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MINI-REVIEW

Biotechnology progress for removal of indoor gaseous formaldehyde

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Abstract

Formaldehyde is a ubiquitous carcinogenic indoor pollutant. The treatment of formaldehyde has attracted increasing social attention. Over the past few decades, an increasing number of publications have reported approaches for removing indoor formaldehyde. These potential strategies include physical adsorption, chemical catalysis, and biodegradation. Although physical adsorption is widely used, it does not really remove pollution. Chemical catalysis is very efficient but adds the risk of introducing secondary pollutants. Biological removal strategies have attracted more research attention than the first two methods, because it is more efficient, clean, and economical. Plants and bacteria are the common organisms used in formaldehyde removal. However, both have limitations and shortcomings when used alone. This review discusses the mechanisms, applications, and improvements of existing biological methods for the removal of indoor gaseous formaldehyde. A combination strategy relying on plants, bacteria, and physical adsorbents exhibits best ability to remove formaldehyde efficiently, economically, and safely. When this combination system is integrated with a heating, ventilation, air conditioning, and cooling (HVAC) system, a practical combined system can be established in formaldehyde removal. Multivariate interactions of biological and non-biological factors are needed for the future development of indoor formaldehyde removal.

Key Points

- *Indoor gaseous formaldehyde removal is necessary especially for new residence.*
- *Biological removal strategies have attracted increasing research attentions.*
- *Combined system of plants, bacteria, and physical adsorbents exhibits best efficiency.*
- *Integrated device of biological and non-biological factors will be potential practical.*

Keywords Formaldehyde · Indoor air · Biodegradation · Biological removal

Introduction

Formaldehyde is a colorless gaseous, flammable, and highly reactive volatile organic compound (VOC) at room temperature. It is often dissolved in water to obtain a 30–50% (by weight) aqueous solution, known as formalin. Formaldehyde is a widespread indoor pollutant and it is listed as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC) (Baan et al. 2009; IARC Working Group on the Evaluation of Carcinogenic Risk to Humans 2012). Formaldehyde is highly reactive and can denature nucleic acids and proteins (Craft et al. 1987; Squire and Cameron 1984). Acute exposure to indoor formaldehyde may result in eye redness, eye irritation, frequent blinking, and irritation in the upper respiratory system. In addition, exposure to formaldehyde can lead to long-term effects such as cancer, childhood leukemia, premature birth, low birth weight, genotoxicity, congenital anomalies, and Alzheimer's disease (2010). The data from the human exposure characterization of chemical substances (HEXPOC) report (Watson 2005) shows that the ratio of indoor to outdoor concentrations of formaldehyde is always greater than one. Therefore, formaldehyde is viewed as a very specific indoor pollutant. The main source of

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formaldehyde in indoor environments is building materials and consumer products (Chen et al. 2018; Jiang et al. 2017; Kelly et al. 1999; Plaisance et al. 2017). These products and materials can release formaldehyde for several months or more because of the large amount of formaldehyde-rich binder, urea-formaldehyde (UF) resin. The continuous hydrolysis of UF resin can release formaldehyde for prolonged times. Some indoor pollutants can react with each other, such as ozone and decene (Nazaroff and Weschler 2004; Uhde and Salthammer 2007). The indoor concentration may be high enough to cause adverse health effects and is the main cause of personal exposure because people spend more time indoors than outdoors. Therefore, the World Health Organization (WHO) recommends a short-term (30 min) guideline with formaldehyde up to 0.1 mg/m^3 to prevent sensory irritation in the general population. At the same time, this guideline also prevents long-term health effects, including cancer (2010).

At present, formaldehyde emission reduction technologies mainly include adsorption (Bellat et al. 2015; de Falco et al. 2018; Lee et al. 2013; Nomura and Jones 2013; Suresh and Bandosz 2018; Wang et al. 2018b), oxidation catalysis (Liu et al. 2019b; Rong et al. 2018; Song et al. 2018; Wang et al. 2018a), and biological removal (García-Pérez et al. 2013; Wolverton and Wolverton 1993; Kim et al. 2008; Wang et al. 2014). The detail comparison of advantages and disadvantages for these methods are shown in Table 1. Physical adsorption using porous materials, such as activated carbon, can provide excellent performance. However, such adsorbents must be replaced or periodically regenerated, to prevent re-emission of the adsorbed compounds (Luengas et al. 2015). Moreover, if these used adsorbents are not subjected to subsequent treatment, they risk transferring pollutants to other sites, the pollution is not addressed. Noble metal-containing catalysts are very effective in decomposing formaldehyde into carbon dioxide and water at room temperature. However, their application is still limited owing to the following: (1) the potential interference of humidity and other contaminants in the indoor environment on catalytic oxidation efficiency (conversion efficiency and by-products); (2) the high cost of precious metal catalysts; and (3) most of these catalysts are harmful to humans (Kim et al. 2018; Liu et al. 2019a; Zhu et al. 2017). Photocatalytic oxidation has received increased attention because the TiO_2 catalyst is relatively harmless. However, photocatalytic oxidation degradation can produce harmful by-products because the specific surface area of this catalyst is small and the redox reaction it catalyzes is often incomplete (Akbarzadeh et al. 2010; Liu et al. 2017; Luo et al. 2016). Compared to the abovementioned strategies, biodegradation offers better potential for widespread application because it is both environmentally friendly and cost-effective (Delhomenie and Heitz 2005).

Table 1 The advantages and disadvantages of different formaldehyde removal methods

Methods		Advantages	Disadvantages	References
Physio-chemical methods	Physical adsorption	Widely used; excellent performance	Remove pollutant incompletely; adsorbents replacement or regeneration required	Bellat et al. (2015); Liu et al. (2019a, b); Na et al. (2019)
	Oxidation catalysis	Effective	High cost of metal catalysts; Most catalysts harmful	Gu et al. (2019); Rong et al. (2018)
	Photocatalytic oxidation	TiO_2 catalyst harmless; Promising and effective.	Harmful by-products; Redox reaction incompletely.	Chen et al. (2005); Liao et al. (2012) Tasbihi et al. (2015)
Biological removal	Phyto-remediation	Clean and safe; High applicability; Environment friendly	Ineffective; Uptake formaldehyde slowly	Khaksar et al. (2016a, b); Pettit et al. (2017); Wood et al. (2006)
	Microbial removal	More effective; Economical & novel.	Bio-safety risk; Leakage possibility	Darlington et al. (2001); Guieysse et al. (2008); Mudliar et al. (2010)

In the past decades, there are a few publications of review on the degradation and removal of formaldehyde. Most of the published reviews are about physical and chemical methods to remove formaldehyde; for examples, TiO_2 photocatalysis degradation of indoor formaldehyde (Liao et al. 2012; Tasbihi et al. 2015), sorptive removal of formaldehyde via high-performance materials like carbon nano tubes, metal-organic frameworks, graphene oxides for effective (Na et al. 2019), and carbon-based materials (Suresh and Bandosz 2018), and catalytic oxidization and chemisorption methods for indoor formaldehyde removal (Pei and Zhang 2011). However, to our knowledge, there is no publication of review on the biodegradation removal of indoor gaseous formaldehyde. Thus, this article is the first review on the principles and applications of biodegradation to summarize the pros and cons and prospects for the future of biological methods for removal of indoor gaseous formaldehyde.

Biological strategy of formaldehyde removal and its mechanism

Phytoremediation

Phytoremediation strategy includes phytoextraction, stomatal uptake, phytodegradation, and phytovolatilization. Both spider plant (*Chlorphytum comosum*) and soybean (*Glycine max* L.) can remove formaldehyde from indoor air. Formaldehyde enters the plant leaves through the stomata and corneum or dissolves in the aqueous solutions and then enters the tissue through the capillary diffusion of the root (Giese et al. 1994; Ugrekhelidze et al. 1997). Formaldehyde in air can be removed by a spider plant (*Chlorphytum comosum*)-soil system and higher plant, and in microorganism metabolism is detected in the daytime resulting in higher, 95% removal (Xu et al. 2011). The different parts of indoor plants make different contributions. The major contributor to volatile formaldehyde removal is the root zone, not the aerial portions as exhibited in a chamber experiment of 2-year-old plants *Ficus benjamina* and *Fatsia japonica* (Kim et al. 2008). Recently, water hyacinth was found to absorb formaldehyde from the water through its roots and transport it to its leaves (Gong et al. 2018), suggesting its potential in an integrated system for indoor air formaldehyde removal.

After formaldehyde enters a plant tissue, it typically enters the C1 metabolism or is transformed into carbon dioxide (Pilon-Smits 2005). Formaldehyde can enter plant C1 metabolism through the pathway shown in Fig. 1. Based on the enzymatic and genomic evidence, one-carbon metabolism in higher plants is divided into four sectors: (1) folic acid mediated reactions; (2) folate-independent reactions; (3) activated methyl cycles; and (4) *S*-methylmethionine (SMM) cycles (Hanson and Roje 2001).

In addition to C1 metabolism through enzymatic reaction, a redox reaction is another possible route for formaldehyde removal by a plant. For example, Zhao et al. reports that the main metabolic mechanisms of two wild plants, *Plantago asiatica* L. and *Taraxacum mongolicum* Hand.-Mazz, are redox reaction and an enzymatic reaction, respectively (Zhao et al. 2019). When plant shoots were exposed to formaldehyde at a concentration of 1.28 mg/m^3 for 24 h, the formaldehyde removal rates of *P. asiatica* and *T. mongolicum* reached 73.18 and 121.20 mg/h/kg FW (fresh weight), respectively. Liang et al. also found that a redox reaction between plant oxidant and formaldehyde may represent the main mechanism of plant formaldehyde decomposition, by comparing the dissipative ability of potted *Chlorophytum comosum* fresh leaves and leaf extracts to the addition of formaldehyde (Liang et al. 2018). When the initial concentration of formaldehyde gradually increased in the extract of leaves, the contribution of the enzymatic reaction (e.g., dehydrogenase catalysis) to remove the added formaldehyde drastically decreased while the corresponding redox reaction increased.

Microbial removal

Microorganisms with formaldehyde removal capacity are very diverse in nature. An increasing number of formaldehyde-degrading microbial strains have been isolated and identified, including bacterial strains such as *Methylobacterium* sp. MF1 (Mitsui et al. 2005) and *Methylobacterium* sp. XJLW (Qiu et al. 2014), yeast strains such as *Debaryomyces vanriji* (Kato et al. 1982), fungi such as *Aspergillus nomius* (Kondo et al. 2002), and marine algae such as *Nannochloropsis* (Yoshida et al. 2009) (Table 2).

Metabolic pathway of formaldehyde microbial removal is similar to that of phytoremediation. For example, in the methanol metabolism pathway of *Methylobacterium extorquens* (Fig. 1), formaldehyde can enter the dissimilation pathway as an internal metabolite of methanol dehydrogenase (Ochsner et al. 2015). Formaldehyde is further oxidized in the H_4MPT-dependent pathway. However, the difference between microbial removal and phytoremediation is that in microorganisms a variety of complex removal methods are involved and a redox reaction is the major one.

In general, the microbial metabolism pathway of C1 compounds, such as formaldehyde and methanol, includes three processes: (1) the oxidation of the C1 substrate to formaldehyde; (2) the oxidation of formaldehyde to carbon dioxide; and (3) assimilation into biomass. The first process is an oxidation reaction of non-formaldehyde C1 compounds, such as methanol, which are transformed to formaldehyde through an H_4MPT-dependent pathway. H_4MPT also participates in formaldehyde detoxification (Chistoserdova 2011; Marx et al. 2003). As a spontaneous reaction, the methyl groups on the formaldehyde and H_4MPT are reacted to form

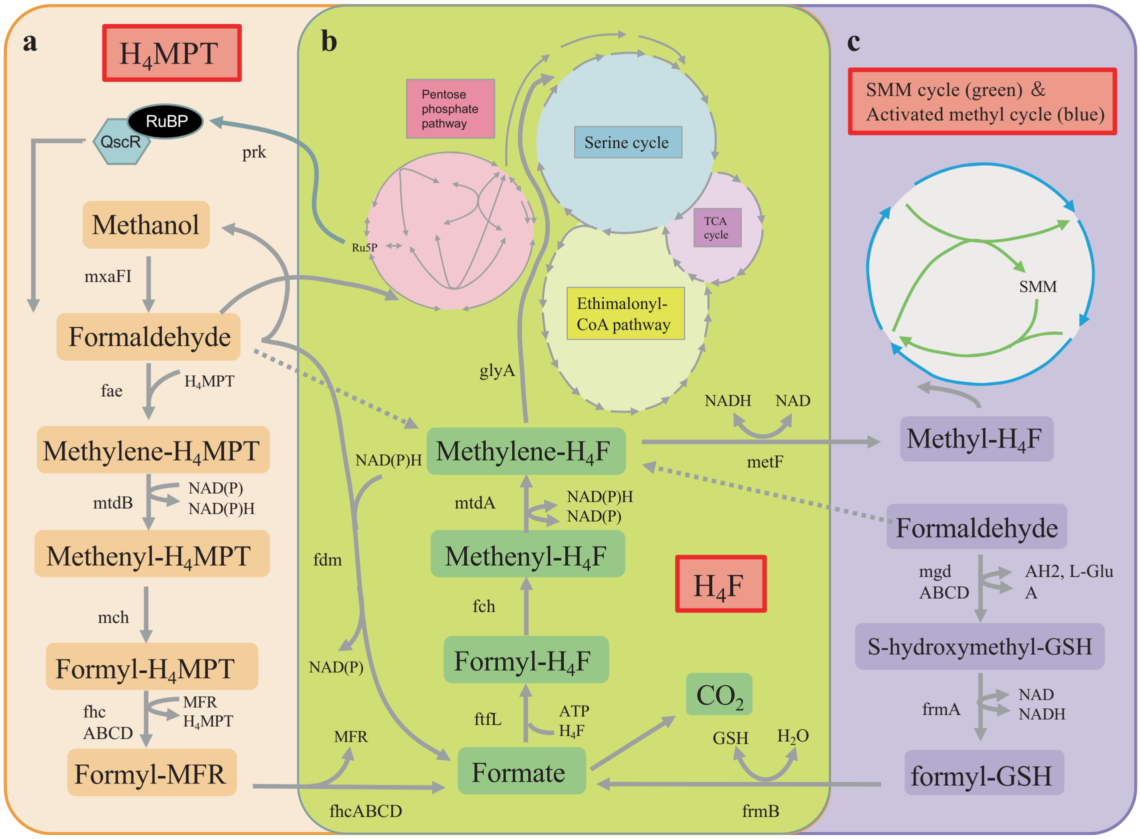

Fig. 1 Main pathways of plant and microbial degradation of formaldehyde. **a** H_4MPT-dependent pathway, mainly found in microorganisms; **b** H_4F-dependent pathway, serine cycle, TCA cycle, and ethinmalonyl-CoA pathway, found both in plants and microorganisms, contain these metabolic pathways; **c** Folate-independent pathway, SMM cycle, and activated methyl cycle, mainly found in plants. (H_4MPT, trahydromethanopterin; H_4F, tetrahydrofolate; RuBP, ribulose-1,5-bisphosphate; QscR, the serine cycle regulator; Ru5P, ribulose-5-phosphate; GSH, glutathione; SMM, S-methylmethionine; MFR, methanofuran. The dashed arrows indicate reactions that can occur spontaneously, but they are inefficient. RuBP and QscR regulate the expression of one-carbon assimilation gene clusters)

methylene-H_4MPT (Chistoserdova et al. 2005), which was catalyzed by formaldehyde-activating enzyme (*fae*). *fae* is indispensable for methylotrophy. If the *fae* gene is deleted, the resulting mutant is sensitive to formaldehyde, indicating that spontaneous reactions are not sufficiently efficient to detoxify formaldehyde (Vorholt et al. 2000). Next, the C1 unit is transformed to formic acid through an H4MPT-dependent pathway. In addition, there are three other ways in which formaldehyde can be converted to formic acid: (1) formaldehyde is produced from methanol and formic acid through catalysis by formaldehyde dismutase (Kato et al. 1986; Park et al. 2010); (2) formaldehyde can be converted by S-(hydroxymethyl) glutathione synthase to generate S-(hydroxymethyl) glutathione (Goenrich et al. 2002) and formic acid is finally produced after enzymatic catalysis (Barber et al. 1996; Gonzalez et al. 2006; Gutheil et al. 1992; Harms et al. 1996; Ras et al. 1995); and (3) the H^+ of formaldehyde is replaced by OH^- under the action of glutathione-independent formaldehyde dehydrogenase (Diab et al. 2006). Formate dehydrogenase can transfer formic acid to eventually become carbon dioxide and water, and the C1 unit can assimilated into the serine cycle by the H_4F-dependent pathway.

In methylotrophic bacteria, formaldehyde can be immobilized on D-ribulose 5-phosphate (Ru5P) to form D-arabinose-3-hexulose-6-phosphate, followed by isomerization to fructose-6-phosphate (F6P). This pathway, called the ribulose monophosphate pathway (RuMP), is ubiquitously used for the detoxification of formaldehyde (Fig. 1). It is noteworthy noting that when driving one-carbon assimilation, ribulose-1,5-bisphosphate (RuBP) is an essential metabolite that induces a transcriptional regulator (Ochsner et al. 2017). This may be the way for methylotrophic microorganisms to respond to one-carbon compounds. For example, methane and methanol can be converted to formaldehyde, which is then fixed to Ru5P through the RuMP pathway. Phosphoribulokinase catalyzes the transformation of formaldehyde and Ru5P to form RuBP that then regulates the expression of one-carbon assimilation gene clusters.

The increasing number of publications on one-carbon metabolic pathways has resulted in an improved knowledge of methyltrophy. However, new insights based on genomics, transcriptomics, and other -omics technologies are beginning to reveal the complete mechanism of these pathways. For example, Ochsner et al. discovered and verified the

Table 2 Comparison of published formaldehyde removal microorganisms

Kind	Strains	Sources	Formaldehyde-degrading performance & application	Sequencing (Genbank #)	References
Bacteria	*Methylobacterium* sp. MF1	Soil	6 $mg \cdot L^{-1} \cdot h^{-1}$	NO	Mitsui et al. (2005)
	Methylobacterium extorquens	Soil and wastewater	1.85–2.96 $g \cdot L^{-1}$	NO	Di Maiuta et al. (2009); Mirdamadi et al. (2005)
	Methylobacterium sp. XJLW	Soil	Rest cells 1.68 $g \cdot L^{-1} \cdot h^{-1}$; Entrapped cells, 0.46 $g \cdot L^{-1} \cdot h^{-1}$	NZ_CP017427	Qiu et al. (2014)
	Pseudomonas pseudoalcaligenes	Soil and wastewater	154 $mg \cdot L^{-1} \cdot h^{-1}$; 58 $mg \cdot L^{-1} \cdot h^{-1}$	NO	Mirdamadi et al. (2005)
	Pseudomonas putida	Industrial wastewater	6.94 $mg \cdot L^{-1} \cdot h^{-1}$	MK645976.1	Adroer et al. (1990); Di Maiuta et al. (2009)
	Pseudomonas cepacia	Wastewater	1.0 $g \cdot L^{-1}$, 18–24 h.	NO	Glancer-Soljan et al. (2001)
	Pseudomonas putida NS15	Soil	57.8 $mg \cdot L^{-1} \cdot h^{-1}$	NO	Zvulunov et al. (2019)
	Pseudomonas putida B1	Waste water	0.0389 $g \cdot L^{-1} \cdot h^{-1}$,	NZ_CP022560.1	Wang et al. (2019)
	Bacillus cereus ERBP	Root of *C. ternatea*	0.01 $mg \cdot L^{-1} \cdot h^{-1}$	NO	Khaksar et al. (2016a, b)
Yeasts	*Debaryomyces vanriji*	Soil	0.15~0.55% coexisting glucose, completely, 96 h	NO	Kato et al. (1982)
	Trichosporon penicillatum	Soil	1.0 $g \cdot L^{-1}$, 8–24 h.	NO	Glancer-Soljan et al. (2001); Kato et al. (1982)
	Hansenula polymorpha	Soil and wastewater	1.75 $g \cdot L^{-1}$	NO	Kaszycki and Koloczek (2000)
Fungi	*Aspergillus nomius*	Soil	4.5 $g \cdot L^{-1}$,	NO	Kondo et al. (2002)
	Aspergillus oryzae	Soil	2.79 $mg \cdot m^{-3} \cdot h^{-1}$	NC_036442.1	Minemura et al. (2017)
	Penicillium chrysogenum	Deep sea	2.9 $mg \cdot L^{-1} \cdot h^{-1}$	JX480902.1	Luo et al. (2014)
	Paecilomyces sp. No. 5	Soil	2.0%, 20 days; 2.4%, 55 days.	NO	Iwahara et al. (2002)
	Fusarium solani B1	Plant	0.2731 $g \cdot L^{-1} \cdot h^{-1}$	KM229694.1	Vergara-Fernande et al. (2018)
Marine algae	*Nannochloropsis oculata* ST-3	Sea water	26.65 $mg \cdot m^{-3}$, 99.3%, 22 days	NO	Yoshida et al. (2009)

abovementioned effect of RuBP on one-carbon metabolism through genomic and transcriptomic methods (Ochsner et al. 2017). Many methylotrophs remove formaldehyde using dismutase which transforms formaldehyde to formic acid and methanol. Many methylotrophs, such as *Methylobacterium* sp. XJLW, contain only H_4MPT-dependent pathways and H_4F-dependent pathways. However, comparative genomic and transcriptomic analysis results on *Methylobacterium* sp. XJLW suggest a novel C1 metabolism pathway containing novel enzymes such as methyltransferase 12,775. XJLW can degrade methanol without intermediate formaldehyde (Shao et al. 2019a). Unfortunately, XJLW accumulates formic acid when it utilizes formaldehyde as a sole carbon source.

Thus, it is important to understand the key regulatory factors in the pathways involved in the microbial degradation of formaldehyde. Further modification of formaldehyde-degrading strains may improve their efficiency and ability to treat indoor gaseous formaldehyde in real environment.

Application of biological treatment for indoor gaseous formaldehyde

Botanical filtration

Due to the characteristics of modern buildings and the fast pace of change in modern societies, simply increasing the ventilation rate of buildings to eliminate the accumulation of pollutants is both tough and uneconomical. Therefore, phytoremediation has received increased attention in recent decades due to its applicability and availability, as well as the environmental, economic, and social benefits of potentially achieving zero emissions.

At present, there are two main strategies for the removal of formaldehyde by plants. The first is to place potted plants directly in the indoor environment to remove continuously formaldehyde. When plants are existed in a room or workplace, the level of comfort, productivity, and mental function can be notably improved, and pain perception can be reduced (Lohr 2010; Mudliar et al. 2010; Soreanu et al. 2013; Zhou et al. 2006). The concentration of VOC in rooms with plants also reduced compared to the same room without plants (Fjeld 2000; Fjeld et al. 1998).

Another strategy is to develop active botanical biofiltration system, which uses air collectors to increase the mass of pollutants exposed to the plant substrate and plant foliage (Darlington et al. 2001; Irga et al. 2017a; Irga et al. 2017b; Pettit et al. 2017). Such a system can potentially replace or enhance a traditional heating, ventilation, air conditioning, and cooling (HVAC) system in maintaining indoor air quality (Chen et al. 2005).

However, most plants have limited ability to remove formaldehyde in the absence of rhizosphere microorganisms (Chen et al. 2010), because the amount of formaldehyde removed by redox is generally limited. In addition, because of their low uptake rate, the accumulation and formaldehyde metabolism in plants are unlikely to be efficient for the purification of indoor. Microorganisms, especially rhizosphere bacteria, are able to efficiently degrade VOC including compounds containing methyl or methyoxy groups (Orita et al. 2005). Thus, the plant root zone and its rhizosphere microorganisms both require consideration for efficient formaldehyde removal through phytoremediation (Aydogan and Montoya 2011; Godish 1989; Kim et al. 2008; Xu et al. 2010).

Application of microorganisms to remove formaldehyde

Above all, it is necessary to select a suitable kind of microorganism for application for formaldehyde removal. As shown in Table 2, an increasing number of formaldehyde-degrading microorganisms have been isolated and identified, including bacteria, yeast, fungi, and marine algae. Besides the efficiency of formaldehyde metabolism, the following three aspects have been taken into consideration for selection of suitable microorganism for application. Firstly, fungi are regarded as unsuitable one for large-scale cultivation and use in biofilters. While fungi have excellent formaldehyde removal ability, they can grow numerous hyphae and form spores at certain temperatures and humidities. These characteristics limit their role in indoor air purification because of their adverse impact on human health. Secondly, pathogenic microorganisms cannot be used in filters because of the possibility of leakage. Therefore, current research on microbial removal of indoor pollutants has mainly focused on non-pathogenic bacteria. Thirdly, for pathogenic bacteria having excellent formaldehyde-degrading ability, their formaldehyde-degrading gene or gene clusters encoding formaldehyde-degrading pathway can be transferred to a genetically engineered host cells for application in indoor air treatment. For example, the formaldehyde dismutase gene of *Methylobacterium* sp. FD1 can be expressed in non-pathogenic *Escherichia coli* and resting cells of resultant recombinant *E. coli* can be used to degrade high concentrations of formaldehyde (Yonemitsu and Kikuchi 2018).

Then, a suitable biofiltration device is required for the microbial treatment of formaldehyde on scale-up level. The commonly used device is trickling biofilter which contains three components: (1) an air compressor for pumping gas into the reaction unit, including humidification device; (2) a biofilter bed with a multi-layered reaction body containing filler and microbes; (3) exhaust gas treated component (Fig. 2) (Guieysse et al. 2008). Trickling biofilters have been widely used in treatment of industrial gaseous pollutants, and recently was also be used for formaldehyde removal (Lu et al. 2012).

When a strain is utilized alone or in combination with other methods for formaldehyde removal in a biofilter, its potential biohazard risk due to leakage of strains still require carefully

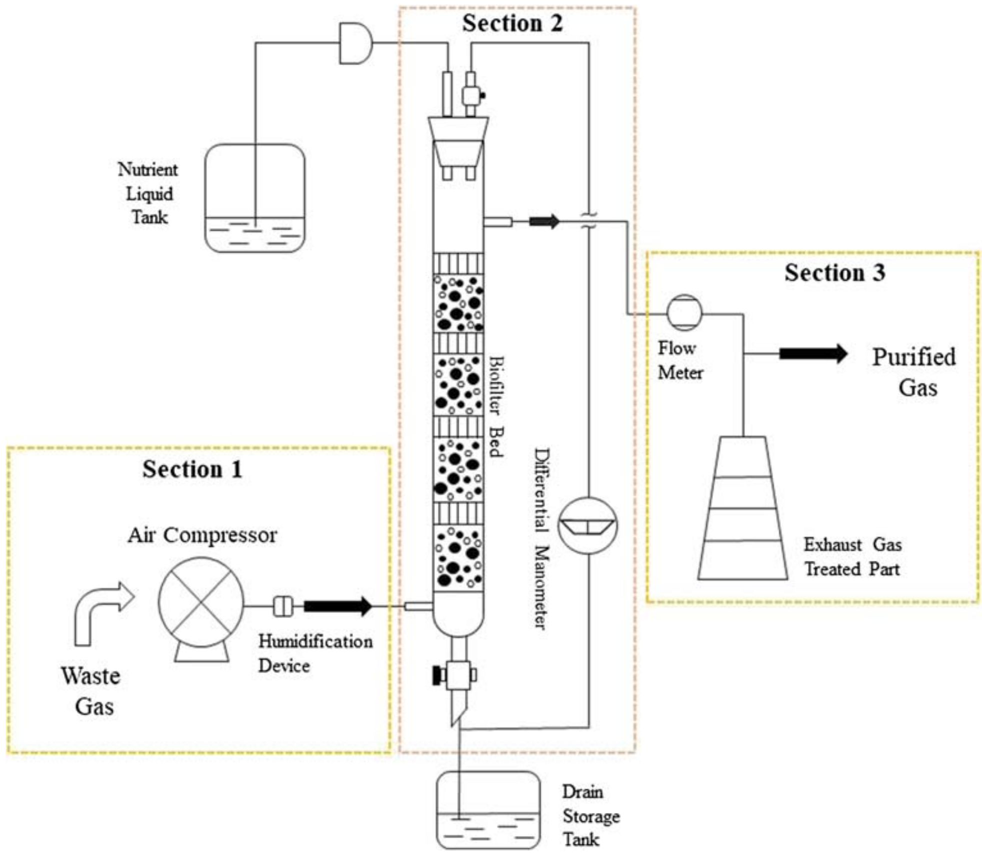

Fig. 2 Schematic of the trickling biofilter system with three sections: (1) an air compressor for pumping waste gas into the reaction unit; (2) a biofilter bed which is a multi-layered reaction body containing filler and microbes to degrade the waste gas; (3) exhaust gas-treated parts to prevent harmful microbes from escaping into the air. In addition, there are a nutrient tank to provide nutrients for microbial growth and a tank to storage waste water

evaluation (Guieysse et al. 2008). Microbial cells can be entrapped within calcium alginate to form microbial granules to reduce the potential biohazard risk. For example, the biofilter containing embedded strains exhibited excellent operating performance for formaldehyde removal from indoor air without microbial leakage (Lu et al. 2012). However, such an approach can also adversely impact the removal efficiency and results in a high pressure drop, which requires energy consumption. Furthermore, the possibility of microbial leakage still exists, even when microbial cells entrapped in a filter with anti-leakage measures. In conclusion, non-pathogenic bacteria are essential for formaldehyde-degrading filters applied in indoor air treatment. Besides, secondary pollution, such as biological aerosols, has also attracted researchers' attention. Biological aerosols include airborne particles (viruses, bacteria, etc.) of all biological origin and are harmful to the ecological environment and human health (Esquivel-Gonzalez et al. 2017). Photocatalytic systems based on Perlite-supported ZnO and TiO_2 is found a potential strategy to process the bioaerosols produced from biofilter (Valdez-Castillo et al. 2019). The main mechanism of bioaerosol inactivation was related to cell death. Thus, the treatment of microorganisms contained in biofilters and exhaust gas should be taken into consideration for a suitable filter devices design before real application for indoor gaseous formaldehyde treatment.

Finally, potential novel biofilter for formaldehyde removal must meet some specific nutritional and environmental conditions requirements. (1) The first is to control pH in the biofilters system. In our previous study, it was found that formic acid may accumulate in the formaldehyde metabolism pathway of *Methylobacterium* sp. XJLW, resulting in decreased pH and decreased efficiency (Qiu et al. 2014). Thus, a pH control strategy is often required in biofiltration technology. For example, the continuously addition of ozone can improve the removal efficiency of a biofilter and maintain its prolonged operation (Maldonado-Diaz and Arriaga 2015), because the addition of ozone helps maintain the optimal biofilter pH between 7.5 and 8.2. Unfortunately, this method is not economical. (2) The second is to improve mass transfer rate for substrates. Current microbial-based biofilters are generally packed tower type filtration devices requiring continuous inlet of indoor air into the filter interior. Moreover, formaldehyde needs to enter and dissolve in an aqueous phase so that microbial cells can adsorb the formaldehyde for intracellular metabolism. To improve mass transfer rate, new technologies for more efficient pollution transfer need to be developed, such as membrane bioreactors may be an effective solution (Guieysse et al. 2008). (3) The third is to supply nutrients enough for the microbial metabolism. Contaminant compounds in indoor air are usually the carbon sources and/or nitrogen sources for microorganisms in biological filters.

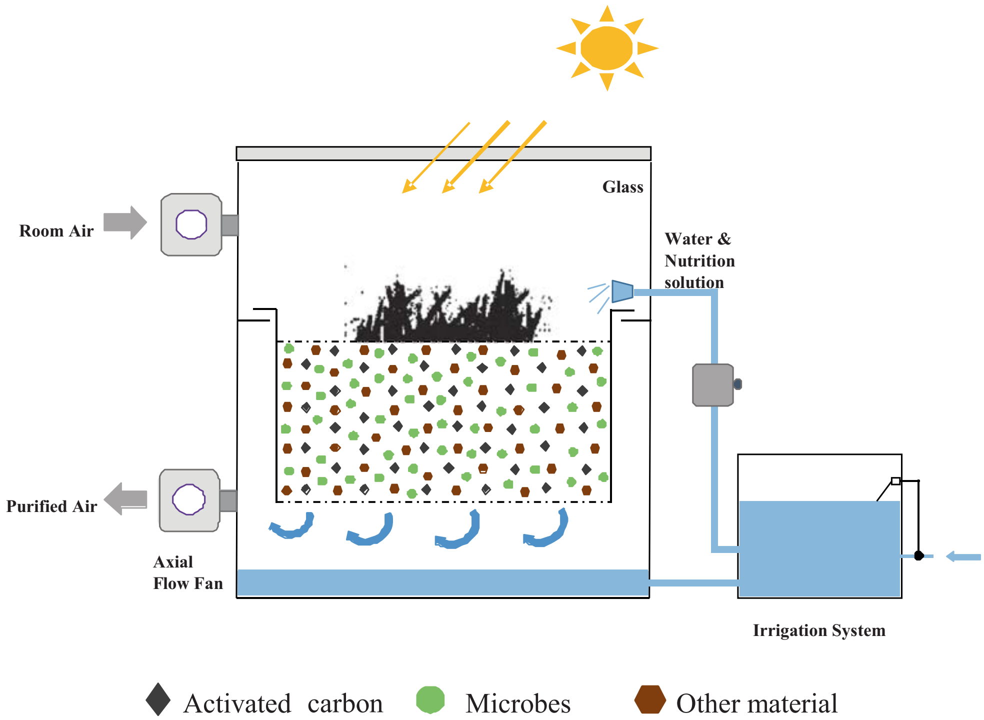

Fig. 3 Schematic of a dynamic botanical air filtration system. The DBAF system used different materials as root bed of selected plants with microorganisms growing in the root system. The filtration system is operated with periodical irrigation and airflow passing through. An axial flow fan is installed to transport room air. The adsorbed and/or absorbed organic compounds are degraded by the microbes, regenerating the sorbent-based root bed. The purified air is released into indoor environment. The irrigation control sensor is buried on the right side of the bed. When the moisture content is below the lower limit, the fan will be stopped, and irrigation system triggered and operated until the moisture content is higher than the higher limit

The concentrations of these indoor pollutants may be too low for microbial growth even though these levels are harmful to humans. If the growth rate of the microorganism is inappropriate, it will inevitably adversely impact the durability and removal efficiency of a microbial filter. There are two strategies to solve the issue of insufficient nutrients for microbial cells in biofilters. One strategy is to use resting cells as an inoculum or to directly use the relevant enzymes isolated from microorganism. For example, the resting cells of *Methylobacterium* sp. XJLW have been used to degrade the high concentrations of formaldehyde in the simulated device and lasted for up to 200 h (Shao et al. 2019b). The resting cells, entrapped within the support materials, could be replaced when their formaldehyde degradation efficiency decreased. Another strategy is to add additional nutrients to the biofilter but this can be uneconomical and cumbersome. Additional research or a clever design strategy is required to solve this problem.

Collaboration between microbe and plant

Botanical biofilters integrate green plants into the structure of biofilters, making them an environmentally friendly technology that uses plants and their rhizosphere microbes (natural microorganisms living near or in the roots of plants) to remove contaminants from the environment. The collaboration between plants and microbes is not limited to the rhizosphere. When an endophytic bacterium, such as *Bacillus cereus*, was inoculated into the roots, non-native host plants exhibit increased resistance to formaldehyde toxicity (Khaksar et al. 2016a; Khaksar et al. 2016b; Plaisance et al. 2017).

In industry, bioreactors are often constrained by required filler materials. Microorganisms can metabolize organic components present in these filler materials as a source of nutrition. The use of such fillers greatly limits the service life of the filler material (Devinny et al. 1999). Moreover, the most effective bioreactors for removing pollution have the shortest filler lifetime. Plants in the botanical biofilters can supplement the organic matter in fillers by secretions from their living roots or organic matter coming from the decomposition of dead roots. This can greatly extend the life of the filler. At the same time, carbon dioxide, the final product of microbial degradation of formaldehyde, is absorbed and fixed by photosynthesis into organic matter in plants. Clearly plants have limited capability to assimilate gaseous formaldehyde (Chen et al. 2010). Although formaldehyde is a natural metabolite of plants, it is usually present in plants in a form of linked cofactors such as tetrahydrofolate and glutathione. Some microorganisms are capable to fix formaldehyde as a cellular component when endogenous formaldehyde was produced or when they exposed to exogenous formaldehyde. In addition to combining the use microorganisms and plants, researchers are also genetically engineering plants to enhance their tolerance and removal formaldehyde. For examples, Chen et al. introduced a 3-hexulose-6-phosphate synthase (HPS)- and 6-phosphate-3-hexuloisomerase (PHI)-dependent formaldehyde fixation pathway into *Arabidopsis thaliana* and tobacco with the ribulose monophosphate synthesis pathways linked to each other (Chen et al. 2010). The RMP pathway is one of the ways in which methylotrophs fix formaldehyde. Song et al. overexpressed HPS/HPI fusion enzymes in the chloroplasts of geranium (Song et al. 2010). In addition, plant-derived genes also enhance the formaldehyde removal capacity of other plants (Lee et al. 2015). These methods provide a new strategy for the removal of formaldehyde contamination as well as additional VOCs contamination problems.

Collaboration between adsorption and microbes

Many studies have described the impact of substrates with different compositions on VOC removal. The innate capability of plant growth substrates to assimilate formaldehyde has also been noted (Godish 1989; Hormann et al. 2017). Aydogan and Montoya reveal the contribution of a substrate to removal efficiency (Aydogan and Montoya 2011). An activated carbon substrate exhibits greater formaldehyde reduction ability than expanded clay and growstone. A substrate with a high adsorption capacity and adequate microbial sites should increase microbial removal efficiency. As further evidence, Irga et al. found that differences in benzene removal efficiency between potted plants grown in different substrates (hydroponics and soil culture) (Irga et al. 2013) resulted from the differences in diversity and density of substrate microbial communities. Zvulunov et al. designed a versatile regenerative system including montmorillonite clay, polyethyleneimine, and formaldehyde-degrading *Pseudomonas putida* for adsorbing formaldehyde in water and degrading formaldehyde by attached bacteria (Zvulunov et al. 2019). The adsorbent not only absorbs formaldehyde but also reduces the cytotoxicity of formaldehyde and can release formaldehyde slowly. Such systems also have the potential in application for formaldehyde removal from indoor air. However, of these systems, redesign is required for accommodating the indoor environment. For example, Wang et al. designed a dynamic botanical air filtration system (DBAF) (Fig. 3) that uses activated carbon, hydroponics, and microbes to decompose formaldehyde, which could be integrated into the HVAC system of the new office space (96.8 m^2) (Wang and Zhang 2011).

Among the abovementioned strategies, the most potential strategy to remove indoor formaldehyde seems to require the combination of biological methods and other methods without resulting in secondary pollution, allowing these combined systems to compensate for each other's defects. Thus, a plant-adsorbent-microbial system and the air conditioning system connected to a biofilter is becoming an accepted strategy for the design of more ideal facilities to remove formaldehyde from indoor air. Thus, the exploration of safe, non-polluting, and economical way to achieve the long-term efficient and safe removal of formaldehyde remains a critical need.

Conclusions

Biological removal of formaldehyde, even removal of other VOCs, has greater applicability than other methods. It is theoretically possible to remove any indoor contaminants, provided suitable microorganism or plant strains are isolated. However, the combination of biological treatment with absorption and air conditioning system is the direction for removal of indoor gaseous formaldehyde pollution.

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Compliance with ethical standards

Conflict of interest All authors declare no conflict of interest in this work.

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