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Engineering green wall botanical biofiltration to abate indoor volatile organic compounds: A review on mechanisms, phyllosphere bioaugmentation, and modeling

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ABSTRACT

Indoor air pollution affects the global population, especially in developed countries where people spend around 90% of their time indoors. The recent pandemic exacerbated the exposure by relying on indoor spaces and a teleworking lifestyle. VOCs are a group of indoor air pollutants with harmful effects on human health at low concentrations. It is widespread that plants can remove indoor VOCs. To this day, research has combined principles of phytoremediation, biofiltration, and bioremediation into a holistic and sustainable technology called botanical biofiltration. Overall, it is sustained that its main advantage is the capacity to break down and biodegrade pollutants using low energy input. This differs from traditional systems that transfer VOCs to another phase. Furthermore, it offers additional benefits like decreased indoor air health costs, enhanced work productivity, and well-being. However, many disparities exist within the field regarding the role of plants, substrate, and phyllosphere bacteria. Yet their role has been theorized; its stability is poorly known for an engineering approach. Previous research has not addressed the bioaugmentation of the phyllosphere to increase the performance, which could boost the system. Moreover, most experiments have studied passive potted plant systems at a lab scale using small chambers, making it difficult to extrapolate findings into tangible parameters to engineer the technology. Active systems are believed to be more efficient yet require more maintenance and knowledge expertise; besides, the impact of the active flow on the long term is not fully understood. Besides, modeling the system has been oversimplified, limiting the understanding and optimization. This review sheds light on the field's gains and gaps, like concepts, experiments, and modeling. We believe that embracing a multidisciplinary approach encompassing experiments, multiphysics modeling, microbial community analysis, and coworking with the indoor air sector will enable the optimization of the technology and facilitate its adoption.

Keywords: bioaugmentation, bioremediation, botanical biofiltration, green wall, multiphysics modeling, volatile organic compounds

Contents

1. INTRODUCTION

1.1 Indoor air pollution

The World Health Organization considers air pollution the most critical environmental threat to public health. In 2019, almost all of the global population (99%) breathed air that exceeded WHO guidelines, especially in low- and middle-income countries. Historically, indoor air pollution has received less attention when compared to ambient air quality, particularly in highly industrialized areas (González-Martín *et al*., 2021). The rapid increase of urbanization impacts outdoor and indoor air pollution levels. It is estimated that the levels of indoor air pollutants could be at least twice as high as the outdoor environment (Kumar *et al*., 2023). Notably, a shift in attention has occurred in recent years since the impacts of indoor air pollution have become more noticeable. This is explained by the time spent indoors by contemporary societies (around 90%), which directly increases exposure to indoor pollution, i.e., approximately 22 hours each day in industrialized countries (González-Martín *et al*., 2021). Furthermore, 3.2 million deaths were reported in 2020 due to indoor air pollution, including ischemic heart disease, stroke, and lower respiratory infection (World Health Organization, 2022).

In developed countries, the increase in building sealing to save heating and cooling energy costs and maintain thermal comfort as a priority linked to the reliance on mechanical ventilation contributes to the accumulation of indoor air pollutants. Moreover, the energy used for mechanical ventilation systems indoors will increase (Son, Elkamhawy and Jang, 2022). The accumulation of indoor air pollutants is associated with sick building syndrome (SBS), under which occupants describe discomfort in indoor settings. People can experience acute health-related effects linked to the time spent in buildings, but no specific illness is identified. Signs and symptoms include headache, dizziness, nausea, dry cough, itching skin, fatigue, sensitivity to odors, and difficulty concentrating (Joshi, 2008). On the other hand, 2.4 billion people in less developed countries use firewood and solid fuels in inefficient combustion technologies, impacting primarily women's and children's health (World Health Organization, 2022).

Indoor air pollution also has economic impacts; for instance, in Europe, US$1.6 trillion is associated with air pollution, and it is estimated that 15% of the reduction in work productivity is due to the same cause (World Health Organization, 2021). A study in France reported that €20 billion in annual costs correspond to indoor air pollution in terms of mortality, loss of productivity, illnesses, and sick leave (Boulanger *et al*., 2017).

Indoor air pollution is the result of both indoor and outdoor sources. Indoor sources include different compounds such as dust, biological agents, and chemical compounds that originate from firewood burning, occupant respiration, and a wide range of products that produce volatile organic compounds (VOCs) (Table 1). Outdoor air brought indoors can be likewise contaminated and contribute to the existing indoor sources. The WHO published specific guidelines to control indoor air pollutants; nevertheless, only pollutants whose effects are known were selected (World Health Organization, 2010). The most researched indoor air pollutants include volatile organic compounds (VOCs), carbon dioxide (CO2), and particulate matter (PM). The primary pollutants addressed in this review are VOCs (Figure 1), which are considered the most hazardous in indoor spaces due to the variety of sources and the effects they cause on human and environmental health.

157 *Table 1. Chemical characteristics of priority indoor air pollutants. MW: molecular weight; WS: water solubility; H: Henry's constant; VP: vapor pressure; CM: complex mixture. Values retrieved*
158 *from Joint Research Centre (2005), Hoge Gezondheidsraad (2017) and PubChem (2022).*

Group	Compound	Formula	MW ($g\ mol^{-1}$)	WS ($mg\ L^{-1}$ at 25 °C)	H ($mol\ m^{-3}\ Pa^{-1}$)	VP (mmHg at 25 °C)	Limit value Indoors - WHO ($\mu g\ m^{-3}$)
Particles	PM < 10 µm	PM_{10}	CM	CM	CM	CM	50 (24-h)
	PM < 2.5 µm	$PM_{2.5}$	CM	CM	CM	CM	25 (24-h)
Non-VOCs	Carbon monoxide	CO	28.01	27.6	9.70E-6	1.55E+8	7000 (24-h)
	Carbon dioxide	CO_2	44.01	1450	3.30E-4	4.83E+4	1000 ppm
	Nitrogen dioxide	NO_2	46.0	Reacts	9.90E-5	9.00E+2	200 (1-h)
	Sulfur dioxide	SO_2	64.0	1.07E+5	1.30E-2	3.00E+3	0.048 ppm (24-h)
	Ozone	O_3	48	0.57 at 20 °C	1.00E-4	4.13E+4 (10.4 ˚F)	120 (8-h)
VOCs (BTEX)	Benzene	C_6H_6	78.1	1.79E+3	1.70E-3	9.48E+1	<1
	Toluene	C_7H_8	92.1	5.62E+2	1.50E-3	2.84E+1	260
	Ethylbenzene	C_8H_{10}	106.2	1.69E+2	1.40E-3	9.60E+0	260
	o-xylene	C_8H_{10}	106.2	1.78E+2	2.40E-3	6.65E+0	200
	m-xylene	C_8H_{10}	106.2	1.61E+2	1.40E-3	8.29E+0	200
	p-xylene	C_8H_{10}	106.2	1.62E+2	1.90E-3	8.84E+0	200
VOCs	Formaldehyde	CH_2O	30.0	1.98E+5	3.20E+1	3.89E+3	100 ppm (30-min)

159

1.2 Volatile organic compounds in the indoor environment

Indoor VOCs are a pollutant group of concern, given the health effects that can result from exposure to low concentrations during short and long periods. They are emitted by building materials, furniture, textiles, and cleaning products (Figure 1). Besides, they are the hardest to remove with conventional methods (section 1.4). For instance, light and highly volatile VOCs like acetaldehyde, formaldehyde, and dichloromethane were reported not to be effectively removed by activated carbon filters. VOCs are generally described as organic chemicals with a low boiling point and high vapor pressure at room temperature (20 ˚C, 100 kPa). The European Union defines VOC as any organic compound with an initial boiling point less than or equal to 250 measured at a standard pressure of 101,3 kPa. They are associated with problems in the respiratory, nervous, and hepatic systems. VOCs such as benzene and formaldehyde are classified as carcinogenic by the International Agency on Cancer Research (Pettit, Irga and Torpy, 2018b).

On the other hand, plants can naturally produce VOCs of less concern. These are called biogenic VOCs, such as isoprene, terpenes, and alkanes. Nevertheless, this review focuses on VOCs of anthropogenic origin, which include various compounds, such as formaldehyde, polycyclic aromatic hydrocarbons (PAHs), benzene, toluene, ethylbenzene, and xylene (BTEX). Studies on removing BTEX and formaldehyde are prioritized, given their abundance and toxicity (Wei *et al*., 2017; Fleck *et al*., 2020).

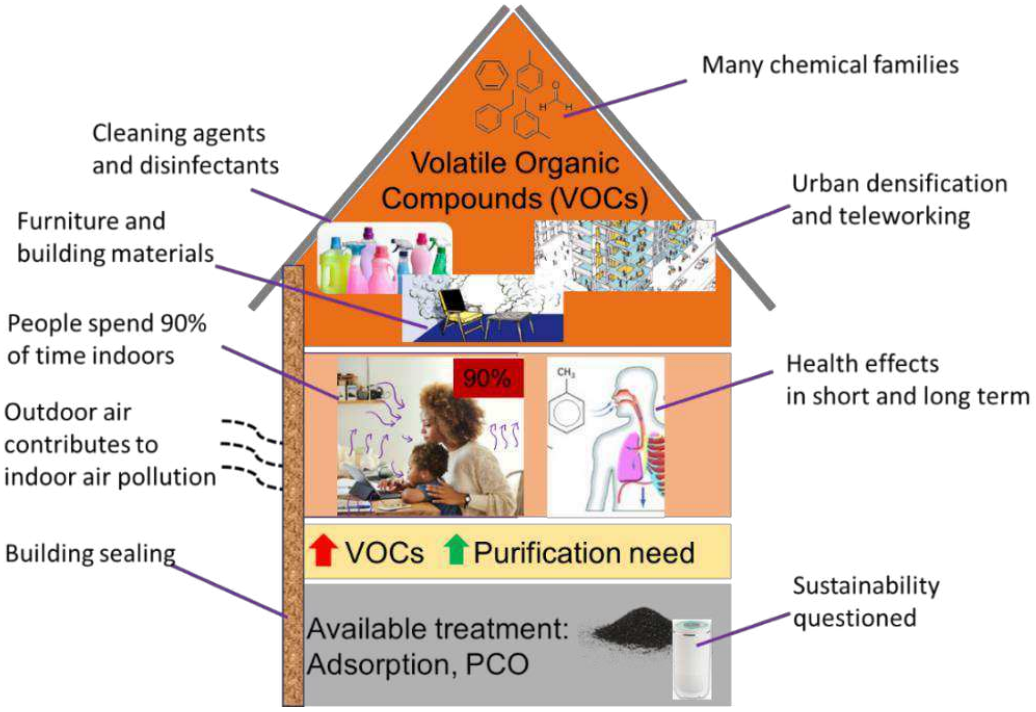

Figure 1. Indoor air pollution, VOC sources, impact, and available technologies.

1.3 Why depart from traditional indoor VOC removal technologies?

The traditional approach to counteract indoor air pollution relies on physical-chemical technologies (US EPA, 2022). Particularly, using ventilation (HVAC) systems is prevalent in buildings under the principle of dilution, i.e., replacing polluted indoor air with outdoor air, which is believed to be less polluted. Thus, by enhancing the ventilation air flow rate, HVAC systems aim to control indoor air quality (Montgomery *et al*., 2012). However, the latter does not eliminate VOCs (Mata *et al*., 2022). Additionally, depending on the location, outdoor air can be several times more contaminated than indoors.

It becomes clear that, in many cases, the indoor air must be purified in situ. Traditional technologies in this regard aim to remove particulate matter and VOCs, such as air filtration, electrostatic precipitation, activated carbon adsorption, ozonation, and photocatalytic oxidation (PCO) (González-Martín *et al*., 2021; Mata *et al*., 2022; Szczotko *et al*., 2022). Mechanical filtration involves using fibrous materials through which air is forced, and particulate matter is retained. Electrostatic precipitation treats the particulate matter as negatively charged particles attracted to a plate with different charges in which they are collected.

On the other hand, adsorption deals with capturing gaseous pollutants like VOCs on the surface of adsorbent material. On the contrary, ozonation treats VOCs by producing ozone from molecular oxygen, reacting with the target compounds. In photocatalytic oxidation, semiconductors are employed to create radicals that then react with the pollutants. However, these technologies mentioned have been criticized as they rely on a single removal principle, often do not degrade VOCs, and need an energy input (Irga, Pettit and Torpy, 2018; Pettit, Irga and Torpy, 2018b). An ideal alternative technique must treat and degrade a wide range of indoor air VOCs with a low environmental impact (González-Martín *et al*., 2021). Biobased technologies have gained interest in the last few years due to their versatility and low environmental impact. Nevertheless, they face criticism due to the complexity of biological mechanisms, which impacts the stability of the air purification process (Table 2).

210 *Table 2. Comparison among indoor VOC removal technologies. Adapted from Luengas et al. (2015); González-Martín et al. (2021); Mata et al. (2022); Szczotko et al. (2022) and US EPA (2023).*

Indoor air purification technique	**HEPA Filter** 	**Photocatalytic oxidation (PCO)** 	**Active carbon filtration** 	**Ozonation** 	**Botanical biofiltration** 
Type	Mechanical	Physical-chemical	Physical-chemical	Physical-chemical	Biological
Main mechanism	Filtration	Oxidation	Adsorption	Oxidation	Biodegradation
VOC removal	No	≈ 75%	≈ 90%	Low	Less stable, according to the design and boundary conditions. Removal range: 50 – 90%
VOC mineralization	No	Yes, but incomplete	No, VOC transferred to another phase.	Yes, but incomplete	Yes, due to multiple pathways
Hazardous by-products	VOCs might resuspend.	Harmful by-products	VOCs might desorb	Harmful by-products	Intermediate compounds can be metabolized
Waste generation	Spent filters.	Catalyst, lamps	Spent adsorbent	-	Organic waste
Energy consumption	Moderate	Moderate	Moderate to high	High	Low
Additional benefits	-	-	-	-	Enhanced productivity, aesthetics, thermal comfort, noise reduction

211

212

1.4 Review objectives and methodology

This review focuses on botanical biofiltration, a bio-based indoor air purification technology that may offer desired advantages compared to the traditional VOC treatment (Table 2): simultaneous VOC removal mechanisms, VOC mineralization, less energy consumption, and additional psychological and economic benefits. Despite its benefits, there is a lack of consensus on botanical biofilters' design, operation, performance, and stability. This review first discusses the VOC-removal principles occurring in botanical biofiltration (section 2) to gain further insights into those areas with significant gaps in the field, which refer to strategies to increase the performance and stability of the technology. These are the bioaugmentation of the botanical compartment and phyllosphere (section 3), the role of the airflow (section 4), and developing a more comprehensive model to understand and optimize the technology (section 5).

The consolidated knowledge since the initial efforts in botanical biofiltration was reviewed by retrieving research and review articles from peer-reviewed journals using constraints to refine the scope of the study. They were (i) year of publication (2000-2023) and (ii) matching one of the following terms: "botanical biofiltration," "botanical biofilter," "active botanical biofilter, "green wall biofilter," "indoor air phytoremediation," "indoor air phylloremediation." For this purpose, the Web of Science engine was employed to retrieve articles from different journals in varied disciplines such as "Applied and Environmental Microbiology," "Air Quality, Atmosphere and Health," "Atmospheric Pollution Research," "Building and Environment," "Chemosphere" and "Journal of Hazardous Materials." As a result, the database contained 70 articles, most of which are original research articles. Also, 61% of the articles retrieved were published during the last seven years (2017 - 2023) out of the 23 years selected (2000 – 2023).

2. BOTANICAL BIOFILTRATION PRINCIPLES

2.1 Mechanisms involved in botanical biofiltration

Bio-based indoor air purification systems like botanical biofiltration and green walls have received attention from built environment professionals. These systems aim to remove indoor air pollutants more sustainably and aesthetically (Llewellyn and Dixon, 2011; Kim *et al*., 2018a). Botanical biofiltration is a technique that employs the action of plants, microbes, and substrates to remove VOCs and other compounds like PM and CO2. It is believed to be a sustainable, cost-effective, energy-efficient, and friendly air purification technology (Ronald A. Wood *et al*., 2006). Besides, it is claimed that botanical biofiltration contributes to thermal comfort and contributing to aesthetics and psychological effects, like enhanced productivity (Llewellyn and Dixon, 2011; Pettit, Irga and Torpy, 2018b). Botanical biofilters can be considered successors of the biofiltration technology employed in industrial VOC treatment, especially by the odor-generating industries (Figure 2) (Kennes, Rene and Veiga, 2009). A traditional biofilter consists of a unit loaded with packing material on the surface of which a biofilm develops, i.e., aggregation of microorganisms (Kennes, Rene and Veiga, 2009). The same biofilm is thought to develop in the substrate of a botanical biofilter.

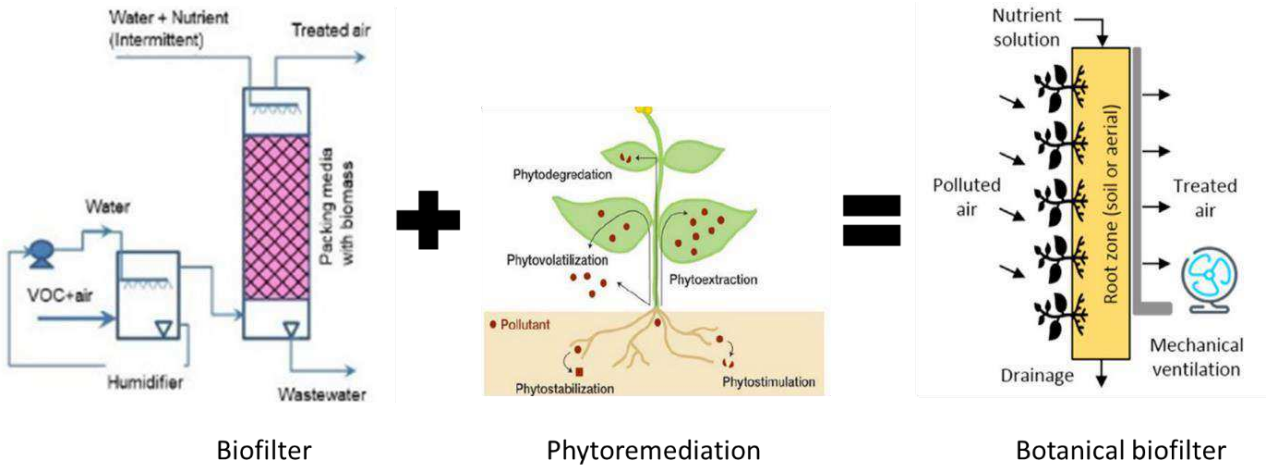

Figure 2. A traditional biofilter configuration is a base onto which a botanical compartment is added to conform a botanical biofilter. The botanical biofilter shown was conceived as a plant-assisted biotrickling biofilter by González-Martín et al. (2021).

Commonly, botanical biofilters are vertical structures where plant species are grown on soil or another medium; they are known by other names such as “phytoremediation systems,” “living walls,” and “active living walls” (Mikkonen *et al*., 2018). The systems tend to be designed vertically to maximize the surface area, translating into higher removal (Kumar *et al*., 2023).

However, assessing which components of a biological system are responsible for pollutant removal is complex since multiple mechanisms occur simultaneously. Very little knowledge has

been gained regarding microbial community changes when degrading indoor air pollutants, particularly VOCs (Irga, Pettit and Torpy, 2018). Moreover, further investigations are needed to comprehend the long-term performance in situ instead of controlled laboratory experiments. To engineer and increase the performance of botanical biofiltration, it is essential first to understand the removal mechanisms, namely, bioremediation, phytoremediation, and substrate remediation, as explained below (Irga, Pettit and Torpy, 2018).

2.2 Bioremediation role

Bioremediation involves the action of plants, substrates, and microorganisms to transform, capture, or biodegrade contaminants, i.e., indoor air pollutants. This review uses the term bioremediation to refer to bacterial degradation. Living organisms use the compounds as an energy or carbon source (Shah, 2020). Pollutants are transformed into less toxic or nontoxic substances. Consequently, bioremediation systems have been widely used to treat contaminated soil, water bodies, and air (Wei *et al*., 2017; Kumar Rahul and Kundu, 2020; Landa-Acuña *et al*., 2020). One of the advantages of employing bioremediation systems is that microbes as heterotrophs are present almost everywhere, including plants and shoots, and they have evolved to obtain energy from almost every compound (Wei *et al*., 2017; González-Martín *et al*., 2021). Hence, bioremediation could be brought and exploited indoors with botanical biofiltration technology. Notwithstanding, bioremediation raises concerns like inoculating bacteria to express and maintain diverse microbial degrader communities (Guieysse *et al*., 2008).

Bioremediation can use indigenous or autochthonous bacterial strains as they are adapted to a certain environment or allochthonous strains isolated from other environments or previously cultured, and their biodegradation capacity has been quantified. If the latter is employed, the bioremediation process is preceded by bioaugmentation or inoculation (Pettit, Irga and Torpy, 2018b). Bacteria that have been employed to perform bioremediation applications and could have potential in botanical biofiltration include *Acinetobacter sp*., *Burkholderia cepacian*, *Deinococcus radiodurans*, *Dehalococcoides ethenogenes*, *Pseudomonas aeruginosa*, *Pseudomonas putida*, *Rhodococcus sp*., *Xanthomonas sp*. (Kennes, Rene and Veiga, 2009; Kumar Rahul and Kundu, 2020; Landa-Acuña *et al*., 2020). These bacteria possess enzymes to catalyze the degradation of pollutants via peripheral and central pathways under aerobic or anaerobic conditions (Dell' Anno *et al*., 2021). General factors limiting bioremediation are the contaminants' toxic effect, bioavailability, environmental conditions, and microorganisms' metabolic restrictions (Landa-Acuña *et al*., 2020).

2.3 Plants and Phytoremediation Role

VOC removal has been documented to differ among different plant species. Likewise, different plant cultivars have exhibited different removal values (Dela Cruz *et al*., 2014). In fact, over 120 plant species have been researched for VOC removal in pot-based passive systems (Soreanu, Dixon and Darlington, 2013). However, the results might not be directly extrapolated to active botanical biofilters due to the shorter residence time between plant leaves and the VOC.

Plant species like *Chlorophytum comosum, Chrysanthemum morifolium, Epipremnum aureum, Ficus benjamina*, and *Sansevieria trifasciata* have been documented to degrade and remove VOCs like BTEX and formaldehyde (Han *et al*., 2022; Kumar *et al*., 2023). Moya *et al*. (2019) recommended opting for plants with a large surface area, as this correlates with higher stomatal density and, theoretically, higher VOC removal. Similarly, plants that resist biotic stresses like drought, cold, shade, and indoor light are preferred (Wei *et al*., 2017). Plants with broad leaves and rough surfaces can capture more VOCs than smoother leaves (Kumar *et al*., 2023). These pollutants can then be assimilated and degraded as part of the plant's metabolism in a process known as phytodegradation (Nowak *et al*., 2006). Additionally, pollutants can be translocated to other parts of the plant (Kumar *et al*., 2023).

Some VOCs like BTX can be converted to phenol or pyrocatechol and transformed into muconic and fumaric acid (Kim *et al*., 2018a). This transformation involves participating enzymes like oxidoreductases, hydrolases, and bioconjugation reactions with sugars or other compounds. It has been suggested that VOCs might enter the Calvin cycle and undergo conversion into amino acids (Figure 3) (Irga, Pettit and Torpy, 2018). Another consideration is the generation of CO2 due to plant respiration, especially under low light conditions indoors. Treesubsuntorn and Thiravetyan (2018) addressed this by combining CAM and C3 plants in a botanical biofilter, effectively managing the CO2 emissions under low light while efficiently removing VOCs.

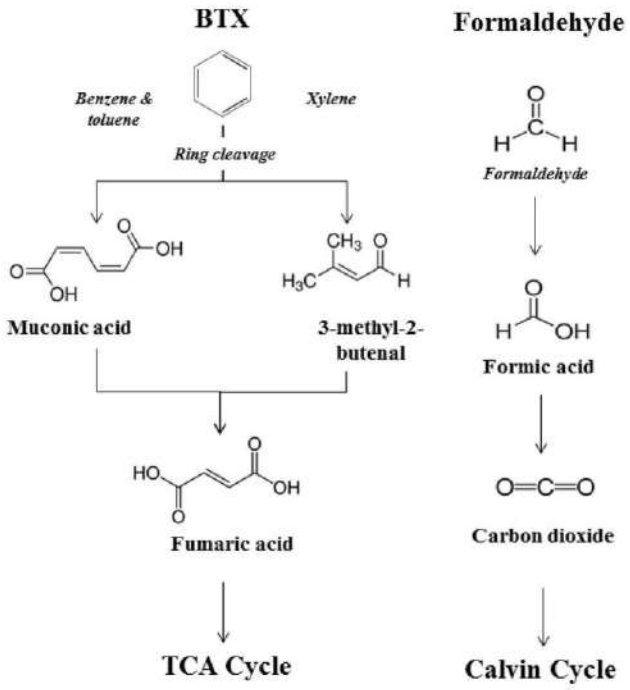

Figure 3. The plants' metabolism of benzene, toluene, xylene (BTX), and formaldehyde. The degradation of BTX begins with ring cleavage, followed by the formation of muconic acid, and further oxidation may lead to fumaric acid, a key intermediate in the Krebs Cycle. The oxidation of formaldehyde produces formic acid that is further oxidized into CO_2. This compound can enter the Calvin Cycle. Taken from Kim et al. (2018a) based on Ciese et al. (no date) and Ugrekhelidze, Korte and Kvesitadze (1997).

Leaves are the photosynthetic organs that consist of an upper surface of a waxy cover called the cuticle, whose primary function is to prevent water evaporation but also serve as a barrier for xenobiotic compounds. Therefore, the chemical structure of the cuticle has been suggested to determine the rate of VOC removal (Treesubsuntorn *et al*., 2013). This waxy cover includes long-chain alkanes, alcohols, aldehydes, ketones, acids, and esters. Therefore, lipophilic compounds of hydrocarbons are easily absorbed and accumulate, especially by younger leaves (Ugrekhelidze, Korte, and Kvesitadze, 1997). This is relevant as botanical biofiltration is intended to cope with various VOCs with different chemical properties, including hydro and lipophilic compounds.

On the other hand, stomata regulate the gas exchange, which is believed to be a primary route for air pollutants like VOCs to enter the leaf, particularly stomata located on the abaxial side would allow VOCs to enter the mesophyll region (Wei *et al*., 2017). It is theoretically assumed that this is the region where biodegradation can occur using xenobiotic metabolism (Joshi and Ghosh, 2014). In other words, the assimilation of aromatic hydrocarbons includes their uptake and transformation steps, which depend on the number of stomata and the structure of the cuticle (Ugrekhelidze, Korte and Kvesitadze, 1997). Moreover, it is sustained that the VOC uptake is

also determined by the stomatal conductance (Tani *et al*., 2007). Mesophyll cells are located underneath the epidermis and occur in palisade and spongy mesophyll cells. Palisade mesophyll cells contain the highest number of chloroplasts in which photosynthesis occurs, which is believed to affect VOC removal directly (Wei *et al*., 2017).

The experiment by Ugrekhelidze, Korte and Kvesitadze (1997) investigated the biodegradation pathways of VOCs by plants, which is momentous in the field of air phytoremediation and botanical biofiltration. The authors demonstrated the crucial role of chloroplasts in the primary process of degradation of hydrocarbons, particularly the hydroxylation of aromatic rings. Notably, VOCs such as benzene and toluene underwent oxidation due to aromatic ring cleavage, incorporating carbon atoms into various fractions of cellular organic compounds. The study introduced photosensitized oxidation occurring in leaves and chloroplasts, a process involving a more pronounced degradation under illuminated conditions. Chlorophyll II was identified as a key player in this mechanism. The authors concluded that the specific oxidizing enzymes present determine the transformation of aromatic hydrocarbons, thus leading to variations in transformation rates among different plant species. Interestingly, continuous removal was observed under dark conditions. This phenomenon was attributed to the absorption of VOCs by the cuticle. Consequently, it is proposed that a botanical biofilter could potentially absorb and transform VOCs even without a light-mediated process.

2.4 Substrate and Rhizosphere Role

A significant part of the VOC removal in botanical biofiltration is attributed to the substrate and the rhizosphere, including the microorganisms (Liu *et al*., 2022). In fact, most of the research has acknowledged the latter, often neglecting the contribution of the plant compartment (Irga, Pettit and Torpy, 2018). This might respond to the lack of studies on the phyllosphere's role in removing VOCs in a botanical biofilter.

The substrate or growing media must not be considered an isolated compartment that can be fully optimized by selecting a new one. It is a complex and interrelated environment where the supporting media, rhizosphere, and microorganisms cohabit and interact synergistically. Furthermore, plant species can affect the rhizosphere and impact the substrate's role in removing VOCs (Irga *et al*., 2019). For instance, Irga *et al*. (2019) suggested that plants could modify the substrate's properties, like hydrophobicity and hydrophilicity, thus altering the VOC's affinity to the medium's binding sites. In addition, the root volume is usually positively correlated with removal efficiency (Irga *et al*., 2019).

Growing media can affect the removal of VOC according to the substrate type, properties, the VOC physicochemical characteristics, and present microorganisms (Dela Cruz *et al*., 2014). A natural growing media is desirable in botanical biofiltration as it may provide plant nutrients and contain indigenous microbial communities. This choice is particularly advantageous if it aligns with sustainable production practices. Factors like porosity, pressure drop, and water retention capacity may affect the mass transport of the VOC into the substrate (Matheson *et al*., 2023). Nevertheless, if the substrate compartment is idealized as a traditional biofilter, the packing material properties may also apply in botanical biofiltration. These include high porosity, water retention capacity, good drainage, and mechanical stability. A high surface area is desired to develop the microbial biofilm and facilitate mass transfer from the gas to the liquid phase.

VOCs can be present in the substrate's liquid, solid, or gas phases (Masi *et al*., 2022). VOCs can firstly physically adhere to the growing media via adsorption, especially when the total residence time is short in the case of active botanical biofilters. Adsorption is thought to occur before the biological processes. In this regard, additional materials have been utilized to enhance the VOC capture in a mixture with horticultural substrates. For instance, the enhanced removal of VOC employing activated carbon as an additional material in the substrate of a botanical biofilter was studied by Wang, Pei and Zhang (2012) and Pettit, Irga and Torpy (2018a). However, this can be challenging from a sustainability point of view as the system would not be biobased, and likely, the removal is greatly due to the sorption media added, undermining the potential of the microorganisms and the botanical compartment.

Secondly, VOCs diffuse through the medium where bacteria can take them and metabolize the compounds as a nutrient or carbon source (Irga, Pettit and Torpy, 2018), as shown in Figure 4. Therefore, sufficient liquid content should be available since the mass transfer from the gas to the liquid phase is the rate-limiting step in the biological treatment of air contaminants. Adequate liquid phase ensures optimal conditions for microbial degradation. Still, irrigation should be minimized to decrease the environmental impact.

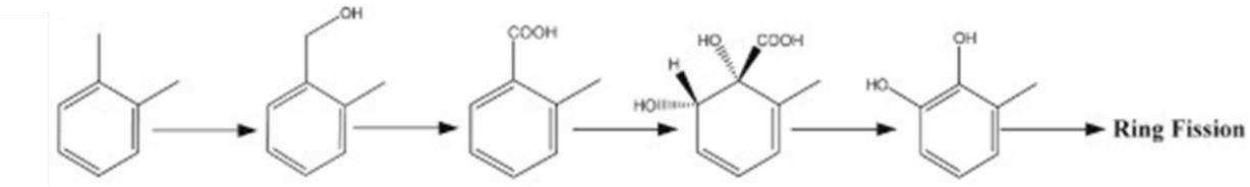

Figure 4. The proposed aerobic metabolic pathway for o-xylene degradation, one of the most recalcitrant BTEX by Rhodococcus sp. ZJUT312 (that can be found in the substrate of a botanical biofilter) involving monooxygenase enzyme. Eventually, 3-methyl catechol (the last compound) would likely be mineralized into CO_2 (You et al., 2018).

Gram-negative bacteria like *Pseudomonas sp*. have been reported to be efficient in VOC removal among microbes (Zhang *et al*., 2013). Gram-negative bacteria are believed to be more tolerant to pollutants like hydrocarbons because of the highly impermeable outer membrane (Lzroaie, 2010). A way to increase the removal rates of VOCs in the substrate compartment is by performing bioaugmentation and biostimulation, as proposed for the botanical compartment. Likewise, the external inoculum should not interfere with the existing community but assist in removing VOCs (Ronald A Wood *et al*., 2006; Pettit, Irga and Torpy, 2018b). The biostimulation of the substrate compartment with benzene increased the total abundance of soil bacteria, particularly the amount and activity of benzene-degrading bacteria (Sriprapat and Thiravetyan, 2016).

3. BIOAUGMENTATION OF THE PHYLLOSPHERE AND PLANT

In a botanical biofilter, it is tough to perfectly separate and quantify the mere contribution of the plant vs. their associated bacteria in the removal, transformation, and biodegradation of VOCs (Figure 5). The approaches taken by Kim *et al*. (2016) and Sangthong, Suksabye and Thiravetyan (2016) were isolating each compartment with Teflon bags and sterilizing plants. Nevertheless, the following sections scrutinize each sole process, emphasizing the phyllosphere's role in degrading VOC. Bioaugmenting or enriching this section can be a strategy to engineer more effective botanical biofilters, and we consider it a gap in this novel field.

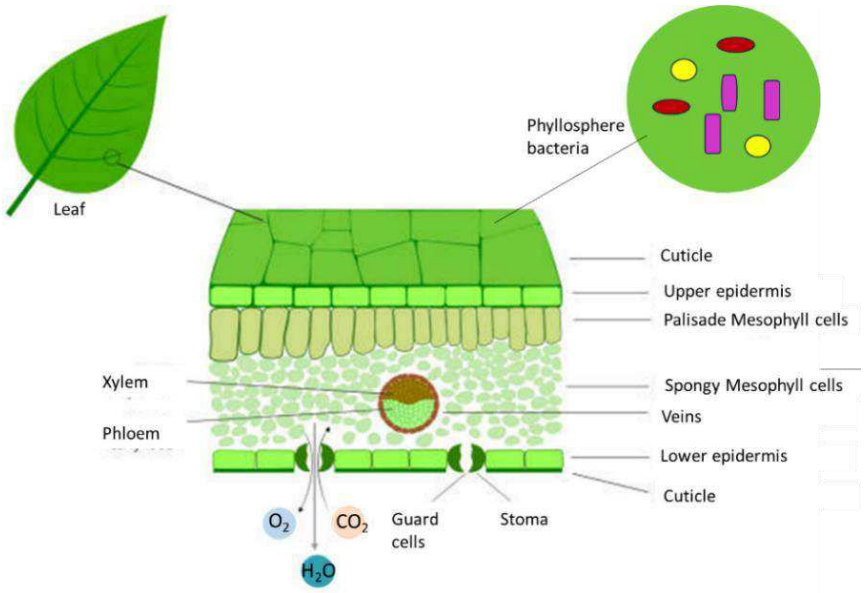

Figure 5. Illustration of the phyllosphere in a cross-section of a leaf. Phyllosphere bacteria can biodegrade volatile organic compounds. Stomata can absorb gaseous pollutants to be degraded by the plant tissue.

3.1 Phyllosphere role in VOC degradation

The process by which air pollutants are bioremediated by plants' leaves and their leaf-associated mechanisms is called phylloremediation (Figure 5) (Wei *et al*., 2017). Therefore, it is logical that phylloremediation accounts for part of the VOC removal in botanical biofiltration. Nevertheless, some studies argue that the contribution of the botanical compartment, including the phyllosphere bacteria, is very low to almost negligible (Jin Kim *et al*., 2008). For instance, Schmitz, Hilgers and Weidner (2000) concluded that using plants is unlikely to be of value for indoor air purification due to a low metabolic rate. Still, a study by Jin Kim *et al*. (2008) showed that the removal ratio or relative contribution between the botanical vs. the substrate compartment of a potted plant reached a ratio of 1:1 in removing formaldehyde during daytime conditions. The results were presumed to depend on the pollutant characteristics, biofilter operation, and design. Hence, we can infer that the relative contribution of phytoremediation to the botanical biofiltration process depends on botanical biofilter design and environmental considerations. The latter also raises the question of optimizing and increasing the removal attributed to the botanical compartment, as explained in the following section.

The phyllosphere is dominated by diverse bacterial communities shaped by plant species, characteristics, and environmental conditions (Vorholt, 2012). However, the phyllosphere is a harsh environment for microbial growth, resulting in a less dense and diverse population of microbes than the rhizosphere (Wei *et al*., 2017; Irga, Pettit and Torpy, 2018). This might be the reason behind the low expectations of their contribution to VOC removal. The composition of the phyllosphere may differ according to geographical location, reflecting the impact of climate and the bacteria deposited on the leaves (Wei *et al*., 2017). Proteobacteria such as *Methylobacterium* and *Shingomonas* have been found to dominate the phyllosphere (Vorholt, 2012). γ-Proteobacteria like *Pseudomonas* can likewise be abundant. However, different genera and strains can be found. Moreover, the complex bacteria-plant interaction makes the phyllosphere a selective environment. The latter might be explained due to adaptation and coevolutionary relationships that allow the close association between plant species and microbes (Wei *et al*., 2017).

3.2 Phyllosphere bioaugmentation for engineering the system

It has been proven that colonized leaves can biodegrade more pollutants than leaves alone (Pettit, Irga and Torpy, 2018b). Since the phyllosphere is typically a carbon-limited environment, it can make the biodegradation of organic pollutants like VOCs a great opportunity for phyllosphere bacteria (Lindow and Brandl, 2003). However, it has been

previously established that indoor plants, like greenhouse plants, usually do not have a well-established, natural, and diverse phyllosphere microbiome, probably owing to limited bacterial sources like soil (Marie *et al*., 2022). Most potted plants and plants grown in botanical biofilters are exposed to similar circumstances, and we can hypothesize they typically lack an adapted phyllosphere microbiome, diversity, and bacterial numbers. Hence, we argue that by bioaugmenting and biostimulating the phyllosphere of a botanical biofilter, the system could be optimized greatly, leading to higher VOC removal and possibly stability in the long term. Nevertheless, little research is found in bioaugmenting a botanical biofilter (Matheson *et al*., 2023), and whenever it is proposed, mainly substrate bioaugmentation is considered (Ronald A Wood *et al*., 2006; Pettit, Irga and Torpy, 2018b).

More details about the steps in bioaugmenting the plant leaves are shown in Figure 6. Furthermore, there is no consensus on the bioaugmentation of plant leaves, the substrate, or both. For instance, are indigenous bacteria preferred over allochthonous? What is the best inoculation method for the highest survivability? Moreover, what would be the method to quantify the inoculated strains over time and see if they become stable? Is inoculation more intended for passive or active botanical biofilters?

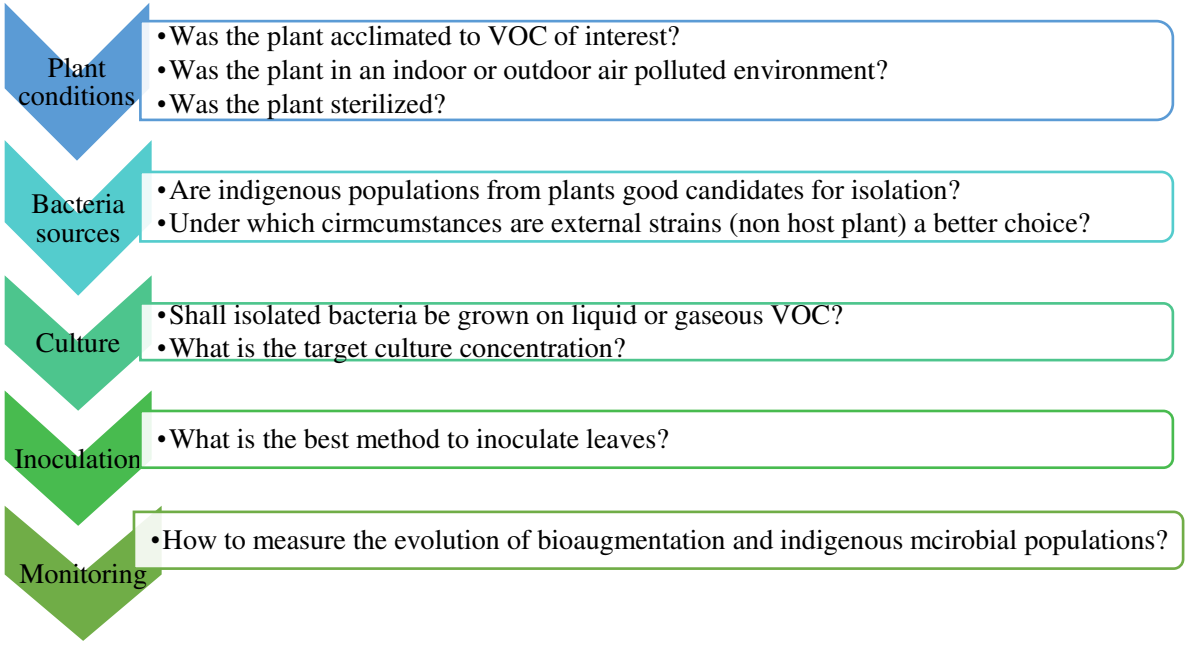

Figure 6. Steps for future research in bioaugmentation of plant leaves and factors to be considered.

The work of De Kempeneer *et al*. (2004) on the bioaugmentation of the phyllosphere for removing toluene has been alluded to in reviews of botanical biofiltration. According to the authors, this was the first study addressing the effect of adding an external inoculum. The authors inoculated *Azalea indica* with *Pseudomonas putida* TVA8 previously culture by using

bubbling reactors to which toluene was given at a rate of 20 mL min-1 with a total residence time of 4.5 days. The strain contained the toluene dioxygenase (tod) operon. Plants were inoculated by spraying the foliage with a suspension containing 107 CFU mL-1. Batch experiments were done by adding 90 ppm of acetaldehyde to a 23 L climate chamber, and the concentration was monitored until 95% of the toluene was removed. A pronounced effect on the toluene removal by bioaugmentation was confirmed against a blank of artificial plants that were likewise inoculated. Furthermore, an adaptation time was suggested as needed by the bacteria in the phyllosphere.

Contrary to the previous study, Sriprapat and Thiravetyan (2016) also performed bioaugmentation but used the plants' indigenous or originally present bacteria after exposure to benzene. Firstly, eight plant species were evaluated in a chamber by injecting 50 µL of acetaldehyde, and from this experiment, only *C. comosum* showed the highest removal. Therefore, the plant was chosen to isolate epiphytic bacteria from the leaf or root, and endophytic bacteria were to be grown in semisolid and gaseous benzene. *Enterobacter sp*. EN2, *Cronobacter* EPL1, or *Pseudomonas* EPR2 were inoculated to non-sterilized plants and placed in a clear serum bottle. The benzene mass added to each treatment was 170 µg. The results showed that the inoculated sterilized plants reduced benzene to a greater extent than the controls. However, non-sterilized plants produced a higher removal than sterilized plants.

Both studies mentioned offer significant insights into the bioaugmentation of the phyllosphere. However, they considered very high pollutant concentrations and batch experiments. Still, there are concerns about how long the microbial communities persist in the phyllosphere, especially if they are non-native to the host plant species. For instance, the study of Sriprapat and Thiravetyan (2016) showed that the *Enterobacter* EN2 strain could colonize inoculated plants. However, when a non-native microbe successfully colonizes, concern arises about what happens to the rest of the indigenous microorganisms. Moreover, bioaugmenting botanical biofilters with bacterial cultures in buildings should be safe for the plant and human health.

The experiment of Sangthong, Suksabye and Thiravetyan (2016) studied the bioaugmentation of *B. buttiana* leaves in removing xylene. The leaves were sterilized, then the culture was sprayed onto the leaves. Phyllosphere bacteria had been previously screened and isolated from the same plant species, and it was shown that the most effective bacteria in the plant-bacteria systems were *E. cloacae* LSRC11, *Staphylococcus sp.* A1 and *P. aeruginosa*. It is not clear whether bacteria were investigated in batch experiments before inoculating the plant leaves. Nevertheless, the plant-bacteria systems were evaluated under 10 ppm of xylene, which could

be considered a high VOC concentration for an indoor environment. Likewise, the authors clarified that the bacteria associated with *B. buttiana* could increase the xylene removal over a shorter period. Moreover, the sterilization procedure can be questioned as it deleted the effect on the indigenous populations.

4. ACTIVE OR PASSIVE AIRFLOW CHOICE

4.1 Can passive botanical biofilters efficiently remove VOCs?

Previous studies by NASA in the 1980s showed that potted plants could decrease the concentration of different VOCs featuring passive botanical biofilters (PBB). The latter led to growing interest in botanical biofiltration (Wolverton, Mcdonald and Watkins, 1984). These very early experiments utilized static climate chambers, in which the VOC removal relied on the passive diffusion of the pollutants into the substrate and plant compartments (Figure 7). Most of these solely evaluated the drawdown of VOCs attributed to the entire activity of both compartments (Fooladi *et al.*, 2019; Jin Kim *et al.*, 2019; Suárez-Cáceres *et al.*, 2021). However, no special attention was paid to the removal mechanisms (Wolverton, Mcdonald and Watkins, 1984). Yet experiments in climate chambers offer an estimation of the VOC removal capacity under static conditions; they have been criticized for not representing realistic conditions and have employed high pollutant concentrations (10-100 times higher than typical indoor environments and very small chamber volumes). The latter contrasts with low VOC concentrations typically found indoors (Llewellyn and Dixon, 2011).

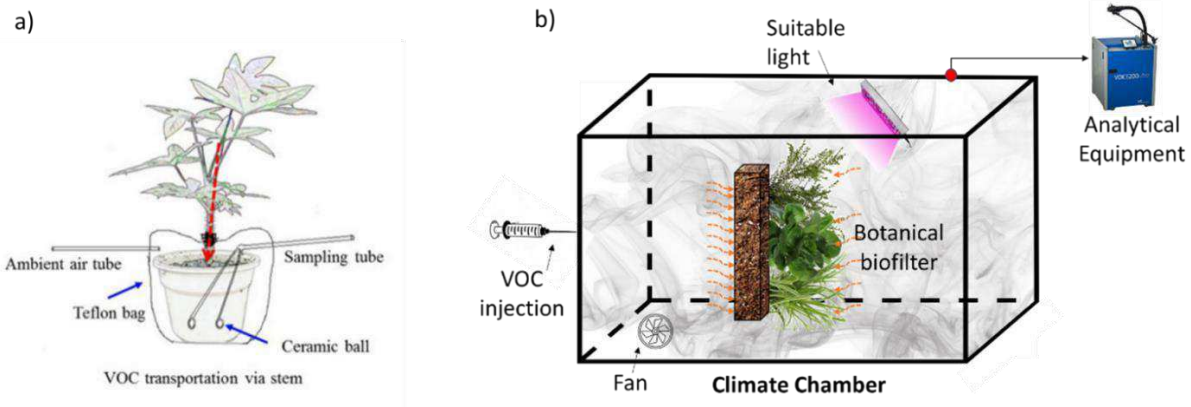

Figure 7. (a) Typical passive botanical biofilter of a potted plant exposed to VOC. The substrate has been isolated using Teflon bags to measure the relative contribution of the plant compartment (Kim et al., 2016). (b) A climate chamber configuration commonly used in assessing the drawdown capacity of passive botanical biofilters where a VOC is injected, and the removal is quantified over time.

548 Likewise, it is sustained that passive botanical biofilters do not create a pressure drop
549 differential necessary for the pollutant to be transported through the substrate or medium
550 (Llewellyn and Dixon, 2011). The mass transfer is the rate-limiting step since the VOC must be
551 in an aqueous phase before entering the microorganisms' cells (Darlington, Dat and Dixon,
552 2001). Furthermore, poor indoor ventilation and low concentrations of pollutants with low
553 diffusivities make the passive process inefficient (Kumar *et al*., 2023). Llewellyn & Dixon
554 (2011) likewise concluded that passive botanical biofilters (PBB) do not offer a solution in the
555 indoor environment because many plants would be needed to observe a change in pollutant
556 concentration. Nevertheless, NASA experiments proved the usefulness of potted plants in small
557 VOC-contaminated volumes, as mentioned above (Wolverton, Mcdonald and Watkins, 1984).

558 The inconsistency in the documented VOC removal capacity of passive botanical biofilters is a
559 returning feature in the literature. Hence, there is potential to research more in-depth passive
560 botanical biofilters, especially since they do not require an additional energy input or could be
561 integrated with current HVAC systems and natural ventilation. We argue that there might be
562 scenarios where a passive botanical biofilter is efficient, for instance, by optimizing the leaf
563 area, plant species combination, a more porous or combination of substrate, or via
564 bioaugmentation of the phyllosphere.

565 **4.2 Is converting into an active flow system the solution?**
566 It is widely spread that passive systems need to be transformed into active botanical biofilters
567 (ABB), also known as active botanical biofilters of active green walls, to be more efficient in
568 removing VOCs. The latter is accomplished if pollutant diffusion and mass transfer are
569 increased (and the pressure drop) via devices like axial fans creating forced mass convection,
570 as shown in Figure 8 (Irga, Pettit and Torpy, 2018). The substrate type, particle size, and water
571 content will determine the active flow effect and residence time. There is no consensus on the
572 airflow direction concerning the botanical biofilter. Often, studies on active botanical
573 biofiltration, like the work of Torpy *et al*. (2018) and Pettit *et al*. (2019), have directed the
574 airflow to the substrate.

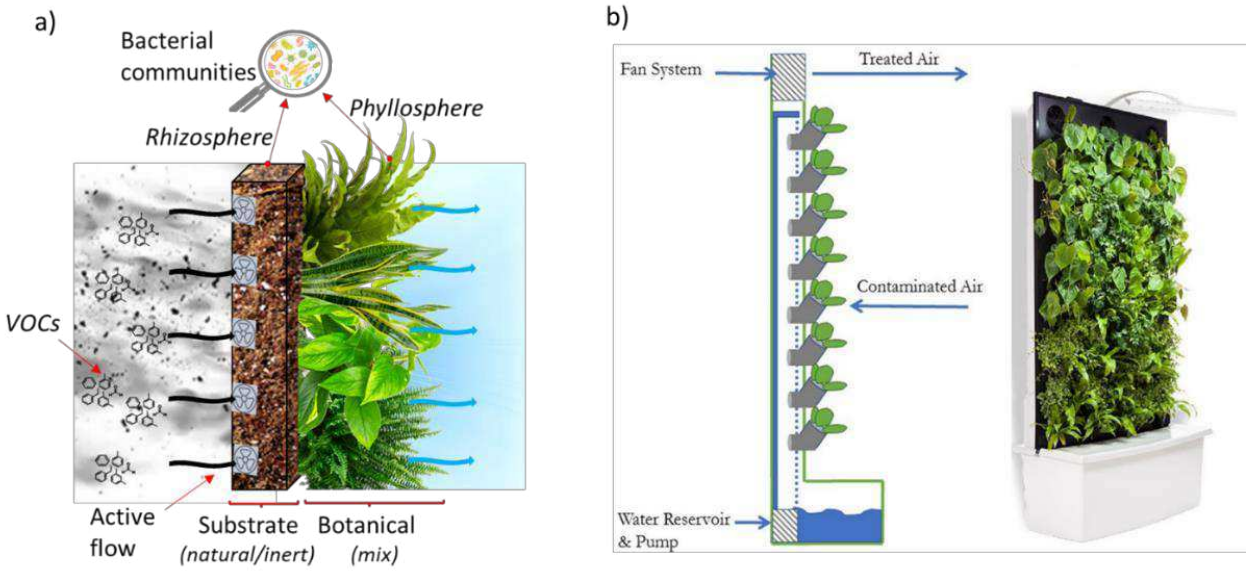

Figure 8. (a) Scheme of the operation and components of an active botanical biofilter (ABB). (b) The commercial active botanical biofilter (Naava One) was evaluated by Torpy et al. (2018) in removing methyl ethyl ketone in a 30 m³ climate chamber.

Increasing the airflow rate has been thought to translate into faster removal of air pollutants, as stated by Mikkonen *et al*. (2018) and Pettit *et al*. (2019). This was also confirmed by Wang and Zhang (2011), who evaluated a botanical biofilter removing formaldehyde and toluene whose substrate was a 50/50 mix of activated carbon and porous shale pebbles and plants grown were *Epipremnum aureum*. The study tested three airflow rates of 250, 600, and 900 m^3 h^{-1} in a climate chamber and found that faster airflow rates transformed into a faster decrease in concentration for both VOCs. However, it was clarified that while the removal speed increases, the single removal efficiency decreases due to short retention time. Nevertheless, a higher clean air flow rate can be delivered during a fixed period.

The forced airflow reduces the contact time between the VOC and the substrate, particularly the microorganisms (Ibrahim *et al*., 2021). This was exemplified by Wu and Yu (2022), who evaluated a 350 × 350 × 350 mm active green wall whose substrate consisted of peat and coconut fiber, and the plants utilized were *C. comosum*, *S. octophylla* and *C. elegeans*. The authors found that the single removal efficiency of formaldehyde decreased proportionally when increasing the airflow rate (from 30 to 50 to 65 m^3 h^{-1}). This was claimed to be the first study assessing the active flow effect on botanical biofiltration, and to this day, results are highly varied among studies. A more comprehensive tool to analyze the effect of airflow and obtain the optimal value is multiphysics modeling.

598 In addition, questioning the effect of active flow in the phyllosphere and rhizosphere microbial
599 communities is valid. To our knowledge, no study in botanical biofiltration has evaluated the
600 long-term airflow effect on the microbiome. For instance, drying of the media is more likely to
601 affect microorganisms or even spread microorganisms in the air directly.

602 On the other hand, Pettit *et al*. (2019) studied the efficiency of an active botanical biofilter
603 concerning the VOC chemical properties. A modular system with an area of 0.25 m^2 was filled
604 with a coconut husk substrate to sustain *Syngonium podophyllum* and was evaluated in a 0.22
605 m^3 climate chamber at an airflow of 0.35 m^3 min^{-1}. Then, 1.27E-05 mol of different families of
606 VOCs, namely, acetone, benzene, cyclohexane, ethanol, ethyl acetate, isopentane, isopropanol,
607 hexane, and toluene, were provided to the botanical biofilter. The average SPRE of all VOCs
608 ranged from 19 – 69 %, and it was concluded that the dipole moment and molecular mass are
609 predictors of VOC removal instead of Henry's law constant, vapor pressure, and octanol-water
610 partition coefficient. This study also suggested that by increasing the airflow, so does VOC
611 water partitioning and dissolution into the liquid phase.

612 Furthermore, the role of plant species selection in active botanical biofiltration has been
613 suggested to be of less influence given the short residence times, as stated by (Irga *et al*., 2019).
614 The latter had also been proposed by Torpy *et al*. (2018), who conducted a relevant study testing
615 a commercial active botanical biofilter (Naava One System) in removing methyl ethyl ketone
616 at 30 ppb in a 30 m^3 climate chamber with controlled conditions. Consistent VOC removal was
617 obtained over an 8-h period with an average SPRE of 57%, implying that plant choice did not
618 influence the removal, given the low levels of VOCs indoors. Notwithstanding, the role of
619 plants in active botanical biofiltration must be studied in the long term under varied airflow
620 rates. A summary of key studies evaluating passive and active botanical biofilters for VOC
621 removal under different designs and operation modes is shown in Table 3.

622 ## 5. HOW CAN MODELING ENGINEER BOTANICAL BIOFILTRATION?

623 ### 5.1 Existing models and gaps

624 A significant gap within botanical biofiltration lies in formulating a comprehensive set of
625 equations that describe complex processes for designing and optimizing a botanical biofilter
626 (Wang, Pei and Zhang, 2012). Although the modeling and simulation of traditional biofiltration
627 have been extensively studied, as presented in the review on biofilter models by Devinny and
628 Ramesh (2005), only the two studies conducted by Masi *et al*. (2022) and Wang, Pei and Zhang
629 (2012) were found containing the terms "botanical biofilter" and "model," which are discussed
630 below. Furthermore, most of the research has been performed in climate chamber experiments,

limited by volume, operation, costs, and conditions that can be tested. Hence, a modeling approach represents a powerful tool for testing a wide range of boundary conditions, like airflow rates, multiple VOC concentrations, chemical reactions, mass transfer coefficients, and biofilter dimensions.

The previous botanical biofilter models overlooked the botanical compartment, assuming that removal solely occurred within the substrate and associated microorganisms. Nevertheless, we have previously elaborated that a significant portion of the removal can occur within the botanical compartment via different mechanisms, and therefore, this compartment needs to be considered and studied in depth to be engineered (Kim *et al*., 2018b; Pettit, Irga and Torpy, 2018b; Irga *et al*., 2019) To our knowledge this is the first work that examines botanical biofilter models to provide guidelines to consider all phenomena in both compartments.

The one-dimensional numerical botanical biofilter model developed by Masi *et al*. (2022) considered the distinct phases, i.e., gas, aqueous, and solid, where transport and biochemical reactions take place simultaneously. The model was based on the idea that a botanical biofilter model is comparable to a traditional biofilter, as previously rationalized by Wang, Pei and Zhang (2012). Therefore, the authors assumed that the contributions of microorganisms could be implicitly considered in the model parameters, arguing that the plant compartment has a minimum role. This last assumption, however, can be questionable since the model would replicate a typical biofilter by not considering the botanical compartment and phyllosphere.

650 *Table 3. Overview of studies of botanical biofiltration, design, experimental conditions, operation, and main findings. ABB: Active botanical biofilter; PBB: Passive botanical biofilter; CSTR: Continuous stirred-tank reactor/dynamic chamber.*

N	Reactor mode	Botanical biofilter type	Dimensions	Plants	Substrate	Experimental conditions	Pollutant concentration	Main results	Identified or theorized mechanisms involved in the pollutants' removal	Reference
1	CSTR	Active green wall (ABB)	0.25 m^2 modules (500 x 500 x 500 mm) each one Climate chamber: Perspex chamber (0.6 m^3)	*Blechnum gibbum, Callistemon citrinus, Dianella caerulea, Eremophila glabra, Lomandra longifolia, Westringia fruticose*	Coconut fiber-based substrate Active airflow: 14.90 L s^{-1}	1505.5 (uppermost foliage) – 111.6 (bottom) µmol m^{-2} s^{-1} Plants watered to field capacity prior to testing Testing conducted between 09 and 17h when natural photosynthetic activity occurs	Benzene: NR PM: NR CO_2: 1000 ppm	-SPRE of benzene: 39 – 59%; *Dianella* had the greatest and *Lomandra* the lowest. -PM SPRE: no differences amongst species and PM size fractions. PM $_{5-10}$: 50 – 60%. -CO_2: None of the species removed this compound.	-Soil microorganisms are believed to be the main site for VOC removal. Plants could have modified the hydrophobicity of the substrate. -No root features nor leaf traits were correlated with high PM SPRE. -Plants were unable to remove CO_2 due to insufficient light supplied levels.	Paull, Irga and Torpy (2019)
2	CSTR	Potted plants in a climate chamber (PBB)	Chamber: 0.37 m3 (84 cm length x 62 cm width x 72 cm height)	*Chamaedorea elegans* Plants acclimated under laboratory conditions for one month.	Loamy soil: 30% sand; 30% silt; 15% clay; 25% humus.	1928.6 ± 197.4 SD lx Temperature and RH controlled using digital hygrothermometers Two potted plants used to increase the plant biomass to chamber ratio	Formaldehyde mean inlet concentration: 7.13 mg m^{-3}	- RE: 65 – 100% - Elimination capacity increased with elevating the inlet concentrations and reach a plateau at 16.4 mg m^{-3} - Total leaf area increased after experimentation.	- Plant and soil surface absorption, roots, degradation by microorganisms and uptake by the stomas	Teiri, Pourzamani and Hajizadeh (2018)
3	CSTR	Active living wall (ABB)	Modular system: 0.25 m^2 front surface area Modules: 500 x 500 x 500 mm, with 16 compartments Climate chamber: Perspex chamber (0.6 m^3)	*Chlorophytum orchidastrum* *Nematanthus glabra* *Nephrolepis cordifolia* *Schefflera arborícola* Plants maintained in a glasshouse at 23.7 C ± 3.6 °C and RH 68.1 ± 16%	Coconut coir-based substrate Active airflow: 14.90 L s-1	Maximum mid-day light level of 90 ± 10 µmol m^{-2} s^{-1} Plants watered once weekly to saturation	Hydrophilic VOC (ethyl acetate): 3.997 ± 0.074 ppm Hydrophobic VOC (benzene): 4.170 ± 0.144 ppm Ambient TVOCs: 35 ppb	- Ethyl acetate SPRE: 32.36 – 91.19%; significant differences amongst plant species - Benzene SPRE: 45.54 – 59.50%; significant differences amongst plant species - TVOCs: 60 – 70%; no significant differences amongst plant species - Plant type does influence the system's capacity for VOC removal. - Less botanical influence is expected under reduced residence time.	-Plant roots may provide hydrophilic adsorbent sites for ethyl acetate -Root exudates may alter the chemical composition of the rhizosphere and influence the capacity of the VOC to adsorb -Stomatal uptake is believed to be a pathway to ethyl acetate -Cuticle diffusion is attributed to benzene -Sorption due to low residence time	Irga et al. (2019)
4	CSTR	Active botanical biofilter (ABB)	Modular system: 0.25 m^2 modules containing 16 holes Climate chamber: Perspex chamber (0.22 m^3)	Single plant species: *Syngonium podophyllum*	Coconut husk coir Porosity: 53.27% Water holding capacity: 41.03% pH: 4.68	Indoor light levels: 6 µmol m^{-2} s^{-1} Modules watered with 2L of water 24h before testing Active airflow: 0.65 – 0.68 m^3/min Inlet temperature: 21 ˚C Inlet RH: 41.6%	1.275 × 10^{-5} mol of each: Acetone Benzene Cyclohexane Ethanol Ethyl acetate Isopentane Isopropanol Hexane Toluene	- Average SPRE of VOCs: 19 – 96% Ethanol SPRE: 96% Acetone SPRE: 72% - A single VOC does not represent the entire class of pollutants. - Dipole moment and molecular mass are predictors of VOC removal.	-Absorption and adsorption	Pettit et al. (2019)

5	CSTR	Active botanical biofilter (ABB)	Pot: height 14.5 cm, width: 13.5 x 13.5 cm Small chamber: airtight glass desiccator (volume 22.3 L) Large chamber: Glass 60 x 60 x 60 cm (volume 0.36 m^3) Airflow rate1 m^3 h^{-1}	*-Epipremnum aureum* *-Davallia fejeensis*	Small chamber pots: Activated carbon and granular constituents Large chamber: Leca: lightweight expanded clay Nmix	1500 – 2000 lux for 12h/d Acclimatization in the chamber: 7 weeks Each plant was irrigated for 15 min every day	Mixture of benzene, toluene, octane, p-xylene, α-pinene, decane, 2-ethylhexanol at a volume ratio of 1:1:2:2:2:4:6 (total VOC 1.7 – 4.3 ppm) The experiment maintained for 16 weeks	- Active airflow decreased the concentration of VOCs below the detection limit. - Immediate VOC dissipation was unlikely a biological process - Bacterial composition diverged in composition but not in diversity - Proteobacteria dominated the rhizosphere - Members of *Hyhomicrobiaceae* may be global green wall inhabitants.	-Active uptake by plants -Partition from the gaseous phase to moist surfaces of plants or solid phases of the growth media	Mikkonen et al. (2018)
6	CSTR	Active living wall biofilter (ABB)	Circular compartment system with an area of 150 x 100 cm Climate chamber: 30 m^3 (4.0 x 3.0 x 2.5 m) Airflow rate: 50 m^3 h^{-1} ACH: 1.67	*Philodendron scandens* *Asplenium antiquum-Syngonium podophyllum*	Media: inorganic growing media with activated carbon Moisture content increased to saturation twice daily	2500 lux (40 µmol m^{-2} s^{-1} PAR) for a 18 h day^{-1} photoperiod Chamber controlled to 21.5 ± 2 °C and RH 37.5 ± 2.5%	Methyl ethyl ketone: 30 ppb	-SPRE: 56.6 ± 0.86% -Consistent VOC removal over an 8-h testing period. -Plant choice does not influence removal, given the low levels of VOCs indoors	-Mechanisms not identified	Torpy et al. (2018)
7	CSTR	Full scale active green wall (ABB)	Modular system holding 16 plants (50 cm x 50 cm x 50 cm). Climate chamber: Perspex chamber (0.6 x 0.6 x 0.6 m; 0.216 m^3)	*Nephrolepis exaltata bostoniensis*	1) Coconut husk-based media: Water content: 72.5% Organic matter: 95% Specific surface area: 0.75 m^2 g^{-1} 2) Carbon-based media: Moisture: 2% Surface area: 1000m^2 g^{-1}	The pressure drop of all substrates was quantified to determine the required vacuum pressure.	PM: NR Benzene: 1.275E-05 mol Ethyl acetate: 1.253E-05 mol	-Coconut husk: There were no significant differences in the removal of ethyl acetate, benzene, and ambient TVOCS -Coconut husk + AC: Highly variable SPREs for ambient and high dose PM -The addition of higher proportions of AC improved the SPREs of the VOCs	-Hydrophilic adsorbent sites are proposed for the plant roots	Pettit, Irga and Torpy (2018a)
8	Batch	Potted plants (PBB)	Potted plants: diameter 20 cm Climate chamber: Plexiglass volume 0.14 m^3	*Ruscus Hyrcanus* *D. racemosa*	Substrate: NR.	Plants were kept indoors for 6 months at 21 – 25 °C and 50 ± 10% RH	Benzene: 10 µL L^{-1} Toluene: 20 µL L^{-1} Ethylbenzene: 20 µL L^{-1} Xylene: 50 µL L^{-1}	*-R. Hyrcanus:* Benzene: 8.5075 mg m^{-3} h^{-1} cm^{-2} Xylene: 86.66 mg m^{-3} h^{-1} cm^{-2} *-D. racemose:* Benzene : 2.14 mg m^{-3} h^{-1} cm^{-2} Xylene: 29.14 mg m^{-3} h^{-1} cm^{-2}	-Plant surface adsorption -Stomata absorption -Microorganisms' absorption	Fooladi et al. (2019)
9	Batch	Potted plants (PBB)	Climate chamber: volume of 1 m^3 (90 x 90 x 123 cm) Potted plants: diameter of 19 cm with a volume of 2.2 L	*Epipremnum aureum* *Dracaena Fragrans*	Substrate: Growing medium: (peat moss, perlite, dolomitic lime, gypsum, and a wetting agent) bark-humus and sand at 5:1:1 v/v/v.	Plants were acclimated >1 month to the indoor environment (T: 2 C, 20 ± 2 µmol m^{-2} s^{-1}, RH: 40% ± 6%). Testing during 12 h photoperiod Plants watered every 3 days.	Acclimation: 2 µL L^{-1} Toluene: 1.0 µL L^{-1} Benzene: 1.0 µL L^{-1} Xylene: 1.0 µL/L	-Toluene's removal was repressed by xylene and benzene -Total VOC removal increased with increasing xylene. Plant species had a major impact on the rate of VOC removal	- Plant metabolism -Microbe metabolism	Jin Kim et al. (2019)
10	Batch	Living wall modules (PBB)	Climate chamber: volume of 0.13 m3 (0.8 m x 0.4 m x 0.4 m) Modules: 0.49 m x 0.36 m	*Nephrolepis exaltata L.*	Substrate: a mixture of coconut fiber and peat	Light level of 6828 Lux	TVOCs: 5.59 – 7.61 mg m^{-3}	-TVOCs reduction rate: 0.07 mg m^{-3} h^{-1} - Highest reductions occurred in the first 15 min -Greater reductions were observed with high initial TVOCs concentrations	No mechanisms were identified	Suárez-Cáceres et al. (2021)

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A moist, porous medium was employed with considerations for advection, dispersion, mass transfer from gas to liquid and solid phases, and biodegradation reactions performed by microbes within a biofilm attached to solid bed particles. The microbes degrade the VOCs following a Haldane-type kinetic model. The model was validated, and VOC concentration profiles were obtained in the biofilter and biofilm. However, light, soil moisture, and plant removal mechanisms were neglected (Masi *et al*., 2022). Despite this model being validated and a sensitivity analysis done, it oversimplified botanical biofiltration. The authors stated that the conditions neglected should be included in a comprehensive model. Moreover, the authors coupled the biofilter model with a continuous stirred tank reactor (CSTR) scheme to be installed offline from an HVAC system. This configuration represented an indoor botanical biofilter arrangement (Figure 9).

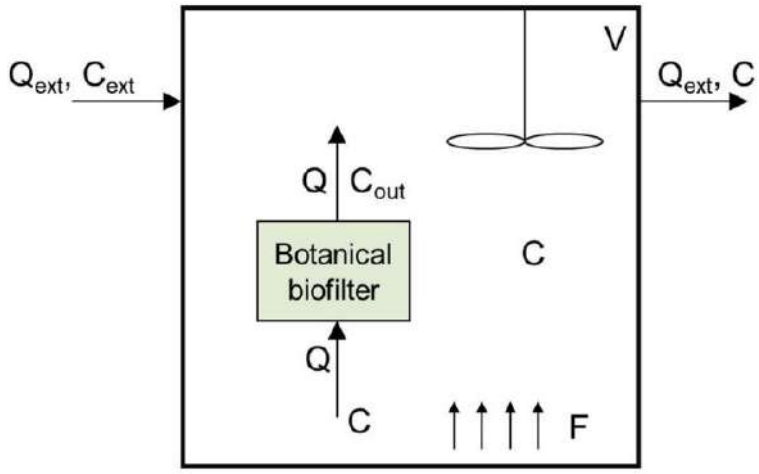

Figure 9. Typical configuration of a botanical biofilter in indoor settings. Taken from Masi et al. (2022).

A distinct approach was conceived by Wang, Pei and Zhang (2012), who used a "Coupled Heat, Air, Moisture and Pollutant Simulation for Building Envelope System" (CHAMP-BES) to evaluate the performance of an activated carbon-based botanical biofilter. The model assumed laminar flow through the bed, identical particles, homogenous density, and porosity. One of the main distinctions compared to the model of Masi *et al*. (2022) was the consideration of explicit sink terms to account for the pollutant removal instead of a bacterial growth model. However, this sink term considered the removal of VOCs by microorganisms, assuming that the VOC was bioavailable in the liquid and solid phase after being captured by the activated carbon. This is questionable since adsorbed VOC (solid phase) would be tough to be taken by microorganisms. The model was fitted with experimental data to obtain a critical k-value, representing the biodegradation rate constant. Basically, a higher k-value translates to a longer time required to

reach a steady state. Still, the model did not account for the removal in the botanical compartment by the plant tissue or phyllosphere bacteria.

5.2 Building a more comprehensive multiphysics model

Based on the above findings, an extended botanical biofiltration model is urgently needed to optimize and engineer the technology according to the needs of the indoor environment. The latter can be accomplished upon state-of-the-art insights into the phenomena underpinning biofiltration, phytoremediation, bioremediation, and indoor airflow. A foundational model shall encompass airflow simulation and the mass transport phenomena within both compartments of a botanical biofilter, including microorganism biodegradation (Figure 10). Therefore, a comprehensive model that aims to study and engineer a botanical biofilter is a multiphysics and challenging problem. Given its complexity, using commercially available Computational Fluid Dynamics (CFD) packages like FLUENT or COMSOL may aid in solving such a model. The contaminated VOC airflow around the botanical biofilter can be considered steady, incompressible, and turbulent. For instance, Hwang, Yook and Ahn (2011) used the standard k-ε model to obtain the flow field around vegetation using an inlet velocity, concentration, and no-slip wall boundary conditions.

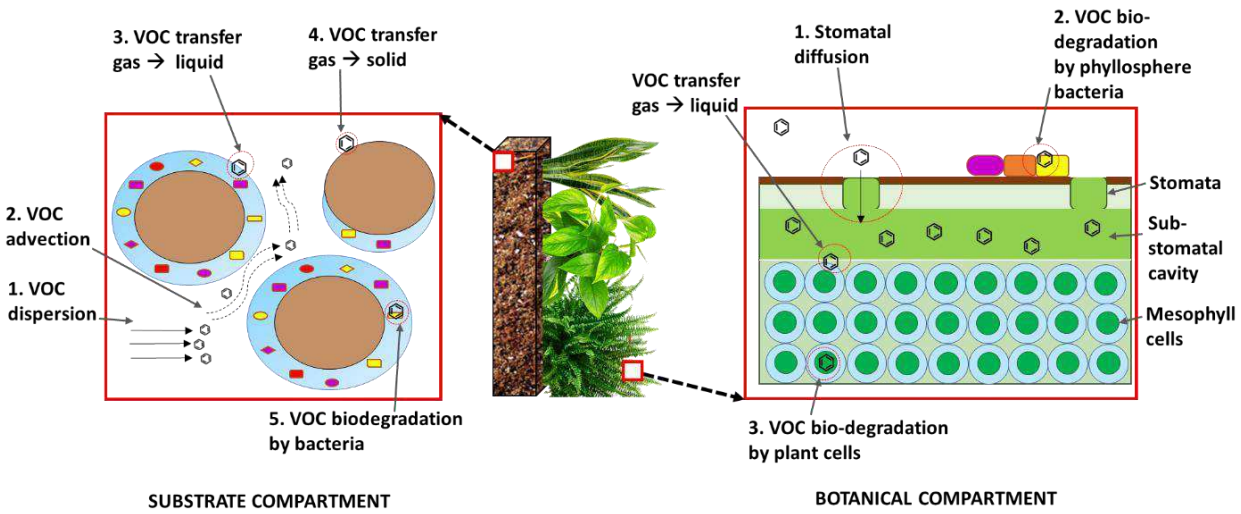

Figure 10. Summary of the transport phenomena and biodegradation steps occurring in the compartments of a botanical biofilter. The substrate compartment is elaborated upon the transport phenomena considered in the biofiltration models of Wang, Pei and Zhang (2012) and Masi et al. (2022).

Firstly, the airflow in an indoor space around and through the botanical biofilter must be solved. For this purpose, the substrate compartment can be built as an unsaturated porous medium with a gas, liquid, and solid phase. Permeability, which involves particle shape and size, and porosity dictate the airflow behavior. Similarly, the botanical compartment can be seen as a porous

medium with given permeability. However, this remains a great challenge due to the aerodynamic characterization of vegetation. Plant morphological parameters determine permeability or how easily the air flows through vegetation. This could be solved by adding a momentum sink term due to the vegetation, considering the leaf area density and a drag coefficient (Ysebaert, Samson and Denys, 2022).

The convection-diffusion equation must be added and coupled with the airflow field. Each compartment's mass transport phenomena will depend on the airflow field and mass transfer coefficients from the gas to the liquid and solid phases. Finally, the sink terms for the VOC removal and biodegradation shall be parameterized. The VOC sink term for the substrate compartment in which microorganisms are present in the biofilm or liquid phase needs to consider the biofilm thickness, the amount of VOC degrading microorganisms, the VOC bioavailability, and the stability of the microbiome. Parallelly, the VOC sink term for the botanical compartment must include the leaf area density as it translates into a higher stomatal number, the biodegradation due to the plant cells (phytodegradation), and the amount of VOC phyllosphere bacteria on the leaf surface. Finally, a sensitivity analysis can be conducted by perturbing the input parameters in a feasible range and conducting simulations. This way, relationships between the botanical biofilter, its compartments, and the specific indoor environment can be obtained to optimize the technology—for instance, the optimal dimensions, airflow rate, water saturation, and bacterial concentration.

6. STANDARDIZATION OF TECHNOLOGY OPERATION AND ASSESSMENT

Botanical biofiltration experiments have successfully evaluated removal capacities, mostly in laboratory climate chambers, assessing pollutant drawdown (batch) or the system's single pass removal efficiency (SPRE), as shown in Table 3. The former offers information on the system's drawdown but lacks quantitative efficiency data. The latter could be more representative of actual indoor environments since VOCs are continuously emitted from sources (Figure 1) at relatively stable concentrations (Dela Cruz *et al*., 2014; Torpy *et al*., 2018). Extrapolating experimental climate chamber results to actual indoor environments proves challenges due to the diverse range of indoor air pollutants and specific room factors such as volume, temperature, airflow, moisture, and ventilation (Pettit, Irga and Torpy, 2018b). Hence, there is an urgent need to develop and integrate experimental procedures to assess botanical biofiltration (Figure 11). These shall include assembling the botanical biofilter, laboratory testing, indoor environment assessment, and final implementation. For instance, guidelines regarding chamber size, VOC type and concentration, airflow, and microbial community analysis are needed.

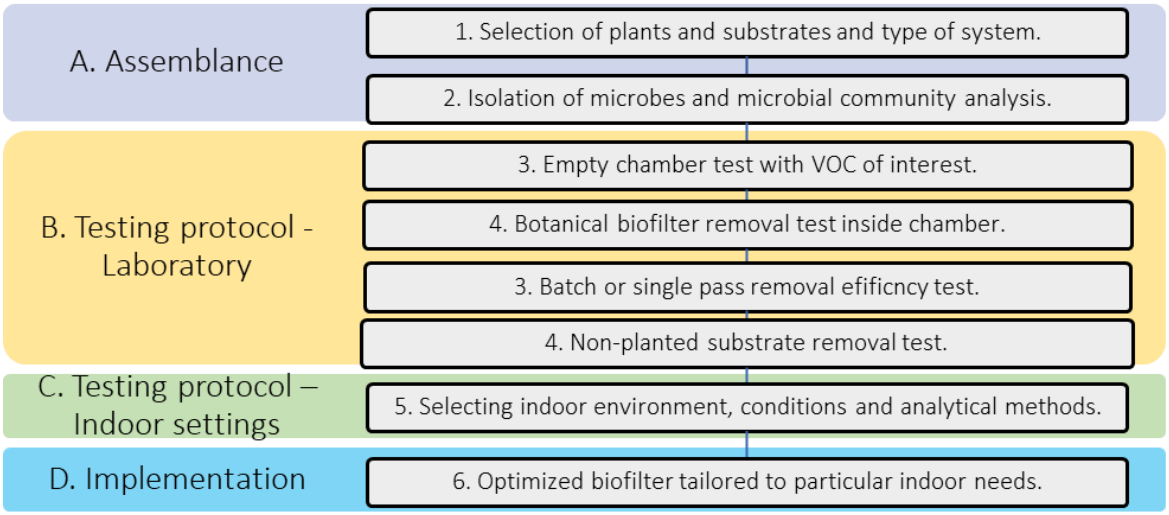

Figure 11. General scheme for developing, testing, and implementing botanical biofiltration technologies. Adapted from Wei et al. (2017) and Torpy et al. (2018).

On the other hand, it is necessary to compare botanical biofiltration performance against alternative systems. A more reliable metric to compare different systems and non-biological purification techniques is the clean air delivery rate (CADR). Metrics used to quantify the efficiency of a botanical biofilter are derived from those employed in traditional biofiltration. In addition, metrics used in conventional air handling systems are added for comparison purposes amongst indoor air pollution control systems. An overview of the operational parameters and metrics that can be used to measure the efficiency of an active botanical biofilter is shown in Table *4*. The dynamics of passive botanical biofilters and potted plants evaluated in climate chambers differ since no inlet or outlet exists. More details to evaluate these are found in the study of Ibrahim *et al*. (2021).

Table 4. Operational parameters of an active botanical biofilter. Adapted from Kennes, Rene and Veiga (2009); Soreanu, Dixon and Darlington (2013) and Matheson *et al*. (2023). V_{BB}: volume of botanical biofilter; Q_{BB}: botanical biofilter airflow rate; $VOC_{in/out}$: inlet/outler VOC concentration.

Metric	Equation	Unit	Description
Residence time (θ)	$\frac{V_{BB}}{Q_{BB}}$	s	Mean time spent by the VOC in the total volume of BB.
Mass inlet load rate (IL)	$\frac{Q_{BB} \times VOC_{in}}{V_{BB}}$	g VOC m^{-3} s^{-1}	Mass of VOC entering the BB per unit time and volume of BB.
Single pass removal efficiency ($SPRE$)	$\frac{VOC_{in} - VOC_{out}}{VOC_{in}} \times 100$	%	The ratio of VOC removed by the BB to the VOC fed.
Elimination capacity (EC)	$SPRE \times IL$	g VOC m^{-3} s^{-1}	Mass of VOC degraded by the BB per unit time and volume of BB.
Clean air delivery rate ($CADR$)	$SPRE \times Q_{BB}$	m^3 s^{-1}	
Biofilter refreshment capacity (BRC)	$\frac{CADR}{V_{room}}$	s	Air exchange rate of the BB.
Required biofilter volume (BRV)	$\frac{BRC \times \theta \times V_{room}}{SPRE}$	m^3	Required BB size to clean a room of a given size

7. CONCLUSIONS AND FUTURE PERSPECTIVES

Botanical biofiltration offers many advantages over traditional systems, particularly because it can biodegrade and break down a broad range of VOCs requiring a low energy input while providing additional societal benefits. These societal benefits encompass decreased indoor health-related costs, enhanced work productivity, and increased well-being.

Overall, innovative efforts have been carried out to analyze the removal capacity and potential of various plant species, potted plants, and passive and active biofilters in reducing indoor air pollutants, particularly VOCs like BTEX and formaldehyde. Less focus has been paid to understanding and quantifying the relative contribution of degradation mechanisms in both compartments. Mostly, research attributes the removal to the substrate compartment and associated microorganisms undermining the botanical part.

Disparities exist regarding the effect of plant and substrate selection, microbiome enrichment, removal stability, and system operation. Results obtained from studies involving potted plants and passive systems may not be directly extrapolated to active botanical biofilters. Moreover, whether laboratory-scale experiments are significant in indoor settings is questioned. Nonetheless, we suggest that further research should be done on passive systems as they may be a solution for specific indoor environments and represent lower environmental impact due to non-energy use and less maintenance. Additionally, for active systems, the effect of the active

flow on the long-term performance and its effect on the microorganisms is not fully understood. Hence, it cannot be ensured that the solution to a botanical biofilter solely increases the flow rate. Further studies could develop hybrid systems where the active flow is activated when necessary.

Future work should study the microbiome's stability, particularly the bioaugmentation of the phyllosphere and plant. Likewise, the botanical compartment needs to be paid attention in optimizing its contribution relative to the substrate. Lastly, multiphysics modeling must be developed to study the technology more in-depth by altering crucial parameters to optimize it according to the indoor environment needs. Incorporating all biological phenomena in a model is a challenge that requires experimental work as input.

This review has shed light on the disparities within the field, the involved mechanisms at play, and the importance of standardized protocols to assess the technology. Botanical biofiltration and green wall filtration research are growing internationally, and so is the interest in industrial applications for indoor environments. We conclude that embracing a multidisciplinary approach encompassing experimental work, multiphysics modeling, and microbial community analysis will aid in better understanding these systems, optimizing them, and facilitating their widespread adoption.

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