The *in situ* pilot-scale phytoremediation of airborne VOCs and particulate matter with an active green wall

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Abstract

Atmospheric pollutant phytoremediation technologies, such as potted-plants and green walls, have been thoroughly tested in lab-scale experiments for their potential to remove air pollutants. The functional value of these technologies, however, is yet to be adequately assessed *in situ,* in 'high value' environments, where pollutant removal will provide the greatest occupant health benefits. Air pollution in countries such as China is a significant public health issue, and efficient air pollution control technologies are needed. This work used pilot-scale trials to test the capacity of potted-plants, a passive green wall and an active green wall (AGW) to remove particulate matter (PM) and total volatile organic compounds (TVOCs) from a room in a suburban residential house in Sydney, Australia, followed by an assessment of the AGW's potential to remove these pollutants from a classroom in Beijing. In the residential room; compared to potted-plants and the passive green wall, the AGW maintained TVOCs at significantly lower concentrations throughout the experimental period (average TVOC concentration 72.5% lower than the control), with a similar trend observed for PM. In the classroom, the AGW reduced the average TVOC concentration by ~28% over a 20 min testing period compared to levels with no green wall and a filtered HVAC system in operation. The average ambient PM concentration in the classroom with the HVAC system operating was 101.18 $\mu g/m^3$, which was reduced by 42.6% by the AGW. With further empirical validation, AGWs may be implemented to efficiently clean indoor air through functional reductions in PM and TVOC concentrations.

Keywords: active green wall; botanical biofilter; living wall; indoor air quality; potted plant; green infrastructure.

1. Introduction

The indoor air quality of urban non-occupational environments, such as residences, schools, child-care facilities and nursing homes, is becoming an important public health issue, as populations susceptible to health effects from air pollutant exposure, such as children and the elderly, spend a considerable amount of time within these settings (Al-Hemoud et al. 2018). Urban areas are often associated with poor air quality (Gulia et al. 2015; Han et al. 2014), as the activities in these areas promote the generation of airborne pollutants, primarily particulate matter (PM) (Guo et al. 2010; Morawska and Clark 2000), which can penetrate and contaminate the urban indoor environment (Perez et al. 2016). Additionally, a range of common household and office materials and products, such as building materials, furnishings, plastics and solvents can emit volatile organic compounds (VOCs) (Aini Jasmin et al. 2012; Cruz et al. 2014a); thus allowing the potential for these pollutants to accumulate within the indoor environment (Weschler 2009). For many VOCs, such as benzene and poly-aromatic hydrocarbons, the World Health Organisation recommends no safe level of exposure (World Health Organization 2010). High concentrations of VOCs are most commonly treated by flushing them from the indoor environment with outdoor air from which a proportion of the PM is filtered as it enters the building. This approach is problematic in urban areas with highly polluted outdoor air, as filter efficiency is highly variable, especially for small particles (Ren et al. 2017).

While poor air quality remains a global issue (World Health Organization 2014), cities within China have experienced unprecedented urban growth in terms of scale and speed. This has led to a corresponding decline in air quality across many of China's cities (Liu et al. 2018), where indoor air pollution mitigation strategies have focused on PM reduction primarily through ventilation filtration technology integrated within heating, ventilation and air conditioning (HVAC) systems (Liu and Liu 2005). Despite the considerable energy that this technology requires (Liu et al. 2017), the success of this approach has been limited (Ren et al. 2017). Additionally, in situations with limited ventilation rates, VOC concentrations are often problematic, as VOC concentration has been shown to negatively correlate with building ventilation (Cheng et al. 2016). Thus, the prevalence of high concentrations of VOCs is becoming an important public health issue in China (Clean Air Alliance of China 2017).

Within China, schools represent a quantitatively important indoor environment, with >90 million students across more than 250,000 primary schools (Hou et al. 2015). Children are highly susceptible to the adverse health effects from air pollutant exposure due to their relatively higher ventilation rates and immature immune systems (Buka et al. 2006). In addition to direct health effects, poor indoor air quality affects student learning performance (Bakó-Biró et al. 2012). Mechanical ventilation systems are not commonly used in public primary schools in China and consequently ambient outdoor particles and other outdoor air pollutants are introduced into the indoor environment as 'fresh air' is brought in through natural ventilation to flush out indoor generated VOCs (Peng et al. 2017). It is thus clear that comprehensive indoor air cleaning technologies that can reduce the high concentrations of ambient particles and VOCs in an energy efficient manner will have high public health and environmental value.

As a possible solution to mitigate poor indoor air quality, a large body of research has tested the capacity of potted-plants to clean VOCs from the indoor environment (Aydogan and Montoya 2011; Cruz et al. 2014a; Cruz et al. 2014b; Hörmann et al. 2017; Hörmann et al. 2018; Irga et al. 2013; Orwell et al. 2004; Sriprapat et al. 2014; Sriprapat and Thiravetyan 2013; Teiri et al. 2018; Treesubsuntorn et al. 2013; Treesubsuntorn and

Thiravetyan 2012; Wood et al. 2002). The use of plants for indoor air remediation offers an economical and sustainable departure from conventional techniques such as adsorption filters, photocatalytic oxidation purifiers, and ozone generators, that are often expensive, remove a constrained range of VOCs, and can produce harmful by-products (Irga et al. 2018). However, the existing experiments on potted-plant VOC removal have most commonly been limited to laboratory-scale chambers, and despite the high VOC removal rates documented in these studies, it has been proposed that their removal rates *in situ* may be of lower practical value (Irga et al. 2013; Llewellyn and Dixon 2011), as the pollutant removal rate is dependent upon the rate at which polluted air can diffuse to the active components of the potted-plant microcosm.

Active botanical biofiltration involves the application of active airflow, through mechanisms such as low power fans, to draw polluted air towards the plant's foliage and substrate. When applied in a green wall format, it is likely that the VOC removal rates of these systems will be significantly higher than those of potted-plants due to the increased rate at which pollutants can be delivered to the system and the increased planting density per unit of floor area possible with these systems (Torpy et al. 2015). Several studies have tested the single pass removal efficiencies (SPREs) of these systems to remove PM (Irga et al. 2017b; Pettit et al. 2017) and a range of VOCs (Darlington et al. 2001), with results that suggest that active green walls have a high air cleaning potential. There is, however, a lack of empirical, *in situ* assessments of air pollutant removal for active green walls, a necessary requirement before this technology can be confidently recommended for functional use (Pettit et al. 2018a). Furthermore, before this technology can be universally applied as an air cleaning solution, it needs to be tested against existing technologies that are used to clean the air of ambient pollutants.

The current work represents two pilot-scale field studies to systematically compare, under realistic in situ conditions, the capacity of major phytoremediation technologies to quantitatively remove generated doses of PM and VOCs. As different biofilter designs, such as green walls and potted-plants, generally use different plant species, plant species typical of each biofilter design were chosen to ensure that the biofilters were representative of their real world application, thus allowing accurate comparisons of the *in situ* air cleaning abilities of different biofilters. The work therefore assesses whether botanical biofiltration designs may be a viable means to realistically clean indoor air. To provide practical outcomes, each biofilter design is representative of its real world application, with plant species typical of each biofilter design used in that application.

2. Methods

2.1 Field study 1: assessment of different forms of phytoremediation technologies

2.1.1 Phytoremediation technologies and room description

This experiment was conducted in a room within a residential building located in a suburban area of Sydney, Australia. The room had a floor area of 8.75 m^2 and a total volume of 22.70 m^3 (Figure 1). There was no HVAC or mechanical ventilation serving this room, and the door and windows were closed to create a sealed environment, representative of the conditions that would be normal in hot or cold seasons in this region. Nonetheless, a control treatment (described later) was used to eliminate any effects associated with the distribution and concentration of pollutants within the test space that might otherwise confound comparisons as per the conservation of mass model (Dockery and Spengler 1981). Experiments were conducted when the room's ambient temperature was between 20 and 24 °C. A ceiling fan in the room operated at a low setting for all trials to promote the distribution and homogenization of pollutants in the room, and to provide turbulence within the experimental space, as would be experienced in an occupied room.

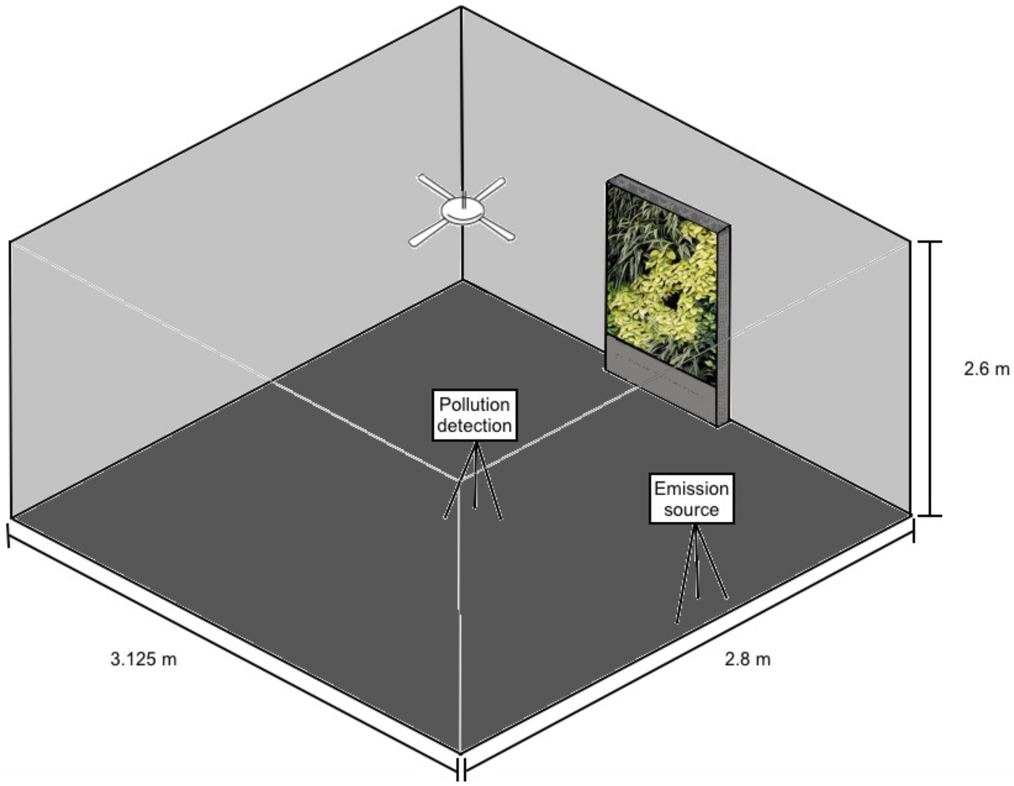

Figure 1. Diagram of the suburban residential test room. The biofilter was located in the centre against the long wall. Emission source was located in the centre against the short wall. Pollution detectors were located in the centre of the room. A ceiling fan was in the centre of the ceiling.

Three different types of botanical biofilters were tested in this room for their capacity to filter VOCs and PM.

Potted-plants: For each experimental replicate, three potted-plants (250 mm internal diameter, with the substrate comprised of commercial potting-mix) were placed in the room, representing a commonly-used planting density for a room of this size. Plant species used in the potted-plant treatment are species that are commonly grown in indoor environments; thus the plant species used in each trial consisted of one *Ficus lyrata* (fiddle leaf fig), one *Schefflera arboricola* (dwarf umbrella), and one *Philodendron tatei* ('Rojo Congo').

Passive green wall: The system tested was as previously described (Torpy et al. 2017). This system consisted of a 1.5 m^2 vertical green wall made up of six 0.25 m^2 modules (Junglefy Pty Ltd; Sydney, Australia), with each module having 16 holes from which plants can grow; thus the passive green wall contained 96 plants grown in a vertical alignment. The plant growth substrate in this system was comprised of coir fibre. The plant species used were *Chamaedorea elegans* (6% of total plants), *Epipremnum aureum* (34%), *Ficus lyrata* (4%), *Neomarica gracillis* (5%), *Peperomia obtusifolia* (10%), *Spathiphyllum wallisii* (21%) and *Syngonium podophyllum* (19%). Different plant species amongst the different treatments was seen as an inherent trait within each biofilter design and was thus left out of analyses, allowing direct comparisons of different biofilters representative of their real world application. Furthermore, the gravitropic effects on the growth and health of green wall species (Burritt 2013), do not allow potted-plant species to be used interchangeably with green wall species. As with the potted-plant treatment, this system had no active airflow, and was thus dependent upon diffusion for pollutant transfer, with assistance provided by the ceiling fan.

Active green wall: The system used (The *Junglefy Breathing Wall*, Figure 2) was as described in Pettit et al. (2017). To allow direct comparison with the passive wall, the active green wall was also a 1.5 m^2 vertical system made up of six 0.25 m^2 modules. The modules were attached to a plywood box forming an external plenum (depth = 100 mm; volume = 0.18 m^3). Two 240 V AC fans (DETA, 200 mm dia., 28 W), each with an open air volumetric flow rate of 320 $m^3 \cdot h^{-1}$, drew ambient air into the external plenum which was then forced through 75 mm ports on the rear face of each module. Within each module, the air was distributed evenly within an internal plenum (20 mm depth), where it then flowed through the plant growth substrate and foliage before returning to the ambient air. Both green wall designs used a substrate consisting of coconut husk, with a water holding capacity of 41.03 ± 1.26 % and an air filled porosity of 53.27 ± 0.98 % (Pettit et al. 2018b). This substrate has been used in other active green wall experiments, and is favourable as it does not contribute to airborne aeromycota (Irga et al. 2017a). The active green wall was comprised of similar plant species to the passive green wall. In both the passive and active green wall treatments, the location of each planted module within the frame structure was randomised amongst replicates to eliminate any bias associated with the orientation of the plant species within the wall.

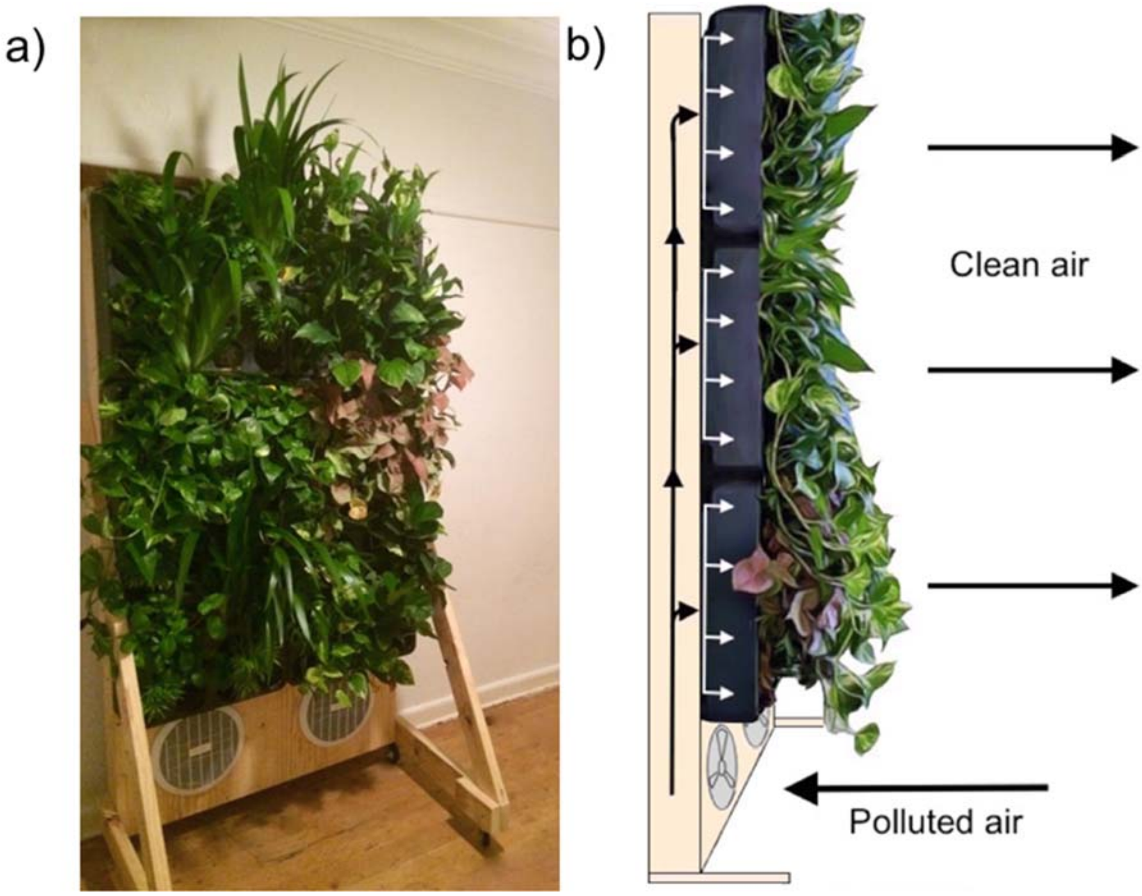

Figure 2. a) The active green wall within the tested residential room. b) Diagram of the active green wall system used in this study. Ambient air is taken in through the fans and pushed upwards through the external plenum. The air passes through an inlet into each green wall module where an internal plenum within the modules further distributes the air before it passes through the substrate and foliage.

Procedural control: As the active green wall treatment utilised mechanically-activated airflow, a procedural control was used to assess whether the influence of air movement facilitated by the use of the device affected the concentration of pollutants detected by the sensors. The procedural control consisted of the external plenum with the two fans operating, but with no green wall modules attached to it; thus mimicking the airflow pathway of the active green wall but without passing the air through any biofiltration matrix.

2.1.2 Pollution generation and sampling procedure

Treatments detailed in *Section 2.1.1* were tested for their capacity to phytoremediate a generated dose of VOCs and PM.

The ambient concentration of TVOCS was negligible ranging from 0-25 ppb Thus, to avoid the release of toxic VOCs in the residential house, lavender oil was used as a surrogate VOC. While lavender oil is usually not regarded as toxic, Chiu et al. (2009) observed that at high ambient temperatures (40 °C), it can emit a range of harmful VOCs such as toluene and *o*-xylene, while Su et al. (2007) report that linalool, D-limonene and terpinene-4-ol are emitted from lavender oil.

VOCs were generated by pipetting 100 µL of lavender oil (Thursday Plantation; Queensland, Australia) onto a 113.1 cm^2 sheet of 536:2012 80 gsm paper. The saturated paper was then suspended 10 cm in front of an axial impeller (FANTECH TEF-100 fan 16W) that was housed on a stand located at one end of the room, keeping the fan and paper 90 cm off the ground. This produced a TVOC concentration gradient in the room that rose from 0 ppb to an average maximum concentration of 120 ppb, representing a maximum concentration similar to the Australian National Health & Medical Research Council's recommended TVOC limit of ~125 ppb (Haag 2005).

A photoionization detector (PID; ppbRAE 3000, RAE Systems, San Jose, CA, USA; detection resolution of 1 ppbv of VOC concentrations ranging from 1 ppbv to 10,000 ppbv), located in the centre of the room on a 90 cm high stand, was used to measure the concentration of TVOCs over the length of each 2200 second (36.66 minute) trial. Pilot data indicated that this was the time required to remediate the entire room of the generated VOC dose or for VOC concentration to asymptote, thus the trial period was applied to all experiments thereafter.

An independent series of trials were performed to assess the capacity of the three phytoremediation technologies to filter suspended PM. In these trials, PM was generated by burning a widely-available incense stick (Meditation incense; S.D. Lovely Incense, Nepal; composition: red sandalwood 20%, sandalwood 15%, spoonpati 10%, Rhododendron 10%, medicinal plants 25%, natural glue 20%). Burning incense is a known particle emission source (Chang et al. 2007; Jetter et al. 2002; Ji et al. 2010; Lung and Hu 2003; See et al. 2007) and has been used previously for indoor plant PM remediation experiments (Panyametheekul et al. 2018). For each trial, a single incense stick was burned for approximately 5 minutes until the room's TSP concentration reached 400 $\mu g \cdot m^{-3}$; at this point, the incense was extinguished and the room's concentration of total suspended particles (TSP) was measured for the following 1900 seconds (31.66 minutes). Although PM concentrations at this level are not usually encountered in indoor environments (however see Huang et al. 2017; Shi et al. 2008; Tian et al. 2009), a high PM dose was selected as the starting concentration to assess how effectively this system can filter PM at levels that have been recorded in Beijing's urban areas on poor air quality days (e.g. TSP = ~400 $\mu g \cdot m^{-3}$ (US

Environmental Protection Agency 2016)); an essential function if these systems are to be a room's primary air cleaning device.

A DustTrak II 8532 nephelometer (TSI Incorporated, Shoreview, Minnesota, USA; detection limit: 0.01 mg/m^3; resolution 0.01 mg/m^3) was used to log the concentration of TSP in each trial. The locations of the PM generation and PM sensor in the room were the same positions as for the VOC generator and sensor respectively.

PM and VOC treatments were replicated eight times each for the potted-plant, passive green wall and active green wall treatments, and also the procedural control along with an empty room trial.

Experimental replication was achieved with time-for-space substitution, with a period of ventilation between samples, thus eliminating carry over effects generated from previous tests. All experimental air samples were taken with the door closed and sealed.

2.1.3 Data analysis

For each VOC trial, the VOC concentration was plotted as a function of time, and the corresponding area under the curve (AUC) was used as a response variable for a single factor ANOVA (IBM SPSS Statistics Ver 21) to test the mean differences amongst treatments. As the PM trials used an initial 'spiked' generation of particles as opposed to a continuous emission of pollutants, the area under the decay curve of TSP as a function of time was used as a response variable in a single factor ANOVA. Pairwise differences were identified using Tukey's HSD test where required. The clean air delivery rate (CADR) was calculated by using the static room test decay curves by taking the log loss function of particle concentration corrected for the rate of natural decay, and factoring in the test room size.

2.2. Field study 2: Active green wall and HVAC system trials

2.2.1 Room description

Before active botanical biofiltration can be applied universally as an air cleaning system, it is important to assess the pollutant remediation effects of these systems in high ambient pollution environments, and to compare these to current technologies such as HVAC systems.

Trials were conducted in a secondary school classroom located in Chaoyang District Beijing, China. The room's ventilation was served by a compartmentalised HVAC system that had 3 influent and 3 effluent ducts providing 2.5 air exchanges per hour. This system included a filter with a MERV H13 rating that filtered out a proportion of the outdoor ambient particles before they enter the indoor environment. As is commonplace in most buildings, this HVAC system removes VOCs from the room's atmosphere solely by flushing with filtered outdoor air. The room had a floor area of 40.07 m^2 and a volume of 120.2 m^3. A pedestal fan was placed in the corner of the room to ensure that air pollutants were distributed homogenously throughout the room. Whilst a fan would not normally be used in the room, occupant movement would lead to significant air mixing; thus the fan does not represent abnormal circumstances. Experimental replication was again achieved with time-for-space substitution, as described in *Section 2.1.2*.

2.2.2 Ambient air pollutant sampling

Ambient samples of suspended PM and TVOCs were taken across eight 30-minute trials in the room prior to active green wall installation. In these samples, the HVAC system was operating, thus this data reflects the concentration of pollutants in the room's normally operational state.

The mass concentration of total suspended particles (TSP) was recorded with a laser nephelometer (DustTrak II 8532), while a second laser nephelometer (Graywolf PC-3016A; Graywolf Sensing Solutions, Connecticut, USA; counting efficiency: 50% at 0.3 µm; 100% for particles >0.45 µm (as per ISO 21501-4) with a concentration limit of 4,000,000 particles / ft^3 at 5% coincidence loss) was used to calculate the size distribution and average concentration for a range of independent particle size fractions. The concentration of TVOCs was recorded with a PID (ppbRAE 3000).

2.2.3 Active botanical biofilter air pollutant sampling

The active green wall was constructed from 36, 0.25 m^2 modules, creating a wall with a surface area of 9 m^2 (Figure 3). These modules, which were of the same type as used in the residential room trial, contained mixed plant species, including *Epipremnum aureum, Nephrolepis exaltata, Peperomia obtusifolia, Schefflera arboricola* and *Spathiphyllum wallisii.* The approximate percentages of each species growing in the green wall were 40 %, 3 %, 10 %, 5 %, 42 % respectively. The modules were attached to a plywood box that was separated into 3 180 mm deep plenums, with each plenum containing 12 modules. Three 12 V DC fans, each with an open air volumetric flow rate of 185 $m^3 \cdot h^{-1}$, drew ambient air from ~40 cm above ground level into each plenum, which was then forced into the rear face of the modules, flowed through the plant growth substrate and foliage before returning to the ambient air. The volumetric flow rate through the green wall was 283.53 $m^3 \cdot h^{-1}$, representing 2.36 air changes per hour for the test room.

Once the active botanical biofilter was installed in the room, the room's HVAC system was turned off and the ducting sealed with plastic sheets to ensure no air exchange between the room and the HVAC ducting. Each trial that tested the active green wall (*n*=3) was conducted for 20 minutes, which was the time taken for the TSP concentration in the biofilter treatment to approach an asymptote. The concentration of TVOCs and the size distribution and concentration of particles were recorded as previously outlined. All samples were taken from a distance of ~2 m away from the active botanical biofilter and ~1 m above the ground.

Figure 3. The active green wall used to filter ambient PM and VOCs installed in a classroom in Beijing, China.

2.2.3 Data analysis

Two separate t-tests were used to compare the mean concentrations of TSP and TVOC between the ambient HVAC system and active green wall treatments. Data was analysed using IBM SPSS Statistics Ver 21.

3. Results

3.1 Field study 1: Sydney Australia suburban residential

A one-factor ANOVA revealed significant differences in the concentration of TVOCs amongst treatments (Figure 4; d.f. = 4 and 36, $F = 89.198$, $p = 0.000$). Subsequent Tukey's HSD *post hoc* tests found that the active green wall treatment was significantly different to all of the other treatments ($p = 0.000$ for all comparisons), while no other significant differences were found amongst any of the other treatments. The active green wall led to considerably lower concentrations of TVOCs throughout the experimental period (Figure 3), in which the active green wall produced an average time-weighted TVOC concentration 72.5% lower than the TVOC concentration present in the empty room.

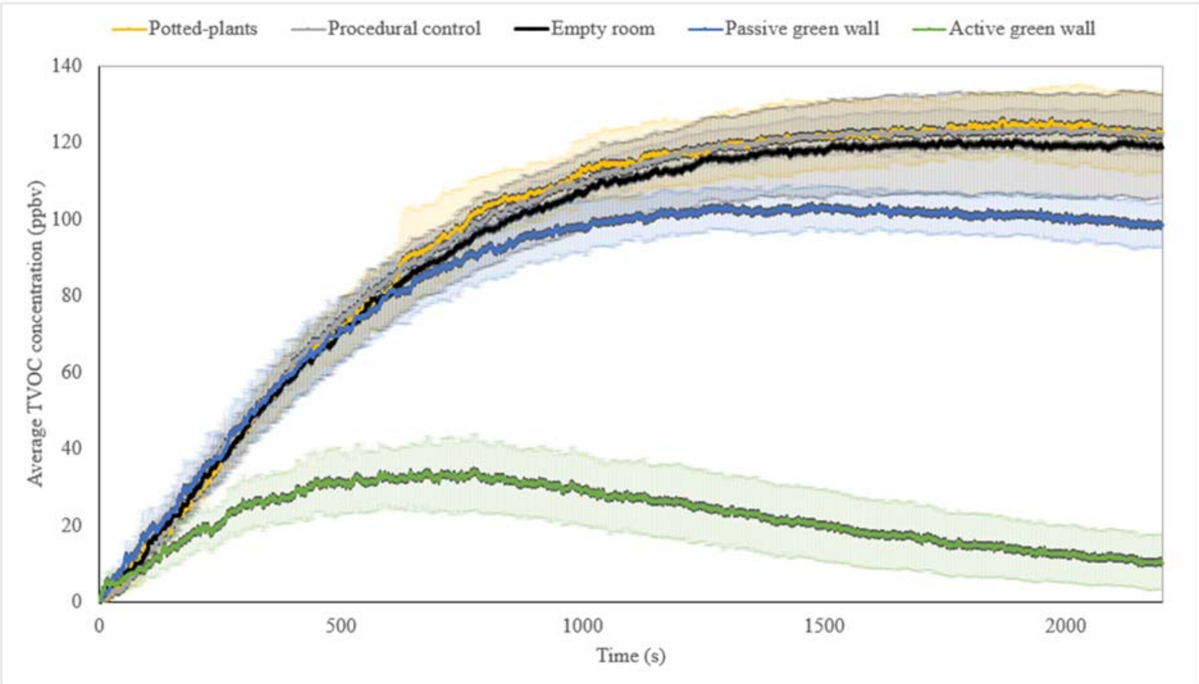

Figure 4. The average concentration of TVOCs for each treatment. Error bars represent the standard error of the mean (*n*=8).

There were significant differences amongst treatments in the AUC of TSP concentration as a function of time (Figure 5; ANOVA: d.f. = 4 and 36, $F = 34.970$, $p = 0.000$). Tukey's HSD *post hoc* tests indicated that AUC of TSP in the active green wall treatment was significantly lower than all other treatments ($p < 0.000$ for all comparisons), while the passive wall had a significantly lower AUC than then empty room ($p = 0.000$) and the potted-plant treatment ($p = 0.004$). The total decay rate constant for the active green wall treatment was 4.53×10^{-4} s^{-1} and the CADR calculated from the decay curves was 21.98 m^3/h.

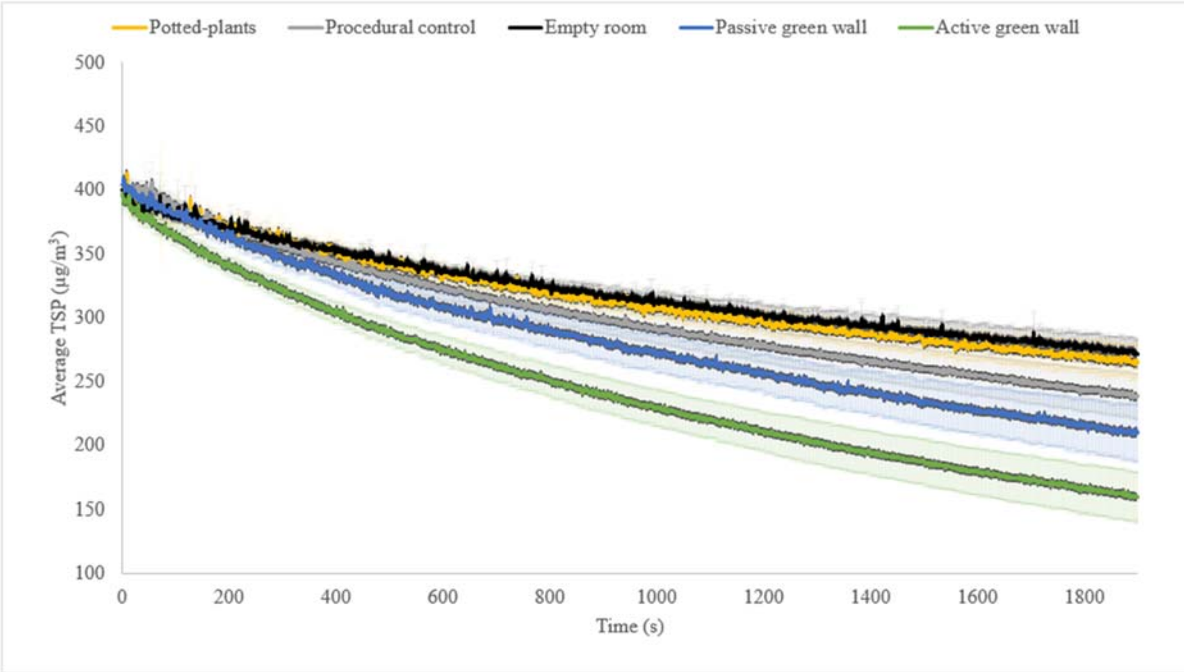

Figure 5. The average concentration of TSP over the trial time. Error bars represent 95% confidence intervals.

3.2. Field study 2: Beijing, China urban classroom trial

Prior to the installation of the active green wall, the average concentration of TVOCs within the room with the HVAC system operating was 300 ± 3.04 ppbv, and this concentration was relatively stable throughout the sampling period. Following the installation of the active green wall, the average concentration of TVOCs was reduced to 217 ± 2.00 ppbv over the 20 min trial period, representing a reduction of ~28%. The average concentration of TVOCs was significantly lower in the active green wall treatment compared to the HVAC ambient air treatment (t = 3.311, d.f. = 7, p = 0.011).

The average ambient concentration of particles (as TSP) in the room with the HVAC operating was 101.18 ± 0.29 $\mu g \cdot m^{-3}$. The mass concentration of particles was distributed relatively evenly over a range of different particle size fractions (Figure 6). Once the active green wall was installed, the mass concentration of all particle sizes was reduced, with relatively rapid removal (Figure 7). The mass concentration of TSP in the room was reduced by 42.6% by the active green wall relative to the building HVAC system over 20 minutes. A t-test revealed that the difference in the TSP concentration between the HVAC ambient air treatment and the active green wall treatment was statistically significant (t = 2.679, d.f. = 7, p = 0.037).

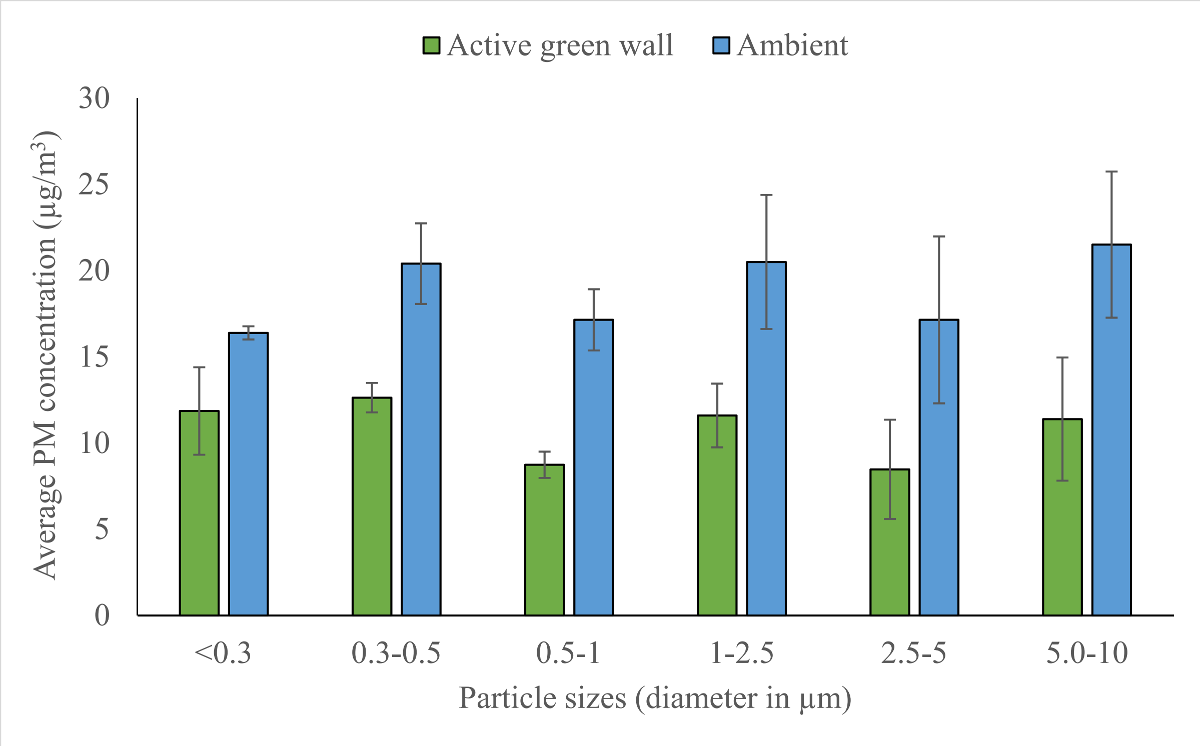

Figure 6. The average particle size fraction concentrations for ambient HVAC and botanical biofilter treatments. Error bars represent standard error of the mean (control: *n*= 8; active green wall: *n*=3).

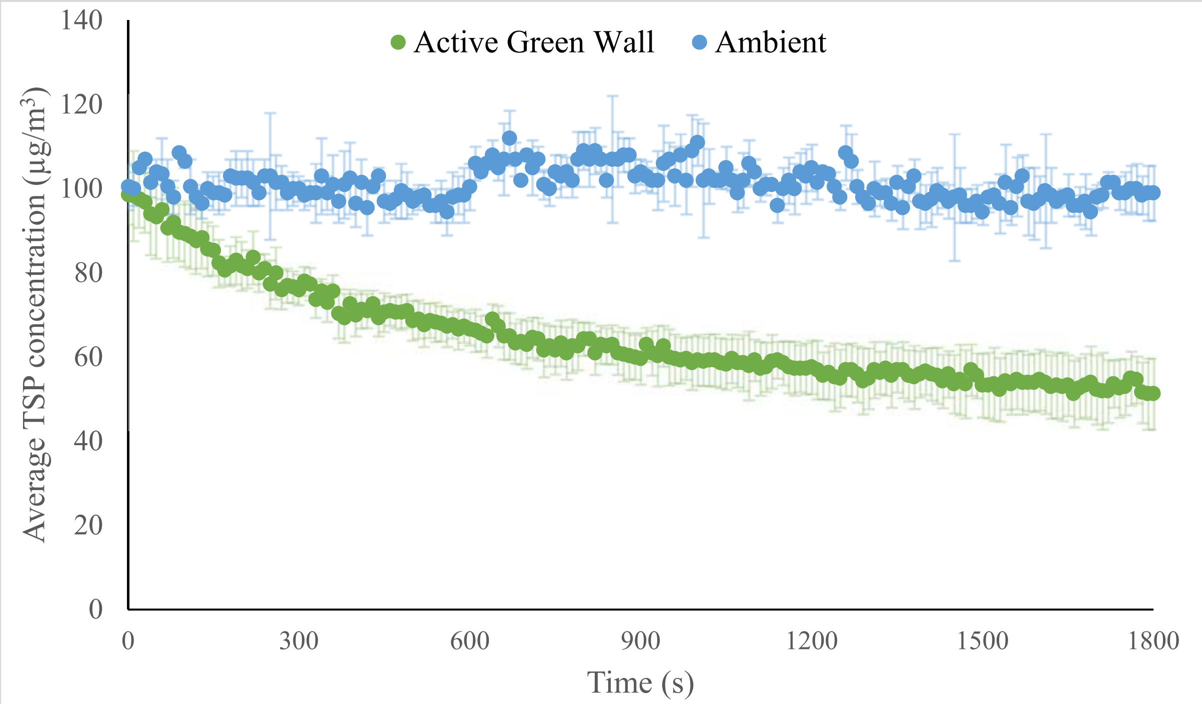

Figure 7. The average concentration of ambient total suspended particles over a 20-minute sampling period with and without an active green wall biofilter present. Error bars represent 95% confidence intervals (control: *n*= 8; active green wall: *n*=3).

4. Discussion

The current study represents the first work conducted to compare the *in situ* VOC and PM removal capabilities of the major phytoremediation technologies to add further evidence to support the use of these systems as plausible solutions for managing indoor air quality. Several previous studies have assessed the capacity of potted-plants (or parts of potted-plants) to adsorb and degrade VOCs (Aydogan and Montoya 2011; Hörmann et al. 2018; Irga et al. 2013; Kim et al. 2016; Sriprapat et al. 2014; Sriprapat and Thiravetyan 2013; Sriprapat and Thiravetyan 2016; Treesubsuntorn et al. 2013; Treesubsuntorn and Thiravetyan 2012), while a lesser number of studies have measured the effects that potted-plants have had on ambient concentrations of VOCs in realistically sized rooms (Wood et al. 2006), and only a very limited number of studies have demonstrated VOC removal by active or passive green walls *in situ* (Darlington et al. 2001). The current work has shown that in a small airtight room with elevated VOC concentrations, a reasonable density of potted-plants or a reasonably sized passive green wall do not provide substantial reductions in the concentrations of VOCs within a relatively short time period (i.e. ~37 minutes in this experiment). Alternatively, the active green wall effectively reduced the concentration of VOCs to levels that are unlikely to have health effects. Towards the end of the trial period, the active green wall began to remove VOCs at a faster rate than they were emitted, so that the concentration of VOCs had almost returned to their starting concentration. This comparison suggests that active green walls can provide practical reductions in VOC concentrations, while other forms of phytoremediation system may not provide equally rapid reductions. While previous *in situ* studies have suggested that potted-plants can reduce in-room VOC concentrations over longer time periods (e.g. 24 h (Wood et al. 2006)), this performance did not extend to the short duration study presented here. Although some laboratory scale experiments have shown that plant tissues are capable of removing VOCs from chambers over several hours (Liang et al. 2018; Parseh et al. 2018; Su et al. 2018), the relatively short trial time in this experiment suggests that VOCs were most likely removed through adsorption processes as opposed to microbial degradation.

Orwell et al. (2004) found that the substrate microbial community's VOC removal efficiency improves with repeated exposure to multiple doses, and it is possible that considerably lengthening the experimental trial period, or testing the VOC removal of repeated VOC doses, may have provided improved removal rates. Alternatively, Inouye et al. (2003) has shown that lavender oil may inhibit the growth of microorganisms, and thus the use of lavender-derived VOCs may have differentially affected the removal rates observed if longer trials were performed. In any case, the demonstration of fast-response VOC removal by the active green wall is indicative of considerable practical value, as the system has the capacity to remove VOCs as they are emitted, maintaining low room VOC concentrations without the need for a lengthy adaptation period where VOCs in an indoor space would still be at high levels.

The accumulation of particles on the plant foliage of passive green walls (Perini et al. 2017; Weerakkody et al. 2017; Weerakkody et al. 2018a; Weerakkody et al. 2018b) and potted-plants (Gawrońska and Bakera 2015) has been noted as a promising potential means for the removal of atmospheric PM. Although it is clear that plant foliage can provide PM deposition sites, it has previously been difficult to determine if this quality corresponds with functional reductions in ambient indoor PM concentrations. The current results comparing TSP removal efficacy across different phytoremediation treatments suggest that passive green walls can reduce the PM concentration of the surrounding air. Furthermore, the active green wall provided significantly lower concentrations of TSP than all other treatments, which is unsurprising as this treatment uses active airflow to treat a greater quantity of polluted air, thus having both the capacity to capture PM on the plant's foliage and to filter PM through the substrate matrix. Importantly, this study only made these comparisons under a relatively high initial PM concentration in a relatively small room, and the removal capacity of such systems may be different under different conditions. Given these findings, it is thus essential that subsequent experiments measure ambient air pollution reductions associated with botanical biofilters to gauge their potential to functionally enhance air quality, rather than to only measure variables that may be associated with providing cleaner air such as particle accumulation.

In the second field study located in China, the active green wall outperformed the tested HVAC system in terms of both VOC and PM mitigation. The MERV H13 filter used in the HVAC treatment has particle size removal efficiencies of >90 % for particles with diameters of 1-10 µm (ASHRAE 2007), and is typically applied in "superior commercial buildings, smoke removal systems and hospitals" (ASHRAE 2007). These metrics notwithstanding, the active green wall system outperformed the HVAC treatment by significant margins for all particle size fractions. The high filtration performance of the active green wall indicates a high air cleaning capacity, and suggests that it may have considerable practical potential. Importantly, these two technologies filter ambient particles with different airflow pathways. HVAC systems most frequently filter particles from outdoor air as the airstream enters the building, while the active green walls (in the form used here) filtered recycling ambient air from within the building. While this characteristic of the active green wall negates the requirement to temperature modulate outdoor air to the desired indoor temperature as is necessary for the HVAC, the overall performance of the system relies on being able to draw air from all regions of the indoor space in which it is situated. Thus, further studies will be needed to understand the airflow dynamics of active green walls in differently sized and shaped rooms, and how this interacts with biofilter dimensions with fan mass airflow rates.

The concentration of VOCs was significantly reduced in the active green wall treatment when compared to the HVAC system treatment. This is a differentiating function of active botanical biofilters, as HVAC systems simply reduce high concentrations of indoor-generated VOCs by flushing with outdoor air. The observed capacity to mitigate high *in situ* concentrations of both PM and VOCs lends support to Darlington et al.'s (2001) proposal that air pollutants can be treated by recirculating and treating the air within a building, thus partially eliminating the energy intensive process of flushing the building with temperature modulated, filtered outdoor air to control problematic concentrations of PM and VOCs. Increasing concentrations of CO_2 resulting from occupant respiration, however, remain difficult for green wall technology to treat with practical removal rates (Torpy et al. 2017), as CO_2 removal is dependent on plant photosynthetic activity; a process largely governed by photon flux density in the wavelength ranges used by plants for photosynthesis, which are typically low in indoor environments (Safe Work Australia 2011). As such, further development is needed before botanical systems can be effectively implemented to offset high CO_2 concentrations in addition to the demonstrated effects on PM and VOCs. Furthermore, while current research suggests that active biofilters do not emit bioaerosols in harmful concentrations (Irga et al. 2017a; Mallany et al. 2002; Zilli et al. 2005), further testing is needed to ensure that these findings remain valid across different indoor environments.

A number of previous studies have assessed the VOC single pass removal efficiencies (SPRE) and calculated clean air delivery rates for phytoremediation technologies. Torpy et al. (2018) tested a botanical biofilter that had the same planted surface area as the active green wall used in the residential experiment in the current study. They found that their active botanical biofilter could remove ~57% of methyl ethyl ketone (MEK) from a constant stream of contaminated air, thus providing a clean air delivery rate (CADR) of 28.4 $m^3 \cdot h^{-1}$. Wang and Zhang (2011) assessed the SPRE of their 'dynamic botanical air filtration' system and recorded SPREs of 50.1–91.7% and 73.2–98.7% for toluene and formaldehyde respectively, depending on soil moisture levels and airflow rate providing CADRs ranging from 232.4–759.7 $m^3 \cdot h^{-1}$ (Wang and Zhang 2011). Comparisons across systems suffer from low validity, however, due to the use of different VOCs, different room sizes and layouts, inconsistent plant species and substrates, and different remediation metrics. Despite the impressive VOC removal rates demonstrated by active botanical biofilters, the influence of system operation on *in situ* PM concentrations is a novel finding that supports the value of active green walls as a technology capable of remediating high concentrations of a range of behaviourally diverse air pollutants.

It is likely that positive health impacts would be associated with the reductions in VOCs and PM observed for the active green wall treatments in both room trials. VOC exposure has been shown to have dose-response relationships for upper and lower respiratory symptoms (Pappas et al. 2000), and research suggests that chronic exposure to relatively small concentrations of certain VOCs is associated with detrimental health effects (Khanchi et al. 2015; Qu et al. 2002). Thus, even small reductions in VOCs are likely to have quantitatively positive health outcomes. Similarly, significant health impacts have been associated with exposure to relatively minor increases in PM (in particular fine particles) concentrations: Wang et al. (2016) found that whole population all natural-cause mortality increased by 3% with each 2 $\mu g \cdot m^{-3}$ increase in $PM_{2.5}$ exposure. There is strong evidence to suggest that a 10 $\mu g \cdot m^{-3}$ incremental increase in the concentration of $PM_{2.5}$ is associated with a detectable increase in total population mortality, specifically that related to cardiovascular disease and respiratory disease risk (Li et al. 2017). The active green wall used in the schoolroom thus potentially produced an indoor environment that could lead to quantifiably improved health outcomes. Before such epidemiological claims can be made, however, this technology needs to be widely implemented over various temporal and spatial scales, with air quality monitoring and health outcome assessment programs.

The schoolroom setting represents an ideal environment for active botanical biofiltration to be implemented, as children are particularly vulnerable to adverse health effects related to air pollution exposure (Buka et al. 2006), notwithstanding the well-documented biophilic satisfaction and increased school performance associated with indoor greening (Daly et al. 2010).

5. Conclusion

Potted-plants, passive green walls and active green walls were tested for their capacity to reduce in room concentrations of VOCs and PM, with active green walls providing significant reductions in VOC and PM concentrations, while passive walls showed a lesser reduction in PM concentration. Active green walls reduced the concentration of PM and VOCs from a classroom to provide greater air quality than that provided by the classroom's current HVAC system. Although these pilot-scale results indicate that active green wall systems are capable of improving indoor air quality, further empirical validation, incorporating long-term studies in varied indoor environments are needed to ensure active botanical biofiltration can be implemented to efficiently and reliably clean indoor air.

Acknowledgements

The authors would like to acknowledge Junglefy Pty for providing the green wall modules. The authors would additionally like to thank Junglefy, Ornatec International Consultancy and Beijing Florascape for their effort and assistance that allowed these trials to occur. This research is supported by The UTS Centre for Technology in Water and Wastewater (CTWW). T Pettit is supported by an Australian Government Research Training Program Scholarship.

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