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Styrene vapor mass transfer in a biotrickling filter: Effects of silicone oil volume fraction, gas-to-liquid flow ratio, and operating temperature

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HIGHLIGHTS

- Effects of oil, flow ratio, and temperature on mass transfer in a BTF were studied.
- Full-factorial experimental design was used to study the interactions.
- Oil fraction and flow ratio were significant factors affecting mass transfer.
- Addition of more than 10% oil had just a slight effect on the mass transfer.
- Mesophilic/thermophilic conditions did not have main effect on mass transfer in BTF.

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ABSTRACT

The addition of silicone oil as a non-aqueous phase (NAP) to the trickling liquid enhances the mass transfer of hydrophobic compounds from gas to liquid phase in biotrickling filters (BTFs). The present study investigated the effect of NAP volume fraction (ϕ; 0–20%), gas-to-liquid flow ratio (7.5, 16 and 32), and operating temperature (30 and 50 °C) on styrene mass transfer in a BTF under abiotic conditions using a full-factorial experimental design. The BTF was packed with a mixture of pumice and steel pall rings. The inlet styrene concentration was 0.5–1.2 g m^{-3} and EBRTs were set at 1 and 2 min. The results were incorporated into a model to calculate the maximum fraction of styrene transferable from the gas to the aqueous phase (β_S^*). The β_S^* was determined to be 0.208–1, depending on the operating conditions. Flow ratio and ϕ were the most significant factors affecting styrene mass transfer (p-value = 0.0348 and p-value = 0.0230, respectively). Temperature had no significant effect on the model (p-value = 0.9322) at a 95% confidence interval. Styrene mass transfer was very sensitive to ϕ when $\phi < 10\%$; at $\phi > 10\%$ the addition of silicone oil had a slight effect on β_S^*.

1. Introduction

Styrene (vapor pressure of 0.6 kPa at 20 °C) is an important volatile organic compound (VOC) emitted from process industries such as paint and plastics factories and refineries. It is volatile and toxic and can cause serious problems for the environment and human health. Chronic exposure to styrene can cause headache, weakness, hearing loss and central nervous system dysfunction as well as gastrointestinal disorders [1–3]. Of the existing waste–gas biological treatment technologies, biotrickling filters (BTFs) offer ease of temperature control, pH control, and lower pressure drop along the reactor because of the presence of a mobile liquid phase. The continuous circulation of a mobile liquid phase in BTFs enables easier control of biological operating parameters (nutrients, pH and temperature) than in biofilters (BFs). In other words, these parameters can be controlled by easily adding acids, bases, and fresh medium to the trickling liquid in desired temperature to maintain optimum conditions for pollutant removal [4]. Because of simultaneous biodegradation and absorption of pollutants, the solubility specifications in BTFs are less drastic than for bioscrubbers [5]. In addition, BTFs represent lower pressure drop along the bed height as the packing used in BTFs are made from inert materials with higher porosity, such as: polyurethane foam, resins, pall rings and lava rock than that used in BFs (compost and soils). BTFs show promise for removal of gas-phase styrene and other VOCs in comparison to different physico-chemical techniques [2,6,7].

Although BTFs have a good history of removal of hydrophilic VOCs such as ethanol, limitations on mass transfer exist for treatment of hydrophobic compounds (hexane, styrene, toluene) because of their low solubility in water. Consequently, very high

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Nomenclature

a	reactor specific interfacial area (m^2/m^3)
a_p	packing specific surface area (m^2/m^3)
$C_{G,in}$	VOC concentration in the gas phase at the inlet of the reactor (kg/m^3)
$C_{G,out}$	VOC concentration in the gas phase at the outlet of the reactor (kg/m^3)
C_W	bulk VOC concentration in the aqueous phase (kg/m^3)
d_p	particle diameter (m)
D_L	diffusion coefficient of VOC in liquid (m^2/s)
EBRT	empty bed residence time (s)
EC	elimination capacity (kg/m^3 s)
g	gravitational constant (m/s^2)
H_{ef}	effective Henry constant for the VOC in the aqueous phase (dimensionless) (–)
H_{mix}	Henry constant for the VOC in the mixture (dimensionless) (–)
H_{NAP}	Henry constant for the VOC in the NAP (dimensionless) (–)
H_w	Henry constant for the VOC in the NaCl solution (dimensionless) (–)
ILR	initial loading rate (kg/m^3 s)
k_{LNAP}	individual VOC mass transfer coefficient at the NAP through the gas-NAP interface (m/s)
k_{Lw}	individual VOC mass transfer coefficient at the water phase through the gas–water interface (m/s)
K_La	volumetric mass transfer coefficient (1/s)
Q_G	gas flow rate (m^3/s)
Q_L	total liquid flow rate in the BTF (m^3/s)
R	volumetric VOC transfer rate (kg/m^3 s)
RE	removal efficiency (%)
Sc_L	Schmidt number for liquid defined by $\mu/\rho D$ (–)
t	time (s)
T	temperature (°C)
u_L	liquid superficial velocity (m/s)
V_c	packing volume (m^3)
β_S^*	maximum VOC fraction transferred from the gas to the aqueous phase including the VOC transferred through the organic phase (dimensionless) (–)
ϕ	volumetric fraction of the organic phase (%)
η	lumped parameter with units of an individual mass transfer coefficient (m/s)
ρ_L	density of liquid (kg/m^3)
μ_L	viscosity of liquid (kg/m s)

elimination capacities (ECs) are not common in BTFs [2]. Under such circumstances, the addition of an organic non-aqueous phase (NAP) to the aqueous phase containing pollutant-degrading microorganisms increases the affinity for dissolving hydrophobic VOCs in the aqueous phase.

The NAP can act as a reservoir to control delivery of VOCs to microorganisms in the aqueous phase [6,8]. The organic solvent should be water-immiscible, non-biodegradable and non-toxic for biocatalysts. Silicone oil has been reported to be an efficient organic solvent from the perspective of chemical engineering and thermodynamics in laboratory-scale two-liquid phase systems [6,8,9]. A recent study showed that solvent concentration affects VOC mass transfer and biodegradation rates; it increases mass transfer interfacial area, bioavailability of VOC, and consequently, the activity of microorganisms [10]. Moreover, The partition coefficient of the liquid mixture (H_{mix}) as a function of organic solvent volume fraction has been reported to have a significant influence on the overall mass transfer coefficient (K_La) [3,11]. Therefore, consideration of mass transfer data in BTF design is critical. In real BTFs, wherein VOC biodegradation by microbial population occurs, it is difficult to study mass transfer. Thus recent research has focused on the role of mass transfer in VOC removal by BTFs under abiotic conditions [12,13].

Gas flow rate (Q_G) can influence the transfer of VOC between phases by changing the contact time between them [5]. The trickling liquid flow rate (Q_L) is also reported to have a significant effect on the mass transfer coefficient [14]. The ratio Q_G/Q_L (flow ratio) can be applied to consider the effects of both parameters on mass transfer. Lebrero et al. found a linear increase for K_La with an increase in flow ratio up to a critical ratio of 200 [12].

Operating temperature also affects biofiltration by affecting the VOC diffusion coefficient, VOC absorption, and the kinetics of biodegradation [15]. Studies have suggested the use of thermophilic (45 or 50 °C), rather than mesophilic (20 or 25 °C), biodegradation of highly volatile hydrophobic pollutants [6,16,17]. An increase in temperature increases volatility of the VOC, which could restrict diffusion of the VOC into the liquid or biofilm phases.

Assessment of the net influence of operating temperature on the mass transfer of the process is complex. The present work aims to study the effect of silicone oil (NAP) concentration on the performance of styrene removal in single- and two-phase partitioning BTFs at mesophilic (30 °C) and thermophilic (50 °C) operating temperatures under abiotic conditions. The effect of gas-to-liquid flow ratio on mass transfer was also investigated. A mathematical model was used to obtain a parameter defining the maximum fraction of styrene transferred from the polluted gas stream to the liquid mixture (β_S^*) to evaluate the mass transfer capacity of the BTF. Probable interactions between operating temperature, silicone oil volume fraction, and flow ratio were investigated using a full factorial approach.

2. Materials and methods

2.1. Theoretical background

Hernandez et al. developed a model framework based on general mass balance and transfer equations that represents an isomorphous expression for all configurations [14] as:

$$\ln\left(1-\frac{C_{G,out}}{C_{G,in}}\right)=\ln\beta_s^*-\left(\frac{Q_G}{V_C}H_{mix}\beta_s^*\right)t \tag{1}$$

In Eq. (1), β_S^* is the maximum fraction of VOC transferable from the gas to the liquid phase and can be determined using Eqs. (2) and (3) in the absence and presence of NAP, respectively [14]:

$$\beta_s^*=\frac{1-\exp\left(-(k_{Lw}aV_C/H_wQ_G)\left(1+H_w\left(\frac{Q_G}{Q_L}\right)\right)\right)}{1-H_w\left(\frac{Q_G}{Q_L}\right)\exp\left(-(k_{Lw}aV_C/H_wQ_G)\left(1+H_w\left(\frac{Q_G}{Q_L}\right)\right)\right)} \tag{2}$$

$$\beta_s^*=\frac{1-\exp\left(-\eta(aV_C/Q_G)\left(1+H_{mix}\left(\frac{Q_G}{Q_L}\right)\right)\right)}{1-H_{mix}\left(\frac{Q_G}{Q_L}\right)\exp\left(-\eta(aV_C/Q_G)\left(1+H_{mix}\left(\frac{Q_G}{Q_L}\right)\right)\right)} \tag{3}$$