, R., Kiang, J., Bashir, S., Spath, N., Stelzle, D., Mortimer, K., Bularga, A., Doudesis, D., Joshi, S.S., Strachan, F., Gumy, S., Adair-Rohani, H., Attia, E.F., Chung, M.H., Miller, M.R., Newby, D.E., Mills, N.L., McAllister, D.A., Shah, A.S.V., 2020. Adverse health effects associated with household air pollution: a systematic review, meta-analysis, and burden estimation study. Lancet Global Health 8, e1427–e1434. https://doi.org/10.1016/S2214-109X(20)30343-0.
Lyngberg, O.K., Ng, C.P., Thiagarajan, V., Scriven, L.E., Flickinger, M.C., 2001. Engineering the microstructure and permeability of thin multilayer latex biocatalytic coatings containing E. coli. Biotechnol. Prog. 17, 1169–1179. https://doi.org/10.1021/bp0100979.
Mandin, C., Trantallidi, M., Cattaneo, A., Canha, N., Mihucz, V.G., Szigeti, T., Mabilia, R., Perreca, E., Spinazzè, A., Fossati, S., De Kluizenaar, Y., Cornelissen, E., Sakellaris, I., Saraga, D., Hänninen, O., De Oliveira Fernandes, E., Ventura, G., Wolkoff, P., Carrer, P., Bartzis, J., 2017. Assessment of indoor air quality in office buildings across Europe – the OFFICAIR study. Sci. Total Environ. 579, 169–178. https://doi.org/10.1016/j.scitotenv.2016.10.238.
Nakaoka, H., Todaka, E., Seto, H., Saito, I., Hanazato, M., Watanabe, M., Mori, C., 2014. Correlating the symptoms of sick-building syndrome to indoor VOCs concentration levels and odour. Indoor Built Environ. 23, 804–813. https://doi.org/10.1177/1420326X13500975.
Pei, J., Yin, Y., Liu, J., Dai, X., 2020. An eight-city study of volatile organic compounds in Chinese residences: compounds, concentrations, and characteristics. Sci. Total Environ. 698, 134137. https://doi.org/10.1016/j.scitotenv.2019.134137.
Quast, C., Pruesse, E., Yilmaz, P., Gerken, J., Schweer, T., Yarza, P., Peplies, J., Glöckner, F.O., 2012. The SILVA ribosomal RNA gene database project: improved data processing and web-based tools. Nucleic Acids Res. 41, D590–D596. https://doi.org/10.1093/nar/gks1219.
R Core Team, 2019. R: A Language and Environment for Statistical Computing. URL. Version 3.6.1. https://www.r-project.org/.
Robert, L., Guichard, R., Klingler, J., Cochet, V., Mandin, C., 2021. Indoor air quality in shopping and storage areas. Indoor Air 31, 1238–1251. https://doi.org/10.1111/ina.12783.
Sander, R., 2015. Compilation of Henry's law constants (version 4.0) for water as solvent. Atmos. Chem. Phys. 15, 4399–4981. https://doi.org/10.5194/acp-15-4399-2015.
Schulte, M.J., Wiltgen, J., Ritter, J., Mooney, C.B., Flickinger, M.C., 2016. A high gas fraction, reduced power, syngas bioprocessing method demonstrated with a Clostridium ljungdahlii OTA1 paper biocomposite. Biotechnol. Bioeng. 113, 1913–1923. https://doi.org/10.1002/bit.25966.
Singh, D., Kumar, A., Kumar, K., Singh, B., Mina, U., Singh, B.B., Jain, V.K., 2016. Statistical modeling of O3, NOx, CO, PM2.5, VOCs and noise levels in commercial complex and associated health risk assessment in an academic institution. Sci. Total Environ. 572, 586–594. https://doi.org/10.1016/j.scitotenv.2016.08.086.
Soreanu, G., Dumont, E., 2020. From Biofiltration to Promising Options in Gaseous Fluxes Biotreatment, from Biofiltration to Promising Options in Gaseous Fluxes Biotreatment. Elsevier. https://doi.org/10.1016/C2018-0-03712-3.
Standard Methods Committee of the American Public Health Association, American Water Works Association, Water Environment Federation, 2018. 2540 SOLIDS, in: Lipps, W., Baxter, T., Braun-Howland, E. (Eds.), Standard Methods For the Examination of Water and Wastewater. APHA Press, Washington DC. https://doi.org/10.2105/SMWW.2882.030.
Talwar, C., Nagar, S., Kumar, R., Scaria, J., Lal, R., Negi, R.K., 2020. Defining the environmental adaptations of genus Devosia: insights into its expansive short peptide transport system and positively selected genes. Sci. Rep. 10, 1151. https://doi.org/10.1038/s41598-020-58163-8.
Tay, S.T.L., Hemond, H.F., Polz, M.F., Cavanaugh, C.M., Dejesus, I., Krumholz, L.R., 1998. Two new Mycobacterium strains and their role in toluene degradation in a contaminated stream. Appl. Environ. Microbiol. 64, 1715–1720. https://doi.org/10.1128/AEM.64.5.1715-1720.1998.
Tsai, W.-T., 2019. An overview of health hazards of volatile organic compounds regulated as indoor air pollutants. Rev. Environ. Health 34, 81–89. https://doi.org/10.1515/reveh-2018-0046.
Van Tran, V., Park, D., Lee, Y.C., 2020. Indoor air pollution, related human diseases, and recent trends in the control and improvement of indoor air quality. Int. J. Environ. Res. Publ. Health 17. https://doi.org/10.3390/ijerph17082927.
Veselova, M.A., Plyuta, V.A., Khmel, I.A., 2019. Volatile compounds of bacterial origin: structure, biosynthesis, and biological activity. Microbiology 88, 261–274. https://doi.org/10.1134/S0026261719030160.
Wickliffe, J.K., Stock, T.H., Howard, J.L., Frahm, E., Simon-Friedt, B.R., Montgomery, K., Wilson, M.J., Lichtveld, M.Y., Harville, E., 2020. Increased long-term health

risks attributable to select volatile organic compounds in residential indoor air in southeast Louisiana. Sci. Rep. 10, 21649. https://doi.org/10.1038/s41598-020-78756-7.

World Health Organization, Regional Office for Europe, 2010. WHO Guidelines for Indoor Air Quality: Selected Pollutants. World Health Organization. Regional Office for Europe, Geneva.

World Health Organization, 2018. Burden of Disease of Household Air Pollution for 2016.

Yang, C., Yu, G., Zeng, G., Yang, H., Chen, F., Jin, C., 2011. Performance of biotrickling filters packed with structured or cubic polyurethane sponges for VOC removal. J. Environ. Sci. 23, 1325–1333. https://doi.org/10.1016/S1001-0742(10)60565-7.

Zhao, H., Geng, Y., Fan, J., Tao, K., Hou, T., 2013. Isolation and identification of Paracoccus sp. FD3 and evaluation of its formaldehyde degradation kinetics. Biotechnol. Bioproc. Eng. 18, 300–305. https://doi.org/10.1007/s12257-012-0449-8.

Zhao, K., Xiu, G., Xu, L., Zhang, D., Zhang, X., Deshusses, M.A., 2011. Biological treatment of mixtures of toluene and n-hexane vapours in a hollow fibre membrane bioreactor. Environ. Technol. 32, 617–623. https://doi.org/10.1080/09593330.2010.507634.

Zhong, L., Su, F.C., Batterman, S., 2017. Volatile organic compounds (VOCs) in conventional and high performance school buildings in the U.S. Int. J. Environ. Res. Publ. Health 14. https://doi.org/10.3390/ijerph14010100.