Does plant species selection in functional active green walls influence VOC phytoremediation efficiency?

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Abstract

Volatile organic compounds (VOCs) are of public concern due to their adverse health effects. Botanical air filtration is a promising technology for reducing indoor air contaminants, but the underlying mechanisms are not fully understood. This study assessed active botanical biofilters for their single-pass removal efficiency (SPRE) for benzene, ethyl acetate and ambient total volatile organic compounds (TVOC)s, at concentrations of *in situ* relevance. Biofilters containing four plant species (*Chlorophytum orchidastrum*, *Nematanthus glabra*, *Nephrolepis cordifolia* 'duffii' and *Schefflera arboricola*) were compared to discern whether plant selection influenced VOC SPRE. Amongst all tested plant species, benzene SPREs were between 45.54–59.50%, with *N. glabra* the most efficient. The botanical biofilters removed 32.36–91.19% of ethyl acetate, with *C. orchidastrum* and *S. arboricola* recording significantly higher ethyl acetate SPREs than *N. glabra* and *N. cordifolia*. These findings thus indicate that plant type influences botanical biofilter VOC removal. It is proposed that ethyl acetate SPREs were dependent on hydrophilic adsorbent sites, with increasing root surface area, root diameter and root mass all associated with increasing ethyl acetate SPRE. The high benzene SPRE of *N. glabra* is likely due to the high wax content in its leaf cuticles. The SPREs for the relatively low levels of ambient TVOCs were consistent amongst plant species, providing no evidence to suggest that *in situ* TVOC removal is influenced by plant choice. Nonetheless, as inter-species differences do exist for some VOCs, botanical biofilters using a mixture of plants is proposed.

Keywords Air pollution; botanical biofilters; phytoremediation; green walls; VOCs

Introduction

Air pollution in urban environments is a growing concern, with exposure directly linked to seven million deaths globally in 2012 (WHO 2014). One major component of urban air pollution is volatile organic compounds (VOCs), which include a range of organic compounds that quickly vaporise at room temperature, such as ethyl acetate (EtOAc) and benzene. VOC exposure can be damaging to human health, with complex and long-term compounding effects that are difficult to treat (Deng et al. 2015, Deng et al. 2017).

Given increasing pollution concentrations and population densities in metropolitan areas, there is a growing need to develop methods to maintain habitable urban living environments (Irga et al. 2018). Urban developers are adopting sustainability frameworks that require the employment of strategies to limit or mitigate pollution, and demonstrate a positive impact on the environment (De Valck et al. 2019). As social and technological changes are leading to increases in the proportion of time individuals spend in indoor environments (Vardoulakis et al. 2015), the quality of indoor air is becoming an increasingly important health factor. Indoor air quality is maintained primarily by heating, ventilation and air conditioning (HVAC) systems, which have variable control over the indoor atmospheric chemosphere (Irga and Torpy 2016).

The combined application of biotechnology, environmental engineering and horticultural science has led to the development of biological air filters as a promising avenue of research for the bioremediation of indoor air (Soreanu 2016). These systems use natural bioagents (plants and/or microorganisms) to remove pollutants from the air through an aerobic process, where the pollutants act as energy, carbon, and other nutritional sources for the bioagents, or are otherwise absorbed into or adsorbed on to the biological materials (Wei et al. 2017). Additionally, these systems can use substrates containing a proportion of activated carbon and a range of other materials, to assist with pollutant filtration or substrate microbial growth (Pettit et al. 2018, Torpy et al. 2018). The efficiency with which biofilters can filter out and degrade VOCs from indoor air indicates that they may be used to reduce inhabitant pollutant exposure (Sriprapat and Thiravetyan 2013, Wolverton et al. 1984, Wood et al. 2006, Brilli et al 2018).

While static systems such as pot plants have been found to be ineffective for high capacity contaminant removal (Llewellyn and Dixon 2011), research indicates that 'active green walls', which utilise mechanical assistance to funnel air into the biofilter substrate, improves their bioremediation efficiency to the extent that functional air remediation is probable (Torpy et al. 2015). These systems may also be practical for large infrastructure use, given that they are accessible, robust, cost-effective and have a low-energy footprint. Although the available types of botanical biofiltration systems differ in design, they all use active airflow facilitated with devices such as impellers that increase the airflow across or through the systems and therefore allow larger volumes of air to be processed by the biofilter. Whilst there is a growing body of literature that demonstrates the air pollutant remediation capabilities of this technology (Pettit et al. 2019), to date, the potential for plant selection to enhance botanical biofilters ability to filter some of the more dangerous air pollutants is required.

Plant selection is known to have an influence on VOC removal efficiency for static, potted-plant systems (e.g. Kim et al. 2010). Whilst the nature of the plant characteristics that determine these effects have yet to be resolved, there may be phylogenetic associations where certain groups of plants are more effective for the removal of certain forms of VOC (Kim et al. 2016). Whilst it has been shown that rhizospheric bacteria are the major agents of removal for some VOCs (eg. Wood et al. 2002), there are clearly plant-associated effects that may or may not (Irga et al. 2017) interact with the substrate microbial community, or even subsume its activity for specific VOCs, as is the case for CO_2 removal (Pettit et al. 2017). An alternate hypothesis is that different plants can affect the abiotic chemical or physical properties of the substrate such that VOC removal is altered (Deng and Deng 2018). Despite many years of research, these patterns have yet to be resolved, and thus objective decisions on the most effective plant species for VOC biofiltration cannot be made.

Previous work that has tested the removal of multiple VOCs has usually tested pollutants with similar physio-chemical properties, for example numerous studies have assessed VOCs focusing on benzene, toluene, ethyl benzene and xylene (BTEX). Darlington et al. (2001) compared the biofilter removal rates of toluene, ethyl benzene and *o*-xylene, finding that all compounds had similar removal rates, and suggested that the limiting factor that affected VOC removal rates was transfer of gaseous pollutants to the liquid phase, rather than microbial degradation. However, for higher concentrations of more soluble compounds, which are easily absorbed into the liquid phase, the rate of microbial degradation may be the primary limiting factor (Pettit et al 2018).

The current study investigates a range of common green wall plant species in an active botanical biofilter to elucidate the influence of plant type on VOC removal efficiency. This is the first study that compares the SPRE of VOCs with the explicit aim to identify the most efficient plant wall species for active green wall VOC biofiltration. Further, assessments were made to determine the correspondence between VOC filtration efficiency and a range of plant and substrate characteristics so as to identify traits that may be associated with increased VOC removal rates.

Methods

Active living wall biofilter design

The current study assessed an active green wall system previously described in Irga et al. (2017; Figure 1). Briefly, the system utilizes assisted aeration by incorporating an axial impeller to both increase gaseous pollutant exposure to the substrate and plant rhizosphere, and to allow for particulate matter removal, which is filtered through the substrate. The system is modular, allowing for flexibility in upscale design, with module dimensions of 500 x 500 x 130 mm, with 16 circular compartments for plant insertion. The module is constructed from polyethylene and contains a coconut coir based substrate. When operational, air is drawn into the system, and flows through the plant substrate matrix (25 L total substrate volume), contained within a tight weave high density polyethylene (HDPE) bag – typically used as shade cloth, and returned to the environment through the planted surface. Total airflow through the 0.25 m^2 front surface area test system was 14.90 L/s (Abdo et al. 2016). This green wall system has been previously demonstrated to be effective in the removal of VOCs, CO_2 and particulate matter in laboratory trials (Pettit et al. 2017).

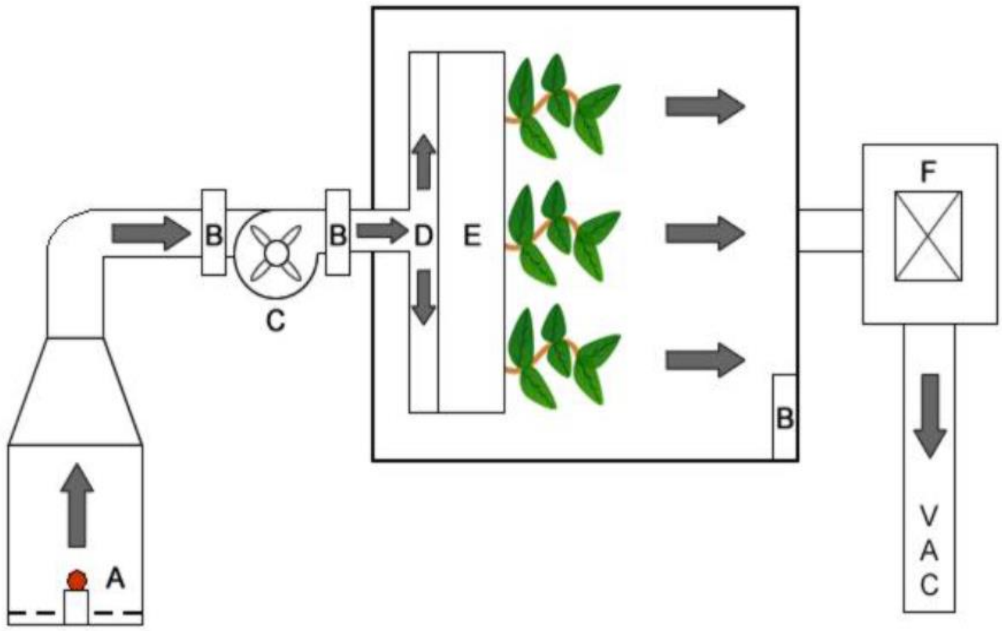

Fig. 1. Schematic of the single pass removal efficiency test apparatus set up: A Combustion chamber; B Digital pressure differential sensor; C Axial impeller; D Plenum within system module; E Biofilter packing medium; F Photoionisation detector; VAC Exhaust vacuum pump.

Plant materials and measurement of plant morphological traits

Four plant species were selected for this study (Figure 2), which are all commonly used vertical biowall plants, that encompass a range of phylogenetic, physiological and morphological variability. These plants were selected as they have previously been used in biowalls for phytoremediation assessments (Pettit et al. 2017). The four species tested were: (i) *Chlorophytum orchidastrum* Lindl. (Fire flash), a monocot; (ii) *Nematanthus glabra* Bailey. (Goldfish plant), a eudicot; (iii) *Nephrolepis cordifolia* (L.) C. Presl. var. ‘duffii’ (Lemon button fern), a monilophyte; and (iv) *Schefflera arboricola* Hayata. (Dwarf umbrella tree), also a eudicot. When not being tested, all plants within their green wall modules were maintained in a glasshouse lined with shade cloth, with an average temperature of 23.7 ± 3.6°C, relative humidity of 68.1 ± 16.0%, and a maximum mid-day light level of 90 ± 10 $\mu mol.m^{-2}.s^{-1}$ (4860 ± 54 lux). Plants were allowed to develop for 8 months under glasshouse conditions after planting and prior to testing. All modules were watered once weekly to saturation, as per industry standards.

Fig. 2. Green wall plant species tested in the current experiment. From top left to right; *Chlorophytum orchidastrum* (Fire flash), *Nematanthus glabra* (Goldfish plant), *Nephrolepis cordifolia* (Lemon button fern), and *Schefflera arboricola* (Dwarf umbrella tree).

At the conclusion of the VOC removal trials, plants were carefully removed from the biofilter. The substrate was gently washed from the plants, and the plants were assessed for several morphological characteristics that could have influenced VOC removal efficiency, with four replicates per plant trait.

Average root diameter was recorded using callipers, by taking four composite measurements from each plant, from four plant replicates per species. Root and leaf mass fresh weights were recorded with scales. Root surface areas were determined by creating plant pressings of the samples between two sheets of clear Perspex. Images of each pressing were taken with a camera (Canon 1100D, 18 mm lens, Canon Australia Pty Ltd, Macquarie Park, Australia) placed ~100 cm vertically above the Perspex sheets containing the leaves and roots. Image analysis software (Fiji Image J 1.50g; National Institutes of Health, Bethesda, Maryland, USA) was utilised to measure root surface area by multiplying the two-dimensional root surface area by π. Leaf area was determined using portable leaf area machine (Licor LI-3000-A, Lincoln, Nebraska, USA).

Plant morphological data is shown in Table 1. For all variables, there were substantial variations in morphology amongst the test species.

Table1. Plant morphological data of each plant species. (Means ± SEM, n =4).

Species	*Chlorophytum orchidastrum*	*Nematanthus glabra*	*Nephrolepis cordifolia*	*Schefflera arboricola*
Root diameter (mm)	3.55±0.32	0.52±0.06	0.68±0.07	3.95±0.79
Root mass fresh weight (g)	13.71±1.51	1.22±0.61	3.27±0.93	16.01±3.36
Leaf mass fresh weight (g)	26±3.34	30.22±16.64	214.42±46.2	245.6±53.81
Root surface area (cm^2)	150.8±20.01	63.71±28.45	6.92±0.35	33.6±4.31
Leaf surface area (cm^2)	731.17±229.5	255.6±121.9	15±0.65	255.63±1.75

Experimental set up

Experiments used a flow-through chamber system to assess the SPREs of benzene, ethyl acetate and TVOCs. This set up used a sealed Perspex chamber (0.6 X 0.6 X 0.6 m; 216 L), of which one of the sides could be removed and resealed using adhesive foam rubber and adjustable metal clamps, thus allowing green wall module placement in the chamber. Ducting was fixed to one side of the chamber, which led to a second chamber in which air pollutants were generated. The generated air pollutants flowed through the fitted ducting with active airflow provided by a 16 W fan housed within the ducting, before flowing through the biofilter. A second fan within the sealed Perspex chamber encouraged mixing of the chamber atmosphere, creating a homogenous concentration of pollutants within the chamber, before exhaust into another ducting system on the opposite side of the chamber. This led to a third chamber that housed a photo-ionisation detector (PID) that was used to monitor the concentration of air pollutants. Air was exhausted to waste through a vacuum exhaust after sampling.

Single pass removal efficiency represents the percentage of a VOC removed from the air stream as it passed through the biofilter, relative to the control treatment. In order to determine the removal efficiency for each VOC, all trials were run independently, i.e. with a single pollutant per run, with three replicates per treatment. Gaseous ethyl acetate (EtOAc) and benzene were chosen to assess how the system comparatively treats hydrophilic VOCs (EtOAc: solubility at 25°C = ~80.3 g/L) and hydrophobic VOCs (benzene: solubility at 25°C = ~1.71 g/L). Each VOC was generated by placing 4.0 mL of the liquid chemical into a 10 mL sealed glass vial and extracting 2.5 mL of the vapour-saturated headspace with a gas chromatograph plunger-in-needle style syringe, which was then injected into the pollutant generation chamber of the flow through system. This process produced a pulse of VOC through the flow through duct. The benzene treatment was thus a ~10 minute pulse, reaching a peak concentration of 4.170 ± 0.144 ppm ~60 seconds after injection. The EtOAc treatment generated a ~8 minute pulse, reaching a peak concentration mean of 3.997 ± 0.074 SD ppm, 45 seconds after injection. Additionally, the system was tested for the SPRE of ambient total VOCs (TVOCs) using laboratory air supplied by the building's HVAC system, thus reflecting the usual concentration of TVOCs in the room's normal operational state (~35 ppb). Given that this experiment was performed in a general use research laboratory, the TVOC concentration would be expected to be greater than that experienced in most other building types. The concentration of the effluent gas was monitored with a PID (ppbRAE 3000, RAE Systems, San Jose, CA, USA), with corrections applied as per the manufacturer's instructions for the two VOCs.

Blank data (chamber with no green wall module present) for all VOC treatments was also collected and used to calculate the background VOC removal efficiency of the flow through system. The empty flow-through chamber was exposed to the stream of gaseous VOC with identical concentration and flow conditions as the biofilter trials. Calculations of specific VOC and TVOC removal efficiencies were thus based on measurements at the same sampling point in the system with or without the biofiltration system present.

Data analysis

After the data was checked for normality with Kolmogorov–Smirnov test and checked for homogeneity of variance with Levene's test; one factor ANOVAs followed by Tukey's *post hoc* tests (IBM SPSS Statistics Version 21, IBM Corp, Armonk, NY, USA) were conducted to compare the SPREs of EtOAc, benzene and TVOCs amongst the different plant species. The presence and strength of the relationship between benzene removal and EtOAc removal across treatments was examined by computing the Pearson correlation coefficient. Further, statistical associations between plant morphological traits across plant species and pollutant removal were also tested with Pearson correlations.

Results

EtOAc and benzene removal rates for each plant species are shown in Figs. 3 and 4. Ambient indoor TVOC removal is shown in Fig. 5. VOC removal was achieved in all treatments, across all plant species.

The system when tested for EtOAc removal recorded removal efficiencies in the range of 32.36–91.19%. EtOAc removal efficiencies were 39.97 ± 5.17% for *N. glabra*, 80.69 ± 5.97% for *S. arboricola*, 64.02 ± 1.06% for *N. cordifolia*, and 82.61 ± 5.97% for *C. orchidastrum*. Significant differences in EtOAc removal were observed among plant species tested (df 3,8 $F=14.19$, $P=0.001$), with *C. orchidastrum* and *S. arboricola* recording significantly higher EtOAc SPREs than *N. glabra* and *N. cordifolia* (Figure 3, $P<0.05$ for all differences mentioned).

The system when tested for benzene removal recorded SPREs between 45.54–59.50%. Benzene single pass removal efficiencies were 58.78 ± 1.07% for *N. glabra*, 51.01 ± 3.03% for *S. arboricola*, 48.00 ± 2.19% for *N. cordifolia*, and 47.65 ± 1.46% for *C. orchidastrum*. Significant differences in benzene removal were observed among the plant species tested (df 3,8 $F=18.61$, $P=0.001$), with *N. glabra* recording significantly greater benzene SPRE than the other plant species (Figure 4, $P<0.05$ for all comparisons).

When comparing ambient TVOC removal efficiencies, no significant differences amongst plant species were observed (Figure 5, df 3,8 F=0.01, P=0.998).

Plant species that were efficient for EtOAc removal demonstrated lower efficiency for benzene removal, and *vice versa*. When removal efficiencies were combined across species, benzene removal rates were significantly negatively correlated with EtOAc removal (r=–0.688, P=0.013).

EtOAc SPRE was significantly positively correlated with root surface area (r=0.694, P=0.005), root mass (r=0.666, P=0.005), root diameter (r=0.479, P=0.05), and leaf area (r=0.664, P=0.005). No associations were found between EtOAc SPRE and leaf mass.

Benzene SPREs were not positively associated with any of the plant traits measured, however they were significantly negatively correlated with root surface area (r=–0.699, P=0.003), root mass (r=–0.318, P=0.036) and root diameter (r=–0.479, P=0.05). No significant associations were found between benzene SPRE and any leaf traits.

It should be noted, whilst many correlations between VOC removal and plant traits were statistically significant, the values of the correlation coefficients obtained were moderately low, with none exceeding r = 0.7.

As no significant differences amongst plant species were observed in TVOC removal efficiencies, no plant trait associations were tested with this treatment.

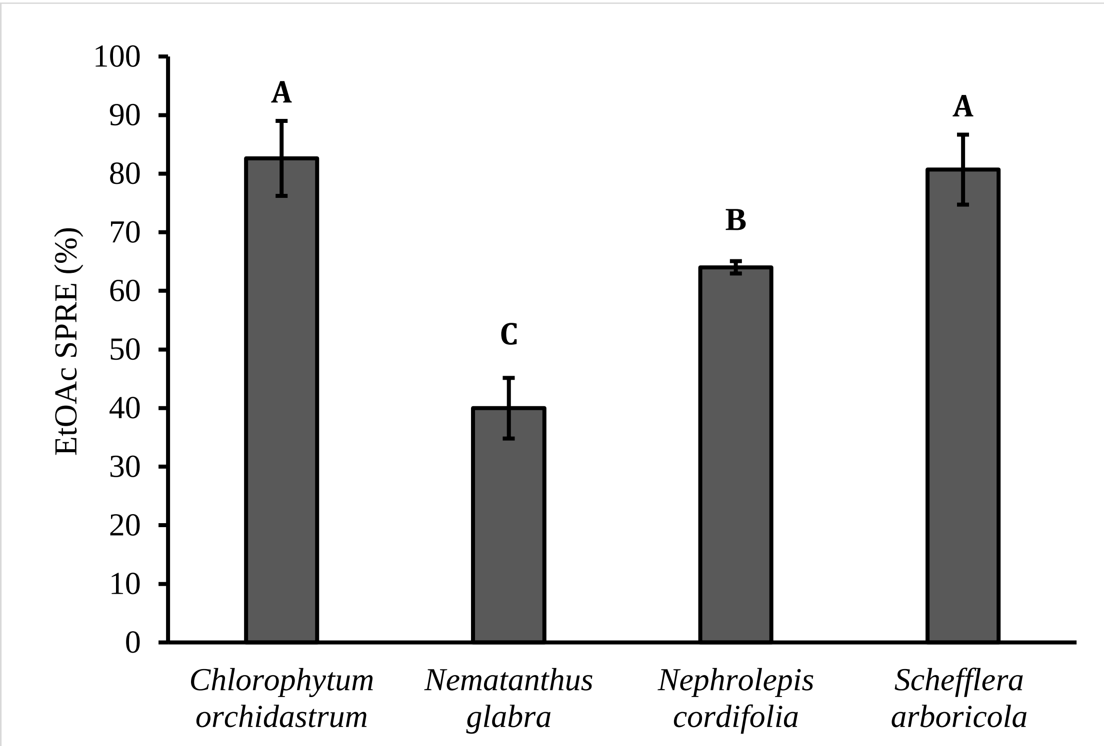

Fig. 3. Average levels of EtOAc removal across plant species (Means ± SEM, n =3). Species sharing the same letter are not significantly different using Tukey's post hoc test, P>0.05.

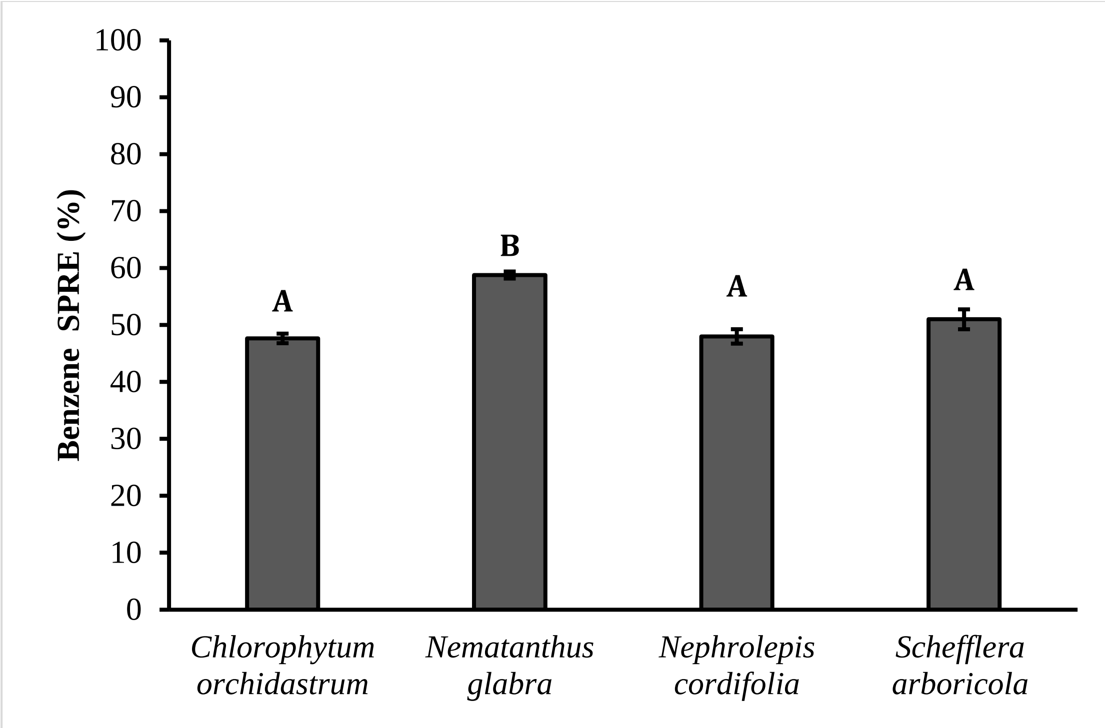

Fig. 4. Average levels of benzene removal across plant species (Means ± SEM, n =3). Species sharing the same letter are not significantly different using Tukey's post hoc test, P>0.05.

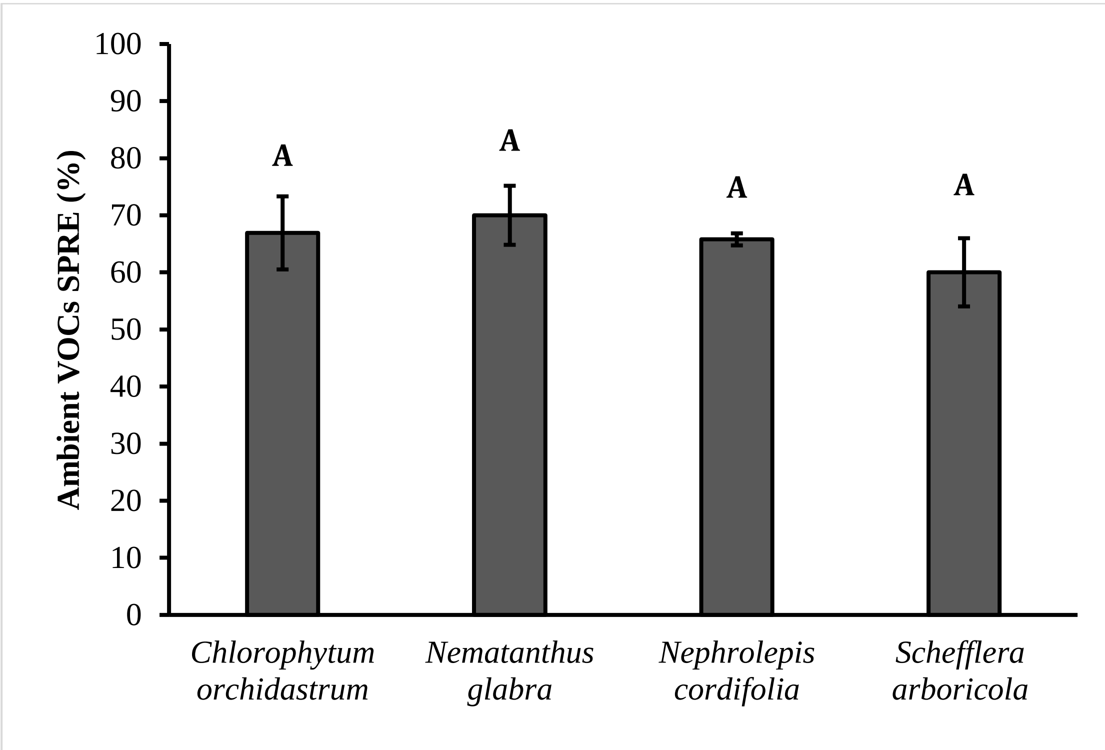

Fig. 5. Average levels of ambient indoor VOC removal across plant species (Means ± SEM, n =3). Species sharing the same letter are not significantly different using Tukey's post hoc test, P>0.05.

Discussion

If active green walls are to be used as functional air cleaning devices, the VOC removal efficiency of these systems must be developed so as to provide maximized air cleaning efficiency, whilst minimising additional energy use. To date, there is a scarcity of literature on the relative contribution of the botanical component to the overall VOC filtration ability of these systems. The primary observation of the current work is that plant type does influence the system's capacity for VOC removal. It is well established that potted plants can effectively improve indoor air quality by reducing hazardous VOCs (Ugrekhelidze et al. 1997, Kim and Jeon 2009, Sriprapat and Thiravetyan 2013, Dela Cruz et al. 2014, Kim et al. 2016, Hörmann et al. 2018). However, it is known that the efficiency of VOC removal varies substantially both among plant species (Kim et al. 2018, Yoo et al. 2006), and with the molecular characteristics of individual compounds. The current work extends this understanding to active botanical biofilters, where less botanical influence might be expected due to the reduced VOC residence time within the biological components of these systems.

Given that increasing root surface area, root diameter and root mass were all associated with increasing EtOAc removal, it is proposed that the plant roots may provide hydrophilic adsorbent sites for EtOAc, or facilitate high microbial activity in the substrate stimulated by root exudates acting as nutrients for soil microorganisms (Kim et al. 2018). Alternatively, it is possible that root exudates may alter the chemical composition of the rhizosphere and thus influence the capacity of specific VOCs to adsorb to the substrate (Pettit et al. 2018). Furthermore, with the substantial air flow inherent in active biofiltration, certain root morphologies may enable increases in exposure at the substrate/root/pollutant interface; potentially elevating the EtOAc removal capacity. Additionally, it is plausible that the aerial plant parts could influence EtOAc removal, with both the stomata and cuticle on the leaves creating pathways for VOC removal, as has been proposed in previous work (Gkorezis et al. 2016, Jindachot et al. 2018), and observed in the current study in the positve correlations between leaf surface area and EtOAc SPRE. However, EtOAc is a relatively hydrophilic VOC, and therefore will not diffuse readily through the cuticle due to its waxy nature, and thus may largely be taken up through the stomata when they are open. This proposal is supported by the current data, as *N. glabra* would not have been capable of stomatal activity during the day as it is a CAM plant, and thus only opens its stomata in the dark, a strategy that has evolved to limit moisture loss (Paull et al. 2018). In any case, the correlations detected between EtOAc removal and plant characteristics in the current study provide evidence that the belowground components of plants are the major regulator of VOC removal in active air phytoremediation systems.

The benzene removal data was reasonably consistent across plant species, with less than 15% variability between the most and least effective plants. In contrast to the EtOAc removal trials, the most effective plant species for benzene removal was *N. glabra*, likely due to the high wax content in its leaf cuticles, although stomatal benzene uptake has also been proposed in previous work (Setsungnern et al. 2017). Alternatively, *S. arboricola* has been shown to have high benzene removal efficiency, which has been previously attributed to its relatively large leaf area (Parseh et al. 2018), along with a significant waxy cuticle comprised of alpha-linoleic acid and dodecyl cyclohexane (Treesubsuntorn and Thiravetyan 2012). Interestingly, in this study, no significant associations were found between benzene SPRE and any leaf traits, and thus we cannot determine the pathway for benzene removal observed for *N. glabra* in the current work, nor can we eliminate effects that this species may have had on the substrate as the means by which enhanced benzene removal was afforded. It is recommended that in future work, quantitative assessments of leaf hydrophobic compounds, such as waxes, be made to determine whether they have a major effect on hydrophobic VOC removal. Work using substrates in which various plant species have been grown, but subsequently removed, would also be of value to elucidate plant-mediated substrate effects on VOC removal.

As this experiment assessed SPRE, the removal of each chemical was dependent upon its residence time within the active green wall system (i.e. the time that the polluted air stream was in contact with the growth substrate and plant foliage). Due to the limited residence times in these single pass experiments, it is likely that the removal processes in these trials were predominantly sorption process as opposed to microbial degradation processes. Whilst several static chamber studies have found that the potted-plants' microbial community plays a significant role in VOC removal (Aydogan and Montoya 2011, Orwell et al. 2006), the very short residence time of pollutants in the current trials (<10 min in all cases) would probably limit the time available for microbial metabolism to occur. Mikkonen et al. (2018) observed a decrease in a green wall system's microbial diversity after it had been exposed to VOCs for 16 weeks, as heterotrophic bacterial groups that could use the VOCs as a nutrient source had been favorably selected and became numerically dominant in the community. Whilst it is thus possible that prolonged exposure to VOCs would increase the bacterial community's VOC degradation capacity, this effect may not affect an active green wall's *in situ* VOC air cleaning efficiency to the same degree, due to the short pollutant residence time (Weyens et al. 2015).

It is clear from the current findings that plant selection will effect VOC removal in active botanical biofilter systems. This may be of value in functional biofilter design, especially if hydrophilic VOCs are problematic in a specific application. The general *in situ* importance of these effects may, however, not be of great magnitude, as all plants tested has considerable VOC removal efficiencies; thus a biofilter of adequate size relative to the concentration of VOCs encountered should lead to major VOC reductions, irrespective of the performance of the individual species selected.

A further consideration in plant selection relates to long-term VOC effects on plant health. Whilst several common green wall plants have been shown to have excellent short term / high concentration pollutant tolerance (Paull et al. 2018), there have yet to be long term trials pollutant exposure trials. The use of active airflow through the plants' substrate may have unexpected effects on plant health, and these conditions must be tested as a key contributor to the whole-of-life costs of botanical pollutant removal systems. Similarly, substrate changes over long-term exposure remain unknown, beyond those effects related to microbial community shift, previously described.

A potential solution for the observed variability in VOC removal amongst plant species is through green wall design, with targeted combinations of different species growing together. Uniform plant types in large scale green walls are rarely encountered, thus the combined removal efficiencies of biodiverse green walls could be used to account for a diversity of VOCs. However, with the introduction of active air flow through green wall systems, very little is known about the potential influence this will have on VOC degrading bacteria in the substrate, or whether they play an important role in VOC degradation, as has been shown to be the case in static systems (Wood et al. 2002). It is thus proposed that experiments using radiolabelled VOCs will be required to test the role of rhizospheric bacteria to utilize and degrade airborne VOCs in constant air flux conditions.

Summary and conclusion

There is a growing body of evidence indicating that botanical biofiltration has major potential for the low energy use removal of a broad range of air pollutants. However the physical, chemical and biological functions of these systems remain poorly described, and thus evidence supporting the design criteria for tailoring or maximising pollutant filtration efficiency is still weak. The current work assessed the capacity of several common green wall plants for removing two major classes of VOC providing a baseline indication of the plant species' removal efficiencies for model hydrophobic and hydrophilic VOCs. The findings suggest that target pollutant dependent botanical biofilter plant selection are possible, as whilst all plant species were successful in removing ambient TVOCs and benzene, there were substantial differences between species in hydrophilic VOC removal. The authors propose that future work should examine plant effects on biofilter substrates to determine the specific physical, chemical or biological processes that are associated with VOC removal.

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