Review

Effects of Water Content and Irrigation of Packing Materials on the Performance of Biofilters and Biotrickling Filters: A Review

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Abstract: Biofilters (BFs) and biotrickling filters (BTFs) are two types of bioreactors used for treatment of volatile organic compounds (VOCs). Both BFs and BTFs use packing materials in which various microorganisms are immobilised. The water phase in BFs is stationary and used to maintain the humidity of packing materials, while BTFs have a mobile liquid phase. Optimisation of irrigation of packing materials is crucial for effective performance of BFs and BTFs. A literature review is presented on the influence of water content of packing materials on the biofiltration efficiency of various pollutants. Different configurations of BFs and BTFs and their influence on moisture distribution in packing materials were discussed. The review also presents various packing materials and their irrigation control strategies applied in recent biofiltration studies. The sources of this review included recent research articles from scientific journals and several review articles discussing BFs and BTFs.

Keywords: biofilters; biotrickling filters; intermittent trickling; VOC; moisture content; packing material

1. Introduction

In various air biological treatment technologies, such as biofilters (BFs) and biotrickling filters (BTFs), the factors that affect the performance of biofiltrations can be divided into physical, chemical, and biological [1,2]. In biological air treatment technologies, microorganisms play a key role in the biodegradation of volatile organic contaminants (VOCs) released into the environment by natural or industrial processes. One of the main physical factors that determines the growth and multiplication of microorganisms is the moisture content of packing material. Water is the most important medium in which the metabolism of microorganisms occurs. In addition, all chemical reactions that occur in living microorganisms require water. Water is necessary not only for the vital activity of microorganisms but also for ensuring the dissolution of VOCs. The biological method of air treatment is based on the use of various types of microorganisms (such as bacteria and fungi) for the oxidation of VOCs. During biofiltration, aerobic heterotrophs oxidize organic compounds carried to the packing material by air stream. The growth of spontaneous or inoculated microorganisms on the surface of the packing material forms a thin liquid film (a biofilm). A biofilm is the main component of a biofiltration device that contains different types of microorganisms [3]. The decomposition of VOCs results in the growth of new microorganisms and organic components are oxidized to carbon dioxide (CO₂) and water (catabolism) [4].

The supply of VOCs to the biofiltration device increases the activity of microorganisms. However, the reduction in the flow rate of the treated VOC reduces the activity of microorganisms, and at the same time, the thickness of the biofilm decreases. The biofilm consists of more than 90% water [5]. Air purification biotechnologies are used to decompose hydrophilic (i.e., water-soluble) compounds with dimensionless Henry’s law coefficient (H) < 0.1 and hydrophobic (i.e., poorly water-soluble) organic compounds with H > 0.1 [5]. Hydrophilic compounds include VOC groups such as alcohols (e.g., butanol, methanol)
and ketones (e.g., acetone, methyl ethyl ketone (MEK)) and other gaseous compounds such as SO\textsubscript{2} and NH\textsubscript{3} \cite{6}. Hydrophobic organic compounds include alkanes such as methane and hexane, and aromatic hydrocarbons such as toluene \cite{7,8}.

Packing materials are one of the most important elements of BFs and BTFs. Vital processes of microorganisms occur in a biofilm formed on the surface of the packing material \cite{9}. To achieve biodegradation of organic compounds, the packing material must be moistened with water saturated with nutrients and buffers \cite{10}. Buffers are needed to maintain pH in biofilters, which should remain close to neutral, i.e., within the range of 6.0 to 7.5 \cite{11}. Buffers that are commonly used in biofilters are CaCO\textsubscript{3} \cite{12}, solutions of NaOH \cite{13,14}, KH\textsubscript{2}PO\textsubscript{4}, KHPO\textsubscript{4} \cite{15}, etc. The moisture content of the packing material affects its porosity and compaction; therefore the pressure drop of BF or BTF \cite{16}. The most frequent range of moisture content for most packing materials provided in the literature is in the interval of 40 to 60\% on a weight basis \cite{17}. The humidity of packing material changes during the air purification process. When gas flows through the packing material, the air is saturated with water from the packing material, causing a reduction in the moisture content of the packing material. On the other hand, during the biodegradation process, organic compounds are converted to carbon dioxide (CO\textsubscript{2}) and water, partially restoring the moisture content in the packing material. However, the amount of water produced is not enough and constant irrigation of the packing materials of BFs and BTFs is required. Insufficient moisture content in the packing material prevents the formation of a biofilm layer. However, the moisture content of the packing material should not be too high, as excessive water can cause anaerobic zone formation, compaction, and clogging of the packing material \cite{2}.

When using BFs and BTFs, the biggest challenge can be maintaining the right amount of moisture throughout the system. Proper irrigation of BF and BTF packing materials is crucial for the stable and robust performance of the biofiltering device. Therefore, various studies have been performed to investigate the optimal level of moisture content in the packing materials of BFs or to find an optimal rate of trickling liquid flow in BTFs to ensure efficient and cost-effective elimination of contaminants. Moreover, various packing material irrigation systems have been developed and tested. The objectives of this review are aimed at (1) reviewing different packing materials applied in BFs and BTFs; (2) presenting the influence of the moisture content of the packing material on the biofiltration efficiency; (3) and providing an overview of various irrigation systems of packing materials. In this review, we analyse two types of biofiltration devices: biofilters and biotrickling filters. The general principles of biofiltration are similar for the bioreactors reviewed; however, they differ in their packing material irrigation systems.

2. Comparison of Biofiltration Processes in Biofilters and Biotrickling Filters

Biofiltration is a pollution control technique that involves three different phases, gas, liquid, and solid \cite{18}. Both BFs and BTFs are packed with various packing materials (a solid phase), where microorganisms are immobilized \cite{19}. The main difference between two types of bioreactors is that the water phase in BFs is stationary, i.e., water is only used to maintain the required humidity of the BF packing material. Biotrickling filters allow the absorption and biodegradation of pollutants to be carried out in a single device. The water flow in BTFs is continuous or occasional and it contains the nutrients necessary for the growth microorganisms \cite{20}.

The schemes of a gaseous pollutant biofiltration process in BF and BTF are shown in Figure 1. To degrade VOCs, the pollutant must be transferred from the gas phase to the biofilm. The contaminant mass transfer occurs due to the concentration gradient between different phases \cite{9,21}. The water in the BTF packing material can be in a free or bound form \cite{22}. In BTFs, the water layer (mobile liquid phase) surrounds the biofilm.
layer, and therefore, an additional contaminant transport step to the biofilm is required. Alonso et al. [25] approximated the thickness of the flowing water layer:

\[ L_{wl} = \left( \frac{Q_w^3 \mu_w}{AH \rho_w g} \right)^{1/3}, \]

where \( Q_w \) is the water flow rate, \( \mu_w \) is the viscosity of water, \( A \) is the surface area of the packing material, \( H \) is the height of the filter, \( \rho_w \) is the density of water, and \( g \) is gravitational acceleration [23].

![Figure 1. Biofiltration process in (a) a biofilter and (b) a biotrickling filter. 1: Contaminant transport in a gaseous phase; 2: contaminant transport in a liquid phase; and 3: contaminant biodegradation.](image)

The removal of pollutants by BF and BTFs strongly depends on the pollutant mass transfer rate from the gas phase to the biofilm [24]. The presence of biomass and metabolites may affect the equilibrium of the gas-biofilm phase, thereby affecting the transfer of the pollutant [25]. Biofilters and BTFs are particularly effective in the treatment of highly water-soluble VOCs [26]. The solubility of VOC in a water or a biofilm phase depends on the pressure. The relationship between VOC pressure (or concentration in treated air) and solubility of a gas in a liquid is described by Henry’s law, which is formulated as follows: at a constant temperature, the solubility of a gas in a liquid is directly proportional to its partial pressure above the liquid. Henry’s law constant is expressed by formula:

\[ H = \frac{C_g}{C_w}, \]

where \( C_g \) is VOC concentration in the gas phase, and \( C_w \) is VOC concentration in the liquid phase.

Since BF does not have a mobile liquid phase, they are better suited for hydrophobic pollutant treatment than BTFs [19]. The contaminant diffusion coefficient in a packing material is very low and can be considered negligible [18].

3. Configurations of Biofilter Irrigation Systems

All biofilters are equipped with irrigation and temperature control systems of packing material which ensure the vital activity of microorganisms. In BF, the irrigation system is one of the key elements. Moisture can be added to the BF system in two ways: humidification of the inlet gas stream in air humidification chambers and/or by irrigating BF packing material.
3.1. Humidification of Inlet Gas and Packing Material by Spraying

Biofilters have two general configurations [27]:

1. Open-type biofilters;
2. Closed-type biofilters.

Open-type BFs are usually fabricated of concrete boxes with upper parts in contact with the atmosphere. Such BFs are exposed to changes in weather conditions, so the moisture content of their packing materials is difficult to control: in rain, the filter material can be too wet, and in hot weather it can be too dry. Closed-type BFs are less susceptible to moisture loss as they are located in steel or plastic containers. Parameters of the humidification process, such as liquid flow, temperature, etc., can be easier controlled [27].

Often open-type or closed-type BFs (Figure 2) have separate inlet gas humidification chambers. Contaminated gases are pre-humidified by spray nozzles in these chambers before entering the BF. At the bottom of the humidification chamber there is a water tank from which water is supplied to the nozzles by a water pump. In some BF constructions, the inlet air is humidified by passing the gas flow through the water layer (bubble saturator) [18,28]. Moisture (water) also absorbs contaminants from the gas phase during the humidification process. The achieved saturation levels of inlet gas usually reach 90–95% [29,30]. The air humidified in the chamber is directed to the lower part of the BF, where it is evenly distributed over the entire cross-sectional area of the packing material. To maintain the activity of inoculated microorganisms, the packing material itself is occasionally irrigated with water or a nutrient solution. In most cases, the packing material of BFs is moistened using spray nozzles installed above the packing material. Due to the presence of moisture in the packing material and large contact surface for moisture transfer from packing material to treated gas, and relatively long gas residence time, treated air often leaves the BF with a relative humidity of 100% [29]. The contaminated gas can also be supplied from the top to the bottom of a BF (descending flow mode) in order to reduce drying of the packing material through airflow [31].

![Figure 2. Closed-type biofilter with inlet gas humidification chamber.](image)

The decrease in relative humidity of inlet gas may have adverse effects on the performance of BF. Bruneel et al. [32] studied the influence of inlet gas humidification on a fungal BF performance. It was found that disconnection of the inlet gas humidification system caused a sudden decrease in relative humidity of inlet gas (from 90% to 30% in lower section of BF) and decrease in hexane removal efficiency [32]. Jin et al. [33] investigated alpha-pinene removal through a fungal biofilter under various humidity of inlet gas (16, 45, 85, and 95%). The optimal humidity of inlet gas was found to be 85% [33]. On the other hand, excessive water accumulation in the pores of packing material results in its clogging and the increase in pressure drop of BFs [34,35]. In many biofiltration studies, the packing material in BF column is divided into several sections (layers) with spaces between them [36,37]. Such BF construction can reduce uneven moisture distribution in
the packing material, improve its physical properties (e.g., temperature distribution), and reduce pressure drop in the system [4]. Several studies have observed that the highest pollutant elimination capacity is obtained in the lowest packing material section (when BF is operated under up-flow air mode) [36,38]. For example, Malakar et al. [36] constructed lab-scale BF which had packing material divided to three sections. The authors found that toluene elimination capacity was greater at the lower section of packing material that had higher moisture content and biomass concentration. Authors also observed that an increased gas flow rate resulted in the increase in the activity of the upper packing material section [36]. The highest elimination capacities of pollutants obtained in the entry section of packing material can be explained by the fact that this section receives the highest pollutant loading. Moreover, due to the effect of gravity, the lower section of packing material can contain a higher amount of moisture and biomass, which may enhance the removal of pollutants.

To increase inlet air humidity and improve VOC removal efficiency, BFs with integrated suspended-growth bioreactors are designed (Figure 3c shows example of such BF construction). Such an approach can be useful for treating VOCs, with $H < 1$ [39]. In such integrated BFs, the inlet air duct is installed at the sump (under the liquid) in order to accelerate the absorption of VOCs in water. The air contaminated with VOCs firstly passes through the water layer, then it is directed evenly towards the packing material using perforated pipe. Microorganisms begin to multiply in the water, breaking down some amount of contaminants [40]. The flow of air through the water layer reduces the concentration of pollutants in the inlet gas. In addition, the use of such inlet air humidification ensures that a high relative humidity of gas (up to 100%) can ensure the humidity of packing material is maintained. Excess water (drained from the packing material) is returned to the lower part of the BF. Such design of humidification systems can reduce the economic costs related with BF humidification [4].

![Figure 3. Closed-type biofilters](image1)

Li et al. [39] designed an integrated bioreactor for gaseous o-xylene treatment. The effective volumes of packed and liquid sections were of 1.2 m$^3$ and 0.3 m$^3$, respectively. The authors observed an average pollutant removal of 91.1% under inlet load range of 529.1–931.5 mg/m$^3$ and maximum pollutant elimination capacity of 53.4 g/(m$^3$ h) [39]. Baltrénas and Zagorskis [4] designed and investigated three different closed-type BF
constructions, which had different humidification systems and different air flow directions (ascending and descending) (Figure 3). The packing materials of those BFs were divided to five layers. In order to maintain the vitality of the microorganisms, nutrient solution was sprayed onto each layer of the packing material by nozzles installed above each layer. On the packing material, water was sprayed from the water tank present at the bottom of each BF. Meshes were installed below and above each layer of the packing material to ensure an even distribution of air and water flow throughout the biofilm. Two of investigated BFs had separate air humidification chambers, in which the air was humidified to 95–100%, while in BF with the integrated humidification (suspended-growth) chamber the air was humidified to 100%. Authors found that the lowest acetone removal efficiency was obtained in BF with descending gas flow, while for the other two BFs, acetone removal efficiency was similar (>90%) [4]. The biofilter with descending gas flow did not have a gas flow distributor installed before the entry layer of the packing material. Therefore, it can be assumed that the lower acetone removal efficiency can be due to the uneven air flow distribution before the entry of the packing material. The uneven distribution of the air flow can result in the formation of areas where the pollutant load per unit volume of the packing material was too high.

3.2. Irrigation of Biofilter Packing Material by Immersing It in Liquid

A typical method of irrigation of BF packing material is the spraying of nutrient solutions to the top of the BF packing material [41]. In contrast, in some BFs, the entire or part of the BF packing material is immersed in liquids containing nutrients. Such a packing material irrigation system can be used to reduce the energy consumption required to irrigate the packing material and therefore, the operation of such BF may be simpler. In addition, the packing material can be irrigated even in the shutdown periods. Baltrėnas et al. [42] designed the BF with packing material consisting of several plates arranged vertically next to each other (Figure 4). To irrigate the packing material, the lower part of the packing material was immersed in a nutrient solution. The study observed that after immersion of the lower part of the packing material, a nutrient solution rose up to the top of the plates, thus moistening the entire volume of the packing material. During the irrigation of the packing material through capillary action, the nutrient solution rose to fill the small capillaries and pores. The use of such an irrigation system helped to avoid an insufficient or excess moisture level in the packing material. In addition, the use of a capillary system to irrigate the packing material helped to avoid possible leaching of biomass from the packing material [42].

![Figure 4. Biofilter with packing material partially immersed to nutrient solution. Reprinted with permission from [42].](image-url)
Only few studies have investigated removal of VOCs using BF with completely immersed (flooded) packing materials. Mansoori et al. [41] compared two different packing material (fabricated of polyvinyl formal) irrigation methods. It was found that complete soaking of packing material in the liquid provided more uniform distribution of supplied nutrients in the filter media compared with nutrient supply by spraying. In addition, soaking provided higher mass transfer for most types of nutrients. High efficiency of MEK removal (above 95%) was achieved by both bioreactors; however, toluene removal in the BF with completely immersed packing material was lower than that in the filter, in which packing material was humidified by spraying [41]. In other study, Lu et al. [43] also compared toluene removal through a typical biofilter and fluidized bed biofilter. Both BFs were packed with wheat bran coated expanded polystyrene and run under the same operating conditions. It was found that average toluene elimination capacity was similar between two studied BFs. Compared with a conventional BF, a fluidized bed BF was more advantageous in controlling microorganism growth, had a lower pressure drop, and better mass transfer performance [43].

4. Biotrickling Filter Irrigation Systems

The disadvantage of BFs is the difficulty in controlling the moisture content of their packing materials. BTFs allow better control of operational parameters such as biofilm thickness, pH, and subsequently the elimination of pollutants can be performed at higher VOC inlet loadings than with conventional BFs [44]. Contrary to BFs, BTFs do not require pre-humidification of inlet gas before its treatment; although, the humidification of inlet gases before entering BTF using humidifiers was performed in some studies [45,46]. To maintain the activity of the microorganisms in BTFs, the packing material is trickled continuously or intermittently by nutrient solution [47]. The most commonly used circulating liquid is a mineral salt solution [48]. Liquid flow through the packing material of a BTF provides humid conditions needed to support the biofilm growth and maintain the microbial activity [3]. The nutrient solution is supplied to the unit by a circulation pump. Khoramfar et al. [48] studied VOC removal by BTF-BF unit and observed that due to continuous nutrient solution spraying, biofilm growth and consequently VOC removal was relatively higher in the BTF unit than in the BF unit [48]. Trickling liquid also washes out excess biofilm as well as degradation products that can hinder biodegradation of VOC [3].

Figure 5 shows schematic representations of BTF configurations. Biotrickling filters use a co-current or counter-current flow of liquid and gas phases [3]. The main option is to use the counter-current flow of different phases, which is usually convenient for absorption of pollutants. The main issue of using this mode is clogging of the lower part of a BTF (owing to biomass over accumulation), so the co-current mode is sometimes preferred [22]. In order to reduce uneven biomass or moisture distribution and improve pollutant elimination, several studies used BTFs with directional-switching flow configurations [49,50]. Wang et al. [50] found that such BTF configuration resulted in a higher toluene elimination capacity and helped to avoid clogging of the packing material. In addition, BTF was more tolerant to the changes in the inlet pollutant loadings [50].

High moisture content can cause clogging in the top of BTF packing material. Therefore, similar to BFs, BTFs with packing material divided to several layers are also often designed [51–53]. This BTF construction can help to distribute contaminated gas and trickling liquid flow more uniformly. Yang et al. [54] studied VOC removal using BTF (counter-current flow mode) with three-layer ceramsite packing material. It was reported that liquid sprayed from the top of the filter resulted in the higher moisture content of the upper layer than of the middle and lower layer [54]. Wu et al. [55] investigated methyl acrylate removal using BTF packed with three-layer ceramic particles. The BTF was operated in counter-current flow and in an up-flow mode. The authors found that the greatest amount of pollutant was removed in the first layer of packing material (the bottom of BTF), where high abundance of bacteria responsible for methyl acrylate degradation were detected [55]. Similar tendencies were also found in the other studies, where BTFs with
several layer of packing material were used [52, 56]. While, Wang et al. [50] reported that toluene elimination was relatively similar between different sections of packing material of BTF which was operated under directional-switching flow mode [50].

![Figure 5. The principal scheme of a biotrickling filter (a), co-current biotrickling filter (b), biotrickling filter with three-layer packing material (c).](image)

Similar to BFs, BTFs can also be designed with integrated scrubbers. This can be performed by placing the inlet gas pipe in the trickling liquid holding tank (under the liquid). Xue et al. [57] constructed such a type of BTF and reported that trimethylamine, which is hydrophilic contaminant, was completely eliminated in the scrubber and in the biotrickling sections. The authors also noted that such construction can help to reduce the pollutant concentration at the inlet area of BTF and improve the distribution of biofilm [57].

The combination of two different microbial growth systems (fixed-film and a suspension) can possibly improve the ability of the microorganisms to degrade VOCs. Flooding of the packing material in a BTF can enhance the absorption of VOCs into the liquid medium, since the surface area of the interface between the gas phase and liquid phase increases [58]. There is a lack of studies combining different microbial growth systems in a single bioreactor [59]. In fact, this type of the bioreactor (in a particular study it was called a 'fixed-film bioscrubber') was designed by Wongbunmak et al. [60] and used for toluene and styrene biodegradation. Employing the single device, authors compared the performance of the fixed-film bioscrubber and BTF by changing the liquid level in the column. The applied packing material used in the study was polyurethane (PU)-based sponge. Authors found that toluene elimination capacity was higher in the fixed-film bioscrubber (83 g/(m³ h)) than that in BTF (50 g/(m³ h)). In addition, the fixed-film bioscrubber contained a larger amount of BTEX-degrading Microbacterium esteraromaticum cells than BTF [60].

5. Influence of Packing Material Moisture Content on Biofiltration Efficiency

5.1. Packing Materials

The purpose of BF/BTF packing materials is to provide a large surface for the growth of microorganisms. Adsorption of VOCs directly on the packing material also occurs after their diffusion through the whole biofilm layer [25]. Generally, packing materials used in air biofiltration systems are divided into organic (biomass-based), inert, and mixed materials. The early BFs were usually loaded with various natural organic packing materials, such as wood chips, tree bark, compost, peat, and soil [61]. Biotrickling filters are usually packed with inert materials; nevertheless, in some studies natural organic materials (e.g., wood chips, pine bark or straw) were also used [62, 63]. Organic packing materials serve as a food source for microorganisms. The biofilm formed on the organic packing material not only absorbs VOCs supplied to the device but also deteriorates the packing material due to consumption of its carbon molecules. Deterioration of organic materials cause uneven
moisture distribution and therefore reduce the microbial activity, leading to a decrease in VOC removal efficiency [25]. As a result, most natural organic packing materials are used for a short period of time and must be replaced after a few years.

In the last decade, ways to extend the life of BF packing materials have been actively sought. To ensure high cleaning efficiency and durability of the device, BFs are packed with inert materials or mixtures of organic and inert materials (Table 1). The inorganic material can support a rigid and uniform structure to avoid compaction and minimize uneven air flow distribution [25,64]. Mixtures of organic and inert packings often include compost as natural organic material, which can be supported by inert materials such as ceramic beads [18,64], granular activated carbon (GAC) [65], bamboo-clay [66], etc. Inert packing materials such as lava [67], ceramsite [53,67], and perlite [12,32,34] are often used in biofiltration studies. Ceramsite is a light porous material which can be fabricated from municipal sludge [68], fly ash [69], and clay. Ceramsite has an inert surface, high porosity, good longevity, and can provide sufficient surface for microorganism attachment [69]. Perlite is also a suitable material for BFs as it has good attachment properties for various microorganisms and good water holding capacity (about 3–4 g water per 1 g perlite) [32,70].

In addition, BFs packed with perlite can be run at a lower pressure drop [70]. One of the most popular inert materials used in recent biofiltration studies is PU foam [37,39,71]. Polyurethane foam is characterised by high porosity (about 95%), good mechanical strength, large surface area, and low density (about 20 kg/m³). However, a smooth and hydrophobic surface of PU foam can result in uneven distribution of moisture and biofilm. Sun et al. [72] modified a PU surface (by using various chemical substances) to make it rougher and create a positive charge. The results showed that such a PU surface enhanced biofilm formation and improved the removal of benzene, toluene, ethylbenzene, and o-xylene (BTEX) by biofilters [72]. In order to improve the performance of BFs or BTFs, composites of different materials can be produced and used as packings. For example, Zhao et al. [73] combined PU and bamboo charcoal powder to form packing material applied in BF for the removal of n-hexane (EC = 12.68 g/(m³ h)) and dichloromethane (EC = 30.28 g/(m³ h)). It was concluded that such packing can be widely used for hydrophobic pollutant treatment [73].

Zhang et al. [74] produced polydimethylsiloxane (PDMS) and foam ceramic composite material and used it as packing media in BTF for toluene removal. The results of the study showed that BTF packed with PDMS/foam ceramic composite showed better performance in toluene removal, and more rapid start-up and restart (after 1 month starvation period) than BTF packed with PDMS [74]. The biochar-based packing material of BF or BTF can act as an adsorbent of pollutants and carrier of microorganisms [75]. Liu et al. [76] used bamboo-biochar beads as packing material in a BTF designed for toluene removal. The authors observed that on average 74.9 ± 17.8% of toluene removal efficiency was achieved by BTF due to the adsorption of the pollutant. When the microorganisms became activated, the removal of toluene increased to 80.3 ± 14.4% [76].

**Table 1.** Examples of packing materials used in recent biofiltration studies.

<table>
<thead>
<tr>
<th>Packing Material; Inoculation</th>
<th>Bioreactor</th>
<th>Pollutant</th>
<th>Humidity Control</th>
<th>EBRT</th>
<th>ILR or IC</th>
<th>EC or RE</th>
<th>Packing Material Properties</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coir pith glucose syrup beads + sterile distilled water + Bacillus megaterium</td>
<td>BF</td>
<td>Benzene</td>
<td>External nutrients were not added</td>
<td>60 s</td>
<td>ILR (max): 22.15 g/(m³ h)</td>
<td>EC (max): 18.82 g/(m³ h)</td>
<td>Air filled porosity: 0.4 m³/m², Moisture content of PM: 53% (day 0); 6.8% (day 215)</td>
<td>[77]</td>
</tr>
<tr>
<td>Mixture of bamboo-clay and turkey litter compost (3:2, w/w); microbial consortium of compost with toluene degraded consortia</td>
<td>BF</td>
<td>TCE</td>
<td>Inlet gas humidification (RH = 70%), periodical spraying of packing material with moisture and nutrients</td>
<td>1.4–2.0 min</td>
<td>8–67 g/(m³ h)</td>
<td>3–38 g/(m³ h)</td>
<td>Porosity of bamboo: 75%; Moisture content of bamboo: 65%; Porosity of clay: 64%; Moisture content of clay: 10%</td>
<td>[66]</td>
</tr>
<tr>
<td>Packing Material; Inoculation</td>
<td>Bioreactor</td>
<td>Pollutant</td>
<td>Humidity Control</td>
<td>EBRT</td>
<td>ILR or IC</td>
<td>EC or RE</td>
<td>Packing Material Properties</td>
<td>References</td>
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<tr>
<td>Mixture of compost and ceramic beads (5:2, v/v)</td>
<td>BF</td>
<td>Toluene</td>
<td>Inlet gas humidification (RH = 45%), periodical spraying of packing material with nutrient solution</td>
<td>1.2–2.0 min</td>
<td>0.052–3.810 g/m^3</td>
<td>EC (max): 93 g/(m^3 h) at ILR of 114 g/(m^2 h)</td>
<td>SSA of ceramic beads: 500 m^2/m^3; Water retention of ceramic beads: 0.3 w/w. Porosity of compost: 50%; Moisture content of compost: 42%</td>
<td>[64]</td>
</tr>
<tr>
<td>Mixture of compost and GAC prepared in the form of spherical beads; activated sludge</td>
<td>BF</td>
<td>Toluene</td>
<td>Inlet gas humidification; periodical spraying of packing material with water (100 ml/day)</td>
<td>19–42 s</td>
<td>160–8750 g/(m^3 h)</td>
<td>RE: 70–96%; EC (max): 6665 g/(m^3 h) at ILR of 8790 g/(m^2 h)</td>
<td>Porosity of PM: 68%; Moisture content of PM: 55% (under wet conditions)</td>
<td>[65]</td>
</tr>
<tr>
<td>Mixture of vermi-compost and wood charcoal (2:1, v/v); Pseudomonas putida PTCC 1694</td>
<td>BF</td>
<td>Toluene</td>
<td>Inlet gas humidification; spraying packing material with nutrient solution every 3 days</td>
<td>21 s</td>
<td>21.27 ± 3.3 g/(m^3 h)</td>
<td>16.23 ± 3.37 g/(m^3 h)</td>
<td>Porosity of compost: 55%; moisture content of compost: 44–53%; porosity of wood charcoal particles: 50%; moisture content of wood charcoal particles after 24 h immersion in water: 78%</td>
<td>[35]</td>
</tr>
<tr>
<td>Mixture of 60% peat and 40% perlite (v/v); Pichia pastoris GS115</td>
<td>BF</td>
<td>Methanol</td>
<td>Inlet gas humidification</td>
<td>60–120 s</td>
<td>81.12–672.12 g/(m^3 h)</td>
<td>80.28–322.74 g/(m^3 h)</td>
<td>Moisture content of PM: 63%; Porosity of PM: 66.8%</td>
<td>[78]</td>
</tr>
<tr>
<td>Perlite; activated sludge</td>
<td>BF</td>
<td>Methanol</td>
<td>Inlet gas saturation in a liquid methanol; daily mineral medium spraying</td>
<td>60–160 s</td>
<td>98.8 ± 4.8–341.5 ± 47.5 g/(m^3 h)</td>
<td>EC (max): 343.8 g/(m^3 h)</td>
<td>Moisture content of PM: 50–73%; moisture level was higher in the lower BF module due to effect of gravity</td>
<td>[34]</td>
</tr>
<tr>
<td>PU foam cubes; Brevibacillus (58.8%)</td>
<td>BF</td>
<td>Total VOCs</td>
<td>Spraying of packing material with nutrient solution</td>
<td>1.5 min</td>
<td>0.5–200 mg/m^3</td>
<td>&gt;85%</td>
<td>Size of 1 cube: 1.0 cm^3; Porosity: 95%</td>
<td>[71]</td>
</tr>
<tr>
<td>Pressmud and cornstack (80%/20%); sludge</td>
<td>BF</td>
<td>MEK</td>
<td>Inlet gas humidification, spraying packing material with nutrient solution</td>
<td>0.7–2.81 min</td>
<td>4.16–100.08 g/(m^3 h)</td>
<td>RE: 58–97%</td>
<td>MEK removal decreased, when the cornstack ratio increased</td>
<td>[79]</td>
</tr>
<tr>
<td>Cell-immobilized bamboo-biochar beads; mixed culture from activated sludge</td>
<td>BTF</td>
<td>Toluene</td>
<td>Tricking liquid rate: 2 L/min</td>
<td>99 s</td>
<td>0.14–99.1 g/(m^3 h)</td>
<td>EC (max): 34.9 g/(m^3 h) at ILR of 46.2 g/(m^2 h)</td>
<td>Water content: 82 ± 1.14%</td>
<td>[76]</td>
</tr>
<tr>
<td>Ceramic Raschig rings; activated sludge</td>
<td>BTF</td>
<td>Styrene</td>
<td>Continuous liquid trickling</td>
<td>34–136 s</td>
<td>12.3–159.8 g/(m^3 h)</td>
<td>EC (max): 126 g/(m^3 h) at ILR of 160 g/(m^2 h)</td>
<td>SSA: 526 m^2/m^3; Porosity: 68.2%</td>
<td>[80]</td>
</tr>
<tr>
<td>Ceramspite; Fusarium oxysporum</td>
<td>BTF</td>
<td>Toluene</td>
<td>Intermittent nutrient solution spraying</td>
<td>37–73 s</td>
<td>20–100 g/(m^3 h)</td>
<td>EC (max): 79.9 g/(m^3 h)</td>
<td>A diameter of ceramic: 4–5 mm; porosity 46.3%; SSA: 1.08 m^2/g</td>
<td>[53]</td>
</tr>
<tr>
<td>HDPE pall rings; Pandoraea prunorum DSM 16536, Rhizomia eutropha PTCC1615</td>
<td>BTF</td>
<td>Toluene; methanol</td>
<td>Tricking liquid rate: 20 mL/min</td>
<td>60 s</td>
<td>18–36 g/(m^3 h) (toluene); 0–226 g/(m^3 h) (methanol)</td>
<td>RE: 30–99% (toluene); EC (average): 220 g/(m^3 h) for an ILR of 226 g/(m^3 h) (methanol)</td>
<td>SSA: 480 m^2/m^3; Pore density of pall rings: 90%; Porosity of PM: 68%</td>
<td>[26]</td>
</tr>
<tr>
<td>PDMS and foam ceramic composite; Cladophialophora fungus</td>
<td>BTF</td>
<td>Toluene</td>
<td>Intermittent nutrient solution spraying</td>
<td>10–57 s</td>
<td>20–265 g/(m^3 h)</td>
<td>EC (max): 264.4 g/(m^3 h)</td>
<td>Porosity of PM: 73.0%</td>
<td>[74]</td>
</tr>
</tbody>
</table>
Table 1. Cont.

<table>
<thead>
<tr>
<th>Packing Material; Inoculation</th>
<th>Bioreactor</th>
<th>Pollutant</th>
<th>Humidity Control</th>
<th>EBRT</th>
<th>ILR or IC</th>
<th>EC or RE</th>
<th>Packing Material Properties</th>
<th>References</th>
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</thead>
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<tr>
<td>Plastic biological ball filters (bioballs); <em>Pseudomonas putida</em>, <em>Rhodococcus aerolatus</em>, and <em>Aquaspirillum annulus</em></td>
<td>BTF</td>
<td>Acetone</td>
<td>Trickling liquid rate: 0.1 L/min</td>
<td>6.3–12.5 s</td>
<td>51.8–230.4 g/(m$^3$ h)</td>
<td>EC (max): 207.8 g/(m$^3$ h) at ILR of 230.4 g/(m$^3$ h)</td>
<td>SSA: 620 m$^2$/m$. Porosity of PM: 75%</td>
<td>[52]</td>
</tr>
<tr>
<td>Polypropylene rings, activated sludge</td>
<td>BTF</td>
<td>Styrene</td>
<td>Intermittent nutrient solution spraying (2.5–3 L per min for 15 min every 2 h)</td>
<td>30–60 s</td>
<td>13–41 g/(m$^3$ h)</td>
<td>11.8–31.8 g/(m$^3$ h)</td>
<td>Nominal diameter: 25 mm, SSA: 207 m$^2$/m$. Porosity: 92%</td>
<td>[81]</td>
</tr>
<tr>
<td>PU pall rings; H$_2$S degraders, <em>Pseudomonas oleovorans</em> DT4, <em>Methylobacterium rhodesianum</em> H13</td>
<td>BTF</td>
<td>H$_2$S, THF, and DCM</td>
<td>Continuous nutrient solution trickling</td>
<td>20–50 s</td>
<td>200, 100, and 100 mg/m$^2$ for H$_2$S, THF, and DCM, respectively</td>
<td>EC (max): 52.5 g/(m$^3$ h) (H$_2$S), 26.7 g/(m$^3$ h) (THF), 17.2 g/(m$^3$ h) (DCM)</td>
<td>SSA: 350 m$^2$/m$. Porosity: 91.3%</td>
<td>[82]</td>
</tr>
</tbody>
</table>

Legend: DCM: dichloromethane; GAC: granular activated carbon; EBRT: empty bed residence time; EC: elimination capacity; HDPE: high-density polyethylene; IC: inlet concentration; ILR: inlet loading rate; MEEK: Methyl ethyl ketone; PDMS: polydimethylsiloxane; PM: packing material; PU: polyurethane; RE: removal efficiency; SSA: specific surface area; TEA: triethylamine; TCE: trichloroethylene; THF: tetrahydrofuran.

Packing materials in biofiltration systems should have suitable porosity for efficient gas flow distribution and high specific surface area for development of microorganisms. Kim and Deshusses [83] determined gas and liquid film mass transfer coefficients for inert packing materials and found that porous ceramic beads had the highest ones followed by lava rock, porous ceramic rings, and PU foam cubes [83]. The use of homogenous, porous, and well-drained packing materials reduces the risks of excessive watering and formation of anaerobic zones [29]. The packing material should also have high water retention properties to avoid its drying [84]. Saravanan et al. [79] reported that agro-waste materials, pressmud and cornstack, have good water holding capacities, and were effective as packing materials in BF designed for MEK removal [79]. In another study, Pérez et al. [85] compared styrene removal by BFs packed with different organic materials, peat, and coconut fibre. It was found that peat BF was more efficient in pollutant removal, because peat had a higher water holding capacity and higher specific surface area than coconut fibre, which favoured microorganism growth [85]. In general, organic packing materials have high water holding capacity, which increases their swelling capacity. Therefore, high moisture content of organic packing material can result in a higher pressure drop of BF [16]. Some packing materials (especially inert) lack the necessary nutrients and need to be moistened with nutrient solutions to ensure high activity of microorganisms [10]. The configuration of the packing material is also important for even air, liquid, and biofilm distribution. Caicedo et al. [86] compared the performance of two of the same BTF units with different configurations of PU foam packing material. It was reported that BTF with structured monolith packing showed better performance in removing treated pollutants compared with BTF with randomly packed PU foam cubes. These results were attributed to improved liquid distribution in the structured packing material [86]. When BFs or BTFs are used with conventional packing materials, the first layer of the packing material often receives the highest inlet load of VOCs. This can result in its clogging and increase in the pressure drop. To solve this issue Baltrénas et al. [42] designed BFs with packing materials consisting of several plates with millimetre gaps between them (Figure 4). In such a plate-type biofilter, air can flow more smoothly and distribute more evenly over the entire volume of the packing material [42].
5.2. Influence of Packing Material Moisture Content on Biofiltration Efficiency

5.2.1. Biofilters

The efficiency of biofiltration depends on the moisture content of the packing material which affects gas–liquid mass transfer and the growth of microorganisms [87]. Water content of the biofilter packing material is an important parameter, which must be controlled in order to optimize the performance of BF [31]. The optimum moisture content for a biofiltration device depends on the characteristics of the targeted contaminant [29]. In general, hydrophilic and easily biodegradable compounds can be effectively removed in biofiltration devices. Xue et al. [30] studied removal of different pollutants (NH$_3$, SO$_2$, and total VOC) by two-stage thermophilic biofilter packed with PU foam cubes. The authors varied moisture content of the packing material from 40 to 85% (on a weight basis) by using different irrigation methods, which was periodical (twice a week) and continuous (at the rate between 0.05 and 0.42 m$^3$/h). It was found that SO$_2$ removal was not affected due to variation in packing material moisture content, while NH$_3$ removal increased from 66.41 to 89.75% with an increase in irrigation rate. Maximum removal efficiency of the total VOC was achieved when the irrigation rate of the packing material was 0.3 m$^3$/h and the moisture content of the packing material was 80% [30]. Similarly, Yang et al. [88] also observed an increase in NH$_3$ removal efficiency from 40 to 80%, when the humidity of the packing material (composed of compost and woodchip) was increased from 35 to 63% (wet basis) [88]. Auria et al. [2] investigated the removal of ethanol in a peat BF under various water contents of packing material (35–70%). Obtained ethanol elimination was similar (about 30 g/(m$^3$ h)) when the water content of the packing material was 49–70%. The lowest ethanol elimination capacity (4 g/(m$^3$ h)) was observed when the moisture of the packing material was decreased to 35% [2]. Sun et al. [17] treated toluene vapour with BF packed with mixture of compost and perlite (7:3, v:v). It was found that toluene degradation increased with the increase in the moisture content of the packing material from 30 to 70% (on a weight basis) [17]. Bagherpour et al. [31] examined the effect of moisture content of the packing materials on alpha-pinene removal through BF packed with compost and wood chips. The study concluded that an excess amount of water reduced the removal efficiency of the hydrophobic pollutant, since the thick water layer around the packing material caused higher resistance of pollutant diffusion [31]. Vergara-Fernández et al. [89] studied n-pentane removal through a fungal BF packed with vermiculite. The moisture content of the packing material varied between 20 and 80%. It was reported that the elimination capacity of n-pentane increased with the increase in moisture content of the packing material [89]. Rene et al. [90] studied the effect of different relative humidity of inlet gas on the moisture content of the packing material and performance of perlite BF. When the relative humidity of inlet gas was 30%, 60%, and >92%, the moisture of the packing material reached 23.4, 44.3, and 56%, respectively. With the decrease in moisture content of the packing material, styrene removal decreased from 100 to 82.3% and from 92.4 to 69.8% at inlet loading rates of 80 g/(m$^3$ h) and 260 g/(m$^3$ h). The drop of styrene elimination was explained by depletion of water and available nutrients in the BF [90]. Pandey et al. [91] treated pyridine (C$_5$H$_5$N) in BF packed with compost and wood chips. The biofilter was operated at different moisture contents of packing material (in the range of 37–95%). The maximum elimination of pollutant was obtained when the moisture content of the packing material was 68% [91]. Based on different studies it can be concluded that optimum moisture content of BF packing material is usually between 40% and 80%.

5.2.2. Biotrickling Filters

The key parameter that controls the moisture content of packing material in BTFs is the flow rate (L/h) of trickled nutrient solution. The liquid flow rate affects the moisture content of BTF packing material, and thus, the mass transfer of treated pollutants to biofilm. An optimum liquid flow rate depends on the solubility of a pollutant and its inlet loading rate [92]. Excessive liquid flow can produce a thick liquid layer that surrounds the biofilm and prevents the mass transfer of some hydrophobic gaseous contaminants, consequently
reducing the efficiency of treatment. In addition, a high trickling rate may increase the pressure drop of the filter and compaction of its packing material. In the case of treating hydrophilic compounds, significant biodegradation of pollutants can occur in the sump. In such cases, the increase in the liquid flow rate can significantly increase pollutant transfer to liquid and therefore its treatment efficiency. On the other hand, low liquid flow rate can cause drying of packing material and reduce activity of microorganisms. If the liquid flow rate is too low, the removal of produced acids and metabolites may not be sufficiently fast, and the pH can be reduced to a level that inhibits the growth of microorganisms [93].

Flow rates or velocities of trickling liquid (m/h) and nutrient solution renewal can be regulated to improve the removal efficiency of pollutants and optimise the costs for the BTF operation. In this sense, Caicedo et al. [86] studied toluene and ethylbenzene removal using BTF packed with PU foam. It was found that the increase in the trickling liquid velocity enhanced gas-liquid mass transport and increased removal efficiencies of pollutants. In addition, authors found optimal liquid renewal rates for efficient performance of differently packed BTFs [86]. Lu et al. [94] investigated BTEX removal by BTF operated under the co-current flow mode. The authors found that the increase in the liquid flow from 3.44 to 8.60 L/m$^3$/h resulted in the increase in BTEX removal efficiency. However, further increase in the nutrient flow rate to 17.2 L/m$^3$/h caused a slight decrease in those pollutant removals [94]. In another study [95], simultaneous removal of methanol (Cin = 140 ± 2 mg/m$^3$) and dimethyl sulphide (Cin = 75 ± 2 mg/m$^3$) through BTF at liquid flow in the range of 0.3–3.0 m$^3$/m$^2$/h was investigated. It was found that the increase in liquid flow rate from 0.3 m$^3$/m$^2$/h to 1.2 m$^3$/m$^2$/h resulted in the increase in removal efficiency of methanol and dimethyl sulphide from 94.9% and 32.7%, respectively, to 98.6% and 92.2%, respectively. There was no obvious effect on the efficiency of removing of methanol and dimethyl sulphide when the liquid flow rates exceeded 1.2 m$^3$/m$^2$/h [95]. Kim and Deshusses [83] studied the effect of liquid velocities on gas film mass transfer coefficients. It was found that mass transfer coefficients increased with an increase in liquid velocity at low liquid velocities (0.1−4 m/h), but the trend changed to constant at the liquid velocity of about 6.3 m/h. Several studies have concluded that trickling liquid velocity has no significant effect on biofiltration efficiency when the packing material is completely wet [83,96].

Intermittent nutrient solution spraying is the other important method for controlling moisture content in BTF packing material and improving removal efficiency of hydrophobic compounds. Such methods have been proposed as an effective strategy to prevent excessive growth of biomass. Such a spraying mode is usually applied in BTFs used in industrial conditions [97]. Yu et al. [98] studied n-hexane removal in BTF that was operated under intermittent nutrient spraying mode (4.5 L per day, and spraying control; 30 s on and 20 min off). It was reported that the elimination of pollutants (ECmax = 46.08 g/(m$^3$/h)) was enhanced due to the development of a thin water layer that facilitated the direct contact between the treated pollutant and the biofilm [98]. Alinejad et al. [99] reported that sudden liquid flow had no significant effect on toluene removal through BTF at the inlet load of 152 g/(m$^3$/h). While, at the inlet load of 320 g/(m$^3$/h), trickling liquid caused the decrease in toluene removal efficiency by 11% within 2 h, after the beginning of the flow [99]. Sun et al. [100] studied styrene removal through BTF in which microbial sludge was inoculated. Authors varied the humidity of ceramic pellet packing material (between 15.5% and 70%) by changing the liquid flow rate and found that styrene removal was the highest at the packing material humidity level of 41.3% [100]. Some studies also reported that the intermittent solution spraying mode helped to improve elimination of highly water-soluble compounds. San-Valero et al. [101] observed effective isopropanol removal using BTF operated under intermittent spraying (15 min every 1.5 h). The authors also reported that the decrease in the spraying frequency to 15 min per 3 h resulted in a decrease in the outlet isopropanol concentration from 86 to 59 mg C/Nm$^3$ [101]. Pérez et al. [97] studied 2-butoxyethanol removal using BTF packed with polyurethane foam. An activated sludge was used as inoculum and BTF was operated under the intermittent trickling mode.
The authors found that change in the irrigation frequency from 1 min every 12 min to 1 min every 2 h resulted in a decrease in the number of emission peaks at the outlet; therefore, the average 2-butoxyethanol removal efficiency increased [97].

Fungi can be more tolerant to dry conditions than bacteria and can be a suitable choice for VOC treatment in BTFs [44]. Zhang et al. [53] studied toluene removal through BTF inoculated with Fusarium oxysporum. It was observed that the decrease in humidity in the BTF column from 100% (spraying frequency 105 mL per 4 h) to 40% (spraying frequency 30 mL per 4 h) did not affect toluene removal efficiency, which remained at 100%. However, authors also observed that a long spraying interval (longer than 4 h) caused a decrease in VOC removal efficiency due to the lack of nutrients [53]. Li et al. [80] investigated styrene removal through BTF inoculated with fungi. High styrene removal efficiency (above 90%) was observed at the packing humidity range of 7.7% to 60.0%, while removal efficiency of this pollutant decreased when packing humidity was above 60% [80].

6. Conclusions

Different packing material irrigation strategies and their influence on various pollutant treatment by BF s and BTFs were presented in this review. The moisture content of BF packing materials can be controlled by changing the relative humidity of inlet gas, occasional spraying of packing, and immersing part or the entirety of the packing material in a nutrient solution. In the case of BTFs, the moisture content of packing material is controlled by changing the flow rate of trickling liquid or by applying the intermittent liquid trickling method. A literature review revealed that an intermittent liquid spraying can be an effective strategy for biofiltration of both hydrophilic and hydrophobic VOCs. A thin layer of water developed around biofilm facilitates contact of VOCs with biofilm.

Overall, the reviewed studies show that the effects of different strategies used for irrigation of packing material can be numerous. Therefore, it is often necessary to experiment with this parameter to adjust the operation conditions. It can be suggested that the optimum packing material irrigation rate should be found experimentally. It seems that passing contaminated gas through the water layer that is present below the packing material can be an effective method for increasing the treatment efficiency of hydrophilic VOCs through both BF s and BTFs. However, more research is needed to determine the feasibility of this strategy. In addition, only few studies have applied flooding as the irrigation method of BF or BTF packing material. Therefore, more future research that compare different packing material irrigation systems (e.g., conventional vs. flooding) should be conducted for both BF s and BTFs.

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