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Determination of local elimination capacities and moisture contents in different biofilters treating toluene and xylene

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Abstract

The removal of toluene and xylene from an artificial waste gas was investigated in two laboratory scale biofilters filled with mixtures of peat, bark and wood. The packed beds differed in the mixture of materials used, so that peat and then bark was the dominant constituent. The biofilters were operated in an upflow mode. Both biofilters showed relatively high removal efficiencies for both pollutants (74–98%). The evaluation of the local elimination capacities in the peat-loaded biofilter revealed that the major part of pollutants was degraded in the middle layer. In this biofilter, larger differences in the removal rates along the bed height were also observed. In the biofilter with bark as dominant material, the major part of pollutants was degraded at the inlet of the bed and also at a relative height of 0.7. Moisture contents of 71–80% and 65–78% were found for the biofilter with peat and bark respectively. When the regular pouring of nutrient solution through the bed was interrupted for 1 month, a decrease in efficiency was observed in the biofilter with bark, whilst the efficiency in the biofilter with peat remained the same.

Abbreviations: Cout/Cin – dimensionless concentration (out = outlet, in = inlet); RE – removal efficiency; EC – elimination capacity; h/H – dimensionless height of the packed bed; OL – organic load; OL_C – organic load calculated as total organic carbon; Ug – superficial gas velocity; VOCs – volatile organic compounds

Introduction

Toluene and xylene belong to the top 50 chemicals manufactured each year (Manahan 1994). They are extensively used as industrial solvents and synthetic intermediates and are significant constituents of fuels. Toluene is included in the list of priority pollutants selected by Environmental Protection Agency (Keith & Telliard 1979). European emissions in 1990 were calculated to be 1.65 million tonnes (Engesser et at. 1996). Treatment of toluene, xylene and other hydrocarbons by biofiltration represents an attractive, economically feasible method, which can be an advantageous alternative to adsorption, catalytic oxidation and incineration, if the pollutants are present in relatively low concentrations (up to 1–2 g m^{-3}).

Initially, biofiltration was developed to treat odours, but recently biofiltration has made great progress and its application extended to the removal of various volatile organic compounds (VOCs). Research in this field is focused on the improvement of process design and the optimisation of biofilter performance. The topics include: the search for optimum packing materials providing good wetting characteristics and preventing high pressure drops (Shareefdeen et al. 1993; Smet et at. 1996; Sabo et al. 1996; Oude Luttighuis 1997), the development of appropriate mathematical models and their experimental validation (Ottengraf 1986; Deshusses et al. 1995a, b; Shareefdeen & Baltzis 1994; Ergas et al. 1994; Zilli et al. 1996; Cox et at. 1997) and the investigation of microbiological aspects of biofiltration, i.e. isolation

and characterisation of the degrading microflora (Mallakin & Ward 1996; Andreoni et al. 1997; Lipski et al. 1997a, b; Reichert et al. 1997), the effect of inoculum (Ottengraf et al. 1986; Mallakin & Ward 1996; Smet et al. 1996; Smet et al. 1997), application of special strains and types of microorganisms (Zilli et al. 1993; Cox et al. 1997) and the effect of external conditions on the microbial activity (Weckhuysen et al. 1993; Kennes et al. 1995).

The important characteristics of biofilters, evaluated in this work, are the concentration profiles of individual compounds throughout the depth of the packed bed. They allow to find more detail about the character of the degradation, to evaluate the local elimination capacities and, together with the other performance characteristics, help to better understand the process. This work was carried out to investigate the long term performance stability and concentration profiles of toluene and xylene using two biofilters filled with different packing materials.

Materials and methods

Biofilters

The experimental apparatus consisted of two laboratory scale biofilters. Tubular packed bed glass reactors with an inner diameter of 50 mm and a bed height of 280 mm were used. As packing materials, mixtures of peat, bark and wood in ratios 7 : 1 : 2 and 1 : 7 : 2 (weight) were used in biofilters 1 and 2 respectively. The packing material was kept sufficiently moist by humidifying the inlet air and by pouring 0.1 L of nutrient solution on the top of the filter bed once a week. The pH of the eluent varied (mainly in the range 6.0–6.5). The humidified air was contaminated with toluene and xylene (mixed isomers) and fed to the biofilters in an upflow mode (each biofilter received gas with the same pollutant concentrations). If not specified the volumetric flow rate was kept at 30 L h^{-1} (i.e. superficial gas velocity of 15.3 m h^{-1}). The experiments were performed at the room temperature (21–23 °C).

Inoculum and media

The biofilters were inoculated with a mixed microbial population resulting from the adaptation of soil microbiota to benzene, toluene and xylene (Páca & Koutský 1994; Páca 1996). The inoculum was prepared growing in Erlenmeyer flasks at 30 °C using mineral medium and oil vapour as a carbon source. After a 5-day cultivation on a rotary shaker, the cell suspension was used to saturate the packing material. The culture was maintained on the same mineral medium with agarose, exposed to oil vapour and stored at 4 °C in a refrigerator.

The composition of the mineral medium was (in g L^{-1}): K_2HPO_4, 4.3; KH_2PO_4, 3.4; $(NH_4)_2SO_4$, 2.0; $MgCl_2$, 0.34; $MnCl_2$, 0.7×10^{-3}; $CaCl_2$, 1.3×10^{-3}; $FeSO_4$, 0.6×10^{-3}; Na_2MoO_4, 1.7×10^{-3}. The nutrient solution that was applied each week (to supply cells with water and nutrients) contained (in g L^{-1}): K_2HPO_4, 0.17; KH_2PO_4, 0.13; $(NH_4)_2SO_4$, 0.71; $MgCl_2$, 0.16; $MnCl_2$, 0.7×10^{-3}; $CaCl_2$, 1.3×10^{-3}; $FeSO_4$, 0.6×10^{-3}; Na_2MoO_4, 1.7×10^{-3}.

Analytical methods

The concentrations of toluene and xylene were determined by gas chromatography. A CHROM 51 gas chromatograph (Lab. pristroje Praha, Czech Republic) was equipped with a packed column filled with Resorb BLK (Lachema, Czech Republic) containing 10% of polyethylenglycoladipate. Argon was used as a carrier gas and pollutants were detected using the flame ionisation detector (the concentration of xylene was evaluated as the total concentration of all isomers). Injection, separation and detection temperatures were all 150 °C. The chromatographs were calibrated using carbon disulphide solutions of xylene and toluene.

Experimental setup

The experiments were carried out over a period of fourteen months in the course of which the effects of various types of loadings and dynamic behaviour of biofilters were examined (results are not included in this paper). During the 8th month, the biofilters were tested to find out the concentration and moisture content profiles along the bed height. Since the biofilters were not fitted with sampling ports, the experiment was arranged as follows: the inlet concentrations of toluene and xylene were kept constant (180 mg m^{-3} of each) and the outlet concentrations were measured step by step after removing the upper part of the packed bed. The water content of packing materials at various bed heights were determined gravimetrically taking the average samples from each bed height (triplicate experiments). After the experiment, the biofilters were refilled with the same packing materials.

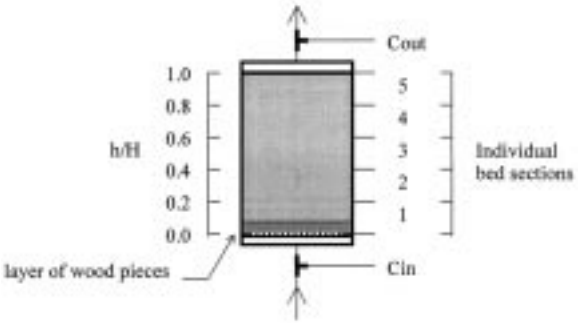

Figure 1. Schematic representation of the experimental setup.

Results and discussion

Acclimation period and history of biofilters

The biofilters were acclimated by increasing the total carbon concentration gradually from 20 to 150 mg m^{-3} (the latter value corresponds to the 12th day of operation). The volumetric flow rate was kept constant at 60 L h^{-1} (superficial gas velocity of 30.6 m h^{-1}). The performance of biofilters over the initial period of 20 days is illustrated in Figure 2. Xylene was applied as the only pollutant in the first part of the acclimation period. On the 3rd day after inoculation, the removal efficiency reached 90% and remained (with slight variations) at the same value after the addition of toluene on the 7th day. Toluene removal efficiencies of around 90% were achieved on the 11th and 12th days using both biofilters. The initial toluene removal effciency of 100% observed in biofilter 2 was probably a consequence of adsorption on the packing material that lasted a longer time than in biofilter 1. Deshusses et al. (1995c) and Morales et al. (1998) have reported the initial adsorption phase of 8 hours and less than 1 hour respectively.

During the experiments, both the removal efficiency and elimination capacity varied depending on the organic load and the manner of loading. Within the 4th–6th month, the biofilters were examined to find the effect of high loading rates, step inlet concentration changes and starvation periods (results not shown). For most of the operation time (except the above experiments) the biofilters were operated at pollutant concentrations not exceeding 200–300 mg m^{-3} of each VOC. Figure 3 shows the comparison of the removal efficiencies in the 1st, 4th and 8th month of operation. The efficient degradation of toluene and xylene remained up to the 8th month, when the removal efficiencies still ranged between 74 and 98%.

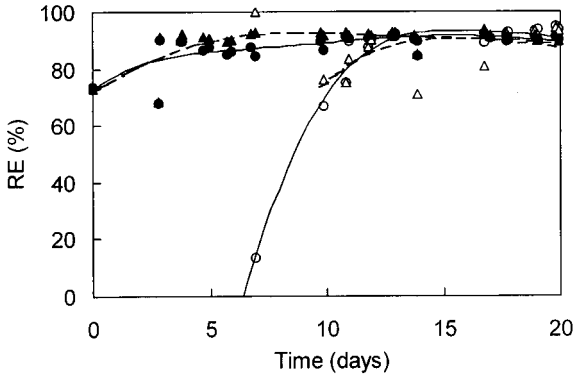

Figure 2. Removal efficiencies during the acclimation period. (Superficial gas velocity: 30.6 m h^{-1}. OL_C ranged from 2 to 22 g m^{-3} h−1; biofilter 1 (solid line, ○, ●), biofilter 2 (dashed line, △, ▲), toluene (○, △), xylene (●), ▲).

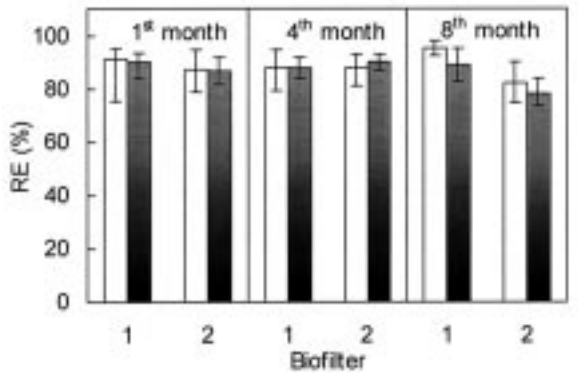

Figure 3. Comparison of degradation efficiencies during 1st (after the acclimation period), 4th and 8th months. OL_C was kept at 14–31 g m^{-3} h^{-1}; toluene (□), xylene (■).

Concentration profiles along the bed

Figure 4 shows the gaseous concentration profiles along the biofilter bed investigated in the 8th month of operation. The contours indicate the simultaneous biodegradation of toluene and xylene. In both biofilters, the decrease of pollutant concentrations was irregular. This irregularity was observed especially in the biofilter 1. The characteristics of the profiles obtained by most authors (Ottengraf & Van den Oever 1983; Ottengraf 1986; Shareefdeen et al. 1993; Ergas et al. 1994; Kennes et al. 1996; Zilli et al. 1996; Cox et al. 1997) showed the regular decrease of concentrations or the decrease with only small irregularities as it was seen in biofilter 2. Some of these authors observed exponential (Ergas et al. 1994; Zilli et al. 1996), linear (Ottengraf & Van den Oever 1983; Cox et al. 1997) or quadratic (Ottengraf & Van den Oever 1983) concentration profiles and used the results to

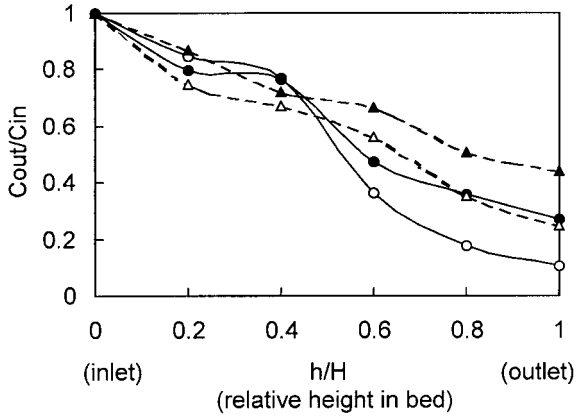

Figure 4. Concentration profiles along the bed height. (Inlet concentration of both pollutants: 180 mg m^{-3} each. Superficial gas velocity: 15.3 m h^{-1}. OL_C = 19.2 g m^{-3} h^{-1}); biofilter 1 (solid line, ○, ●), biofilter 2 (dashed line, △, ▲), toluene (○, △), xylene (●, ▲).

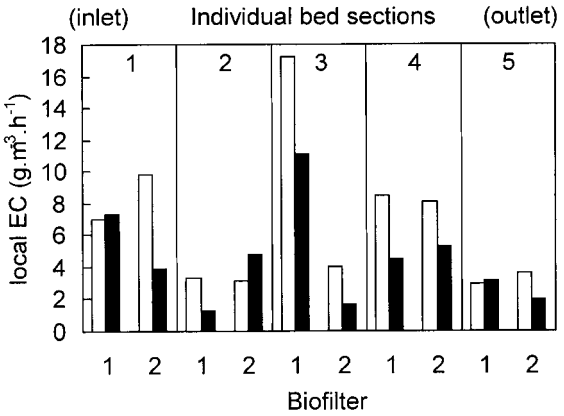

Figure 5. Local capacities of individual sections of the bed. Bed heights are numbered from bottom to top. Each section represents 20% of the total height. Toluene (□), xylene (■); the other parameters are the same as in Figure 3.

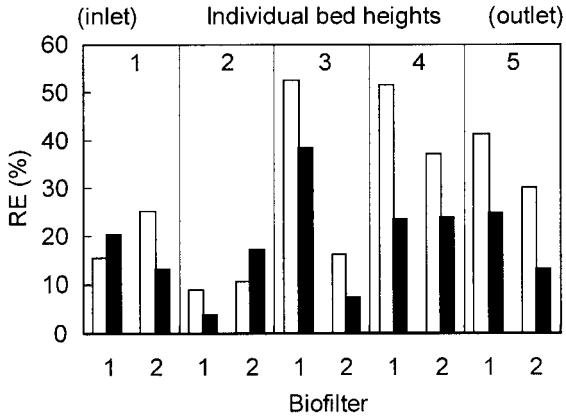

Figure 6. Local removal efficiencies of individual sections of the bed. The legend is the same as in Figure 5.

validate the biofilter model proposed by Ottengraf (Ottengraf & Van den Oever 1983; Ottengraf 1986). Thus, the authors assumed the degradation kinetics to be first-order, zero-order with reaction rate limitation and zero-order with diffusion limitation respectively. The results similar to those found in biofilter 1 (irregular decrease of concentration) were described by Deshusses and Hamer (1993). The differences in removal rates are illustrated in Figure 5 where the local elimination capacities of individual sections of the biofilter are calculated. In Figure 6 the results are presented in the form of local removal efficiencies. Unlike the scale of h/H in Figure 4, the description of axes in Figures 5 and 6 were chosen according to Figure 1 to characterize the degradation effect of the individual sections of the bed's total height.

As expected, Figures 5 and 6 have similar shapes, but the removal efficiencies observed for the 4th and 5th sections of the bed were higher in comparison to the elimination capacities (Figure 5). This was because the pollutant concentrations entering these parts were lower than those entering the lower parts of the bed (Figure 4). Hence, even though the elimination capacities were the same or even lower than in the sections 1–3 the efficiency was relatively high. In biofilter 1, the major part of pollutants was removed in the middle layer (35% of all carbon), whereas in biofilter 2, layer 1 closely followed by layer 4 showed the removal 19% of all carbon. The lower removal rates in the lower parts of biofilter 1 may be explained by a local channelling effect (especially in the layer 2). The high moisture content (cf. h/H = 0.1 and 0.3 in Figure 7) and the wash out of small particles from the upper parts of the bed probably resulted in changes of local packing homogeneity. Despite this small particle accumulation in the layer 2, the pressure drop increased from 15 mm to 17 mm of water only. Nevertheless, the lowermost parts of biofilters (section 1 in Figure 5) showed, in comparison with the others, relatively high elimination capacities. This could be due to the slightly higher presence of wood pieces (laid out close to the perforated plate in order to improve the air distribution cf. Figure 1). Cross laid wood pieces could avoid the channelling at the bottom of these parts of biofilters. The decrease of elimination capacities at the top of the biofilter was accompanied by decrease of removal efficiencies. Thus, the removal rates of the upper parts of the bed could be limited by the concentration of pollutants (with the inlet concentrations of 180 mg m^{-3} of

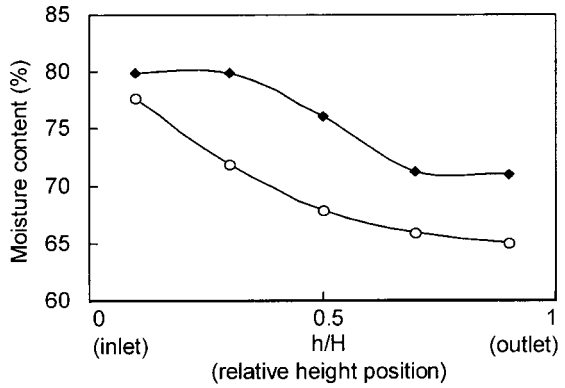

Figure 7. Moisture content of the packing material (% of wet mass) along the biofilter bed. Biofilter 1 (peat : bark : Wood = 70 : 10 : 20 wt %, ♦); biofilter 2 (peak : bark : wood = 10 : 70 : 20 wt %, ○).

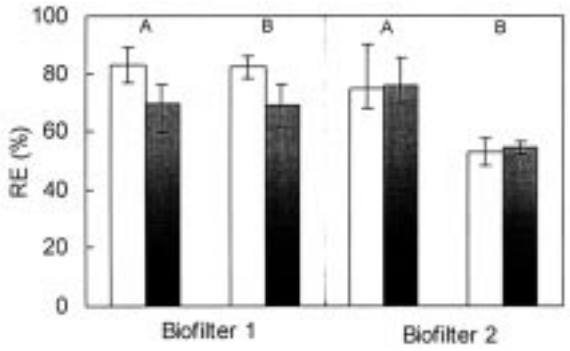

Figure 8. Effect of elimination of nutrients addition. Removal efficiency before (A) and after (B) the period without the addition of nutrients. (□) toluene, (■) xylene.

each only). To investigate this, the upflow air direction was changed to a downflow mode of operation for a couple of days and the individual elimination capacities were measured. The results achieved were the same as before. This finding suggested that the low elimination capacities in the highest part of the bed (section 5 in Figure 5) were not caused by the carbon source transfer rate into the cells. (Local elimination capacities and CO_2 production were measured by Morales et al. (1998) in peat biofilter after the addition of gaseous ammonia into the airstream). They reported that at the beginning, the highest elimination capacity was in the middle zone. Subsequently, this zone showed a gradual decrease of the capacity, while the inlet zone showed an increase of the capacity. Hence the highest capacity was found in the inlet zone. This change was assumed to be a consequence of the retention of the highest amount of ammonia in the inlet zone (and the elimination of nitrogen limitation). Low elimination capacities at the outlet of biofilter were explained by nitrogen limitation.

Moisture content of the packing material

Figure 7 illustrates the moisture content profiles along the packed bed of both biofilters. The moisture content of biofilter 1 containing the peat as the major constituent of packing material varied in the range of 71–80% (weight). This is in agreement with the range 70–80% recommended for peat biofilters by Martin et al. (1996). However, a moisture content of 40–60% and especially near to the lower value of 40% for the treatment of hydrophobic VOCs is proposed by van Lith et al. (1997) to achieve optimum and sustained biofilter performance. The distribution of the humidity in biofilter 1 and 2 was different. In biofilter 1, two zones, in the lower and upper halves of the column, with a transient zone in the middle of the biofilter could be distinguished.

The moisture content of the biofilter 2 (with bark as a dominating packing material) changed steadily from 78 wt % at the bottom to 65 wt % at the top of the column. Tahraoui and Rho (1998) found a gradual decrease of the moisture content from 64% at the relative height of 0.1 to 53% at the relative height of 0.8 in a biofilter filled with a blend of composted chicken manure and peat moss, when the bed was sprinkled with water. If the biofilter was not watered, the pattern of the moisture content along the bed height was opposite (the moisture content of 53% and 60% at the relative heights of 0.1 and 0.8 respectively).

Nutrient input

In the 9th and 10th months of performance, the regular weekly pouring of nutrient solution through the bed was interrupted for 30 days. Figure 8 shows the comparison of removal efficiencies before and after this 30-day period. Whilst in biofilter 1, no decrease of removal efficiencies was found, the removal efficiencies of biofilter 2 declined from about 75% to 53% for both pollutants. If the removal efficiencies in 8th month (cf. Figure 3) are compared with those in 9th month (Figure 8 – removal efficiencies before the nutrient solution elimination), the decrease is evident. The lower removal efficiencies in 9th month result from a loss of packing material due to sampling for the moisture content measurement and, therefore, a shorter mean residence time of the pollutants in the bed.

Conclusions

Both biofilters demonstrated a good stability during the period of fourteen months. Despite the starvation periods and changes in loadings, the removal efficiencies remained in the range approximately 80–90% up to the 8th month of operation. The evaluation of concentration profiles showed simultaneous biodegradation of toluene and xylene. An irregular decrease of both toluene and xylene concentrations along the bed was observed. The biofilter with peat as a dominant packing material showed larger differences in local elimination capacities. The major part of pollutants was degraded in the middle layer of the bed where some 35% of all carbon was removed. The biofilter filled with compost as a major constituent showed smaller differences between local elimination capacities; the highest removal rates were observed in the layers 1 and 4 (20 and 19% of all carbon was removed in these layers respectively). The moisture contents of peat and compost dominant packing materials ranged in the intervals of 71–80% and 65–78% respectively. A higher moisture content was found at the bottom of biofilters. The removal efficiency of the biofilter with peat was not influenced by the 1-month elimination of nutrients, but the biofilter with bark showed a decrease of efficiency from about 75 to 53% for both pollutants.

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