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Research article

Effect of surfactant type and concentration on the gas-liquid mass transfer in biotrickling filters used for air pollution control

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ABSTRACT

Volatile organic compounds (VOCs) emitted into the atmosphere negatively affect the environment and human health. Biotrickling filtration, an effective technology for treating VOC-laden waste gases, faces challenges in removing hydrophobic VOCs due to their low water solubility and therefore limited bioavailability to microorganisms. Consequently, the addition of (bio)surfactants has proven to be a promising strategy to enhance the removal of hydrophobic VOCs in biotrickling filters (BTFs). Yet, up to now, no single study has ever performed a mass transfer characterization of a BTF under (bio)surfactants addition. In this study, the effect of (bio)surfactant addition on the gas-liquid mass transfer characteristics of two BTFs was measured by using oxygen (O_2) as a model gas. Through an empirical correlation, the mass transfer coefficients (k_La) of two hydrophobic VOCs, toluene and hexane, which are of industrial and environmental significance, were estimated. One BTF was filled with expanded perlite, while the other with a mixture of compost and wood chips (C + WC). Both BTFs were operated under different liquid velocities (U_L: 0.95 and 1.53 m h^{-1}). Saponin, a biological surfactant, and Tween 80, a synthetic surfactant, were added to the recirculating liquid at different critical micelle concentrations (CMCs: 0–3 CMC). The higher interfacial and surface area of the perlite BTF compared to the C + WC BTF led to higher k_LaO_2 values regardless of the operational condition: 308 ± 18–612 ± 19 h^{-1} versus 42 ± 4–177 ± 24 h^{-1}, respectively. Saponin addition at 0.5 and 1 CMC had positive effects on the perlite BTF, with k_LaO_2 values two times higher compared to those at 0 CMC. Tween 80 exhibited a neutral or slightly positive effect on the mass transfer of both BTFs under all conditions. Overall, the CMC, along with the physical characteristics of the packing materials and the operational conditions evaluated explained the results obtained. This study provides fundamental data essential to improve the performance and design of BTFs for hydrophobic VOCs abatement.

1. Introduction

The emission of volatile organic compounds (VOCs) into the atmosphere represents an environmental and human health threat. Biofiltration is recognized as a cost-effective and sustainable biological treatment technique for the removal of VOCs from waste gas streams (Rybarczyk, 2022). Biotrickling filters (BTFs) are of particular interest because of the advantages they offer, including low operating costs, excellent operational control and robustness (Estrada et al., 2014).

BTFs are fixed-bed bioreactors where active microorganisms grow as a biofilm on a packing material and degrade the VOCs present in the waste stream. The pollutants serve as a source of carbon and energy to the microorganisms and, under aerobic conditions, they are typically converted into biomass, carbon dioxide (CO_2) and water (H_2O). An

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aqueous solution containing the essential nutrients for the microorganisms is trickled over the bed and continuously recirculated at a specific velocity (Rybarczyk, 2022). The removal of VOCs in BTFs involves a combination of various physical-chemical and biological phenomena, such as absorption, adsorption, diffusion and biodegradation. Initially, the VOCs present in the gas phase and passing through the bed must be transferred to the aqueous and biofilm phases. Subsequently, biodegradation occurs (Lamprea Pineda et al., 2021). Therefore, the removal of VOCs in BTFs is governed by (i) the mass transfer rate of VOCs from the gas phase to the biofilm, and (ii) the biodegradation reaction. In this sense, depending mainly on the type of VOC and operational conditions, the process can be mass-transfer or kinetically limited (Ferdowsi et al., 2016; Sarkar et al., 2022).

Hydrophobic VOCs play an important role in the production of a wide range of products and are released into the atmosphere by multiple industries. They are often used as solvents and raw materials or as intermediates in the pharmaceutical, textile, printing and coating, petrochemical, and cosmetics and personal care industries, among others (Wang et al., 2013). Moreover, hydrophobic VOCs such as toluene and xylene have the potential to cause neurological harm. In contrast, others such as benzene, ethylbenzene and trichloroethylene are known to be toxic and carcinogenic (David and Niculescu, 2021). The low aqueous solubility of hydrophobic VOCs is translated into limited bioavailability to the microorganism (i.e., mass transfer limitation) and, therefore, poor removal performances in BTFs. In the last decades, different strategies to enhance the removal of hydrophobic VOCs in BTFs have been evaluated (Cheng et al., 2016; Rybarczyk, 2022). Among them, the addition of surfactants has shown promising results in terms of removal performance by increasing the removal efficiencies (REs) and elimination capacities (ECs) of hydrophobic pollutants (Lamprea Pineda et al., 2021). Surfactants are amphiphilic compounds (i.e., with both a hydrophobic -tail- and hydrophilic -head- moiety). They reduce surface and interfacial tensions between two immiscible phases (e.g., liquid-gas, liquid-liquid and/or solid-liquid) and significantly increase the solubility of a compound in a medium when added at concentrations beyond their critical micelle concentration (CMC). The CMC corresponds to the concentration at which micelles are formed, and is characteristic for each type of surfactant. Micelles are aggregates of surfactant monomers with their hydrophobic heads arranged on the outer periphery and the hydrophilic tails positioned in the center (Singh et al., 2007). As an example, the addition of Tween 80 (100 mg L^{-1}; CMC: 15.7 mg L^{-1}), a synthetic surfactant, to a BTF filled with polyurethane foam (PUF) and treating a gaseous stream of m-xylene (inlet load (IL): 216 g m^{-3} h^{-1} at an empty bed residence time (EBRT) of 20 s) increased the m-xylene RE and EC by 20% and 24%, respectively (Wang et al., 2018). The authors concluded that Tween 80 could enhance the bioavailability of m-xylene and therefore improve its degradation efficiency.

The biofiltration studies carried out so far to evaluate the effect of surfactants addition have mainly focused on the efficacy of the process, and a comprehensive compilation of these studies can be found in Lamprea Pineda et al. (2021). In such studies, complementary measurements of Henry's law constants (or gas-liquid partitioning coefficients) (Dewidar and Sorial, 2021; Wu et al., 2022) and/or surface tension (He et al., 2020; Yang et al., 2022) are usually conducted, although typically under ideal conditions far from real biofiltration experiments, i.e., using mostly water instead of a mineral medium and not considering the presence of packing material. However, despite this strategy aiming to alleviate the mass transfer limitation of hydrophobic VOCs, to date, no study has conducted a mass transfer assessment of a BTF under surfactant addition. To the best of our knowledge, mass transfer studies involving the addition of surfactants have only been conducted in bubble columns (Belo et al., 2011; Jia et al., 2015; Mcclure et al., 2015; Nekoeian et al., 2019; Painmanakul et al., 2005; Sardeing et al., 2006), stirred tanks (Alves et al., 2004; Garcia-Ochoa and Gomez, 2005; Hebrard et al., 2009) and airlift reactors (Moraveji et al., 2012a, 2012b). All of them, except for Garcia-Ochoa and Gomez (2005), employed synthetic surfactants. Moreover, in each case, the evaluation focused on the effect of surfactants addition on the mass transfer of either CO_2 or oxygen (O_2) as model compounds, with the former presenting safety concerns due to the use of caustic water (Estrada et al., 2014). Therefore, enhancing the performance and design of BTFs for hydrophobic VOCs abatement requires a thorough understanding of the degree to which the VOC mass transfer rate improves with the addition of both synthetic and biological surfactants. The latter are particularly important due to their recognized biodegradability, environmental friendliness, and lower toxicity in comparison to their synthetic counterparts (Mulligan et al., 2001; Souza et al., 2014; Tu et al., 2015).

In this study, we evaluated the effect of surfactant addition by using O_2 as a model gas for which the volumetric mass transfer coefficient (k_La) was experimentally measured in two non-inoculated BTFs, i.e., one filled with expanded perlite and one with a mixture of compost and wood chips (C + WC), both operated under different trickling liquid velocities (U_L). To this aim, the oxygen transfer rate (OTR) method, based on the oxidation of sulfite in a liquid medium, was implemented. This method is considered as a reliable and rapid technique for the determination of k_La coefficients (Muñoz et al., 2018). Two surfactants were chosen, i.e., a synthetic (Tween 80) and a biological (saponin) one. They are both commercially available and have already been evaluated in biofiltration studies, but only in terms of performances, biofilm characteristics, and microbial growth and identification ((Lamprea-Pineda et al., 2024; Qian et al., 2018; Tu et al., 2015; Wang et al., 2018)). Moreover, the k_La values of two common hydrophobic VOCs (i.e., toluene and hexane) – widely investigated under surfactants' addition and of industrial and environmental significance– were estimated from the experimental values obtained for O_2.

2. Materials and methods

2.1. Biotrickling filters (BTFs) set-up

The mass transfer characterization was performed in two identical lab-scale BTFs (Fig. 1). Each BTF consisted of a cylindrical PVC column with a working packed bed volume of 2 L (12 cm of internal diameter and 18 cm of height). One BTF was packed with a mixture of compost (5% v/v) (Compo Sana, COMPO IBERIA S.L., Spain) and wood chips

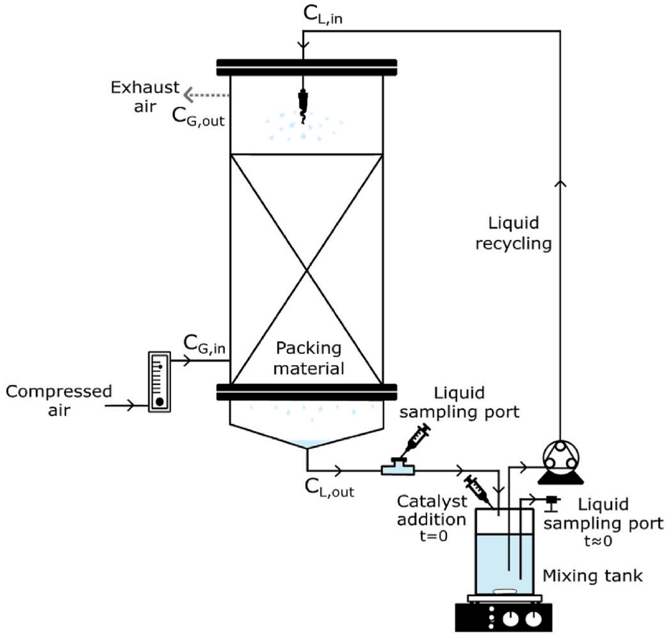

Fig. 1. Biotrickling filter (BTF) set-up. C_G stands for gas concentration, while C_L for liquid concentration.

(95% v/v) (pine bark; granulometry: 15–20 mm) (GEOLIA S.L., Madrid, Spain) (C + WC), while the other BTF was packed with expanded perlite (granulometry: 2–6 mm) (MASSÓ Garden, Barcelona, Spain). An air stream supplied by an air compressor (ABAC B2500-50 2, Robassomero, Italy) flowed from bottom to top at a constant flow rate of 0.8 L min^{-1}, corresponding to an EBRT of 153 s and a gas velocity (U_G) of 4.24 m h^{-1}. A 1 L liquid solution containing sodium sulfite 0.05 M (Na_2SO_3 assay = 98–100%; Sigma-Aldrich, Madrid, Spain) was continuously agitated at 600 rpm in an external 1.5 L mixing tank and recirculated through the BTFs at a U_L of 0.95 or 1.53 m h^{-1}. A long EBRT and low U_Ls were selected to mimic the desired conditions for the removal of hydrophobic VOCs. These compounds require extended contact times for efficient removal, while low water percolation is desired to reduce gas-liquid mass transfer limitation (Estrada et al., 2014; Rybarczyk, 2022). To guarantee a uniform distribution of the liquid solution over the packed bed, a PVC cone nozzle (S.S. CO. B ¼ HHSJ-PVC6007; Spaying Systems Co., Madrid, Spain) was installed in the upper part of the BTF. Two sampling ports allowed the collection of liquid samples at the bottom of the BTF and in the mixing tank. Tween 80 (assay: not reported; Sigma-Aldrich, Madrid, Spain) and saponin (Saponin Quillaja sp. - Sapogenin content: 20–35%; Sigma-Aldrich, Madrid, Spain) were added to the mixing tank at different CMCs (Table S1) and tested at each U_L. Each test was performed in triplicate (Table 1) and the experimental set-up was kept at a constant temperature of 25 °C.

The packing materials were characterized in terms of bulk density (ρ_b), moisture content (Mc), water holding capacity (WHC) and interparticle porosity (Φ) according to Lamprea Pineda et al. (2023) and TMECC (2002) (Table 2).

2.2. Surface tension measurements

To evaluate the effect of surfactant addition at different CMCs on the surface tension of the liquid solutions employed in this study (i.e., 0.05 M Na_2SO_3 and 0.05 M Na_2SO_3 with saponin or Tween 80), surface tension measurements were carried out by the pendant drop method using a FTA200 instrument (First Ten Ångstroms Inc., Portsmouth, VA, USA). Additionally, to get some insights into the interaction of the packing materials with the (bio)surfactants, measurements of the remaining liquid (with and without (bio)surfactant) after contact with the packing material were conducted. To this aim, samples of C + WC and expanded perlite were mixed in 150 mL plastic containers with liquid solutions containing either 0.05 M Na_2SO_3 or 0.05 M Na_2SO_3 with 0.5–3 CMC saponin or Tween 80. The packing material-liquid volume ratio (2:1) used was the same as in the BTFs. The solution was mixed at 180 rpm for 2 min and settled for 1 h. Afterward, a sample of the supernatant was taken and measured in the FTA200. In both cases, liquid densities were experimentally measured at room temperature by weighing a precise volume of the solution to later correct the surface tension measurements. The surface tension of each sample was measured five times, and reported values represent their arithmetic average.

Table 1
Set of experiments carried out in this study. CMC stands for critical micelle concentration, U_L for liquid velocity and U_G for gas velocity.

Surfactant	Surfactant concentration (CMC)[a]	U_L (m h^{-1})	U_G (m h^{-1})
Tween 80	0	0.95	4.24
	1		
	3		
Saponin	0		
	0.5		
	1		
	3		
Tween 80	0	1.53	
	1		
	3		
Saponin	0		
	1		
	3		

[a] The CMC value in pure water and at room temperature (25 °C) is used in this manuscript and corresponds to 15.7 mg L^{-1} for Tween 80 and 1000 mg L^{-1} for saponin (see Table S1).

Table 2
Packing material (PM) characteristics. The table shows average values ± standard deviations (SD) for measurements carried out in triplicate.

Property	Expanded perlite	Mixture C + WC
Bulk density - ρ_b (g mL^{-1})	0.107 ± 0.004	0.32 ± 0.01
Moisture content - Mc (%)	0.63 ± 0.13	5.7 ± 1.0
Water holding capacity - WHC (g H_2O g PM^{-1})[a]	3.7 ± 0.4	0.496 ± 0.002
Interparticle porosity - Φ (%)[b]	61 ± 1	58 ± 2
Specific surface area (m[b] g^{-1})	1.2–6.5[c]	Unknown

[a] Expressed per unit of packing material at ambient conditions.

[b] Measured for each packing material "as received" i.e., without drying or water saturation.

[c] Range of values reported by Bruneel et al. (2020), Celik et al. (2013) and Hernández-Meléndez et al. (2008) for expanded perlite.

2.3. Determination of the volumetric mass transfer coefficients

The OTR method was used to determine the $k_La_{O_2}$. The OTR method is based on monitoring the oxidation of sulfite (SO_3^{2-}) in the mixing tank according to Muñoz et al. (2018).

At the beginning of each experiment, new packing material was placed in the BTF. A nitrogen (N_2) stream (1 L min^{-1} for ~10 min) was used to eliminate the dissolved O_2 present in the mixing tank that contained Na_2SO_3 with or without a (bio)surfactant (see Table 1). Afterward, 5 mL of a catalyst solution containing 0.05 M $CoSO_4 \cdot 7H_2O$ (assay ≥99%; Sigma-Aldrich, Madrid, Spain) was injected to support the instantaneous oxidation of SO_3^{2-} according to Eq. (1).

$$SO_3^{2-} + \frac{1}{2}O_2 \xrightarrow{Co^{2+}} SO_4^{2-} \tag{1}$$

The injection of the catalyst solution was considered as time zero. Simultaneously, an aqueous sample of 3 mL was taken from the bulk phase of the mixing tank, and the recirculation of the liquid solution through the BTF was started. Subsequently, 3 mL samples were taken at the bottom of the BTF every 2–3 min (see Fig. 1), and the kinetics of O_2 absorption were recorded until complete SO_3^{2-} depletion. The SO_3^{2-} concentration in each sample was determined by iodometric back-titration according to Zhao et al. (1999). Briefly, each sample was placed in a 50 mL Erlenmeyer containing 1 mL of an iodine solution (i.e., a titrant solution containing: 0.08 M KIO_3 (assay: 99.5%; Sigma-Aldrich, Madrid, Spain), 0.06 M KI (assay: 99%; Sigma-Aldrich, Madrid, Spain) and 0.3 M H_2SO_4 (assay: 96%; Sigma-Aldrich, Madrid, Spain)). The sample was titrated with 0.03 M sodium thiosulfate ($Na_2S_2O_3 \cdot 5H_2O$) (assay ≥99.5%; Sigma-Aldrich, Madrid, Spain) using a solution of 1% on a mass basis of potato starch (assay: not reported; Panreac, Barcelona, Spain) as indicator. Each sample was immediately titrated after sampling to minimize contact with ambient air. Moreover, a sample of distilled water was processed as a blank control every day. The absorbed O_2 (mg L^{-1}) was estimated from Eq. (1) and Eq. (2) considering the sample volume. By plotting the mg O_2 L^{-1} versus time, the OTR (mg O_2 L^{-1} min^{-1}) was calculated from the slope of 4–8 data points.

$$SO_3^{2-} + I_2 + H_2O \rightarrow 2I^- + SO_4^{2-} + 2I^- + 2H^+ \tag{2}$$

Next, the $k_La_{O_2}$ was determined using Eq. (3):

$$k_La_{O_2} = \frac{OTR}{\frac{C_{O_2,G}}{H_{O_2}} - C_{O_2,L}} \tag{3}$$

where H_{O_2} stands for the dimensionless Henry's law constant of O_2 at 25 °C (i.e., 32.08 (Rettlch et al., 1981)), $C_{O_2,G}$ for the O_2 concentration in the exhaust gas (i.e., O_2 concentration in the air stream), and $C_{O_2,L}$ for the liquid O_2 concentration in the mixing tank (i.e., 0 due to N_2 flushing at the beginning).

Finally, the k_La of two model hydrophobic VOCs (i.e., toluene and hexane) was estimated from the k_La of O_2 (i.e., reference compound, ref) according to Estrada et al. (2014) and Eq. (4).

$$\frac{kLa_x}{kLa_{ref}} = \frac{\left(1/V_{m,x}\right)^{0.4}}{\left(1/V_{m,ref}\right)^{0.4}} \tag{4}$$

where $V_{m,x}$ and $V_{m,ref}$ stand for the molecular volume at the boiling point of the model hydrophobic VOC and the reference compound, respectively. In this study, V_m values of 25.6, 118.2 and 140.6 mL mol^{-1} were used for O_2, toluene and hexane, respectively (Mackay et al., 2006a, 2006b; Welty et al., 2008). All experiments (OTR determinations) were carried out in triplicate.

3. Results and discussion

3.1. Surface tension measurements

The addition of saponin and Tween 80 induced a decrease in the surface tension of the aqueous solutions compared to the surface tension of pure water, i.e., 0 CMC (72.5 ± 0.2 mN m^{-1}) (Fig. 2). A sharp decrease was observed when saponin was added, with an almost constant surface tension at surfactant concentrations above 1 CMC. The highest saponin concentration tested (3 CMC) resulted in a surface tension of 44.6 ± 0.1 mN m^{-1}, more than 1.6 times lower than that of pure water. On the contrary, the addition of Tween 80 resulted in only a slight decrease in surface tension when compared to pure water, with no significant differences between 2 (60.2 ± 0.8 mN m^{-1}) and 3 CMC (58.1 ± 0.9 mN m^{-1}). The presence of 0.05 M Na_2SO_3 did not influence the surface tension of pure water and showed no significant differences for all Tween 80 concentrations tested. In the aqueous solutions containing saponin, a slight difference was observed upon the addition of 0.05 M Na_2SO_3, but only at 0.5 and 1 CMC.

Previous publications have already reported a notable decrease in the surface tension of aqueous solutions with the addition of (bio)surfactants, as well as its subsequent stabilization regardless of further (bio) surfactant increments. For example, Mitra and Dungan (1997) observed a surface tension decrease from ~50 up to ~36 mN m^{-1} when Quillaja saponin was added at a concentration of 0.5 g L^{-1}, while higher biosurfactant concentrations (e.g., up to 10 g L^{-1}) did not lead to any further decrease in surface tension. Similar findings have been achieved by Patist et al. (2000) for Tween 20, 22, 40, 60 and 80; Triton X-100; Brij 35, 58 and 78; pentaethyleneglycol mono n-dodecyl ether, and octaethyleneglycol mono n-dodecyl ether.

The CMC, which is a characteristic value for each surfactant, is in fact represented by the break in the plot of the measured physical property (surface tension in this case) against surfactant concentration. However, depending on the measuring device (e.g., surface tention versus dye solubilization) and surfactant type (e.g., (bio)surfactant source and impurities present), a clear break is not always observed and some authors prefer to report a range of CMC instead (Garidel et al., 2021; Mitra and Dungan, 1997; Qazi et al., 2020).

The sharp decreases in surface tention observed for both Tween 80 and saponon in Fig. 2 indicate that the CMC value of each (bio)surfactant is within the order of magnitude of the values reported in the literature and used in this study (see Table S1). However, the purpose of these measurements was not to identify the CMC of both (bio)surfactants but merely to corroborate the decrease in surface tension when (bio) surfactants are added and, more importantly, to quantify the effect of the addition of Na_2SO_3 in the surface tension of the aqueous solutions. The latter is of particular interest because salts are known to modify significantly the CMC of (bio)surfactants. Yet, this depends on the (bio)surfactant (i.e., the hydrophobic and hydrophilic groups), the salt and its concentration (Miyagishi et al., 2001). For instance, Qazi et al. (2020) have experimentally demonstrated that the addition of NaCl up to 6.5 M to aqueous solutions of Tween 80 does not affect the CMC. In contrast, Mitra and Dungan (1997) observed that the addition of 0.1–1 M NaCl notably decreases the saponin CMC (up to 8.5 times lower), although this decrease appeared less pronounced for NaCl concentrations below 0.2 M (1.5 times lower).

The interaction between the packing material (perlite and C + WC) with the aqueous solutions containing Na_2SO_3 and saponin or Tween 80 altered the surface tension of the aqueous solutions (Fig. 3). At 0 CMC, the surface tension recorded for the perlite supernatant was the same as that of pure water (Fig. 3 A), while for the C + WC supernatant, a slight decrease (8.4% lower) was observed. Such a decrease has been reported before for soil water, due to the presence of dissolved organic matter (Tschapek et al., 1978). At saponin and Tween 80 concentrations of 0.5, 1 and 3 CMC, both the perlite and C + WC supernatant showed slightly higher surface tensions compared to the aqueous solutions without perlite (4–20% higher) and C + WC (max. 13% higher), respectively (Fig. 2). This observation suggests a potential interaction between the surfactant and the packing material, such as adsorption.

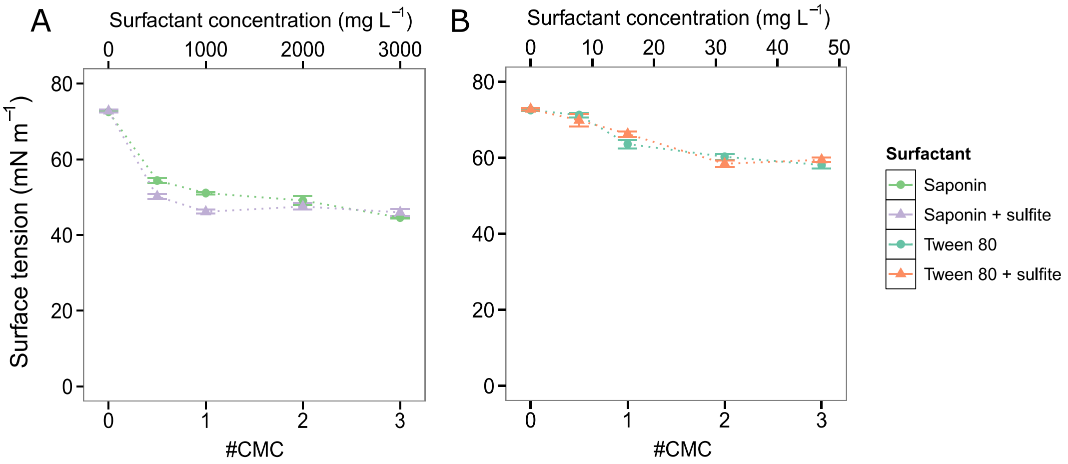

Fig. 2. Surface tension as a function of surfactant concentration (critical micelle concentration (CMC) and mg L^{-1}) for saponin (A) and Tween 80 (B) in pure water (dots) and in water with 0.05 M sodium sulfite (Na_2SO_3) (triangles). The error bars represent the standard deviation (n = 5) of the measured surface tension (see Section 2.2). Note: the dotted lines do not represent continuous values of surface tension for the #CMC but merely connect the measured points to guide the eye.

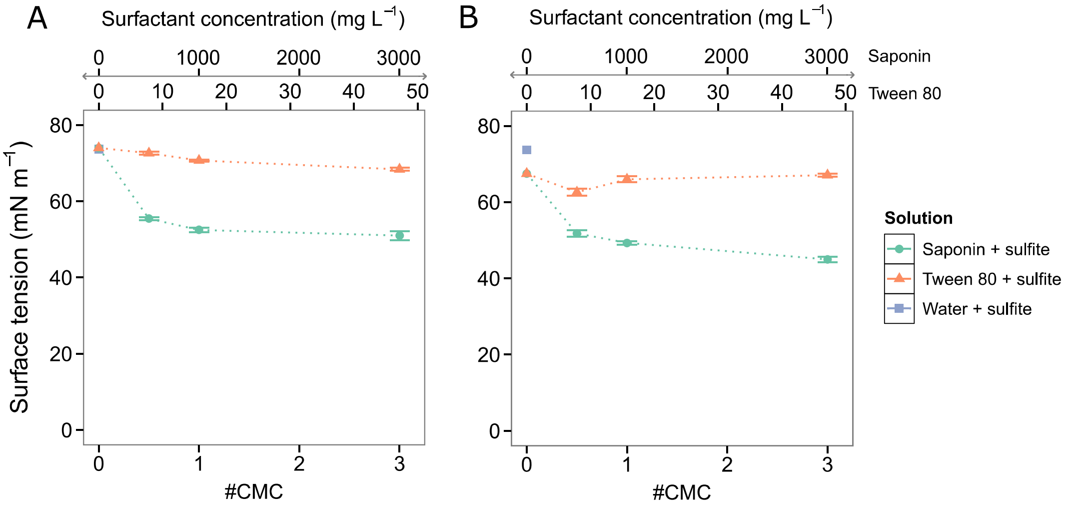

Fig. 3. Surface tension as a function of surfactant concentration (critical micelle concentration (CMC) and mg L^{-1}) of the supernatant of expanded perlite (A) and the mixture of compost and wood chips (C + WC) (B) with saponin and 0.05 M sodium sulfite (Na_2SO_3) (green dots), Tween 80 and 0.05 M Na_2SO_3 (orange triangles), and pure water and 0.05 M Na_2SO_3 (purple squares). The error bars represent the standard deviation (n = 5) of the measured surface tension (see Section 2.2). Note: the dotted lines do not represent continuous values of surface tension for the #CMC but merely connect the measured points to guide the eye. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

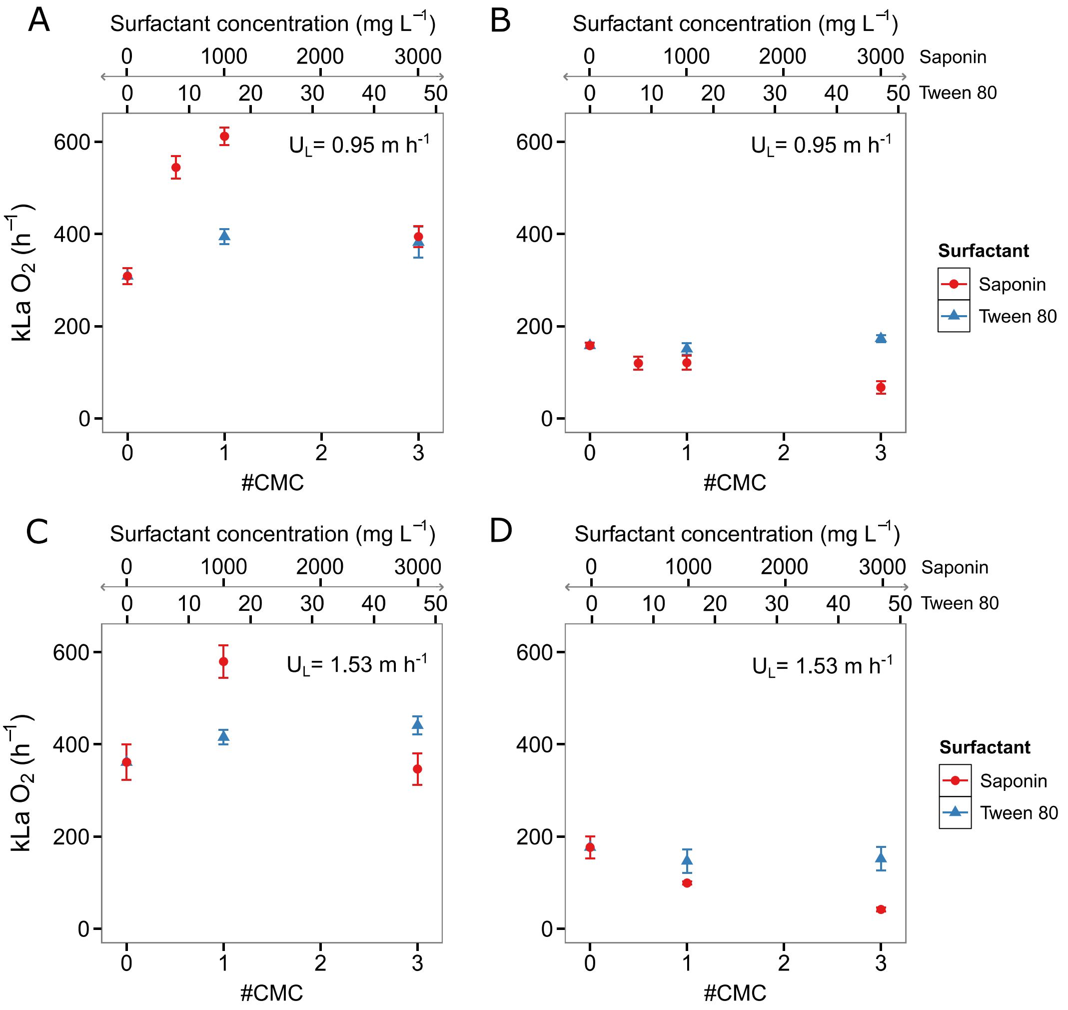

Fig. 4. Measured volumetric mass transfer coefficients of O_2 (k_LaO_2) in the BTF filled with expanded perlite (A, C) and with a mixture of compost and wood chips (C + WC) (B, D) and as a function of saponin (red dots) and Tween 80 (blue triangles) concentrations. Measurements were carried out at two different liquid velocities (U_L) and in triplicate. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.2. *Mass transfer characterization*

3.2.1. *Oxygen mass transfer coefficients*

At 0 CMC and both liquid velocities, the k_LaO_2 values for the perlite BTF were 2 times higher than those obtained for the C + WC BTF (Fig. 4 and Table S2). A k_LaO_2 increase of 1.2 times was observed in both BTFs when the U_L increased from 0.95 m h^{-1} (Perlite: 308 ± 18 h^{-1}; C + WC: 158 ± 5 h^{-1}) to 1.53 m h^{-1} (Perlite: 361 ± 39 h^{-1}; C + WC: 177 ± 24 h^{-1}). The lower k_LaO_2 values observed in the C + WC BTF are attributed to the lower interfacial and surface area of this packing material compared to the expanded perlite. Additionally, as highlighted by Kim and Deshusses (2008), the heterogeneous nature of the C + WC bed can be translated into air channeling, leading to a subsequent decrease in the surface area for mass transfer due to compost relocation in the bed. On the other hand, the increase in k_LaO_2 obtained with increasing U_L is consistent with the results observed in other studies (Estrada et al., 2014; Kim and Deshusses, 2008; Moreno-Casas et al., 2020). An increase in the liquid trickling velocity over the bed results in higher O_2 concentration differences between the two phases, and therefore in a higher concentration gradient for mass transfer. Moreover, higher turbulence at higher liquid velocities increases the contact between both phases (Kim and Deshusses, 2008; Moreno-Casas et al., 2020).

The k_LaO_2 values here obtained at 0 CMC are higher than those reported in the literature for BTFs filled with PUF and operated under similar conditions. For example, Moreno-Casas et al. (2020) experimentally measured a k_LaO_2 of 113 ± 3 h^{-1} for a PUF BTF at an U_L of 2 m h^{-1} and an EBRT of 240 s. The highest recorded k_LaO_2 value of 381 ± 30 h^{-1} was achieved at 17 m h^{-1} and 17 s, comparable to that of the perlite BTF in our study. Estrada et al. (2014) reported values of ~140 h^{-1} and 30 h^{-1} in a PUF BTF operated at 120 s and 5 m h^{-1}, and at 240 s and 0.6 m h^{-1}, respectively, with k_LaO_2 values of up to ~300 h^{-1} when operating at 12 s and 5 m h^{-1}. In addition, Kim and Deshusses (2008) reported k_LaO_2 values as high as ~275 h^{-1} for a BTF filled with porous ceramic beads and operated at an EBRT of 1.8 s and a U_L of 10 m h^{-1}. This indicates a high mass transfer potential for our BTFs configurations and packing materials, particularly for the perlite BTF. According to the literature (see Table 2), perlite exhibits a specific surface area that can be between 50 and 3000 times higher than those reported for common packing materials used in BTFs such as PUF, pall rings and porous ceramic beads (Kim and Deshusses, 2008).

The addition of (bio)surfactants to the mixing tank solution showed different k_LaO_2 trends depending on the BTF packing material and the (bio)surfactant (Fig. 4 and Table S2). In the perlite BTF, adding saponin up to 1 CMC improved the k_LaO_2 by a factor of 2.0 (612 ± 19 h^{-1}) and 1.6 (579 ± 36 h^{-1}) compared to 0 CMC at 0.95 and 1.53 m h^{-1}, respectively. On the contrary, a further increase in saponin concentration up to 3 CMC led to a substantial decrease in the k_LaO_2 value (394 ± 23 h^{-1} at U_L: 0.95 m h^{-1} and 346 ± 34 h^{-1} at U_L: 1.53 m h^{-1}) compared to 1 CMC. In the case of Tween 80 and at both U_Ls, the k_LaO_2 increased by only a factor of ~1.2 from 0 to 1 CMC, and no significant differences were observed between 1 and 3 CMC. The C + WC BTF, on the other hand, exhibited a decrease in k_LaO_2 with increasing saponin concentrations, reaching values as low as 67 ± 13 h^{-1} and 42 ± 4 h^{-1} at 3 CMC and 0.95 and 1.53 h^{-1}, respectively, i.e., 2.4 and 4.2 times lower than without surfactant addition. The addition of Tween 80 at each CMC showed a very limited effect at both U_Ls.

By plotting the k_LaO_2 as a function of the U_L (Fig. 5), for the perlite BTF, it is possible to observe a slight, although not significant, decrease in k_LaO_2 at 1.53 m h^{-1} when saponin was added at both 1 and 3 CMC, compared to 0.95 m h^{-1} (Fig. 5 A). The opposite trend (increasing k_LaO_2 with increasing U_L) was observed for Tween 80 (Fig. 5 C), with significant differences only at 3 CMC (382 ± 34 h^{-1} at 0.95 m h^{-1} versus 441

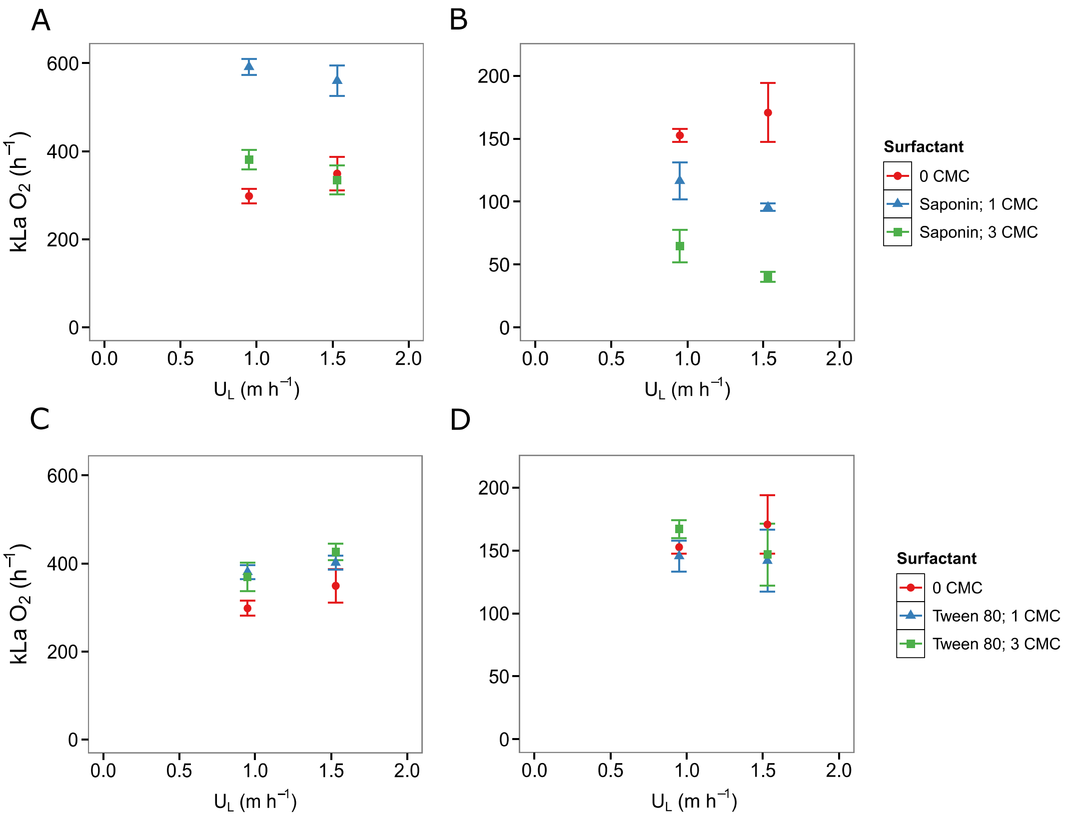

Fig. 5. Measured volumetric mass transfer coefficients of O_2 (k_LaO_2) in the BTF filled with expanded perlite (A, C) and with the mixture of compost and wood chips (C + WC) (B, D) and as a function of the liquid velocity (U_L) when saponin and Tween 80 are added at 0 (red dots), 1 (blue triangles) and 3 (green squares) critical micelle concentrations (CMC). Measurements were carried out in triplicate. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

± 19 h^{-1} at 1.53 m h^{-1}). The increase in U_L in the C + WC BTF resulted in slight decreases in the k_LaO_2 when either saponin or Tween 80 were added, although these changes were not statistically significant (Fig. 5B–D).

Overall, the results demonstrate that, under the conditions here evaluated (i.e., U_L: 0.95 and 1.53 m h^{-1} EBRT: 153 s and U_G: 4.24 m h^{-1}), there is an optimal saponin concentration (1 CMC in this study) that boosted the mass transfer in the perlite BTF. On the contrary, negligible and even negative effects are obtained at higher CMCs and/or other packing materials. For example, in the C + WC BTF, a decrease in mass transfer is observed after the addition of saponin at 0.5–3 CMC. In the case of Tween 80 and at both 1 and 3 CMC, only a slight enhancement was observed in the perlite BTF compared to the C + WC BTF where the effect was almost negligible.

The dissimilarity between both BTFs might be explained by the differences in their packing materials and the amount of (bio)surfactants dosed. At the highest U_L tested, both the perlite and the C + WC BTFs showed foaming when saponin was added, either at the top of the bed (perlite BTF) or within the packed bed (C + WC BTF). This was associated with the higher compaction of the perlite bed compared to the C + WC bed, and the non-uniform distribution of wood chips, leading to significant void spaces in the C + WC BTF that resulted in preferential flow and channeling. Besides this, in terms of mass, higher amounts of saponin (~64 times higher) were added compared to Tween 80 (see Table S1), which likely contributed to the foaming observed at 3 CMC in the perlite BTF and at 1 and 3 CMC in the C + WC BTF, at both U_Ls, and compared to 0–1 and 0 CMC, respectively. This could indeed explain the decrease in the k_LaO_2 value in both BTFs at the corresponding CMCs. Based on this rationale, it is evident that a higher U_L would result in increased foaming and thus lower k_LaO_2 values, as illustrated in Fig. 5.

3.2.2. *Estimation of mass transfer coefficients for different gas pollutants*

The k_La estimation of the two model hydrophobic VOCs, i.e., toluene and hexane, using the experimental k_LaO_2 values shows that toluene mass transfer is slightly higher (1.1 times), although not significantly different, than that of hexane (see Fig. S1). For toluene, the estimated k_La values range between 86 ± 3 (C + WC BTF, U_L: of 0.95) and 196 ± 21 h^{-1} (perlite BTF, U_L of 153 m h^{-1}) at 0 CMC, and an EBRT of 153 s. These are in the same order of magnitude than those reported in literature for toluene in PUF BTFs ranging between 35 and 113 h^{-1} (EBRTs: 50-7 s and U_L: 0.6 m h^{-1}) (Lebrero et al., 2012) and 12-258 h^{-1} (EBRTs: 100-6 s and U_L: 2 m h^{-1}) (Dorado et al., 2009). On the contrary, the estimated hexane k_La values correspond to 80 ± 3 (C + WC BTF, U_L: 0.95) and 183 ± 20 h^{-1} (perlite BTF, U_L: 153 m h^{-1}) at 0 CMC, and an EBRT of 153 s, which are considerably higher than those reported in the literature by for example Hernández et al. (2011) in a PUF BTF (from 25 to 60 h^{-1} at EBRTs between 40 and 120 s and a U_L of 2 m h^{-1}).

As this estimation (Eq. (4)) is based on the molecular volume and not the hydrophobicity of the compounds of interest, O_2 shows a higher mass transfer than toluene (i.e., almost two times higher), even though its dimensionless Henry's law constant is more than 100 times higher than that of toluene (i.e., 0.27 at 25 °C (Howard and Meylan, 1997). This approach represents a simplified method for estimating the mass transfer of model VOCs relative to O_2 and attention should be paid to more complex systems where external factors could influence the expected values.

4. Conclusions and recommendations

The results of the evaluation of (bio)surfactants addition showed a high mass transfer potential for both BTFs, i.e., expanded perlite and a mixture of C + WC, compared to other commonly used packing materials such as PUF. Different trends were observed depending on the packing material, the type of (bio)surfactant and its concentration. Overall, the perlite BTF exhibited higher k_LaO_2 values (308-612 h^{-1}) than the C + WC BTF (42-177 h^{-1}) regardless of the operating condition. In this BTF, an increase in the U_L from 0.95 to 1.53 m h^{-1} resulted in an enhanced mass transfer in case of no (bio)surfactant addition. The addition of saponin at 0.5 and 1 CMC had clearly positive effects on the perlite BTF, with the k_LaO_2 increasing up to 2 times compared to 0 CMC. However, saponin addition at 3 CMC resulted in foaming in the perlite BTF and at any CMC in the C + WC BTF, which was then translated into a decreased mass transfer. An increase in foaming was observed with increasing U_L. In contrast, the addition of Tween 80 at both U_Ls had a positive effect in the perlite BTF (e.g., 1.3 times higher k_LaO_2 at 1 CMC and 0.95 m h^{-1} compared to 0 CMC) and a neutral effect in the C + WC BTF. Although saponin decreased the surface tension of aqueous solutions to a greater extent, the CMC of each (bio)surfactant (Tween 80: 15.7 mg L^{-1} versus saponin: 1000 mg L^{-1}), together with the physical characteristics of the packing materials (e.g., higher surface area in the perlite BTF and non-uniform void spaces in the C + WC BTF) seemed to play an important role in the mass transfer experiments.

The results here obtained are of great importance to (i) improve the understanding of surfactants addition in BTFs as a strategy to enhance the removal of hydrophobic VOCs, and (ii) facilitate the selection of packing materials for future studies. Additionally, this fundamental data holds significant value for future modeling tools aiming for the design and optimization of bioreactors for air pollution control. Future studies should evaluate the effect of the presence of microorganisms in the trickling solution. For instance, nutrients added to the medium and substances excreted by the microorganisms affect the physical-chemical properties of the liquid solution, and could therefore have an impact on the mass transfer of pollutants. In this study, relatively high EBRTs and low U_Ls were selected to represent operational conditions that favor the removal of hydrophobic VOCs. Future studies could expand these conditions and evaluate the effect of both parameters on their mass transfer. In particular, lower EBRTs are important to reduce the footprint of future installations.

CRediT authorship contribution statement

Paula Alejandra Lamprea-Pineda: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Francisco Javier Carmona:** Writing – review & editing, Validation, Methodology. **Kristof Demeestere:** Writing – review & editing, Supervision, Methodology. **Jose Joaquin González-Cortés:** Writing – review & editing, Supervision, Methodology. **Herman Van Langenhove:** Writing – review & editing, Supervision, Methodology. **Christophe Walgraeve:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Raquel Lebrero:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Formal analysis, Conceptualization.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2024.121968.

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