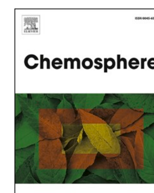
Chemosphere 287 (2022) 132182

Contents lists available at ScienceDirect

Chemosphere

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



Optimization of acrylic-styrene latex-based biofilms as a platform for biological indoor air treatment

Javier González-Martín [a,b], Sara Cantera [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, Stippeneng 4, , Wageningen, the Netherlands

HIGHLIGHTS

- A latex-based biofilm on polyurethane foam was developed for indoor air treatment.
- Latex-biomass mixtures reached removal efficiencies over 95% for toluene and pinene.
- Activated carbon-pretreated latex achieved the highest toluene and pinene removals.
- Water & nutrient content was a key factor in biocoating VOC abatement performance.
- Latex-based biofilms exhibited a superior robustness at high VOC loadings.

ARTICLE INFO

Handling Editor: Chang-Ping Yu

Keywords:
Biofiltration
Indoor air quality
Latex-based biofilm
VOCs

ABSTRACT

Biotechnologies have emerged as a promising solution for indoor air purification with the potential to overcome the inherent limitations of indoor air treatment. These limitations include the low concentrations and variability of pollutants and mass-transfer problems caused by pollutant hydrophobicity. A new latex-based biocoating was herein optimized for the abatement of the volatile organic compounds (VOCs) toluene, trichloroethylene, *n*-hexane, and α-pinene using acclimated activated sludge dominated by members of the phylum Patescibacteria. The influence of the water content, the presence of water absorbing compounds, the latex pretreatment, the biomass concentration, and the pollutant load was tested on VOC removal efficiency (RE) by varying the formulation of the mixtures. Overall, hexane and trichloroethylene removal was low (<30%), while high REs (>90%) were consistently recorded for toluene and pinene. The assays demonstrated the benefits of operating at high water content in the biocoating, either by including mineral medium or water absorbing compounds in the latex-biomass mixtures. The performance of the latex-based biocoating was likely limited by VOC mass-transfer rather than by biomass concentration in the biocoating. The latex-based biocoating supported a superior toluene and pinene removal than biomass in suspension when VOC loading rate was increased by a factor of 4.

1. Introduction

Indoor air quality has received an increasing attention worldwide in the past decade. The fact that indoor air is often more polluted than outdoor air and that people spend as much as 90% of their time indoors, makes improving indoor air quality a priority in terms of occupational health (European Comission, 2003). Indoor air pollution can cause severe health problems, such as sick building syndrome, chemical sensitivity, and severe cardiorespiratory or cancerous diseases (Burge, 2004; World Health Organization and WHO Regional Office for Europe, 2010). Indeed, the World Health Organization has estimated that up to 4.3 million deaths are attributable to indoor air pollution every year (World Health Organization, 2014). In addition, poor indoor air quality entails large economic losses derived from increased healthcare expenses, damage to buildings, and losses of worker productivity (World Bank; Institute for Health Metrics and Evaluation, 2014). Indoor air

* 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.chemosphere.2021.132182
Received 30 April 2021; Received in revised form 10 August 2021; Accepted 4 September 2021
Available online 7 September 2021
0045-6535/© 2021 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

contains hundreds of different pollutants, including particulate matter, volatile organic (VOC) and inorganic compounds (Leung, 2015; Mandin et al., 2017). The sources and health effects associated with some of these pollutants, such as formaldehyde or benzene, are widely known (World Health Organization and WHO Regional Office for Europe, 2010). However, new pollutants (pesticides, phthalates, etc) are continuously being identified, which limits the impact of indoor air pollution prevention strategies (Garcia-Jares et al., 2009). In addition, the current COVID-19 pandemic has raised the attention on biological air pollutants such as virus, bacteria, etc. Indoor air quality is expected to deteriorate in the coming years because new building construction regulations and 'green' certifications are focused on near-zero energy consumption. This will result in tighter buildings with reduced natural and forced ventilation and increased indoor pollutant concentrations (European Parliament, 2018).

In this context, air purification is needed in order to provide consistent and optimal indoor air quality. Traditionally, indoor air quality has been based on ventilation, temperature, humidity control, and filtration, the latter providing effective removal of particulate matter. In addition, physical-chemical technologies such as adsorption-based filters, ozonizers or photocatalytic devices are nowadays commercially available to treat indoor air (Luengas et al., 2015). However, these technologies do not provide a satisfactory pollutant abatement due to the complex nature of indoor air, the release of secondary pollutants, and high operating costs (González-Martín et al., 2021; Zhang et al., 2011). On the other hand, biotechnologies have recently emerged as a promising alternative for indoor air purification, especially for VOC removal. Biotechnologies can cope with the low concentrations and the variability in type and concentration of VOCs better than conventional physical-chemical technologies. Additionally, biotechnologies can increase the cost effectiveness, robustness, sustainability, and energy efficiency of indoor air treatment while also reducing operating costs. (Cheng et al., 2016; González-Martín et al., 2021). Unfortunately, biotechnologies present some drawbacks associated with the mass transfer of hydrophobic VOCs (Kraakman et al., 2011) and the potential release of microorganisms into the indoor atmosphere (Cheng et al., 2016), which requires further research in the field.

To overcome the above-mentioned limitations of biotechnologies, a latex-based biocoating is proposed. This novel bioreactor design consists of a porous polymer matrix where microorganisms are trapped, in which a non-growing steady state is achieved (Flickinger et al., 2017). The latex-based biocoating allows for direct contact between the gas pollutants and the attached cells, thus improving the bioavailability of hydrophobic pollutants, while preventing any release of microorganisms to the indoor air (Cortez et al., 2017; Estrada and Quijano, 2020). The operating and investment costs would decrease since biocoating is expected to remain active in a non-growing state for long periods of time and the treatment capacity per unit of volume is expected to increase (Estrada et al., 2012). For example, a latex-based biofilm containing photosynthetic bacteria was tested for hydrogen production. In this study the biofilm experienced an improved and reproducible hydrogen production even after long periods of dry or frozen storage (Piskorska et al., 2013). Likewise, Estrada et al. (2015) reported a toluene biodegradation rate in a *Pseudomonas putida* F1 biofilm embedded in latex which was 10-times higher than that of agarose biofilms (after drying and rehydration of the biofilm). The research in this area is scarce and limited to simple and small-scale experiments, in which the conditions are far from a real operating indoor air treatment device (Estrada et al., 2015). Therefore, this study aimed at engineering latex-based biofilms for the continuous treatment of indoor air in a configuration representative of real indoor air biofiltration units. Polyurethane foam (PUF) was selected as packing material of the biocoating, as it was successfully used in biofiltration in previous studies (Dobslaw and Engesser, 2018; Jia et al., 2020). *n*-hexane, α-pinene, trichloroethylene (TCE) and toluene were selected as model hydrophobic VOCs. These pollutants were selected to cover a wide range of VOCs (alkanes, terpenes, halogenated, aromatics, respectively) that can be typically found in indoor environments. Hydrophilic compounds were not considered as their absorption and degradation by microorganisms is often simple. This work was conducted in an experimental set-up that mimicked a biofilter and represented a step forward compared to previous works conducted in flat biofilms on paper. The influence of different preparation processes and operating parameters of the latex-based biofilm on VOC abatement was evaluated, including the presence of nutrients in the mixture, the concentration of biomass, the degree of drying of the polymer and biomass and the inlet VOC loading rate.

2. Materials and methods

2.1. Chemicals

n-hexane (CAS-110-54-3), α-pinene (CAS-80-56-8), toluene (CAS-108-88-3) and trichloroethylene (CAS-79-01-6) were used as model indoor air pollutants in a liquid mixture at 30%, 25%, 30% and 15%, respectively. The mineral salt medium (MSM) used was composed of 0.7 $g\ L^{-1}$ KH_2PO_4, 0.92 $g\ L^{-1}$ $K_2HPO_4 \cdot 3H_2O$, 3.0 $g\ L^{-1}$ KNO_3, 0.2 $g\ L^{-1}$ NaCl, 0.35 $g\ L^{-1}$ $MgSO_4 \cdot 7H_2O$ and 0.026 $g\ L^{-1}$ $CaCl_2 \cdot 2H_2O$. Micronutrients were provided by adding 2 $mL\ L^{-1}$ of a trace element solution as described elsewhere (Muñoz et al., 2008). MSM salts were purchased from Panreac® (Barcelona, Spain). *n*-hexane, TCE and toluene were also supplied by Panreac® (Barcelona, Spain), while α-pinene was supplied by Sigma-Aldrich (Madrid, Spain).

PRIMAL™ SF-208 ER (Dow Chemical, Germany), kindly supplied by Brenntag Química (Barcelona, Spain), was used in this study as latex for biocoatings formulations (alkylphenol ethoxylates and biocide free, acrylic-styrene copolymer; solids content 48.05%; pH 8.0–9.5). Hydrogel beads (acrylate-acrylamide copolymer) were obtained from Concorde Ibérica S.L. (Madrid, Spain).

2.2. Microorganisms

The microbial consortium used in the biodegradation tests consisted in an enriched consortium which primary source was fresh activated sludge from Valladolid wastewater treatment plant. The enrichment was acclimated to the selected VOC mixture in a 3 L stirred tank bioreactor for 4 months (Fig. S1). An aliquot of the bioreactor cultivation broth was centrifuged at 10,000 rpm for 10 min at 4 °C. The resulting biomass pellet was resuspended in fresh MSM, centrifuged as above described and supplied to the corresponding chambers as described below.

2.3. Experimental set-up

The experimental set-up consisted of four chambers made of transparent PVC with internal dimensions of $70 \times 10 \times 2\ cm^3$, resulting in a working volume of 1.4 L (Fig. 1). The chambers were packed with 1.4 L of polyurethane foam (PUF) (Filtren TM 25280, Recticel Ibérica S.L., Spain). The PUF exhibited a density of 0.01 $g\ mL^{-1}$, a specific surface area of 1000 $m^2\ m^{-3}$, a porosity of 96% and a water retention capacity of 0.12 $L_{water}\ L_{PUF}^{-1}$ (Lebrero et al., 2014). Air was compressed (ABAC B2500-50 2, Italy), filtered through a PTFE filter (0.22 μm pore size, MilliporeSigma SLFG05010) and humidified in a 1 m water bubble column to prevent the desiccation of the biocoating. A liquid VOC mixture was injected at 1 $\mu L\ h^{-1}$ using a syringe pump (Chemyx Fusion 100, USA) and a 100 μL liquid syringe (SGE 100R-GT-LC, Australia). This resulted in *n*-hexane, TCE, toluene and α-pinene gas concentrations of 9.6 ± 2.4, 13.7 ± 3.0, 14.7 ± 3.5 and 21.3 ± 3.9 $mg\ m^{-3}$, respectively. The injection rate of 1 $\mu L\ h^{-1}$ using a 100 μL syringe matched the operation range of the syringe pump and therefore ensured continuous dosage. No effect of pulsation or retaining droplets was observed neither in the previous studies nor in abiotic experiments. The polluted air stream was fed into the chambers at 50 $mL\ min^{-1}$, resulting in a EBRT of

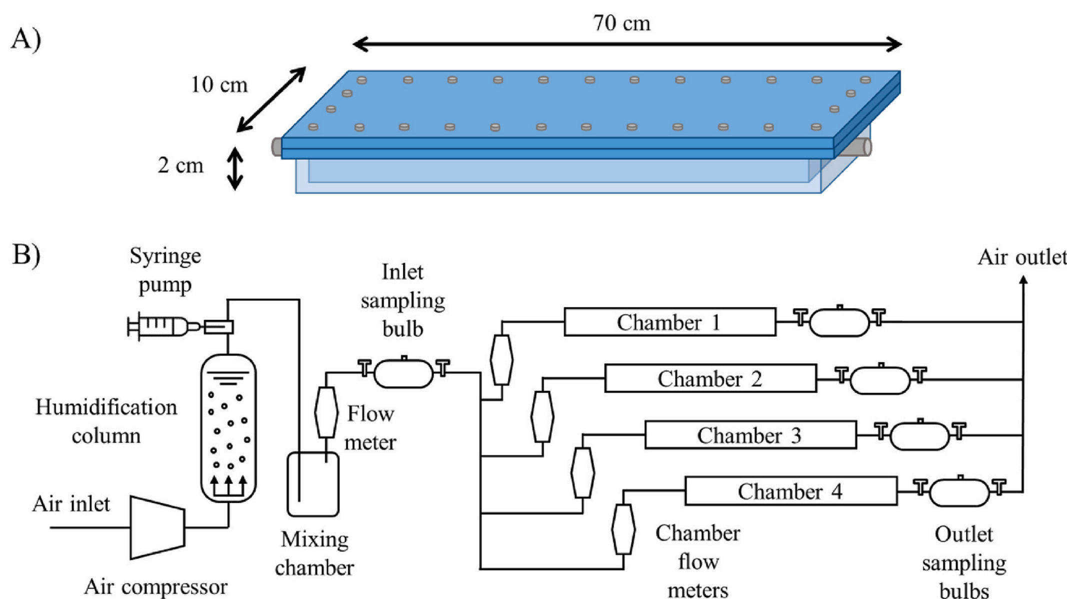


Fig. 1. Schematic representation of a flat PVC chamber used in the biodegradation assays A) and overview of the experimental set-up B).

28 min. VOC concentrations were measured by solid-phase micro-extraction (SPME) GC-FID in 250 mL glass bulbs (Sigma-Aldrich, Madrid, Spain) at the inlet and outlet of the chambers. The optimization assays were carried out at ambient temperature. The optimization assays were maintained until a clear and stable steady state in removal efficiency was observed.

2.4. Optimization of latex-based biocoatings operational conditions

Test series 1 - Influence of the presence of latex in the biocoating on VOC removal: Chamber 1 was inoculated with 40 mL of biomass suspension homogenously distributed in the PUF packing. Chamber 2 was inoculated with a mixture of a wet cell pellet and 60 mL of latex. An abiotic control with 60 mL of latex spread over the entire PUF packing was set in chamber 3. Finally, a mixture of 40 mL of biomass suspension and 60 mL of latex was inoculated in chamber 4. Chambers 1, 2 and 4 contained 0.5 g of biomass.

Test series 2 - Influence of the presence of hydrogel in the biocoating on VOC removal: Chamber 1 was inoculated with 50 mL of a fresh biomass suspension distributed homogeneously along the PUF packing material. A mixture of 60 mL of latex and a wet cell pellet was spread in the PUF packed bed of chamber 2. Chamber 3, packed with non-inoculated PUF, was used as abiotic control to assess VOC adsorption or photolysis in the experimental set-up. A mixture of a wet cell pellet, 60 mL of latex and 50 hydrogel beads (previously hydrated in MSM overnight and crushed) was used as inoculum in chamber 4. Chambers 1, 2 and 4 contained 0.5 g of biomass.

Test series 3 - Influence of the water content of the latex-biomass mixture on VOC removal: Chamber 1 was inoculated with 50 mL of biomass suspension distributed homogeneously along the PUF packing material. A wet cell pellet was mixed with 60 mL of latex and distributed in the PUF of chamber 2. A 50 mL suspension of biomass in MSM was mixed with 60 mL of latex, and then spread in the packed bed of chamber 3. Chamber 4 was inoculated with a mixture of a wet cell pellet and 60 mL of pre-dried latex (flushed with air for 24 h at 37 °C). All chambers contained 0.5 g of biomass.

Test series 4 - Influence of latex pretreatment and MSM content in the latex-biomass mixture on VOC removal: Chamber 1 was inoculated with a wet cell pellet (resuspended in the minimum amount of water to ensure a homogeneous distribution) and used as control. Chamber 2 was provided with a mixture of 40 mL of biomass suspension and 60 mL of pretreated latex (100 mL of latex and 10 g of previously washed active carbon (AC) were stirred for 24 h and subsequently filtered prior mixing with the biomass suspension). A mixture of 60 mL of latex and 20 mL of biomass suspension was inoculated in chamber 3, while 40 mL of biomass suspension and 60 mL of latex were used in chamber 4 mixture. All chambers contained 0.5 g of biomass.

Test series 5 - Influence of biomass concentration and VOC loading rate on VOC removal: Chamber 1 was inoculated with 40 mL of a biomass suspension (containing 0.5 g of biomass) homogenously distributed in the PUF packed bed. Chamber 2 was inoculated with a mixture of 40 mL of biomass suspension (0.5 g of biomass) and 60 mL of latex. Chamber 3 was used as an abiotic control with 60 mL of latex coating the PUF packing. Chamber 4 was coated with a mixture of 40 mL of biomass suspension (2 g of biomass) and 60 mL of latex. After 160 h of experiment, the air flowrate was increased from 50 mL min^{-1} to 100 mL min^{-1} in each chamber and the syringe inlet flowrate was increased from 1 μL h^{-1} to 2 μL h^{-1}.

2.5. Analytical procedures

The concentrations of VOCs were measured by SPME-GC-FID. Sampling was carried out by 10 min preconcentration of the pollutants in 250 mL glass bulbs by using 85 μm CAR/PDMS SPME fibers (Supelco, Bellefonte, USA). The samples were then injected in a GC-FID (Varian 3900) equipped with an Agilent HP-5MSI capillary column (30 m × 0.25mm × 0.25 μm). Injector and detector temperatures were set at 150 and 200 °C, respectively. The oven temperature was set at 40 °C for 1.5 min, increased at 10 °C min^{-1} to 50 °C (held for 1 min), and finally increased at 40 °C min^{-1} to 250 °C (held for 1 min). Nitrogen was used as carrier gas (2.5 mL min^{-1}), and as make-up gas (25 mL min^{-1}). Hydrogen and air flowrates were set at 30 and 300 mL min^{-1}. The fibers were initially conditioned at 300 °C for 1 h prior to calibration, and a cleaning run was performed before sampling with the above-described GC-FID method. External standards of the VOCs, prepared in 250 mL glass bulbs, were used for quantification using the preconcentration conditions above-mentioned (Fig. S2). Biomass concentration was quantified according to Standard Method 2540 D (American Public Health Association, 2017).

2.6. Bacterial community sequencing analysis

A sample from the enriched culture used as inoculum was withdrawn for microbial community analysis. The biomass was centrifuged at 13,000×g for 10 min. The resulting pellet was used for DNA extraction using the MasterPure™ Complete DNA Purification Kit (Epicenter Biotechnologies, USA) according to the manufacturer's instructions. DNA was purified and quantified with Qubit dsDNA broad-range (BR) assays in a QFX Fluorometer (DENOVIX, USA). The extracted DNA was sequenced using the Illumina MiSeq platform at the Foundation for the Promotion of Health and Biomedical Research of the Valencia Region (Valencia, Spain). Amplicon sequencing 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 (Klindworth et al., 2013). Illumina adapter overhang nucleotide sequences were added to the gene-specific sequences. After 16S rRNA gene amplification, library construction was carried out using the Nextera XT DNA Sample Preparation Kit (Illumina, San Diego, CA). Libraries were then normalized and pooled prior to sequencing. Samples containing indexed amplicons were loaded onto the MiSeq reagent cartridge for automated cluster generation sequencing using a 2 × 300 pb paired-end run (MiSeq Reagent kit v3 (MS-102-3001)) according to manufacturer's instructions (Illumina). The 16S rRNA gene sequences were processed and quality filtered using Mothur v1.44.3 following the Mother SOP (date of access 2021 Apr) (Kozich et al., 2013). Sequences were then clustered at 97% identity threshold into Operational Taxonomic Units (OTUs) using the SILVA 16S rRNA gene reference database (Version: 138.1). The nucleotide sequence dataset obtained in this study has been deposited at GenBank with accession code KESS00000000. The results obtained from the prokaryotic classification were further processed using R version 1.4.1. (R Core Team, 2019). The main genera of the prokaryotic population are shown in a heatmap plotted using the package *pheatmap* (Kolde, 2019).

2.7. Data treatment

VOC removal efficiency (%RE) was calculated as follows:

$$\%RE = 100 \times \left(1 - \left(C_i / C_{inlet}\right)\right)$$

where C_i and C_{inlet} are the outlet and inlet concentrations of the target pollutant measured in a chamber. The values of %RE and standard deviation are calculated as average of daily REs after achieving a steady state of pollutant abatement. A *t*-test comparing averages of unequal variances was performed in test series 5 (p = 0.05). REs before and after the change in the inlet VOC loading rate were compared.

3. Results

Test series 1 - Influence of the presence of latex in the biocoating on VOC removal (Fig. 2): Very low removals of hexane and TCE (<25%) were measured in chamber 1, which contained an aqueous suspension of biomass. On the contrary, chamber 1 achieved optimal degradations for toluene (90.6 ± 4.9%) and pinene (99.7 ± 0.7%) from the beginning of the experiment. Chamber 2, provided with a mixture of latex and a wet cell pellet, only achieved a moderate degradation for toluene (35.7 ± 11.9%) and pinene (33.8 ± 15.7%), while the removal of hexane and TCE was lower (29.8 ± 19.1 and 22.3 ± 12.3%, respectively). Chamber 3, which contained PUF coated with latex, effectively eliminated toluene (70.3 ± 8.2%) and pinene (89.0 ± 13.9%) throughout the experiment, while the removal of hexane and TCE was not significant. Chamber 4, which contained a mixture of latex and aqueous biomass suspension, achieved similar results than those of chamber 1. Hence, toluene and pinene experienced consistent removals of 90.6 ± 3.0% and 97.1 ± 6.6%, respectively, while hexane and TCE removals remained low (<17%).

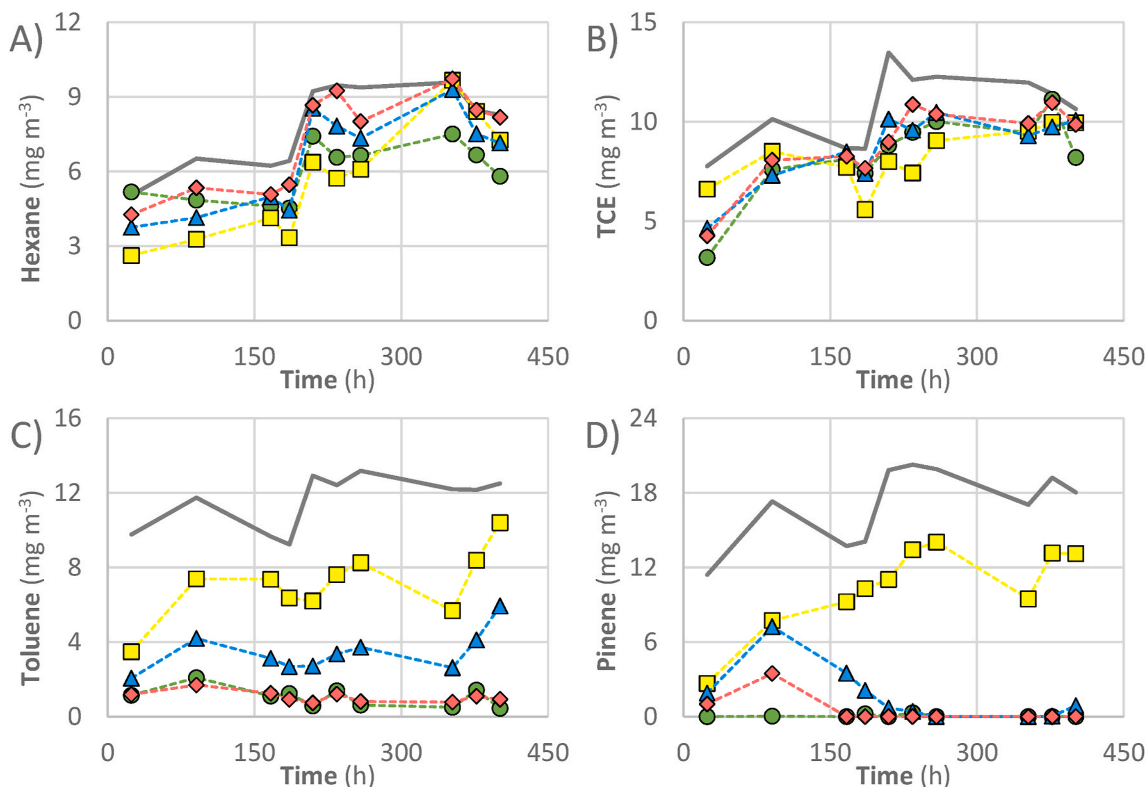


Fig. 2. Time course of the concentrations of A) hexane, B) TCE, C) toluene and D) pinene at the inlet (——), and outlet of chamber 1 (--●--, 40 mL biomass suspension), chamber 2 (--■--, mixture of a wet cell pellet and 60 mL latex), chamber 3 (--▲--, 60 mL latex) and chamber 4 (--◆--, mixture of 40 mL biomass suspension and 60 mL of latex).

Test series 2 - Influence of the presence of hydrogel in the biocoating on VOC removal (Fig. 3): TCE abatement was negligible in all chambers. Chamber 1, containing the acclimated aqueous biomass suspension, was able to degrade toluene and pinene with high efficiencies during the entire experiment (93.2 ± 4.1% and 99.6 ± 0.8%, respectively). Interestingly, hexane biodegradation increased from 6.2% by hour zero to 91.3% by hour 137 and remained at 98.4 ± 3.3% during the rest of the experiment. Chamber 2, which contained a mixture of latex and a wet cell pellet, had limited degradation of all VOCs. The abiotic experiment carried out in chamber 3 showed no substantial adsorption of hexane or TCE in the uncoated PUF, but a basal adsorption of toluene and pinene (26.7% and 34.3% of the inlet concentration, respectively). Finally, a moderate degradation of toluene of 46.3 ± 12.1% was obtained in chamber 4 (cell pellet + hydrogel + latex), along with a consistent pinene degradation of 98.6 ± 2.2% from hour 137 onwards, and negligible hexane and TCE removals.

Test series 3 - Influence of the water content of the latex-biomass mixture on VOC removal (Fig. 4): The removal of TCE was again very low under all tested conditions (<20%). Chamber 1, which contained an aqueous biomass suspension spread on the PUF, degraded toluene (89.3 ± 5.5%) and pinene (98.8 ± 2.2%) remarkably throughout the experiment. Hexane degradation, which was low at the beginning of the experiment, gradually increased from hour 208 onwards, to finally achieving steady removals of 64.2 ± 13.2%. The removal of hexane and toluene was low in chamber 2 (inoculated with a mixture of a wet cell pellet and latex). However, pinene degradation increased from 28.4 ± 7.5% to 62.2 ± 16.9% from hour 232 to 400 and decreased from hour 498 onward. Chamber 3, which was inoculated with an aqueous biomass suspension mixed with latex, supported hexane degradations <20%, moderate toluene removals (60.2 ± 13.9%) and high pinene removals (98.0 ± 4.0%) from hour 96 onwards. Finally, VOC removals in chamber 4, which contained a wet cell pellet mixed with pre-dried latex, remained lower than 25%.

Test series 4 - Influence of latex pretreatment and MSM content in the latex-biomass mixture on VOC removal (Fig. 5): No significant removal of TCE and hexane (<20% in all chambers) was recorded in this test series. The removal of toluene and pinene in chamber 1, inoculated with a wet cell pellet, was effective at the beginning of the experiment, but decreased to 21.7 ± 9.9% and 21.2 ± 8.2%, respectively, from hour 64 onwards. The aqueous biomass suspension and AC pre-treated latex in chamber 2 achieved optimal toluene (87.5 ± 4.0%) and pinene (91.2 ± 13.6%) degradations. Chamber 3, inoculated with 20 mL of biomass suspension mixed with latex, supported steady state average toluene and pinene degradations of 56.4 ± 3.9% and 94.4 ± 4.8%, respectively. Chamber 4, which contained a mixture of 40 mL of biomass suspension and latex, supported steady state toluene and pinene removals of 72.9 ± 5.1% and 96.1 ± 4.6%, respectively.

Test series 5 - Influence of biomass concentration and VOC loading rate on VOC removal (Fig. 6): Hexane and TCE removals were very low in all chambers (<20%). Chamber 1, which contained an aqueous biomass suspension, supported average degradations of 93.9 ± 3.5% for toluene and 95.9 ± 7.5% for pinene before the inlet VOC loading rate was increased by hour 160. This increase resulted in a deterioration of toluene and pinene removals to average values of 18.2 ± 8.1% and 18.6 ± 4.6%, respectively. Chamber 2, which was inoculated with a mixture of latex and an aqueous biomass suspension (containing 0.5 g of biomass), achieved average toluene removals of 86.4 ± 5.61%, and pinene removals of 93.6 ± 7.1%, despite the increase in VOC loading rate. A slight degradation of toluene and pinene (28.6 ± 17.2% and 27.5 ± 16.3%, respectively) was observed in the abiotic test performed in chamber 3. Chamber 4, where a mixture of latex and an aqueous biomass suspension (containing 2.0 g of biomass) was used, supported average removals of toluene and pinene of 94.4 ± 3.0% and 99.0 ± 2.0%, respectively, regardless of the VOC loading rate.

Bacterial community structure (Fig. 7): Regarding the microbial community, the inoculum was dominated by members of the phylum

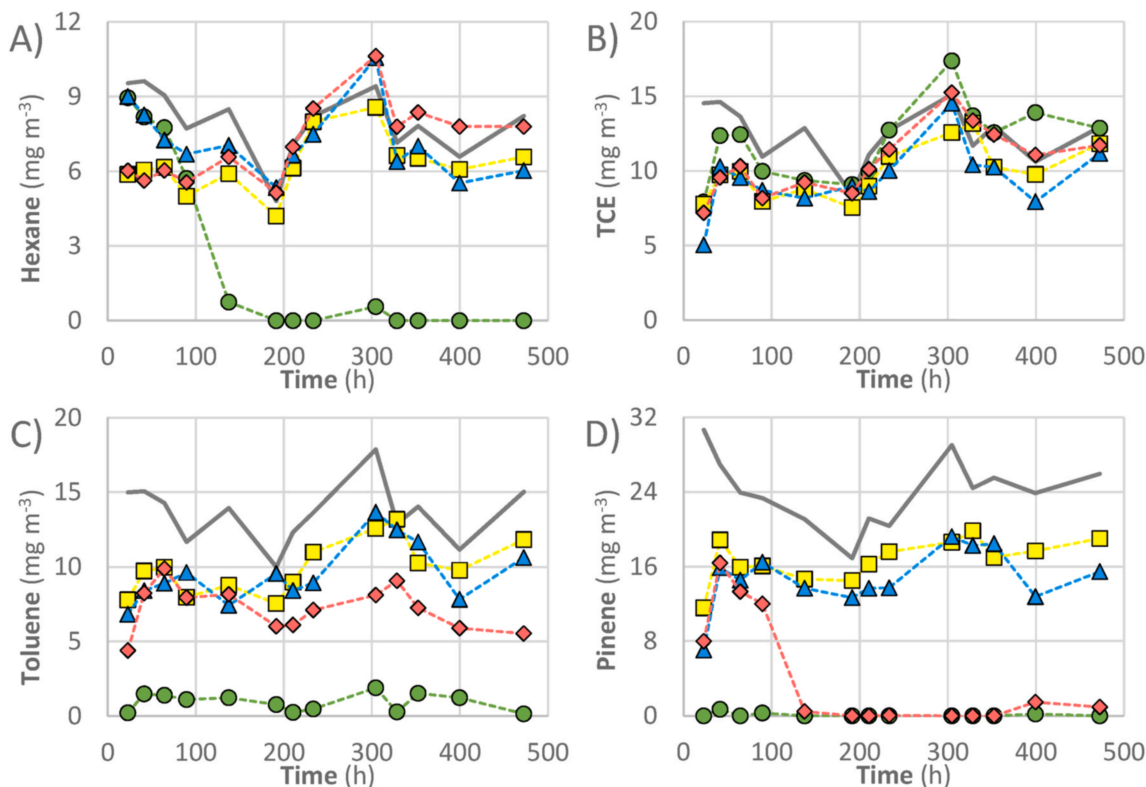


Fig. 3. Time course of the concentrations of A) hexane, B) TCE, C) toluene and D) pinene at the inlet (——), and outlet of chamber 1 (--●--, 50 mL biomass suspension), chamber 2 (--□--, mixture of wet cell pellet and 60 mL latex), chamber 3 (--▲--, uncoated PUF) and chamber 4 (--◆--, mixture of wet cell pellet, 60 mL of latex and 50 hydrogel beads).

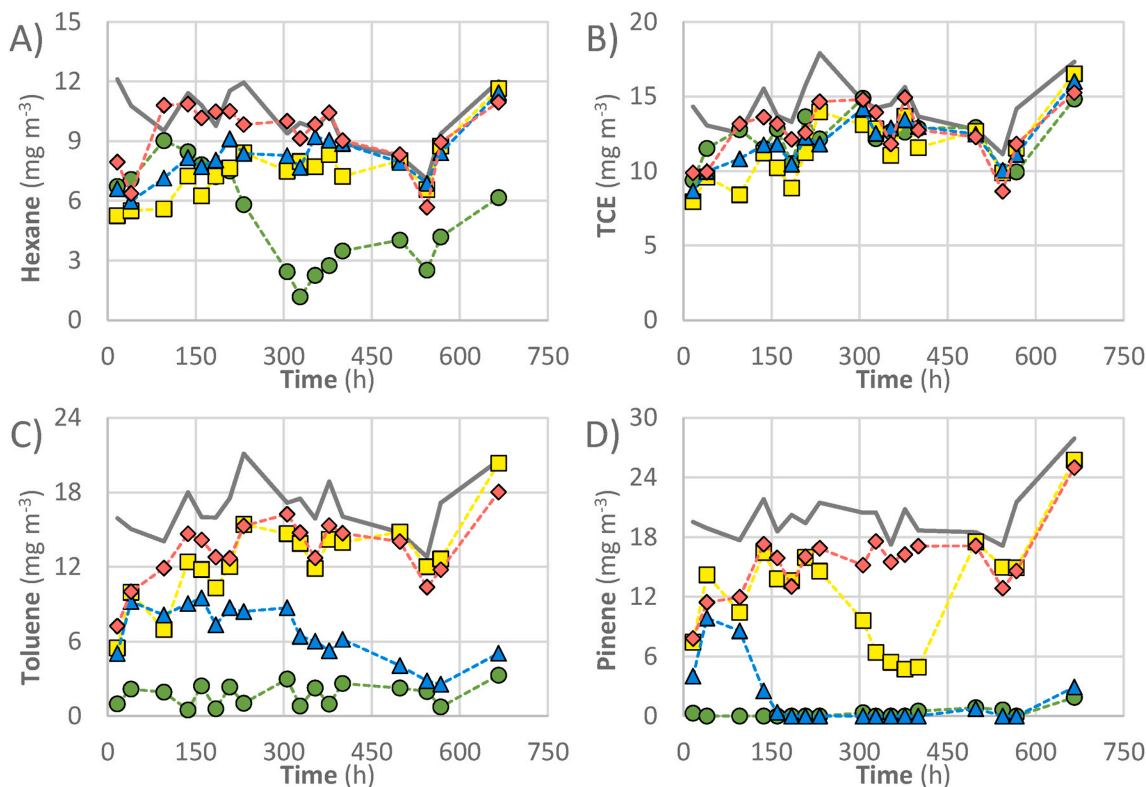


Fig. 4. Time course of the concentrations of A) hexane, B) TCE, C) toluene and D) pinene at the inlet (——), and outlet of chamber 1 (--●--, 50 mL biomass suspension), chamber 2 (--■--, mixture of wet cell pellet and 60 mL latex), chamber 3 (--▲--, mixture of 50 mL of biomass suspension and 60 mL of latex) and chamber 4 (--◆--, mixture of wet cell pellet and 60 mL of pre-dried latex).

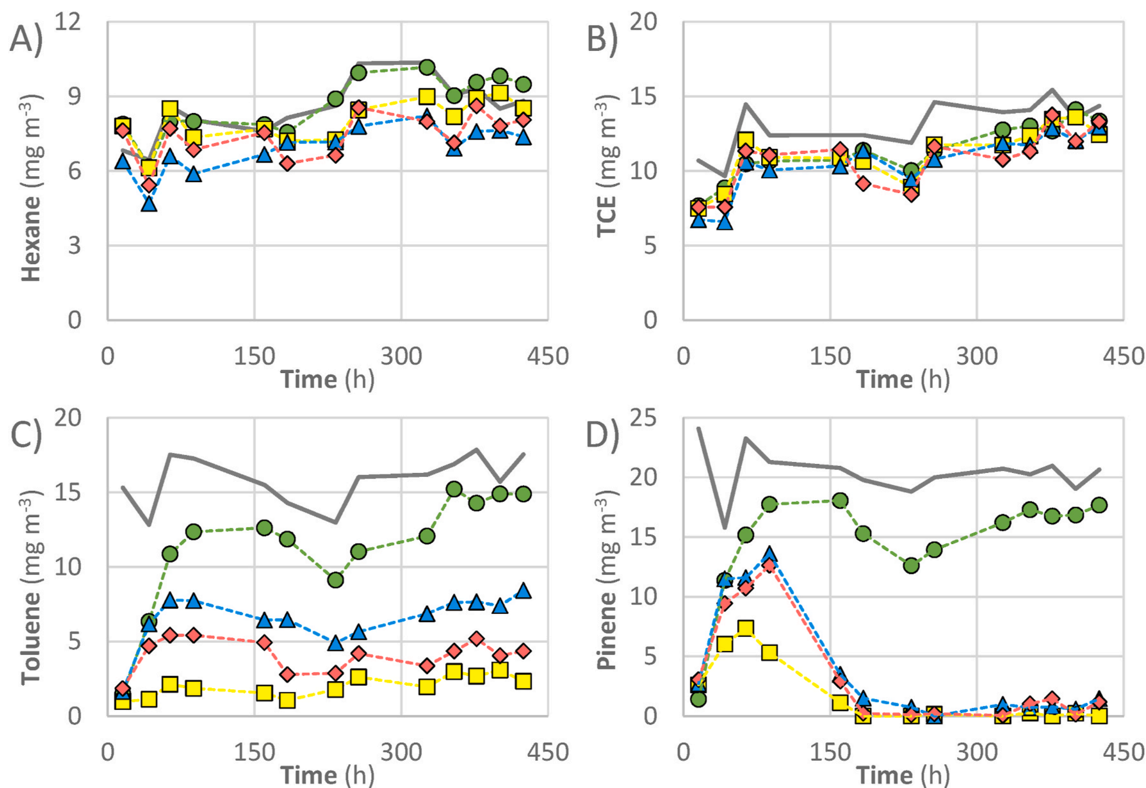


Fig. 5. Time course of the concentrations of A) hexane, B) TCE, C) toluene and D) pinene at the inlet (——), and outlet of chamber 1 (--●--, wet cell pellet), chamber 2 (--■--, mixture of 40 mL of biomass suspension and 60 mL of AC-treated latex), chamber 3 (--▲--, mixture of 20 mL of biomass suspension and 60 mL of latex) and chamber 4 (--◆--, mixture of 40 mL of biomass suspension and 60 mL of latex).

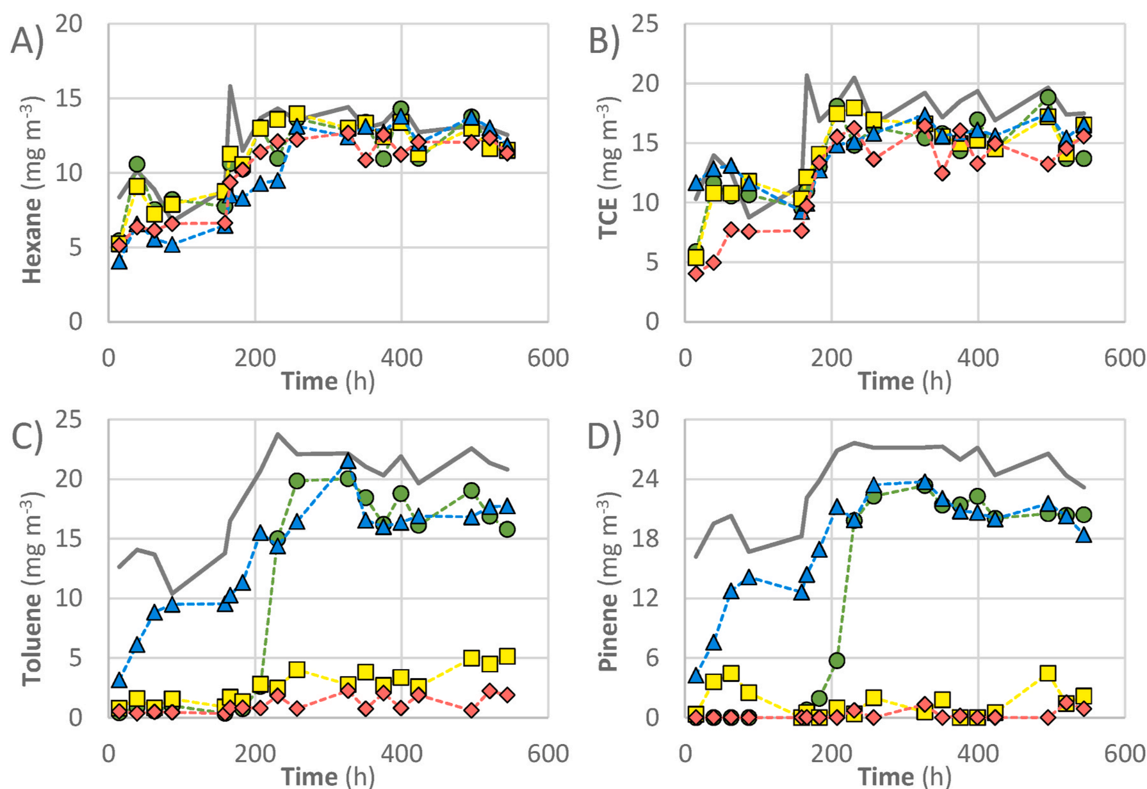
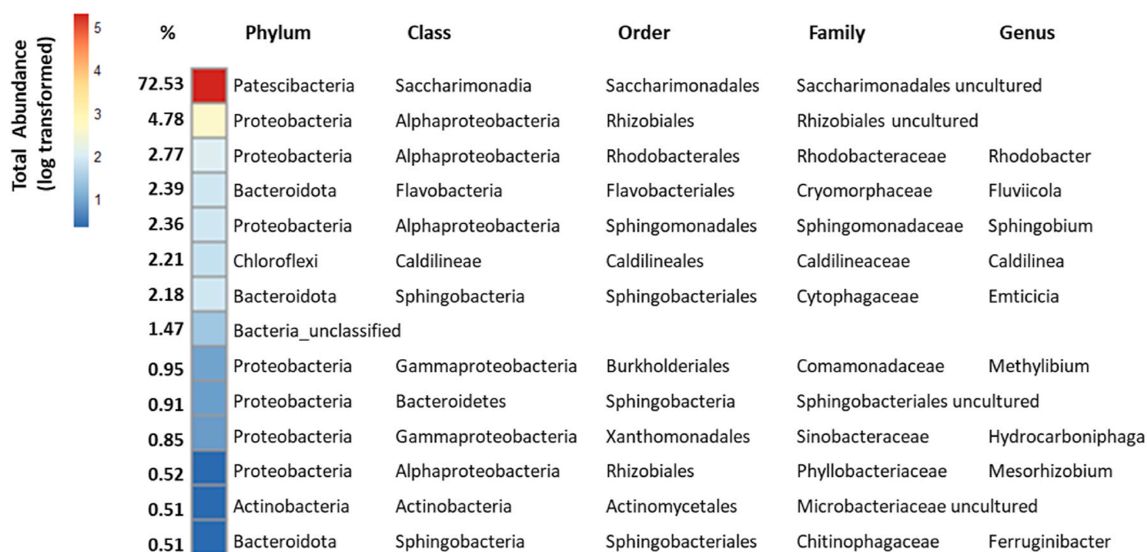


Fig. 6. Time course of the concentrations of A) hexane, B) TCE, C) toluene and D) pinene at the inlet (——), and outlet of chamber 1 (--●--, 40 mL biomass suspension, 0.5 g biomass), chamber 2 (--■--, mixture of 40 mL biomass suspension and 60 mL of latex, 0.5 g biomass), chamber 3 (--▲--, 60 mL latex) and chamber 4 (--◆--, mixture of 40 mL of biomass suspension and 60 mL of latex, 2 g of biomass).



%	Phylum	Class	Order	Family	Genus
72.53	Patescibacteria	Saccharimonadia	Saccharimonadales	Saccharimonadales uncultured	
4.78	Proteobacteria	Alphaproteobacteria	Rhizobiales	Rhizobiales uncultured	
2.77	Proteobacteria	Alphaproteobacteria	Rhodobacterales	Rhodobacteraceae	Rhodobacter
2.39	Bacteroidota	Flavobacteria	Flavobacteriales	Cryomorphaceae	Fluviicola
2.36	Proteobacteria	Alphaproteobacteria	Sphingomonadales	Sphingomonadaceae	Sphingobium
2.21	Chloroflexi	Caldilineae	Caldilineales	Caldilineaceae	Caldilinea
2.18	Bacteroidota	Sphingobacteria	Sphingobacteriales	Cytophagaceae	Emticicia
1.47	Bacteria_unclassified				
0.95	Proteobacteria	Gammaproteobacteria	Burkholderiales	Comamonadaceae	Methylibium
0.91	Proteobacteria	Bacteroidetes	Sphingobacteria	Sphingobacteriales uncultured	
0.85	Proteobacteria	Gammaproteobacteria	Xanthomonadales	Sinobacteraceae	Hydrocarboniphaga
0.52	Proteobacteria	Alphaproteobacteria	Rhizobiales	Phyllobacteriaceae	Mesorhizobium
0.51	Actinobacteria	Actinobacteria	Actinomycetales	Microbacteriaceae uncultured	
0.51	Bacteroidota	Sphingobacteria	Sphingobacteriales	Chitinophagaceae	Ferruginibacter

Fig. 7. Heatmap of the most representative bacterial genera (95% of the total genera) in the inoculum. Data is presented as the log transformation of the total abundance per sample.

Patescibacteria (72.5%), followed by members of the phyla Proteobacteria (13.1%), Bacteroidota (5.5%) and Chloroflexi (2.2%) (% absolute abundance). The most abundant organism (72.5%) consisted of uncultured bacteria from the order Saccharimonadales. Other relevant bacteria were uncultured Rhizobiales (4.8%), and members of the genera *Rhodobacter* (2.8%), *Fluviicola* (2.4%), *Sphingobium* (2.4%), *Caldilinea* (2.2%) and *Emticicia* (2.2%).

4. Discussion

VOC removal due to adsorption onto the PUF, and to a larger extend onto the latex-coated PUF was observed during the abiotic test (Fig. S3). However, in both assays, VOC removal due to adsorption gradually decreased with time. Absorption of the treated VOCs in the water phase or in latex was considered not significant when a steady state in RE was reached under long term operation. In addition, if pollutant absorption in the water phase was significant, this would have supported higher REs

for TCE, as its Henry partition coefficient ($H = 9.7 \times 10^{-4}$ mol m^{-3} Pa^{-1}) is higher than that of the other pollutants used in the study ($H_{TCE} > H_{pinene} > H_{toluene} \gg H_{hexane}$).

The limited abatement performance of TCE (REs<23.2%) confirmed that this is a very recalcitrant indoor air pollutant. Indeed, even if the Henry's law coefficient of TCE ($H = 9.7 \times 10^{-4}$ mol m^{-3} Pa^{-1}) is very similar to that of pinene ($H = 2.1 \cdot 10^{-4}$ mol m^{-3} Pa^{-1}) (Sander, 2015), the TCE removals remained below 20% under all tested conditions. The low TCE degradation performance could be attributed to physiological limitations of the bacterial community, which probably preferred to feed in other pollutants. There is still scarce information about the metabolic pathways involved in TCE degradation under aerobic conditions. However, it has been demonstrated that some microorganisms are able to co-metabolize TCE, by the action of non-specific oxygenases, using other compounds such as toluene or methane as primary substrates (Dolinová et al., 2017; Gafni et al., 2020).

Similarly, hexane removal was low-moderate in the majority of developed experiments (0–30%). Hexane is generally easily biodegradable (McGenity, 2019), however in this study low RE of hexane were observed. This low hexane removal could be explained by the high hydrophobicity of this indoor air pollutant, which decreases its bioavailability ($H = 6.0 \times 10^{-6}$ mol m^{-3} Pa^{-1}) (Sander, 2015). Unexpectedly, the adsorptive capacity of latex did not significantly increase hexane removal, which could be attributed to the high moisture content that was present in the experiments. Interestingly, hexane degradation was only clearly observed after an acclimation period in the chambers which were operated with an aqueous biomass suspension (98.4% and 64.3%), as shown in Figs. 3A and 4A. This effect may be attributed to the partial drying of the surface of the biofilm. Although partially dried biofilms can benefit from the direct uptake of highly hydrophobic chemical species by the direct contact cell-pollutant, this dryness can hinder the metabolic activity of the cells. TCE and hexane showed an unexpected basal removal, which could be attributed to pollutant solubilization in water and latex.

Recent research findings suggest the feasibility and superior performance of this direct pollutant gas-cell transport mechanism. For example, Lebrero et al. (2010) observed an increase in α-pinene RE after an irrigation system failure in a biofilter treating H_2S and other VOCs, and Popat and Deshusses (2010) observed optimum biodegradation performance in a biotrickling filter treating trichloroethane when the liquid recycling was temporarily stopped.

Overall, toluene and pinene were effectively degraded in all the experiments. Both pollutants are moderately hydrophobic ($H = 1.5 \times 10^{-4}$ and 2.1×10^{-4} mol m^{-3} Pa^{-1}, respectively) (Sander, 2015) and some microorganisms are able to biodegrade them. Indeed, in bioreactors for VOCs treatment inoculated with bacteria from the *Pseudomonas* (Estrada et al., 2015; López De León et al., 2019; López de León et al., 2020) and *Burkholderia* (Morya et al., 2020; Sun et al., 2013) genera were able to degrade toluene under aerobic conditions. In addition, some *Pseudomonas* (Chen et al., 2013a; Widhalm et al., 2016) and *Bursaphlenchus* (Chen et al., 2013b) strains seem to biodegrade pinene under aerobic conditions.

In this study, high eliminations of toluene and pinene were obtained in all the test series with aqueous biomass suspensions. This fact suggests the ability of the enriched microbial community to biodegrade these indoor air pollutants. The higher water activities likely resulted in a minimal microbial deactivation, maintaining a higher toluene and pinene biodegradation performance in the test series with aqueous biomass suspensions (56.4–94.4% for toluene; 93.6–99.7% for pinene). Opposite results were observed when a wet cell pellet with or without latex was used (18.2–35.7% for toluene; 18.6–33.8% for pinene). High REs have been previously reported for toluene and pinene in bioreactors inoculated with activated sludge. For example, Caicedo et al. (2018) reported toluene REs of 96% treating inlet concentrations of 0.82 ± 0.04 mg m^{-3} in a PUF-BTF. Cheng et al. (2011) reported REs of 25–76% treating pinene in a PUF-BTF, using inlet concentrations of 600–1500 mg m^{-3} and EBRTs of 58-35 s. Lebrero et al. (2011) investigated the simultaneous removal of toluene (inlet concentrations 4.3–6.3 mg m^{-3}) and pinene (inlet concentrations 0.40–0.60 mg m^{-3}) in a compost BF. Despite REs >99% were achieved for toluene at an EBRT of 32 s, only a RE of 7.3% was obtained for pinene. Toluene and pinene were also simultaneously treated in a compost BF and a PUF-BTF by Lebrero et al. (2014). REs >98% were achieved in both systems for toluene and pinene at EBRTs of 18 s. Interestingly, high hexane REs were also achieved under these conditions (96 ± 1.9 for BF; 91.8 ± 3.9 for BTF). Inlet concentrations ranged between 0.6 and 1.0 mg m^{-3} for the three VOCs. The REs of pinene and toluene achieved in this study with high water activity were as high as others found in the literature, but hexane and TCE abatement was generally very low. However, the results need to be carefully compared to others as the ranges of EBRT and inlet concentrations are different.

The influence of the water activity on the microbial biodegradation was confirmed by the higher average REs obtained in those mixtures with increased water content (test series 3; Fig. 1). Thus, the control in chamber 1, using a biomass suspension as inoculum, achieved high toluene and pinene REs (90.6% and 99.7%, respectively), while the mixture with 40 mL of biomass suspension and latex in chamber 4 achieved similar results (90.6% and 97.1% for toluene and pinene, respectively). On the contrary, the mixture with a wet cell pellet and latex in chamber 2 only reached REs of 35.7% for toluene and 33.8% for pinene. This phenomenon was also observed in test series 4, in which different amounts of MSM were added to the latex-biomass mixtures. Indeed, the wet cell pellet used as control in chamber 1 rapidly became dry (despite of the use of prehumidified air) and only supported average REs of 21.7% for toluene and 21.2% for pinene. On the other hand, the mixture of latex and a biomass suspension in 20 mL of MSM achieved REs of 56.4% for toluene and 94.4% for pinene, while when biomass was resuspended in 40 mL of MSM the REs increased up to 72.9% for toluene and 96.1% for pinene (Fig. 5). In this context, Estrada et al. (2015) reported specific elimination capacities of toluene in latex-based biocoatings (inoculated with *Pseudomonas putida*) 10-fold times higher than those of agarose-based biofilms. These toluene elimination capacities were slightly lower than those achieved in suspended cultures of *Pseudomonas putida* under the same operating conditions. In this work, toluene and pinene removals using aqueous biomass suspensions were very similar to those obtained using mixtures of biomass suspensions and latex. Finally, the addition of hydrogel beads as moisture preservative in the latex-cell pellet mixture supported a higher degradation of toluene (46.3%) and pinene (84.1%) in chamber 4 compared to chamber 2 (28.3% for toluene and 29.2% for pinene), where only latex and a wet cell pellet were used (Fig. 3C and 3D). These results suggested a positive effect of hydrogel beads on microbial activity based on their water retention capacity, although it did not enhance operation performance as much as the direct addition of MSM to the biomass-latex mixture.

The different evaluated latex pretreatments resulted in a wide range of indoor air pollutant abatement performance. Thus, in test series 3 (Fig. 4), very low REs were obtained when pre-dried latex mixed with the cell pellet was used (chamber 4) compared to the assays where regular latex was mixed with the cell pellet (chamber 2). The lower VOC removals were likely caused by the lower content of water in the biomass-latex mixture of chamber 4. However, when latex was pretreated with AC and mixed with 40 mL of biomass suspension, very high average REs for toluene (87.5%) and pinene (91.2%) were observed. This combination outperformed the indoor air pollutant abatement efficiencies of similar mixtures with regular latex (chamber 4). The increased performance was likely due to either the residual AC (which could have facilitated the adsorption of pollutants and increase their bioavailability) or to the removal of inhibitory solvents present in the latex during the pre-treatment stage (Fig. 4).

The influence of biomass concentration used in the latex-biomass-MSM mixture can be observed in test series 5. Chamber 2, inoculated with 0.5 g of biomass, achieved REs of 9.5, 15.7, 86.4 and 93.6% for

hexane, TCE, toluene and pinene, respectively, while chamber 4 (inoculated with 2.0 g of biomass) attained slightly higher removals for all indoor air pollutants (REs of 17.4, 23.2, 94.4 and 99.0% for hexane, TCE, toluene and pinene, respectively), as shown in Fig. 6. This 4-fold increase in biomass concentration did not entail a 4 times higher hexane degradation, which suggests that hexane removal was limited by mass-transfer, rather than by the metabolic activity of the cells at the tested biomass contents.

Finally, test series 5 showed that the presence of latex in the mixture provided an enhanced robustness towards increases in air flowrate and VOC concentration. Indeed, only minimal variations in the removals of toluene and pinene were observed when the VOC loading rate was increased by hour 160 in chambers 2 and 4, which included latex (Fig. 6). However, the degradation of toluene and pinene significantly decreased from 99.3 to 95.9% to steady state values of 18.2 and 18.6% in the chamber inoculated with biomass suspension likely due to a severe deterioration of metabolic activity caused by biomass desiccation. This finding confirmed the additional benefit entailed by the presence of latex, which minimized water evaporation from the active biofilm and preserved biological activity.

The concentrations of VOCs herein used were in the range of 20 mg m^{-3}, which indeed are much higher than the levels of VOC typically found in indoor air. However, these high concentrations were selected in order to provide sufficient carbon and energy source for the microorganisms embedded in the latex since this was a preliminary proof of concept study of the technology. In addition, the use of relatively high concentrations allowed a more accurate determination of the VOC concentrations in this preliminary study. Further studies using significantly lower concentrations (μg m^{-3}) must be conducted in latex-based biofilters to validate the technology in real indoor environments.

Based on the prokaryotic analysis we can assume that the members of the order Saccharimonadales (phylum Patescibacteria) had an important role in VOCs catabolism. However, little is known about their ecophysiology and metabolic capabilities (Lemos et al., 2019). Recent studies have pointed that Patescibacteria encompasses mostly unculturable bacterial taxa with relatively small genome sizes with potential for co-metabolism interdependencies (Lemos et al., 2019). Moreover, other studies have found that the members of the phylum Patescibacteria have streamlined many functions, regardless of their small genome, and they can rapidly acquire metabolic and physiologic advantages to adapt to different environments (Tian et al., 2020). In fact, they have been previously identified in benzene-degrading communities (Toth et al., 2021) and in petroleum-associated seabed sediments (Dong et al., 2019). Thus, they could be involved in the degradation of the persistent compounds treated in this study in co-metabolism with other abundant organisms.

Together with the order Saccharimonadales, other interesting bacterial orders were found. In fact, Rhizobiales, Xanthomonadales, Sphingomonadales and Burkholderiales have been previously reported as able to degrade alkanes, and mono and polycyclic aromatic hydrocarbons (McGenity, 2019). Rhizobiales and Burkholderiales have been frequently related with toluene degradation in aerobic conditions (Khammar et al., 2005; Lünsmann et al., 2016; Morya et al., 2020). The specific genus *Methylibium* from the order Burkholderiales has been described as a potential tool in the biodegradation of organosilicon and volatile compounds (Boada et al., 2020). Moreover, the genus Burkholderia has been identified as able to degrade mixtures of toluene, benzene, and mono-halogen benzenes to catechols (Dobslaw and Engesser, 2015). Similarly, the genera *Sphingobium* (Sphingomonadales) and *Hydrocarboniphaga* (Xanthomonadales) have been related to the degradation of alkane and aromatic hydrocarbons (Kertesz and Kawasaki, 2010; Palleroni et al., 2004). Thus, it seems that several bacterial members could have been involved in the degradation of the persistent compounds treated in this study in collaboration with the abundant organisms from the order Saccharimonadales.

5. Conclusions

The latex-based biofilms herein developed degraded toluene and pinene at low concentrations, while TCE and hexane removals were always low as a result of either mass transfer limitations from the gas phase to the cells or microbiological constraints. The residual elimination noticed could be attributed to absorption in water. This work represents the first latex-based biocoating study assessing the degradation of low concentrations of VOCs under continuous mode using an enriched consortium as inoculum. The beneficial influence of water activity in the biocoating was herein demonstrated. AC-treated latex showed the highest REs for toluene and pinene, while pre-drying of latex exerted a considerable negative impact on biofilm activity. The concentration of biomass slightly increased the degradation of toluene and pinene, although hexane and TCE remained low. These facts suggests that mass transfer or metabolic limitations hinder the degradation of hexane and TCE. The VOC loading rate increase revealed the superior robustness of the latex-based biofilm compared to aqueous biomass suspensions, which rapidly suffered desiccation. The experiments have successfully demonstrated the performance of latex-based coatings on VOC removal. However, additional investigation is needed to improve the removal efficiency of other pollutants and increase the air treatment capacity of the system.

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.

Acknowledgements

This work was supported by the Ministry of Science, Innovation and Universities, Spain [project RTI2018-0-096441-B-I00]. The Regional Government of Castilla y León and the EU-FEDER program [grant number CLU 2017-09] are also gratefully acknowledged.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2021.132182.

Credit author statement

Javier González-Martín: Conceptualization; Investigation; Writing – original draft; Writing – review & editing **Sara Cantera**: Methodology; Writing – review & editing; Supervision **Raquel Lebrero**: Conceptualization; Methodology; Writing – review & editing; Funding acquisition; Supervision **Raúl Muñoz**: Conceptualization; Methodology; Writing – review & editing; Funding acquisition; Supervision; Project administration

References

American Public Health Association, 2017. American Water Works Association, Water Environment Federation. Standard Method. Exam. Water Wastewater, 23rd ed.

Boada, E., Santos-Clotas, E., Bertran, S., Cabrera-Codony, A., Martín, M.J., Bañeras, L., Gich, F., 2020. Potential use of Methylibium sp. as a biodegradation tool in organosilicon and volatile compounds removal for biogas upgrading. Chemosphere 240, 124908. https://doi.org/10.1016/j.chemosphere.2019.124908.

Burge, P.S., 2004. Sick building syndrome. Occup. Environ. Med. 61, 185–190. https://doi.org/10.1136/oem.2003.008813.

Caicedo, F., Estrada, J.M., Silva, J.P., Muñoz, R., Lebrero, R., 2018. Effect of packing material configuration and liquid recirculation rate on the performance of a biotrickling filter treating VOCs. J. Chem. Technol. Biotechnol. 93, 2299–2306. https://doi.org/10.1002/jctb.5573.

Cheng, Z.W., Zhang, L.L., Chen, J.M., Yu, J.M., Gao, Z.L., Jiang, Y.F., 2011. Treatment of gaseous alpha-pinene by a combined system containing photo oxidation and aerobic

biotrickling filtration. J. Hazard Mater. 192, 1650–1658. https://doi.org/10.1016/j.jhazmat.2011.06.092.
Cheng, Z.W., Chen, J.M., Yu, J.M., Li, S.S., Jiang, Y.F., 2013a. Structural characterization and property analysis of surface-active substance accumulated during biodegradation of hydrophobic α-pinene by pseudomonas fluorescens. Water. Air. Soil Pollut. 224 https://doi.org/10.1007/s11270-013-1457-9.
Cheng, X.Y., Tian, X.L., Wang, Y.S., Lin, R.M., Mao, Z.C., Chen, N., Xie, B.Y., 2013b. Metagenomic analysis of the pinewood nematode microbiome reveals a symbiotic relationship critical for xenobiotics degradation. Sci. Rep. 3, 1–10. https://doi.org/10.1038/srep01869.
Cheng, Y., He, H., Yang, C., Zeng, G., Li, X., Chen, H., Yu, G., 2016. Challenges and solutions for biofiltration of hydrophobic volatile organic compounds. Biotechnol. Adv. 34, 1091–1102. https://doi.org/10.1016/j.biotechadv.2016.06.007.
European Comission, 2003. Indoor air pollution: new EU research reveals higher risks than previously thought. Eur. Community. IP/03/1278.
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.
Dobslaw, D., Engesser, K.-H., 2015. Degradation of toluene by ortho cleavage enzymes in B urkholderia fungorum FLU 100. Microb. Biotechnol. 8, 143–154. https://doi.org/10.1111/1751-7915.12147.
Dobslaw, D., Engesser, K.-H., 2018. Biodegradation of gaseous emissions of 2-chlorotoluene by strains of Rhodococcus sp. in polyurethane foam packed biotrickling filters. Sci. Total Environ. 639, 1491–1500. https://doi.org/10.1016/j.scitotenv.2018.05.278.
Dolinová, I., Štrjsová, M., Černík, M., Němeček, J., Macháčková, J., Ševců, A., 2017. Microbial degradation of chloroethenes: a review. Environ. Sci. Pollut. Res. 24, 13262–13283. https://doi.org/10.1007/s11356-017-8867-y.
Dong, X., Greening, C., Rattray, J.E., Chakraborty, A., Chuvochina, M., Mayumi, D., Dolfing, J., Li, C., Brooks, J.M., Bernard, B.B., Groves, R.A., Lewis, I.A., Hubert, C.R. J., 2019. Metabolic potential of uncultured bacteria and archaea associated with petroleum seepage in deep-sea sediments. Nat. Commun. 10, 1816. https://doi.org/10.1038/s41467-019-09747-0.
Estrada, J.M., Quijano, G., 2020. Bioremediation of air using microorganisms immobilized in bedding nanomaterials. Nanomater. Air Remediat. 211–225. https://doi.org/10.1016/B978-0-12-818821-7.00011-7.
Estrada, J.M., Kraakman, N.J.R., Bart), Lebrero, R., Muñoz, R., 2012. A sensitivity analysis of process design parameters, commodity prices and robustness on the economics of odour abatement technologies. Biotechnol. Adv. 30, 1354–1363. https://doi.org/10.1016/j.biotechadv.2012.02.010.
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.
European Parliament, 2018. DIRECTIVE (EU) 2018/844. Official Journal of the European Union.
Flickinger, M.C., Bernal, O.I., Schulte, M.J., Broglie, J.J., Duran, C.J., Wallace, A., Mooney, C.B., Velev, O.D., 2017. Biocoatings: challenges to expanding the functionality of waterborne latex coatings by incorporating concentrated living microorganisms. J. Coating Technol. Res. 14, 791–808. https://doi.org/10.1007/s11998-017-9933-6.
Gafni, A., Siebner, H., Bernstein, A., 2020. Potential for co-metabolic oxidation of TCE and evidence for its occurrence in a large-scale aquifer survey. Water Res. 171, 115431. https://doi.org/10.1016/j.watres.2019.115431.
Garcia-Jares, C., Regueiro, J., Barro, R., Dagnac, T., Llompart, M., 2009. Analysis of industrial contaminants in indoor air. Part 2. Emergent contaminants and pesticides. J. Chromatogr. A 1216, 567–597. https://doi.org/10.1016/j.chroma.2008.10.020.
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. https://doi.org/10.1016/j.chemosphere.2020.128376.
Jia, T., Sun, S., Chen, K., Zhang, L., Peng, Y., 2020. Simultaneous methanethiol and dimethyl sulfide removal in a single-stage biotrickling filter packed with polyurethane foam: performance, parameters and microbial community analysis. Chemosphere 244, 125460. https://doi.org/10.1016/j.chemosphere.2019.125460.
Kertesz, M.A., Kawasaki, A., 2010. Hydrocarbon-degrading sphingomonads: sphingomonas, Sphingobium, novosphingobium, and sphingopyxis. In: Timmis, K.N. (Ed.), Handbook of Hydrocarbon and Lipid Microbiology. Springer Berlin Heidelberg, Berlin, Heidelberg, pp. 1693–1705. https://doi.org/10.1007/978-3-540-77587-4_119.
Khammar, N., Malhautier, L., Degrange, V., Lensi, R., Godon, J.J., Fanlo, J.L., 2005. Link between spatial structure of microbial communities and degradation of a complex mixture of volatile organic compounds in peat biofilters. J. Appl. Microbiol. 98, 476–490. https://doi.org/10.1111/j.1365-2672.2004.02474.x.
Klindworth, A., Pruesse, E., Schweer, T., Peplies, J., Quast, C., Horn, M., Glöckner, F.O., 2013. Evaluation of general 16S ribosomal RNA gene PCR primers for classical and next-generation sequencing-based diversity studies. Nucleic Acids Res. 41 https://doi.org/10.1093/nar/gks808 e1–e1.
Kolde, R., 2019. Pheatmap: Pretty Heatmaps. R Package, Version 1.0.12.
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., 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.
Lebrero, R., Rodríguez, E., Martin, M., García-Encina, P.A., Muñoz, R., 2010. H2S and VOCs abatement robustness in biofilters and air diffusion bioreactors: a comparative study. Water Res. 44, 3905–3914. https://doi.org/10.1016/j.watres.2010.05.008.
Lebrero, R., Rodríguez, E., García-Encina, P.A., Muñoz, R., 2011. A comparative assessment of biofiltration and activated sludge diffusion for odour abatement. J. Hazard Mater. 190, 622–630. https://doi.org/10.1016/j.jhazmat.2011.03.090.
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.
Lemos, L.N., Medeiros, J.D., Dini-Andreote, F., Fernandes, G.R., Varani, A.M., Oliveira, G., Pylro, V.S., 2019. Genomic signatures and co-occurrence patterns of the ultra-small Saccharimonadia (phylum CPR/Patescibacteria) suggest a symbiotic lifestyle. Mol. Ecol. 28, 4259–4271. https://doi.org/10.1111/mec.15208.
Leung, D.Y.C., 2015. Outdoor-indoor air pollution in urban environment: challenges and opportunity. Front. Environ. Sci. 2, 1–7. https://doi.org/10.3389/fenvs.2014.00069.
López De León, L.R., Deaton, K.E., Deshusses, M.A., 2019. Miniaturized biotrickling filters and capillary microbioreactors for process intensification of VOC treatment with intended application to indoor air. Environ. Sci. Technol. 53, 1518–1526. https://doi.org/10.1021/acs.est.8b05209.
López de León, L.R., Deaton, K.E., Junkin, J., Deshusses, M.A., 2020. Capillary microbioreactors for VOC vapor treatment: impacts of operating conditions. Chemosphere 258, 1–9. https://doi.org/10.1016/j.chemosphere.2020.127286.
Luengas, A., Barona, A., Hort, C., Gallastegui, G., Platel, V., Elias, A., 2015. A review of indoor air treatment technologies. Rev. Environ. Sci. Biotechnol. 14, 499–522. https://doi.org/10.1007/s11157-015-9363-9.
Lünsmann, V., Kappelmeyer, U., Taubert, A., Nijenhuis, I., von Bergen, M., Heipieper, H. J., Müller, J.A., Jehmlich, N., 2016. Aerobic toluene degraders in the rhizosphere of a constructed wetland model show diurnal polyhydroxyalkanoate metabolism. Appl. Environ. Microbiol. 82, 4126–4132. https://doi.org/10.1128/AEM.00493-16.
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.
McGenity, T.J., 2019. Taxonomy, Genomics and Ecophysiology of Hydrocarbon-Degrading Microbes. Springer. https://doi.org/10.1007/978-3-319-60053-6.
Morya, R., Salvachúa, D., Thakur, I.S., 2020. Burkholderia: an untapped but promising bacterial genus for the conversion of aromatic compounds. Trends Biotechnol. 38, 963–975. https://doi.org/10.1016/j.tibtech.2020.02.008.
Muñoz, R., Díaz, L.F., Bordel, S., Villaverde, S., 2008. Response of Pseudomonas putida F1 cultures to fluctuating toluene loads and operational failures in suspended growth bioreactors. Biodegradation 19, 897–908. https://doi.org/10.1007/s10532-008-9191-5.
Palleroni, N.J., Port, A.M., Chang, H.-K., Zylstra, G.J., 2004. Hydrocarboniphaga effusa gen. nov., sp. nov., a novel member of the gamma-Proteobacteria active in alkane and aromatic hydrocarbon degradation. Int. J. Syst. Evol. Microbiol. 54, 1203–1207. https://doi.org/10.1099/ijs.0.03016-0.
Piskorska, M., Soule, T., Gosse, J.L., Milliken, C., Flickinger, M.C., Smith, G.W., Yeager, C.M., 2013. Preservation of H2 production activity in nanoporous latex coatings of Rhodopseudomonas palustrisCGA009 during dry storage at ambient temperatures. Microb. Biotechnol. 6, 515–525. https://doi.org/10.1111/1751-7915.12032.
Popat, S.C., Deshusses, M.A., 2010. Analysis of the rate-limiting step of an anaerobic biotrickling filter removing TCE vapors. Process Biochem. 45, 549–555. https://doi.org/10.1016/j.procbio.2009.11.017.
R Core Team, 2019. R: A Language and Environment for Statistical Computing. https://www.r-project.org/.
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.
Sun, D., Li, J., Xu, M., An, T., Sun, G., Guo, J., 2013. Toluene removal efficiency, process robustness, and bacterial diversity of a biotrickling filter inoculated with Burkholderia sp. Strain T3. Biotechnol. Bioproc. Eng. 18, 125–134. https://doi.org/10.1007/s12257-012-0253-5.
Tian, R., Ning, D., He, Z., Zhang, P., Spencer, S.J., Gao, S., Shi, W., Wu, L., Zhang, Y., Yang, Y., Adams, B.G., Rocha, A.M., Detienne, B.L., Lowe, K.A., Joyner, D.C., Klingeman, D.M., Arkin, A.P., Fields, M.W., Hazen, T.C., Stahl, D.A., Alm, E.J., Zhou, J., 2020. Small and mighty: adaptation of superphylum Patescibacteria to groundwater environment drives their genome simplicity. Microbiome 8, 1–15. https://doi.org/10.1186/s40168-020-00825-w.
Toth, C.R.A., Luo, F., Bawa, N., Webb, J., Guo, S., Dworatzek, S., Edwards, E.A., 2021. Anaerobic benzene biodegradation linked to the growth of highly specific bacterial clades. Environ. Sci. Technol. 55, 7970–7980. https://doi.org/10.1021/acs.est.1c00508.
Widhalm, B., Ters, T., Srebotnik, E., Rieder-Gradinger, C., 2016. Reduction of aldehydes and terpenes within pine wood by microbial activity. Holzforschung 70, 895–900. https://doi.org/10.1515/hf-2015-0243.
World Health Organization, WHO regional office for Europe, 2010. WHO Guidelines for Indoor Air Quality: Selected Pollutants. Geneva.

World Bank, Institute for Health Metrics and Evaluation, 2014. The Cost of Air Pollution. Strengthening the Economic Case for Action.

World Health Organization, 2014. Burden of disease from household air pollution for 2012. Summary of results. World Heal. Organ. 35, 2012–2014.

Zhang, Y., Mo, J., Li, Y., Sundell, J., Wargocki, P., Zhang, J., Little, J.C., Corsi, R., Deng, Q., Leung, M.H.K., Fang, L., Chen, W., Li, J., Sun, Y., 2011. Can commonly-used fan-driven air cleaning technologies improve indoor air quality? A literature review. Atmos. Environ. 45, 4329–4343. https://doi.org/10.1016/j.atmosenv.2011.05.041.