Towards practical indoor air phytoremediation: a review

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Abstract

Indoor air quality has become a growing concern due to the increasing proportion of time people spend indoors, combined with reduced building ventilation rates resulting from an increasing awareness of building energy use. It has been well established that potted-plants can help to phytoremediate a diverse range of indoor air pollutants. In particular, a substantial body of literature has demonstrated the ability of the potted-plant system to remove volatile organic compounds (VOCs) from indoor air. These findings have largely originated from laboratory scale chamber experiments, with several studies drawing different conclusions regarding the primary VOC removal mechanism, and removal efficiencies. Advancements in indoor air phytoremediation technology, notably active botanical biofilters, can more effectively reduce the concentrations of multiple indoor air pollutants through the action of active

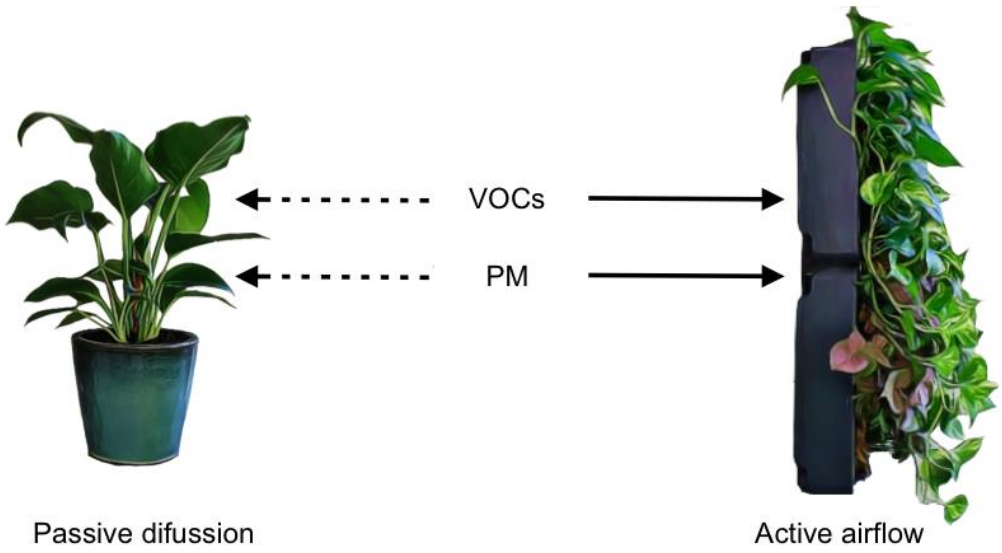
airflow through a plant growing medium, along with vertically aligned plants which achieve a high leaf area density per unit of floor space. Despite variable system designs, systems available have clear potential to assist or replace existing mechanical ventilation systems for indoor air pollutant removal. Further research is needed to develop, test and confirm their effectiveness and safety before they can be functionally integrated in the broader built environment. The current article reviews the current state of active air phytoremediation technology, discusses the available botanical biofiltration systems, and identifies areas in need of development.

Keywords: green wall; botanical biofilter; indoor air quality; living architecture; VOC; potted plant.

Highlights

- Indoor air pollution is associated with a range of detrimental health outcomes.
- The VOC phytoremediation mechanism is dependent on the type of VOC.
- Active botanical biofilters have overcome several limitations of potted-plants.
- Appropriately designed botanical biofilters may functionally clean air pollutants.

45 **Graphical abstract**



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1.0 Introduction: Air pollution exposure in the indoor environment

Due to the significant amount of time contemporary Western populations spend indoors, human exposure to most air pollutants is often quantitatively higher indoors than within the outdoor environment (Colbeck and Nasir, 2010; Molloy et al., 2012). In industrialised countries, people spend approximately 22 hours each day indoors (Klepeis et al., 2001), where chronic exposure to low levels of indoor air pollutants over time places them at risk of adverse health effects (Bernstein et al., 2008). With a rapidly increasing shift towards greater urbanisation globally (World Health Organisation, 2016), it is likely that population exposure to air pollutants within the indoor environment will continue to increase.

Indoor air pollution results from contaminated air entering buildings from outdoors, which is augmented by pollutants sourced from indoors such as carbon dioxide (CO_2) from occupant respiration, particulate matter (PM) released from occupant activities, such as cooking (Buonanno et al., 2009), and a range of volatile organic compounds (VOCs) off-gassed from a broad array of synthetic materials and cleaning products (Torpy et al., 2013). Air pollutants that are primarily generated within the indoor environment, such as VOCs, often accumulate in indoor air due to the reduced building ventilation rates that accompany contemporary building designs (Weschler, 2009). While power generation, urbanization and rapid industrialization are generally perceived as major factors contributing to poor air quality, minor sources of air pollution can produce significant health effects when a large fraction of the source's emitted pollution enters the occupants' breathing zones (Saksensa and Smith, 2012). Although the concentration of indoor air pollutants is often 2 to 4 times higher than that of outdoor air pollutants (Jafari et al., 2015), overall occupant exposure to indoor air pollutants can be 100 times higher than their exposure to

outdoor air pollutants (Jafari et al., 2015). This high exposure, therefore, affects a large portion of the population in urbanised regions. In the United States, between 800,000 and 1.2 million buildings may be associated with building-related illnesses, exposing 30 – 70 million workers to unhealthy working conditions (Fiedler et al., 2005).

During the 1970's, a desire to reduce building energy consumption initiated a trend where building ventilation rates were reduced (Seppänen, 2002), as *The American Society of Heating, Refrigerating and Air-Conditioning Engineers* (ASHRAE) reduced their standard for building ventilation rates (Burroughs and Hansen, 2004; Persily, 2015). Simultaneously, the utilisation of items that can act as indoor air pollution sources, such as new office equipment, has increased (Jafari et al., 2015). While policy-makers and environmental managers have generally ignored the role of minor sources of air pollution, particularly as they do not contribute substantially to ambient emissions (Saksensa and Smith, 2012), these trends, along with a growing body of evidence documenting pollutant-associated symptoms (Colbeck and Nasir, 2010), have led to the emergence of indoor air pollution as a consideration in public health (World Health Organization, 2010).

Management of indoor air quality is therefore critical to alleviate the burden of disease resulting from exposure to indoor air pollutants. It has now been known for decades that botanical systems are capable of cleaning air to some extent (Wolverton et al., 1984), while concurrently providing many other benefits such as biophilic satisfaction (Tifferet and Vilnai-Yavetz, 2017). Although this technology has been criticised for its estimated low *in situ* efficiency in modelling experiments (Llewellyn and Dixon, 2011), technological developments have introduced promising improvements in air cleaning capacity through the application of active airflow across

biofilter systems, however it is likely that this technology will need further improvements for successful *in situ* application.

This review aims to:

1. Assess the need for effective botanical biofiltration.
2. Determine the primary mechanism of VOC removal by botanical biofilters.
3. Uncover how active botanical biofilter design varies and how this affects system practically.
4. Investigate the challenges faced by active botanical biofiltration before these systems can be applied *in situ*.

2.0 Common Indoor air pollutants

There are notable differences in the types and concentrations of pollutants emitted from and found in indoor environments, and these are closely linked to differences in socio-economic development around the globe (Colbeck and Nasir, 2010). In developed countries, the most prominent and well-researched pollutants include VOCs (Wolkoff, 2013), CO_2 (Ramalho et al., 2015) and PM (Morawska et al., 2013). Although other pollutants have been detected in the indoor environment, such as NO_2 (Bozkurt et al., 2015) and O_3 (Wisthaler and Weschler, 2010), this review primarily focuses on the mitigation of VOCs, CO_2 and PM.

2.1 Volatile organic compounds

VOCs are of particular concern in indoor air, as almost all human VOC exposure occurs indoors (Arulneyam and Swaminathan, 2004). VOCs are primarily sourced within the indoor environment from off-gassing from building materials, furnishings (Zhang et al., 1996), solvents (de Gennaro et al., 2015), plastics and cleaning products (Schlink et al., 2010). Building ventilation rates will have a major influence on the levels of VOCs in a building, and due to the heterogeneity amongst buildings and indoor activities, the composition and levels of VOCs are highly variable amongst indoor environments (Cooke, 1991).

Exposure to VOCs in the indoor environment has been associated with several health effects, such as contributing to sick building syndrome (Brinke et al., 1998) and exacerbating asthma symptoms (Fuentes-Leonarte et al., 2009; McGwin Jr et al., 2010), while several common VOCs, such as acetyl aldehyde, have been identified as endocrine disruptors (Kawano et al., 2012), whilst others have been linked to problems with the nervous, hepatic, renal and respiratory systems (Sriprapat et al., 2014). Several VOCs, such as benzene and formaldehyde, are known carcinogens (Khanchi et al., 2015).

Health effects from VOC exposure can result from exposure to low concentrations of VOCs over both short and long temporal periods. Pappas et al. (2000) found that a mixture of common indoor VOCs had dose-response relationships for upper and lower respiratory symptoms, with exposure to concentrations of 25–50 mg/m^3 of total VOCs over only 4 hours causing detectable effects. Other studies have reported evidence of hematotoxicity after exposure to less than ~12 mg/m^3 of benzene over a 4 week period (Qu et al., 2002). These levels are indicative of poor indoor air quality, yet are still at lower concentrations than those typically found in industrial settings. Conversely, life time exposure to certain VOCs, such as benzene and 1,3-

146 butadiene, at concentrations as low as 1 μg/m^3, has been associated with increased
147 cancer risk (Khanchi et al., 2015).

148 *2.2 Particulate matter*

149

150 Aerosolized PM includes a broad range of suspended particles of mixed
151 chemical composition. It is generally classified based on aerodynamic diameter, as
152 this largely dictates the extent to which particles can penetrate the respiratory system
153 (Xing et al., 2016). Coarse particles are termed PM_{10} and include all particles with an
154 aerodynamic diameter less than 10 μm. Fine particles, with an aerodynamic diameter
155 less than 2.5 μm ($PM_{2.5}$), are able to penetrate deeper into the lung's gaseous
156 exchange region whereby they can enter the circulatory system, and thus have greater
157 health effects (Xing et al., 2016). This effect is compounded by the larger specific
158 surface area of smaller particles, which promotes the transfer of toxic compounds.
159 Coarse particles are also an important health concern, for example, black carbon
160 generated from incomplete combustion processes, such as diesel exhaust, has been
161 linked to more significant health effects when particles are of greater size (Janssen et
162 al., 2011). There is also a growing body of evidence regarding the negative health
163 effects of ultra-fine particles (Oberdörster and Utell, 2002; Bräuner et al., 2007;
164 Stölzel et al., 2007; Weichenthal et al., 2016), which have an aerodynamic diameter
165 less than 0.1 μm. In comparison to larger particles, ultra-fine particles do not
166 contribute significantly to the airborne PM mass concentration, yet they represent the
167 largest size fraction in terms of particle numbers (Slezakova et al., 2015). This may
168 partly result from vehicle emissions regulations in which mass output of particles is
169 regulated, therefore allowing a considerable number of low mass, ultra-fine particles
170 to be emitted (Oberdörster and Utell, 2002). In comparison to larger particles, the

significant health effects from ultra-fine particles have been hypothesized to result from their (i) increased reactivity, (ii) larger specific surface area, (iii) higher deposition rate in the pulmonary region and (vi) increased likelihood to penetrate epithelial tissues thus reaching interstitial sites (Stölzel et al., 2007).

Exposure to high concentrations of ambient PM is associated with increased morbidity and mortality due to cardiovascular, respiratory and venous thromboembolic disease (Bari et al., 2014). Relatively minor increases in PM concentration have been linked with significant health impacts, for example, a New Jersey, USA study found a 3% increase in all natural-cause mortality for the whole population for each 2 $\mu g/m^3$ increase in annual $PM_{2.5}$ exposure (Wang et al., 2016). A Canadian study found long-term exposure to relatively low concentrations of $PM_{2.5}$ (average concentration = 8.7 $\mu g/m^3$) was associated with increased mortality from cardiovascular disease (Crouse et al., 2012), while Li et al. (2017) report similar trends from several international cities.

Adverse health effects resulting from PM exposure are becoming increasingly prominent (Wyzga and Rohr, 2015; Feng et al., 2016; Maji et al., 2017), as is the documented presence of problematic PM levels in some indoor environments (Morawska et al., 2001; Morawska et al., 2003; He et al., 2004; Fromme et al., 2007; Branco et al., 2014; Challoner and Gill, 2014; Tunno et al., 2015; Stabile et al., 2017). PM concentrations are often high in urban environments, where it is commonly sourced from diesel emissions as solid black carbon associated with harmful hydrocarbons (Rohr and Wyzga, 2012). Indoors, suspended PM is generally sourced from the outdoor environment, and enters the building through mechanical ventilation systems or natural ventilation, such as windows and doors. Consequently, the prevalence of health effects resulting from PM exposure generally correlate strongly

with outdoor PM concentrations, despite the fact that most human PM exposure occurs in the indoor environment (Riley et al., 2002). High PM concentrations can also be sourced from within the indoor environment, with particles generated by activities such as cooking (Buonanno et al., 2009), smoking and use of office printers (He et al., 2007), while cleaning and increased building occupancy can lead to the re-suspension of previously precipitated particles (Long et al., 2000; Wheeler et al., 2011). Indoor generated PM contributes to 10–30% of the total burden of disease from PM exposure (Morawska et al., 2013).

2.3 Carbon dioxide

Whilst not always considered a pollutant, the quantitatively predominant indoor gaseous pollutant is CO_2, which can accumulate indoors through occupant respiratory emissions (Llewellyn and Dixon, 2011), coupled with insufficient building ventilation. The average global atmospheric concentration of CO_2 is ~400 ppm (World Meteorlogical Organization, 2016), however outdoor levels as high as 500 ppm have been recorded in urban areas (Persily, 1997), while concentrations inside office buildings typically range from 350–2500 ppm (Daniels, 2007). Elevated levels of CO_2 have been associated with 'sick building syndrome' (Jafari et al., 2015), decreased work place productivity (Milton et al., 2000; Seppänen et al., 2006) and decreased student school attendance (Gaihre et al., 2014), while Satish et al. (2012) found statistically significant decrements in decision-making performance as CO_2 concentrations increased from 600 to 1000 and 2500 ppm. A room's CO_2 concentration is often used as a surrogate indicator for the adequacy of building ventilation, and hence it is used to reflect the capacity of a building ventilation system to flush other indoor air pollutants (Fisk et al., 2006). Despite this, even with good ventilation that achieves low CO_2 concentrations, the complex relationship between

the concentrations of individual pollutants and their sources in some buildings makes specific systems for reducing indoor VOCs and particles necessary to achieve satisfactory indoor air quality (Ramalho et al., 2015).

3.0 HVAC systems for indoor air quality maintenance

The ventilation components of heating, ventilation and air conditioning (HVAC) systems are widely used in the developed world to control indoor air quality by replacing polluted indoor air with outdoor air (Wargocki et al., 1999; Lin and Chen, 2014), alongside their temperature and humidity modulation functions. Ventilation rates may be increased to maintain acceptable indoor air quality, and while this may be the simplest and currently most effective method of improving indoor air quality (Torpy et al., 2015), it uses significant amounts of electrical energy, especially when there is a large temperature differential between outdoor air and the required indoor conditions. The cumulative energy usage in industrialized countries is substantial: in the USA, ~26.5% of all energy use is attributed to HVAC systems (Ben-David and Waring, 2016). Although HVAC systems have the potential to bring outdoor sourced pollutants into the indoor environment, nearly all mechanically ventilated buildings use filters that reduce inlet air PM concentrations (Quang et al., 2013; Ben-David and Waring, 2016). Nonetheless, in numerous cases, indoor PM concentrations closely correlate with proximal outdoor PM concentrations (Morawska and Clark, 2000; Guo et al., 2010; Morawska et al., 2013) due to the inefficiencies of HVAC filter systems, as common commercial systems can only filter a proportion of PM from influent air. HVAC PM filters commonly used in building ventilation, such as MERV (minimum efficiency reporting value) 4, 6, 10 and 11 filters, have removal efficiencies of less than 20% for all particle sizes (Stephens and Siegel, 2013), and

246 although more efficient filters are available (Quang et al., 2013), increased efficiency
247 is met with greater airflow resistance and thus energy use, higher maintenance, and
248 reduced sustainability (Montgomery et al., 2012), while remaining incapable of
249 capturing gaseous pollutants: HVAC systems reduce indoor VOC concentrations
250 solely by dilution with outdoor air. While naturally ventilated buildings (those that
251 rely on building openings to allow the entry of outdoor air) generally use less energy
252 than mechanically ventilated buildings, they have the potential to provide greater
253 exposure to PM of outdoor origin, as they lack integral PM filtration (Ben-David and
254 Waring, 2016). Furthermore, natural ventilation is not always possible due to outdoor
255 climatic conditions (Guieysse et al., 2008).

256 In an attempt to limit the energy cost of ventilation, the rate of infusion of
257 outdoor air into the indoor environment has been reduced over time, as buildings have
258 increasingly been designed to be as 'air tight' as possible. However, this has a
259 detrimental effect on the concentrations of indoor sourced pollutants (Darlington et
260 al., 2000). Consequently there is an emerging need to purify the air inside buildings
261 (Guieysse et al., 2008). Several air purification methods may be integrated into a
262 building's ventilation system, such as combinations of air filtration, ionization,
263 activated carbon absorption, ozonation and photocatalysis (Chen et al., 2005;
264 Guieysse et al., 2008; Luengas et al., 2015). Although each of these techniques can
265 effectively ameliorate a single pollutant type, the numerous pollutants with different
266 physio-chemical properties in a typical indoor environment makes joint effective
267 treatment difficult (Luengas et al., 2015), and all mechanical methods are expensive,
268 potentially hazardous (notably ozonation), and use energy themselves. There is
269 therefore a crucial need to develop air-cleaning technologies that are energy efficient
270 and capable of treating a wide range of air pollutants efficiently.

4.0 Bioremediation of VOCs with potted-plants

As an alternative to mechanical systems, several studies have revealed the potential of passive botanical biofilters, such as potted-plants, to phytoremediate several indoor air pollutants, in particular a range of VOCs (Godish and Guindon, 1989; Wolverton and Wolverton, 1993; Wood et al., 2002; Orwell et al., 2004; Wood et al., 2006; Aydogan and Montoya, 2011; Torpy et al., 2013). A series of studies by NASA during the 1980s demonstrated the capacity of indoor potted-plants to enhance air quality by reducing the ambient concentration of numerous VOCs (Wolverton et al., 1984; Wolverton et al., 1985; Wolverton, 1988). The majority of early botanical air cleaning research assessed VOC removal by simple potted-plants (Godish and Guindon, 1989; Wolverton et al., 1984; Wolverton and Wolverton, 1993; Porter, 1994; Wood et al., 2002; Orwell et al., 2004; Orwell et al., 2006; Wood et al., 2006), demonstrating significant removal of high concentrations of VOCs from sealed chambers, with reductions ranging from 10–90% over 24 hours (Llewellyn and Dixon, 2011). Possibly due to the variances in conditions amongst different experiments such as the use of different plant species, VOCs, pollutant concentrations, chamber sizes and light levels (see Table 1), it is difficult to ascertain which components of the potted-plant system are responsible for VOC removal. Most of our understanding of the mechanisms of VOC removal is derived from experiments that have used aluminum foil or Teflon bags to isolate a particular part of the potted-plant microcosm (Aydogan and Montoya, 2011; Treesubsuntorn and Thiravetyan, 2012; Sriprapat et al., 2014; Kim et al., 2016), or experiments that have assessed VOC removal under different lighting conditions (Porter, 1994; Kondo et al., 1995; Wood

et al., 2002; Orwell et al., 2004; Yoo et al., 2006; Kim et al., 2008; Aydogan and Montoya, 2011; Xu et al., 2011; Treesubsuntorn and Thiravetyan, 2012; Hörmann et al., 2018; Teiri et al., 2018), while several experiments have simply assessed VOC drawdown without testing removal mechanisms (Cornejo et al., 1999; Orwell et al., 2006; Liu et al., 2007; Yang et al., 2009; Kim et al., 2010; Kim et al., 2014; Mosaddegh et al., 2014). A thorough understanding of the removal mechanism is crucial if these systems are to be optimized with the intention of enhancing the VOC removal rate.

4.1 Removal mechanisms of VOCs

4.1.1 Potting substrate material and substrate microorganism effects

While it was initially assumed that VOC removal was primarily an activity performed by the plant foliage, along with small contributions from the soil, roots and rhizospheric microorganisms (Wolverton et al., 1985; Wolverton 1988), this concept was not explicitly tested. In following experiments, Godish and Guindon (1989) and Wolverton et al., (1989) both independently compared the VOC removal efficiencies between ordinary potted-plants and potted-plants with their foliage removed. Both studies concluded that a significant portion of uptake must occur through the potting substrate. Wolverton et al.'s, (1989) comparison between a potted plant and a pot containing only soil led to the inference that plants must be growing in the soil in order for the potted-plant system to remove VOCs efficiently, and that microorganisms within the rhizosphere contribute to considerable VOC removal.

The contribution of the microbial community has further been demonstrated by experiments that have assessed VOC removal under light and dark conditions and have found no significant differences in the VOC removal under these two conditions (Wood et al., 2002; Orwell et al., 2004; Hörmann et al., 2018), suggesting that stomatal uptake by the plant is negligible for the VOCs that were tested. Several studies have found that the VOC removal efficiency of the potted-plant microcosm increases when the system is exposed to repeated doses of a pollutant (Wood et al., 2002; Orwell et al., 2004; Torpy et al., 2013), with these authors suggesting that this response is due to biostimulation of the substrate's microbial community. It is currently generally thought that as indoor air passes over a potted-plant and its substrate, pollutants are drawn into the substrate by diffusion and become a carbon nutrient source for some members of the microbial community (Wood et al., 2006).

While many experiments have assessed the potential for the bioremediation of single VOCs independently, hundreds of VOCs may be present simultaneously in a typical indoor environment (Meciarova and Vilcekova, 2016). Simultaneous biodegradation of multiple VOCs provides the opportunity for substrate interactions to occur. For example, the simultaneous microbial biodegradation of benzene and toluene has been shown to exhibit competitive inhibition, which limits the rate of the simultaneous biodegradation of the two pollutants (Yu et al., 2001). Orwell et al. (2006), however, observed a synergistic effect between the biodegradation of toluene and *m*-xylene. Orwell et al. (2006) suggested that this was a result of toluene supporting a specific microbial population and inducing within that population the activity of the enzyme catechol 1,2 dioxygenase which is used for the biodegradation of both pollutants, however when toluene concentrations become limiting, *m*-xylene was then more effectively biodegraded.

345 Numerous studies have noted the innate ability of plant growth substrates to
346 adsorb VOCs (Godish and Guindon, 1989; Hörmann et al., 2017), and consequently
347 substrates of different compositions have been trialled in experiments for their
348 capacity to influence VOC removal. Aydogan and Montoya, (2011) noted the
349 substrate's contribution to removal efficiency as their activated carbon substrate
350 treatment demonstrated larger reductions in formaldehyde in comparison to expanded
351 clay and growstone substrates, and concluded that substrates that have high adsorption
352 capacities and provide sufficient microbial sites could lead to increased VOC
353 removal. Further evidence for this claim came from Irga et al. (2013) who found
354 differences in the benzene removal efficiency between potted-plants grown in soil and
355 hydroculture, suggesting that differences in the density and diversity of the substrate's
356 microbial community were responsible for the differences in benzene removal
357 efficiency.
358
359 *4.1.2 Plant foliage and aerial part effects*
360
361 Several experiments have compared VOC removal efficiencies under different
362 lighting conditions (Porter, 1994; Kondo et al., 1995; Wood et al., 2002; Orwell et al.,
363 2004; Yoo et al., 2006; Aydogan and Montoya, 2011; Xu et al., 2011; Treesubsuntorn
364 and Thiravetyan, 2012; Hörmann et al., 2018; Teiri et al., 2018). These experiments
365 used light intensity as a surrogate for foliage uptake under the assumption that
366 increased light intensity increases stomatal conductance and plant metabolic activity
367 (Porter, 1994). Of these experiments, there is no clear consensus on whether light
368 intensity influences the removal rate of VOCs, however recent work by Hörmann et
369 al. (2018), has suggested that the possible primary removal mechanisms may be both

370 VOC and plant species dependent. Hörmann et al. (2018) tested toluene and 2-
371 ethylhexanol degradation under light and dark conditions and found no differences in
372 the removal rate of toluene between these treatments, yet found that some of their
373 tested plant species exhibited differences in the removal efficiency of 2-ethylhexanol
374 depending on light or dark conditions. Work that has assessed benzene removal has
375 generally found no difference in benzene removal under different light conditions
376 (Orwell et al., 2004; Wood et al., 2002). Alternatively, experiments that have assessed
377 formaldehyde removal efficiency by potted-plants amongst different light intensities
378 have generally found that increased light intensity is associated with increased
379 removal (Kondo et al., 1995; Xu et al., 2011; Teiri et al., 2018) however Aydogan and
380 Montoya (2011) found that all of their tested plant species demonstrated quicker
381 formaldehyde removal under dark as opposed to light conditions.

382 Several studies have isolated aboveground plant parts from the root zone and
383 substrate with the use of physical barriers, and have concluded that leaves are capable
384 of VOC removal (Lin et al., 2007; Tani et al., 2007; Tani and Hewitt, 2009;
385 Treesubsuntorn and Thiravetyan, 2012; Sriprapat and Thiravetyan. 2013;
386 Treesubsuntorn et al., 2013; Sriprapat et al., 2014a; Sriprapat et al., 2014b). While
387 stomatal uptake offers one possible means of VOC removal by the aerial part of the
388 plant, some VOCs are also able to become adsorbed to or diffuse across the cuticle
389 (Baur and Schönherr, 1995; Treesubsuntorn et al., 2013), with some authors
390 suggesting that removal by the cuticle is depenendent on wax quanity and chemical
391 structure (Treesubsuntorn et al., 2013). Although this work has suggested that plant
392 leaves are capable of some VOC removal, these studies have not directly made
393 comparisons between removal by the root zone component and the aerial component
394 of the potted-plant system. Alternatively, Aydogan and Montoya, (2011) tested the

formaldehyde removal efficiency of the root zone and aerial parts independently and found that while the aerial parts of plants were capable of VOC removal, removal by the root zone occurred at a substantially faster rate. Kim et al., (2016), used Teflon bags to contain certain plant parts, and suggested that although the root zone is important for toluene and xylene degradation, uptake and transport of the pollutants by the stem tissue is critical for transferring the pollutant to the root zone. Setsungnern et al., (2017) measured removal rates by a potted *Chlorophytum comosum* with its roots covered with aluminium foil and measured benzene degradation within plant cells, and concluded that *C. comosum* was capable of removing 68.77% of an initial 500 ppm concentration of benzene over an eight day period.

In support of the Green Liver Model, where plants use metabolic processes to manage toxic compounds inside the plant cell, Setsungnern et al., (2017) found that after plant uptake, benzene was oxidised to phenol within plant tissues by the cytochrome P450 monooxygenase system, before being catalysed to catechol and then cleaved to produce cis, cis-muconic acid. In comparison, Orwell et al. (2004) found that a potted-plants microbial community was responsible of removing ~97% of benzene within 24 h after 3 days of exposure to an initial concentration of 25 ppm that was 'topped up' to the starting concentration every 24 hours.

Hörmann et al. (2017) covered the potted-plant's substrate with foil to assess removal by the plant's aerial parts and compared this to a 'potting soil' treatment, noting similar VOC removal rates between the treatments. While this method revealed that both soil and the plant's aerial components are independently capable of degradation, it has been suggested that the plants play a key role in promoting substrate microbial VOC degradation both through VOC transport (Kim et al., 2016) and microbial

biostimulation through the release of root exudates (Wood et al., 2002; Xu et al., 2010; Wang et al., 2014). Regardless of the primary removal mechanism, it is probable that the entirety of the potted-plant system is needed for quantitatively effective VOC removal, as the root zone and aerial components support each other to maintain mutual health (Wood et al., 2002; Xu et al., 2010; Aydogan and Montoya, 2011, Wang et al., 2014).

At the current state of research, it is difficult to determine the exact removal mechanism for a range of behaviourally different VOCS when applied *in situ*. The trialling of the multiple VOC removal performance of botanical bioremediation systems in real indoor environments, using highly sensitive apparatus so as to quantify the very low level pollutants present, will be required to reveal the true value of these systems.

4.1.3 Effects of biostimulated microbial communities

After establishing that microorganisms within the potted-plant system play a role in VOC removal, some experiments have looked at optimizing the system through biostimulating or bioaugmenting the microbial community for enhanced VOC degradation. Torpy et al. (2013), who compared the removal of benzene between ordinary potted-plants and potted-plants with a stimulated substrate microbial community, found that specifically enhancing the growth of the benzene-degrading components of the bacterial community increased benzene removal. Similarly, Sriprapat and Thiravetyan (2016) identified benzene degrading bacteria in the phyllosphere and found that potted-plants with sterilized leaf surfaces exhibited decreased benzene removal rates, while plants inoculated with the identified

endophytic benzene-degrading bacteria showed an increased benzene removal efficiency in comparison to ordinary potted-plants. De Kempeneer et al., (2004) showed that toluene removal rates could be increased by inoculating the leaf surface with a culture of toluene-degrading bacteria. Notably both of these studies used high concentrations of VOCs, and it remains largely unknown how inoculated phyllospheric microbial communities could be sustained in *in situ* conditions (De Kempeneer et al., 2004). Alternatively, Khaksar et al., (2016) inoculated two non-native host plant species, *Zamioculcas zamiifolia* and *Euphorbia milii*, with an endophytic species of bacterium (*Bacillus cereus),* and found that plants with the endophytic *Bacillis cereus* inoculation experienced increased tolerance to formaldehyde phytotoxicity. An endophytic *Bacillis cereus* inoculation has also been shown to enhance *Clitoria ternatea* seed germination and sapling growth under formaldehyde stress and simultaneously enhance gaseous formaldehyde removal (Khaksar et al., 2016b). These methods of system optimisation need to be thoroughly explored for their potential application in long term *in situ* scenarios, while it is also necessary to accurately uncover these removal methods' comparative efficiency in relation to substrate mediated removal effects.

464 **Table 1. A summary of static chamber experiments that have assessed VOC drawdown.**

Author	Pollutant(s)	Starting concentration(s)	Removal rate/efficiency	Experimental light conditions	Suggested removal mechanism/conclusion	Was removal mechanism indirectly tested?
Aydogan and Montoya, 2011	Formaldehyde	~2.038 mg/m^3	81-96% over 24 hours	Cycles of ~28–70 µmol/m^2/s for 12 hours followed by a dark period	Removal by the root zone was faster in comparison to the aerial parts of the plants. Furthermore, there was no discontinuity in removal rate with a transition from light to dark. Concluded that both rhizosphere and aerial parts contribute to removal.	Tested differences in removal rates by comparing the removal rate of the entire plant; rhizosphere + substrate (by surgically removing aerial parts); aerial parts (by sealing the rhizosphere and substrate in a Teflon bag). Tested removal rates under light and dark conditions.
Cornejo et al., 1999	Benzene, pentane, toluene, trichloroethylene	33.176 mg/m^3 for benzene; not stated for other VOCs	0.6-8.5 µg/g/24h	Ambient light and incandescent lamps	Did not identify mechanisms. Only suggested that species morphology and physiology, such as stomatal density and enzymatic activity, may affect pollutant uptake.	No

De Kempeneer et al., 2004	Toluene	339 mg/m^3	7-76 hours to remove 95% of the initial dose	Not stated	Did not identify mechanism, but found that removal efficiency could be increased by bioaugmenting the leaves with an inoculum of toluene-degrading bacteria. Uninoculated plants were also capable of removing toluene	Potting soil was covered by polyethylene to prevent sorption by the roots/substrate, but no comparisons were made comparing uptake between substrate, roots or aerial parts.
Hörmann et al., 2017	Toluene, 2-ethylhexanol	14.6-20.0 mg/m^3	~1-9.5 mg/m^3 over 48 hours	180 $\mu mol/m^2/s$	The potting soil has a similar removal rate to the aerial plant parts.	Covered the substrate with foil to assess removal by aerial parts and compared this to a 'potting soil' treatment.
Hörmann et al., 2018	Toluene, 2-ethylhexanol	14.6-20.0 mg/m^3	1.4 to 5.7 $L/h/m^2$ of leaf area	A light treatment of 180 $\mu mol/m^2/s$ and a dark treatment	Aerial plant parts have no major impact on VOC removal. No significant differences in toluene removal under light and dark conditions, and different physiological differences amongst species did not influence removal rate. 2-ethylhexanol removal varied among	Compared removal rates under light and dark conditions.

					lighting conditions in some species.	
Irga et al., 2013	Benzene	80 mg/m^3	739-1444 $\mu g/m^3/h$ per pot	20 $\mu mol/m^2/s$	Suggested that differences in VOC removal amongst substrate treatments were due to differences in the density and diversity of the substrate's microbial community.	Compared VOC removal efficiencies amongst a potting mix treatment, 'virgin' soil treatment, and a hydroculture treatment.
Kim et al., 2010	Formaldehyde	2.472 mg/m^3	0.13-6.64 $\mu g/m^3/cm^2$ of leaf area	20-60 $\mu mol/m^2/s$	Did not identify mechanisms, yet found differences amongst plant species.	No
Kim et al., 2014	Toluene and xylene	1.236 mg/m^3	~15-170 $\mu g/m^3/m^2$ of leaf area	20 $\mu mol/m^2/s$	VOC removal by plants increased as the root zone volume increased. No relationship between leaf surface area or above ground plant tissue volume and removal efficiency.	Tested the VOC removal efficiency amongst plants in different sizes of pots.
Kim et al., 2016	Toluene and xylene	1.303-1.884 mg/m^3	7.0-13.3 $\mu g/m^{3/}m^2$ leaf	20 $\mu mol/m^2/s$	The root zone is a significant contributor to VOC removal, but VOC	Teflon bags were used to seal aboveground

			area over a 24 h period		transportation to the root zone via the stem plays an important role.	parts from rhizosphere and substrate.
Lin et al., 2017	Formaldehyde	≥6.25 mg/m^3	~4 ppm over 17.1 hours	16.2 μmol/m^2/s	Suggested VOC removal can occur through foliar pathways.	No: the authors covered the substrate with foil to exclusively test foliar removal.
Mosaddegh et al., 2014	Acetone, acetonitrile, benzene, ethylbenzene, methanol, toluene, xylene	2.62-8.68 mg/m^3	0.24-4.42 mg/m^3/day	Cycles of 12 hours of darkness and 12 hour of light at undescribed levels	Did not identify mechanisms.	No
Orwell et al., 2004	Benzene	79.75 mg/m^3	12–27 ppm/d	~120 μmol/m^2/s	Substrate micro-organisms play a major role in VOC removal; plants may contribute to biostimulation of substrate microbes, assistance in VOC diffusion to substrate, or adsorption	Tested removal rates under light and dark conditions. Tested for VOC removal efficiency after plants had been removed from the pots.

					onto plant foliage.	
Orwell et al., 2006	Toluene, *m*-xylene	0.758-437 mg/m^3	0.68-1014 mg/m^2/day	~120 µmol/m^2/s	Suggested that removal is by substrate microbes.	No
Porter, 1994	Toluene, benzene	0-1200 mg/m^3	5.11-35% in 3 h	35-90 µmol/m^2/s	VOC removal efficiency was light dependent.	Tested removal rates under different light levels
Setsungnern et al., 2017	Benzene	1595 mg/m^3	343.85 ppm over 8 days	50 µmol/m^2/s	VOC removal rate was dependent on red or blue light; benzene uptake affected plant gene expression.	No: the authors covered the substrate with foil to exclusively test foliar removal.
Sriprapat and Thiravetyan, 2013	Benzene, ethylbenzene, toluene, xylene	63.8-81.67 mg/m^3	0.86-0.96 mmol/m^2 of leaf area at 72 h	Natural light-dark cycles	Removal can occur through stomatal uptake and diffusion into the cuticle.	No: the authors covered the substrate with foil to exclusively test foliar removal.
Sriprapat and Thiravetyan, 2016	Benzene	1416 mg/m^3	25.30-34.00 µmol/h/m^2 of leaf area	Fluorescent light with a 16 hour photoperiod at	Non sterilised plants removed benzene at a faster rate. Phyllospheric bacteria play a role in VOC removal.	Phyllospheric benzene degrading bacteria were identified, inoculated and applied as a treatment

				undescribed levels		
Sriprapat et al., 2014b	Toluene, ethylbenzene	70.88-81.67 mg/m^3	10.17 µmol/72 h of toluene; 11.11 µmol of ethylbenzene over 72 h	12 h photoperiod at undescribed levels	VOCS may be taken up by adsorbing onto the cuticle. This is influenced by cuticle composition.	No: the authors covered the substrate with foil to exclusively test foliar removal.
Sriprapat et al., 2014a	Xylene	81.6 mg/m^3	59.14-88.20% at 72 hours; 0.66-0.86 mmol/m^2 of leaf area after 72 hours	Natural light-dark cycles	Removal can occur through stomatal uptake and diffusion into the cuticle.	No: the authors covered the substrate with foil to exclusively test foliar removal.
Su and Liang, 2015	Formaldehyde	30, 60 or 120 mg/L applied as a solution	135 µg/h^1 per plant (maximum)	14 hours of light at 260–350 µmol/m^2/s	Suggested primarily by shoot	Not clearly tested, although tissue samples were taken from different plant parts

Torpy et al., 2013	Benzene	80 mg/m^3	Biostimulation increased removal rates by ~27%	120 $\mu mol/m^2/s$	Suggested that removal is due to substrate microbes.	Biostimulation of microbes increased removal efficiency
Treesubsuntorn and Thiravetyan, 2012	Benzene	63.8 mg/m^3	43-77% in 72 h	Light and dark periods at undescribed levels	Removal can occur through stomatal uptake and diffusion into the cuticle.	Roots covered in aluminium foil; light and dark testing
Treesubsuntorn et al., 2013	Benzene	63.8 mg/m^3	1.10-23.46 µmol/g over 3 d	Undescribed	High quantities of cuticle wax was associated with high benzene removal efficiency.	Did not test removal mechanism; only tested leaf removal- did not test pot effects
Wood et al., 2002	Benzene and *n*-hexane	79.75-353 mg/m^3	367-4032 $mg/m^3/day/m^2$ of leaf area	Light and dark conditions (light = 120 $\mu mol/m^2/s$)	Substrate microbes are the primary 'rapid response' agents of VOC removal.	Compared removal efficiencies between light and dark conditions as well as hydroponic and soil treatments.

Xu et al., 2011	Formaldehyde	1-4 mg/m^3, increasing by 0.5 mg/m^3 every 5 d depending on visible foliar injury	14-95% / 3 d; 0-2.2 mg/h	12 h cycles of darkness and light with light at 80, 160, 240 $\mu mol/m^2/s$	Both soil and leaves	Tested removal rates amongst different light levels.
Yang et al., 2009	Benzene, octane, α-pinene, toluene, trichloroethylene	31.9-55.7 mg/m^3	0.34-1.03 $\mu g/m^3$/3 h; 0.38-1.21 $\mu g/m^3$/6 h	~5.45 $\mu mol/m^2/s$	Did not identify mechanisms	No
Yoo et al., 2006	Benzene, toluene and a mixture of both	3.204-3.779 mg/m^3	18.8-220.2 $ng/m^3/cm^2$ of leaf area/h	100 $\mu mol/m^2/s$ before experiment; unknown during VOC exposure	Day time results were higher in some cases, however removal mechanism not exclusively tested.	Compared removal rates between day and night time.

Zhou et al., 2011	Formaldehyde	15 mg/m^3	2.21-4.60 mg/m^3 over 7 d	Undescribed	Did not identify mechanisms	Used a plantless control treatment consisting of a pot containing only soil.

4.1.4 Problems with static chamber experiments

The vast majority of the knowledge regarding the efficacy of potted-plants to remove VOCs comes from static chamber trials, in which a high concentration of a pollutant is spiked into a small sealed chamber containing potted-plants, with VOC concentrations within the chamber headspace monitored over time (Llewellyn and Dixon, 2011). Given the nature of these trials, generalizing their results to realistic indoor air concentrations in larger rooms has been subject to controversy (Llewellyn and Dixon, 2011; Aini Jasmin et al., 2012). Furthermore, there has been uncertainty regarding how the substrate's active microbial populations will be sustained if exposed to fluctuating concentrations of the VOCs that they catabolize, a situation that is likely in real-world situations (Guieysse et al., 2008). These issues have led to work that has experimented with airflow systems that attempt to expose the microbial population to a constant pollutant flux (Wang and Zhang, 2011). This development has made botanical air cleaning more feasible process for the indoor environment (Wang and Zhang, 2011).

5.0 Indoor plants and the bioremediation of other air pollutants

Indoor plants are capable of ameliorating several indoor air pollutants other than VOCs through several different mechanisms.

In the process of photosynthesis, green plants take up CO_2 through their stomata (Torpy et al., 2014), and thus have an innate potential to reduce ambient CO_2 concentrations. The key limitation to the utilization of this capacity in the built environment, however, is the need for substantial light energy to facilitate meaningful photosynthetic rates. Torpy et al. (2014) found marked differences in the CO_2 removal

rate of different plant species due to different photosynthetic capacities at indoor light levels, with high CO_2 removal rates only being detected at light levels higher than those usually found indoors. When these low photosynthetic rates are coupled with decomposing organic matter that releases CO_2 as well as plant and microbial respiration, it may be unrealistic to expect that a reasonable density of indoor potted-plants will be able to significantly reduce indoor CO_2 concentrations (Llewellyn and Dixon, 2011). It is possible that plants with high VOC removal efficiencies may emit CO_2 as an end product of VOC degradation, thus low CO_2 emitting plants grown under low light conditions need to be screened, or placed in combination with C3 and CAM plant species to limit CO_2 emissions (Treesubsuntorn and Thiravetyan, 2018).While many other gaseous pollutants are known to commonly occur indoors, such as nitrogen dioxide (Lawson et al., 2011), sulfur dioxide, and ozone (Wisthaler and Weschler, 2010), botanical systems have not been widely studied for their potential to reduce these pollutants in indoor environments. Limited work has suggested that the potential for plants to remove these gases through stomatal uptake is realistic, however these pollutants can be harmful to plants, with some species demonstrating high sensitivities (Esguerra et al., 2007; Soreanu et al., 2013).

Relatively few studies have assessed the ability of potted-plants to phytoremediate PM in the indoor environment. Lohr and Pearson-Mims (1996) found that potted-plants were able to accumulate PM through foliar interception, and suggested that plants with rough leaf structures such as trichomes may be more efficient at intercepting PM than smooth-surfaced vegetation. Gawronska and Bakera (2015) showed that the foliage of *C. comosum* was capable of collecting PM across a range of particle sizes, and concluded that more than simple gravitational forces influence PM accumulation on foliage. Neither of these studies, however, measured

515 the effect that this accumulation had on ambient air quality and are thus of limited
516 value as predictors of likely phytoremediation capabilities.
517
518 **6.0 Active botanical biofiltration with functional green walls**
519
520 Although the potted-plant has produced promising results for air quality
521 maintenance, its *in situ* application is constrained by its reliance upon the diffusion of
522 pollutants to permit removal, along with inefficient microbial degradation associated
523 with the very low concentrations of pollutants normally found indoors (Llewellyn and
524 Dixon, 2011). This has led to the development of active botanical biofiltration
525 systems that use active airflow through the plant substrate and across the plant foliage
526 to increase the mass exposure of pollutants to the substrate. These systems normally
527 take the form of active green walls (Darlington et al., 2001; Irga et al., 2017a; Irga et
528 al., 2017b; Pettit et al., 2017). Green walls have advantages over potted-plants due to
529 increased plant density, vertical alignment and the efficiency with which polluted air
530 can be passed through the substrate with the use of mechanically assisted ventilation
531 (Soreanu, 2016). This has led to the possibility of maintaining indoor air quality
532 through the biofiltration of air recirculating within the building rather than through the
533 traditional approach of ventilation through HVAC systems (Chen et al., 2005),
534 however performance development is required to meet this goal. Torpy et al. (2015)
535 suggested that the capacity of biofiltration systems could be enhanced by processing
536 the largest possible volumes of air in conjunction with optimizing the exposure time
537 of the polluted air to the biological material. Thus, the airflow rate, substrate matrix
538 depth and air path tortuosity of active biofilters should influence their pollutant
539 removal efficiency. Supporting evidence came from Darlington et al. (2001), who

found that the single pass VOC removal efficiency of their biofilter was greater at low airflow rates.

The use of active airflow in conjunction with increased plant density allows these systems to simultaneously treat indoor generated gaseous air pollutants as well as filtering PM, a function inherent in the industry standard HVAC PM filters.

6.1 VOC removal by functional green walls

Numerous experiments have tested the performance of microbial air biofilters that do not use plants. In these systems, VOCs are either degraded by the system's microbial community, or become adsorbed onto the substrate (Rene et al., 2017) and consequently, related research has targeted substrate enhancement to allow improved airflow, distribution, retention time and management of the microbial community to maximize VOC filtration (Elmrini et al., 2001; Lee et al., 2013). Lee and Heber (2010) concluded that hydrophilic media with a low specific surface area are disadvantageous for the removal of nonpolar VOCs removal due to poor water retention and minimal sites for microbes. This poses a problem for botanical biofilters, as water is an essential component of the substrate for plant growth. It is likely that the performance criteria of microbial systems will equally apply to active botanical systems, as the airflow assistance will reduce the contact time between airborne pollutants and plant materials, resulting in the activity of these systems becoming strongly influenced by the physiochemical and microbial properties of the media.

In comparison to passive potted-plants, it is likely that the quantitatively significant removal mechanism of active botanical biofilters is through a combination

565 of substrate adsorption and microbial degradation rather than uptake through plant
566 foliage activities (Wang et al., 2014). Similarly to potted-plants however, the plants in
567 active biofilters may support and stimulate the rhizospheric microbial community
568 allowing efficient pollutant removal (Xu et al., 2010). As botanical biofilters require
569 nearly water saturated substrates, biodegradation or adsorption of VOCs is a step-wise
570 process, where a VOC must transfer into the aqueous phase before diffusion to a
571 microbial cell where it is degraded, or to an adsorbent site within the substrate
572 (Darlington et al., 2001; Karanfil and Dastgheib, 2004). It is therefore likely that the
573 degradation of VOCs in the absence of active airflow ('passive' biofiltration) will be
574 limited by each VOCs' Henry's Constant (Guieysse et al., 2008). Active biofilters
575 create a pressure drop across the substrate membrane (Wang and Zhang, 2011; Irga et
576 al., 2017b; Pettit et al., 2017), and this increased pressure should theoretically increase
577 the rate at which VOCs are able to dissolve into the aqueous phase. Although
578 dissolved VOCs will exit the aqueous phase and return to ambient air at a rate
579 determined by each VOC's Henry's Constant, the use of substrates that act as
580 effective adsorbents should promote increased retention within the substrate, thus
581 limiting the rate at which gases simply pass through the aqueous phase. Wang and
582 Zhang's (2011) use of activated carbon as a highly adsorbent substrate component
583 was likely responsible for the substantial single pass removal efficiencies (SPREs) of
584 toluene and formaldehyde (91.7% and 98.7% respectively) demonstrated by their
585 optimized active botanical biofilter. Apart from these findings, substrate development
586 of active botanical biofilters remains a relatively unexplored field, and thus there is a
587 need to critically and comprehensively evaluate biofilter substrates for their capacity
588 to filter both VOCs and PM, as well as their effects on plant health and system
589 practicality, such as cost.

590 *6.2 PM removal by functional green walls*

591

592 The plant growing media in an active botanical system has many of the
593 properties of a filter. Unlike potted-plant systems, where PM removal is limited to
594 deposition on plant foliage, active systems pull air through the plant growth substrate,
595 which can filter out a portion of the PM from the air stream (Irga et al., 2017b). Irga et
596 al. (2017b) and Lee et al. (2015) revealed the potential for active green walls to
597 effectively filter and reduce PM. While the use of highly adsorbent substrates has the
598 potential to improve removal efficiency for VOCs, it is largely unknown how biofilter
599 substrate design affects PM removal. Pettit et al. (2017) revealed that the PM SPRE of
600 active botanical biofilters could be enhanced through appropriate plant species
601 selection, as the different root structures characteristic of each plant species alters the
602 substrate pressure drop properties differentially to influence PM removal efficiency. It
603 is thus likely that the alteration of other substrate properties that influence pressure
604 drop, along with many other physio-chemical characteristics, could affect the PM
605 removal performance of botanical biofilter systems.

606

607 *6.3 CO_2 removal by functional green walls*

608

609 Although it is likely that an impractical number of potted-plants will be
610 needed to offset all CO_2 occupant emissions from most built environment applications
611 (Irga et al., 2013), green walls provide a greater density of plants for a given area of
612 floor space, and thus may provide greater value in this regard. Su and Lin (2015)
613 showed that a 5.72 m^2 indoor plant wall could reduce the CO_2 concentration of a
614 38.88 m^3 room from 2000 to 800 ppm within an hour. Notably, however, each plant's

substrate was covered with aluminium foil to eliminate the effect of substrate respiration, which would not be possible for longer-term plant health, and thus largely negates the practical value of this study. Similarly, Torpy et al. (2016) showed that a 1 m^2 active green wall was capable of significant room CO_2 reductions, but only with the provision of considerable supplementary lighting (250 μmol m^{-2} s^{-1}, whilst indoor light levels typically range between 5–12 μmol m^{-2} s^{-1}: Torpy et al., 2016). While, Torpy et al. (2014) found CO_2 removal could be improved through suitable plant species selection, the most efficient CO_2 sequestering plants identified (*Howea fosteriana* and *Dypsis lutescens*) are not suitable for use in current green wall designs. It is not known whether suitable green wall species can be identified that have the ability to reduce CO_2 concentrations efficiently.

6.4 Other functions

Active green walls have further benefits related to human comfort, for example, in warm climates, temperature reductions of 4–6°C have been observed in proximity to an indoor wall (Fernández-Cañero et al., 2012), showing potential for reduced reliance upon air conditioning leading to possible energy savings. Similarly, Wang and Zhang (2011) predicted that an active botanical biofilter integrated into a HVAC system could reduce energy usage by 25%, while still maintaining equivalent indoor air quality.

Active green walls can remove a range of air pollutants and provide other benefits relating to comfort. Their current removal efficiencies show that their implementation in full-scale rooms has the potential to provide significant benefits for indoor air quality. There is still, however, a need for realistic, *in situ* tests of these

systems, along with an integrated approach to optimise these systems for both improvements in indoor air quality and energy reduction.

7.0 System design

There is range of active botanical biofilters available, each with its own design and claims for air quality remediation. The range of system designs available offers the potential to enhance the technology by selecting the best traits from each system. While all active systems use plants and increased airflow to promote aesthetic appeal and air quality remediation, systems differ with regards to substrate, size, alignment of plants, modularity, as well as water and air supply.

7.1 Substrate

Substrate depth and composition are important metrics of biofilter design, as they are key determinants of the plant growing conditions, required watering regimes, pressure drop, microbial composition and pollutant filtration capacity. While some commercially available systems such as the *AgroSci Aerogation™* green wall system (AgroSci, 2018) make claims about effects on air quality and using an 'engineered soil structure', there is often little information disclosing what substrate is in use.

There is an abundance of research related to the differential capacity of a range of substrates to filter VOCs in non-botanical biofilters, yet it is unknown how plants tolerate such substrates, and furthermore, how these substrates filter out PM. It is, however, known that substrate choice will influence botanical biofilter performance. Darlington et al. (2001) found substantial removal rates of VOCs in a

665 hydroponic system, while Wang and Zhang (2011) found high removal rates of VOCs
666 utilising a substrate consisting of a 50:50 mix (by volume) of activated carbon to shale
667 pebbles. It is thus likely that research that compares the relative performance of
668 different substrates will identify characteristics that will lead to performance
669 development of botanical air filtration systems.
670
671 *7.2 Air supply*
672
673 A number of different airflow orientations exist amongst different
674 commercially available systems, which may have implications for their air cleaning
675 abilities. Only a limited number of these systems have had experimental results
676 published in the literature. Torpy et al. (2017) experimented with the NAVAA ONE
677 system (Naturvention Pty, Jyväskylä, Finland; Figure 1), in which contaminated air is
678 drawn through the planted face of the green wall before flowing vertically upwards
679 through the substrate, thus passing the polluted air stream over a great length of the
680 system's substrate. The treated air then returns to the ambient air via the top surface of
681 the system.

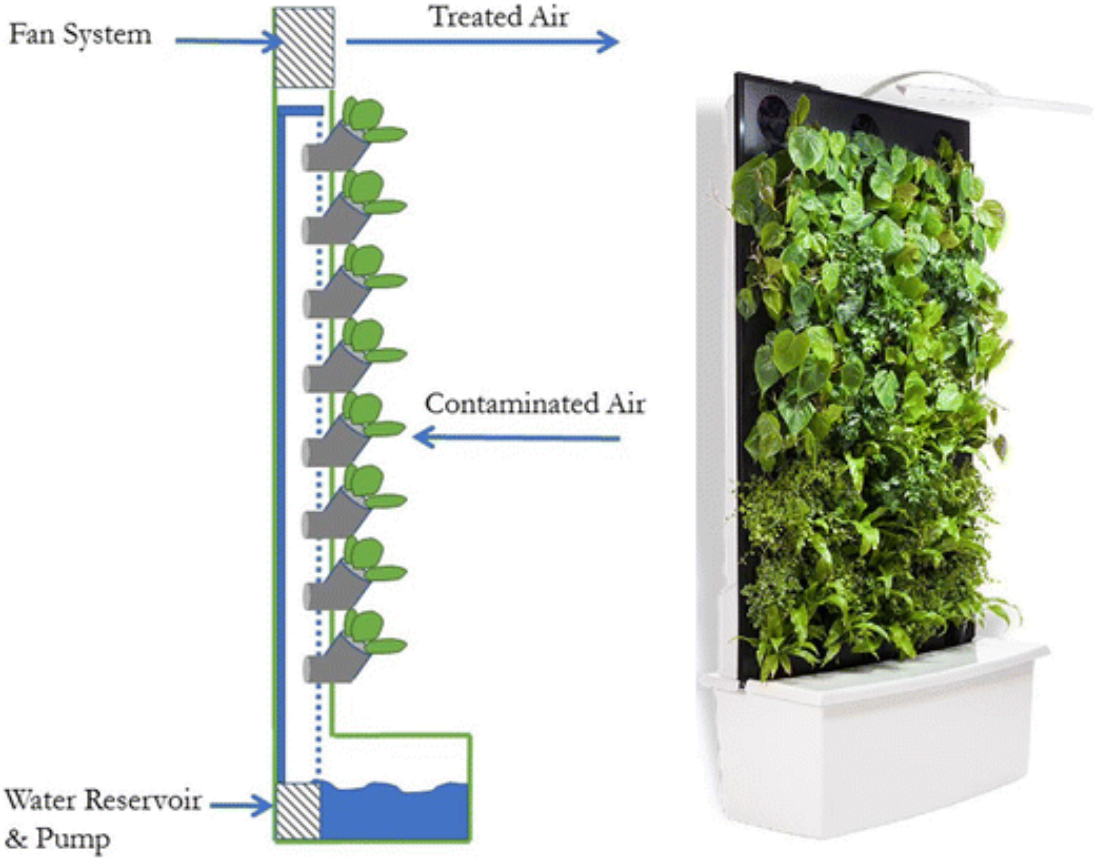

Figure 1. An active botanical biofilter system in which contaminated air is drawn through the planted face and migrates upwards through the substrate before returning to ambient air. Image from Torpy et al., (2017).

Alternatively, other studies have tested systems in which airflow is directed horizontally along the width of the substrate, passing through a duct on the rear side, flowing through the substrate and out through the planted side to ambient air. This application is generally associated with modular systems such as the *Junglefy Breathing Wall* (Junglefy, 2016; Figure 2) and the *AgroSci Aerogation Green Wall* (AgroSci, 2018), which allow designs to be highly customised. Wang and Zhang (2011) and Treesubsuntorn et al. (2017) tested botanical biofilters (Figure 3) in which contaminated air was pulled through the planted surface of a horizontally aligned plant bed before flowing downwards through the substrate and returning to ambient air or an HVAC system. Due to a reduced path length of airflow, these systems likely experience less resistance to airflow and this may enable them to process larger

volumes of air, accompanied with a shorter filtration path length, which may reduce filtration efficiency.

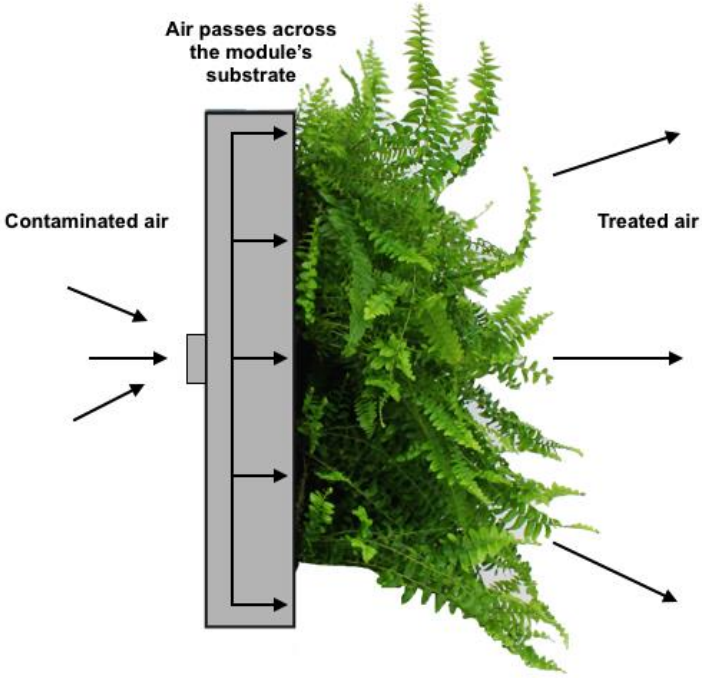

Figure 2. An example of airflow passing through the width of the substrate in an active green wall.

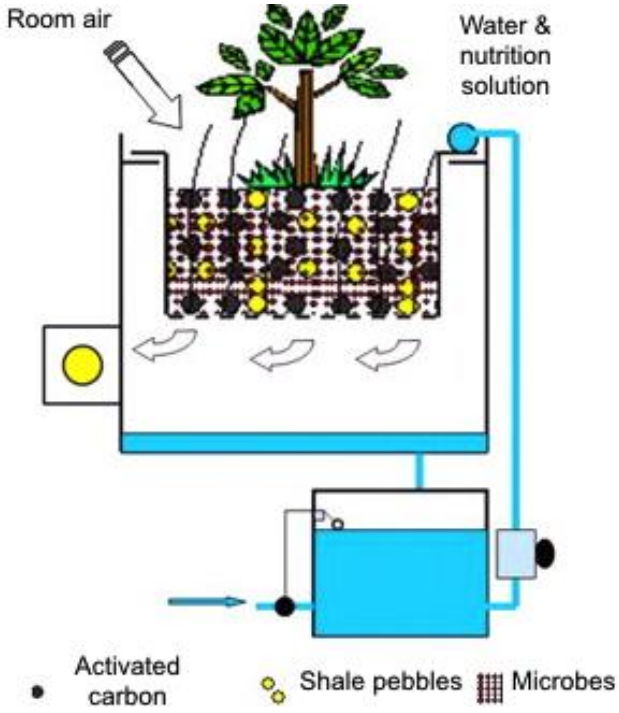

Figure 3 A botanical air filter system in which airflow flows through a horizontal planted surface and downwards through the substrate depth. Image adapted from Wang and Zhang (2011).

A promising innovation is the integration of botanical biofiltration with HVAC systems (Wang and Zhang, 2011), in which HVAC systems distribute clean air while providing an air filtering role, while the air cleaning ability of the botanical biofilter can reduce the load of the HVAC system, as well as removing VOCs and possibly CO_2.

Volumetric airflow rate is another characteristic that has not been compared amongst systems, and this also likely has ramifications for air pollutant filtration. Darlington et al. (2001) found the highest biofilter VOC removal occurred at the lowest tested airflow rate, as this increased the contact time between the polluted air and the substrate. Reduced airflow rates however compromise a system's capacity to process high volumes of air. Although Wang and Zhang (2011) found the lowest single pass removal efficiencies of VOCs at their highest airflow rate, this airflow rate achieved the highest clean air delivery rate (CADR), and thus may be most beneficial when applied *in situ*, depending on energy use.

7.3 Water supply

Water is essential for plant life in botanical biofilters, and watering design and regime may be utilised to influence pollutant removal. Some systems, such as that used by Darlington et al. (2001), incorporate biotrickling water regimes that provide a constant trickle of water that runs down the substrate where it is captured at the bottom and recirculated (Figure 4). Biotricklers allow higher surface volumetric loading rates, and through the provision of constant water, ensure that the water source does not reach the pollutants' saturation point (Guieysse et al., 2008). Another innovation is the use of a combined irrigation and air supply system such as that used

by AgroSci, which uses a wick to provide a constant water supply while air is delivered through a central channel within the wick (Figure 5).

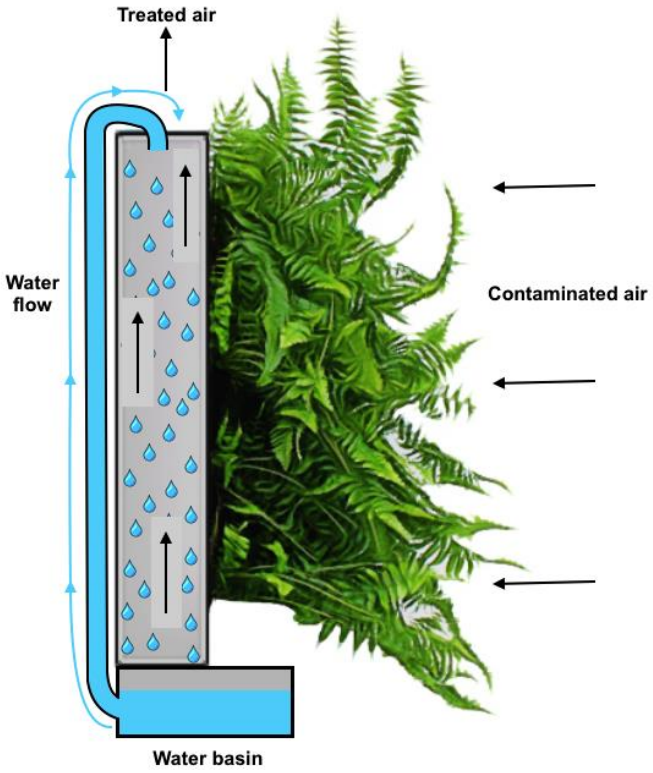

Figure 4. A botanical biotrickler. Within the substrate, contaminated air flows upwards, while a constant supply of water drips downwards where it is caught in a basin.

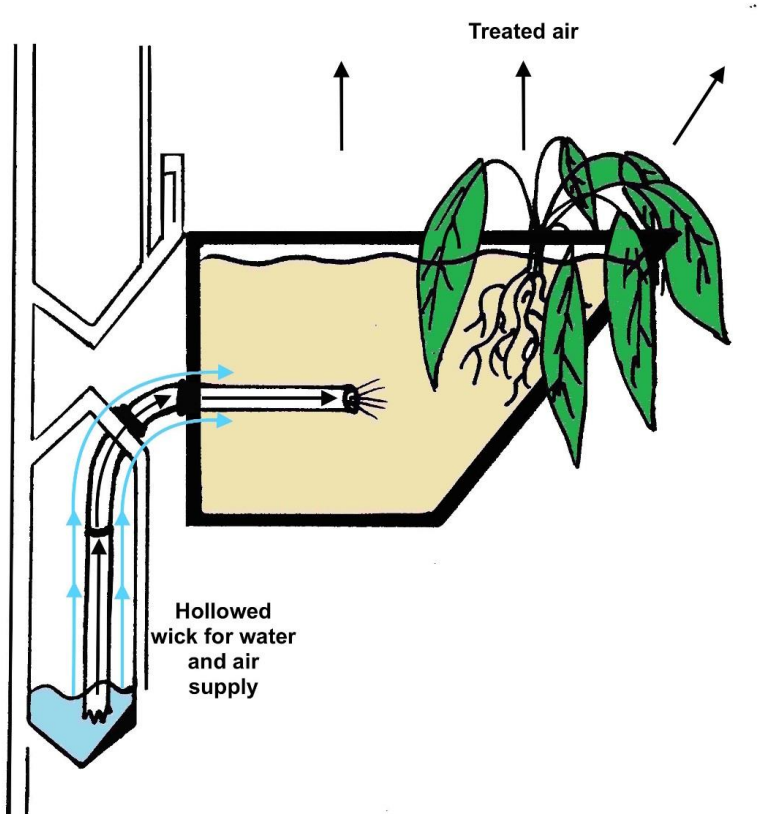

Figure 5. The *AgroSci Aerogation* system that uses a hollowed wick to supply water as needed from a reservoir, while air is supplied through a central channel within the wick.

There may be, however, considerable potential to optimize these systems for air quality improvement: to date there has been little developmental work, with commercial development based almost solely around plant health. Thus potential improvements may relate to airflow rate, position of airflow source and direction of airflow, along with substrate composition and depth, lighting, irrigation regimes, plant selection and management of the microbial population.

8.0 Botanical biofiltration challenges and limitations

One of the most challenging issues associated with the use of biological air filtration relates to the levels of maintenance required for the persistence of healthy plants and their associated microbial populations. The absorption of pollutants into the

substrate and plant tissues may differentially compromise the health of different plant species, and thus in conjunction with species-specific lighting requirements, plant selection may become a crucial aspect for the development of biofiltration technologies (Soreanu et al., 2013). There is limited evidence of plant tolerance to long term air pollution exposure, particularly PM, however limited evidence suggests that relatively short-term (5 weeks) exposure to high concentrations of PM is unlikely to severely affect plant health (Paull et al., 2018). Furthermore, vertically aligned plants may require specialised irrigation regimes due to substrate drainage and increased drying due to airflow across the substrate. While some studies have suggested that certain species are more efficient at phytoremediating certain pollutants than others (Kim et al., 2010; Torpy et al., 2014; Pettit et al., 2017), it is likely that all plant species and their innate microflora have some pollutant removal capabilities, and thus in some cases the capacity of the species to thrive in active botanical biofilter conditions may be a more important consideration than pollutant removal capacity.

The presence of large numbers of plants and aerated, moist substrates clearly presents the potential to increase building relative humidity (Guieysse et al., 2008). Wang and Zhang (2011) noted an increase of up to 18% in their room-sized experimental chamber containing an active botanical biofilter. Increased relative humidity may promote mould formation and deterioration of building materials, and therefore should be kept below 65% (Soreanu et al., 2013), which may be achieved through a balance between appropriate irrigation, air flow rates and substrate selection, as these are all factors likely to influence effluent air humidity. Once again, further research in this area will be required before biofiltration can be effectively used in particular types of buildings and locations.

It has been proposed that indoor plants could act as a significant source of fungal inocula (Staib et al., 1978; Botzenhart et al., 1984; Summerbell et al., 1989; Hedayati et al., 2004; Engelhart et al., 2009), and it is a logical inference that active airflow may promote the emission of fungal spores and bacteria from botanical biofilters into ambient air. However to date, no work has found evidence to support this hypothesis and conversely, Irga et al. (2017a) compared airborne bioparticle densities is offices with active biofilters to those without and concluded that active biofilters are unlikely to make hazardous contributions to indoor fungi. Similarly, experiments conducted by Darlington et al. (2000) and Mallany et al. (2002) both found that botanical biofilters did not increase the concentration of culturable fungal bioaerosols. While bacterial emissions have not been thoroughly studied in active botanical biofiltration, Zilli et al. (2005) found that the bacterial aerosols in the effluent air from laboratory scale biofilters were only slightly denser than those found in the ambient air. The evidence combined thus far suggests that properly maintained active botanical biofilters are unlikely to emit aerosolized fungi or bacteria in concentrations or community compositions that differ from the ambient indoor air.

9.0 Active botanical biofilter experimental design

Although there is significant literature examining the capacity of a range of passive and active botanical biofilters to remediate different air pollutants, the vast majority of these experiments have been conducted on a laboratory scale, generally using small (<1 m^3) sealed chambers and often with unrealistically high concentrations of pollutants. There is therefore difficulty extrapolating these results to a building scale due to the comparatively low pollutant concentration and potentially

reduced diffusion effects found in larger rooms. Due to the wide range of pollutants generally found in buildings, in conjunction with room-specific factors such as moisture, temperature, size, ambient airflows etc., experiments that comprehensively assess the capacity of biofilters to enhance indoor air quality with reproducible testing conditions, controls and independent replication, are difficult to achieve (Guieysse et al., 2008). Furthermore, many studies have focused on assessing the short term or single pass pollutant removal efficiency (e.g. Darlington et al., 2001; Lee et al., 2015; Irga et al., 2017b; Pettit et al., 2017), thus there is a paucity of research relating to the CADR of these systems; although the CADR achieved by Wang and Zhang (2011) provides a promising insight into their potential. Similarly, the short-term experimental approach has largely left long-term effects on plant health unknown.

With numerous active botanical biofilter designs in existence, the use of inconsistent experimental approaches makes it difficult to compare systems. The use of differently sized walls, different VOCs, different doses of pollutants, and different time frames confounds valid comparisons. There is a clear need to standardise experimental procedures to some degree to allow comparisons across studies so different system aspects can be accurately evaluated for the technology to progress.

10.0 Future directions

Whilst the active biofiltration of indoor air is expected to become a growth industry (Torpy et al., 2015), there remains much unknown regarding the value of these systems in the indoor environment for enhanced air quality. Among the current vertical green wall active biofilter systems, there are marked differences in system design, notably direction of airflow. No study is yet to compare the effectiveness of

current designs, and although studies have compared airflow velocities (Darlington et al., 2001), no study has assessed whether airflow direction has any influence on effectiveness.

Current research suggests that the emission of bioaerosols from active biofilters may be negligible (Mallany et al., 2002; Zilli et al., 2005; Irga et al., 2017a), however previous findings have tested biofilters only in their optimal condition. Further work is needed to determine how bioaerosol emissions vary with temperature, airflow, plant species, substrate differences, moisture levels, system age and planting densities (Irga et al., 2017a) before these systems can be widely and safely implemented.

A potentially significant problem when biofiltration is used in full-scale rooms is the rate of pollutant transfer from air to the active components of the biofilter (Irga et al., 2018). The rate of pollutant transfer to biofilter systems has yet to be empirically quantified, and may be affected by different moisture levels, substrates, airflows, and plant types. These combinations will need to be optimised to allow increased airflow and high exposure of pollutant to the biological material, while simultaneously supporting a healthy microbial community capable of significant biodegradation.

To validate the air cleaning potential of active botanical biofilters, *in situ* studies are needed to quantify the effects of the variances in system design and those inherent in indoor room design such as size, layout and natural ventilation effects (Wu et al., 2011). Finally, the gross air remediation capacity of these systems is still largely unknown, and thus their potential for use as standalone air treatment systems, or simply energy reducing adjuncts to HVAC, has yet to be determined.

11.0 Conclusion

Since first recognising the potential of potted-plants to enhance indoor air quality over three decades ago (Wolverton et al., 1984; Wolverton et al., 1985), there has been a progressive increase in research that has measured the air treatment capabilities of the potted-plant, as well extending this capacity in the form of active botanical biofilters. While this work has produced promising findings, and the industry is expected to grow significantly, there is still a need for both further research and a means to increase public awareness so as to promote the value of these systems, before this technology will become widely adopted and implemented in the indoor environment.

Conflict of interests

Declarations of interest: none

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