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Tubular biofilter for toluene removal under various organic loading rates and gas empty bed residence times

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HIGHLIGHTS

- A TBF was developed with tubular structure and tubular sponge media in 3 cm thick.
- An area of the outermost surface medium bed per specific medium volume is much larger than most existing biofilters.
- VOC could be effectively removed by the TBF even at short gas EBCTs.
- No apparent biomass accumulated in the TBF during 391 days' operation.

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ABSTRACT

A tubular biofilter (TBF) which consisted of a closed chamber, a polyurethane sponge tube and a nutrient solution distributor was developed and evaluated under organic loading rates (OL) ranging from 18.7 to 149.3 g m^{-3} h^{-1} and gas empty bed residence times (EBRTs) of 30–5.0 s. Using toluene as model VOC, the startup of the TBF lasted approximately 7 weeks. The removal efficiency decreased from 99% to 52.2% when OL was increased from 18.7 to 149.3 g toluene m^{-3} h^{-1} at 15 s, but did not decline significantly when the EBRT was reduced from 30 to 5.0 s at 18.7 g m^{-3} h^{-1}. Biomass concentration did not increase significantly within the sponge tube during the 391 days' operation as observed through the Plexiglas pipe of the TBF. The TBF is suitable for treating waste gases with low toluene concentrations even at high gas flow and over long periods.

1. Introduction

Biofiltration has been successfully employed for the removal of volatile organic compounds (VOCs) from waste gas streams due to its cost effectiveness and environmental friendliness (Melse and van der Werf, 2005; Montes et al., 2010; Popat and Deshusses, 2009; Yang et al., 2011). However, two problems still hamper the widespread application of traditional biofilters and biotrickling filters at full-scale. On one hand, a large footprint of the biofilters is usually required due to the fact that a longer gas empty bed residence time (EBRT) should be allocated for a biofilter than for other reactors such as chemical scrubbers (Gabriel and Deshusses, 2003; Gallastegui et al., 2011; Kennes et al., 2009; Rene et al., 2011). On the other hand, removal performance of a biofilter often declines due to excessive accumulation of biomass within the filter media over extended periods of operation (Omri et al., 2011; van Groenestijn and Kraakman, 2005; Yang et al., 2010).

To avoid these problems, rotating drum biofilters (RDBs) were successfully developed in previous studies (Yang et al., 2003, 2004, 2008). The RDBs could achieve high and stable removal efficiencies (REs) even at high organic loading rates over long-term operation periods. The cylindrically wrapped polyurethane sponge medium through which gas streams was passed from the outermost layer to the innermost layer is helpful for promoting bioflter performance even under high organic loading rates over long-term operation periods. Unfortunately, disadvantages of high construction and operation costs still exist for RDBs. Although drum rotation might contribute to high performance, it also significantly increases the construction and operation costs of RDBs. Nevertheless, various patterns of biomass accumulation including surface, shallow, in-depth and reverse patterns have been proposed on

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the basis of removal mechanisms (Yang et al., 2003, 2004). Studies have demonstrated that mass loading along the height of a filter bed declines as contaminants are removed (Alonso et al., 1998; Thullner and Baveye, 2008). A high filter bed is needed when the pollutant in waste gases penetrates through the filter bed of biofilters more readily. Some waste gases with biodegradable contaminants at low concentrations can penetrate only a small way into the filter bed when the dominant removal mechanism is the surface or shallow pattern. A thin tubular layer of polyurethane sponge is ideal for biofilters to overcome the problems mentioned above when the surface or shallow pattern is the dominant removal mechanism.

In the present study, a tubular biofilter (TBF) was developed. The TBF consisted of a closed chamber with a tubular layer of polyurethane sponge and a nutrient solution distributor. Toluene was used as the model VOC. The performance of the TBF was evaluated continuously for 391 days under various organic loading rates and gas EBRTs at either a constant VOC loading or a constant VOC influent concentration.

2. Methods

2.1. Tubular biofilter system

The biofilter consisted of a closed chamber, a module polyurethane sponge tube and a nutrient solution distributor. The closed cylindrical chamber had an inner diameter of 16 cm and a height of 15 cm; it consisted of a Plexiglas pipe and two perforated Plexiglas plates at both ends. The nutrient solution distributor included two nutrient feeding channels and a horizontal perforated plate for uniform distribution of liquid, and was impermeable for waste gases (Fig. 1).

A clean air stream and a neat toluene (99.5%, Changsha Antai Chemical Industry Co. Ltd., Hunan, China) vapor stream were used to synthesize the waste gas streams. The mixed gases entered the TBF through the gas inlet on the top of the chamber and passed through the sponge tube where biodegradation took place by microorganisms attached to the polyurethane sponge. The purified gas was emitted through the gas outlet located at the bottom face of medium tube. The nutrient solution was pumped into the TBF at a rate of 6.0 L d^{-1} and uniformly distributed throughout the sponge, and the effluents collected were circulated to the medium tube.

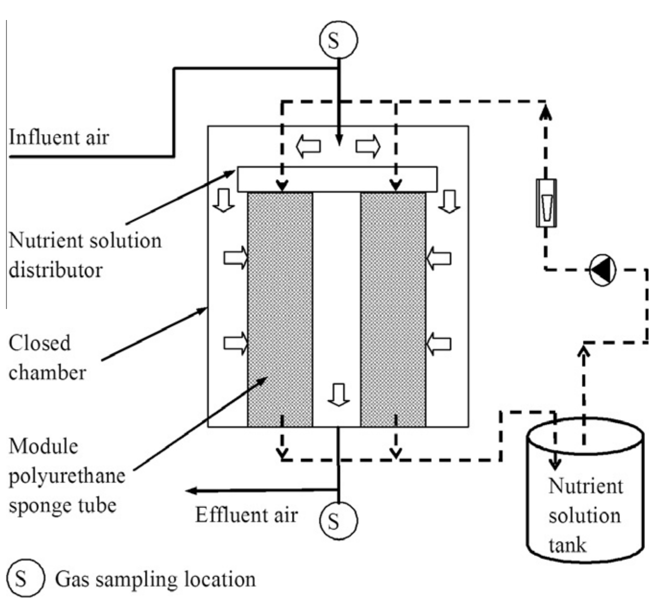

Fig. 1. Schematic of the tubular biofilter.

2.2. Filter material

The material on which the biofilm grew in the TBF consisted of open-pore reticulated polyurethane sponge. A module polyurethane sponge tube produced from the Shenzhen Jiechun Filter Material Co. Ltd. (Guangdong, China) was mounted in the center of the closed cylindrical chamber located between the nutrient solution distributor and the bottom of the chamber. The porosity and pore size of the sponge medium were 98% and approximately 12 pores per centimeter (30 PPI), respectively. The characteristic parameters of the module polyurethane sponge tube are listed in Table 1.

2.3. Seed cultures

Fresh activated sludge was taken from a secondary sedimentation tank at Changsha Guozhen Wastewater Treatment Co. Ltd., Hunan, China and used for seeding the TBF.

2.4. Chmemical reagent

Analytically pure toluene (99.5%, Changsha Antai Chemical Industry Co. Ltd., Hunan, China) was used as the target contaminant in the preparation of the model waste gases.

Analytically pure reagents that are common in the commercial market in China were used for preparing the nutrient solution including $NaNO_3$ 490 mg L^{-1}, K_2HPO_4 28.2 mg L^{-1}, $NaHCO_3$ 24.0 mg L^{-1}, KH_2PO_4 9.3 mg L^{-1}, $CaCl_2$ 2.7 mg L^{-1}, $MgSO_4$ 4.4 mg L^{-1}, $CoCl_2{\cdot}6H_2O$ 0.86 mg L^{-1}, $MnCl_2{\cdot}4H_2O$ 0.14 mg L^{-1}, $FeCl_3$ 0.09 mg L^{-1}, $CuCl_2{\cdot}2H_2O$ 0.06 mg L^{-1}, and that Riboflavin, 4-aminobenzoic acid, Pyriodoxine HCl, Folic acid, Nicotinic acid and Thiamin HCl were all less than 0.01 mg L^{-1}.

2.5. Analytical methods

Toluene concentrations in both the influent and the effluent were measured using a gas chromatograph (GC) (HP 6890N, Series II, Hewlett–Packard, Palo Alto, California, USA) equipped with a flame ionization detector. A HP-VOC capillary column (60 m × 320 μm ID × 1.8 μm, Agilent, USA) was used for the analysis. High purity nitrogen gas (99.9%) was the carrier gas and supplied at a flow rate of 30 mL min^{-1}. The temperatures at the GC injector, oven, and detector were set at 120, 120, and 250 °C, respectively. The samples from a same gas sampling location were taken and measured in triplicate, and the average values and the corresponding standard deviations of the values are reported.

2.6. Operation of the TBF

A new module polyurethane sponge tube was immersed into clean water for about 2 days, then placed into the activated sludge for the TBF microbial inoculation. After about a week, the sponge tube was taken out from the activated sludge and mounted on the TBF. Subsequently, the synthetic waste gas and nutrient solution were introduced into the TBF. The moment at which the media was introduced into the biofilter was counted as the initial time of TBF operation (the 1st day).

The reference conditions were set at gas EBRT of 15 s and organic loading rate of 18.7 g toluene m^{-3} h^{-1} was used to track the reproducibility and pseudo steady-state condition of the TBF. During operation, the TBF was adjusted to the reference level for about 1 week before and after a change in an operating parameter.

Table 1
Characteristics of the module polyurethane sponge tube.

Porosity (PPI)	Radial thickness (mm)	Outer diameter (mm)	Inner diameter (mm)	Height (mm)	Outer lateral surface area (m^2)	Inner lateral surface area (m^2)	Volume (L)
30	30	140	80	100	0.044	0.025	1.04

The concentration of toluene in the synthetic waste gases can be adjusted by changing the flow rate of the clean air stream and the amount of neat toluene vapor, while the gas EBRT can be altered by changing the flow rate of the clean air stream. When the organic loading rate was doubled, the inlet gas flow rate remained constant but the toluene concentration in the influent doubled. The gas EBRT was decreased by increasing the flow rate of the clean air stream, so that the inlet VOC concentration decreased while the organic loading rate remained unchanged. When the effect of gas EBRT on the TBF performance at a constant incoming toluene concentration was studied, the gas flow rate and organic loading rate were increased and the gas EBRT was decreased. While the organic loading rate, gas EBRT, or gas flow rate was changed, all other parameters including nutrient solution concentration, nutrient solution feed rates and temperature, etc., remained constant.

The TBF was operated for 391 days. Days 1–52 was considered as the initial startup stage and the TBF was kept at the reference conditions. Throughout days 53–202, 203–338, and 339–391, the effects of organic loading rate and gas EBRT at a constant VOC loading or at a constant VOC incoming concentration on TBF performance were evaluated, respectively. The operation conditions after the initial startup stage are listed in Table 2. The biomass that accumulated in the filter of the TBF was not removed.

3. Results and discussion

3.1. TBF performance on the initial startup stage

The TBF was started and operated for 52 days at the reference operation condition. When inlet toluene concentration was maintained at 77 mg m^{-3} in the initial startup stage, the outlet toluene concentration decreased while the RE increased with time (Fig. 2). Toluene REs were less than 50% in the first 7 days, and reached 95% at day 52.

However, a relatively longer start-up period was needed for the TBF than for other types of biofilters since approximately 7 weeks was necessary and the RE did not exceed 90% until day 45. Although a long period for the start-up of biofilters is a major problem in biofilter operation, it could be overcome by modifying a hydrophobic polymer with additives containing hydroxyl groups to improve water retention of the material, microbial growth,

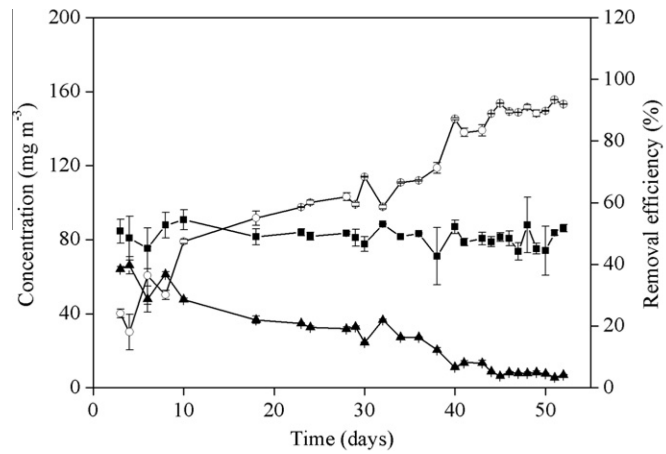

Fig. 2. Performance of the tubular biofilter on the startup stage. (■) Inlet toluene concentration, (▲) outlet toluene concentration, (○) toluene removal efficiency. The single data point is representing the mean value of the data, while error bars representing the overall distribution of the data.

and attachment of microorganisms (Gutirrez-Acosta et al., 2010). That biofilm rapidly and effectively formed on a solid support is helpful for the bioprocess (Prado et al., 2009). Besides, Vergara-Fernández et al. (2011) investigated four different carbon sources (glycerol, 1-hexanol, wheat bran, and n-hexane) to reduce the startup period and obtained a short adaptation period of 7 days when wheat bran was as nutrient source used for *n*-hexane removal in a fungal biofilter.

3.2. Effect of organic loading rate

Following the startup stage, the TBF was operated at organic loading rates of 18.7, 37.3, 74.6 and 149.3 g toluene m^{-3} h^{-1} from day 53 to day 202, and the corresponding average REs were 99%, 84.5%, 72.0% and 52.2%, respectively (Fig. 3).

The TBF could achieve a higher toluene RE than a peat biofilter (Alvarez-Hornos et al., 2007) or an agricultural by-product-based

Table 2
Operation conditions during the whole experiment. For time periods not listed, the TBF operated under the reference condition at gas EBRT of 15 s and organic loading rate of 18.7 g toluene m^{-3} h^{-1}.

Time (d)	OL (g m^{-3} h^{-1})	C_0 (mg m^{-3})	EBRT (s)
61–105	37.3	155	15
113–150	74.6	310	15
159–194	149.3	620	15
224–249	18.7	55	10
273–288	18.7	37.5	7.5
300–309	18.7	25	5.0
318–326	18.7	150	30
339–347	37.3	77	7.5
354–381	74.6	77	3.8

OL, organic loading rate, g m^{-3} h^{-1}; C_0, inlet toluene concentration, mg m^{-3}; EBRT, empty bed residence time, s.

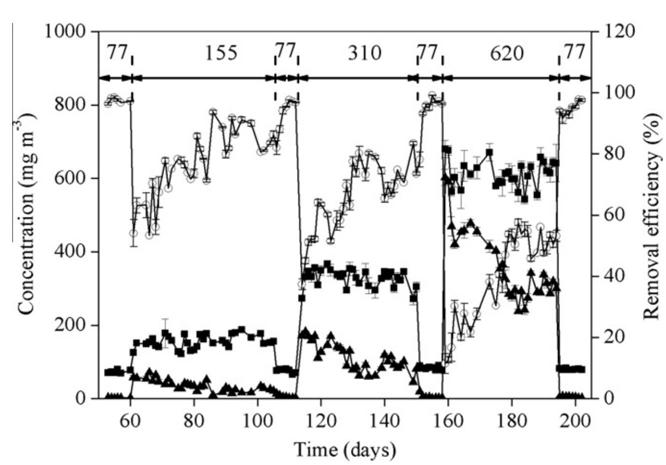

Fig. 3. Toluene removal efficiencies at various organic loading rates. (■) Inlet toluene concentration, (▲) outlet toluene concentration, (○) toluene removal efficiency. The single data point is representing the mean value of the data, while error bars representing the overall distribution of the data.

biofilter (Krishnakumar et al., 2007) at a lower organic loading rate and a shorter gas EBRT. After the organic loading rate was doubled, it took the TBF approximately 30 days to reach a relatively stable RE (Fig. 3), while it only took 2–4 days for the TBF to recover its RE at the reference operation condition after changing from a high organic loading rate. Cai et al. (2007) also reported that the re-acclimation of biofilter performance was delayed by increasing influent concentrations for trickle bed air biofilters. Therefore, the TBF is recommended for treating waste gases with a stable organic loading and a moderately high or low VOC concentration.

As shown in Fig. 3, the ECs were 18.3, 33.5, 58.5, and 83.0 g toluene $m^{-3}\,h^{-1}$ at organic loading rates of 18.7, 37.3, 74.6, and 149.3 g toluene $m^{-3}\,h^{-1}$, respectively. Though it increased with an increase in the toluene loading rate, the EC was lower than the load at the highest organic loading rate. The EC of BTF packed with 30 cm high structured synthetic polyurethane sponges achieved 34.0, 49.2 and 113.0 g toluene $m^{-3}\,h^{-1}$ at influent loading rate of 34.4, 70 and 140 g toluene $m^{-3}\,h^{-1}$, respectively (Yang et al., 2011). It can be seen that toluene removal performances have differed little between RDB and TBF under low influent loading rates.

3.3. Effect of 200 gas EBRT at constant toluene loading rate

The TBF performance at gas EBRTs of 30, 15, 10, 7.5, and 5.0 s, was evaluated from day 203 to day 326 when the organic loading rate remained at 18.7 g $m^{-3}\,h^{-1}$ (Fig. 4). The corresponding toluene concentrations in the incoming gas streams were 155, 77, 52, 39, and 26 mg m^{-3}, respectively.

The RE did not change dramatically when the gas EBRT was reduced at an organic loading rate of 18.7 g $m^{-3}\,h^{-1}$. At a gas EBRT of 5 s from day 300 through 309, the RE was only a little lower than that stabilized at the other gas EBRTs, but still achieved a high and stable performance even at a gas EBRT of 5 s. The outlet toluene concentration was otherwise uniformly less than 7 mg m^{-3} in the evaluation period from day 203 to day 338. Under a constant organic loading rate of 18.7 g $m^{-3}\,h^{-1}$, the calculated EC was 18.5, 18.3, 18.4, 18.2, and 18.6 g $m^{-3}\,h^{-1}$ at gas EBRT of 15, 10, 7.5, 5, and 30 s, respectively. The EC of BTF packed with 30 cm high structured synthetic polyurethane sponges achieved 15.2, 14.1, 9.5 and 8.2 g $m^{-3}\,h^{-1}$ at a toluene loading rate of 16 g $m^{-3}\,h^{-1}$ and gas EBRTs of 30, 15, 10 and 7.5 s (Yang et al., 2011). It can be seen that the TBF can achieve a high removal performance at a short gas EBRT.

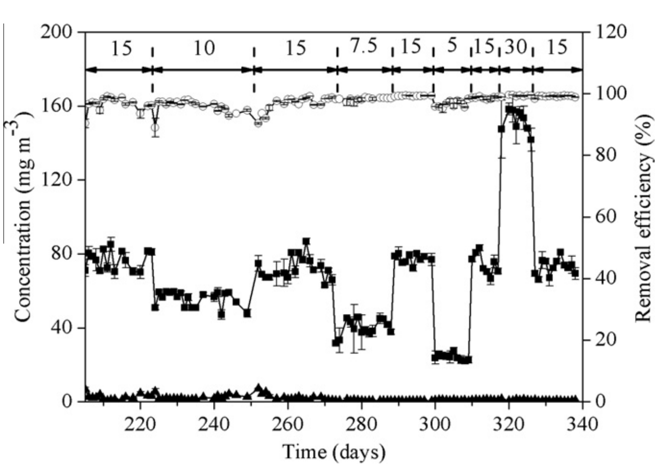

Fig. 4. Toluene removal efficiencies at various gas empty bed residence times and a constant toluene loading rate. (■) Inlet toluene concentration, (▲) outlet toluene concentration, (○) toluene removal efficiency. The single data point is representing the mean value of the data, while error bars representing the overall distribution of the data.

Fig. 4 also shows that the acclimation times needed for the TBF performance recovery were only 1–3 days, such as on day 224 and day 318. In contrast during the period of days 250–272 when the TBF was at an unsteady state, the TBF needed a relatively long period of about 5 days to reach a high RE over 95.4%. Compared to a time of approximately 1 month needed for achieving a high and stable TBF performance when the organic loading rate was doubled (Fig. 3), a much shorter period of time was needed when the gas EBRT was changed in this study. So, the TBF performance is steady at various gas EBRTs from 30 to 5 s over a long period of operation, and a short gas EBRT of less than 7.5 s would be enough for the TBF to achieve a high and stable performance at middle to low toluene concentrations.

Although the small thickness of the filter media is helpful for decreasing the footprints of the bioreactors, problems exist such as a longer duration for the TBF startup and deteriorating performance at high organic loading rates or short gas EBRTs or over a long period of operation. In this study, a long period of time was needed for successful start-up of the TBF or stabilization at a high performance, and the RE dropped when the organic loading rates were increased. It seems that a polyurethane sponge thickness of 3 cm in the TBF is not enough to buffer load variations. However, changes in operating conditions can also alleviate this influence (Wang et al., 2009). Moe et al. (2007) and Nabatilan and Moe (2010) proposed a strategy for managing this problem by installing a column packed with granular activated carbon (GAC) to achieve load equalization prior to biofilter treatment.

According the biofiltration mechanisms proposed by Yang et al. (2003, 2004, 2008), the removal mechanisms of the depth pattern and reverse pattern will be applied to waste gases at high concentrations. However, the TBF was developed for VOC removal when the surface and the shallow patterns were the dominant removal mechanisms. Therefore, TBFs are considered not suitable for the treatment of waste gases at high pollutant concentrations.

3.4. Effect of gas EBRT at a constant incoming pollutant concentration

The selection of a reasonable gas flow rate at fixed pollutant concentrations is important for the design of a full-scale biofilter system. Therefore, the effect of gas EBRT on TBF performance under a constant incoming toluene concentration of 77 mg m^{-3} was evaluated at gas flow rates of 0.25, 0.5 and 1.0 $m^3\,h^{-1}$, and the corresponding gas EBRT were 15, 7.5 and 3.8 s and the toluene loading rates were 18.7, 37.3 and 74.6 g $m^{-3}\,h^{-1}$, respectively. The TBF achieved 99.2%, 97.4% and 83.5% toluene removal when the gas EBRT was 15, 7.5 and 3.8 s, respectively, and the corresponding EC were 18.6, 36.4 and 63.6 g $m^{-3}\,h^{-1}$ (Fig. 5). The EC of 63.6 g $m^{-3}\,h^{-1}$ at incoming concentration of 77 mg m^{-3} and gas EBRT of 3.8 s was a little higher than that of 58.5 mg m^{-3} at incoming concentration of 310 mg m^{-3} and gas EBRT of 15 s (Fig. 3).

When the reference condition was changed as shown in Fig. 5, the TBF recovered to the reference level in a period of about 1–2 days, which is shorter than that reported in Figs. 3 and 4. That the incoming toluene concentration as illustrated in Fig. 5 was maintained constant, might contribute most to this phenomenon.

Tubular biofilters were developed to try to reduce construction and maintenance costs and to maintain the advantages of sponge materials with high porosity and high specific areas of the outermost medium bed and consequent low approaching gas velocity (Yang et al., 2003, 2004, 2008, 2011). Biomass accumulation and toluene degradation primarily occur in the filter near the inlet of waste gases (Song and Kinney, 2010; Kim and Sorial, 2010). The outermost surface of the sponge in RDBs could be considered to contribute more significantly to VOC EC due to the fact that most of the pollutants are biodegraded and most of the biomass is accumulated in this region (Yang et al., 2003). Like the RDBs with

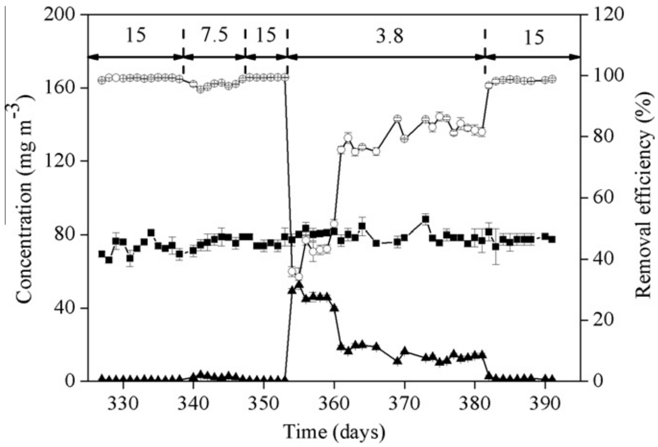

Fig. 5. Toluene removal efficiencies at various gas empty bed residence times and a constant incoming pollutant concentration. (■) Inlet toluene concentration, (▲) outlet toluene concentration, (○) toluene removal efficiency. The single data point is representing the mean value of the data, while error bars representing the overall distribution of the data.

cylindrical drums, the TBF can also provide a larger outermost surface area of the tubular material than a conventional compost biofilter or biotrickling filter with the same footprint.

With the decrease in sponge tube thickness, the volume of the filter of a TBF is greatly decreased. The height of the bed of a traditional biofilter can be over 1 m, and that of a biotrickling filter is usually around 1 m (Kennes et al., 2009; van Groenestijn and Kraakman, 2005; Yang et al., 2010). Vinage and von Rohr (2003a, 2003b) developed a modified rotating biological contactor (RBC) containing 20 biofilm support discs with a diameter of 40 cm. For the RDBs packed with polyurethane sponges, the total thicknesses of the wrapped filter medium layer were 11.4 and 14.96 cm for single-layer and multi-layer RDBs, respectively (Yang et al., 2003, 2004, 2008).

However, the polyurethane sponge tube in the TBF in this study was only 3 cm thick. Therefore, the footprint for TBFs could be significantly reduced.

This study confirmed that the TBF could effectively treat a waste gas with middle to low toluene concentrations even at short gas EBRTs of 3.8–7.5 s. The RDBs could achieve an even higher RE than the TBF when operated at similar organic loadings (Yang et al., 2003, 2004), but the gas EBRT of the TBF during operation was much shorter than that of the RDBs. When toluene was used as the model VOC, the RE of the modified RBC was lower than that of the TBF even at longer EBRTs (Ravi et al., 2010; Vinage and von Rohr, 2003a,b). Therefore, the TBF showed its advantages over other biofilters for the treatment of waste gases with low toluene concentration at a high gas velocity.

Throughout the 391 days of operation, no obvious excessive biomass accumulation in the filter media was observed through the Plexiglas pipe of the TBF. Excess biomass within sponges of RDBs can be removed by repeatedly squeezing the sponge media in a nutrient solution periodically to maintain a stable performance for the RDBs. Biomass control strategies were summarized by Yang et al. (2010), and the costs for construction and operating of biofilters with such strategies were often greatly increased. As for TBFs, biomass control strategies were not necessary due to its much larger area of the outermost bed. Therefore, low cost is another advantage of TBFs.

4. Conclusions

A tubular biofilter consisting of a closed chamber, a module polyurethane sponge tube and a nutrient solution distributor could be successfully started in 7 weeks for toluene removal. The RE decreased when increased OLs but did not decline significantly when reduced gas EBRTs. High ECs at high organic loading rates could be obtained under low pollutant concentrations. Excessive biomass accumulation was singularly occurred on the TBF. The TBF is suitable for treating waste gases with low toluene concentrations even at high flow rates over long periods of operation, reduces the footprint of gas biofilters, and overcomes the deterioration in removal observed with traditional biofilters.

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