

Removal of NH_3 , H_2S and Toluene by Biofilters Packed with Rock Wool-Compost Media

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Abstract: Biofiltration is carried out in packing materials where attached microorganisms degrade the passing contaminant. In this work, a new packing material was introduced and evaluated for the treatment of malodorous gases-ammonia and hydrogen sulfide and a volatile organic compound-toluene in three biofilters, BF1, BF2, and BF3, respectively. The new composite rock wool-compost media consisted of rock wool and compost in 70:30 weight ratios. Above 95 % removal efficiencies were obtained as the inlet concentrations of ammonia, hydrogen sulfide and toluene were increased up to 155, 150 and 260 ppm_v, respectively in the biofilters at an empty bed residence time of ~ 65 s. The removal efficiencies in the hydrogen sulfide (BF2) and toluene (BF3) biofilters immediately decreased when the frequency of water irrigation was reduced. However, hydrogen sulfide removal in the BF2 could be improved in some extent by maintaining pH of above 5. Parameters such as pH, microbial count, and pressure drop along the biofilters were also evaluated. The highest elimination capacities obtained from the study were 6.4 g- $\text{NH}_3/\text{m}^3/\text{h}$, 12.1 g- $\text{H}_2\text{S}/\text{m}^3/\text{h}$, and 57.6 g-toluene/ m^3/h , indicating that the rock wool-compost media can be suitably and effectively applied for biofiltration.

Keywords: biofilter, rock wool, compost, ammonia, hydrogen sulfide, toluene

Introduction

Ammonia (NH_3) and hydrogen sulfide (H_2S) are the most prevalent malodorous compounds that can be produced from petrochemical plants, food preparation, paper manufacturing, sewage and wastewater treatment plants, composting works, and livestock farms. These toxic, colorless gases have strong repellent and offensive odors with thresholds of 1.1 and 37 ppb for hydrogen sulfide and ammonia, respectively [1,2]. On the other hand, volatile organic compounds like toluene, for instance, can be emitted extensively when used as solvent and in the production of resins, plastics, explosives, agrochemicals and pharmaceuticals [3]. These compounds are often emitted in significant concentration posing both olfactory nuisance and health-related problems. An effective waste gas control technology for these compounds is biofiltration.

In comparison to other physical and chemical methods like incineration, wet scrubbing and adsorption, biofiltration has significant economical and operational advantages [4,5]. It requires low maintenance, has appropriate applicability for large gas volume of complex yet easily degradable compounds, produces harmless by-products and usually has above 90 % removal efficiency. The process simply utilizes microorganisms fixed to support media to break down the contaminants that are transferred from an air stream to the biofilter media [6].

The selection of suitable packing material is an important factor to achieve high removal efficiencies and sustain effective biofilter performance. Packing materials may be organic or inorganic. Organic media such as compost and peat have high removal efficiency but may have problems of high pressure drop and bed clogging, compaction, acidification and short life. On the other hand, inorganic materials such as ceramics and polypropylene rings usually have good mechanical properties but are limited by insufficient or no water content and

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Table 1. Comparison of Media Properties

Packing material	Bulk density (g/mL)	True density (g/mL)	Porosity (%)	Water holding capacity (g-H ₂ O/g-media)	pH	Reference
Rock wool-compost	0.35	0.98	64.1	0.72	7.38	This study
Rock wool	0.20	1.26	84.2	-	7.15	[8]
Wood chip	0.31	0.65	52.3	-	6.68	[8]
Compost	0.40	1.12	64.3	-	7.05	[8]
Compost	0.51	1.48	65.2	-	8.90	[9]
Porous ceramics	0.56	-	-	0.85	9.63	[10]
Coarse bark + zeolite	0.74	-	52.0	0.51	5.30	[11]
Yellow-gram stems	0.24	-	55.0	-	-	[12]
Lava rock	0.63	1.04	-	0.38	8.25	[13]
Lava rock	0.63	1.67	62.5	-	-	[14]
Expanded clay	0.58	1.05	44.4	-	-	[14]

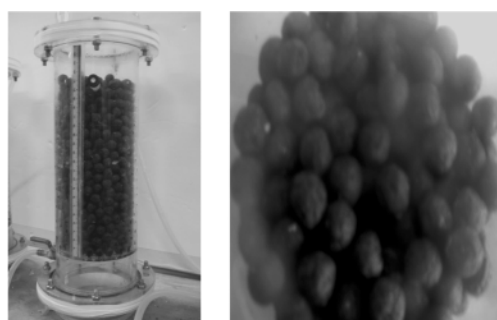
nutrients for microorganisms [4]. Fibrous materials have also been investigated as filter bed for biofiltration process [5,7,8]. Chan, in particular, made use of rock wool in treating restaurant emissions [7]. Rock wool is a man-made mineral fiber that is commonly used for insulation purpose and as hydroponics' substrate. It offers several advantages as a packing material including: (i) high water holding capacity; (ii) high porosity; (iii) large surface area; (iv) high chemical persistence; (v) low density; (vi) low cost; and (vii) a large buffering capacity. In a previous study, a biofilter with rock wool packing showed high hydrogen sulfide removal efficiencies [8]. However, the tendency of rock wool to compact easily when wetted was a disadvantage. Also, nutrients were needed to be added to rock wool to sustain viability of the attached microorganisms.

In this study, the potential of rock wool as packing material for biofilter was further studied by developing a composite rock wool-compost media. The main objective of this work was to evaluate the feasibility and performance of biofilters packed with this media in treating waste air streams contaminated with ammonia, hydrogen sulfide and toluene as single gas. The study also investigated the significance of moisture addition to biofilter performance.

Materials and Methods

Packing Material

Food waste compost, initially screened between 1.19 and 2.00 mm sieve openings, was mixed with the fibrous rock wool (UR Company, Korea) in weight ratio of 70:30 rock wool to compost. It was mixed with activated carbon (Charcoal activated powder, DSP Grade, Duksan Chemicals, Korea) at 2.5 % (w/w) to increase adsorption capacity, and was wetted with water for thorough mixing. It was blended with an optimized proportion of

**Figure 1.** Rock wool-compost media.

organic and inorganic binding solutions. The mixture was molded into ball-shape pellets of 0.8~1.0 cm diameter which were then placed in a drying oven at 60 °C for 4 h. Figure 1 shows the rock wool-compost media. A comparison of physical properties with other media is shown in Table 1. The rock wool-compost media is comparably light and has good porosity, water holding capacity and suitable pH for microbial viability.

Microbial Seeding

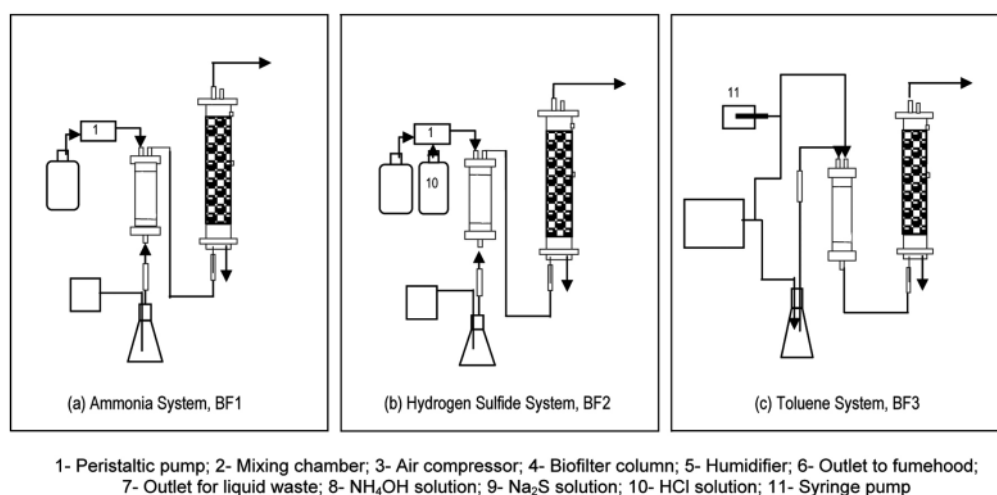
Table 2 lists the composition of the mineral solutions in which the specific strains for inoculation were grown. The strains were previously isolated from activated sludge taken from Yongin wastewater treatment plant in Korea. For the ammonia-and sulfur-oxidizing strains (AMM and *Pseudomonas sp.* SUL4, respectively), cultivation was performed at 28 °C and 150 rpm for 3 days. In the case of the toluene-degrading strain (*Bacillus sp.* TOL 1), incubation period was 5 days in a flask sealed with Teflon-coated silicon plug to prevent toluene loss in the gas phase. The culture media were sprayed onto the rock wool-compost packing media which were autoclaved at 121 °C for 15 min prior to seeding.

Experimental Setup and Operating Conditions

The schematic diagram of the three biofilter systems is

Table 2. Mineral Medium Composition for Cultivation of Different Strains

NH ₃ -oxidizing (AMM strain)		Sulfur-oxidizing (<i>Pseudomonas sp.</i> SUL4)		Toluene-degrading (<i>Bacillus sp.</i> TOL1)	
Na ₂ HPO ₄	1.0 g/L	KH ₂ PO ₄	2.0 g/L	KH ₂ PO ₄	5.0 g/L
CH ₃ COONa	1.0 g/L	K ₂ HPO ₄	2.0 g/L	K ₂ HPO ₄	4.5 g/L
KH ₂ PO ₄	0.3 g/L	NH ₄ Cl	0.4 g/L	(NH ₄) ₂ SO ₄	2.0 g/L
NH ₄ Cl	26.8 g/L	MgCl ₂ · 6H ₂ O	0.2 g/L	Mg SO ₄ · 7H ₂ O	0.34 g/L
Yeast extract	5.0 g/L	FeCl ₃ · 6H ₂ O	0.02 g/L	Trace elements	200 µL/L
Deionized water	fill to 1L	Na ₂ S ₂ O ₃ · 5H ₂ O	8.0 g/L	Deionized water	fill to 1L
		Yeast extract	5.0 g/L	300 ppm toluene ^a	
		Deionized water	fill to 1L		

^a Add after autoclaving**Figure 2.** Schematic diagram of the biofilter for single odorous gas removal.

shown in Figure 2. The ammonia, hydrogen sulfide and toluene biofilters were designated as BF1, BF2 and BF3, respectively. Each column has diameter and height of 10 and 30 cm, respectively. In Figure 2(a), ammonia solution (NH₄OH) was delivered by a peristaltic pump into a mixing chamber where humidified air was directed countercurrently. The contaminated air stream from the mixing chamber was directed into the biofilter column via an air flow meter in an upflow mode. Hydrogen sulfide, on the other hand, was supplied by the reaction of disodium sulfide (Na₂S) and hydrochloric acid (HCl) solutions as shown in Figure 2(b). The toluene-contaminated stream was produced as illustrated in Figure 2(c). Toluene (Sigma-Aldrich, 99.8 % HPLC Grade) was introduced by syringe pump (Model 200 KD Scientific, USA) into a T-type stainless connector where a low flow air stream was flowing and directed towards the mixing chamber. The empty bed residence time (EBRT) was set to ~65s. Initial bed temperature and moisture content were 26 °C and 50 %, respectively. Water irrigation was performed daily from day 1 to 35 by spraying 100 mL of water on top of the biofilters. From day 36 to 54, the frequency of water irrigation was reduced to every other

day. On specified days, basic irrigation water was prepared by adding 25 mL of 0.33 N NaOH solution to 75 mL water, and sprayed on top of the BF2.

Analytical Methods

Liquid impingement method using an improvised ammonia sampling train was initially performed for the analysis of ammonia gas. Outlet air from the BF1 was directed into a glass impinger containing 100 mL of 0.1 N H₂SO₄ solution for 1 h. This solution was transferred into a 200 mL volumetric flask and was filled to mark with deionized water. The ammonia concentration was determined spectrophotometrically by Bran+Luebbe Automatic Analyzer at 660 nm. Hydrogen sulfide concentration in the BF2 was measured using an automatic gas monitor (Multi-RAE PLUS PGM-50, USA) with 0-100 ppm_v detection range. For higher ammonia and hydrogen sulfide concentrations, gas detection tubes (Gastec, Tokyo, Japan) with 0 ~ 200 ppm_v measurable range were used. Toluene concentration from inlet and outlet ports of the BF3 was determined by a gas chromatograph equipped with a flame ionization detector (HP 6890 Series GC-FID System). The GC carrier gas was nitrogen

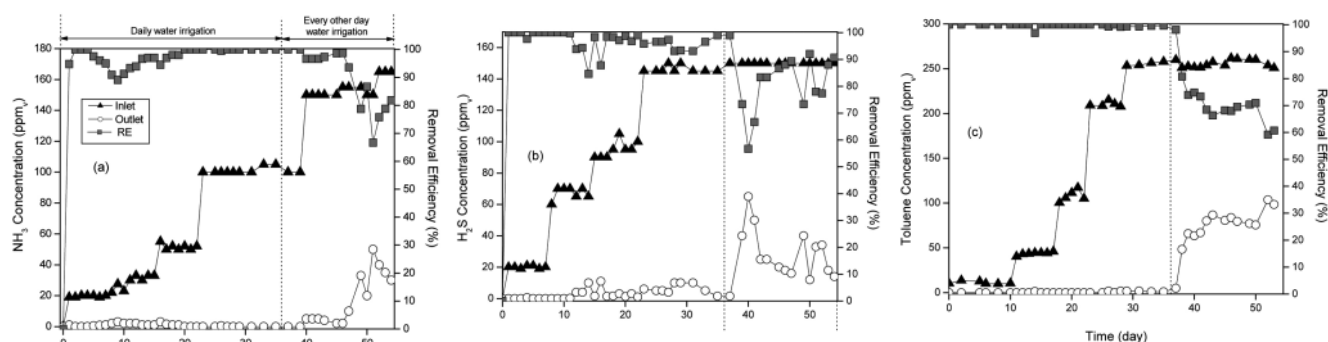


Figure 3. Biofilter response to increasing contaminant concentration: (a) ammonia, BF1; (b) hydrogen sulfide, BF2; and (c) toluene, BF3.

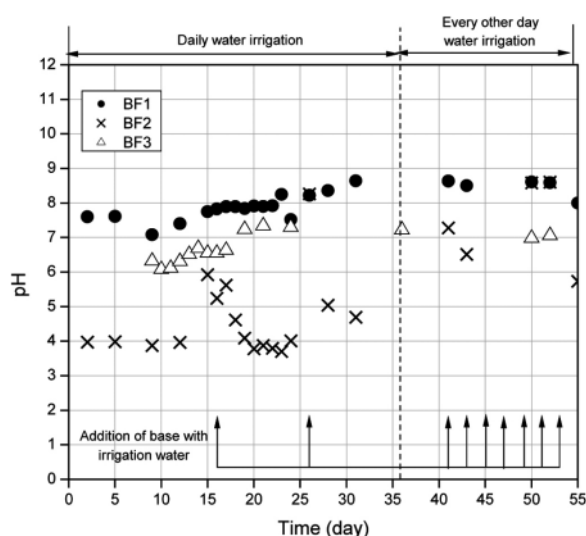


Figure 4. Variation in pH of percolated liquid from the biofilters.

and the operational conditions were: inlet temperature, 200 °C; initial oven temperature, 80 °C; final oven temperature, 150 °C; oven ramp, 10 °C/min; oven post-run temperature, 200 °C; and detector temperature, 250 °C. Gas sample volume was 500 μ L and toluene retention time was at 4.97 min.

Percolated liquid in the column was collected whenever available. The pH of the liquid was measured with a digital pH meter (ThermoOrion Model 250A+). After the necessary dilution of percolated liquid, the sulfate ion concentration was determined by ion chromatography (Waters). The moisture content of biofilter media samples taken regularly from each column was determined according to APHA standard methods [15]. Microbial count in the sample media was estimated by the number of colony forming units per gram of medium (CFU/g_{medium}) following the SSSA method for recovery and enumeration of viable bacteria [16]. Pressure drop along the biofilter beds was determined using a digital manometer (Dwyer Series 477, USA).

Results and Discussion

Ammonia, Hydrogen sulfide and Toluene Removal Efficiencies

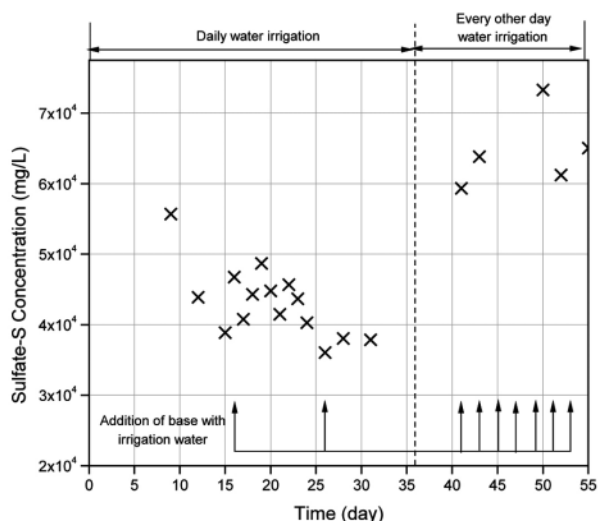
The performance of the biofilters with rock wool-compost packing material for the removal of ammonia (BF1), hydrogen sulfide (BF2) and toluene (BF3) are shown in Figure 3. The responses of the biofilters to (i) increases in pollutant concentration and (ii) reduction of water supply frequency were evaluated in terms of removal efficiency (RE). Removal efficiency refers to the fraction of the pollutant removed by the biofilter, expressed in Equation (1) where C_{Gi} = inlet concentration (ppm_v, g/m³); C_{Go} = outlet concentration (ppm_v, g/m³) [4].

$$RE = \left(\frac{C_{Gi} - C_{Go}}{C_{Gi}} \right) \times 100 \quad (1)$$

The initial inlet concentrations of ammonia and hydrogen sulfide in the BF1 and BF2, respectively, were ~ 20 ppm_v while toluene concentration were ~ 10 ppm_v in the BF3. This corresponded to initial loading rates of 0.79 g-NH₃/m³/h, 1.62 g-H₂S/m³/h and 2.16 g-toluene/m³/h. Initial high removal efficiencies (~ 100 %) obtained implied that the inoculated microorganisms in each biofilter were easily acclimated to the low inlet concentration of target pollutants. As the concentrations of ammonia and hydrogen sulfide were increased stepwise in the BF1 and BF2, respectively, gradual decreases in the removal efficiencies were observed. The removal decrease in the BF1 was probably due to microbial acclimation as the pollutant concentration was increased to the next concentration level. The removal efficiency of the BF1 gradually increased again from day 10 and achieved stable and complete removal from day 20 to 39 at 100 ppm_v inlet ammonia concentration. In the case of the BF2, the removal efficiency substantially decreased to about 85 % on day 14 at inlet hydrogen sulfide concentration of around 90 ppm_v. Analysis of the percolated liquid from

Table 3. Moisture Content and Microbial Count of Rock Wool-Compost Media in the Biofilters

Moisture Content (%)				Microbial Count (CFU/g _{media})				
Day	BF1	BF2	BF3	Day	BF1	BF2	Day	BF3
1	50.2	50.5	50.1	1	2.98E6	2.30E6	1	3.99E6
8	46.4	49.0	50.0	8	4.05E6	4.03E6	9	1.51E7
16	50.3	51.4	50.9	16	2.01E7	1.29E7	18	7.51E8
24	50.2	53.4	51.4	24	1.50E8	9.93E7	38	3.57E8
45	25.0	30.4	30.0	45	1.83E8	4.93E8		

**Figure 5.** Sulfate-S concentration of drained liquid (leachate) from H₂S column.

the BF2 taken when available showed lower pH values relative to those of the BF1 and BF3 (Figure 4). This resulted from the production of sulfates when hydrogen sulfide was oxidized as shown in Figure 5. Although *Pseudomonas sp.* SUL4 in the BF2 acclimated easily at lower hydrogen sulfide concentration and probably adjusted to low pH (~ 4) or acidic environment [8], the sudden increase in inlet concentration to 90 ppm_v resulted in the decrease in removal rate corresponding to low sulfate production as measured on day 15. Addition of base with the irrigation water (0.33 N NaOH, day 16) increased the pH (5 \sim 6), and regained the high hydrogen sulfide removal of above 98 % on day 17, resulting in the increased of the sulfate production in the percolated liquid from the BF2. When the inlet hydrogen sulfide concentration was increased to 150 ppm_v on day 23, the removal started to decrease but to only about 93 % suggesting that *Pseudomonas sp.* SUL4 could tolerate the acidic environment. Re-addition of base (day 26) in the BF2 gradually increased the removal rate to 99 % obtained on day 35. It apparently indicates that *Pseudomonas sp.* SUL4 could be more active at pH of above 4, resulting in the improvement of removal efficiency even at high loading of hydrogen sulfide. On the other hand, consistent performance was observed in the BF3 attain-

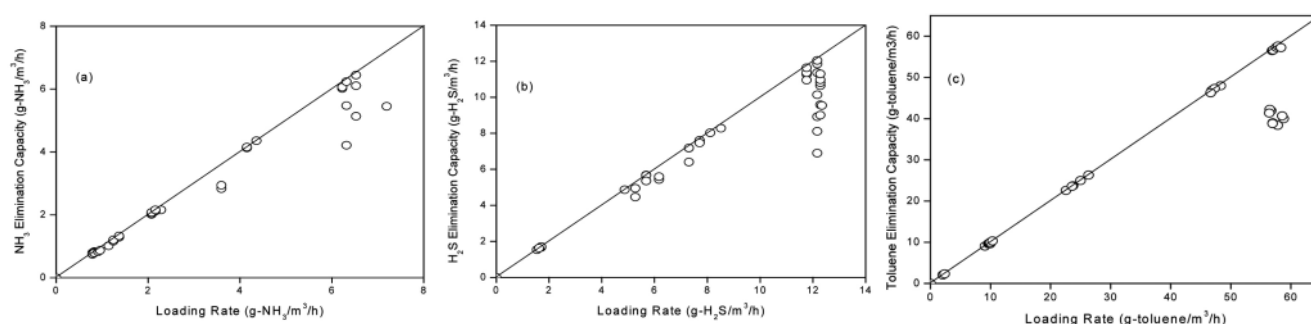
ing almost complete removal of toluene up to inlet concentration of 260 ppm_v on day 35.

During the latter part of the experiment (day 36 \sim 54), decrease in removal efficiencies were observed from the BF1, BF2 and BF3 as result of the reduction in the frequency of water irrigation from daily to every other day. As shown in Table 3, the bed moisture content in all three biofilters decreased to around 25 \sim 30 % on day 45 from the initial 50 % during the first phase of the run. The optimum range of moisture content suitable for microorganisms to carry out normal metabolic activities is from 40 to 60 % [18]. Therefore, the reduction in the removal efficiencies may be due to the low moisture content. However, the BF1 initially maintained high removal efficiencies of about 97 % during the first 10 days after reducing the water irrigation frequency, even after the inlet concentration was increased from 100 to 155 ppm_v. Table 3 shows that there was no significant change between the estimated microbial counts of the sampled BF1 media on day 24 and 45, which indicates the tolerance of the ammonia-oxidizing bacteria to some degree of low moisture content. However, after day 46, the removal efficiency started to decrease and became unstable possibly due to slower growth rate and/or decreasing activity of the BF1 microorganisms under a low moisture content of 25 %.

BF2 and BF3, on the other hand, showed apparent sensitivity to moisture content as evident from the decrease in their removal efficiencies to 55 (day 40) and 68 % (day 42), respectively. In the BF2, the water in the media bed served not only to provide moisture for microorganism but also contributes to the media's buffer capacity by diluting the sulfate concentration and minimizing the decrease in pH that may decrease microbial activity. The pH and sulfate concentration in the percolated liquid from the BF2 were not measured during the first 5 days after reduction of irrigation frequency, due to a slow liquid percolation. However, it could be presumed that pH and sulfate concentration might be lower and higher, respectively, compared to those obtained during daily water irrigation. Addition of 0.33 N NaOH with the irrigation water from day 41 to 53 gradually increased the H₂S removal efficiency up to about 90 % on day 47. According to Chung and coworkers [17], the optimum

Table 4. Comparison of Elimination Capacities for NH₃, H₂S and Toluene

Removal rate	Packing material	Reference
6.4 g-NH ₃ /m ³ /h	Rock wool-compost	This study
1.0 g-NH ₃ /m ³ /h	Compost, bark mulch, wood chips	[19]
7.6 g-NH ₃ /m ³ /h	Calcinated cristobalite	[5]
12.0 g-NH ₃ /m ³ /h	Compost	[20]
10.1 g-NH ₃ /m ³ /h	Granulated sludge	[20]
10.6 g-NH ₃ /m ³ /h	Compost, oyster shells, perlite	[21]
12.1 g-H ₂ S/m ³ /h	Rock wool-compost	This study
2.4 g-H ₂ S/m ³ /h	Compost, bark mulch, wood chips	[19]
10.0 g-H ₂ S/m ³ /h	Peat	[22]
15.0 g-H ₂ S/m ³ /h	Compost	[23]
10-45 g-H ₂ S/m ³ /h	Pig manure, sawdust	[24]
57.6 g-toluene/m ³ /h	Rock wool-compost	This study
28.1 g-toluene/m ³ /h	Compost with perlite	[25]
45-55 g-toluene/m ³ /h	Compost with perlite	[26]
45, 90, 180 g-toluene/m ³ /h	Compost-based media pellet	[27]

**Figure 6.** Elimination capacities of the biofilters: (a) ammonia, BF1; (b) hydrogen sulfide, BF2; and (c) toluene, BF3.

pH for the removal of hydrogen sulfide by *Pseudomonas putida* CH11 in biofilter was in the range of 6~8. Figure 4 indicates that with regular pH adjustment (0.33 N NaOH), the media became less acidic than the pH 4 even at high sulfate production (Figure 5). This might have promoted microbial growth as shown by the increased in the microbial count of *Pseudomonas sp.* SUL4 in the BF2 (Table 3) from day 24 to day 45 even at low moisture content of only about 30 %. However, in terms of hydrogen sulfide removal, the low moisture content with the reduction of water irrigation demonstrated unstable removal efficiency in the range of 70~90 %. Therefore, hydrogen sulfide removal efficiency could be improved to some extent by pH adjustment but it was affected by moisture content as well.

In the case of the BF3, there was a significant increase in the microbial count of *Bacillus sp.* TOL1 as shown in Table 3 when inlet toluene concentration was around 105 ppm_v on day 18, indicating that high concentration induced higher biodegradation activity for the microor-

ganisms. However, with environmental stress brought by insufficient moisture on the latter phase of the study, the activity of the *Bacillus sp.* TOL1 might decrease and thus became more susceptible to inhibition at higher toluene loading (250 ppm_v). This was evident from the significant decrease in microbial count in BF3 media samples on day 38 (Table 3). Contrary to the BF1 and BF2, the removal efficiency of the BF3 for toluene declined further to 61 % which may be indicative of nutrient limitation in the system which is a more common occurrence in biofiltration of organic compound.

Ammonia, Hydrogen Sulfide and Toluene Elimination Capacities

The dependence of the pollutant elimination capacity with loading rate is shown in Figure 6. Elimination capacity refers to the mass of contaminant degraded per unit volume of the filter material per unit time, defined by Equation (2) where C_{Gi} = inlet concentration (ppm_v, g/m³); C_{Go} = outlet concentration (ppm_v, g/m³); V_f = filter

bed volume (m³); and Q = air flow rate (m³/h) [4].

$$EC = \left(\frac{C_{Gi} - C_{Go}}{V_f} \right) \times Q \quad (2)$$

The highest elimination capacities obtained in the BF1, BF2, and BF3 were 6.4 g-NH₃/m³/h, 12.1 g-H₂S/m³/h and 57.6 g-toluene/m³/h, respectively, obtained at 99 % removal efficiencies. The points below the 100 % RE diagonal line were those obtained when the rate of water addition was decreased. These results imply that with proper water irrigation control, the rock wool-compost biofilter system has potential for treating higher loadings than the applied in this study. Nonetheless, the values obtained were comparable to some EC values reported in literature as listed in Table 4. Contrary to the single strain inoculation used in this study, utilization of microbial consortium [5] or adapted microbial source (like sludge or compost) [20,23,24,27] in those studies with significantly higher EC implies that a more heterogeneous microbial population may be necessary to treat high loadings of ammonia, hydrogen sulfide or toluene.

Pressure drop

The pressure drop or head loss along the biofilter is an important operating parameter and an indirect measure of bed permeability. The pressure differences along the biofilter columns of the three systems generally increased with time but only to about 10.0 mm H₂O/m bed (data not shown). These were lower than the reported 10~41 mm H₂O/m bed and 5~30 mm H₂O/m bed pressure drops for compost and granulated sludge-packed biofilters, respectively, by Chen and coworkers [20]. The rock wool-compost media did not compact due to its firm structure and relatively low density. The spherical shape could also result in better air flow and distribution along the columns.

Conclusion

Biofiltration experiments were carried out to evaluate the capacity of rock wool-compost media in removing representative odorous and volatile organic compounds from an air stream. From the study, it was found out that the media had good removal efficiencies towards ammonia, hydrogen sulfide and toluene. High removal efficiencies of 95~100 % were achieved as the inlet concentrations of ammonia, hydrogen sulfide and toluene were increased up to 155, 150, and 260 ppm_v, respectively. The rock wool-compost media were not subjected to high pressure drop or to significant bed compaction. The pressure drop was ≤ 10 mm H₂O/m bed for the media during the study. Decline in removal efficiencies were found to

be at days when moisture content dropped to as much as 30 % particularly in the hydrogen sulfide and toluene biofilters. With proper water control (to about 50 % moisture content), the biofilters may have the potential to treat higher loadings than the applied in this study. Nonetheless, the highest elimination capacities obtained were 6.4 g-NH₃/m³/h, 12.1 g-H₂S/m³/h, and 57.6 g-toluene/m³/h for the BF1, BF2, and BF3, respectively. Results showed that the rock wool-compost media offers viable application for biofilter use in treating malodorous and volatile organic compounds.

Acknowledgments

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