Environmental Technology
Vol. 30, No. 6, May 2009, 621–627

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The effects of a lower irrigation system on pollutant removal and on the microflora of a biofilter

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(*Received 22 December 2008; Accepted 20 February 2009*)

Moisture control is one the most important parameters in biofilters for air pollution control. Biofilters tend to experience drying at the air inlet port, which causes decreased pollutant removal over time. In this study, the installation of an irrigation system within the lower part of the biofilter bed was proposed, and its effect was quantified in a laboratory scale biofilter operated side by side with a control biofilter. The removal of toluene vapours at short gas residence time (13.5 s) served as a model system. The results showed that the rate of toluene elimination in the biofilter with the lower irrigation system was 1.2–1.7 times greater than the rate of toluene elimination in the control biofilter. At the completion of the two-month experiment, a detailed examination was conducted of the packing materials with the immobilized pollutant-degrading culture. The results highlighted the effects of bed drying on cell viability in the control biofilter. They also revealed that the bottom segment of the biofilter with the lower irrigation system had a higher moisture content, a higher biomass density and a larger fraction of active biomass than the corresponding segment in the conventional biofilter. These detailed examinations explained why an increased toluene removal was observed in the system equipped with a lower irrigation system. Overall, this study demonstrates enhanced pollutant removal in biofilters equipped with a lower irrigation system through a better control of moisture.

Keywords: biofilter; moisture control; biological activity; air pollution control; biodegradation

Introduction

Biofiltration is an increasingly accepted technology for the control of odours and emissions of volatile organic compounds (VOCs) from stationary sources [1]. Biofilters work by passing polluted air through a packed bed on which a pollutant-degrading biofilm develops [1–3]. Under optimum conditions, the micro-organisms forming a biofilm on the packing rapidly degrade absorbed pollutants to harmless end-products. Biofilters are effective and environmentally friendly; they require low energy input and unlike conventional air pollution control technologies, they do not produce secondary pollutants such as spent activated carbon, or CO_2 from the burning of fossil fuel.

The selection of a suitable biofilter packing material is important in order to obtain high pollutant removal efficiencies and to maintain performance over the long-term [1,3]. Factors such as bed porosity and air permeability, water permeability and absorption capacity, compression strength and long-term stability play an important role in the mechanical suitability of a material for use as a support in biofilters. On the other hand, the specific interfacial area, surface properties, water absorption capacity and the ability to slowly release nutrients affect the suitability of a material to host a thriving culture of pollutant-degrading bacteria. Other properties such as pollutant adsorption capacity also have an impact on the future performance of a packing material [1]. For field application of biofiltration, economic factors have to be considered as well.

While packing selection is essential for the success of a biofilter, proper reactor design and operation are also extremely important. One of the most critical operating parameters is the control of moisture in biofilters [1,3,4]. Drying of the packing is a common problem in biofilters [5–8], which results in both structural problems in the bed, such as bed shrinking, heterogeneous air flow and generation of fine particles, and reduced biological activity [9]. In a previous study [10], four biofilter packing materials were compared for the removal of low concentrations of toluene vapours at short retention times (13.5 s). A key observation was that incomplete humidification of the influent air caused drying of the packing material at the inlet of the reactor

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ISSN 0959-3330 print/ISSN 1479-487X online

DOI: 10.1080/09593330902838106
http://www.informaworld.com

and possibly reduced the performance of the biofilter. VanLith *et al.* [4] proposed an evaluation criterion to determine the risk of bed drying in biofilters depending on the operating conditions. The criterion illustrated that drying is more likely to occur in high performance biofilters (i.e. short gas residence times or high pollutant concentrations). It also demonstrated that drying can occur even when the inlet air is close to 100% humidity which was the case in the Sakuma *et al.* study [10] and in other studies [11,12]. In the majority of cases, drying starts at the air inlet port. In biofilters operated with air in a downflow mode, additional moisture can easily be applied using sprinklers above the bed [1,4]. However, in biofilters operated with the air in upflow mode, the solution is more difficult as water applied from the top of the bed will need to percolate through the entire bed to reach the section susceptible to drying, posing possible process control challenges and waterlogging issues within the bed. Another approach is needed.

In this paper, the beneficial effects of installing an irrigation system within the lower portion of a biofilter bed are demonstrated. Two laboratory-scale upflow biofilters were operated in parallel: a conventional biofilter that served as a control and one with an irrigation system installed within the bed at one third of the bed height. The focus was on demonstrating the effectiveness of such a system for the control of bed drying and quantifying the effects on pollutant removal and on the density and activity of the pollutant-degrading bacteria in the biofilter.

Materials and methods

Biofilter and packing materials

Two biofilters (thereafter referred to as conventional biofilter and lower irrigation biofilter, respectively) were used in this study. The biofilters were constructed from clear polyvinyl chloride pipe and were 1.2 m in length and 10 cm in internal diameter. The lower irrigation biofilter had a drip system consisting of a ring-shaped perforated tubing installed within the bed at one third of the bed height, a pump controlled by a timer, and a water collection tank. Figure 1 provides a schematic of the experimental set-up.

The biofilters were packed to a height of 60 cm with Cattle Bone Porcelite (CBP) beads (4 mm diameter, Aisin Takaoka Co. Ltd, Toyota, Japan) which had proven to be a very good packing for biofilters [10]. Cattle Bone Porcelite is a porous ceramic (37% microporosity; 0.8 g cm^{-3} bulk density) manufactured with 20% vol. of cattle bone powder in the raw material. During the making of the ceramic beads, part of the cattle bone powder burns leaving pore space and ashes, whereas the remainder of the cattle bone is believed to act as a slow-release nutrient source for microorganisms [10]. The biofilters were initially inoculated with activated sludge and then re-inoculated with several mixed cultures of toluene-degrading microorganisms from another reactor in our laboratory [13] on day 24. The initial moisture content of the packed bed was about 22% (weight by bed vol. basis).

Set-up and operating conditions

Compressed air was passed through a humidifier consisting of a 40 L carboy filled with water and then passed over vials containing toluene in a mixing chamber. The concentration of toluene was adjusted by changing the number of vials and partially obstructing the openings of the mouths of the vials. The humidified contaminated air stream (relative humidity of about

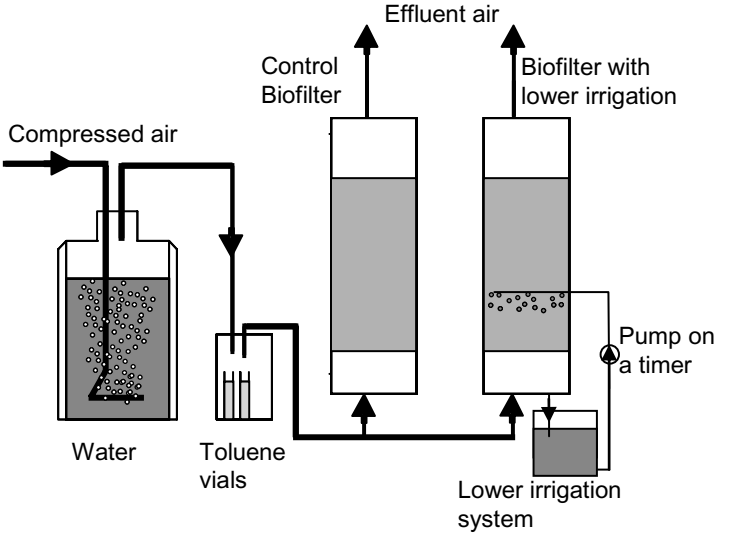

Figure 1. Schematic of the experimental set-up (not to scale).

95%) was supplied to the bottom of the biofilters. The airflow rate in each reactor was 20 L min^{-1}, corresponding to an empty bed residence time of 13.5 s. The toluene concentration in the influent gas stream ranged from 0.07 to 0.81 g m^{-3} resulting in loadings of 19–217 g m^{-3} h^{-1}. These concentrations match the needs of many air pollution control systems. All experiments were carried out at a room temperature (21–24 °C). One hundred and twenty milliltres of mineral medium were manually supplied to the top of each biofilter twice per week using a simple drip system. The mineral solution contained (L^{-1}): 2.0 g KH_2PO_4, 2.0 g K_2HPO_4, 2.0 g KNO_3, 2.0 g NaCl, 0.04 g $MgSO_4$, 0.04 g $CaCl_2$, and 2 mL of a trace elements solution. The trace element solution contained (L^{-1}): 1.5 g $FeCl_2{\cdot}4H_2O$, 0.06 g H_3BO_3, 0.1 g $MnCl_2{\cdot}4H_2O$, 0.12 g $CoCl_2{\cdot}6H_2O$, 0.04 g $ZnCl_2$, 0.025 g $NiCl_2{\cdot}6H_2O$, 0.015 g $CuCl_2{\cdot}2H_2O$, 0.025 g $NaMoO_4{\cdot}2H_2O$, 18.27 mL 37% HCl and 5.2 g EDTA $Na_4.4(H_2O)$.

For the lower irrigation biofilter, in addition to above mentioned supply of mineral medium, water was used to periodically irrigate the lower third of the filter bed. Deionized water was trickled through the bottom third segment using a peristaltic pump and a timer at a rate of 5 L h^{-1} for 20 minutes, five times per day. The leachate (pH 6.7–7.4) was collected and reused in the subsequent irrigation events. Irrigation water was replaced by fresh deionized water once a month. With the exception of the lower irrigation system, the design and operation of the two biofilters were identical.

Analytical methods

Gaseous toluene concentrations were measured on an HP 5890 Series II gas chromatograph fitted with an HP-5 capillary column and a flame ionization detector. Online monitoring of CO_2 in the reactor influent and effluent air was performed using a non-dispersive infrared probe and a data logger from Vernier Instruments (Beaverton, OR). Pressure drop was measured by a U-tube water manometer.

At the end of the experiments, the packed bed of each biofilter was gently removed and the packing was split into three sections of equal volume (top third, middle, and bottom third of each reactor). Each section (1.5 L of packing) was independently mixed, and subsamples were taken for several measurements described below.

For moisture and dry biomass determinations, subsamples of about 30 cm^3 of packing from each section were placed in an oven at 80 °C for 48 hours until they reached a constant weight. The moisture content in each section of the biofilter was determined by measuring the weight loss after drying. The dried samples were then placed in a furnace at 800 °C for four hours, and the dry biomass content per volume of packing material was determined by measuring the weight loss and correcting for the loss of an abiotic control.

For protein analysis and microscopy observation, subsamples of about 60 cm^3 of packing were each mixed with 180 mL of double-concentrated mineral medium (i.e. twice the concentrations listed above) and shaken on a rotary shaker at 300 rpm for 10 min, and the resulting suspension used for the analyses. Protein analysis was performed using a BCA Protein Assay Kit from Pierce (Rockford, IL, USA) following the manufacturer's instructions.

Live and total cell counts were conducted. Staining with 5-cyano-2,3,-ditolyl tetrazolium chloride (CTC) for live cells and 4′,6-diamidino-2-phenylindole, dihydrochloride (DAPI) for total counts was performed using a procedure modified from Bhupathiraju *et al.* [14] and Rodriguez *et al.* [15]. Staining was performed in 2 mL microcentrifuge tubes containing 0.4 mL of suspended bacteria produced as described above. The CTC was added to a final concentration of 5 mM from a stock solution (50 mM) prepared in deionized water. Samples tubes (wrapped in aluminium foil to protect from light) were incubated for four hours in the dark at room temperature before counterstaining. All of the staining assays were performed in duplicate. Cultures treated with sodium azide (3.2% final concentration, treated 15 min) were used as killed controls. After CTC incubation, DAPI was added to a final concentration of 0.01%. Samples were first mixed for three minutes; then one drop of the samples was transferred to a clean microscope slide and a coverslip was immediately laid on the slide. Fluorescence microscopy was performed with an Olympus BX51 microscope at 400× magnification. Two different excitation and barrier filters were used to simultaneously observe CTC and DAPI fluorescence. A total of three fields per slide were counted. The ratio of active bacteria was determined by:

$$Ac = (N_{act}) / (N_{tot}) * 100 \quad (1)$$

where Ac is the ratio of active bacteria (%), N_{act} is the number of live bacteria per field determined by CTC staining and N_{tot} is the total number of bacteria per field determined by DAPI staining.

Results and discussion

Performance of the biofilters

The results of the continuous operation of the two biofilters are reported in Figure 2. Throughout the 58 days of operation, the inlet concentration was varied to determine the performance of the biofilters over a range

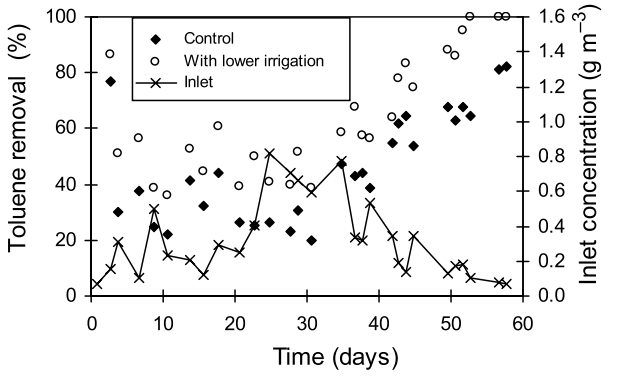

Figure 2. Toluene inlet concentration and steady-state removal efficiencies of the control biofilter and the biofilter fitted with the lower irrigation system during the experiment. The empty bed residence time was kept constant at 13.5 s.

of conditions. Pressure drop remained low for both biofilters during the entire study and ranged from 0.2 to 0.5 cm of water gauge most of the time, with a few scattered data reaching 2 cm water gauge towards the end of the study (detailed data not shown). During the initial 24 days, the toluene removal was at most 60%, which was below the expected value based on earlier studies [10]; hence both reactors were re-inoculated with mixed cultures from other toluene-degrading bioreactors in our laboratory [13]. The inlet toluene concentration was also increased to ensure sufficient feeding for biomass growth. This resulted in improved toluene removal, and the performance of both biofilters could then be compared.

At pseudo-steady state, toluene was mineralized to CO_2 in both biofilters and a carbon balance was conducted. The recovery of the carbon-toluene degraded as carbon-CO_2 decreased with increasing the inlet concentration (Figure 3). As was discussed earlier [16], this decreasing trend is due to the fact that when pollutant-substrate ratio is low, the majority of the pollutant degraded is used for cell maintenance purposes, while at a high pollutant-substrate ratio, a significant portion of the degraded pollutant is incorporated into biomass with net growth of the process culture. Differences in CO_2 recovery between the two biofilters were not statistically significant indicating that the biomass yields in the two reactors were approximately the same. This is consistent with the fact that the pollutant and nutrient loadings were identical in the two reactors. The degraded toluene recovered as carbon dioxide was greater than 100% at toluene inlet concentrations lower than 0.1 g m^{-3} for both biofilters. This indicates that cell death was faster than cell growth at the lowest toluene inlet concentrations and suggests that long-term operation at these low concentrations may not be stable. It also explains the slow start-up and low biomass build-up in the first 24 days due to the low inlet toluene concentrations.

A plot of the steady-state toluene elimination capacity (EC) vs. loading is shown in Figure 4 whereas the maximum elimination capacities and critical loadings are summarized in Table 1. At an empty bed residence time of 13.5 s, the critical load for the lower irrigation biofilter was 47 g m^{-3} h^{-1}. In contrast, the conventional biofilter never reached a removal of 95% under the same conditions. A paired comparison of both biofilters revealed that the difference in performance between the two bioreactors was statistically significant and that the biofilter fitted with the lower irrigation system had a toluene elimination capacity on average 1.44 times higher than that of the conventional biofilter. Thus, the results demonstrate that the lower irrigation system improved biofilter performance. This was most probably achieved by avoiding dry zones and increasing bacterial activity in the lower third segment of the biofilter. Experimental evidence to support this conclusion is presented in the next sections.

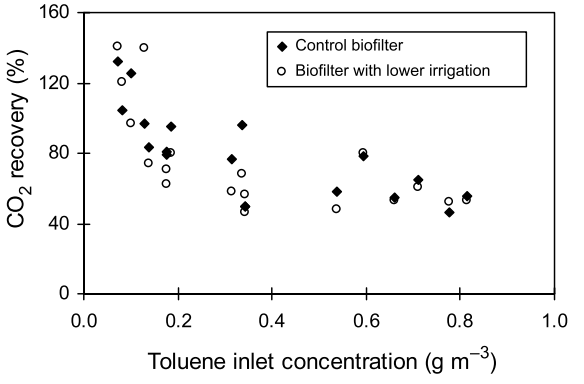

Figure 3. Recovery of the degraded carbon-toluene, as carbon-CO_2, as a function of the inlet toluene concentration for all data after day 24.

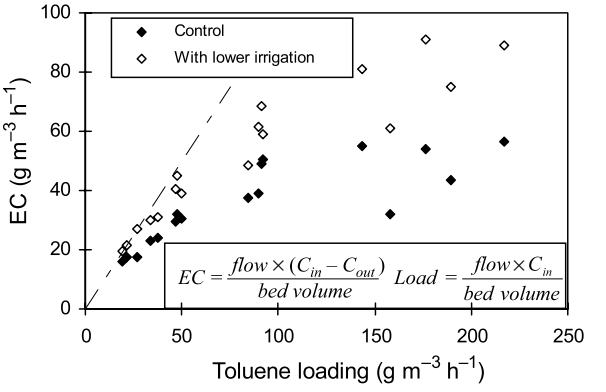

Figure 4. Toluene elimination capacity (EC) vs. load (data after day 24).

Table 1. Summary of the maximum elimination capacity (EC_{max}) and critical loading (defined as the maximum load at which 95% removal occurs) for toluene removal at a 13.5 s empty bed gas residence time.

	Conventional biofilter	Lower irrigation biofilter
EC_{max} (g m^{-3} h^{-1})	56	90
Critical load (g m^{-3} h^{-1})	95% removal not reached	47

Detailed packing and bacterial analysis

The differences in toluene removal between the two biofilters stimulated further analysis of the packing and detailed examination of the attached microbial culture. The results are presented in Figures 5–8. All analyses were conducted at the end of the experiment (day 58). The packing moisture data (Figure 5) show that the bottom segment of the biofilter fitted with the lower irrigation biofilter had a higher water content than that of the control biofilter. Figure 5 also reveals signs of bed dry-out in the bottom segment of the conventional biofilter. When dry biomass content (Figure 6) was analysed, the lower irrigation biofilter showed a slightly higher value in the lower segment compared with the conventional biofilter, although the differences were modest. Protein analyses (Figure 7) revealed that the biomass content in the lower irrigation biofilter was 1.5–3 times that of corresponding packing segments in the convential biofilter. Altogether, this indicates that irrigation of the lower segment of the test biofilter led to a greater moisture content and more homogeneous distribution of water in the biofilter, which then resulted in a greater density of bacteria. This is consistent with the investigations of Sun *et al.* [17] and Prado *et al.* [18] on the effect of moisture, which both showed that

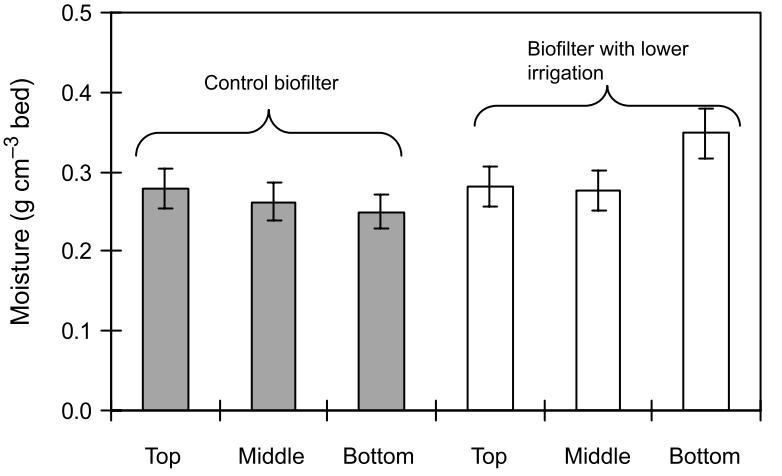

Figure 5. Moisture content in the different sections of the biofilters (day 58). Error bars show standard deviations.

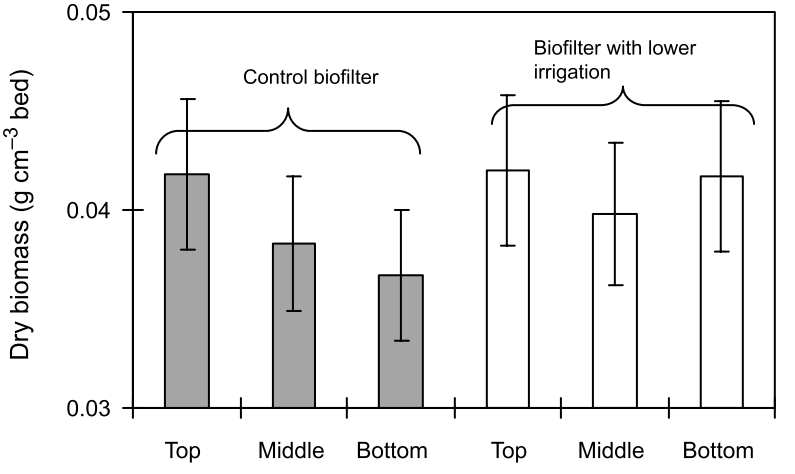

Figure 6. Dry biomass content in the different sections of the biofilters (day 58). Error bars show standard deviations.

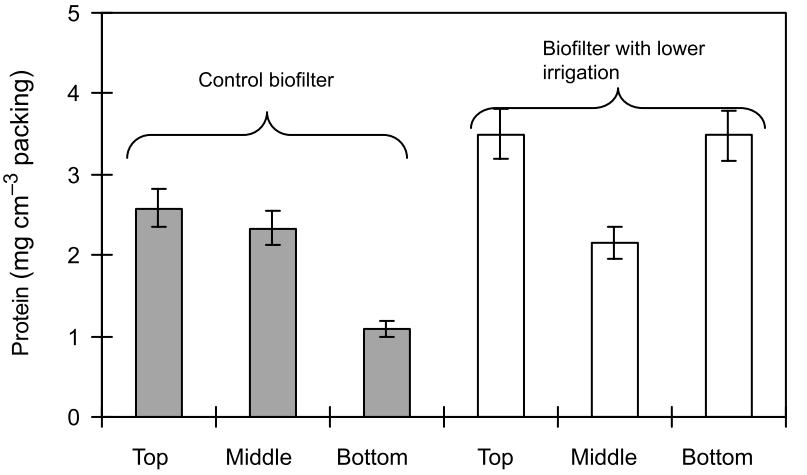

Figure 7. Specific protein content in the different sections of the biofilters (day 58). Error bars show standard deviations.

packing material with a higher moisture content was more favourable for the growth of microorganisms and for a better distribution of water and nutrients.

The modest differences in dry biomass content compared with the large differences in protein content (Figure 6 vs. Figure 7) between the reactors suggested that there must be very significant differences in the viability and the activity of the culture between the two biofilters. Cell activities have often been determined by pollutant-induced oxygen uptake rate [19]. Here, live and dead cell counts were conducted, since toluene biodegradation had already been determined during the biofiltration experiments. A summary of the results is presented in Figure 8. Clearly, the bottom part of the conventional biofilter was experiencing drying-out and the process culture was under significant stress, as indicated by the lowest of all live cell counts and the lowest fraction of active cells. Comparatively, the bottom segment of the biofilter fitted with the lower irrigation system had a much higher absolute count of active cells and the highest fraction of active cells. The latter can be explained by the fact that the air inlet port of a biofilter is exposed to the highest pollutant concentrations, which stimulates active bacterial growth. This is consistent with toluene mineralization data presented in Figure 3.

Overall, the combined results of Figures 5–8 provide a strong explanation as to why the biofilter equipped with the lower irrigation system exhibited better performance than the conventional biofilter. Irrigation of the bottom of the bed provided better moisture and environmental conditions in the bottom

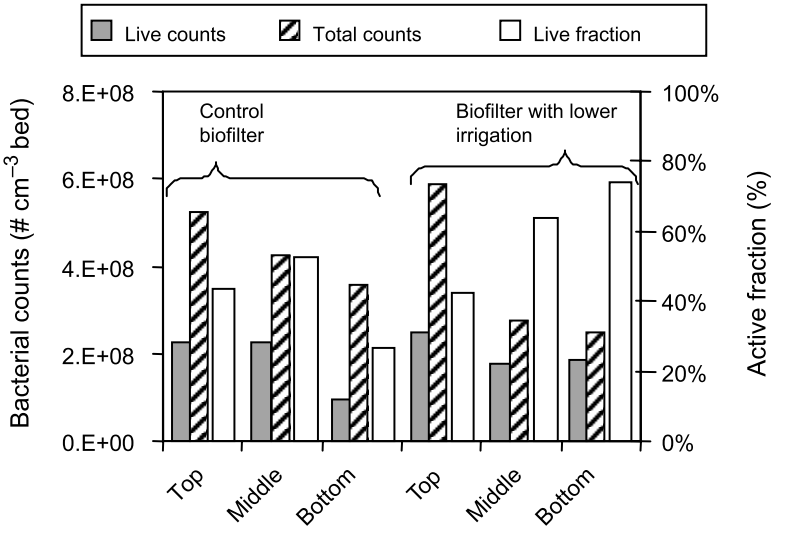

Figure 8. Live and total bacterial counts on the packing and fraction of live bacteria in the different sections of the two biofilters (day 58).

segment of the reactor; hence a higher cell density and a higher fraction of viable cells were able to develop. Drying-out of the bed was avoided and greater toluene removal activity ensued in the bottom of the biofilter.

Conclusion

This study demonstrated that the commonly observed drying-out of biofilter packing at the air inlet port and resulting losses in pollutant removal could be mediated with the installation of an irrigation system in the lower part of the biofilter bed. Detailed experiments showed that the lower irrigation system maintained adequate moisture content in the packing and provided better environmental conditions for the microorganisms. This resulted in greater density and activity of microorganisms, which in turn resulted in 1.2 to 1.7 times greater toluene elimination capacity depending on the conditions. Additional advantages of a lower irrigation system, not explored within this study, include the ability to flush out possible biodegradation metabolites, the establishment of differentiated pH zones, for example for the combined treatment of H_2S or NH_3 together with volatile organics, and possibly the washing out of excess biomass. While these ideas have been around for some time, side-by-side studies such as the one presented here to quantify the benefits are warranted.

Acknowledgement

The financial support of Aisin Takaoka Co., Ltd, Japan, who funded this study, is greatly acknowledged.

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