



Toluene mass transfer characterization in a biotrickling filter

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ABSTRACT

Biotrickling filters (BTFs) devoted to air pollution control often present mass transfer limitations for hydrophobic volatile organic compounds (VOCs). Under such limiting conditions, BTF design and scale-up should be based on mass transfer data. A general and simple model was developed to characterize the VOC transfer by means of the overall mass transfer coefficient ($K_L a$). The $K_L a$ values were obtained by fitting the model to experimental data of toluene absorption obtained at empty bed residence times (EBRT) from 7 to 50 s. The model fitted well the experimental data ($r^2 = 0.97$) and the resulting $K_L a$ values ranged from 35 to 113 h^{-1} . These values are similar to those reported in the literature for BTF despite the lower liquid recycling velocity here used (0.6 m h^{-1}). A critical gas-to-liquid flow rate ratio (Q_G/Q_L) of 200, above which $K_L a$ was poorly increased, was observed. In addition, the individual film coefficients were estimated from the Van Krevelen and Hofstijzer correlations, which revealed that the main resistance for toluene mass transfer was in the liquid film regardless of the EBRT used. Finally, the high mass transfer potential of BTFs was confirmed by estimating the mass transfer capacity under varied operating conditions.

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1. Introduction

Volatile organic compounds (VOCs) are widely used in industrial processes and consequently, large amounts of these compounds are daily emitted to the atmosphere and wastewaters [1,2]. This entails a severe threat to both human health and natural ecosystems. Nowadays, the treatment of VOCs emitted by industries and waste treatment facilities is a key environmental issue due to the stricter environmental regulations [3]. In this context, biological systems for air pollution control are becoming increasingly popular for the removal of VOCs. Recent research has focused on biotrickling filters (BTFs) due to their key advantages over their biological counterparts: (i) effective treatment of acid-producing pollutants and (ii) lower pressure drop during long-term operation [4]. A high removal performance for hydrophilic VOCs such as ethanol and acetone has been recorded in BTFs [5]. However, mass transfer limitations may arise when treating moderately and highly hydrophobic VOCs. In this scenario, the design and scale-up of BTFs should be based on mass transfer data. Unfortunately, VOC mass transfer data in BTFs are scarce in the literature, research being mainly focused on biodegradation kinetics [6].

The mathematical modeling approaches used so far to characterize the mass transfer of hydrophobic VOCs are either complex

or can be applied only under particular experimental conditions. For instance, Pedersen and Arvin [7] developed a model to characterize the toluene mass transfer using an overall mass transfer coefficient ($K_L a$). However, the model was valid only for high recirculation velocities of the liquid phase. Later, Heymes et al. [8] developed a steady-state model to characterize the toluene mass transfer in a BTF operated with a viscous recycling liquid (di(2-ethylhexyl)adipate). Kim and Deshusses [9] recently developed a simple and robust method to characterize mass transfer in BTFs with several packing materials. Nonetheless, the mass transfer characterization required the addition of high amounts of NaOH in the recycling liquid phase (40 g L^{-1}). Such high salt concentration, much higher than that normally used in culture media, may lead to the overestimation of the mass transfer coefficients due to the high ionic strength established under these conditions [10]. Therefore, there is a lack of simple and general models to assess the VOC mass transfer in BTFs under normal operational conditions.

The aim of this work was to develop a simple and reliable model able to characterize the mass transfer in a BTF operated under typical VOC treatment conditions. The $K_L a$ value was obtained from experimental data using toluene as model VOC. Based on the agreement between the experimental $K_L a$ values and the Van Krevelen and Hofstijzer correlations, the individual gas and liquid film coefficients (k_G and k_L , respectively) were estimated. Finally, the application of the model in the design and optimization of BTFs was further discussed.

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Nomenclature

A_C	Cross sectional area of the packing column (m^2)
a_e	Effective specific interfacial area, defined as $a_p \varepsilon^{-1}$ (m^{-1})
a_p	Packing specific surface area (m^{-1})
C	Toluene concentration (g m^{-3})
D	VOC diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)
d_C	Column inner diameter (m)
g	Gravitational constant (m s^{-2})
H	Henry's law constant
k	Individual mass transfer coefficient (m h^{-1})
$K_L a$	Overall mass transfer coefficient (h^{-1})
$K_L^* a$	Theoretical overall mass transfer coefficient (h^{-1})
Q	Flow rate ($\text{m}^3 \text{h}^{-1}$)
u	Superficial velocity (m s^{-1})
V_C	Packed column volume (m^3)
V_T	Holding tank volume (m^3)

Greek letters

ρ	Density (kg m^{-3})
ε	Packed bed porosity
μ	Viscosity ($\text{kg m}^{-1} \text{s}^{-1}$)

Subscripts

G	Gas phase
L	Liquid phase
in	Packed column inlet
out	Packed column outlet

2. Materials and methods

2.1. Chemicals

Toluene (99.9% purity) was purchased from Sigma–Aldrich (Madrid, Spain). The liquid phase consisted of a sulphate-free mineral salt medium (MSM) prepared according to Lebrero et al. [11], which is often used as a culture medium. All chemicals for MSM preparation were purchased from PANREAC (Barcelona, Spain) with a purity of at least 99.0%.

2.2. Bioreactor set up

The mass transfer characterization was performed in a laboratory scale BTF at empty bed residence times (EBRTs) of 7, 11, 30 and 50 s. The BTF consisted of a cylindrical jacketed PVC column with a working packed bed volume of 4 L (0.08 m inner diameter, 1 m height). The BTF was packed with 1 cm^3 polyurethane-foam (PUF) cubes (Filtren TM 25280, Recticel Iberica S.L.) with a net density of 20–24 kg m^{-3} and a specific surface area of 1000 $\text{m}^2 \text{m}^{-3}$. The BTF was operated in countercurrent mode at a constant temperature of 20 °C and an inlet toluene concentration of $\sim 5 \text{ g m}^{-3}$. The liquid phase volume (1.4 L of MSM) was recycled at a velocity of 0.63 m h^{-1} (corresponding to a flow rate of $4.8 \times 10^{-3} \text{ m}^3 \text{h}^{-1}$) and continuously agitated at 200 rpm in an external 1.5-L holding tank. A detailed diagram of the experimental set-up is shown in Fig. 1.

2.3. Porosity of the packing material

The porosity of the packing material was experimentally determined by introducing a known mass of dry PUF in a test-tube. The

PUF cubes were then filled with water and the amount of water was determined by weight difference. The porosity can be expressed as:

$$\varepsilon = \frac{V_L - m_s / \rho_s}{V_L} \quad (1)$$

where V_L represents the water volume added, m_s is the mass of the packing material and ρ_s its density. The effective porosity for the wet packing material was determined using the same experimental procedure.

2.4. Overall mass transfer coefficient

A simple mathematical approach based on mass balances over the whole packed column was here proposed to characterize the toluene mass transfer in the BTF. The mass balance for the gas phase, which considers the overall mass transfer coefficient ($K_L a = H \times K_G a$), can be written as follows:

$$\frac{dC_{G,\text{out}}}{dt} = \frac{Q_G}{V_C} (C_{G,\text{in}} - C_{G,\text{out}}) - K_G a (C_{G,\text{out}} - H C_{L,\text{out}}) \quad (2)$$

where H represents the Henry's law constant, $C_{G,\text{out}}$ is the VOC concentration at the outlet of the packed column and $C_{G,\text{in}}$ is the VOC concentration at the inlet of the column (Fig. 1). On the other hand, the mass balance for the liquid phase at the outlet of the BTF is as follows:

$$\frac{dC_{L,\text{out}}}{dt} = K_L a \left(\frac{C_{G,\text{out}}}{H} - C_{L,\text{out}} \right) + \left(\frac{Q_L}{V_C} \right) C_{L,\text{in}} \quad (3)$$

A mass balance over the holding tank is also necessary to describe the toluene concentration in the liquid phase at the inlet of the BTF. This balance must consider that the liquid phase is first diluted in the holding tank volume (V_T) and then recycled at a constant rate (Q_L). Thus, the mass balance for the VOC in the liquid phase at the inlet of the BTF can be expressed as:

$$\frac{dC_{L,\text{in}}}{dt} = \frac{Q_L}{V_T} (C_{L,\text{out}} - C_{L,\text{in}}) \quad (4)$$

The model represented by Eqs. (2)–(4) is a simplified description of the system and allows for the characterization of the mass transfer performance in BTFs considering the following assumptions: (i) the liquid phase is perfectly mixed and (ii) conditions are isothermal throughout the experimental system (reactor and holding tank), so H remains constant during all experiments. In order to obtain the overall mass transfer coefficient from experimental data, the kinetics of toluene saturation in the recycling MSM were assessed in duplicate at each EBRT. In brief, $C_{G,\text{in}}$ and $C_{G,\text{out}}$ were continuously monitored until liquid phase saturation was reached ($C_{G,\text{out}}/C_{G,\text{in}} \geq 0.97$). At the same time, liquid samples (5 mL) from the inlet and outlet of the packed column were periodically drawn and injected into 10.5 mL gas-tight serum bottles (closed with butyl septa and sealed with aluminum caps). Toluene in the gas-tight bottles was allowed to equilibrate and then analyzed in the headspace to determine the corresponding values of $C_{L,\text{in}}$ and $C_{L,\text{out}}$ by means of the Henry's law (liquid concentrations at equilibrium).

The experimental measurements of C_G and C_L were used to determine the mass transfer coefficients by solving simultaneously Eqs. (2)–(4), $K_L a$ being the fitting parameter. The 4th order Runge–Kutta method was used to solve the differential equations, whereas the Levenberg–Marquardt method was used for parameter fitting according to Jia et al. [12]. ModelMaker[®] software (Cherwell Scientific, UK) was used to solve the model.

2.5. Analytical methods

Toluene gas concentration was determined in a GC-FID (HP 6890 Series, Hewlett Packard, USA) equipped with a SupelcoWax

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