Journal of Environmental Management 349 (2024) 119362

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Research article

Indoor air VOCs biofiltration by bioactive coating packed bed bioreactors

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ARTICLE INFO

Handling Editor: Raf Dewil

Keywords:
Bioactive coating
Biofiltration
Indoor air
Latex
VOCs

ABSTRACT

Bioactive coatings are envisaged as a promising biotechnology to tackle the emerging problem of indoor air pollution. This solution could cope with the low concentrations, the wide range of compounds and the hydrophobicity of some indoor air VOCs, which are the most important bottlenecks regarding the implementation of conventional biotechnologies for indoor air treatment. A bioactive coating-based bioreactor was tested in this study for the abatement of different VOCs (n-hexane, toluene and α-pinene) at different empty bed residence times (EBRT) and inlet VOC concentrations. The performance of this reactor was compared with a conventional biofilm-based bioreactor operated with the same microbial inoculum. After an acclimation period, the bioactive coating-based bioreactor achieved abatements of over 50% for hexane, 80% for toluene and 70% for pinene at EBRTs of 112–56 s and inlet concentrations of 9–15 mg m^{-3}. These results were about 25, 10 and 20% lower than the highest removals recorded in the biofilm-based bioreactor. Both bioreactors experienced a decrease in VOC abatement by ~25% for hexane, 45% for toluene and 40% for pinene, after reducing the EBRT to 28 s. When inlet VOC concentrations were progressively reduced, VOC abatement efficiencies did not improve. This fact suggested that low EBRTs and low inlet VOCs concentration hindered indoor air pollutant abatement as a result of a limited mass transfer and bioavailability. Metagenomic analyses showed that process operation with toluene, hexane and pinene as the only carbon and energy sources favored an enriched bacterial community represented by the genera *Devosia*, *Mesorhizobium*, *Sphingobacterium* and *Mycobacterium*, regardless of the bioreactor configuration. Bioactive coatings were used in this work as packing material of a conventional bioreactor, achieving satisfactory VOC abatement similar to a conventional bioreactor.

1. Introduction

Nowadays, indoor air quality (IAQ) is one of the major globally recognized concerns as stated by the IAQ guidelines of the World Health Organization (World Health Organization. Regional Office for Europe, 2010), along with other national regulations and recommendations (Wei et al., 2015). The growing attention given to indoor air pollution is based on the increasing amount of time that people spend indoors (up to 90% of their life) together with the often higher level of pollutants that indoor air has in comparison to outdoor air (Kelly and Fussell, 2019). Indoor air pollution is responsible for mortal illnesses like acute lower respiratory and pulmonary diseases, ischemic heart disease or cancer (Ahmed et al., 2019; Lee et al., 2020), causing about 3.8 million annual deaths worldwide (World Health Organization, 2018). Other severe non-fatal effects in human health can also arise from indoor pollution, such as allergies and asthma, as well as the so-called sick building syndrome, which consists of a group of non-specific symptoms like eye, nose, throat and skin irritations, headaches, dizziness or nausea. Building regulations, focused on energy savings and, consequently, air tightness, contribute to raising indoor pollution levels (European Parliament, 2018). Although source-elimination and/or prevention measures are preferred to tackle indoor pollution, purification systems are often needed.

Indoor air pollution is strongly associated with the presence of volatile organic compounds (VOCs) in indoor environments. This group of pollutants includes a wide variety of compounds and is constantly growing, as new sources of pollution are continuously identified (Tsai, 2019). VOCs are found at very low concentrations indoors, usually in the

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https://doi.org/10.1016/j.jenvman.2023.119362
Received 30 June 2023; Received in revised form 29 September 2023; Accepted 14 October 2023
Available online 27 October 2023

range tens of μg m^{-3}, although pollutant emission peaks of up to several mg m^{-3} can be recorded when performing common activities such as cooking or cleaning (Pillarisetti et al., 2022). Traditional physical-chemical technologies are neither efficient nor cost-effective for indoor air treatment due to the specific characteristics of indoor pollution, thus biotechnologies have emerged as a potential solution to poor IAQ (González-Martín et al., 2021).

Biotechnologies have been successfully applied to industrial gas treatment (Dobslaw and Ortlinghaus, 2020). High removals have been achieved even for the treatment of very hydrophobic compounds such as n-alkanes. Wu et al. (2022) reported removals of 77 ± 1 and 35 ± 6% for C6 and C1, respectively, which improved to 99 ± 1 and 74 ± 3% after the addition of the surfactant SDBS. However, some adaptations are needed to implement biotechnologies for indoor air purification. The trace level concentrations, the complex mixtures of compounds and unknown interactions between them, and the sometimes-high hydrophobicity of indoor VOCs often hinder an optimal performance of biotechnologies when treating indoor air. In this context, new configurations in which the mass transfer of hydrophobic, diluted VOCs is enhanced, such as bioactive coatings, are being investigated (Kraakman et al., 2021).

A bioactive coating is formed by a thin layer of porous polymer matrix in which the microorganisms are attached or embedded (Lyngberg et al., 2001). In such configuration, the microorganisms are ideally isolated from each other, and the natural formation of an aqueous-rich biofilm is then limited. VOC biodegradation in bioreactors involves a first mass-transfer step through a wet biofilm, which represents the main limitation for the degradation of hydrophobic VOCs in different bioreactor configurations (Wu et al., 2018b; Wu et al., 2023a). If the formation of a biofilm is avoided, the microorganisms would be in direct contact with the pollutants, thus mass transfer would be greatly enhanced. Additionally, bioactive coatings support a high density of microorganisms, as well as a high surface-volume ratio (Cortez et al., 2017; Lamprea Pineda et al., 2021).

The concept of bioactive coatings has already been tested, but only in small scale experiments, which are far from the operational conditions of an indoor air purification device. A first bioactive coating preparation procedure was presented by Gosse et al. (2007) for the immobilization of the H_2-producing photosynthetic bacterium *Rhodopseudomonas palustris*; a method further described in Gosse and Flickinger (2011). A similar procedure was used by Bernal et al. (2014) in the photosynthetic production of O_2 from CO_2 using bioactive coatings containing several cyanobacteria strains; and also by Fidaleo et al. (2014) for the bioconversion of D-sorbitol to L-sorbose by *Gluconobacter oxydans*. Bioactive coatings have also been tested for VOC abatement at low concentrations. Estrada et al. (2015) reported a *Pseudomonas putida* bioactive coating providing toluene degradation rates 10 times higher than traditional agarose degradation assays. These previous studies support the potential for bioactive coatings to be implemented in traditional bioreactors and further applied to indoor air purification, a concept that remains largely unexplored to date. Indeed, a limited number of works have been carried out exploring the potential of acrylic-styrene latex-based biofilms for the simultaneous removal of different VOCs (Cantera et al., 2022; González-Martín et al., 2022).

In this paper, the application of bioactive coatings was proposed for the first time in laboratory-scale conventional bioreactor configurations and tested in a continuous, long-term experiment in order to assess the potential of this new biotechnology for the abatement of indoor VOCs. To this aim, a bioactive coating, supported on polyurethane foam, was used as the packing material of a conventional biofilter. A simulated indoor air stream, contaminated with VOCs, was used to assess the influence of different empty bed residence times (EBRTs) and VOC inlet concentrations on pollutant removal. n-hexane, toluene and α-pinene were selected as model pollutants, since these VOCs have been consistently reported in indoor air and cover a wide range of chemical groups (alkanes, aromatics and terpenes, respectively) and hydrophobicities (Henry's law constants of $6.0{\cdot}10^{-6}$, $1.5{\cdot}10^{-3}$ and $2.1{\cdot}10^{-4}$ mol m^{-3} Pa^{-1}, respectively) (Sander, 2015). Hydrophilic VOCs were not considered as model pollutants in this work since their removal is generally not limited by mass transfer (Wu et al., 2023a).

2. Materials and methods

2.1. Chemicals

A mixture of model VOCs, namely α-pinene (CAS 80-56-8), toluene (CAS 108-88-3) and n-hexane (CAS 110-54-3), was utilized as feed for the bioreactors, containing 22% of α-pinene, 34% of toluene, and 44% of n-hexane (v/v). The macro- and micronutrient composition of the mineral salt medium (MSM) can be found elsewhere (González-Martín et al., 2022). D (+)-saccharose and glycerol were included in bioactive coating formulations as osmoprotectancts, as previously described in Gosse and Flickinger (2011). PRIMAL™ SF-208 ER (acrylic-styrene copolymer; biocide/alkylphenol ethoxylate free; solids content 48.05%; pH 8.0–9.5; Dow Chemical, Germany), kindly supplied by Brenntag Química (Barcelona, Spain), was used as polymer in the preparation of the bioactive coatings. α-pinene was purchased from Sigma-Aldrich (Madrid, Spain). MSM salts, n-hexane, toluene and glycerol were purchased from Panreac® (Barcelona, Spain); and D (+)-saccharose was purchased from Labkem (Barcelona Spain).

2.2. Microorganisms and coating

Fresh activated sludge was obtained from a local wastewater treatment plant (Valladolid WWTP, Spain) and further used as inoculum for both bioreactors. Aliquots of activated sludge were centrifuged (10,000 rpm, 10 min, 4 °C) to obtain biomass pellets of ≈2.3 g (dry weight) for each bioreactor. Bioreactor A, which was set as a conventional biofilm bioreactor, was used as a control, while a bioactive coating was used in bioreactor B. The bioactive coating was prepared with the proportions of polymer and osmoprotectancts described in Gosse and Flickinger (2011). The formulation included 200 mL of polymer, 70 mL of a saccharose solution (0.58 g L^{-1}) and 30 mL of glycerol (100% v/v). The polymer was replaced by distilled water in the formulation of bioreactor A, in order to facilitate the distribution of the cells throughout the packing. The packing material was homogeneously coated with each formulation and then dried overnight prior to the start of the experiment. No pretreatment was done to the packing material prior to coating.

2.3. Experimental set-up

Two transparent PVC column reactors (12.2 cm internal diameter; 40 cm height; 4.7 L working volume), namely BF A and BF B, were used for the experiments (Fig. 1). Polyurethane foam (PUF) (Filtren TM 25280, Recticel Ibérica S.L., Spain), with 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}, was used as packing material. Four PUF pieces of 8 cm height, separated by 2 cm empty spaces, were used in each reactor.

The air stream was provided by an air compressor (ABAC B2500-50 2, Italy), then humidified in two 1 m water columns connected in series, and regulated by calibrated air rotameters (Aalborg, New York, USA) before entering the reactors. The VOC mixture was fed to the air stream by using a syringe pump (Fusion 100, Chemyx Inc., USA) and glass syringes (Hamilton, USA). Different air flows, syringe pump flows and syringe volumes were selected to achieve the desired experimental conditions, as shown in Table 1. For the irrigation system, 1 L MSM reservoir bottles, a multichannel peristaltic pump (model 205S; Watson-Marlow Limited, Falmouth, UK) and a timer were used. Intermittent irrigation was set in stages 1–3, with a frequency of 100 mL day^{-1} distributed in 8 injections of 5 min duration. From day 17 of stage 3

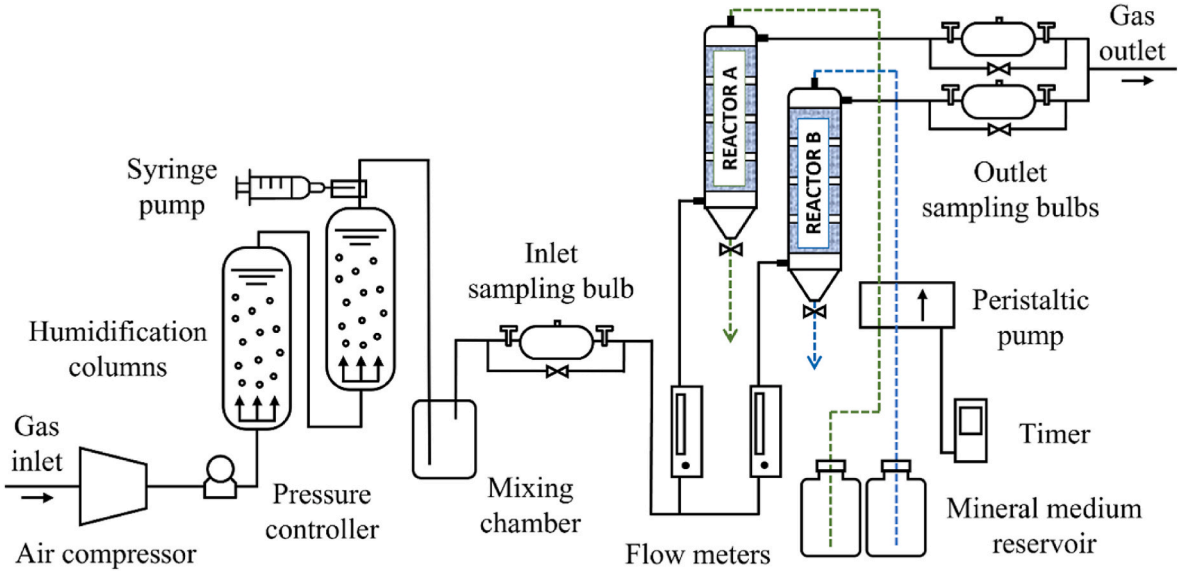

Fig. 1. Schematic overview of the experimental set-up.

Table 1
Operational parameters and average inlet VOC concentrations (standard deviation) in each stage.

Stage	Elapsed time (days)	Q_{gas} (L min^{-1})	EBRT (s)	Irrigated volume (frequency)	C_{Hex} (mg m^{-3})	C_{Tol} (mg m^{-3})	C_{Pin} (mg m^{-3})
1	24	2.5	112	100 mL MSM day^{-1}	9.7 (0.8)	15.0 (1.4)	11.1 (1.3)
2	47	5.0	56	(8 × 12.5 mL)	9.3 (1.7)	12.6 (1.5)	12.4 (3.4)
3	31	10	28		10.6 (1.3)	18.7 (1.5)	15.5 (1.4)
4	25	10	28	200 mL MSM day^{-1}	7.0 (0.8)	8.5 (0.9)	6.7 (0.7)
5	31	10	28	(8 × 25.0 mL)	2.9 (0.7)	4.1 (0.6)	3.7 (0.5)
6	33	10	28		1.7 (0.3)	2.3 (0.4)	1.7 (0.3)
7	21	10	28		1.1 (0.1)	1.4 (0.2)	1.2 (0.2)
8	7	2.5	112		1.4 (0.2)	1.8 (0.3)	1.4 (0.2)

onwards, the pumping period was doubled (10 min) to achieve a total irrigation volume of 200 mL day^{-1}. Fresh MSM was utilized throughout the experiment to avoid nutrient limitation and metabolite accumulation.

The empty bed residence time was reduced from 112 s in stage 1–56 s and 28 s in stages 2 and 3, respectively, while inlet concentrations of hexane, toluene and pinene were maintained at average values of 10.2 ± 2.0, 15.4 ± 3.0 and 13.1 ± 3.0 mg m^{-3}. From stage 4–7, the EBRT remained constant at 28 s and inlet hexane, toluene and pinene concentrations were reduced from 10.6 ± 1.3, 18.7 ± 1.5 and 15.5 ± 1.4 mg m^{-3} in stage 3, to 7.0 ± 0.8, 8.5 ± 0.9 and 6.7 ± 0.7 mg m^{-3} in stage 4, 2.9 ± 0.7, 4.1 ± 0.6 and 3.7 ± 0.5 mg m^{-3} in stage 5, 1.7 ± 0.3, 2.3 ± 0.4 and 1.7 ± 0.3 mg m^{-3} in stage 6, and 1.1 ± 0.1, 1.8 ± 0.3 and 1.4 ± 0.2 mg m^{-3} in stage 7. Finally, in stage 8, the EBRT was increased again to 112 s maintaining inlet hexane, toluene and pinene concentrations at 1.4 ± 0.2, 1.8 ± 0.3 and 1.4 ± 0.2 mg m^{-3}, respectively (Table 1). The temperature was controlled at 22 ± 2 °C throughout the 219 days of experiment.

Sampling of VOCs was performed by solid-phase microextraction (SPME) in 250 mL glass bulbs (Sigma-Aldrich, Madrid, Spain) located at the inlet and outlet of the reactors. VOCs were then analyzed and quantified by GC-FID.

2.4. Analytical procedures

The concentrations of the three VOCs were measured daily at the inlet and the outlet of BF A and BF B by SPME-GC-FID. SPME preconcentration consisted of a 10 min exposure of the adsorbent fiber (85 μm CAR/PDMS; Supelco, Bellefonte, USA) to the contaminated air in closed 250 mL glass bulbs. Then, analysis and quantification were performed by a GC (Varian 3900), equipped with an Agilent HP-5MSI capillary column (30m × 0.25mm × 0.25 μm) and FID detector. GC-FID method parameters are described in González-Martín et al. (2022). External standards of VOCs were used for SPME calibration in 250 mL glass bulbs. A conditioning step (300 °C, 1 h exposure) was performed before using a new SPME fiber. A blank run was performed daily prior to VOC measurements to ensure a clean SPME fiber.

The dry-weight biomass concentration in the inoculum was calculated as described in Standard Method 2540 D (American Public Health Association et al., 2017).

2.5. Bacterial community sequencing analysis

Samples of 20 mL of the inoculum, BF A and BF B were withdrawn in duplicate for metagenomic amplicon sequencing. To obtain representative samples of BF A and BF B the cells were detached at the end of the experiment (day 219) from the polyurethane foam by intense rinsing with deionized water. The DNA was extracted from each biological replicate with a FastDNA™ SPIN Kit for Soil (MP Biomedicals, USA). Library preparation 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) according to Pascual et al. (2021). Sequence processing and cleaning was developed with Mothur v.1.47.0 (Kozich et al., 2013). Clustering of the sequences into Operational Taxonomic Units (OTUs) was based on the gene reference database SILVA (Version: 138.1) and the ribosomal database project (version 18) (Cole et al., 2014; Quast et al., 2012). The sequences obtained have been deposited in Genbank as Bioproject PRJNA929218. Bar graphs showing the main genera (>5% relative abundance) were plotted with R using the package *ggplot2* (Wickham, 2009). The relative abundance of all the genera found was plotted in a heatmap using R *pheatmap* (Kolde, 2019).

2.6. Data treatment

Removal efficiency (%RE) was calculated using daily VOC concentration measurements as described in Eq. (1):

$$\%RE = 100 \times \left(1 - \left(\frac{Cout_i}{Cin_i}\right)\right) \quad (1)$$

where Cin_i and $Cout_i$ are the inlet and outlet concentrations (in mg m^{-3}), respectively, of each VOC. RE values are presented as the average and standard deviation after a steady removal has been achieved in each stage (Table S1).

3. Results and discussion

An initial acclimation period was performed to achieve optimal REs before the start of the experiment (Fig. S1). In this 67-day acclimation period, EBRT and irrigation rate were maintained at 112 s and 100 mL day^{-1}, respectively. Inlet concentrations were progressively increased to enhance the mass transfer of VOCs to microorganisms (by increasing the concentration gradient) and thus achieving a good adaptation to the bioreactors' conditions. At the end of this period, the experimental conditions of stage 1 were set, and the experiment was initiated when a steady VOC removal was observed. Both adsorption and absorption phenomena were considered negligible, as demonstrated by the abiotic test performed prior inoculation of the biofilters (not shown) and by the low VOC removals recorded at the beginning of the acclimation period (Fig. S1). In this initial phase of acclimation, hexane removals were similar in both bioreactors until day 55, when BF A outperformed BF B (74.5 ± 5.7% vs. 41.1 ± 5.3%). Toluene and pinene REs followed a similar trend. From day 20 onwards, the removal of toluene in BF A was higher than that recorded in BF B, to reach final steady removals of 93.7 ± 2.5% and 76.6 ± 3.0%, respectively. Pinene removal was similar in

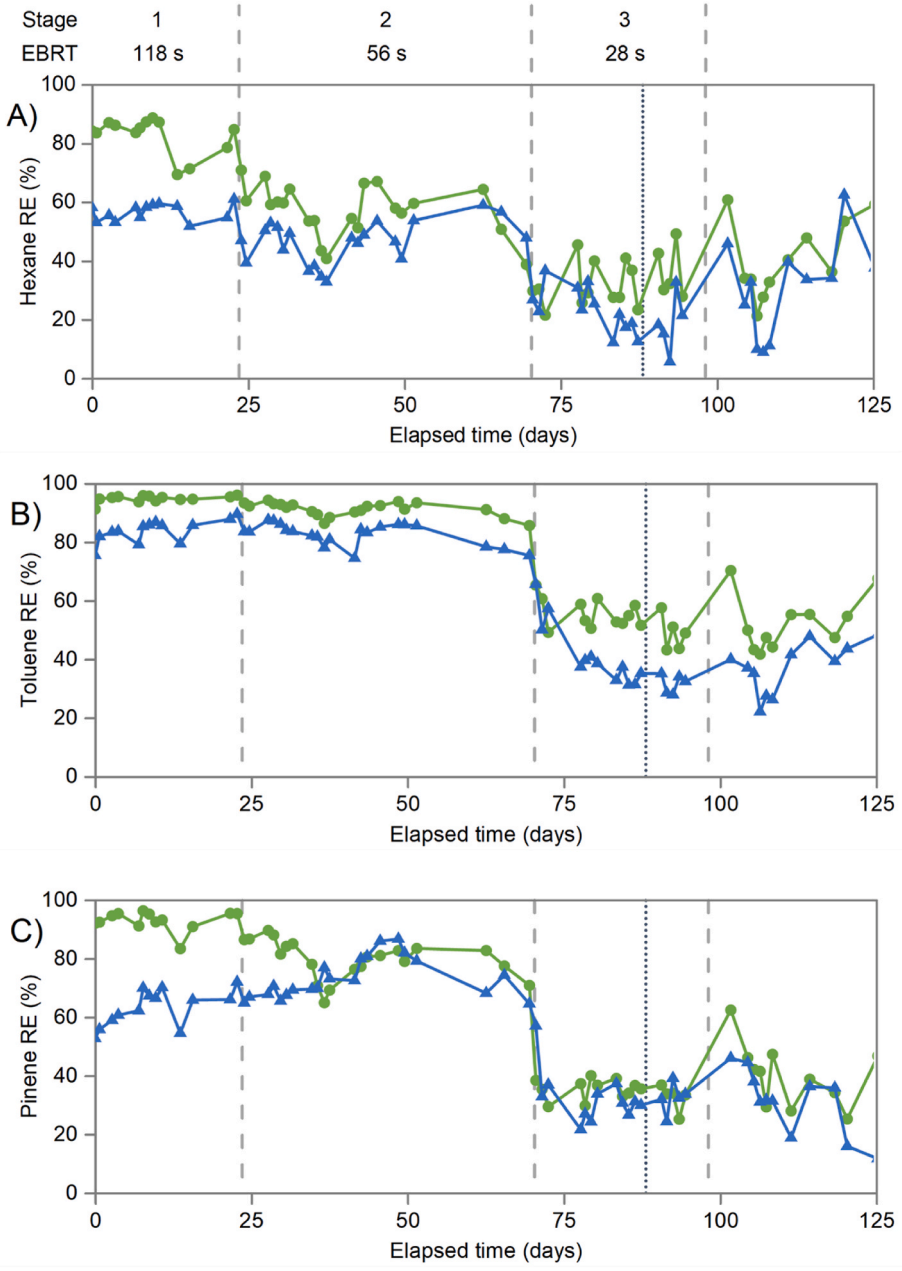

Fig. 2. Time course of the removal efficiency of A) hexane, B) toluene and C) pinene achieved in BF A (—●—) and BF B (—▲—) at different EBRTs in stages 1, 2 and 3. Dashed lines represent the different stages, and the dotted line in stage 3 represents the change in the irrigation volume.

both bioreactors until day 35, when an increase in BF A performance was observed. Final steady REs of 86.6 ± 7.0% in BF A and 49.7 ± 4.1% in BF B were reached. These results pointed out the higher difficulty of microorganisms in adapting to new environments and sources of carbon when entrapped in a bioactive coating rather than in a naturally-formed biofilm.

3.1. EBRT influence on VOC removal

Stage 1 was initiated following the adaptation period, thus similar steady REs were recorded: 82.7 ± 6.1%, 94.8 ± 1.3% and 92.9 ± 3.4% in BF A and 56.2 ± 2.9%, 83.7 ± 4.0% and 63.1 ± 6.4% in BF B for hexane, toluene and pinene, respectively (Fig. 2). Overall, BF A showed an excellent performance, which was especially notable for hexane, a very hydrophobic and persistent compound and thus very difficult to transfer and assimilate by microorganisms.

In stage 2, the EBRT was reduced from 112 s to 56 s. Hexane REs immediately decreased following EBRT reduction, and did not recover the previous values, with average removals of 56.9 ± 8.9% in BF A and 46.1 ± 7.3% in BF B. Interestingly, the removal of toluene was not influenced by the decrease in gas residence time, remaining at steady state values of 91.1 ± 2.4% and 82.5 ± 3.8% in BF A and BF B, respectively. On the contrary, pinene REs clearly decreased in BF A (79.7 ± 3.4%), while the abatement performance remained similar in BF B (73.0 ± 6.9%).

Further reduction in the EBRT to 28 s in stage 3 resulted in a significant deterioration in the performance of both bioreactors. A slight decrease in hexane REs was recorded (≈25%), reaching steady state REs of 32.5 ± 8.1% in BF A and 21.5 ± 8.4% in BF B, with a high variability in both reactors. On the contrary, the reduction in toluene RE was much more pronounced (≈40% in BF A and ≈50% in BF B), decreasing to steady values of 53.4 ± 6.0% and 34.1 ± 4.0%, respectively. A similar deterioration in pinene abatement (≈45%) was observed in both reactors, stabilizing at 34.2 ± 3.8% in BF A and 30.4 ± 5.0% in BF B. The decrease in all VOC REs was attributed to the shorter gas residence time, which entailed a reduction in the contact time between microorganisms and VOCs, thus reducing their bioavailability. Surprisingly, the decrease in hexane removal, the compound with the highest hydrophobicity among the VOCs here tested, was less pronounced than that of toluene and pinene. No consistent explanation could be found for the different impact of EBRT decrease on hexane and toluene/pinene removals. In some studies, the cell surface hydrophobicity was reported to progressively increase after a prolonged exposure to hydrophobic compounds such as surfactants, non-aqueous phase liquids (NAPLs), solvents or some VOCs (Wu et al., 2023). In this case, hexane-degraders could have experienced an increase in cell surface hydrophobicity, which could have helped mitigate the effects of the EBRT reduction. However, other unknown factors could be involved in this different behavior. Additionally, the irrigation rate was increased from 100 to 200 mL day^{-1} at day 88 in stage 3 (Fig. 2, solid line). However, no clear effect was observed regardless of the reactor, as the average REs before and after the change were statistically similar.

Instantaneous deterioration of the biofiltration performance when reducing the EBRT is commonly reported in biofiltration, although the previous abatement performance can be retrieved after a certain acclimation period if operational conditions are favorable enough. Lebrero et al. (2014) observed this phenomenon in a BF and a BTF when the EBRT was reduced during VOC treatment. Yang et al. (2011) also reported a decrease in the removal of toluene from 98% to 64% and 74% in two polyurethane sponge BTFs (structured vs. random) when the EBRT was reduced from 30 to 5 s. Similarly, a toluene RE reduction from 99% to 83% was observed by Chen et al. (2012) in a tubular BF, when EBRT was reduced from 15 to 3.8 s at a constant inlet concentration of 77 mg m^{-3}. However, our bioreactors were not able to recover the preceding steady-state REs achieved before each EBRT decrease. Interestingly, both bioreactors experienced a similar decrease in performance with decreasing EBRTs, which could indicate that the bioactive coating offers a robustness at least as high as the natural-formed biofilm.

3.2. Reduction of inlet VOC concentrations

From stage 3 onwards, the inlet concentration of VOCs was progressively reduced by lowering the rate of the syringe pump while maintaining the EBRT at 28 s (Fig. 3). However, no change in the removal performance was observed. Thus, the first decrease in inlet concentrations exerted no significant effect in the REs of toluene and pinene (<5%). Only hexane REs slightly increased in both bioreactors, but higher fluctuations were also recorded. In stage 5, reducing the inlet concentration only had a visible effect on pinene removal, which increased from 37.5 ± 8.3% to 64.1 ± 10.3% in BF A and from 30.6 ± 11.3% to 47.8 ± 12.1% in BF B. A high variability in hexane removal was also observed in this stage. During stage 6, a slight stabilization, concomitant with a slight decrease in hexane REs (≈7%), was recorded in both bioreactors. Toluene REs were similar to those recorded in stage 5, while pinene REs decreased again to reach values similar to those of stage 4. The last inlet concentration reduction was performed in stage 7, resulting in a minimal influence on VOC REs regardless of the bioreactor. In BF A, hexane removal remained stable at 31.7%. Toluene removal experienced a slight decrease to average values of 47.1%, whereas RE values 6% higher were recorded for pinene to reach 48.1%. In BF B, REs slightly increased to reach steady removals of 30.9% for hexane, 40.8% for toluene and 34.2% for pinene.

Overall, a reduction in the inlet VOC concentrations did not trigger significant variations in any of the bioreactors. In this sense, hexane REs did not change from stage 3 (32.5%) to stage 7 (31.7%) in BF A, while in BF B a slight increase was observed (from 21.5% to 30.9%). In the case of toluene, an average RE of 52.1 ± 7.5% was achieved in BF A in stages 4–7, while in BF B, toluene RE increased from 34.1% in stage 3–40.8% in stage 7. The most significant effect of the progressive decrease in the inlet concentration was observed for pinene removal in BF A. Thus, pinene RE increased from 34.2% in stage 3–48.1% in stage 7, while in BF B the removal only increased from 30.4% to 34.2%.

Contrary to our study, higher removal performances have been reported for the simultaneous treatment of these VOCs at lower EBRTs. For instance, Lebrero et al. (2012) observed steady removals of ≈70% for hexane and ≈96% for toluene and pinene at an EBRT of 11 s, although at lower inlet concentrations (0.22–0.28 mg m^{-3}). However, an important deterioration of the system performance was observed at an EBRT of 7 s, REs decreasing to minimum values of 25%, 40%, 14% for hexane, toluene and pinene, respectively. Similarly, Lebrero et al. (2014) assessed the performance of a biofilter and a biotrickling filter for the abatement of these pollutants. Steady-state REs over 95% were achieved in the biofilter for toluene and pinene at EBRTs as low as 8 s, while hexane removal was slightly lower (≈94%). The biotrickling filter also achieved REs over 95% for toluene and pinene when operating at EBRTs of 8 and 4 s, whereas hexane abatement decreased from 93% to 88% at EBRTs of 8 and 4 s, respectively. These studies were conducted at lower inlet concentrations than those here tested, ranging from 0.75 to 0.91 mg m^{-3}. Based on these previous reports, it was hypothesized that a decrease in the inlet VOC concentrations could facilitate their removal (Valenzuela-Reyes et al., 2014; Yang et al., 2008), despite lower concentrations also entail a decrease in the mass transfer to the degrading microorganisms. The impossibility to recover previous RE levels was attributed to the short EBRTs at the beginning of the experiment. This EBRT reduction likely mediated a severe deterioration in the activity of the microbial community previously alive and active.

On the other hand, lower REs have also been reported in biofiltration systems. For instance, Lebrero et al. (2010) observed steady state REs for pinene of less than 15% in a compost/perlite biofilter during the simultaneous treatment of H_2S, butanone, toluene and α-pinene at an EBRT of 50 s, while REs >90% were achieved for toluene, and REs >95% were achieved for butanone regardless of the EBRT. Similarly, the REs

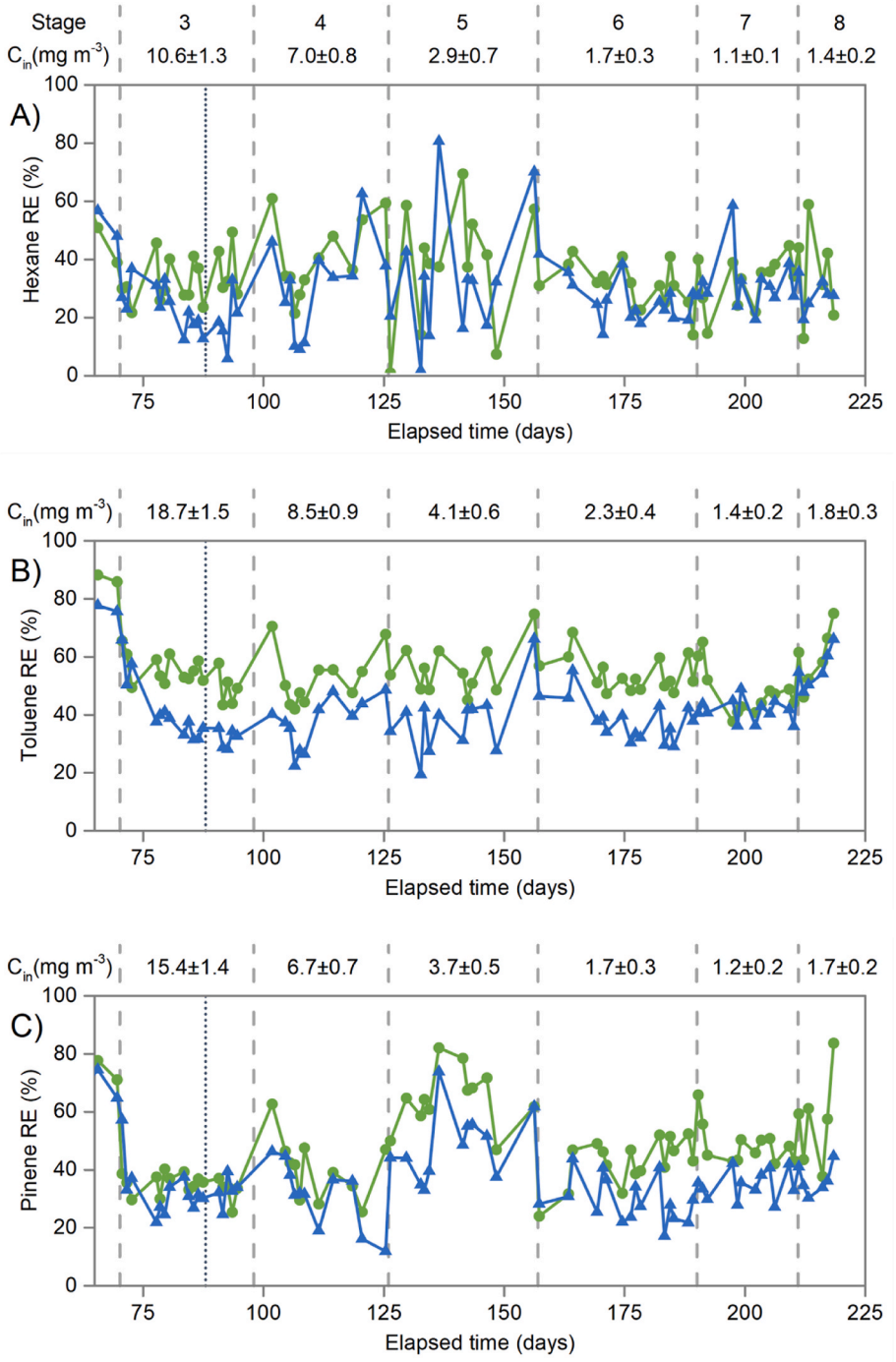

Fig. 3. Time course of the removal efficiency of A) hexane, B) toluene and C) pinene achieved by BF A (—●—) and BF B (—▲—) during inlet VOC concentration reduction in stages 3–8. Dashed lines represent the different stages, and the dotted line in stage 3 represents the change in the irrigation volume.

obtained in this work (especially low for n-hexane) are directly linked with the hydrophobicity of the compound, which severely affects the mass transfer and thus the bioavailability of these VOCs (Cheng et al., 2016). Additionally, other VOC-depending effects have been observed in biofiltration (Yang et al., 2018). For instance, antagonistic interactions between VOCs have been reported to decrease the biofiltration performance. Hence, Amin et al. (2017) recorded hexane REs of 76% as a single pollutant, which decreased to 21% when BTEX were introduced in the BF. Similarly, Hassan and Sorial (2010) and Hu and Wang (2015) observed a decrease in hexane biofiltration performance when benzene was present in the polluted air. A similar antagonistic interaction was reported by Wu et al. (2023b) when the addition of propane and n-hexane decreased the removal of methane in a bioreactor. On the contrary, the addition of methane and propane promoted the removal of n-hexane. Another factor influencing the degradation capacity of the system may be substrate competition, especially when more hydrophilic (and thus more bioavailable) compounds are present (Balasubramanian et al., 2012). Knowledge related to the interactions during the biodegradation of multiple VOCs is still scarce for complex mixtures such as those found in indoor air and need further research.

Finally, at stage 8, the EBRT was restored to the initial value of 112 s without changing the inlet VOCs concentration. This final adjustment aimed at assessing whether the system could recover the initial abatement performance. Although no improvement was observed in hexane

removal, toluene and pinene REs increased by 10–15% after the change, with a more pronounced effect in BF A. In this sense, REs of toluene and pinene achieved steady values of 59.4 ± 10.3% and 56.7 ± 16.2% in BF A, and 55.1 ± 6.7% and 36.1 ± 5.2% in BF B.

Since the progressive reduction of inlet concentrations did not entail a sustained recovery of VOC removal, it could be inferred that the EBRT was the determining factor in VOC removal regardless of the inlet concentrations. The partial recovery of the system when initial EBRT values were retrieved (i.e. 112 s) suggests that, although a possible damage could have occurred, the microbial community is able to recover when operational conditions are more favorable, as previously reported in biofiltration systems (Yu et al., 2021). Overall, little difference in VOC removals was observed between the bioactive coating-based bioreactor and the conventional biofilm bioreactor. Several factors can explain this difference. First, the preparation process of the bioactive coating is much more complex than the inoculation of a biofilm bioreactor, which can lead to uneven distribution of the coating and therefore inadequate supply of the gaseous pollutants to the microorganisms. Moreover, the literature on bioactive coatings is limited to small-scale, batch experiments, whereas in this experiment the bioactive coating was used as bioreactor packing material in a continuous, long-term experiment. The practical application of bioactive coatings involves different new challenges (scale-up of the coating, long-term activity of microorganisms, operating conditions of the bioreactor, etc.) that need to be further investigated.

3.3. Bacterial community structure

At the end of the experiment, the bioreactors contained a more specialized and less diverse community than the inoculum. Bacterial richness and evenness of both specialized communities in BF A and BF B decreased significantly in comparison to the inoculum (AMOVA <0.05). However, BF A and BF B had similar number of OTUs and their beta diversity did not differ significantly between samples. In Fig. 4, a clustered heatmap shows the similarity between the main bacterial orders of BF A and BF B and the clear differentiation between the inoculum and the specialized communities of the BFs by the end of operation. This shows that the tailored communities in both reactors were most likely involved in the degradation of the treated α-pinene, toluene and n-hexane.

Fig. 5 shows the most representative bacterial genera found in each reactor upon inoculation and after long term operation. The enrichment under the target VOCs favored the representation of members from the genera *Devosia* (11.9 ± 0.7 and 10.5 ± 0.4% in BF A and BF B, respectively), *Mesorhizobium* (7.9 ± 0.5 and 4.5 ± 0.3% in BF A and BF B, respectively), *Sphingobacterium* (7.5 ± 0.7 and 7.3 ± 0.1% in BF A and BF B, respectively) and *Mycobacterium* (4.1 ± 0.1 and 5.1 ± 0.1% in BF A and BF B, respectively). In fact, these genera had almost negligible representation in the inoculum (Fig. 4) and their population specialized and dominated along operation (total bacterial members detected are represented in Fig. S2).

Devosia and *Mesorhizobium* belong to the order *Hyphomicrobiales* that represented the most dominant order of the BFs. *Hyphomicrobiales* is

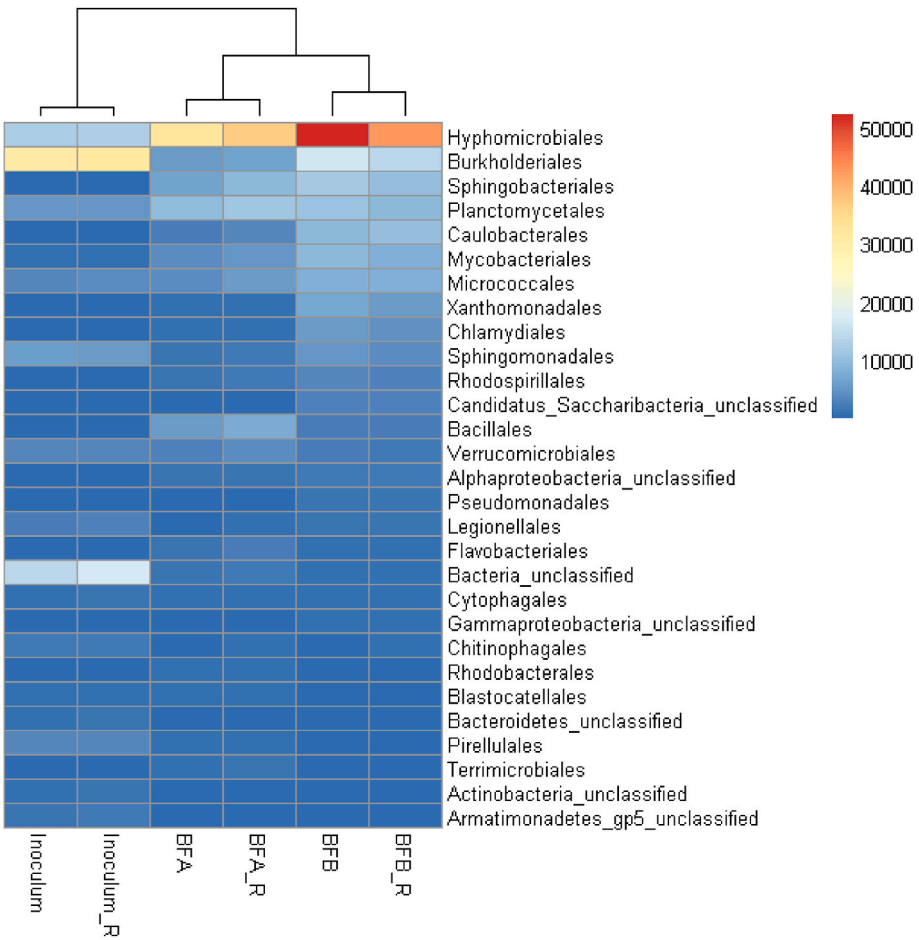

Fig. 4. Heat map of the most abundant bacterial orders in BF A, BF B and original inoculum (99% of the total abundance). Data are presented as the total reads. The dendrogram on top represents similitude between samples according to their communities. Inoculum_R, BFA_R and BFB_R stand for biological replicates during sequencing.

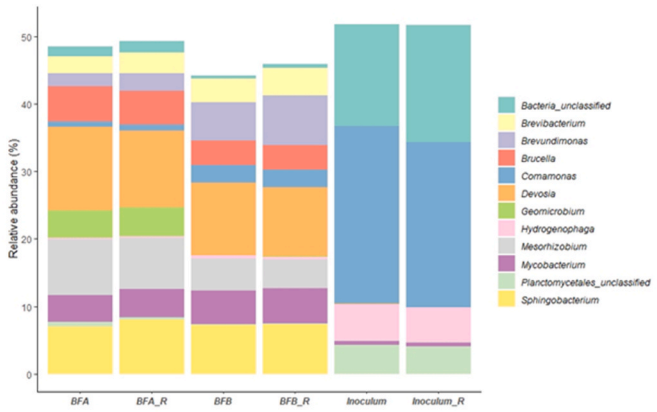

Fig. 5. Bar plot of the main bacterial genera found in each reactor (the R stands for the replica). Only bacterial genera that represented more than 5% relative abundance in one of the stages are represented.

repeatedly found in reactors treating persistent compounds, specifically VOCs (Carvajal et al., 2018; Deng et al., 2017; Yoshikawa et al., 2017). *Devosia* is a bacterial genus dominant in contaminated soil with hydrocarbons. Recent metagenomic analyses have revealed that members from this genus have the metabolic pathway to degrade toluene and some terpenes (Lu et al., 2018; Talwar et al., 2020; Wu et al., 2018a). In the case of *Mesorhizobium* members, genes related to the degradation of toluene, xylene, and other persistent VOCs have not been identified. However, this bacterium appears in several studies as a representative genus in BTEX-degrading microbial consortia (Deng et al., 2017; Weelink et al., 2007). On the other hand, the genus *Sphingobacterium* from the order *Sphingobacteriales* (third most representative order in the BFs) has the capacity to biodegrade polycyclic aromatic hydrocarbons and was probably involved in the bioconversion of the aromatic VOCs fed (Alias et al., 2022). *Mycobacterium* is a recognized degrader of ethene and oxidizes C_2 and C_4 alkanes and alkenes from the order *Mycobacteriales*. Usually, *Mycobacterium* species' role is to metabolize and/or detoxify by-products during BTEX degradation, although some *Mycobacterium* species are also able to degrade toluene (Martin et al., 2014; Tay et al., 1998; Yoshikawa et al., 2017).

Nevertheless, some differences in the relative abundance of the most dominant bacterial members were found between BF A and BF B. Bacteria from the genera *Brucella* (5.0 ± 0.7%), order *Hyphomicrobiales,* and *Geomicrobium,* (4.1 ± 0.2%), order *Bacillales,* were favored in BF A, while in BF B the genera *Brevundimonas* (6.5 ± 1.1%) and *Comamonas* (2.7 ± 0.2%), from the order *Caulobacterales* and *Burkholderiales*, were more representative. This difference found among populations could be related to the use of a latex-based bioactive coating in BF B. The influence of the latex coating on the bacterial population dynamics has been previously detected (Cantera et al., 2022). Bioactive coatings increase mass transfer and pollutant load and therefore can enhance biomass growth, although they also restrain nutrient and water availability for the biofilm. Thus, bioactive coatings most likely favor those bacteria that are more resistant to adverse growth and stressing environmental conditions (Estrada et al., 2015).

4. Conclusions

Bioactive coatings are a promising biotechnology to address the problem of indoor pollution, which involves very low pollutant concentrations and the presence of hydrophobic VOCs. Bioactive coatings have been herein applied on the surface of the packing material (polyurethane foam) of a traditional bioreactor. The VOC abatement performance of the bioactive coating-based bioreactor was similar to that of a traditional biofilm-based bioreactor inoculated with the same microbial community. Removals over 45%, 80% and 70% were obtained for hexane, toluene and pinene at EBRT of 56 s. When the EBRT was decreased to 28 s, removals decreased to 21%, 4% and 30%, respectively. The progressive decrease in inlet VOC concentrations did not clearly improve the abatement performance. In the final stage, low inlet VOC concentrations were maintained while EBRT was shifted back to the initial 112 s. A slight recovery of the abatement performance in the bioactive coating-based bioreactor was finally observed, achieving steady REs of 27%, 55% and 36% for hexane, toluene and pinene, respectively. Therefore, the loss of removal capacity was attributed to the low EBRT, which limited the gas-liquid transfer of VOCs, while the inability to recover the previous performance could be associated with a permanent damage of the metabolic activity of the bacterial community. Future research should focus on the development of new bioactive coatings that can overcome the mass transfer problems and other difficulties associated to long-term indoor VOC abatement at even lower concentrations, and therefore improve the performance of other conventional biotechnologies.

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. Raúl Muñoz: Conceptualization; Writing - Review & Editing; Supervision; Funding acquisition; Project administration. Raquel Lebrero: 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 (Spain) [project RTI 2018-0-096441-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 J. 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.jenvman.2023.119362.

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