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Review

Simultaneous Removal of Multicomponent VOCs in Biofilters

Chunping Yang,[1,4,*] Hui Qian,[1,4] Xiang Li,[1,4] Yan Cheng,[2,4] Huijun He,[2,4] Guangming Zeng,[1,4] and Jinying Xi[3,4,*]

Volatile organic compounds (VOCs) are significant atmospheric pollutants that cause environmental and health risks. Waste gases polluted with multiple VOCs often need to be purified simultaneously in biofilters, which may lead to antagonistic, neutral, or synergistic effects on removal performance. Antagonism limits the application of biofilters to simultaneous treatment of multiple VOCs, while synergism has not yet been fully exploited. We review the interactions among multiple target pollutants and the changes in the bioavailability and biodegradability of substrates that are responsible for substrate interactions. Potential strategies for enhancing biofilter performance are then discussed. Finally, we propose further efforts to alleviate antagonism by enhancing bioavailability and biodegradability, and discuss possible challenges to take advantage of synergism.

Practical Applications and Limitations in Simultaneous Biofiltration of Multiple VOCs

Volatile organic compounds (VOCs; see Glossary), that are emitted in large quantities from industries producing paper, paint, solvents, and wood products, as well as from fuel/petroleum industries and pharmaceutical operations, threaten the environment and human health [1–6]. Biological techniques based on microbial metabolism are usually more effective than other techniques for controlling VOC emissions [7–9]. Overall, the characteristics of target VOC pollutants, principally their hydrophobicity (Box 1), govern their removal performance in biofilters [1,2,7,10]. There are numerous publications on the removal of different types of VOCs, and several excellent review papers focusing on single-component VOC removal are currently available [7,10–13]. In practice, waste gases emitted from industrial processes and municipal facilities are complex, and are often composed of multiple VOCs that have different characteristics from single-compound VOCs [7,14–16]. Several recent publications have addressed the removal of multiple VOCs by gas biofilters, but no comprehensive review has so far addressed the complexity of the processes and the poor removal performance for hydrophobic VOCs.

The removal efficiency of a single-component VOC during biofiltration is only related to its own **bioavailability**, while for multicomponent VOC mixtures the characteristics of each compound in the mixture are also relevant. Substrate interactions between target pollutants can alter the biodegradation rate of an individual compound, either antagonistically, neutrally, or synergistically [17–19]. Therefore, we describe these as antagonistic, neutral, and synergistic interactions. Antagonistic interactions have become the primary obstacle to the effective biodegradation of these pollutants. By contrast, synergistic interactions have shown potential as an alternative process for reluctant VOC removal with biofiltration. Understanding the possible mechanisms that underlie whether a substrate interaction is detrimental, neutral,

Highlights

The structure of microbial populations plays an important role in the interactions between hydrophobic and hydrophilic VOCs, and the application of specific single species or mixed microorganisms may alter substrate interactions and consequently enhance removal performance.

Enhancing the bioavailability of reluctant VOCs can better offset the negative interactions exerted by the cosubstrates.

Strategies to alleviate the negative interactions among multiple VOCs will make it possible to employ biofilters for full-scale removal of multiple VOCs.

Biofilter performance for hydrophobic VOCs can be enhanced by exploiting the synergistic interactions of hydrophilic substrates. Regulating operational parameters, such as changing the feeding loading rate for every component and alternating the use of some hydrophilic compounds, may be promising strategies.

[1]College of Environmental Science and Engineering, Hunan University and Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha, Hunan 410082, China
[2]College of Environmental Science and Engineering, Guilin University of Technology, Guilin, Guangxi 541006, China
[3]School of Environment, Tsinghua University, Beijing 100084, China
[4]These authors contributed equally to this paper.

*Correspondence:
yangc@hnu.edu.cn (C. Yang) and
xijinying@tsinghua.edu.cn (J. Xi).

Box 1. Classification of VOCs and Their Removal Performance in Biofilters

VOCs are usually classified into two groups – hydrophilic VOCs and hydrophobic VOCs – according to their Henry's law constant, H, at 25°C [7]. Generally, substrates with H values under 0.1 are classified as hydrophilic VOCs (examples include alcohols, ketones, aldehydes, and volatile fatty acids), and substrates with values of H under over 0.1 are classified as hydrophobic VOCs (examples include alkanes, alkenes, and aromatics) [98]. This classification is confirmed by empirical observations and can approximately evaluate the removal performance of a target pollutant in a biofilter. The H value is a proportionality constant that describes the ratio of VOC concentration in the air phase and the water phase, and this governs the mass-transfer rate from the air phase to the water phase [7]. Hydrophilic substrates can be more effectively biodegraded in biofilters because high gas–liquid transfer rates make target pollutants more available to microorganisms [7]. Unfortunately, when hydrophobic VOCs are fed into biofilters, the lower mass-transfer rate from the air phase to the liquid phase limits the supply of compounds to the microorganisms, which leads to poor removal performance [10,20,73]. These low mass-transfer rates are the main limitations to removal performance for hydrophobic VOCs.

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or beneficial to the removal of target pollutants can provide significant insight to permit simultaneous and efficient treatment of multiple VOCs in biofilters. Therefore, we focus on how substrate interactions affect biofilter performance, and we suggest techniques to enhance biofilter performance by alleviating antagonistic interactions among multiple VOCs or by regulating complex substrate interactions.

Antagonistic Interactions Among Multiple VOCs

Simultaneous Biofiltration for Removal of Multiple Hydrophobic VOCs

Hydrophobic VOCs are faced with a poor removal rate because their low gas–liquid transfer rates limit the uptake for metabolism by microorganisms [20–22]. When multiple hydrophobic VOCs are concurrently degraded in a biofilter, the elimination capacity for each compound is generally suppressed significantly in comparison with a single compound [23]. Usually, the removal of hydrophobic compound with a higher Henry's law constant, H, is suppressed by other components in the mixtures [15,24]. For example, both toluene and chlorobenzene decreased the removal rate of methane when methane biofiltration was evaluated in the presence of either toluene or chlorobenzene [25]. The degradation of *n*-hexane was inhibited significantly in the presence of **aromatic compounds** in a biofilter, and competitive inhibition kinetic analysis accurately described the performance of the biofilter [15,24].

Notably, in hydrophobic VOC mixtures with similar chemical structures such as benzene (B), toluene (T), ethylbenzene (E) and xylenes (X), the removal performance for each component is lower than that of single-VOC biofiltration at an identical total feeding load [26–28]. Generally, benzene and xylenes are more recalcitrant substrates, and their processing is likely to be inhibited more than for toluene or ethylbenzene [3,26]. Furthermore, Rene and colleagues found an inhibitive effect wherein xylene removal was decreased with an increased concentration of toluene [3]. This phenomenon may be attributed to the small differences in hydrophobicity and substituent groups [17]. In a biofilter, toluene and ethylbenzene are likely to be more easily captured for metabolism by microbes growing on the **packing media**, which results in better biodegradation [27]. In addition, this preferential metabolism could also lead to lag phases before other components are consumed [29]. Interestingly, in a benzene/toluene mixture, benzene exhibited a larger maximum elimination capacity than toluene, and the presence of benzene antagonistically affected the removal of toluene [30]. The presence of xylenes can also hinder toluene metabolic processes, while toluene has a neutral effect on xylene removal [31]. Similarities among the physicochemical properties and chemical structures of BTEX compounds (benzene, toluene, ethylbenzene, and xylene) are considered to result in similar metabolic processes and enzymatic systems [32]. Therefore, competitive inhibition effects take place among substrates in biofilters simultaneously fed with multiple BTEX compounds [33]. Alternatively, **catabolite repression** may be responsible for the inhibitory effect [17,34].

Glossary

Aromatic compounds: organic compounds with benzene rings. Their stable chemical structures make them resistant to disintegration and toxic to microorganisms and human beings.

Bioavailability: the accessibility of a chemical compound for assimilation. Contact between microorganisms and the compound is an essential prerequisite for bioavailability.

Biodegradability: the capacity of a chemical compound to be degraded by microbes. Biodegradability is a significant factor affecting the removal performance of a compound in a biofilter.

Biofilms: composed of microbes and their secreted extracellular polymeric substances. The characteristics of biofilms are related to the amount of biomass, which is partially responsible for biofilter performance.

Catabolite repression: a phenomenon where the metabolites of some compounds can repress the synthesis of some enzymes.

Cometabolism: a phenomenon where one compound cannot be biodegraded unless another particular compound is present. Cometabolism plays an essential role in the biodegradation of organic compounds.

Induction effect: a phenomenon where the presence of some compounds induces or enhances the synthesis of particular enzymes.

Polar compound: a hydrophilic compound, one that is soluble in an aqueous phase.

Packing media: the solid media packed in a biofilter and on which the microbes grow and metabolize. It plays a significant role in biofiltration for gas pollutants.

Volatile organic compounds (VOCs): organic compounds whose boiling points are less than 250 °C and whose melting points are below indoor temperature at standard pressure. Some examples include methanol, acetone, *n*-hexane, chlorobenzene, and benzene.

For simultaneous biofiltration of hydrophilic and hydrophobic VOCs, hydrophilic compounds have better removal efficiency than hydrophobic compounds [35,36]. Generally, the removal of hydrophobic VOCs is adversely affected by the presence of hydrophilic VOCs, while the presence of hydrophilic VOCs has little inhibitory effect on the removal of hydrophobic VOCs. For example, the introduction of ethyl acetate clearly suppressed the biodegradation rate of toluene whereas, conversely, ethyl acetate removal was negligibly affected in the presence of toluene [37]. In addition, the assimilation of hydrophobic pollutants is more sensitive to changes in operational conditions in biofilters than that of hydrophilic compounds. Generally, antagonism in the removal of recalcitrant compounds is more pronounced for increasing concentrations of **polar compounds**. Chan and Wu found that the biodegradation rate of toluene was inversely proportional to the loading rate of the hydrophilic methyl ethyl ketone (MEK), but an increasing concentration of toluene had no pronounced effect on MEK removal [38]. Step-changes in the inlet loading rates of methanol were significantly inhibitory to the assimilation of α-pinene, while the removal of methanol was negligibly affected by fluctuations in α-pinene concentrations in the inlet airstream [5]. Álvarez-Hornos and colleagues hypothesized that the difference in the biodegradation of target pollutants could be related to their solubility in water and the specific growth rate of degrading microorganisms, and found that polar compounds had higher concentrations in **biofilms** owing to their high hydrophilicity, which led to increased biodegradation rates unless self-inhibition levels were achieved [39]. However, overloaded acetone not only significantly inhibited the removal performance of styrene but also triggered a decrease in acetone biodegradation, which might be caused by significant cell damage at high concentrations [40]. Kinetic analysis of biofiltration for toluene showed that both microbial growth and biochemical reactions were inhibited by the introduction of MEK [38]. The inhibition of toluene removal might be interpreted as a failure of toluene-degraders competing for O_2 [41]. In addition, the accumulation of toxic metabolites or acidic byproducts could hinder the assimilation of hydrophobic substrates [42]. When hydrophobic toluene, α-pinene, and n-hexane were fed into a biofilter together with methyl mercaptan, the gradual accumulation of toxic metabolites irreversibly damaged the activity of functional species, leading to a decrease in the removal efficiency of hydrophobic substrates [43].

Simultaneous Biofiltration for Multiple Hydrophilic VOCs

Compared to the biodegradation of hydrophobic VOCs, hydrophilic VOCs can be more effectively degraded in biofilters [44]. For mixtures of hydrophilic VOCs with similar chemical structures, during biofiltration microorganisms can more readily consume compounds with either lower carbon content or a main chain without side groups. For example, when isomers of a butanol mixture were simultaneously biodegraded, n-butanol was readily biodegraded by the microbial community [45]. Similarly, MEK achieved a higher removal efficiency when MEK was fed into a biofilter together with methyl isobutyl ketone (MIBK) [46]. Moreover, fluctuant MEK vapor decreased the removal efficiency for both ketones, while no significant changes on removal rates were observed with a fluctuant MIBK vapor [46]. According to Datta and Philip [36], the antagonism between MEK and MIBK might be attributed to competitive inhibition.

Generally, the degradation rates of hydrophilic VOCs are related to biodegradation kinetics [47–49]. Biodegradation processes consist of two main components: microbial growth and the biochemical reaction itself [47]. These microbial growth and biochemical reaction processes for each pollutant may change due to the introduction of other pollutants, variations in the pollutant concentration, or both [45,48]. The adverse effect on MEK was more pronounced at low concentrations, and the adverse effect on methyl isopropyl ketone (MIPK) was more pronounced at high concentrations in a biochemical reaction process. However, both had similar antagonistic influences on the microbial growth process [48]. In a study by Chan and Su [49],

the inhibitory effect of ethyl acetate was equivalent to that of amyl acetate on the microbial growth process, but was stronger on the biochemical reaction process. Unfortunately, the influences of such changes on biodegradation kinetics are generally complex and unpredictable, and experimental assessments are therefore necessary.

For hydrophilic VOC mixtures with different chemical structures, the removal rates and their interactions are dependent on the stability and **biodegradability** of the target pollutants [50]. For example, the adverse effect on acetone removal exerted by isopropyl alcohol was more pronounced than that on isopropyl alcohol by acetone when the two compounds were simultaneously degraded [51]. In a biofilter cotreating a gas stream polluted by a mixture of formaldehyde and methanol, formaldehyde removal was suppressed by a high inlet loading of methanol [52].

Synergistic Interactions Among Multiple VOCs
Simultaneous Biofiltration of Multiple Hydrophobic VOCs
For simultaneous biofiltration of mixtures of waste gases with similar chemical structures (e.g., BTEX), substrate interactions can synergistically alter the removal rates of individual contaminants. For example, the preferential biodegradation of toluene in biofilters can promote microbial growth to enhance the metabolism of other cosubstrates [53]. Strauss and colleagues used a toluene-acclimatized pine-bark biofilter under thermophilic conditions (50°C) to treat paired mixtures of toluene and BEX, and the assimilation of BEX was stimulated in the presence of toluene [19]. An **induction effect** where one BTEX compound can induce the production of enzymes by a microbial population to biodegrade other BTEX compounds can also lead to synergistic effects in the removal of cosubstrates [29]. In addition, the removal of xylenes and benzene can be altered synergistically when particular microbial species are present, as well as by other operational parameters. For example, some xylene isomers are partially biodegraded or cannot be biodegraded by some strains even though BTE can be metabolized [54]. However, when they were simultaneously fed to biofilters inoculated with particular strains, the xylene isomers could be **cometabolized** with toluene, while an inhibitory effect on toluene removal could be also observed [55]. Generally, metabolic processes of multicomponent BTEX mixtures likely involve competitive or non-competitive inhibition, induction, and cometabolism in biofilters [17,56,57]. For example, the presence of toluene competitively inhibits the biodegradation of xylene; in turn, xylene can improve the removal efficiency of toluene [56]. Guo and colleagues reported that the biodegradation of benzene was positively affected by the introduction of toluene and xylenes, but was inhibited by ethylbenzene [57]. These results imply that antagonistic effects on removal performance for each component can be avoided to some extent via synergistic interactions.

Chlorinated hydrophobic VOCs such as trichloroethylene (TCE) and chloroform are generally poor sources of carbon or energy for microbes because of their highly oxidized state [58], and these compounds can be only biodegraded via cometabolism [59,60]. VOCs such as toluene and o-xylene have all been proven to make TCE more available to microorganisms and support its biodegradation when they are supplied to a biofilter together with TCE [59,61]. Of those, toluene has better performance for cometabolic TCE degradation [60]. Toluene can be preferentially utilized by a microbial population and can help to stimulate the release of oxidizing enzymes for degrading TCE, such as monooxygenase and dioxygenase, to enhance the TCE removal rate. However, competitive inhibition exists between toluene and low-concentration TCE [62–64]. Moreover, high-concentration TCE and continuous operation can also lead to decreased biofilter performance as a result of toxic effects of metabolites on microbial growth and enzyme activity [65].

Simultaneous Biofiltration of Multiple VOCs with Different Hydrophobicities

For simultaneous biodegradation of multicomponent VOC mixtures with different hydrophobicities, the removal of hydrophobic VOCs is not always inhibited by the hydrophilic substrates, and the presence of some hydrophilic substrates can stimulate the removal of reluctant substrates in biofilters. The presence of hydrophilic methanol had a positive effect on biofiltration for dimethyl sulfide (DMS) removal [66]. Nevertheless, the biodegradation of hydrophobic VOCs is sensitive to changes in the concentrations of hydrophilic substrates, and synergistic interactions are generally observed with low concentrations of hydrophilic substrates. Dixit and colleagues [67] observed a synergistic effect on the assimilation of toluene with a low concentration of n-propanol in the biofilters, but an antagonistic effect at high concentration. Zehraoui and colleagues [68] found that a low concentration of methanol could promote the growth of microbial populations and the accumulation of biofilms within biofilters, and consequently make n-hexane more accessible to the microbial population. However, high concentrations of methanol led to inhibition of n-hexane removal because methanol acted as the dominant carbon source [68]. In addition, methanol was observed to improve TCE removal because cometabolism helped TCE to become more available to microorganisms, while competitive inhibition was also found at higher methanol concentrations [59]. High concentrations of chlorinated compounds also led to a reduction in cosubstrate degradation because of the accumulation of toxic metabolites [60].

Even though synergistic effects on the removal of some reluctant target substrates are only observed under specific operational conditions, synergism has significant potential for reluctant VOC removal with biofiltration. Adding hydrophilic VOCs into a biofiltration system is regarded as one significant strategy to enhance the bioavailability of hydrophobic substrates [7,10,69]. In addition, synergism, especially cometabolism, allows reluctant compounds such as TCE and xylenes to be treated by biofiltration [59,60]. Most importantly, synergistic interactions in a biofiltration system may reduce operational costs while improving biofilter performance. Synergism of reluctant target pollutants and cosubstrates has been widely applied in the bioremediation of soil and groundwater [70–72]. Hypothetically, it could be also used to enhance biofilter performance for multiple VOCs.

Mechanisms for Different Types of Interactions

The removal performance of target pollutants in biofilters depends principally on their bioavailability and biodegradability (Box 2) [7,12]. Changes in the removal rate of one compound in the presence of other cosubstrates can be attributed to consequent changes in bioavailability and biodegradability. When multiple VOCs are simultaneously fed to a biofilter, the following aspects are considered to lead to substrate interactions (Figure 1, Key Figure).

Characteristics of Target Pollutants

The characteristics of the target pollutants in waste gases, including the hydrophobicity of the substrates, their toxicity to microorganisms, and chemical structure, are significant factors affecting biofilter performance and substrate interactions. Compounds with lower H constants are more readily soluble in liquid phase, and higher concentrations in the liquid phase lead to higher availability to the microorganisms, while the opposite is true for compounds with higher H constants [73,74]. Microorganisms then consume these substrates in the liquid phase as carbon sources. Competitive effects for microorganisms may occur among target pollutants in the liquid phase, and these are related to the hydrophobicity and chemical structure of the substrates. Generally, compounds with higher hydrophilicity or more active groups are preferentially utilized by microbial populations, which may result in competitive or non-competitive inhibition in the removal of reluctant compounds. However, the presence of reluctant

compounds may or may not affect the biodegradation process of hydrophilic substrates. Some studies have observed that hydrophilic compounds can act as solvents to enhance the transfer rates of hydrophobic compound from the gas to the liquid [7]. Unfortunately, only a few investigations have demonstrated this effect. Furthermore, some substrates or their metabolites may be poisonous to microorganisms, which decreases the biodegradation rates of cosubstrates [60,66]. Generally, more reluctant compounds withstand adverse effects on degradation, while some preferred substrates may escape the adverse effects [65,75].

Substrate Interactions in the Microbial Population and Metabolic Processes

Effective co-elimination of multiple VOCs in air biofiltration systems is based on the presence of active microorganisms and biofilms [69]. Generally, mixed cultures are inoculated into biofilters for simultaneous treatment of multiple VOCs [7]. Different substrates exhibit different inhibitory effects on different microorganisms [76]. In addition, specific single species and microbial communities display differences in both microbial growth and substrate utilization rates [13]. Therefore, mixed cultures exhibit greater diversity and versatility in simultaneously treating VOC mixtures in biofilters [37,50]. Furthermore, a large number of strains in a biofilter can offer various pathways for degradation of different substrates, consequently increasing the stability of microbial population and contributing to a better response to changes in environmental conditions [77,78]. However, compared to the biodegradation of single substrates, the microbial composition, microbial interactions, dynamics, and abundance of microorganisms may be altered by introducing other substrates into the biofilter, leading to antagonistic, synergistic, or neutral interactions among target pollutants [41]. Changes in a microbial community usually reflect the characteristics of the compounds in the mixtures [43,79]. For example, a microbial community in a biofilm may experience enormous changes in the presence or absence of


Box 2. Mechanisms of Biofiltration

A biofilter is a bioreactor fed with packing media where the microorganisms grow in suspension or attached [99]. Pollutants cannot be treated directly in the gas phase, and the substrates must first be transferred to the liquid phase [100]. Therefore, biofiltration for VOCs is a complex process that includes diverse physicochemical and biological interactions. Figure I describes the removal processes for VOCs in a biofilter. Although the process mechanisms for VOC biodegradation are complex, they can be grouped into the gas–liquid transfer process, physical and chemical processes in the liquid phase and biofilm, and biological processes.

Waste gases including contaminants and oxygen are first dissolved in the aqueous phase containing the microorganisms [7]. Generally, the mass-transfer rates of the substrates in this process depend on their H values, where compounds with lower H values have higher mass-transfer rates [3,13,18]. When the contaminants and oxygen are transferred into the aqueous phase, physicochemical processes such as absorption and adsorption decrease the concentration of pollutants in the gas phase and substantially increase the concentration in the liquid phase [15,18]. The compounds may or may not then be available to the microorganisms. Once the substrates are made bioavailable to microorganisms, the target pollutants are utilized as carbon sources for microbial growth and maintaining biological activity, and the compounds are simultaneously completely or partially mineralized [7,15,101]. Metabolites are created in the process of VOC biodegradation, and they also undergo physicochemical and biological processing [7].

The microorganisms present in biofiltration systems are the biocatalysts in the treatment process for VOCs [32,81]. Both pure and mixed cultures have been used for gas treatment. However, mixed cultures enriched from activated sludge or organic packing media such as compost or peat are more commonly applied in biofilters [10,24,102–104]. Bacteria, fungi, yeasts, algae, and protozoa compose a complex ecosystem, and the complexity of the microbial population ensures the stability of the biofiltration system [7,50]. Generally, the microbial composition, biokinetics, richness, and microbial interactions are most important for biofilter performance [77,105].

Different species exhibit different affinities for different target pollutants [7]. A biofilter inoculated with a pure culture can better remove some reluctant VOCs [33,97]. Microbial growth and metabolism depend on suitable environmental conditions. Therefore, biofilter performance is also related to the nature of the inlet waste gases, the type of packing material, bed temperature, moisture content, and pH [10,13].
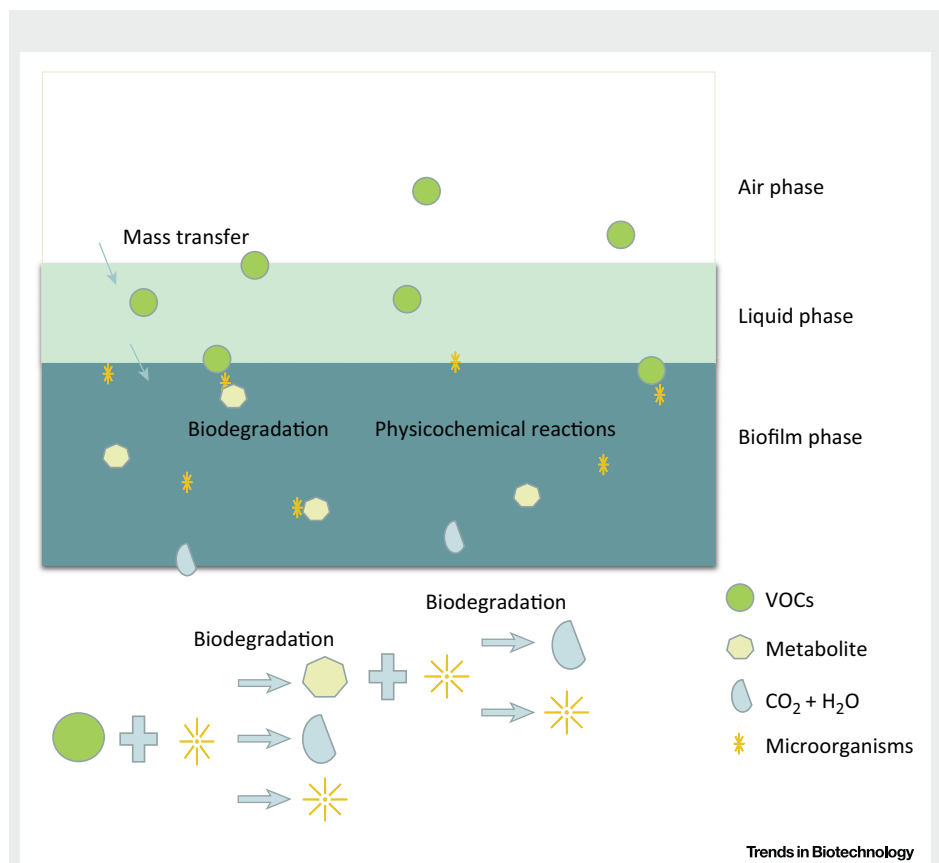
Figure I. Biofiltration Processes in VOC Removal. There are three phases – gas, water, and biofilm – in a biofilter. A VOC is transferred from the gas phase to the liquid phase and then to the biofilm phase. Complex physicochemical processes and substrate biodegradation takes place in the biofilm. The target pollutant is utilized as a carbon source for microbial growth and metabolized; the resulting metabolites also undergo physicochemical and biological processing.

desirable compounds because microbes that degrade the most desirable substrates are the most dominant population in biofilters [80]. This phenomenon may result in competitive or non-competitive inhibition interactions in the removal of cosubstrates, especially the reluctant VOCs [19,67]. By contrast, the preferential biodegradation of some hydrophilic compounds may promote biomass growth and accumulation to increase the bioavailability of reluctant compounds [66–68]. By contrast, a change in reluctant substrates may not significantly alter the microbial community because of the lower number of biodegraders.

Target pollutants are transformed into biomass and b-products through metabolic processes [7,13,81]. Some compounds can induce or facilitate the production of microbial enzymes catalyzing substrate biodegradation, which leads to positive effects [29]. In addition, some compounds such as xylenes and chlorinated hydrocarbons are negligibly utilized by microorganisms because they lack appropriate oxidizing enzymes [60,64], but these molecules can be cometabolized by introducing a cometabolite that induces the microorganisms to secrete nonspecific oxygenase systems that catalyze the oxidation of the substrates [54,55]. However, substrates and co-substrates must compete for metabolism by these enzymes [60], and the presence of preferred substrates can repress the secretion of inducible enzymes that are

Key Figure

Substrate Interactions and Their Possible Mechanisms
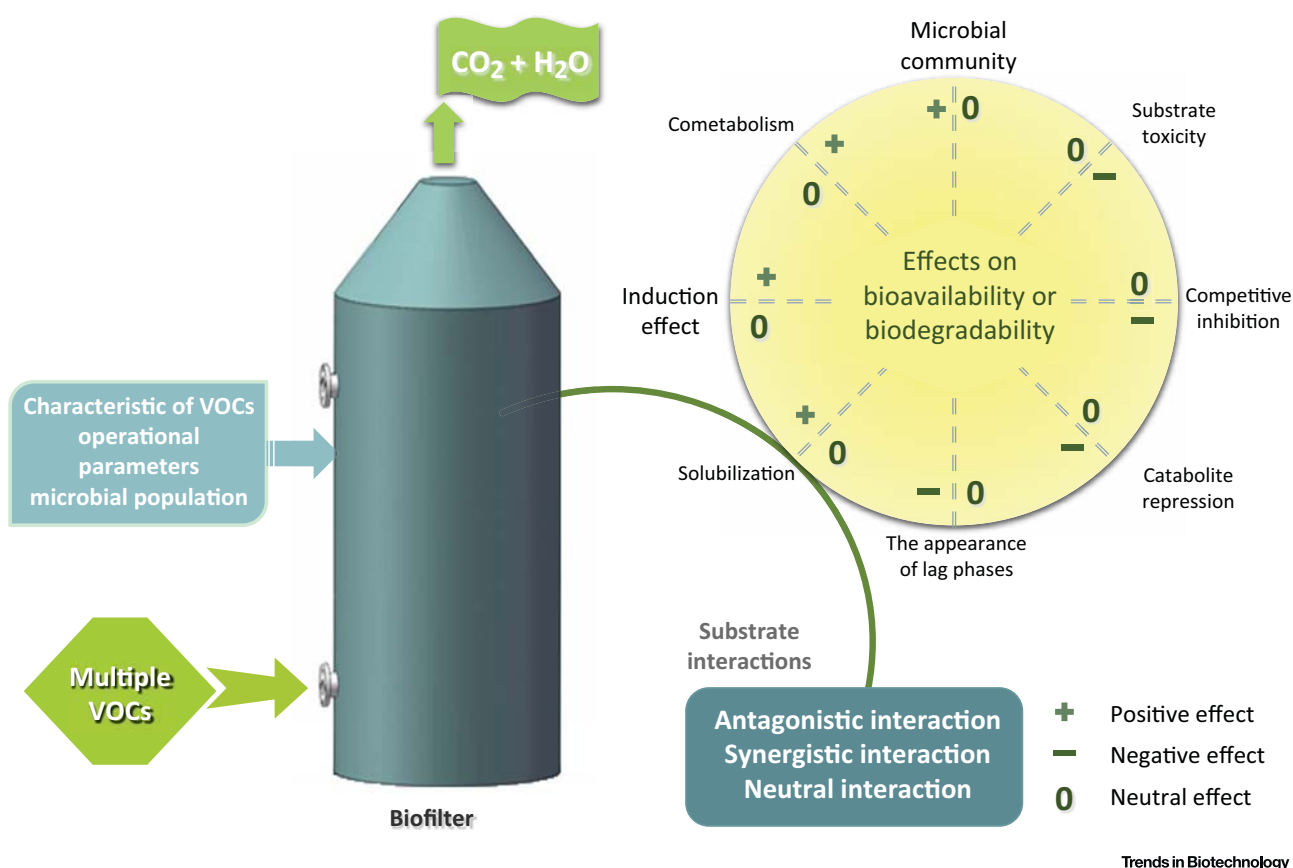
Figure 1. When multiple volatile organic compounds (VOCs) are simultaneously fed to a biofilter, the performance of the biofilter mainly depends on the characteristics of the target pollutants, the microbial population, and the operational parameters. Interactions among multicomponent VOC mixtures during biofiltration can alter the biodegradation rate of an individual compound synergistically, neutrally, or antagonistically. The bioavailability and biodegradability of one compound may be affected negatively, positively, or not at all by toxicity to the microorganisms, competitive inhibition, catabolite repression, the appearance of a lag phase, and the microbial population, as well as by solubilization, cometabolism, or induction effects. Synergistic neutral or antagonistic interactions on the removal performance can be attributed to the consequent change in the bioavailability and biodegradability of target pollutants.

needed to initiate the degradation of other substrates, causing longer lag phases [29]. In addition, catabolite repression resulting in antagonistic interactions should not be ignored [17,29]; some metabolites of prior substrates may also act as catabolite repressors in the biodegradation of cosubstrates [34]. Unfortunately, little information evaluating substrate interactions between the metabolic pathways and enzyme activity is available, except for BTEX and mixtures of TCE and cosubstrates.

Mutual effects on metabolic processes are complex, and positive and negative effects may be found simultaneously [57,82], strongly depending on the microbial population, the target

pollutants, and the operational conditions. Notably, Vergara-Fernández and colleagues found that the benzo[α]pyrene, toluene, and formaldehyde were independently metabolized [21]. Marek and colleagues observed a similar phenomenon [83]. Therefore, there may be no correlation among the metabolic processes of some target pollutants.

Potential Strategies for Process Optimization
Two-Stage Biofilter

For VOC mixtures where the removal of different components is antagonistically affected during biofiltration, combining two biological reactors, such as biofilter–biofilter, biotrickling filter–biofilter, and bioscrubber–biofilter systems, is one efficient method to alleviate antagonistic substrate interactions [14,84,85]. If the different pollutant metabolic processes can be separated, this can decrease antagonistic interactions by mitigating the toxicity of target pollutants, catabolic repression, and the appearance of lag phases because of preferential biodegradation. For example, when mixtures of VOCs with different hydrophobicities are biodegraded in a two-stage reactor, most hydrophilic compounds are assimilated in the first stage, and the highest elimination capacity for the hydrophobic substrates is achieved in a second stage [86]. There are unique configurations, modes of operation, and reaction conditions for each reactor, which can provide optimal conditions for the degradation of different target pollutants [87–89]. Therefore, it is necessary to select an appropriate type and order of bioreactors and operational parameters according the composition of the mixed gases so as to biodegrade every component effectively [87].

Two-Phase Biofilter

Two-phase biofilters are frequently considered as innovative biofilters to enhance the removal of hydrophobic VOCs by enhancing gas–biofilm mass-transfer rates [12]. Therefore, for simultaneous biofiltration of VOC mixtures with hydrophobic compounds, a two-phase biofilter might be the best option to mitigate antagonism by enhancing bioavailability. Lebrero and colleagues found that a two-phase system could better adjust to differing inlet concentrations, and improved the removal performance of hydrophobic substrates during simultaneous biofiltration of multiple VOCs [90]. Volckaert and colleagues reported that the presence of toluene and DMS did not significantly inhibit the biodegradation of toluene in two-phase biotrickling filters [91].

Table 1. Substrate Interactions and Removal Performance of Different Microorganisms

Target pollutants	Microorganism	Removal performance	Substrate interactions	Refs
Benzene, toluene	Mixed culture	Toluene > benzene	Antagonistic effect on toluene degradation in the presence of benzene	[30]
BT, BE, and BX	Pure culture	Not determined	Synergistic effect on benzene removal in the presence of toluene and xylene Antagonistic effect in the presence of ethylbenzene	[57]
Benzene, xylene	Mixed culture	Benzene > xylene	Antagonistic effect on benzene removal in the presence of xylene	[28]
BTEX	Mixed culture	Toluene > ethylbenzene > benzene > xylene	Synergistic effect on BEX removal Antagonistic effect on toluene degradation	[19]
BTEX	Pure culture	Toluene > ethylbenzene > xylene > benzene	Cometabolism and competitive inhibition among BTE	[55]
Acetone, styrene	Mixed culture	Acetone > styrene	Antagonism on styrene removal, neutral effect on acetone removal	[40]
Acetone, styrene	Pure culture	Styrene < acetone	Antagonism on acetone removal, neutral effect on styrene removal	[95]

Biofiltration with Pretreatment

Pretreatment such as UV oxidation or dielectric barrier discharge (DBD) can enhance the removal of recalcitrant contaminants in biofiltration systems by breaking down the compounds to more easily soluble and biodegradable compounds [92,93]. Combining these techniques with biofiltration may be beneficial in removing VOC mixtures containing recalcitrant contaminants, such as aromatic compounds and chlorinated compounds, and may be the most efficient strategy in the future. Moussavi and colleagues [94] reported that a UV-biofilter system could not only accelerate the treatment of toluene and o-xylene but also relieve the inhibitory effects. Recently, Jiang and colleagues [23] used a DBD-biofilter system to degrade mixtures of styrene and o-xylene, and the biodegradation rates of the both compounds were improved.

Alternative Use of Preferred Compounds

For the biofiltration of VOC mixtures that can be affected synergistically, the alternative use of preferred compounds may be a promising strategy to alleviate negative interactions and strengthen positive interactions. Methanol is one potential substrate that can stimulate the removal of reluctant VOCs. For example, Hayes and colleagues found that the alternative use of methanol could enhance removal rates of DMS [16]. The use of methanol was also demonstrated to be an alternative means to enhance n-hexane removal [95,96] and to alleviate the inhibition of benzene on n-hexane removal [96]. The introduction of methanol can stimulate the growth of specialized species that can assimilate the both compounds, and the abundance of these species will be increased after the addition of methanol [16,79]. This reflects a new approach to changing microbial populations to alleviate negative interactions by regulating operational parameters.

Application of Specialized Microbes

Even though mixed communities are more suitable for inoculation into biofilters because of their higher diversity and versatility to simultaneously treat mixed VOCs, some reluctant compounds have poor removal performance when fed together with hydrophilic substrates. Generally, one microbial strain has different affinities for different target pollutants [7], but some specialized microbes that can digest multiple pollutants may have a higher overall removal performance [80]. For example, the fungus *Sporothrix variecibatus* was inoculated into a biofilter to simultaneously treat styrene and acetone, and the removal rates for the both were satisfactory. Notably, the hydrophobic styrene was preferentially utilized and had an antagonistic effect on acetone processing, a totally different behavior from that observed in mixed culture [97]. Other examples of similar phenomena are listed in Table 1. Unfortunately, a biofilter inoculated with a pure culture cannot properly respond to fluctuating waste-gas inputs, and consequently cannot be used industrially. To overcome this limitation, it is possible that two or more specialized strains that can effectively assimilate the target pollutants might be inoculated into the same biofilter.

Concluding Remarks and Future Perspectives

We have reviewed the simultaneous biodegradation of multiple VOCs in a biofilter, and have provided an overview of removal performance for target pollutants and their interactions. Different VOCs can display antagonistic, synergistic, and neutral interactions. Antagonism in the biodegradation of more reluctant substrates makes it more difficult to achieve simultaneous efficient removal of mixed VOCs in gas biofilters as a result of toxicity, competition among microorganisms, catabolic repression, lag phases, and substrate competition for enzymes. Occasionally, low concentrations of some compounds can improve the removal of reluctant VOCs by solubilization, by increasing the abundance of some species, the induction effect, and/or by cometabolism, even though antagonism takes place at high concentration

Outstanding Questions

Are the metabolic pathways for various types of VOCs independent, and how do substrates interact with different metabolic pathway and enzyme systems?

How can the mechanisms underlying substrate interactions best be exploited for simultaneous removal of multiple VOCs in a biofilter?

Can we screen, identify, and utilize the dominant species or microbial populations for enhanced simultaneous biodegradation of some types of multiple VOCs?

How can we effectively utilize the synergistic effect to enhance the removal performance of hydrophobic VOCs?

because of competition. Interactions in removal performance are usually attributed to the general bioavailability and biodegradability of target pollutants, which in turn depend on the characteristics of the compounds, the microbial population, and the operational parameters. Unfortunately, knowledge regarding mutual effects on the metabolic process is very limited, and further systematic investigations will be necessary to better understand substrate interaction mechanisms (see Outstanding Questions). Strategies to alleviate negative interactions among multiple VOCs will make it possible to apply biofilters to multiple VOC removal at full scale. These potential strategies should also take into account how substrate interactions affect biofilter performance, but, unfortunately, the information available so far is insufficient to provide significant insights that can be applied industrially.

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