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Review Article

Bio-filters for the Treatment of VOCs and Odors - A Review

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ABSTRACT

Excessive amounts of volatile organic compounds (VOCs) and odorants discharged into the environment are highly dangerous to human health as well as to ecological systems. Biological treatments of waste gas streams, called biofiltration, containing VOCs and odorous compounds has gained much attention because biofilters are more cost effective and environmentally friendly than conventional air pollution control technologies. This review provides an overview of biotrickling filtration, which is a type of biofiltration including continuous trickled-water flow inside filter media, for VOC and odor abatement. The configuration, design, cost effectiveness, removal capacity and environmental impact of this techniques and the future research and development needs in this area are all considered.

Key words: Biofilter, VOC, Odor, Biotrickling filtration, Biological treatment

1. INTRODUCTION

Various types of anthropogenic activities including kraft pulping, animal rendering, and wastewater treating/composting are sources of reduced sulfur compounds (RSCs: e.g., hydrogen sulphide (H_2S), dimethyl sulphide (DMS), methanethiol (MT), and dimethyl disulphide) (Syed *et al*., 2006; Sercu *et al*., 2005). These compounds are well known for their unpleasant smell and low odor thresholds (e.g., DMS 1.2 ppb and MT 2.4 ppb) (Verschueren, 2001). With ever-increasing global population and industrialization levels, the demand for sustainable VOC odor control technologies thus becomes more important in order to ensure nuisance-free air in and around the emission sources. However, in order to establish a better control strategy for VOCs, it is imperative to accurately characterize their atmospheric behavior and emissions.

It is widely acknowledged that VOCs can play a crucial role in the formation of surface ozone as well as secondary organic aerosols (SOAs) (Yue *et al*., 2017; Kroll and Seinfeld, 2008; Seinfeld and Pandis, 2006). These properties of VOCs have been brought into the lime light of the recent atmospheric research as most urban regions around the world are facing severe pollution associated with their production and emission (Sun *et al*., 2014). In this respect, a proper VOC emission inventory is the need of the hour, but current VOC emission inventories suffer from large uncertainties (e.g., as high as 100%) (Zhong *et al*., 2017). Zheng *et al*. (2017) attempted to quantitatively assess the industrial VOC emissions in China. These autors presented a sectoral VOC emission contributions with four processes using the spatial distribution of VOCs using GIS based emission factors and related data inventory (Fig. 1(a)). These authors pointed out that the VOC-containing products are the fastest growing sector towards the emission of VOCs with an average annual growth rate of 57.2%. They have also projected the average industrial VOC emission from 2000 to 2050 (Fig. 1(b)) using the currently available emission inventory. Accordingly, surface coating industries are also known to be a major culprit for the atmospheric emission of VOCs. Fig. 1(c) provides a comparison of VOCs emission in autocoating industry. Interestingly, VOCs released from the vehicle evaporative emissions also contribute significantly to photochemical air pollution, with toluene, isopentane/n-pentane, and 2,2,4-trimethylpentane as the dominant components (Yue *et al*., 2017).

In recent years, the sludge generated due to the mechanical, biological, and chemical treatment of wastewater has often been viewed as a useful raw

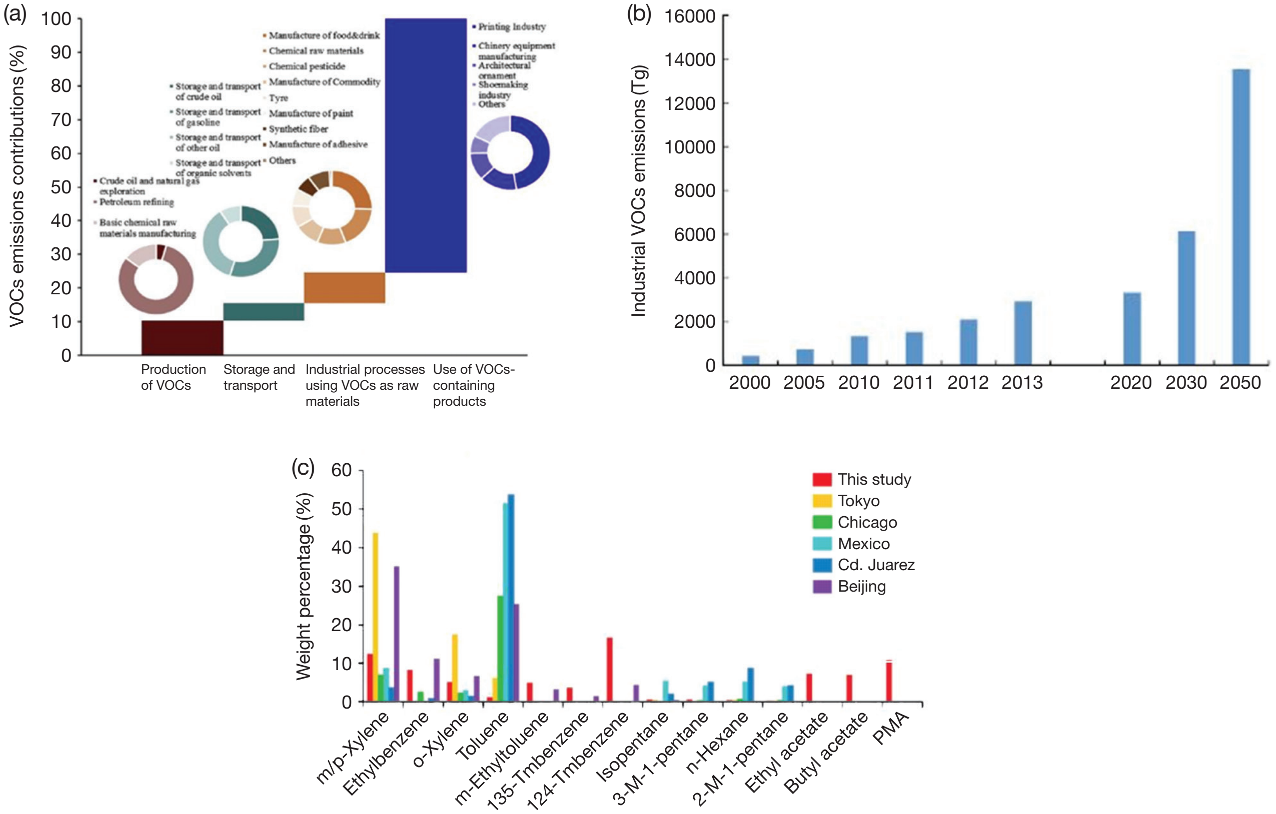

Fig. 1. (a) Sectoral VOC emission contributions with four processes (2013), (b) Industrial VOC emissions from 2000 to 2050, and (c) Comparison of VOCs emissions in auto coating industry [Adapted from Zheng *et al.* (2017) and Zhong *et al.* (2017)].

material to be composted owing to its high content of organic matter, nutrients, and other micro elements (Kosobucki *et al.*, 2000). However, sludge treartment facilities have faced a wide social rejection due to the emission of unpleaseant odors during composting (e.g., VOCs and ammonia) (Komilis *et al.*, 2004; Goldstein, 2002). The introduction of more stringent environmental regulations implemented by government agencies has forced polluters to adopt more effective air pollution treatments (Giri *et al.*, 2010). As a consequence, development of biological techniques which are more environmental friendly and have higher pollution removal efficiency, has been gaining great attention as it may overcome various limitations in conventional techniques (Munoz *et al.*, 2012). For instance, the traditional physicochemical processes such as incineration, employed for treating VOCs and other organic sulfur compounds require relatively high energy with high chemical use and disposal costs (Wani *et al.*, 2008).

Biofiltration relies on aerobic microorganisms immobilized on solid particles in a bed media such as peat, compost, wood chips, or polyurethane foam packed in a column (Kumar *et al.*, 2011). The biofilter is generally a fixed-film bioreactor that provides a large contact area between the gas stream and the microorganisms attached to porous media surface. As the polluted gas stream passes through the filter media, VOCs or odorous compounds in the gas are partitioned into the biofilm where biological oxidation occurs under aerobic conditions (Kumar *et al.*, 2011). The main advantage of biofiltration is that the pollutants are converted into harmless end-products. Relatively low costs and excellent operational stability are also recognized as the advantages of biofiltration approaches (Rene *et al.*, 2012).

Although various configurations exist, the main types of conventional gas-stream biological reactors include biofilters, biotrickling filters, and bioscrubbers. Among the recently developed reactors, membrane reactors have been used for the abatement of VOC and odor (Kumar *et al.*, 2008a, b; Shareefdeen and Singh, 2005). Although the basic mechanisms of pollutant removal are similar to each other, differences exist in water flow types inside packing media. Fig. 2 presents an overview of the broad range of pollutants and applications for which the biological techniques are being used at present.

In this review, we focus particularly on the biotrickling filter (Fig. 3). The continuous supply of trickled-water provides a suitable treatment of hydrophilic VOCs. Nevertheless, as intimate contact exists between microorganisms and the pollutant stream, solubility restrictions are less stringent than for other biofilters such as bioscrubbers (e.g., dimensionless Henry's law coefficient ($[C]_a/[C]_g$) <0.1, where $[C]_a$ and $[C]_g$ are the aqueous-phase and gas-phase concentration of the species respectively) (Cox and Deshusses, 1999; Van Groenestijn and Hesselink, 1993). Also the continuous infusion (Fig. 3) of the nutrient solution facilitates control of the microbial activities and other operating parameters such as pH buffering.

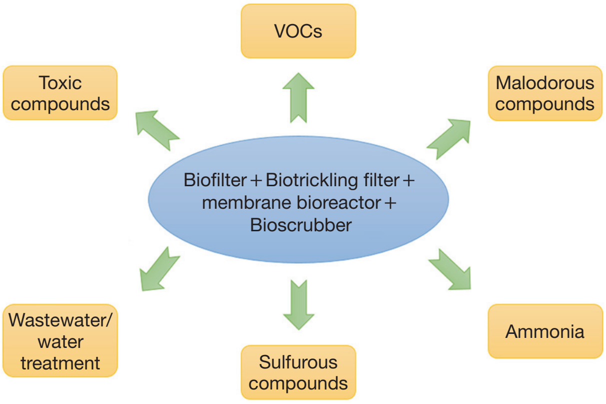

Fig. 2. Application of biological techniques for the mitigation of various pollutants.

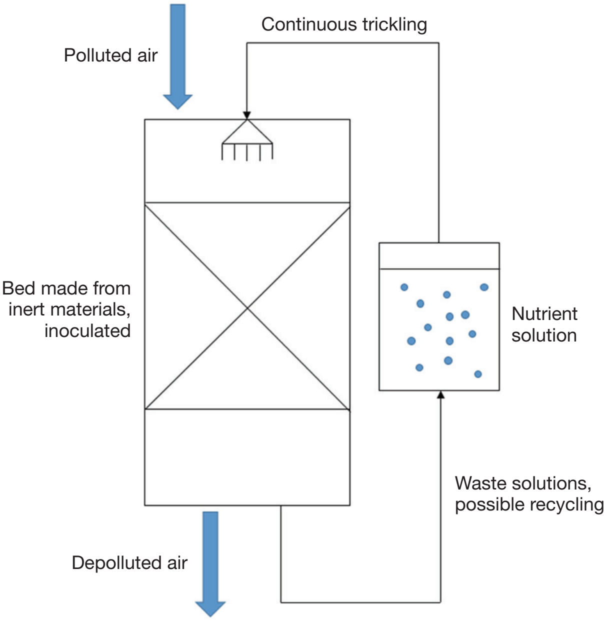

Fig. 3. Schematics of a biotrickling filter unit [Adapted from Delhomenie and Heitz (2005)].

This review has been organized to provide an overview of the biotrickling filter employed for the control of VOCs and odors, its merits and drawbacks, its important operational parameters, and future research and development needs in this area.

2. BIOLOGICAL GAS TREATMENT TECHNIQUES

The biological techniques for the treatment of VOCs include biofilters, biotrickling filters, bioscrubbers, and membrane bioreactors. In these methods, the pollutants are biologically degraded by aerobic microorganisms to stable end products like CO_2, H_2O, sulfate, microbial biomass, etc. (Delhomenie and Heitz, 2005; Kim and Deshusses, 2005) (Figs 4-6 show the schematics of a Membrane Bioreactor, a Biofilter and a Bioscrubber, respectively).

2.1 Biofilters

Biofiltration is the most typical type of biological air pollution control process, initially developed in the late 1970s (Leson and Winer, 1991). It is now emerging as a sustainable alternative for the treatment of air contaminated with VOCs and odorous compounds. In biofiltration, the polluted air is forced through a bed of packing media covered with a layer of aerobic microorganisms. The microorganisms are immobilized on the surface of the packing media. The primary role of the packing material in biofilter media bed is to support the microbial community through the attachment of microbial biofilm to the surface of packed media. Bohn (1992) established that an ideal biofilter bed

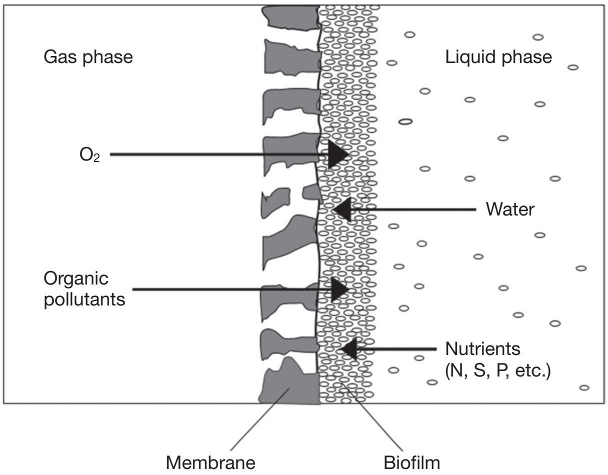

Fig. 4. Schematics of a membrane bioreactor [Adapted from Mudliar *et al.* (2010)].

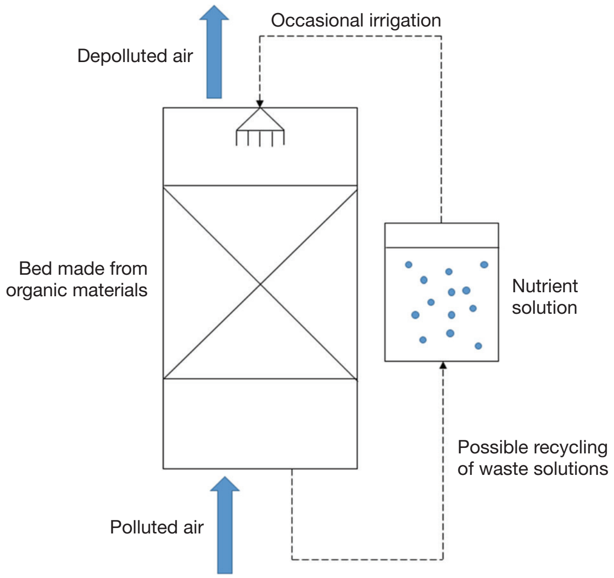

Fig. 5. Schematics of a Biofilter [Adapted from Mudliar *et al.* (2010)].

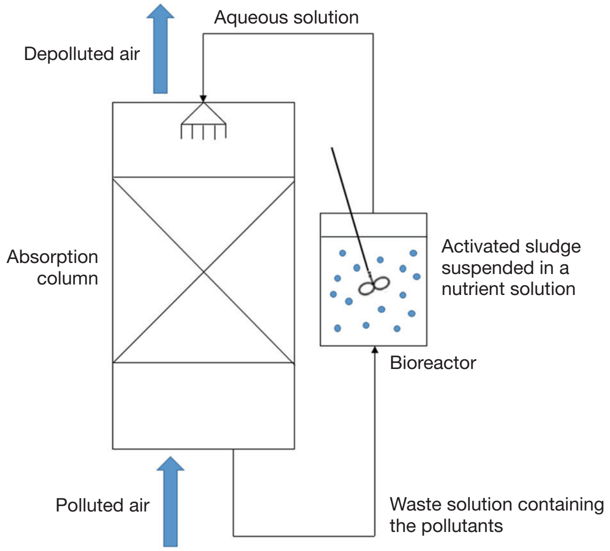

Fig. 6. Schematics of a Bioscrubber [Adapted from Mudliar *et al.* (2010)].

should have a high specific surface area ($>300\ m^{-1}$) for the proper development of the micro flora which can induce high gas-to-biofilm mass transfer. However, specific surface area as high as 1000 m^{-1} has also been reported for polyurethane-based beds (Mudliar *et al.*, 2010). High porosity and water retention capacity are also highly desirable to facilitate homogenous distribution of gas flow and avoid media drying, respectively. The widely used packing materials for biofiltration include soil, compost, and wood chips. These materials are adavantageous, as they satisfy the basic requirements stated above and are cost effective (Mudliar *et al.*, 2010). In order to avoid bed crushing and compaction and to improve many other properties such as moisture hold up and microbial growth, several authors have suggested the use of advanced packing materials with complex blending such as a mixture of compost and wood chips or compost mixed with hard plastic (Taghipour *et al.*, 2008). The pollutants are transferred from air to the water layer adhering to the bacterial growth on the media to be biologically metabolized (Upadhyay and Kumar, 2004). Biofiltration is energy efficient and cost effective while producing minimal quantities of toxic end-product. This technology has been successfully used for removing a wide range of pollutants such as VOCs, ammonia, mercaptans, and sulfurous compounds (Giri *et al.*, 2010; Galera *et al.*, 2008; Ho *et al.*, 2008). The drawbacks of this technique are excessive pressure drops and gradual accumulation of acidic by-products due to dry-out, rapid degradation, and clogging; difficulty in controlling the biological operating parameters; clogging due to the accumulation of large amount of biofilm and reduced treatment efficiency at high pollutant concentrations (Mudliar *et al.*, 2010). A typical biofilter has been observed to operate with a removal efficiency in the range of 50-95% (Park *et al.*, 2001).

The removal efficiencies of toluene were reported to exceed 80% with an inlet concentration less than 1,274 ppm using an agro waste based biofilter (Singh *et al.*, 2006). Moreover, Jaber *et al.* (2016) studied the removal of H_2S using a biofilter under extremely acidic conditions. A maximum H_2S removal capacity of 24.7 $g\ m^{-3}\ h^{-1}$ was reported for a 0.07 m^3 reactor at an inlet flow rate of 4 $m^3\ h^{-1}$ with a removal efficiency of 78% up to 360 ppm (v/v).

2.2 Bioscrubber

A bioscrubber essentially consists of a two stage unit in which absorption occurs in one stage and biodegradation by suspended microbes occurs in the other stage. Bioscrubbers are usually used for the treatment of readily soluble VOCs in the waste air stream (alcohols, ketones, etc.) having a concentration less than 5 $g\ m^{-3}$ (Kellener and Flauger, 1998). Reported removal efficiencies are in the range of 50-99% (Webster and Devinny, 1995). Bioscrubbers are stable enough to allow a better control of operating parameters. Also, they produce a lower pressure drop across the microbe suspension than other filter types. The major problem associated with bioscrubbers is the generation of excess sludge and liquid waste which over time reduces the efficiency of the process considerably.

2. 3 Membrane Bioreactors

Membrane bioreactors have been investigated by various researchers for VOC and odor abatement (Mudliar *et al*., 2010; Kumar *et al*., 2008a, b; Shareefdeen and Singh, 2005; Ergas and McGrath, 1997). They were designed as an alternative to conventional bioreactors for waste gas treatment. In a membrane bioreactor, the mass transfer of VOCs from the gas phase to a microbially active liquid phase occurs through the microporous hydrophobic hollow fiber membranes. The selective permeation of the pollutant across the membrane occurs due to the concentration difference between the gas phase and the biofilm phase, which provides the driving force according to Henry's law. The advantages of membrane bioreactors are the absence of moving parts, ease of scale-up, and the ability to vary the flow of gas and liquid independently without the problems of flooding, loading, or foaming. Disadvantages associated with membrane bioreactors are the high investment cost and possible clogging of the liquid channels due to the formation of excess biomass. Removal efficiencies have been reported in the range of 50-99% (Hartmans *et al*., 1992).

3. BIOTRICKLING FILTER

A schematic description of a typical biotrickling filter (BTF) is provided in Fig. 1 (Delhomenie and Heitz, 2005). For this filter, the gas percolates through a packed bed, which is continuously irrigated with an aqueous solution containing essential nutrients required by the bio-organisms. It was reported that neither a co- or counter-current configuration for liquid and gaseous phases has any influence on the biodegradation performance (Cox and Deshusses, 1999). Microorganisms can grow as a biofilm on the packing material of the filter bed. The biodegradation takes place within the biofilm, as the target pollutants are absorbed on the aqueous film. The filtering material of BTF should facilitate the flow of both gas and liquid through the bed and the development of the microflora while resisting crushing and compaction (Giri *et al*., 2010).

The contact between the microorganisms and the pollutants occurs after the diffusion of the pollutant in the liquid film. Hence, the liquid flow rate and the recycling rate are recognized to be critical parameters for BTF operation. Removal efficiencies for trichloroethylene have been reported in the range of 50-90% (Govind and Bishop, 1994). Likewise, removal efficiencies of H_2S were estimated in the range of 95-98% for gas streams containing H_2S in the range 0-255 ppm (v/v) (Vikromvarasiri and Pisutpaisal, 2016).

3. 1 Biotrickling Filtration Capital Costs

Capital costs for biotrickling filters vary widely according to filter size and construction materials. The required size of the biotrickling filter is a determined by such variables as air flow rate, the nature and concentration of the pollutant treated, and the required removal efficiency. The presence of corrosive gasses (e.g., H_2S) or solvent vapors is the main factor in determining construction materials (polyethylene, fiberglass, perlite, etc.) (Popoola *et al*., 2013). The cost of operating a biotrickling filter is increased by, the presence of dust or fine particles, excessively high or low temperatures, highly fluctuating pollutant concentrations, etc. Hence, before commencing reactor design and construction, a detailed analysis and characterization of the reactors input air stream needs careful consideration.

A simple relationship (Equation 1) was proposed to estimate the capital cost of a biotrickling filter based on bed volume (Deshusses and Cox, 1999). The costs include basic components (e.g., pumps and level switches) for a simple biotrickling filter constructed out of inexpensive materials. The cost estimated by Equation 1 has a ±20% accuracy for reactor volumes 5 to 1000 m^3. For more expensive materials such as stainless steel, the pre-exponent term (13,000) needs to be increased in equation 1.

$$\text{Biotrickling Filter Capital Cost (USD)} \\ = 13{,}000 \times \text{ Bed Volume}^{0.757} \tag{1}$$

The reactor volume can be determined from knowledge of the pollutant concentration, the intended removal efficiency, and the air flow rate. For the reactor capacities of 10, 100, and 1,000 m^3, the unit cost decreases substantially with increasing reactor size, viz., 7,500, 4250, and 2400 (USD m^{-3}), respectively. Equation 1 is then used to estimate the capital cost (Table 1). The final installed cost is somewhat vendor dependent. Other costs such as: land, site preparation, assessment, maintenance, operating, financing, taxes, insurance, other overheads, etc. are also needed to determine the final installation and operational costs.

3. 2 Biotrickling Filtration Operating Costs

The operating cost estimation of a biotrickling filter should include: 1) Electricity for the blower and the recycle pump along with other electrical equipment, 2) cost of the water and nutrients, 3) maintenance, 4) costs associated with biomass growth control, 5) capital costs associated with amortization (Deshusses and Cox, 1999).

Electricity for the blower is often a major fraction of the total operating expenses. In contrast, water, nutrients, and chemicals needed for the control of moisture

Table 1. Estimated capital costs, footprint and treatment capacity of biotrickling filters of various sizes[a].

Order	Bed volume (m^3)	Capital cost (Equation 1) (USD)	Approximate footprint[b] (m^2)	Approximate air flow rate[c] ($m^3\ h^{-1}$)	Reactor unit cost (USD m^{-3})
1	5	45k	1-2.5	300-3,600	9000
2	10	75k	2-5	600-7,200	7500
3	20	125k	4-10	1,200-14,400	6250
4	50	250k	10-25	3,000-36,000	5000
5	100	425k	20-50	6,000-72,000	4250
6	200	720k	40-100	12,000-144,000	3600
7	500	1.4M	100-250	30,000-360,000	2800
8	1,000	2.4M	200-500	60,000-720,000	2400

[a] Not adjusted for inflation. 45% inflation from 1999 to 2016 (http://www.bls.gov/home.htm)
[b] Estimated using a 2-5 m bed height; to convert to sq. ft. multiply by 11.
[c] Calculated using EBRT of 5 s to 1 min; to convert to cfm. Multiply by 0.59.

and pH are a relatively small fraction (10-30%) of the total operating costs. Inspection of spray nozzles for possible clogging is the most important task during maintenance, as it could lead to inadequate wetting of the bed. If the biotrickling filter is subject to clogging problems, the costs of controlling the biomass growth can be significant - up to half of the total operating costs (Deshusses and Cox, 1999). Note that there are various approaches to control biomass growth such as the use of ozone to curb biomass accumulation (Zhou *et al.*, 2016) or by protozoan predation (Cox and Deshusses, 1999). However, those methods have not yet been reliably developed for the applications at the industrial scale. Careful evaluation of the various options is recommended. Since biotrickling filter operation is relatively inexpensive, capital cost amortization will be significant compared to other costs. Assuming the lifetime of a filter plant is 10-20 years, the amortization of capital costs represents on average 20 to 40% of the total treatment cost. This stresses the importance of careful selection of materials and proper sizing to minimize the actual capital costs.

Usual operating costs range from \$0.05 to \$1.5 per 1000 m^3 of air treated excluding capital costs and increases to \$0.1 to \$3 per 1000 m^3 if capital amortization is included (Deshusses and Cox, 1999). These cost estimates are inflation dependent and in the USA, there has been 45% inflation since 1999 to 2016 (http://www.bls.gov/home.htm)). The operating costs need to be carefully considered at the planning stage based on possible applications and biotrickling filter size.

4. APPLICATIONS

A widespread application of BTF has been for the treatment of VOC and odor. This is a significant development over the use of a conventional biofilter (BF) that has generally been limited to the elimination of odorous compounds and non-chlorinated volatile organic compounds. This is due to the permanent trickling mechanism in BTF, which ensures the continuous distribution of the nutrient solution. As a result, BTF can favorably control the biological operating conditions (viz. pH). Also, BTFs are known to be capable of treating the acid degradation products of VOCs.

4.1 Application of BTFs for VOC Abatement

Lu *et al.* (2001) achieved removal efficiencies as high as 95% for a mixture of acetone (20 g m^{-3} h^{-1}) and methyl acetate (27 g m^{-3} h^{-1}) using a bench-scale biotrickling filter. The filter comprised of an acrylic cylinder packed with *phanerochaete chrysosporium* immobilized on glass beads. Clogging of the medium, the complex filter structure and operation of the biotrickling filter, were the only shortcomings reported.

The effect of low dose ozonation was also investigated to prevent excess biomass accumulation and to maintain high removal efficiencies of toluene over extended BTF operation (Zhou *et al.*, 2016). To optimize the biomass control strategy, the relative performance of five parallel BTFs was monitored at different ozone doses. The BTF was constructed from a Perspex pipe with a height of 0.95 m and an internal diameter of 9 cm. The active bed height was 48 cm with the bed volume of 3.1 L. The BTFs were packed with pelletized polyurethane foam (PUF) with a diameter of 10-15 mm. The pelletized PUF had an initial porosity of 91.0% with a specific surface area of 380 m^2 m^{-3}. The ozone-free BTF performance declined after 150 days due to excess biomass accumulation, the buildup of extracellular polymeric substances (EPS) excreta, and a decline in metabolic activity of the biofilm. An optimized dose of ozone (e.g., 5-10 mg m^{-3}, or 2.55-5.09 ppm) was sufficient to maintain stable operation (for 300 days) at a consistently high removal efficiency ($>93\%$) at 400 mg m^{-3} (106 ppm) toluene in the air; this prevented excess biomass accumulation. On the other hand, ozone above 20 mg m^{-3} (10 ppm) inhibited excessive biomass growth to prevent poor BTF performance.

The biodegradation of toluene vapor was investigated by a lab-scale biofilter impregnated with *pseudomonas putida* DK-1 (Park *et al*., 2001). Removal efficiencies in the range of 75-90% were observed for an inlet loading ranging from 250-350 g h^{-1} m^{-3}. They used a fiberglass column with an inner diameter of 50 mm and height of 200 mm. The pressure drop in the bed was 20-100 (Pa m^{-1} of packing) and had limited impact on the process efficiency. At the bottom of the reactor, a perforated sieve plate was to support the medium. The medium (wood chips) was impregnated with *pseudomonas putida* DK-1 and positioned over samples of contaminated soil. The problems associated with the filter were the difficulty in controlling the moisture and pH in order to maintain the optimum environment for the growth of degrading microorganisms and also the relatively high rates of clogging and deterioration of the medium. From these two studies. Biofilters have more drawbacks compared to biotrickling filters. Moreover, biotrickling filters have been shown to reach higher removal efficiencies for relatively low concentrations of VOCs and odorous compounds.

First ever reported laboratory scale BTF for the elimination of nitrobenzene vapors was reported by Oh and Bartha (1997). They used a stable microbial consortium enriched by sewage sludge and immobilized on dry perlite (the reactor occupied 59% (0.4 L) of the total column volume (1.5 L)). During the startup period of four weeks, the inlet nitrobenzene concentration was kept relatively low (<16 ppm) to avoid poisoning of the culture, after which high and sustained nitrobenzene elimination was observed with 80-90% degradation for inlet concentrations ranging from 100 to 300 mg m^{-3} and an empty bed gas contact time of 21 seconds. The resultant elimination capacity was of 50 g m^{-3} h^{-1} at a stream flow rate of 200 m^3 h^{-1}. This is a significant removal rate that could lead to an economically viable process. A nitrogen balance showed that 98% of the removed nitrobenzene was converted into ammonia while a small amount of nitrite was also produced.

Also, noteworthy is the study by (Sun and Wood, 1997). They immobilized a pure culture of *Burkholderia Cepacia* $PR1_{23}$, a Tn5transposon mutant of *B. cepacia* G4 that constitutively expresses the trichloroethylene (TCE) degrading enzyme and toluene orthomonooxygenase (TOM). Aerobic biodegradation of TCE only occurs through co-metabolism with the addition of a growth substrate (usually toluene, methane, propane, phenol or ammonia). This is required to induce the expression of the appropriate TCE-degrading enzyme. However, the bacterium strain *Burkholderia Cepacia* $PR1_{23}$ expresses toluene ortho-monooxygenase at a constant rate, regardless of physiological demand. This circumvents the problem of competitive inhibition of TCE oxidation by the usual inducers during the growth phase. They used glucose as a carbon and energy source and observed TCE eliminations up to 200 times higher than previously reported 90% TCE removal at an inlet loading of 2.4-100 mg TCE L^{-1} together with 95 mg toluene L^{-1} (Guo *et al*., 2001). As observed previously in other bioreactors for TCE aerobic cometabolism, rapid inactivation of the TCE-degrading enzyme by TCE breakdown products (e.g. TCE-epoxide) still remains to be resolved (Guo *et al*., 2001; McFarland *et al*., 1992).

In the past few years, much research has been done to improve the BTF technology (Valero *et al*., 2017; Zhou *et al*., 2016; Tsang *et al*., 2015). Several successful conversions of full-scale chemical scrubbers to biotrickling filters have been demonstrated (Gabriel and Deshusses, 2003; Kraakman, 2003, 2001).

4. 2 Application of BTFs for Odor Control

There are over 16,000 publicly owned treatment works (POTW) in the United States serving 75 percent of the total population (U.S. DHS, 2016). The POTWs treat 32 billion US gallons (120 gigalitres) of wastewater every day (EPA, 2014). Emission of objectionable odors from these facilities is a major problem. POTW off-gases contain a wide range of odorous compounds, air toxics and volatile organic compounds (VOCs). These include volatile sulfur compounds, ammonia, benzene, toluene, chloroform, dichloromethane, and trichloroethylene (Lewkowska *et al*., 2016; Zhou *et al*., 2016). H_2S is the principal odorous component that causes nuisances even at volume fractions as low as 8 ppb (Smet *et al*., 1998). In addition to its unpleasant odor, H_2S gas is highly toxic (Roth, 1993). Continuous exposure to low (15-50 ppm) concentrations will generally cause irritation to mucous membranes, and may also cause headaches, dizziness, and nausea. Higher concentrations (200-300 ppm) may result in respiratory arrest leading to coma and unconsciousness. Exposure for more than 30 minutes at concentrations greater than 700 ppm have been fatal (MSDS, 1996). Concerns about the odor nuisance to the surrounding communities as well as the implementation of more stringent regulations are forcing POTWs to treat their off-gases.

Controlling H_2S is usually achieved by wet or chemical scrubbers. Chemical scrubbing in a packed tower is an established technique and is effective at gas contact times as short as 1.3-2 s (Gabriel and Deshusses, 2003). However, chemical scrubbing suffers from important drawbacks such as high operating costs, generation of halo-methanes that are known air toxics and can contribute to global warming (Wenhai *et al*., 2016), and the requirement for hazardous chemicals that pose serious health and safety concerns. Hence, more research is being done to convert chemical scrubbers into biotrickling units.

Gabriel and Deshusses (2003) demonstrated the conversion of a chemical scrubber located at the Orange County sewerage treatment facility in California. The converted bioscrubber was 9.75 m high, 1.82 m in i.d., and made of fiber-glass reinforced plastic with a nominal packing bed height of 2.8 m and bed volume of 7.3 m^3. The water trickling flow rate was 4.5 m^3 h^{-1} and the foul air was fed to the reactor at atmospheric pressure with an average flow rate of 16,300 m^3 h^{-1}. The researchers operated a laboratory pilot unit under conditions similar to those expected at a publicly owned treatment work and tested selected packing materials for their sustainability for biotrickling filtration. The conversion consisted of (i) replacing the existing packing, which had a low interfacial area and was not suitable for microorganism attachment; (ii) replacing the liquid recycle pump with a smaller one; (iii) disconnecting the chemical feeds; and (iv) modifying controls of the reactor. Before startup, the reactor was impregnated with 0.8 m^3 of activated sludge from wastewater-treatment plant. The pH started to decline after three days of operation to reach pH 2, in seven days after startup. The decline in pH (from the production of H^+ and sulfate from the oxidation of H_2S) was correlated with the increase in H_2S removal efficiency. Acclimation lasted for about ten days, after which H_2S removal was more than 99% for H_2S inlet volume fractions ranging from 5 to 25 ppm and remained high for the rest of the operation.

Vikromvarasiri and Pisutpaisal (2016), studied the removal of H_2S in a biotrickling system using a new bacterial strain of obligately chemolithoautotrophic, *Halothiobacillus neapolitanus* NTV01 (HTN). The biotrickling filter column was made from glass (0.475 m inner diameter and 0.72 m height). The column was packed with randomly structured packing media (GEA2H Water Technologies GmbH) to its working height of 0.282 m. The packing media had a random structure and made from high-density polyethylene (HDPE) 12 mm beads, surface 859 m^2 m^{-3}, and density 150 kg m^{-3}. A mixed gas stream containing CH_4, CO_2, and H_2S in the air was supplied to the inlet of the biotrickling filter; and [H_2S] varied between 0-255 ppm (v/v). The system could completely remove 45 ppm H_2S within 70 min of operation. The removal efficiency was in the range of 95-98%, for 225 ppm (v/v) H_2S.

The removal of H_2S in high-performance biotrickling filters was investigated by Kim and Deshusses (2005) using a differential biotrickling filter. The filter was designed to reach high gas velocities through a miniature packed bed. A small differential filter was used in this study. It was filled with a single cube (64 mL) of open-pore polyurethane foam packing. The biotrickling filter was operated in a counter-current mode. The air flow was circulated in a closed loop from an 85 L Tedlar bag to the differential biotrickling filter by a 0.2 horsepower blower up to a maximum linear velocity of 3 m s^{-1}. Pure H_2S was injected into the differential biotrickling filter system using a 20 mL syringe. Within a short time, gas velocity was varied to determine its effect on H_2S elimination capacity. The H_2S elimination capacity (35 to 125 g m^{-3} h^{-1}) was achieved at a liquid trickling velocity of 1.5 m h^{-1} and inlet [H_2S] was between 50 and 65 ppm. Interestingly, the addition of dissolved sodium sulfide (1-3.5 mg L^{-1}) resulted in reduced H_2S gas degradation at pH 1.9-2.1. H_2S is utilized by the microbial population as an S source. Hence, the addition of sodium sulfide competes with H_2S gas for biodegradation as the only ionic species (from dissolved H_2S and Na_2S), i.e. SH-, are metabolized by the bacteria.

5. ADVANTAGES AND LIMITATIONS

Conventional waste air treatment technologies such as absorption, adsorption, chemical scrubbing, and oxidation are generally not cost effective for the treatment of VOCs in dilute waste air streams (Kamal *et al*., 2016). Moreover, hazardous by-products such as NO_x, CO, dioxins, and furans are also generated whose treatment further adds to the operating cost. In the case of other conventional techniques like catalytic oxidation or adsorption, the development of specific adsorbents and the regeneration of catalysts adds to the total cost (Kamal *et al*., 2016). Also, these treatment techniques produce hazardous by-products also and require the addition of chemicals or fuels that may require further treatment or disposal, thereby creating additional environmental problems (Shareefdeen and Singh, 2005).

As compared to other biological removal techniques, biotrickling filters pose certain advantages over other methods. While they are cost effective (See Table 1) and have a low-pressure drop, but as is the case in other biofilters, biological operating parameters such as pH and moisture can be controlled by continuous trickling. Moreover, biotrickling filters are capable of treating acid degradation products of VOCs which

Table 2. Typical characteristics of biotrickling filters [Source: Deshusses and Cox (1999)].

Order	Characteristics	Values
1	Biotrickling filter bed height	1-5 m
2	Biotrickling filter cross section area	1-3,000 m^2
3	Air flow treated	100-1,000,000 $m^3 h^{-1}$
4	Packing void volume[a]	
	• Plastic rings, foam, random or structured packing	90-95%
	• Lava rock	~50%
5	Empty bed gas retention time[b]	2-60 s
6	Pressure drop	<100 Pa per m bed depth
7	Operating temperatures	15-50°C
8	Trickling rates[c]	0.01-10 $m h^{-1}$
9	Liquid dilution rate[d]	0.1-2 day^{-1}
10	Usual pH of the recycle liquid	
	• Removal of VOCs or compound difficult to degrade	~7
	• Removal of H_2S	1-2
11	Inorganic nutrient supply (N, P, K, traces)	Usually 0.05 to 1 times the amount calculated using biodegradation stoichiometry
12	Inlet pollutant concentration	
	• VOCs	0.01-10 $g m^{-3}$
	• Odors	500-50,000 odor units
13	Typical pollutant removal efficiencies	60-99.9 + %

[a] Value at reactor startup; over time, biomass growth will decrease bed porosity, typically by 10-30%
[b] The empty bed gas retention time (EBRT) is defined as the bed volume / air flow
[c] Trickling flow rate / bed cross section area
[d] Liquid feed rate / recycle liquid volume

gives an edge over other methods (Lu *et al*., 2001).

Despite many advantages, biotrickling filters also have some disadvantages. With the continuous supply of nutrient, the biofilm accumulates which leads to clogging. The accumulating biofilm is one of the major obstacles faced at present for the proper functioning of biotrickling filters over extended periods. Clogging increases pressure drop across the reactor thereby decreases pollutant removal rate (Okkerse *et al*., 1999; Cox and Deshusses, 1999). Biomass growth can be controlled by reducing the overall rate of biomass accumulation by either reducing the specific growth rate or increasing death and lysis (Alonso *et al*., 1998). Other options include increasing predation, washing-out or periodically removing the excess biomass (Cox and Deshusses, 1999).

Long acclimation periods, complex construction and operation, and secondary waste streams are some of the other problems faced by the biotrickling filters. Moreover, the presence of microorganisms in the media has raised concern over their potential release into the treated air and resultant human exposure to pathogens (Ottengraf and Konings, 1991). Table 1 lists the estimated capital costs, footprint and treatment capacity of biotrickling filters of various sizes presents. Table 2 presents the typical characteristics of biotrickling filters. Table 3 presents the recent investigations on biological treatment techniques for VOCs and odorants. Table 4 presents a comparison between various VOC control techniques including cost estimation.

6. FUTURE RESEARCH AND DEVELOPMENT NEEDS

Although much work has been done on biotrickling filters since 2005, almost all are primarily focused on the microbiology of pollutant-degrading microorganisms and the methods to control the biomass accumulation in the biofilter. The fundamental principles of biotrickling filters still need to be understood more clearly. Key questions to be addressed are mainly concerned with the complex ecology of biofilms. Future research work should concentrate on understanding the fundamentals of the degradation process through in situ analysis using modern tools in biotechnology. This is essential to establish baseline information (presently not available) for logical reactor design and optimum process operation. In particular, studies are needed to understand the overall role of secondary processes (i.e. those processes not directly associated with the elimination of the primary pollutant) and how these can be controlled in practice. In the future, the ability to control the ecology of biofilms in BTFs may enable optimal and limited biomass growth, so that

Table 3. Recent investigations of biological treatment techniques for VOCs and odorants.

Order	Scale	Process type	Type bed	Bed volume	Pollutants treated and inlet gas concentration	Gas flow rate and empty bed residence time (EBRT)	Bacteria	Removal efficiency	Inoculation	Reference
1	Laboratory	Biotrickling	Polypropylene pall rings	2.8 L	12-100 ppm H_2S, 450-700 ppm CH_3OH, balance air	7 L min^{-1}, 24 s	Methanol-degraders, sulfide oxidizing bacteria	>99% (H_2S), >95% (CH_3OH)	Yes (biomass from a H_2S-degrading biotrickling filter)	Jin *et al.*, 2007
2	Laboratory	Biotrickling	Polypropylene pall rings	10 L	170 ppm H_2S, 2.2 g m^{-3} toluene, balance air	1 m^3 h^{-1}, 36 s	Toluene-degraders, sulfide oxidizing bacteria	100% (H_2S), 25-75% (toluene)	Yes (biomass from a toluene-degrading biotrickling filter)	Cox and Deshusses, 2001
3	Full	Biofiltration	Polyurethane foam	1000 L	4.81-27.48 ppm H_2S, balance air	60 m^3 h^{-1}, 60 s	sulfide oxidizing bacteria	>90%	Yes (culture from odor treatment bioreactor)	Li *et al.*, 2013
4	Laboratory	Biofiltration	Granular activated carbon	1 L	100-4000 ppm H_2S, balance air	15 L h^{-1}	sulfide oxidizing bacteria	>98%	Yes (culture from concentrated latex wastewater)	Rattanapan *et al.*, 2009
5	Bench	Biofiltration	Compost and wood chips	12 L	0.31-1.44 ppm dimethyl sulfide (DMS)	1.5-2.5 L min^{-1}, 360 s	*Bacillus sphaericus*	71%	Yes (seed culture of *B. sphaericus* isolated from garden soil)	Giri and Pandey, 2013
6	Laboratory	Biofiltration	Sugarcane bagasse	0.98 L	3.13 ppm benzene	Superficial velocity of 30.6 m h^{-1}	*Pseudomonas* sp. NCIMB 9688	100%	Yes (culture of *Pseudomonas* sp. NCIMB 9688)	Sene *et al.*, 2002
7	Laboratory	Biotrickling	High-density polyethylene	0.5 L	0-255 ppm H_2S	0.5 L min^{-1}, 60 s	*Halothiobacillus neapolitanus* NTV01 (HTN)	95-98%	Yes (sludge from a full-scale activated suldge system)	Vikromvarasiri and Pisutpaisal, 2016
8	Laboratory	Biofiltration	Compost and lava rock	4 L	296.88-857.66 ppm n-butanol	4 L min^{-1}, 60 s	n-butanol degraders	>73%		Eshraghi *et al.*, 2016
9	Full	Bioscrubbing (absorption + aerobic biooxidation) + separate biofiltration	Alakaline solution for scrubbing, compost for biofiltration		Natural gas: 2000 ppm H_2S	322,000 nm^3 d^{-1}	Indigenous	>99.8% (H_2S)		Benschop *et al.*, 2002

Table 4. Comparison of various VOC control techniques [Source: Khan and Ghoshal (2000)].

Order	Techniques	Annual operating cost[a] (USD/cfm)	Removal efficiency (%)	Positive and negative remarks
1	Absorption	25-120	90-98	Product recovery can offset annual operating costs Requires rigorous maintenance Requires pretreatment of the VOCs
2	Adsorption (Activated carbon)	35-10	80-90	Recovery of compounds, which may offset annual operating costs Susceptible to moisture, and some compounds (ketones, aldehydes, and esters) can clog the pores
3	Biofiltration	15-75	60-95	Requires less initial investment, less non-harmful secondary waste, and non-hazardous Slow, and selective microbes decomposes selective organics, thus require a mixed culture of microbes
4	Condensation	20-120	70-85	Product recovery can offset annual operating costs Requires rigorous maintenance
5	Catalytic oxidation	15-90	90-98	Energy recovery is possible (maximum up to 70%) Efficiency is sensitive to operating conditions
6	Thermal oxidation	15-90 (Recuperative) 20-150 (Regenerative)	95-99	Energy recovery is possible (maximum up to 85%) Halogenated and other compounds may require additional control equipment
7	Zeolite	15-40	90-96	Effective in more than 90% RH, Recovery of compounds offsets annual operating costs High cost of zeolite, restricted availability
8	Membrane separation	15-30	90-99	No further treatment, recovery of solvent may offset the operating costs Membranes are rare and costly

[a] Not adjusted for inflation. 45% inflation from 1999 to 2016 (http://www.bls.gov/home.htm)

reactor stability can be assured over a very long period (greater than 150 days) (Zhou *et al*., 2016). Additional research is needed for better understanding of the kinetic relationships for pollutant biodegradation. Particularly, understanding the biodegradation of mixtures of pollutants, the role and impact of oxygen and ancillary nutrients on the rate of biodegradation and the biomass yield, and the influences of various stresses, such as changing conditions and mass transfer limitations, is important. This, together with a number of pilot-scale operation and demonstration of techno-economic viability, would transfer this technology from lab to the field (Valero *et al*., 2017; Cox and Deshusses, 1999).

7. CONCLUSION

This review provides an overview of the biotrickling reactors being used for the treatment of various VOCs and odor laden waste gas streams, limitations of the already existing BTFs, future prospects, and avenues which can be explored for a better understanding and development. A summary of some of the other biological treatment methods has also been provided before describing the BTF in detail. Clearly, the design of the BTF still requires improvement, and demonstration of significantly better performance compared to existing designs. Further, developments of innovative combined bioreactor designs remain a high priority. Development in reactor design and development will require similar advances in understanding the fundamentals of bioprocesses so that a more logical, creative and focused approach to BTF design can be implemented.

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