

Paint solvent components degradation from their mixture along the biotrickling bed height

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ABSTRACT. A biotrickling filter was used for the treatment of waste gas. Pollutants tested included aromatic hydrocarbons – toluene, xylenes, oxygenated compounds - methyl ethyl ketone, methyl isobutyl ketone and ester - n-butyl acetate. A defined mixed culture containing four primary aromatic hydrocarbon degraders was applied as the biocatalyst and immobilized on the packing material.

After an acclimation phase - one week (Paca *et al.*, 2004), the loading tests were carried out. In the first phase of the experiments the biotrickling filter was loaded by increasing the inlet concentration of aromatic hydrocarbons in the pollutant mixture from 6 g_C.m³.h⁻¹ to 16 g_C.m³.h⁻¹. It resulted in a decrease of the degradation efficiency of toluene and xylenes to 60%. Almost no effect on the removal efficiency of hydrophilic compounds was observed. Secondly, the effect of an increasing amount of hydrophilic compounds in the mixture of all solvents on the degradation efficiency was investigated. The loading by ketones and n-butyl acetate caused not only a quite rapid drop of RE_{AROM}, but also a slight decrease of RE_{KET}.

1 INTRODUCTION

Vapor-phase bioreactors (VPBs) have drawn increased interest for the treatment of volatile organic compound (VOC) mixtures such as those emitted from painting operations. The paint solvents contain both hydrophilic and hydrophobic compounds.

Some researchers have investigated the degradation of hydrophobic or hydrophilic compounds from waste air in a biotrickling filter (Kim *et al.*, 2004a; Porter *et al.*, 2004; Qi *et al.*, 2002; Kim *et al.*, 2004b; Delhoménie *et al.*, 2001; Cox *et al.*, 2002). However, only recently have studies been initiated concerning the degradation of hydrophobic and hydrophilic compound mixtures using a biotrickling filter (Paca *et al.*, 2004; Song *et al.*, 2002a; Song *et al.*, 2002b; Park and Kinney, 2002).

The objectives of this study were: (1) To evaluate the effect of increasing the content of hydrophobic pollutants in the mixture on biotrickling filter performance, (2) To evaluate system performance while increasing the content of hydrophilic pollutant in the mixture.

2 MATERIALS AND METHODS

A schematic diagram of the bench-scale biotrickling filter is shown in Figure 1. The bioreactor was constructed of three cylindrical units. The two upper units were made of glass and the sump was made of polypropylene. The height of the reactor was 1.7 m and the internal diameter was 0.15 m. The sump was separated from the column by a perforated plate. The packing material consisted of Pall rings made of hydrophilized polypropylene. The column was packed with Pall rings having the following characteristics: 15 x 15 x 1 mm, void volume of 0.8624, specific surface area of 350 m².m⁻³ and a bulk density of 120 kg.m⁻³. The packed bed height was 1 m.

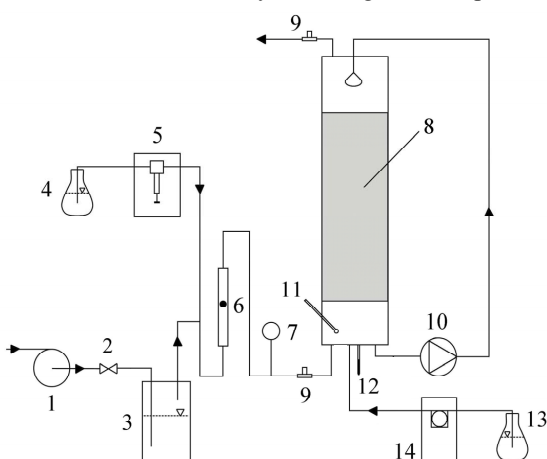


Figure 1. Schematic of the experimental set-up

1- blower, 2 - needle valve for air-flow rate control, 3 - humidifier, 4 - vessel with pollutant, 5 - syringe pump, 6 - rotameter, 7 - manometer, 8 - packing, 9 - sampling ports, 10 - membrane pump, 11 - pH electrode, 12 - thermometer, 13 - NaOH solution, 14 - peristaltic pump.

The mixed microbial culture used to inoculate the biotrickling filter included the following bacterial strains: *Sphingobacterium multivorum* (G⁻ rods), *Comamonas testosteroni* (G⁻ rods), *Pseudomonas putida* (G⁻ rods) and *Chryseobacterium indologenes* (G⁻ rods). The medium composition for cultivation consisted of (g.L⁻¹): K₂HPO₄ 4.3; KH₂PO₄ 3.4; KNO₃ 2; MgCl₂ · 6 H₂O 0.34; FeSO₄ 6.10⁻⁴; Na₂MoO₄ 1.10⁻⁵.

Key biodegradation experimental conditions included: temperature of 22°C, a mineral medium (MM) for growth which contained 0.75 g/L (NH₄)₂SO₄, 0.37 g/L KNO₃, 0.1 g/L NaCl, 0.8 g/L

K₂HPO₄, 0.6 g/L KH₂PO₄, 0.34 g/L MgCl₂.6H₂O, and 1 mL/L trace elements. The MM was present during the first month of operation. After this time period, tap water was used during the following months of experiments and the MM was added in small quantities when necessary. The pH of the circulating water phase was maintained at 7.0. The hydraulic loading rate was 0.0064 m.h⁻¹.

The synthetic waste gas influent included toluene, xylene (technical grade), methyl ethyl ketone (MEK), methyl isobutyl ketone (MIBK), and n-butyl acetate (n-BA). The VOCs in the gas phase were measured using an Agilent 6890 N gas chromatograph equipped with an ultra alloy-5 (5 % phenylmethylsilicone) capillary column of 30 m in length, the inner diameter of 0.53 mm, and film thickness of 1.5 μm (Quadrex Corp., UA5 - 30V - 1.5 F, New Haven, CT). The carrier gas used was ultra high purity argon at a flow rate of 5 mL.min⁻¹. Detection was accomplished with a flame ionization detector (FID) with ultra high purity hydrogen and breathing quality air at flow rates of 28 mL.min⁻¹ and 315 mL.min⁻¹, respectively. GC operating conditions were as follows: column temperature, 80 °C; FID temperature, 250 °C.

3 RESULTS AND DISCUSSION

3.1 Effects of increasing hydrophobic compound concentrations in the influent mixture

Figure 2 shows the organic loading (OL) and removal efficiency (RE) of the bioreactor. This experiment was carried out using tap water as the circulating medium. The biotrickling filter was loaded simultaneously with all the pollutants listed above. On day 64 the toluene concentration was increased from 50 mg.m^{-3} to 70 mg.m^{-3} in one step. The second change of the organic load was carried out on day 67 by increasing the inlet

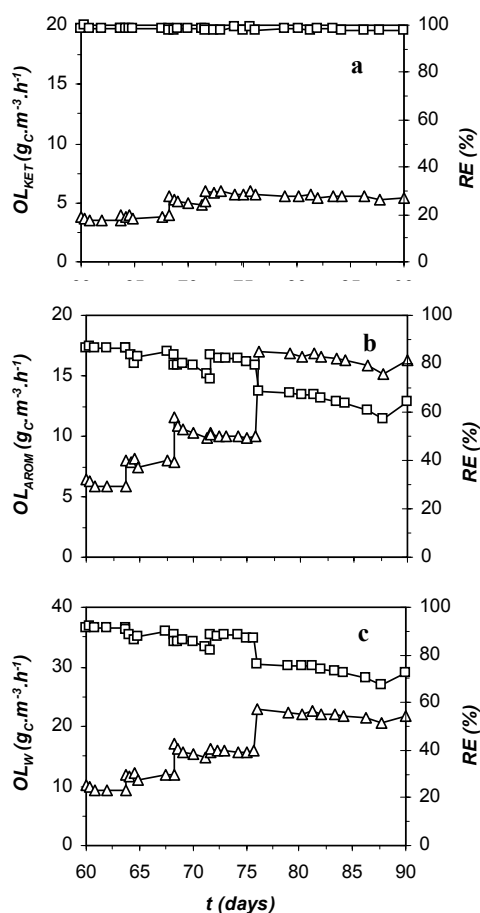


Figure 2. Loading by increasing inlet concentration of hydrophobic compounds in the pollutant mixture.

a – Values for hydrophilic compounds,
 b – Values for hydrophobic compounds,
 c – Overall values,
 Δ - OL, \square - RE

concentration (C_{in}) of both the aromatic compounds (toluene from 70 mg.m^{-3} to 80 mg.m^{-3} , xylenes from 50 mg.m^{-3} to 80 mg.m^{-3}), and the third increase on day 76 (toluene from 70 mg.m^{-3} to 110 mg.m^{-3} , xylenes from 80 mg.m^{-3} to 100 mg.m^{-3}). With these adjustments, the value of the organic loading of aromatics (OL_{AROM}) was increased from $6 \text{ g}_C.\text{m}^{-3}.\text{h}^{-1}$ to $16 \text{ g}_C.\text{m}^{-3}.\text{h}^{-1}$.

Increasing the content of the hydrophobic compounds in the mixture resulted in a drop of the removal efficiency of hydrophobic compounds from 90% to 60% (Fig. 2b). However, the value of RE_{KET} remained constant.

3.2 Effects of increasing hydrophilic compound concentrations in the influent mixture

In this phase of the experimental work, the biotrickling filter was loaded with a mixture of VOC solvents with gradually increasing inlet concentrations of the hydrophilic compounds. Prior to this experiment, salts for 3L of MM were added in order to improve the degradation efficiency of toluene and xylenes. The salts were added on the 355th day of operation, and within 24 hours after addition the total degradation efficiency of 85% was restored.

The inlet concentration of hydrophilic compounds was increased in two steps, from $5 \text{ g}_C.\text{m}^{-3}.\text{h}^{-1}$ to $9 \text{ g}_C.\text{m}^{-3}.\text{h}^{-1}$ and subsequently to $14 \text{ g}_C.\text{m}^{-3}.\text{h}^{-1}$. Simultaneously, the concentration of influent aromatic compounds was kept constant.

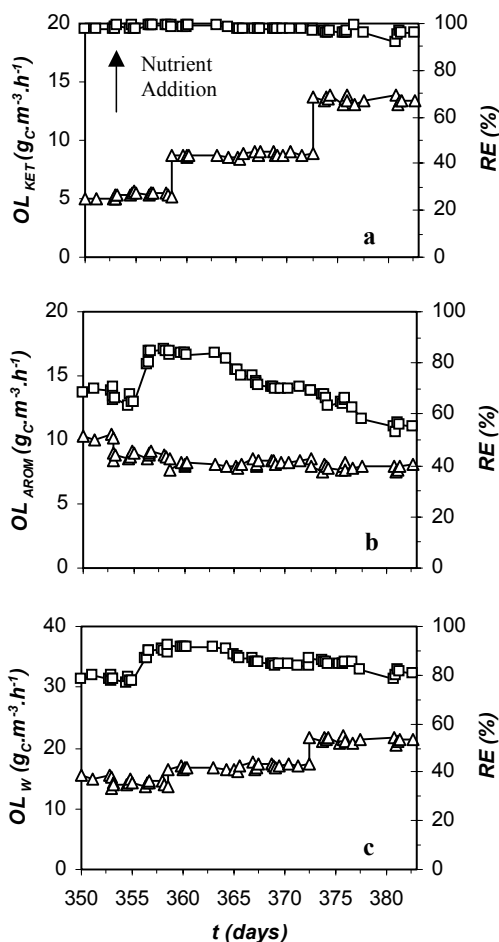


Figure 3. Loading by increasing inlet concentration of hydrophilic compounds in the pollutant mixture.

a – Values for hydrophilic compounds,
 b – Values for hydrophobic compounds,
 c – Overall values,
 Δ - OL, \square - RE

types of the pollutants (hydrophilic and hydrophobic), unless the reactor is overloaded.

The results of this experiment are shown in Figure 3. After the first increase, the degradation efficiency of the aromatic compounds decreased to 70%, while the effect on the degradation ability of ketones was minimal. The final value after the second loading increase was $RE_{AROM} = 55\%$, while RE_{KET} decreased only slightly, approximately to 96%. The slight RE_{KET} decrease was probably a consequence of the mass transfer (desorption) from the water droplets sprayed into the counter current air flow at the reactor head.

4 CONCLUSIONS

The results of this study suggest that the increasing concentrations of toluene and xylenes in the influent solvent mixture during the loading experiments had no effect on the degradation efficiency of hydrophilic compounds, but caused a significant drop in the removal efficiency of aromatic compounds. For the case of increasing concentrations of hydrophilic compounds in the mixture, the degradation efficiency of aromatic compounds was strongly suppressed.

The biotrickling filter system is able to completely remove both

5 ACKNOWLEDGEMENT

This study was financially supported by the Czech Science Foundation, Joint Project 104/05/0194 and the Ministry of Education of the Czech Republic, Research Project MSM 6046137305.

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