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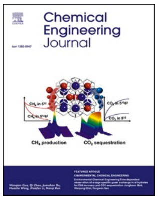

Review

Volatile organic compound removal via biofiltration: Influences, challenges, and strategies

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ABSTRACT

Biofiltration is an established technology for the removal of volatile organic compounds (VOCs) from waste gas streams due to its cost-effectiveness, process safety, and environmentally soundness. However, the performance and applications of biofilters still need improving. In biofiltration for VOC removal, the performance could be affected by several design and operating factors, and there are some prevailing problems which need overcoming. In this article, the state of the art and development trends of biofiltration for VOC removal were comprehensively reviewed and commented on. First of all, the factors affecting the removal performance of VOCs by biofiltration were reviewed and presented which include the properties of VOCs and design and operating parameters of biofilters. Then, the prevailing problems in biofilters were summarized and discussed, such as the uneven distributions of biomass and VOC loads as well as nutrients, unsatisfactory removal for VOCs at high organic loads and with hydrophobicity, and inconsistent interactions among multicomponent VOCs. At last, the corresponding solutions for the problems were proposed and commented on. This review could be referred for investigations and applications of biofiltration in future.

1. Introduction

Volatile organic compounds (VOCs) are widely derived from oil refining [1], wastewater treatment [1,2], consumer products [3], chemical and printing industries [4], paint and coating industries [5], drying processes, agricultural stocks, textile industries [6], etc. The massive emission of VOCs has caused severe pollution to the atmospheric environment and posed a serious threat to human health [4,5,7]. With ultraviolet (UV) light as the energy source, VOCs can react with nitrogen oxides to produce ozone and secondary aerosols in the atmosphere, leading to haze [8–11]. Much worse, VOCs could damage the nervous system of the human body, with carcinogenic, teratogenic, and mutagenic effects [5,11–15]. Therefore, VOC pollution has attracted the attention of the government and citizens [16,17].

Developed countries and regions, such as the United States, the European Union, and Japan, have earlier carried out the work of VOC emission control. The traditional treatments for VOC removal mainly include biological methods and physical–chemical methods [6,18–21], such as biofiltration [11,18], condensation [22], absorption [23], membrane processes [24], thermal combustion [25], non-thermal plasma [26,27], and UV-oxidation [28]. Among them, thermal combustion is an optional technology for the efficient treatment of high-concentration VOCs, which can completely convert VOCs into carbon dioxide, but it also needs to consume a lot of energy, with certain potential safety hazards for some specific industries, such as petroleum and chemical industries [4,29]. In biological methods, microorganisms can use VOCs as a carbon source and energy to convert them into water, carbon dioxide, and biomass. Compared with the physical–chemical methods, the biological methods have the advantages of economy, energy conservation, and process safety and there are different approaches to optimize performance and operation costs [2,6,29–31]. More importantly, the biodegradation of VOCs is of great significance for

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mitigating the greenhouse effect.

Biofiltration is the most widely used biological treatment technology in VOC removal [32,33]. In biofilters, VOCs in waste gas streams could pass a thin liquid phase covering the porous biofilm to get in contact with the bacterial cells itself, and then VOCs would be degraded and removed by microorganisms in the biofilm (Fig. 1) [34,35]. In the 1950s, biofilter was applied to waste gas treatment [30]. The United States and Germany submitted the first application and patent for biofilters. In 1977, Bohn and Bohn [30] designed the first soil biofilter for removing organic waste gases. Therefore, gas-phase biofiltration has developed for more than 70 years in waste gas treatment [30,36,37] (Fig. 2). Many studies and reports on the removal performance and corresponding mechanisms of biodegradation of VOCs by biofiltration have emerged. It could be found that the removal of VOCs by biofiltration was easily affected by many factors, such as the operating conditions including empty bed residence time (EBRT), temperature, nutrient pH, and salinity [38–41]. Besides, the structure and properties of VOCs, and the microbial community structure could also affect the VOC biodegradation [21,34,42,43]. Cheng et al. [43] analyzed the impacts on the hydrophobic VOC removal by biofiltration as early as 2016 from the four main factors of pollutant mass transfer, packing media, microbial community, and water. However, many pieces of research on the removal of VOCs by biofiltration show that the removal performance of VOCs by biofiltration is not only affected by the VOC properties but also closely influenced by the structure, operation mode of the biofiltration system, as well as operating conditions [14,44–47]. Unfortunately, there is still a lack of comprehensive and detailed reviews in this area.

In addition, in the development of biofiltration, some problems are exposed. Among them, the excessive accumulation of biomass is the most common problem in traditional biofilters [43,48]. It would lead to the blockage of the packed bed, and increase the pressure drop and energy consumption [43]. Moreover, it would also result in the decline of VOC removal and the instability of the biofilter system [44]. In 2010, Yang et al. [48] reviewed the excessive biomass accumulation model and proposed the corresponding control measures in detail. However, there are still other problems in the biofiltration process, such as uneven distribution of biomass and nutrients, not being suitable for treating high-concentration organic gases, etc. Unfortunately, a detailed review of these common problems and their corresponding solutions is rare. As a result, the public lacks a comprehensive understanding of biofiltration, which leads to the failure to achieve the desired purpose when using biofiltration to remove VOCs.

Therefore, according to the research results obtained by many domestic and foreign scholars in VOCs removal by biofiltration, we comprehensively summarized and reviewed the influencing factors of VOCs removal by biofiltration, systematically analyzed the common problems in the process of gas-phase biofiltration, and put forward corresponding solutions to these existing problems. Hopefully, the public can have a more comprehensive understanding of VOC removal by biofiltration. In addition, this article can be referred for the future design, investigation, and practical application of gas-phase biofiltration.

2. Main factors affecting removal of VOCs by biofiltration

The ideal removal performance of biofilters for VOCs is often determined by multiple factors. Understanding the main factors that affect the removal performance of biofilters is critical for the rational design of bioreactors, optimization of the operation process, and improvement of VOC removal performance. Generally, the main factors affecting the removal of VOCs by biofiltration include the inherent properties of VOCs, characteristics of the packing materials, microbial populations the operation mode and operating conditions of the biofiltration system, etc. (Table 1, Fig. 3).

2.1. Properties of VOCs

2.1.1. Hydrophilicity of VOCs

At normal temperatures, the organic matter with a boiling point from 50 to 250 ℃ is defined as VOC by the World Health Organization. There are a wide variety of substances that satisfy this definition, such as alkanes, alkenes, alcohols, aldehydes, ketones, etc. The properties of different kinds of VOCs vary greatly. The removal performance of biofilters for different types of VOCs also presents a big difference. Therefore, the physico-chemical properties of VOCs are usually important factors influencing the removal performance of biofilters [47,49]. In biofiltration, the mass transfer process from gas to liquid and biofilm, and the VOC specific degradation kinetics in microorganisms are the two main processes for VOC degradation. The hydrophilicity of VOCs, Henry's Law constant, saturated vapor pressure, dipole moment, and other properties characterizing the water solubility and bioavailability of VOCs play a vital role in the mass transfer and reaction kinetics of biofiltration [1,43].

Generally, the hydrophilic VOCs have higher water solubility since they have a smaller dimensionless Henry's Law constant at 25 ℃ ($H \leq 0.1$) or their saturated vapor pressure is lower than 30 kPa [43,50]. As a result, the mass transfer resistance of hydrophilic VOCs in biofiltration is significantly lower than that of hydrophobic VOCs. It implies that the bioavailability of hydrophilic VOCs is higher than that of hydrophobic VOCs, which can lead to a faster mass transfer rate of hydrophilic VOCs in biofiltration than that of hydrophobic VOCs [51,52]. Thus, under the same conditions, the removal performance of biofilters for hydrophilic

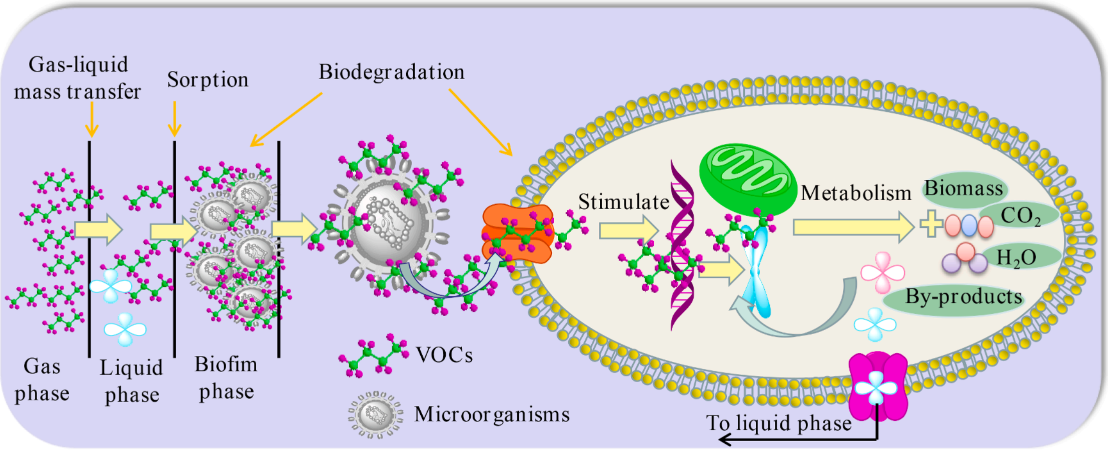

Fig. 1. The migration and transformation process of VOCs in biofilters.

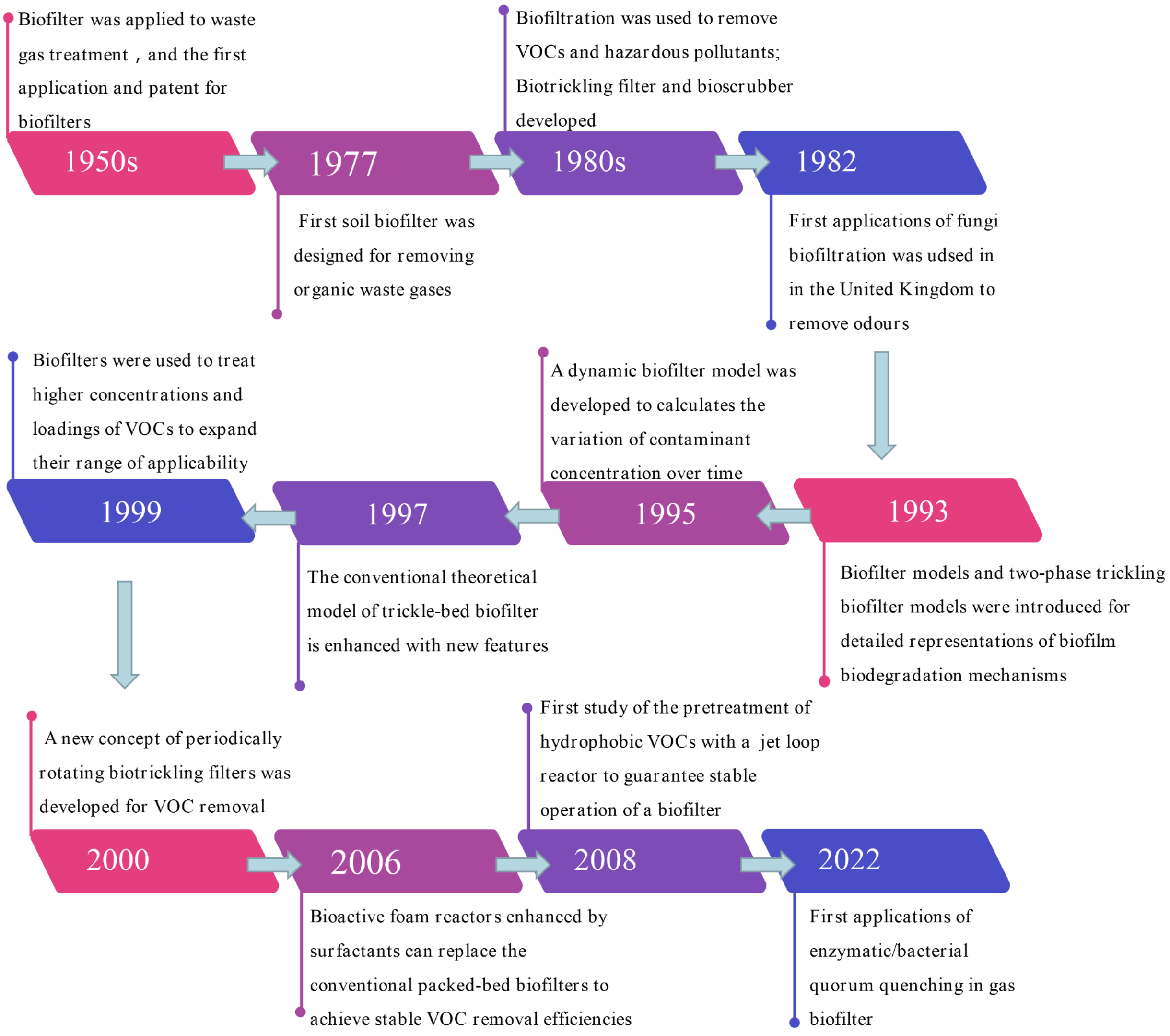

Fig. 2. A timeline showing milestones in the development of biofiltration techniques.

VOCs can be better than hydrophobic VOCs. In the investigation of Cheng et al. [44], they observed that the removal efficiency (RE) of hydrophilic VOC could be maintained at 100% after two days of a startup, while the corresponding RE of hydrophobic *n*-hexane fluctuated between 38 and 43%. In addition, Pittit et al. [49] found that the removal performance of hydrophilic VOCs with larger dipole moments (acetone: 2.88, ethanol: 1.69, ethyl acetate: 1.78, and isopropanol: 1.66) was significantly better than that of hydrophobic VOCs with smaller dipole moments (benzene: 0.00, cyclohexane: 0.33, isopentane: 0.13, hexane: $\leq$0.10 and toluene: 0.36) in plant biofiltration. Moreover, through multiple stepwise linear regression, it was determined that dipole moment and molecular weight are important factors affecting VOC removal, accounting for 54.6% of the variability in VOC removal efficiency [49].

The remarkable advantage that hydrophilic VOCs are almost not limited by mass transfer resistance makes biofiltration a good application prospect in hydrophilic VOC removal. Sun et al. [53] reported that the removal performance of biofilters for hydrophilic VOCs was significantly higher than that of hydrophobic VOCs when employing biofilters for deodorization. Among them, the elimination capacity (EC) of biofilters for hydrophilic propanethiol was as high as 204.1 g/m^3·h, and it was 2.25 times higher than that of moderately hydrophilic toluene (90.9 g/m^3·h) [53]. In the study of Cheng et al. [44], they found that toluene could be completely removed by biofiltration when toluene and *n*-hexane co-existed and that the removal performance of *n*-hexane was lower in the presence of toluene with a high proportion than that single *n*-hexane as a pollutant. These results indicate that the hydrophilicity of VOCs has an important influence on VOC removal in biofilters. Especially when the waste gas streams contain hydrophobic VOCs, as the presence of the higher gas–liquid mass transfer resistance, the removal performance of biofilters for hydrophobic VOCs often cannot meet the satisfaction of the researcher. Therefore, appropriate measures should be taken to promote the removal of hydrophobic VOCs.

2.1.2. *Chemical structure of VOCs*

In addition to the hydrophilicity of VOCs, the chemical structure of VOCs can also affect the removal performance of biofilters. It is well known that VOCs with different chemical structures have different physical and chemical properties. Wu et al. [1] have reported that the *n*-alkane removal in biofilters was related to their chain lengths. Since the octanol–water partition coefficient of *n*-alkanes decreases with the chain length decreases, the absorption capacity of microorganisms for short-chain alkanes reduces.

Except for the influence of chain length, side chain or ring structure could also affect the removal of VOCs by biofiltration. Cheng et al. [54] reported that when the EBRT was 30 s, the EC of the bacterial biofilter filled with polyurethane sponge for *n*-hexane can achieve 67.6 g/m^3·h. While under similar conditions, the bacterial biofilter using cyclohexane

Table 1

Effects of different factors on the removal of VOCs by biofiltration.

Target pollutant	Hydrophilic performance[a]	Packing material	Inoculum	Operation conditions	Gas-liquid flow direction	Removal performance	Reference
n-hexane	hydrophobic	polymeric sponge	activated sludge	IL: 58 ± 3.0 $g/m^3 \cdot h$; EBRT: 30 s	counter-current	RE: 77 ± 1.3%; EC: 45 $g/m^3 \cdot h$	[1]
methane						RE: 35 ± 5.6%; EC: 20 $g/m^3 \cdot h$	
n-hexane	hydrophobic	polymeric sponge	filamentous bacteria from previous experiments	IL: 62 ± 4.3 $g/m^3 \cdot h$; EBRT: 30 s	co-current	RE: 64 ± 3.3 %; EC: 40 $g/m^3 \cdot h$	[57]
cyclohexane	hydrophobic	polymeric sponge	*Candida albicans* and *Candida subhashii*	IL: 45 $g/m^3 \cdot h$; EBRT: 60 s	counter-current	RE: 20%-35%; EC: 9–16 $g/m^3 \cdot h$	[45]
ethanol	hydrophilic			IL: 37 $g/m^3 \cdot h$; EBRT: 60 s		RE: 99%; EC: 37 $g/m^3 \cdot h$	
n-hexane	hydrophobic	polymeric sponge	activated sludge	IL: 16 $g/m^3 \cdot h$; EBRT: 60 s	co-current	RE: 82%; EC: 13 $g/m^3 \cdot h$	[44]
methyl isobutyl ketone	hydrophilic			IL: 4–32 $g/m^3 \cdot h$; EBRT: 60 s		RE: 100%; EC: 4–32 $g/m^3 \cdot h$	
toluene	moderately hydrophilic			IL: 5–58 $g/m^3 \cdot h$; EBRT: 60 s		RE: 100%; EC: 5–58 $g/m^3 \cdot h$	
hexane	hydrophobic	peat-perlite and ceramic Raschig rings	without special inoculation (naturally present diversified microflora in peat-perlite)	IL: 35 $g/m^3 \cdot h$; EBRT: 60 s	counter-current	RE: 90%; EC: 32 $g/m^3 \cdot h$	[60]
ethanol	hydrophilic			IL: 18 $g/m^3 \cdot h$; EBRT: 60 s		RE: > 99%; EC: 18 $g/m^3 \cdot h$	
n-hexane	hydrophobic	compost and Kaldnes rings	a hydrophobic bacterial consortium from the reactor treating *n*-hexane	IL: 30 $g/m^3 \cdot h$; EBRT: 75 s	counter-current	RE: 52%; EC: 16 $g/m^3 \cdot h$	[62]
methanol	hydrophilic	steel pall rings and pumice grains	activated sludge acclimated with methanol	IL: 7.5–453 $g/m^3 \cdot h$; EBRT: 120 s	counter-current	RE: 90–100%; ECmax: 411 $g/m^3 \cdot h$	[90]

[a] The hydrophilic performance is divided according to the dimensionless Henry's Law constant (H) of VOCs at 25 ℃: the H lower than 0.1 is hydrophilic VOCs, the H between 0.1 and 1.0 is moderately hydrophilic VOCs, and the H higher than 1.0 is hydrophobic VOCs.

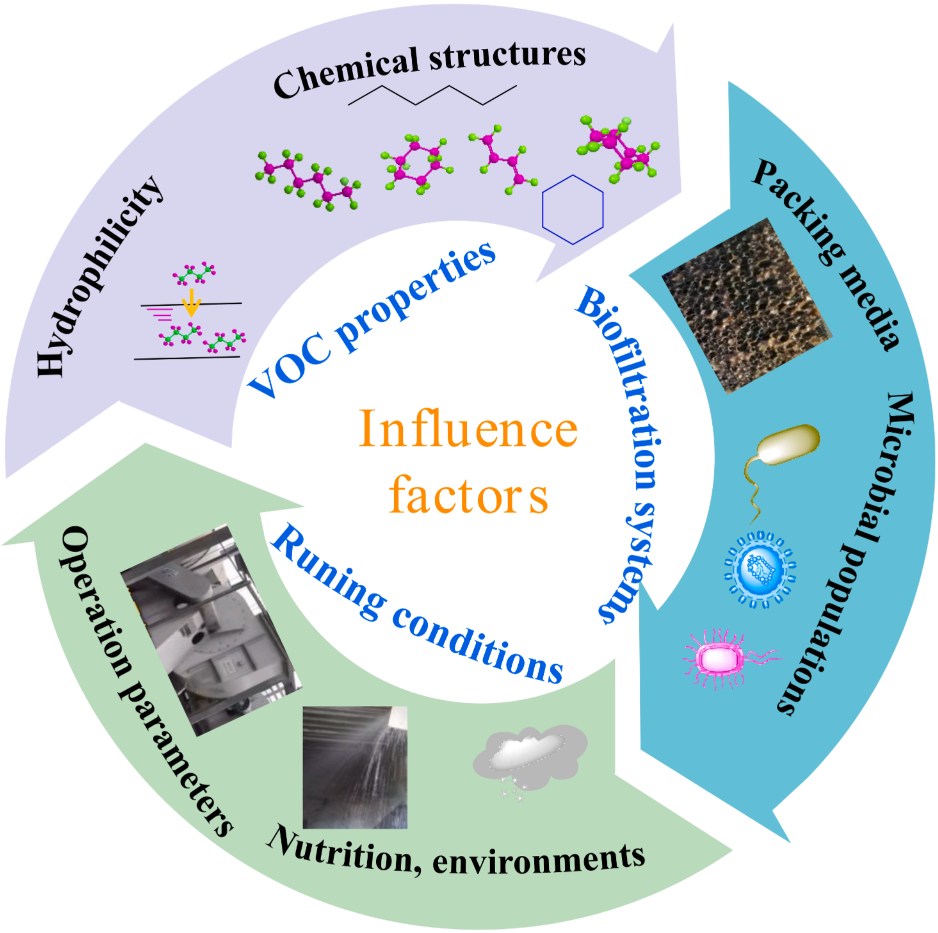

Fig. 3. Main influence factors in VOC removal by biofilters.

as the substrate obtained a maximum EC of 38.0 g/m^3·h, which is only 56% of the EC of *n*-hexane [55]. The main reason for this result is that the biodegradability of cyclohexane is lower than that of *n*-hexane. Compared with *n*-hexane, cyclohexane molecules are arranged more closely, the intermolecular force is higher, and the surface tension is greater (*n*-hexane: 20.3 dyne/cm, cyclohexane: 25.9 dyne/cm). These properties resulted in more energy consumption by microorganisms to realize the utilization of cyclohexane. Moreover, bacteria need some additional enzymes to degrade the cyclohexane in comparison to the *n*-hexane, i.e. cyclohexane monooxygenase, cyclohexanol dehydrogenase, and a Baeyer-Villiger monooxygenase [56].

In a word, the properties of VOCs are crucial to their removal from waste gas by biofiltration. Therefore, in actual industrial production, it is very important to conduct a comprehensive analysis and monitoring of pollution sources. Mastering the inherent properties of pollutants is not only conducive to the selection of appropriate waste gas treatment but also beneficial to adopt reasonable strengthening measures to improve their removal.

2.2. *Biofiltration systems*

Biofilters are the main places where VOCs are degraded by microorganisms. Different structures, compositions, and operation modes of biofilters will produce diverse influences on VOC removal.

2.2.1. *Packing media*

Packing media can support microorganisms to form biofilm in bioreactors [57]. According to the properties of the packing media, the packing media usually used in biofiltration can be divided into organic and inert packing media [29,58,59]. Among them, packing beds filled with organic packing media, such as compost peat, humus, bark, etc., generally contain rich microbial species and some nutrients [31,34,57,59]. This type of biofilter does not require additional inoculation of microorganisms. Because biofilters filled with organic packing media can realize the mineralization and removal of VOCs through the metabolism of indigenous microorganisms [60]. Since organic packing media are economical and easy to obtain, thus, organic packing media are widely used in biofiltration [59,61,62]. Lebrero et al. [62] built a compost-based biofiltration system to remove *n*-hexane with an inlet load of 30 g/m^3·h and EBRT of 75 s, the RE and EC of *n*-hexane reached 52% and 16 g/m^3·h, respectively, and the rapid generation of carbon dioxide was monitored in the biofilters.

However, with the development of biofiltration, the problems of the organic packing media-filled biofilter are exposed, such as the biodegradation of the organic packing media, the compaction of the medium beds, and with high-pressure drop [43,48,59]. Those problems can lead to a reduction in VOC removal performance in biofilters [43]. In order to solve the above problems, inert fillers with porous structures or enhanced physical stability, such as perlite [63], shells [64], lava rock [65], or even hybrids inert/organic packing materials [21,66] are applied. Besides these materials, inert fillers with high porous structure or network structure, such as polyurethane sponge, Bauer ring, plastic hollow ball, glass bead, and modified polystyrene scaffolds [57,58,67,68] are also used, whereby the transition to biotrickling filter technologies is hereby achieved and the process differentiation is then solely limited to irrigation parameters. Compared with biofiltration based on organic packing media, biotrickling filtration is easier to control pH, temperature, water content, and other parameters [30,50]. In addition, the selection of inert fillers with high mechanical strength can often keep the process stable for a long time [35,59]. Merouani et al. [2] used biofilters filled with inert fillers with different particle sizes and investigated the removal performance of biofilters for the mixed pollutants of methane and ethylbenzene or xylene. They observed that the biofilter ran for nearly 300 days, and achieved a satisfactory removal effect of methane and VOCs [2]. It is because that the appropriate reticular structure or porous structure is conducive to promoting the uniform growth of microorganisms in the packed beds and the uniform distribution of airflow and nutrients. Yu et al. [57] investigated the removal of *n*-hexane in biofilters filled with reticulated polyurethane sponge, they found the RE of *n*-hexane remained stable even at a low EBRT (7.5 s). It was attributed to the good buffering capacity of reticulated filler for high-speed airflow. Meanwhile, the reticular structure of polyurethane sponge promoted the uniform distribution of microorganisms on the packing beds and improved the mass transfer efficiency between microorganisms and VOCs. Nevertheless, it should be pointed out once again that the use of polyurethane sponge or other plastic scaffolds in particular marks a transition from the basic principle of the biofilter into biotrickling filter technologies.

Besides the influence of the type and structure of packing media on the removal of VOCs by biofiltration, the density, size, humidity, retention, porosity, specific surface area, and other surface properties of packing media also have a significant impact on the removal of VOCs [50,62,69]. Applicable packing media should generally have the following characteristics: 1) High mechanical strength to prevent performance decrease caused by compaction of packing beds; 2) Good biological compatibility and high specific surface area to promote the formation of biofilm and shorten the start-up time of reactors; 3) Poor-biodegradability to prevent the collapse of the biofilters caused by microbial degradation; 4) Low price to save construction cost [57,58,69].

In the practical application of biofiltration, the mixed media composed of organic media materials and inorganic materials may present a better performance for VOC removal [66,70]. Because on the one hand, it can use the indigenous microorganisms and nutrients in the organic media to accelerate the spread of microorganisms and the formation of biofilm. On the other hand, it also can use porous inert material with high mechanical strength to promote the uniform distribution of biofilm, prevent the compaction of packing beds, and improve the removal of VOCs by biofiltration.

2.2.2. *Microbial populations*

Microbial populations are the main force of VOC degradation in biofiltration [18,30]. VOCs in the gas streams can reach the microbial cells through the mass transfer from a gas phase to the liquid phase and biofilm and are then degraded through the assimilation of microorganisms [30,71].

In the studies of biofiltration, many researchers use specific strains to carry out biofiltration research on VOC removal [2,72–74], which also might be an adequate approach for large-scale systems, even though of high costs, in case of biodegradation of severely xenobiotic compounds like methyl *tert*-butyl ether, or o-chlorotoluene [75,76]. However, it can be observed that the biodegradation of VOCs is commonly the result of the joint action of multiple microorganisms when dealing with common industrial waste gas [29,77–80]. Therefore, when biofilters are filled with inert fillers, the microorganisms inoculated are always activated sludge from the municipal sewage treatment plant or from other biofilters that treat similar pollutants [16,30,44]. This is because the high microbial diversity in the mixed sludge is beneficial for biofilters to resist the impacts of adverse environments [18,81].

In addition, microorganisms that contribute a lot to the biodegradation of VOCs are the dominant microbe. Although some microorganisms can degrade multiple VOCs, the corresponding dominant microorganisms may present a difference due to the wide variety of VOCs. The VOC removal performance of biofilters with different microbial populations would also be diverse. In conventional biofiltration, the most dominant microorganisms for VOC degradation are mixed culture bacteria [29,77,80,82]. However, in the studying of hydrophobic VOC removal by biofiltration, many researchers usually inoculate fungal strains to biofilters to obtain better removal performance of biofilters for hydrophobic VOCs [72,83–85]. On the one hand, fungi can improve the resistance of biofilters to adverse environments, such as acid and dry conditions [43,83]. Furthermore, fungi show lover substrate yields than bacteria, and the risk of clogging is severely reduced.

On the other hand, fungal aerial mycelia can improve the mass transfer efficiency of hydrophobic VOCs in biofiltration [30,43]. Moreover, higher extracellular polymeric substance (EPS) content and the synthesis and release of surfactant are conducive to promoting the biodegradation of hydrophobic VOCs [30]. Rybarczyk et al. [45] inoculated fungal strains (*Candida albicans* and *Candida subtilis*) in biofilters, making biofilters obtain a high cyclohexane removal. The EC of cyclohexane reached 67.5–89.0 $g/m^3 \cdot h$, which is significantly higher than the EC of biofilters inoculated with bacteria [45,55]. For example, Salamanca et al. [55] inoculated the biofilter with the bacteria *Acidovorax* sp. CHX100 which can degrade short-chain alkanes, while the EC of cyclohexane was only 5.4–38 $g/m^3 \cdot h$. However, the negative aspects of fungal biofilters should also be noticed, such as secondary emissions of odor (fungal smell), and stronger degradation of the organic biofilter materials, resulting in a shorter lifetime and higher pressure losses when a dryer operation process occurred.

Although fungi have good potential to remove VOCs in biofiltration, the microbial community, whether inoculated with fungi or bacteria or a mixture of them, will change with time and environmental conditions during the operation of biofilters. Over long periods of time, the share of secondary and ternary degraders will increase and VOC degraders (that are the primary ones) decline in share lower than 10 % in the mixed culture. That is, the microbial community would develop toward a community structure conducive to VOC degradation. Moreover, microorganisms are also sensitive to changes in environmental conditions and operating conditions. The variation of VOCs types or the inlet concentrations, loads, the introduction of additional additives, and the change of environmental temperatures or nutritional conditions all will significantly change the microbial community structure so that the microbial community structure can maintain a dynamic balance during the operation of biofilters. Wu et al. [1] found that under aerobic conditions, among the inoculated sludge, the abundance of anaerobes-like microorganisms (including *norank_f_Saprospiraceae*, *Terrimonas*, and *Ferruginibacter*) declined sharply. During the biofiltration with different short-chain *n*-alkanes as the target pollutants, the abundance of *Bacillus* increased significantly and *Bacillus* became the main dominant microorganism for the biodegradation of short-chain *n*-alkanes [1]. *Bacillus* contains the cytochrome P450 gene, the alkane environment is beneficial to improve the activity of alkane hydroxylase, then improve the removal performance of biofilters for *n*-alkanes. In addition, the microbial community structure along medium bed heights would be different, as the nutrient conditions, VOC concentrations, and VOC loads along medium bed heights are varied. Through the analysis of the microbial communities in the packing layers of different heights, Wu et al. [18] found that the abundance of *Sporosarcina* increased significantly along different medium bed heights. This is because the surfactant sprayed down from the top of the packing bed will present a toxic effect on microorganisms, while *Sporosarcina* is a microorganism with spores, which can protect microorganisms from adverse environmental effects. In addition, many studies have shown that usually most of the hydrophilic VOCs can be removed near the entrance, and the removal of hydrophobic VOCs is mainly concentrated in the middle and the outlet part of the biofilters when hydrophobic and hydrophilic VOCs were simultaneously removed by biofilters [44,61,86,87]. This is due to that hydrophilic VOCs are easy to compete with hydrophobic VOCs and inhibit the biodegradation of hydrophobic VOCs at the entrance, resulting in the microbial community being more adapted to the removal of hydrophilic VOCs at the inlet, while the microbial community at the middle and outlet is mainly adapted to hydrophobic VOCs removal [61].

In the process of VOC removal by biofiltration, it is necessary to analyze the microbial community and identify the dominant microorganisms and the corresponding microbial characteristics [73,77,80]. The microbe is the decisive factor for VOC degradation by biofiltration. According to the types and properties of VOCs in the waste gas, targeted inoculation of VOCs efficient degrading bacteria can shorten the start-up time of biofilters and achieve efficient and rapid removal of VOCs. In addition, the operating conditions can be better controlled according to the living habits of the dominant microorganisms to achieve efficient and stable operation of biofilters.

2.2.3. *Operation modes*

In addition to packing media and microbial populations, the operation mode of biofilters is another vital factor affecting the removal of VOCs by biofiltration. However, the impact of operation mode on biofiltration has not been specifically involved or discussed in previous studies. In the experimental research, most biofilters used for VOC removal are cylindrical, the inlet is basically set at the top or bottom of biofilters, and the nutrient solution supply is generally sprayed from the top of the biofilter [44,57,61]. Therefore, there are two types of contact between the waste gas and liquid nutrient solution and they are counter-current and co-current [61]. Although few scholars have set up experiments to study the differences between the counter-current and co-current operation modes, by comparing similar research results, it can be found that the operation mode of counter-current can make the gas–liquid contact more sufficient than the co-current, which may be more conducive to the mass transfer of VOCs in biofiltration. Thus, the counter-current operation modes may be more conducive to the removal of VOCs by biofiltration [44,57]. Yu et al. [57] investigated the removal of *n*-hexane by biofilter with the co-current operation mode and found that the RE of *n*-hexane could reach 63.8 ± 3.3% when the EBRT was 30 s, and the inlet load of *n*-hexane was 61.59 $g/m^3 \cdot h$. Under similar biofiltration conditions (polyurethane sponge as the packing medium, *n*-hexane inlet load of 58 ± 3.0 $g/m^3 \cdot h$, and EBRT of 30 s), Wu et al. [1] achieved a RE of 77 ± 1.3% for *n*-hexane by a biofilter with the counter-current operation mode.

Additionally, when studying the mixed VOC removal by biofiltration, some investigators observed that the different startup modes of biofilters also affected the removal of VOCs. Rybarczyk et al. [45], respectively, compared the effects of a startup mode introducing ethanol after initial startup with cyclohexane as the sole VOC to the situation starting up directly with mixed ethanol and cyclohexane on the removal of the mixed VOCs. Their research results show that, for the former startup mode, although the removal of cyclohexane enhanced after the introduction of ethanol, compared with the latter startup mode, the enhancement effect of ethanol on cyclohexane removal was far less than that of the mode started by mixed VOCs. As under the same steady-state conditions, the EC of cyclohexane in the latter startup mode was as high as 89.0 $g/m^3 \cdot h$, which was 1.32 times higher than that of the former startup mode (67.5 $g/m^3 \cdot h$) [45].

From the above cases, it can be observed that both the modes of contact between gas and liquid and the startup modes would affect the removal performance of the biofilter for VOCs. In contrast, for the modes of gas–liquid contact, the effect of the counter-current operation mode is usually better than that of the co-current operation mode. In addition, the staged gas inlet should also be a good choice. Especially when inlet VOC loads are high, the staged gas inlet can make the organic load distribution in biofilters more uniform, mitigate the impact of high organic load on biofilters, and maintain the stability of biofilters. Unfortunately, in the current studies, there are few reports on the effect of the staged gas inlet on VOC removal. For the startup mode of biofilters, the actual industrial waste gas is a mixture composed of multiple VOCs. Starting the biofilter directly with mixed VOCs is not only more practical but also conducive to the co-metabolism of mixed VOCs, as well as more efficient for the removal of VOCs. This phenomenon is especially significant in the presence of mixed VOCs with different hydrophilicity. Because the presence of hydrophilic VOCs can improve the bioavailability of hydrophobic VOCs, and it is beneficial to realize the co-metabolism of hydrophilic and hydrophobic VOCs [44,45,88]. In addition, the existence of hydrophilic VOCs can also improve metabolic activity and increase the demand of microbes for carbon sources and energy, which can further improve hydrophobic VOC removal in biofiltration [44,89].

2.3. Operation conditions

2.3.1. VOC inlet concentrations, loads, and EBRT

The diffusion rate of VOCs in biofiltration is not only related to the properties of VOCs but also the concentration or load of VOCs in the exhaust gas. For hydrophilic VOCs, low inlet concentrations or loads have little effect on gas–liquid mass transfer [44]. Therefore, it usually shows good performance by biofilters for the removal of low-concentration hydrophilic VOCs [2,45,90]. In the investigation of methanol removal by biofiltration, Ramezani et al. [90] found that the low-concentration methanol (250 ± 50 mg/m^3) was completely removed in the start-up stage. For hydrophobic VOCs, when the inlet concentration or load is low, the removal performance of biofilters can be limited by the diffusion mass transfer rate [43,84], which can increase by enhanced VOC inlet concentrations or loads within a certain range. Zhai et al. [84] studied the performance of biofilters for toluene removal and observed that the RE of toluene was about 80% when the EBRT was 45 s and the inlet concentration of toluene was 271.4–293.3 mg/m^3. However, when the concentration of toluene increased to 946.7–1086.8 mg/m^3, the RE of toluene increased to about 92 ± 1.0%. This may be due to the biofiltration was still mass transfer-limited when the inlet concentration increased from 271.4 to 293.3 to 946.7–1086.8 mg/m^3 [84]. Generally, typically RE would drop with VOC loads increasing when biofiltration is kinetic-limited.

However, whether it is hydrophilic VOCs or hydrophobic VOCs, the inlet VOC concentrations or loads should not be too high, because high VOC concentrations are likely to have toxic effects on microorganisms or exceed the biodegradability of microorganisms [3,10,14,90]. Merouani et al. [2] reported that the removal performance of biofilters for ethylbenzene decreased by 55% when the inlet load increased from 12 ± 0.2 to 32 ± 0.4 g/m^3·h due to it exceeding the biodegradability of microorganisms. In addition, high VOC concentrations or high loads may also easily cause excessive accumulation of microorganisms, which may lead to blockage of the reactors and performance degradation [43,68,91]. Given the study of Ramezani et al. [90], it could see that when the inlet concentration of methanol was up to 4500 ± 330 mg/m^3, the complete removal of methanol was achieved, while when the inlet concentration increased to 9160 ± 910 mg/m^3, excessive accumulation of biofilm occurred, and the pressure drop of the biofilter increased, and thus the RE of methanol reduced to 52 ± 5%. For hydrophobic VOCs with high inlet loads, especially hydrophobic VOCs with low biodegradability, although the diffusion rate can be accelerated under high inlet loads, the removal of VOCs would be limited by the assimilation reaction of microorganisms, and the removal performance of biofilters still would be reduced [14,57,84]. From the perspective of the study of Yu et al. [57], they found the RE of *n*-hexane was obviously reduced from 92.6 ± 0.6% to 63.8 ± 3.3% when the inlet *n*-hexane load increased from 15 to 60 g/m^3·h.

The above cases showed that the inlet concentration or inlet load of VOCs is an important parameter affecting the performance of biofilters for VOC removal. Therefore, before biofiltration is adopted to treat VOCs in the actual industrial waste gas, it is crucial to monitor the VOC concentration in the waste gas in order to reasonably design a suitable biofilter to achieve efficient removal of VOCs.

In addition to the influence of inlet VOC concentrations and loads, the EBRT is another important factor affecting the performance of biofilters in removing VOCs [10,44,92]. The relationship between the EBRT and the inlet concentration and load is shown in Equation (1), and the relationship between the EBRT and concentration and EC is shown in Eq. (2) and Fig. 4.

$$IL = C_iQ/V = C_i/EBRT \tag{1}$$

$$EC = Q(Ci - Co)/V = (Ci - Co)/EBRT \tag{2}$$

Where, IL is inlet load, g/m^3·h; EC is the elimination capacity of biofilter for VOCs, g/m^3·h; Ci is inlet concentration, mg/m^3; Co is outlet concentration, mg/m^3; Q is inlet flow rate, m^3/h; V is the volume of packing media, m^3; EBRT is the empty bed retention time, h.

Generally, the larger the EBRT is, the more conducive to the removal of VOCs under the condition of keeping the inlet VOC concentrations unchanged [71,93]. Fig. 4 shows that the outlet concentration of hydrophobic VOCs significantly decreases with the increase of EBRT when the inlet concentration of hydrophobic VOCs remains unchanged and biofiltration is mass transfer-limited. However, under kinetic-limited, the outlet concentration gradually stabilizes with the increase of EBRT. As for the RE and EC of hydrophobic VOCs by biofilters, they significantly increase with the increase of EBRT when biofiltration is mass transfer-limited but tends to stabilize in kinetic-limited. Therefore, increasing the EBRT is beneficial for promoting the removal of hydrophobic VOCs in the mass transfer-limited stage. Because increasing the EBRT is conducive to the sufficient contact of VOCs with packing media and microorganisms, and can promote the bioavailability of

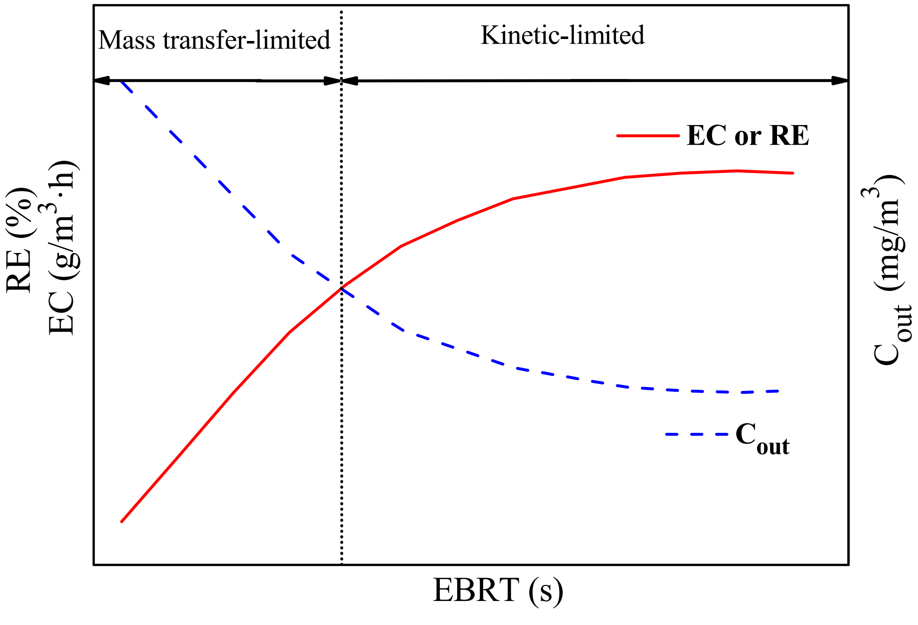

Fig. 4. The relationship between the EBRT and outlet concentration, EC and RE when the inlet concentration of hydrophobic VOC remains unchanged.

hydrophobic VOCs [61]. Yu et al. [57], for instance, investigated the effect of EBRT on the removal of *n*-hexane and found that when the EBRT was 30 s, the RE of the pollutant was 1.75 times higher than that when the EBRT was 7.5 s. This result was similar to the report of Cheng et al. [44], who observed that during the biofiltration of *n*-hexane, the reduction of the EBRT (60 s down to 15 s) reduced the probability of microbial capture of *n*-hexane, leading to a significant decline in the removal of *n*-hexane. For hydrophilic VOCs with low concentration, such as methyl isobutyl ketone, methanol, and acetone, etc. the reduction of the EBRT has little effect on their removal because they are almost not limited by the mass transfer resistance, and their removal mainly occurs in the packing beds near the inlet of the biofilters [44,90,92]. Therefore, in the actual application, the EBRT should be reasonably designed according to the properties of the pollutants to prevent the designed EBRT from increasing the floor area of the biofilters and increasing the construction cost; meanwhile, the EBRT should not be too small to obtain a satisfactory pollutant removal.

2.3.2. Nutritional conditions and operating environments

VOCs in waste gas streams can be utilized as a carbon source and energy by microorganisms to achieve the degradation and removal of VOCs [1,18]. But besides the carbon sources, other nutrients including nitrogen, phosphorus, and some trace elements are also necessary for microorganisms [11]. Therefore, the nutrient solution needs to be provided to microorganisms to maintain the removal performance of biofilters for VOCs. Once the nutrient supply is stopped, the microbes in biofilters will enter the endogenous respiration, which will reduce the activity and easily cause the biofilm to fall off, thus reducing the removal performance of biofilters for VOCs [44].

In addition, the operating environments, such as temperature and humidity, are also crucial conditions affecting the performance of biofilters [94]. Generally, a humid environment is conducive to the growth and reproduction of microorganisms, especially bacteria, which show lower tolerance to the dry environment than fungi [30,43]. Therefore, proper humidity should be executed during the operation of biofilters to keep the activities of microorganisms. During gas-phase biofiltration, the drying effect of high-speed gas flow, friction, and temperature rise caused by the degradation of organic package material, will take away a large amount of water, causing a decline of humidity in the biofilters, and would do harm to microorganisms. Then, it would result in the deterioration of the removal performance of biofilters for VOCs. Spraying nutrient solutions into biofilters is an effective measure to solve this problem. It cannot only provide nutrients and energy necessary for microorganisms but also help keep the biofilters at an appropriate humidity and maintain good conditions for microorganisms to remove VOCs. Thus, on the lab scale, waste gas streams are often passed into the humidifier before flowing into biofilters, while in actual industrial applications, waste gas is often pre-humidified by scrubbers before biofiltration [18,44,61]. On the one hand, this operation can improve the humidity of biofilters, and prevent the problems of channeling [61]. On the other hand, it can also remove water-soluble substances in the waste gas streams, and reduce the inlet loads for the subsequent biofilters. Besides the effect that VOC removal might be interesting in case of harsh limit values for clean gas, the aqueous phase of these scrubbers is commonly recirculated and only small volume flows are changed per day, week, or even month. Many research results showed that the humidity of biofilters should be kept at 40–60% [43,69,72,94]. Humidity higher than 60% may also be detrimental to VOC removal because the high humidity is easy to form anaerobic areas in biofilters and cause some operational problems [43].

The influence of temperature on VOC removal is mainly reflected in the influence on microbial activity and water solubility of VOCs [32,44,93]. Generally, 25–28 ℃ is the appropriate temperature for microbial growth and metabolism. In addition to mesophilic and thermophilic microorganisms, which can withstand temperatures ranging from 40 to 42 ℃ and above 42 ℃, respectively, most other microbes cannot survive in such high temperatures and would die out after domestication. Additionally, lower temperatures, such as those lower than 20 ℃, will reduce the enzyme activities of microorganisms and affect the removal of VOCs by biofiltration [32]. Cheng et al. [44] found that when the temperature was reduced from 25 ℃ to 10 ℃, the maximum RE of *n*-hexane by biofiltration decreased by 42%. When the temperature rises to 25 ℃ again, the RE of *n*-hexane increased remarkably compared with that at 10 ℃ [44]. Sun et al. [32] studied the removal of toluene by biofiltration and also observed that the low ambient temperature of 12 ℃ resulted in the average RE of toluene being only 50% under the condition of EBRT was 60 s. It is far lower than the research results of Cheng et al. [44], who showed that the RE for toluene approached 100% after the treatment of biofilters at 25 ℃ and the EBRT of 60 s. That is, as the lower operating temperature, it more easily leads to the reduction of VOC removal. Therefore, it is vital to maintain an appropriate ambient temperature during biofiltration. When biofiltration is adopted to remove VOCs in winter or high-latitude areas, corresponding strengthening measures (except for heating the biofilters) should be taken to enhance the VOC removal in low temperatures.

3. Prevailing problems in biofiltration

3.1. Uneven distributions of biomass, VOC loads, and nutrients

In biofiltration, VOCs pass through the packed beds, and gas is discharged after microbial actions [1,18]. Therefore, the VOC concentration gradually decreases with the height of the packed beds. Namely, the VOC load near the inlet is the highest, and near the outlet is the lowest [18,44]. In the study of Çelikten et al. [34], they reported that there is no homogenous distribution in the biofilters, and there will be short-cuts at the corners of the rectangular-shaped biofilter walls. Moreover, they also observed that the sampling position close to the inlet pipe can measure higher VOC concentrations [34]. Since VOCs are absorbed by microorganisms as a carbon source and energy to promote microbial metabolism, the packing medium near the inlet often has more microorganisms and thicker biofilm [44,48]. Moreover, the nutrient solution for microbial metabolism is often sprayed down from the top of biofilters. Therefore, in a counter-current operation mode, the concentration of biofilm near the outlet, i.e. nearly at the top of the package, is again higher as the nutrient supply is sufficient. While in a co-current operation mode, the packing medium near the outlet has a small amount of biofilm as the available nutrients for microorganisms gradually decrease from top to bottom. In addition, it should be noticed that the microorganism in the top packing may also be difficult to form a uniformly distributed biofilm because it is difficult to resist the scouring effect of water during irrigation. All these reasons lead to uneven distribution of biomass in biofilters.

The uneven distribution of biomass in different packing layers is a common problem in biofilters because different packing heights have different VOC loads. Cheng et al. [44] studied the simultaneous removal of hydrophilic and hydrophobic VOCs by biofiltration and observed that more microorganisms accumulated on the packing media near the entrance, and hydrophilic acetone could be 100% degraded after the microbial action near the entrance. The uneven distribution of biomass in biofilters is likely to cause some operational problems, such as blockage, short circuits, increasing pressure drop, and deterioration of VOC removal [48]. During the 180 days of operation, Cheng et al. [44] observed many times that the pressure drop of biofilters suddenly rose due to biofilm accumulation, which affected the removal performance of biofilters for VOCs.

Therefore, controlling biomass, VOC loads, and a nutrient uniform distribution is the key to realizing the long-term stable operation of biofilters. However, in the current research and applications, most of the reported solutions are those for the removal of excessive biomass after biofilter clogging occurs, such as chemical washing, mechanical washing, limiting nutrient solution (continuously or for time intervals), and biological predation [7,48,68,95,96]. These methods are easy to damage microbial activities or need taking a long time for biofilters to restore the removal performance for VOCs. While there are few pieces of research on how to prevent the excessive accumulation of biomass. Except for the quorum quenching by enzymes and the introduction of bacteria with group quenching function, such as *Rhodococcus* BH4, proposed by Wang et al. [7,97]. More effective novel strategies to prevent the excessive accumulation of biomass need to further investigate.

3.2. Unsuitable removal VOCs at high organic loading rates

Biofiltration has good removal performance for low VOC concentrations or low loads [98]. Especially for low-concentration hydrophilic VOCs, because there is no mass transfer restriction, microorganisms in biofilters can fully utilize hydrophilic VOCs as a carbon source and energy to achieve complete mineralization [44,90]. However, in actual production, many industries generate high organic load waste gas streams, such as surface spraying, oil refining, raw material manufacturing, etc. [5]. Unfortunately, biofiltration is not suitable for treating high-load organic waste gas. When the incoming gas contains high-concentration hydrophilic VOCs, although good treatment performance can be achieved in the early stage, sufficient carbon sources are likely to cause excessive accumulation of biofilm and even toxic effects on the biocatalyst [50,90,99]. It would lead to the collapse of biofilters in the later stages of operation and cause difficulties in the operation and management of biofilters. Therefore, there is a critical inlet load, also known as the safety threshold, for the removal of hydrophilic VOCs in biofiltration. When the inlet VOC load is lower than the safety threshold, the degradation rate of hydrophilic VOCs by biofiltration can be as high as 100% [44,90]. Once this threshold is exceeded, the removal of VOCs will be limited by the assimilation reaction of microorganisms, leading to a decline in VOC removal [50].

For example, it is reported that when the inlet load of methanol is higher than 300 g/m^3·h, it can have a toxic effect on the biocatalyst and thus have a negative effect on the removal of methanol [50]. Ramezani et al. [90] showed that when a load of imported methanol increased to 453 g/m^3·h, the RE of methanol decreased due to the inhibition of high-load methanol on the biocatalyst. More interestingly, after reducing the inlet methanol load, the methanol concentration at the outlet is even higher than that at the inlet [90]. This result showed that although there is no mass transfer limitation for hydrophilic VOCs, biofiltration is still not suitable for the treatment of high-load hydrophilic VOCs. Because high-load hydrophilic VOCs will not only have toxic effects on microorganisms but also high-concentration hydrophilic VOCs are easy to peel off from the aqueous phase and enter the gas phase again after being absorbed by the aqueous phase [50]. For high-load hydrophobic organics, due to their poor bioavailability, it is more difficult to obtain a satisfactory purification effect [57,58,100]. Taking the study of Vergara-Fernández et al. [101] as an example, they observed that the removal of *n*-pentane by biofiltration was poor with the maximum EC of only 110 g/m^3·h when the IL was higher than 500 g/m^3·h and even if the EBRT was up to 3.7 min, while with the IL lower than 100 g/m^3·h, the RE of *n*-pentane can achieve 90%-100%. Yu et al. [57] also reported a similar phenomenon, they observed that when the EBRT was 30 s, the RE of biofilters for low load *n*-hexane (15.67 g/m^3·h) could be as high as 92.6 ± 0.9%, while when the *n*-hexane load increased to 61.59 g/m^3·h, the RE of biofilters for *n*-hexane decreased to 63.8 ± 3.3%.

Therefore, no matter whether it is treating the hydrophilic or hydrophobic VOCs, the inlet pollutant load in biofilters should not be too high. That is, biofiltration has a good application prospect for the removal of low-load VOCs. When the VOC load in the waste gas streams is high, biofiltration should be coupled with other pre-treatment processes to reduce the VOC loads for biofilters, or effective strengthening measures should be selected to improve the bioavailability of hydrophobic VOCs to achieve efficient purification of high-load organic waste gas streams.

3.3. Inconsistent interactions in biofiltration for multicomponent VOCs

Usually, research on the removal of single VOC by biofiltration was very general. However, the waste gas generated in actual production is often a mixture of multiple VOCs [3,61]. Therefore, it is essential to verify the removal performance and interactions of the multicomponent VOCs in specific industrial waste gas. Based on this, in recent years, research on the removal of multicomponent VOCs by biofiltration is also increasing [42,53,102].

Many studies have investigated the removal of multicomponent VOCs by biofiltration and also discussed the interactions between multicomponent VOCs in biofilters [18,45,60,103]. Considering the removal performance of multicomponent VOCs in biofilters, Yang et al. [42] found that the interactions of multicomponent VOCs in biofiltration can be summarized as synergistic, antagonistic, and neutral effects. Usually, the removal of VOCs with stronger hydrophobicity is more likely to be inhibited by other components when removing mixed VOCs through biofiltration [42,104]. For example, the presence of toluene or chlorobenzene can produce an antagonistic effect on the removal of methane during simultaneous biofiltration of toluene or chlorobenzene and methane [105]. The report by Amin et al. indicated that the presence of aromatic compounds can inhibit the biofiltration removal performance of *n*-hexane [89,104]. Khabiri et al. [58] also reported the interactions between methane and styrene in the counter-current biofilter and noticed that styrene has an antagonistic effect on methane removal, and the higher the styrene load, the stronger the antagonistic effect on methane removal. Moreover, the degree of inhibition was different among media heights. The closer to the inlet, the stronger the inhibition effect of styrene on methane removal. The closer to the outlet, the inhibition effect of styrene on methane removal was weaker. Interestingly, the interaction of methane (7–60 g/m^3·h) on styrene (9–32 g/m^3·h) removal is neutral [58].

However, it is more interesting that the interactions between multicomponent VOCs are not invariable, and the interactions may vary with the change in reaction conditions [42]. Cheng et al. [44] observed the presence of *n*-hexane had a neutral effect on methyl isobutyl ketone removal. But when the load ratio of *n*-hexane to methyl isobutyl ketone was less than 6:1, methyl isobutyl ketone produced an antagonistic effect on *n*-hexane removal due to the existence of higher concentrations of hydrophilic VOCs easy to form a competitive inhibition with hydrophobic VOCs. However, after reducing the dosage ratio of methyl isobutyl ketone, the antagonistic effect on *n*-hexane removal decreased, and finally, methyl isobutyl ketone presented a synergistic effect on *n*-hexane removal [44]. Similar phenomena have also been reported in the study of removing other two-component VOCs (toluene and *n*-hexane, methanol and *n*-hexane) by biofiltration [106]. In other words, in the biofiltration process of mixed VOCs with different hydrophobicity, the interactions between multicomponent VOCs are complicated and can be affected by the ratio of mixed VOCs [91]. When the proportion of hydrophilic VOCs is high, the hydrophilic VOCs are easily absorbed by microorganisms and have an antagonistic effect on hydrophobic VOC removal. When the proportion of hydrophilic VOCs is relatively low, the existence of hydrophilic VOCs can moderately stimulate the growth of microorganisms, improve the demand of microbes for carbon sources and energy, and achieve the collaborative removal of hydrophilic and hydrophobic VOCs.

3.4. Unsatisfactory removal performance for hydrophobic VOCs

The low bioavailability of hydrophobic VOCs results in high mass transfer resistance in gas-phase biofiltration [42,43,50]. Therefore, hydrophobic VOCs are usually difficult to obtain a satisfactory removal performance in traditional biofilters, which affects the practical application potential of biofilters.

In the study of simultaneous removal of hydrophilic and hydrophobic VOCs by biofilters, Cheng et al. [44] observed that the RE of *n*-

hexane was only about 50%, and the RE of hydrophilic VOCs reached 100%. Moreover, the influence of EBRT on the removal of hydrophilic VOCs was significantly lower than that on hydrophobic VOC removal under the conditions of the inlet loads of hydrophilic VOCs and hydrophobic VOCs was similar. In other words, biofilters could still maintain a RE of nearly 100% for the hydrophilic VOCs with low load and low EBRT (15 s) [44]. For hydrophobic VOCs, relatively longer EBRT was often needed to obtain better removal performance, and sometimes even maintaining longer EBRT still cannot achieve a satisfactory purification effect. The research of Khabiri et al. [58] showed that the RE of methane with an inlet load of 7–60 $g/m^3 \cdot h$ by biofilter could not exceed 80% even if the EBRT was up to 360 s. Additionally, when simultaneously removing methane and styrene, Khabiri et al. [58] observed that the RE of biofilters for styrene with an inlet load of 9–22 $g/m^3 \cdot h$ could also reach 100%, and most of the styrene could be degraded and removed after the microbial metabolism in the bottom packing media near the inlet. It is due to the solubility of styrene in the biofilm phase being higher than that of hydrophobic methane since the Henry's Law constant of styrene is 0.113 and that of methane is 28 [50]. Therefore, under the same or similar operating conditions, the removal performance of biofilters for hydrophobic VOCs is far lower than that for hydrophilic VOCs.

Therefore, it is an enormous challenge for biofiltration to improve the degradation performance of hydrophobic VOCs, and it has become the focus of many researchers at home and abroad. A variety of strengthening measures aiming to promote the bioavailability of hydrophobic VOCs came into being, such as co-metabolism with hydrophilic VOCs, adding surfactants and other exogenous additives, coupling pretreatment process, etc.

4. Strategies to solve prevailing problems and improve removal performance of biofilters for VOCs

Many factors can affect the VOC removal performance of biofilters, and the general problems summarized above often occur during the operation of biofilters. Solving these common problems is the key to maintaining the stable operation of biofilters and improving the application of biofiltration in VOC treatment. Therefore, this chapter mainly focuses on these common problems and summarizes the current effective measures to solve these problems, aiming to provide a reference for the research and applications of biofiltration to remove VOCs (Table 2, Fig. 5).

4.1. *Strategies for mitigating VOC loads and uneven biomass distribution*

The problems of uneven distribution of biomass, VOC loads, and nutrients generally exist in biofilters. Those problems are particularly serious in biofilters filled with organic packing media, which will result in the decline of the removal performance of biofilters for VOCs and the increase of the pressure drop during the operation periods. Improving the structure of biofilters and innovating the operation mode of biofilters are effective ways to alleviate those problems [107–109].

4.1.1. *Step-feed and step-spray*

Some studies have shown that the removal of VOCs in biofilters is mainly concentrated in the first 30% of the bed height, which means that biomass accumulation mainly occurs in the first 30% of the bed height near the inlet, while the remaining nearly 70% of the bed height plays a weak role but requires a lot of energy consumption [107,110]. This phenomenon further shows that the biomass and VOC loads are extremely unevenly distributed in traditional biofilters. To make the distribution of biofilm and VOC loads in biofilters more uniform and reduce the energy demand. Estrada et al. [107] designed a step-feed biofilter for VOC removal, which divided the biofilter into three independent modules for the gas inlet. Compared with traditional biofilters, with a similar EC of toluene (EC: 80 $g/m^3 \cdot h$), the step-feed biofilter showed a lower pressure drop. After 60 days of operation, the pressure drop of the step-feed biofilter was only 300 Pa/m, while that of the traditional biofilter rose to 4000 Pa/m on day 30 [107]. Therefore, step-feed of the inlet gas is beneficial for maintaining a lower pressure drop and better VOC removal performance of the biofilters. However, in large-scale applications, the adoption of the step-feed mode should also

Table 2
Strategies and applicable conditions for improving the removal performance of VOCs by biofiltration.

Strategies	Applicable situations	Advantages	References
Step-feed of the inlet gas	a: Uneven distribution of biomass, VOC loads; b: High VOC inlet loads	Low pressure drop; The distribution of biofilm and VOC loads is more uniform	[107,110]
Step-spray the nutrient solution	Uneven distribution of biomass, and nutrients	Low pressure drop; More uniform distribution of biofilm and VOC loads; Lower mass transfer restrictions	[108,109]
Innovative bioreactors	a: Uneven distribution of biomass, VOC loads, nutrients; b: The removal of hydrophobic VOCs	More uniform distribution of biofilm, VOC loads, and nutrients; Relieve biomass accumulation; Better hydrophobic VOCs removal performance	[43,92,111,112,114–116]
Two-phase bioreactors	High VOC inlet loads	Relieve the toxic effects of high concentration hydrophilic VOCs on microorganisms; Improve the mass transfer of hydrophobic VOCs; Improve the bioavailability of hydrophobic VOCs	[42,78,90,117,118]
Coupling pretreatment processes	a: VOCs with high inlet concentration; b: The removal of hydrophobic VOCs	Improve the bioavailability of hydrophobic VOCs; Reduce the inlet loads of biofiltration; Avoid excessive accumulation of biomass	[9,14,119–123]
Co-metabolism with hydrophilic VOCs	The removal of hydrophobic VOCs	Shorter startup time; Faster biofilm formation; Can improve the removal performance of hydrophobic VOCs	[44,45,53,106,124,125]
Addition of surfactants		Reduce the surface tension of solutions; Reduce the apparent Henry's Law coefficient of hydrophobic VOCs; Reduce the mass transfer resistance of hydrophobic VOCs	[1,18,43,46,71,126–128]
Introduction of bacterial quorum sensing molecules		Mediate the production of soluble microbial products and the secretion of extracellular enzymes; Enhance the adhesion and EPS content of biofilm, and promote the formation of biofilm; Increase the solubility of hydrophobic VOCs in liquid; Improve the removal performance of VOCs by biofiltration in adverse operating conditions	[32,129–132]

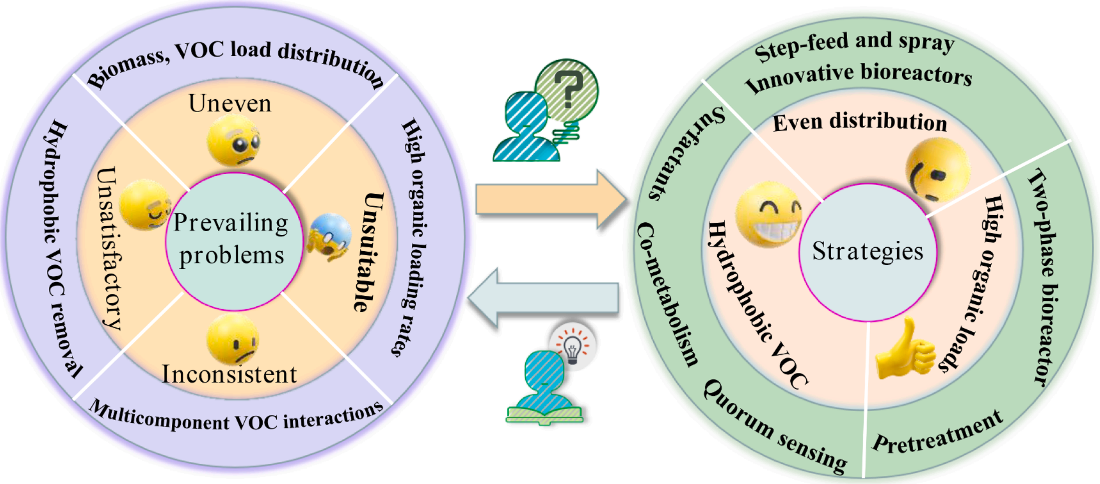

Fig. 5. Prevailing problems and corresponding strategies in VOC removal by biofilters.

consider how to reduce investment and operation costs. Because the use of step-feed of the inlet gas may lead to an increase in the volume and space requirements of the biofilters.

Except for the step-feed of the inlet gas, step-spray the nutrient solution is also an effective strategy to promote the uniform distribution of biofilm and reduce the pressure drop of biofilters. Chaiprapat et al. [108] used the method of step-spray to provide an oxygen-rich nutrient solution to biofilters, and they found that the biofilter fed with step-spray had a higher purification efficiency of biogas, the EC of hydrogen sulfide increased by nearly 10% compared with direct feeding from the top of biofilters. It was consistent with the research results of Rodriguez et al. [109], who reported that only using a single inlet to transport oxygen and the nutrient solution was always inefficient for fully realizing the biological oxidation of hydrogen sulfide, especially when the inlet hydrogen sulfide load was high and the EBRT was short. The distributed feed recirculation flow exposes the top and middle bed layers to less nutrient liquid, which can avoid the entire bed layer from encountering excessive liquid, thus reducing the impact of pollutants in mass transfer. Moreover, it can also reduce the blockage of the packing media, prevent the compaction of the packing beds, and reduce the mass transfer restrictions.

Both step-feed and step-spray biofiltration can promote the uniform distribution of biofilm and microbial nutrients in biofilters, reduce the pressure drop during the operation of biofilters, and save energy. More importantly, they can also improve the EC of biofilters for gaseous pollutants. Thus, a better removal effect may be obtained if the biofilters with step-feed inlet gas and step-spray nutrients are used for VOC removal simultaneously. Unfortunately, there is no related report on gas-phase biofiltration with this operation mode.

4.1.2. *Innovative bioreactors*

In addition to the innovative operation mode, improving the bioreactors is another feasible means to promote the uniform distribution of biomass, VOC loads, and nutrients in bioreactors, such as tubular biofilters and rotary drum biofilters, etc. [43,92,111,112]. When the multi-tube biofilter is running, the waste gas containing VOCs flows through

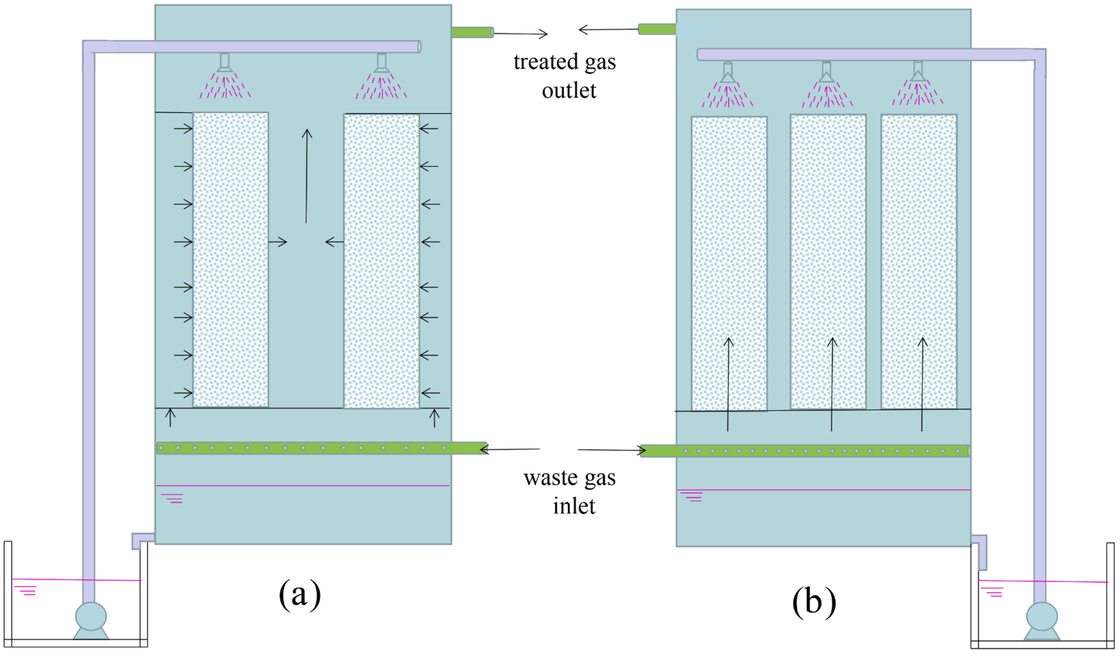

Fig. 6. Structures of innovative biofiltration reactors. (a) Tubular biofilter; (b) Multi-tube biofilter.

the inlet and enters the bioreactor, then it can be divided into several small gas streams and enters the corresponding independent biofilters filled with packing media (Fig. 6b). After being absorbed and degraded by the microorganisms, the small gas streams are combined and discharged through the exhaust port. Similar to the mode of step-feed inlet gas, the unique structure of the multi-tube biofilter can make the inlet VOC loads more evenly distributed in each independent biofilter.

Chen et al [92,111]. adopted tubular biofilters for the investigation of VOC removal, the exhaust gas flowed through the packing media from all directions of the cylindrical sponge packing in tubular biofilters, greatly increasing the cross-sectional area for the inlet gas (Fig. 6a), effectively improving the distribution of the inlet VOC loads. Therefore, taking the methyl isobutyl ketone as the substrate with the inlet load of 20 $g/m^3 \cdot h$, the average RE of methyl isobutyl ketone reached 98.7% even when the EBRT was very small (5–10 s), and no excessive biofilm accumulation occurred during the periods of methyl isobutyl ketone removal capacity less than 80 $g/m^3 \cdot h$ [111]. In addition, the tubular biofilter also has good removal performance for moderately hydrophobic VOCs. When the EBRT was 15 s and the inlet load of toluene was 18.7 $g/m^3 \cdot h$, the RE of toluene could be as high as 99%. The whole research stage has been running for 391 days without significant biomass accumulation [92]. Besides the tubular biofilter, Yang et al. [112] also developed a multi-layer rotary drum biofilter in view of the uneven VOC load and biomass distribution in biofilters. In the removal process of ether, no significant biomass accumulation was observed when the inlet load of ether was lower than 128 $g/m^3 \cdot h$, and the RE of ether reached 99% [112]. However, how to overcome the shortcomings of rotary drum biofilters, like blocking of bearings by wooden parts or fibers, high mechanical abrasion, loss of package material, and high loads of small particles blocking effluents, is also a direction that still needs to be studied in the future.

In addition, other new biofilters, such as air diffusion bioreactors, rotary rope biofilters, and improved rotary biological contactors, also can present better VOC removal performance than traditional biofilters [113–116]. Because these new types of biofilters also can promote the uniform distribution of biofilm, liquid nutrient solution, and inlet VOC loads. Therefore, the innovative bioreactor is a crucial way to solve the problem of uneven VOC loads and biomass distribution in traditional bioreactors, and also a momentous basis for the practical application of biofiltration. Of course, these innovative bioreactors also need to comprehensively consider the factors of energy consumption, operation costs, operability, stability, and maintenance in large-scale applications.

4.2. Strategies for promoting removal of VOCs with high concentrations

Biofiltration is a competitive technology for the removal of low-concentration VOCs. However, for high-concentration organic waste gas, single biofiltration often has the problem of low removal efficiency or excessive biomass accumulation. Therefore, efficient biological treatment of high-concentration organic waste gas is one of the significant challenges in biofiltration.

4.2.1. Two-phase bioreactors

For high-concentration hydrophilic VOCs, it is necessary to avoid excessive accumulation of biomass in biotrickling filters and the toxic effects on microorganisms. Therefore, the key to solving these problems is to avert direct contact between highly concentrated hydrophilic VOCs and biofilm in biotrickling filters. Based on this, the two-phase bioreactor is a favorable strategy to remove a high concentration of hydrophilic VOCs. By introducing a non-soluble organic phase into the liquid phase in biotrickling filters, when the waste gas streams containing high-concentration hydrophilic VOCs enter the biotrickling filters, VOCs can be absorbed by the non-soluble organic phase, and then gradually transported to the biofilm phase for microbial absorption and degradation. The slow-release effect of the non-soluble organic phase for high-concentration VOCs can reduce the toxicity of high-concentration VOCs to microorganisms, and avoid excessive accumulation of biomass due to excessive absorption of hydrophilic VOCs by microorganisms. Ramezani et al. [90] found that the maximum EC of methanol by biotrickling filtration at high concentrations could reach 416 $g/m^3 \cdot h$ in the presence of silicone oil. Compared with single aqueous phase biotrickling filters, although the silicone oil might have a slight negative impact on the removal of high-concentration methanol, the silicone oil could effectively reduce the toxicity effect of methanol on biofilm, and provide a more suitable living environment for microorganisms.

On the contrary, the two-phase bioreactors can play a positive role in the removal of hydrophobic VOCs by biotrickling filtration [78,117,118]. Because hydrophobic VOCs are mainly limited by mass transfer in biotrickling filtration, and the presence of a non-soluble organic phase is conducive to promoting the mass transfer of hydrophobic VOCs and improving the bioavailability of hydrophobic VOCs [42,117]. San-Valero et al. [78] also used silicone oil as the non-soluble organic phase and adopted two-phase biotrickling filters to remove styrene. Compared with traditional biotrickling filtration, they observed that the two-phase biotrickling filters with low silicone oil dosage (25 mL/L) had a faster-starting speed and better styrene removal performance, and could maintain stable styrene removal performance when the inlet waste gas was in an unstable state [78]. Ramezani et al. [90] also strongly suggested that two-phase biotrickling filters can be used to mineralize both high-concentrated hydrophilic and hydrophobic VOCs simultaneously. Because when hydrophilic VOCs and hydrophobic VOCs exist together, the slight inhibition of two-phase biotrickling filters on a high concentration of hydrophilic VOCs can be almost ignored. Therefore, the usage of the two-phase biotrickling filters is effective to remove high concentrations of hydrophilic VOCs and hydrophobic VOCs and their mixtures. In order to improve the practical application potential of two-phase biotrickling filters, it is also necessary to pay attention to the problems resulting from the existence of a non-soluble organic phase and solve these problems, such as deposition, creeping effects, stable emulsions, etc.

4.2.2. Coupling pretreatment processes

Since biofiltration has good removal performance for low-concentration VOCs, efficient measures should be taken to reduce the high concentration of organic waste gas to a suitable degree before it is introduced into biofilters. Combined pretreatment processes (such as ozone oxidation, low-temperature plasma, two-stage biofiltration, magnetic fields, UV oxidation, etc.) are valid means to achieve this goal [6,9,14,119–122].

To prevent excessive biomass accumulation during the long-term operation of biofilters for high-load VOCs. Zhou et al. [122] used ozone oxidation and biofiltration to jointly treat toluene in exhaust gas and they found that the performance of biofilters decreased due to the excessive accumulation of biofilm when the traditional biofilters ran for 150 days. The biofilters jointly treated with ozone could effectively keep the biofilters running stably and maintain high toluene removal performance under the condition of low ozone concentration (5–10 mg/m^3). Among them, when the EBRT was 60 s, the RE and EC of biofilters for toluene can be maintained at 91.1–97.7% and 45.7–72.3 $g/m^3 \cdot h$, respectively [122]. Moreover, the uneven distribution of biomass along the bed height was significantly improved because ozone has a bactericidal effect. Combined ozonation can not only effectively reduce the VOC load for biofilters, but also effectively prevent the excessive accumulation of biofilm in biofilters. At the same time, ozone oxidation can also decompose hydrophobic VOCs, improve the bioavailability of hydrophobic VOCs, and thus enhance hydrophobic VOC removal in biofiltration [119]. In addition, Dobslaw et al. [123] designed a combined treatment process consisting of non-thermal plasma, zeolite adsorption, and biofiltration to treat VOCs containing waste gas streams generated during sludge centrifugation in the sewage treatment plant. They found that the RE of VOCs reached 60.3–93.9% and the bioavailability of VOCs was improved after the actual waste gas streams were pretreated by non-thermal plasma and zeolite adsorption. As a result, after combined treatment with biofiltration, the total RE of the mixed VOCs was as high as 95% [123].

The waste gas containing VOCs generated in the actual productions is often a mixture containing a variety of VOCs. The properties and concentrations of VOCs generated by different industries are various. Therefore, it is often difficult to achieve the ideal treatment effect by using a single treatment facility. An appropriate combined other treatment process is a good choice. It can couple the advantages of each technology to gradually reduce the concentration of VOCs and achieve the effective removal of high-concentration organic waste gas finally.

4.3. Strategies to improve hydrophobic VOC removal

The removal of hydrophobic VOCs becomes an enormous challenge faced by biofiltration and a principal factor limiting the industrial application of biofiltration. Therefore, it is vital to improve hydrophobic VOC removal by biofilters, and it is also a hotspot and focuses in the research field of biofiltration. Many scholars have also proposed several strategies to enhance biofiltration to remove hydrophobic VOCs. In addition to the two-phase biofilters and coupling pretreatment processes mentioned in 4.2, other methods such as co-metabolism with hydrophilic VOCs, surfactant enhancement, and the addition of exogenous substances to utilize quorum sensing are also alternative measures to promote the removal of hydrophobic VOCs by biofiltration [32,53,97,102].

4.3.1. Co-metabolism with hydrophilic VOCs

Hydrophilic VOCs have a nice mass transfer effect in biofiltration and are easily used by microorganisms as carbon sources and energy materials. Therefore, hydrophilic VOCs are beneficial to the growth of microorganisms. When hydrophilic VOC and hydrophobic VOC coexist in the waste gas streams, the presence of hydrophilic VOCs can stimulate the growth of microorganisms and increase the demand for microorganisms for carbon sources. Thereby, the existence of hydrophilic VOCs can improve the removal performance of biofilters for hydrophobic VOCs [44,45,53]. Finally, it could realize the collaborative removal of hydrophilic and hydrophobic VOCs.

Although some studies have shown that the introduction of hydrophilic VOCs can improve the removal performance of biofilters for hydrophobic VOCs, the dosage of the introduced hydrophilic VOCs must be well controlled. The introduction of excessive hydrophilic VOCs would compete with hydrophobic VOCs, which is unfavorable to hydrophobic VOC removal [106]. Cheng et al. [44] found that when the mixing ratio of hydrophobic *n*-hexane and hydrophilic methyl isobutyl ketone was 1:1 or 2:1, the removal of *n*-hexane was inhibited due to the excessive methyl isobutyl ketone competed with *n*-hexane and the microorganisms are more likely to degrade hydrophilic VOCs than hydrophobic *n*-hexane. With the mixing ratio of *n*-hexane and methyl isobutyl ketone increased to 6:1, the existence of methyl isobutyl ketone improved the bioavailability of *n*-hexane and promoted the removal of *n*-hexane [44]. A similar phenomenon also occurred during the simultaneous biofiltration removal of toluene and *n*-hexane. When introducing toluene with a high inlet load, the biofiltration performance of *n*-hexane reduced to less than 35% due to the inhibition of a high toluene inlet load, while with the mixing ratio of *n*-hexane and toluene decreased to 6:1, the antagonistic effect of toluene on *n*-hexane removal disappeared and instead by the synergistic effect, where the RE of *n*-hexane reached 90% under the condition of inlet *n*-hexane load was 29 $g/m^3 \cdot h$ [44]. In addition, many other previous studies have also reported that the existence of high-concentration hydrophilic VOCs is not conducive to the removal of hydrophobic VOCs [124,125]. Because the high-concentration hydrophilic VOCs not only would compete with hydrophobic VOCs but also easily lead to excessive accumulation of biomass.

Therefore, when hydrophilic VOCs are introduced to promote the removal of hydrophobic VOCs, loads of hydrophilic VOCs should not be too high. Controlling the amounts of hydrophilic VOCs is the key to realizing the co-metabolism of hydrophilic and hydrophobic VOCs.

4.3.2. Addition of surfactants

Surfactants are amphoteric substances with hydrophilic and hydrophobic groups in their molecular structures, which can significantly reduce the surface tension of solutions [1,126–128]. For the hydrophobic VOC removal by biofiltration, adding surfactants in nutrient solution can effectually reduce the apparent Henry's Law coefficient (H*) of hydrophobic VOCs and improve the solubility of hydrophobic VOCs in the liquid phase [1,71]. Yang et al. [128] compared the effects of different types of surfactants (sodium dodecyl sulfate, cetyltrimethylammonium bromide, Tween 80, and Triton 100) on the H* of *n*-hexane. They found that sodium dodecyl sulfate can reduce the H* of *n*-hexane to the lowest when the four surfactants were at the same critical micelle concentration, and Triton 100 can minimize the H* of *n*-hexane when the four surfactants were at the same mass concentration [128]. It is because biosurfactants (such as Tween 80, rhamnose, saponin, Triton 100, etc.) usually have lower surface critical micelle concentrations than chemical surfactants (sodium dodecyl sulfate, and cetyltrimethylammonium bromide, etc.). However, both chemical and biological surfactants can reduce the H* of hydrophobic VOCs. It means that when removing hydrophobic VOCs by biofiltration, adding surfactants can effectively reduce the mass transfer resistance of hydrophobic VOCs between the gas phase and liquid phase, which is beneficial to improve the bioavailability of hydrophobic VOCs, and thus improve the removal performance of biofilters for hydrophobic VOCs [43,71].

In a recent study, sodium dodecylbenzene sulfonate (SDBS) was introduced to the nutrient solution to feed a biofilter for the removal of different short-chain *n*-alkanes. The addition of a low-concentration SDBS significantly improved the removal performance of the biofilter for different short-chain *n*-alkanes in the investigation of Wu et al. [1]. Under the conditions of inlet load was 58 ± 3.0 $g/m^3 \cdot h$ and EBRT was 30 s, the RE of methane increased from 35 ± 5.6% to 74 ± 3.3% by adding 15 mg/L SDBS, while that of *n*-hexane increased from 77 ± 1.3% to 99 ± 0.7% [1]. Meanwhile, they found that adding SDBS not only effectually reduced the H* of *n*-alkanes, and improved the cell surface hydrophobicity of microorganisms, but also promoted the evolution of microbial community in favor of the short-chain *n*-alkane removal [18]. Qian et al. [115] found that introducing saponin (40 mg/L) promoted the EC of ethylbenzene from 95.6 to 131.9 $g/m^3 \cdot h$ when the inlet concentration of ethylbenzene was 1300 mg/m^3 and the EBRT was 30 s. Dewidar and Sorer reported that adding surfactin (50 mg/L) enhanced the bioavailability of 2-ethyl-hexanol, making the RE of 2-ethyl-hexanol reach as high as 99% at the inlet load of 32 $g/m^3 \cdot h$ [46].

The addition of surfactants has great potential in enhancing the hydrophobic VOC removal performance of biofilters. It is crucial to select the appropriate surfactants to promote VOC removal and control the cost. Because different kinds of surfactants have disparate effects on the H* of hydrophobic VOCs, and different types of surfactants have diverse effects on biodegradability and microbial growth characteristics. Therefore, when surfactants are introduced to biofilters to remove hydrophobic VOCs, the selection of appropriate surfactant type and dosage should be based on the testing results of the effects on apparent Henry's Law coefficient of hydrophobic VOCs and the growth of microorganisms.

4.3.3. Introduction of bacterial quorum sensing molecules

Except for adding surfactants and co-metabolism with hydrophilic VOCs, the introduction of bacterial quorum sensing (QS) molecules is another innovative way to enhance the removal performance of biofilters for hydrophobic VOCs in recent years. Bacteria produce QS function through the release of chemical signal molecules. The concentration of these molecules will increase with the increase in cell density or activity [32]. Subsequently, the expression of specific genes is induced to produce specific functions. According to the reports, it can mediate the production of soluble microbial products and the secretion of extracellular enzymes, enhance the adhesion and EPS content of biofilm, and promote the formation of biofilm [129,130]. In addition, the introduction of bacterial quorum-sensing molecules can also increase the solubility of hydrophobic VOCs in liquid. Therefore, under adverse operating conditions, such as short EBRT, high hydrophobic VOC loads, low temperature, and shock loads, the introduction of bacterial quorum sensing molecules has significant advantages in improving the removal performance of VOCs by biofiltration.

Usually used bacterial quorum sensing molecules mainly include self-inducible peptide, N-acylhomoserine lactone, N-(3-oxododecanoyl)-L-homoserine lactone (OHL), N-acyl-homoserine lactones (AHLs) and quinolone [32,131]. To solve the problem that short EBRT reduced the removal performance of biofilters for high-concentration toluene, Chen et al. [132] introduced a low concentration (20 uM) quorum sensing molecule OHL into biotrickling filters, and they found that the RE of toluene reached as high as 99.2% under the condition that the inlet concentration of toluene was 2210–3830 mg/m^3 and the EBRT was 30 s. Additionally, they also observed that when the operating temperature was higher than 40 ℃, the removal performance of biotrickling filters for toluene without adding OHL decreased significantly, while the biotrckling filter added OHL could increase the cell density and activate the toluene degrading enzyme by increasing the spray frequency, which improved the removal performance of biotrickling filters for toluene under high temperatures [132]. In addition, adding quorum sensing molecules can also promote the removal performance of biofilters for VOCs under low temperatures by optimizing the potential of biofilm. Considering that the high latitude area and the low environment temperature in winter are likely to lead to poor VOC removal performance of biofilters, Sun et al. [32] introduced exogenous AHLs into the biofilter and inoculated bacteria which can produce AHLs. The removal of toluene by biofilters at an average temperature of 12 ℃ was carried out. Results showed that compared with the removal performance of the biofilter without quorum sensing molecules, the EC of the biofilter with exogenous AHLs and the biofilter inoculated with AHLs-producing bacteria increased by 39% and 26% respectively in the first 24 days of operation, while the average mineralization rate of toluene increased by 25% and 47% respectively [32]. That is, both the addition of exogenous AHLs and the inoculation of bacteria producing AHLs can improve the toluene removal by biofiltration under low-temperature conditions. Moreover, it can be found that the biomass distribution in the biofilter inoculated with bacteria producing AHLs was more uniform. It promoted the continuous production of extracellular proteins under low-temperature conditions and promoted the adsorption and absorption of hydrophobic VOCs. Thus, the removal performance of biofilters for hydrophobic VOCs improved.

In a word, QS has good application potential in biofiltration, and the introduction of quorum sensing molecules is an effective measure to promote the stable and efficient operation of biofilters. However, the current investigations on QS in biofiltration mainly focus on the improvement of the removal performance for hydrophobic VOCs under adverse reaction conditions. While the migration and transformation of QS molecules in biofiltration and the molecular mechanisms of improving the mass transfer effect of hydrophobic VOCs still need further investigation.

5. Conclusions and prospects

(1) Biofiltration is an alternative green, economic, safe, and effective process for treating VOCs from waste gas streams. The VOC removal in biofilters is influenced by many factors, mainly the natural properties of VOCs, biofiltration system, and operating conditions. A proper biofiltration system should be built according to the properties of VOCs in waste gas streams and operated under optimized conditions to achieve efficient purification of VOCs by biofiltration.

(2) Uneven distribution of biomass, VOC loads, nutrients, and excessive biomass accumulation are the most common problems in biofiltration. The operation modes of step-feed and step-spray can effectively alleviate those problems to a certain extent. In addition, innovative bioreactors, such as multi-tube biofilters, are also effective measures to promote the uniform distribution of biofilm, VOC loads, and nutrients. For the problem of excessive biomass accumulation, quorum quenching may be a better choice than chemical washing and mechanical extrusion.

(3) Biofiltration has a good purification capacity for low-concentration VOCs, but it is not suitable for the treatment of high-concentration VOCs. To improve the removal performance of biofilters for VOCs with high concentrations or high loads, two-phase biofilters can be employed. On the one hand, it can reduce the toxic effect of high concentrations of hydrophilic VOCs on microorganisms, and on the other hand, it can effectively prevent the excessive accumulation of biofilm. For high-concentration hydrophobic VOCs, other pretreatment processes, such as ozone oxidation and low-temperature plasma, can be combined to improve the bioavailability of hydrophobic VOCs and reducing gas–liquid mass transfer resistance.

(4) Hydrophobic VOCs often have poor removal performance in biofiltration due to the restriction of mass transfer resistance. How to improve the removal performance of hydrophobic VOCs is an essential challenge faced by biofiltration at present, and needs to be solved urgently to realize the wide industrial application of biofiltration. Although co-metabolism with hydrophilic VOCs is a feasible scheme, it is usually necessary to strictly control the proportion of hydrophilic VOCs in the exhaust gas to obtain a better synergistic effect. The actual exhaust gas is a mixture with multiple VOCs, but the composition and concentration of VOCs in the exhaust gas often fluctuate significantly. It seems impractical to accurately control the composition and concentration of various VOCs in practical applications. Adding surfactants and introducing bacterial quorum sensing molecules may be alternative schemes to enhance the removal performance of hydrophobic VOCs in biofiltration. Because surfactant can reduce the mass transfer resistance of hydrophobic VOCs in biofiltration, and the introduction of quorum sensing molecules can improve the solubility of hydrophobic VOCs in liquid, and even improve the stability of biofiltration under adverse operating conditions such as impact loads and low temperatures. Moreover, the hydrophobic VOC removal could be promoted by adding a low concentration of surfactant or bacterial quorum-sensing molecules to biofilters. Therefore, adding surfactants or introducing bacterial quorum sensing molecules may be a better-enhanced measure with economic and environmental effects to improve the removal performance of hydrophobic VOCs by biofiltration. However, the migration and transformation of surfactants or QS molecules in biofiltration and the molecular mechanisms of improving the mass transfer effect of hydrophobic VOCs should be investigated in depth.

CRediT authorship contribution statement

Xin Wu: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. **Yan Lin:** Conceptualization, Resources, Validation, Writing – original draft, Writing – review & editing. **Yongyi Wang:** Resources, Writing – original draft, Writing – review & editing. **Shaohua Wu:** Writing – original draft, Writing – review & editing. **Chunping Yang:** Conceptualization, Supervision, Project administration, Funding acquisition, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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