https://doi.org/10.1016/j.jhazmat.2021.125566

Effective reduction of roadside air pollution with botanical biofiltration

Thomas Pettit [1], Fraser R. Torpy [1], *, Nicholas C. Surawski [2], Robert Fleck [1], and Peter J. Irga [2]

[1] Plants and Environmental Quality Research Group, School of Life Sciences, Faculty of Science, University of Technology Sydney, Sydney, NSW 2007, Australia;

[2] Centre for Green Technology, School of Civil and Environmental Engineering, University of Technology Sydney, Sydney, NSW 2007, Australia;

Correspondence: Fraser R. Torpy

Email: Fraser.Torpy@uts.edu.au

Phone: +61 2 95144150

Address: 04.06.550 University of Technology Sydney

15 Broadway, Ultimo, NSW, Australia, 2007

Thomas Pettit: 0000-0003-2707-7764

Fraser R. Torpy: 0000-0002-9137-6948

Nicholas C. Surawski: 0000-0003-2584-1044

Robert Fleck: 0000-0002-2045-1656

Peter J. Irga: 0000-0001-5952-0658

Abstract

Currently no sustainable, economical and scalable systems have been developed for the direct removal of roadside air pollutants at their source. Here we present a simple and effective air filtering technology: botanical biofiltration, and the first field assessment of three different botanical biofilter designs for the filtration of traffic associated air pollutants – NO_2, O_3 and $PM_{2.5}$ – from roadside ambient air in Sydney, Australia. Over two six month research campaigns, we show that all of the tested systems filtered NO_2, O_3 and $PM_{2.5}$ with average single pass removal efficiencies of up to 71.5%, 28.1% and 22.1% respectively. Clean air delivery rates of up to 121 m^3/h, 50 m^3/h and 40 m^3/h per m^2 of active green wall biofilter were achieved for the three pollutants respectively, with pollutant removal efficiency positively correlated with their ambient concentrations. We propose that large scale field trials of this technology are warranted to promote sustainable urban development and improved public health outcomes.

Key words: green infrastructure; green wall; living wall; air quality; traffic pollution; urban greening

Highlights

- Botanical biofiltration of NO_2, O_3 and $PM_{2.5}$ was achieved at roadside environments.
- NO_2 was removed most efficiently, with a single pass removal efficiency of 71.5%.

- Pollutant clean air delivery rates of 40–121 m^3/h per 1 m^2 plenum were achieved.
- All pollutant removal rates were positively correlated with ambient concentrations.

1. Introduction

Ambient air pollution is the most significant current environmental risk to human health, with approximately 4.2 million deaths around the globe each year attributed to exposure to ambient air pollution (WHO 2019). Urban air pollution is particularly concerning, where vehicle exhaust and industrial emissions lead to elevated air pollution levels in environments inhabited by the majority of the world's population (WHO 2019). Urban air pollution is comprised of a complex mixture of suspended particles, (particulate matter; PM), and gaseous pollutants, including nitrogen oxides (NO_x), ozone (O_3), amongst other pollutants (Venkatram and Schulte 2018). Vehicular emissions, particularly in locations with high traffic densities, are the main source of harmful air pollutants in many urban areas (European Environmental Agency, 2011). Because traffic related pollutants are emitted close to ground level, elevated pollution concentrations frequently occur in 'on-road' or 'near-road' environments, whereby the urban population, including drivers, commuters, pedestrians and occupants of nearby buildings, is exposed to heightened pollution concentrations (Karner et al, 2010; Pasquier and André, 2017). Furthermore, the dispersion of ground level traffic emissions may be limited by urban geometries and structures, such as buildings, and in some cases, tree canopies (Abhijith et al. 2017; Venkatram and Schulte 2018), thus promoting the accumulation of air pollution in zones where people are likely to be exposed.

The health effects resulting from exposure to urban air pollution are associated with huge economic impacts (Pascal et al. 2013). Therefore, work directed towards air pollutant mitigation is of the greatest importance, as are effective new technologies aimed at reducing the concentration of air pollutants in environments where human exposure is at its highest.

Botanical biofilter technology, which generally takes the form of active green walls, has been developed from an extension of the concept of phytoremediation (Irga et al. 2018). These systems have plants arranged along a vertical pane and use ‘active airflow’ to mechanically force an airstream through the plant foliage and growth substrate, where it exits to the ambient air (Pettit et al. 2018a). In this process, PM is mechanically filtered by the growth matrix, and gaseous pollutants such as VOCs, O_3 and NO_2 can be biodegraded by the microorganisms contained in the growth substrate or removed from the airstream by adhering to substrate adsorbents (Pettit et al. 2018b). Several studies have suggested that such systems (or similar botanical biofilters) can make functional improvements to the air quality of indoor environments (Darlington et al. 2001; Ibrahim et al. 2019; Pettit et al. 2019; Wang and Zhang 2011).

Although active green wall research has been limited to laboratory studies and indoor air quality investigations, traditional urban forestry, such as street trees, hedges and shrubs, have been thoroughly studied for their capacity to remove urban air pollutants (Abhijith et al. 2017; Petrova 2020). Nowak et al. (2006) suggested that urban trees and shrubs remove 711,000 metric tons (US$ 3.8 billion value) of air pollution (O_3, PM_{10}, NO_2, SO_2, CO) across the United States of America each year, whereby pollutants are removed through foliar processes such as stomatal uptake and wet and dry deposition. Several studies however, have noted that in some cases, particularly in street canyons, there is potential for urban tree canopies to limit the diffusion of air pollution from sources such as traffic, and thus, increase the concentration of air pollution at ground level (Gromke et al 2008; Jeanjean et al. 2017; Salmond et al. 2013; Vos et al. 2013). Alternatively, passive green walls may be used in both street canyons and open road settings to provide improvements to air quality, primarily through hindering the dispersion of pollutants from reaching relevant exposure zones (Abhijith et al. 2017; Abhijith and Kumar 2019). Nonetheless, current technologies that attempt to mitigate ground level air pollution exposure in urban contexts, including roadside vegetation barriers and solid barriers (Gallagher et al. 2015; Tong et al. 2016), primarily work through altering pollutant dispersion rather than reducing the pollutant load from the ambient air through filtration and bioremediation.

The use of airflow in botanical biofiltration promotes the rate at which substrate-associated pollutant removal processes operate, whilst adding the effects of bioremediation and filtration; thus removing air pollution from the ambient air rather than simply shifting pollutant dispersion, and thereby providing a promising means to considerably improve urban air quality. Additionally, the small ground and canopy footprint of green walls allows these systems to be installed in spatially constrained urban areas (Abhijith et al. 2017). Due to the extensive range of environments in which this technology can be applied and the vast range of adjunct benefits provided, including urban stormwater management, temperature reductions, acoustic attenuation and enhanced scenic landscape (Attal et al. 2017; Horoshenkov et al. 2011; Manso and Castro-Gomes 2015); the assessment of botanical biofilters for air quality enhancement is of major value for sustainable urban design, and is of international scope. Botanical biofilters have been shown to make functional improvements to the air quality of indoor environments (Darlington et al. 2001; Pettit et al. 2019a; Wang and Zhang 2011), and are beginning to be built for this purpose in urban areas, however their efficacy in outdoor environments remains untested. Here, we aim to build on indoor and laboratory research to evaluate the use of this technology as a solution to improve air quality alongside major roads.

In this investigation, we firstly assess the single pass removal efficiency (SPRE) of traffic associated air pollution achieved by botanical biofiltration. This was accomplished by conducting extensive air quality monitoring across several independent botanical biofilter arrays to assess the biofiltration efficiency for $PM_{2.5}$ (fine suspended particles with an aerodynamic diameter less than 2.5 µm), NO_2 and O_3 from the ambient air of two roadside environments in Sydney. Secondly, we consider the contribution of cleaned air produced by three biofilter designs by evaluating removal efficiencies in conjunction with airflow characteristics to determine the clean air delivery rate (CADR) for the systems when trialled *in situ*. Finally we explore the relationship between removal efficiency and ambient pollutant concentration for each of the pollutants. The combined findings demonstrate the potential for the implementation of this new technology to promote sustainable urban development areas and improved public health outcomes.

2. Methods

2. 1 Site description and botanical biofilter orientation

Botanical biofiltration arrays were installed at two different roadside environments in Sydney. Sydney is Australia's most populous urban centre, with an estimated population of 5.2 million residents (Australian Bureau of Statistics 2019). Emissions from motor vehicles are a major source of air pollution in Sydney (NSW Health 2014; Paton-Walsh et al. 2019) and are the largest contributors of NO_x (Cowie et al. 2019) and $PM_{2.5}$ pollution (Crawford et al. 2017). Motor vehicles also emit VOCs, which are important precursors in the formation of ozone (NSW Health 2014). Traffic counts at both sites during the experimental period were sourced from Transurban (2020).

2.1.1 Site 1: Eastern Distributor

Two biofilter arrays were installed alongside the Eastern Distributor, situated immediately adjacent to north bound traffic so that the biofilter arrays were flush against the traffic barrier closest to the road. The Eastern Distributor Motorway (33°52'12.2"S 151°13'05.8"E) is located in the City of Sydney local government area, and is one of Australia's busiest roads and is located in one of Australia's most densely populated areas (Roads and Maritime Services 2018). To provide spatial independence, the biofilter arrays on site were separated from each other by 30 m. Sampling took place from June 2019 to November 2019.

2.1.2 Site 2: Hills Motorway

The Hills Motorway (M2; 33°46'09.6"S 151°06'58.4"E) site was located approximately 13 km north-west of Sydney's central business district within the local government area of the City of Ryde. This installation was positioned on an unused asphalt area between Southeast bound traffic on the Hills Motorway and the Christie Rd exit ramp. This area is separated from the Southeast bound traffic on the Hills Motorway by concrete ('Jersey') barriers. Three biofilter arrays were situated immediately adjacent to southeast bound traffic, on the immediate edge of the Hills Motorway's southeast bound lanes. At this site, there was at least 50 m between biofilter arrays to ensure that the effects of one biofiltration array would not confound measurements at the others. Sampling at this location took place from November 2019 to May 2020.

As it was hypothesized that the ambient pollution profile and concentration would affect filtration efficiency, the two sites were selected due to their different pollution characteristics. The Hills Motorway is comparatively more open (i.e. less urban development adjacent to the road) than the Eastern Distributor, and thus the dispersion of pollutants at this site may not be hindered to the same degree as that on the more developed Eastern Distributor. Different traffic speeds between the sites may also influence the associations between traffic volume and ambient air pollution concentration at each site. Although traffic speed was not measured in this study, the speed limit on the Hills Motorway is higher than that of the Eastern Distributor (100 km/h and 60 km/h respectively), and it is possible that faster traffic on the Hills Motorway promoted increased pollutant dispersal on the Hills Motorway (Venetsanos et al. 2001).

2.2 Botanical biofilters

Each of the five biofilter arrays (1 x 5 m wall surface area) held 20 biofilter modules (Breathing Wall; Junglefy Pty Ltd, Sydney, Australia) across five independent 1 m^2 plenums per array, as described in Pettit et al. (2020). Each module (0.5 x 0.5 x 0.15 m) was made from recyclable low-density polyethylene, with a front face area of 0.25 m^2 that contained 16 holes from which plants can grow. The biofilter arrays contained the following species of plants: *Westringia fruticosa* (coastal rosemary), *Myoporum parvifolium* (dwarf native myrtle), *Strobilanthes anisophyllus* (goldfussia) and *Nandina domestica* (heavenly bamboo). These species were selected for their survivability in Australian roadside environments. The internal space within the module was filled with a coconut husk-based plant growth substrate. A sheet of high-density polyethylene shade cloth lined the internal surfaces of the module to hold the plant roots and growth substrate within the module. The rear face of each module contained an opening in its centre (63.6 cm^2 cross sectional area), which was used to pull an airstream through the openings in the front face and through the growth substrate, after which it exited the module through this opening. A baffle plate was located against the internal rear face of the module to promote uniform airflow through the front face of the module. Each biofilter array was irrigated via a drip line with ~11 litres of water every 2 days. In addition to this irrigation, biofilter arrays were also exposed to rain and would have received supplementary irrigation through natural rainfall. Each biofilter module contained drainage holes allowing water to drain from each module if they were watered beyond field capacity.

To isolate the effluent airstream, modules were fixed to steel plenums, which contained fans to generate airflow. Each plenum was 1 x 1 m x 0.15 m (1 m^2 front face area) and held four botanical biofilter modules. The airstream passed into the plenum through four openings on the plenum’s front face; these openings corresponded to the opening on the rear face of each of the modules. Two fans (NF-F12, Noctua; Austria) with an internal diameter of 120 mm, a volumetric flow rate of 186.70 m^3/h at 0.00 Pa of static pressure, and rated power consumption 4.32 W, were arranged in parallel on the rear face of the plenum. These generated active airflow that pulled air through the plant foliage and the front face of the module, through the opening in the rear face of the module, where the airstream then entered the plenum and exited the plenum to ambient via to vents adjacent to the fans in the rear face of the plenum. The vents matched the area of the fan outlet and used louvers to prevent rainwater from entering the plenum. Five plenums and their corresponding modules were arranged horizontally, creating 1 x 5 m biofilter arrays, which were supported on frames so that the base of the walls were ~ 1 m above the ground (Figure 1).

Figure 1. A botanical biofilter array. A) the rear view of a biofilter array showing five plenums arranged horizontally to form a 5 m^2 active green wall; B) a side view of the support structure with biofilter modules attached to the plenum; C) the front face of the biofilter array.

2.3 Botanical biofilter design comparisons

As this was the first time botanical biofilters had been assessed for traffic-associated air pollution removal in outdoor environments, it was unclear how some system aspects, such as variations in airflow, would affect the overall performance. Thus, three different design iterations were used to investigate traits associated with optimum *in situ* performance (Table 1). In addition to the design iteration described above, one plenum on each biofilter array contained 4 granular activated carbon (GAC) cassettes housed within the four openings of the plenum's front face. In this design, the airstream would firstly pass through the biofilter module and then through a small cylinder (44 mm internal radius, 20 mm depth) containing GAC (GAC; EA1000 4 mm; Activated Carbon Technologies Pty Ltd, Melbourne, Australia). Although previous work has suggested that GAC can be used to enhance the SPRE of gaseous pollutants (Pettit et al. 2018b), it is unknown how it would influence the CADR in roadside environments, and for a range of behaviourally different pollutants. Lastly, one plenum on each biofilter array contained two fans with a larger volumetric flow rate (NF-A14, Noctua, Austria; 140 mm internal diameter, volumetric flow rate of 269.3 m^3/h at 0.00 Pa of static pressure, and a rated power consumption of 6.6 W. This treatment was included to test the effect of increasing volumetric airflow rate on CADR.

Table 1. The different botanical biofilter design iterations that were trialled in roadside environments.

Botanical biofilter iteration	Fan type	Fan diameter	Fan flow rate at 0 Pa static pressure (m^3/h)	Filtration components
1	NF-F12, Noctua	120 mm	186.70	Coconut husk-based plant growth substrate + 64 plants per 1 m^2
2	NF-F12, Noctua	120 mm	186.70	As for #1 with the addition of granular activated carbon cassettes
3	NF-A14, Noctua	140 mm	269.3	As for #1

2.4 Air quality measurement

The air velocity through each of the louvers was multiplied by the area of the louver opening to calculate the volumetric flow rate through each of the plenums. The airflow through each of the plenums was measured with a VelociCalc Air Velocity Meter 9545 (TSI Incorporated; Shoreview, Minnesota, USA).

The concentrations of NO_2, O_3 and $PM_{2.5}$ were measured with a series of AQY1 – micro air quality monitoring systems (Aeroqual Limited; Auckland, New Zealand). Although Sydney is considered to have relatively ‘good’ air quality, $PM_{2.5}$ and O_3 are the air pollutants that most frequently occur in high levels (Paton-Walsh et al. 2019), while traffic emissions of NO_x account for 61.8% of the total annual NO_x emissions in the Sydney region (NSW EPA 2012). Two AQY1 instruments were located at each end of each biofilter array. These provided measurements of the proximal ambient air quality for each biofilter array. For assessment of air pollutant removal efficiency, AQY1 instruments were placed in each of the plenums, and thus detected the concentration of NO_2, O_3 and $PM_{2.5}$ in the isolated effluent airstream. Although these instruments have high detection resolutions (see Aeroqual Limited 2019) and were factory-calibrated before use, any systematic differences in the calibration of each instrument could potentially influence the accuracy of any comparisons amongst air pollution concentrations between the ambient and filtered effluent air. Thus, the locations of the instruments were randomly rotated several times throughout the experiment, both amongst plenums and ambient air detecting locations.

Average air pollution concentrations were calculated for each 5-minute period from 6:00 am to 6:00 pm. A 12-hour period overnight without fan operation provided temporal independence for each composite daily replicate of pollutant concentrations.

2.5 Data and statistical analysis

In order to make comparisons across treatments, the average ambient and average air pollution concentrations in the plenums of each treatment were calculated. The SPRE was calculated for each pollutant by comparing the average ambient air pollutant concentrations to the average air pollution concentrations detected in the isolated effluent airstreams of each biofilter.

Unlike assessments of air pollutant removal provided by passive vegetation, whereby phytoremediation of air pollution is usually measured as mass of pollutant removed, the use of active airflow in botanical biofiltration allows removal rates to be expressed as clean air delivery rates (CADRs). This metric is a function of the proportion of influent pollution that has been removed on a single pass through the biofilter, multiplied by the volumetric airflow rate through the biofilter. The CADR of each pollutant thus describes the volume of 'clean' air produced by the biofilters, and is generally considered to be the best metric to evaluate air cleaning potential (Zhang et al. 2011). Further, converting the SPREs for each pollutant to CADRs facilitated valid comparisons of the treatments with different airflow rates. Differences in the CADR amongst treatments were statistically compared through ANOVA (IBM SPSS Statistic Ver 25).

Additionally, the SPRE of each pollutant was considered as a function of the ambient pollutant concentration to assess the relationship between removal efficiency and pollutant concentration. The average pollutant concentrations and biofilter SPREs from both sites at each time sample were included in this correlation, thus ensuring bivariate normality of each data point.

The ambient concentration of $PM_{2.5}$ at each site was used as a surrogate pollutant to test associations between air pollution and the traffic densities at each site. A Pearson's correlation analysis was used to test the association between the average ambient $PM_{2.5}$ concentration at each 15-minute interval and the volume of passing cars and trucks at each site.

The presence of the *Black Summer* bushfires between November 2019 – February 2020 considerably altered the ambient air quality, and thus, the contribution of traffic related emissions to the overall ambient pollution load and the corresponding temporal fluctuation of the pollutants throughout each day. Consequently, days where air quality was strongly influenced by bushfire emissions were eliminated from the data. These days were identified by using the ambient $PM_{2.5}$ concentration as an indicator variable in a time series analysis, whereby the daily variation in the $PM_{2.5}$ concentration was broken down into 'trend', 'cyclical' and 'random' components. As $PM_{2.5}$ is strongly associated with traffic emissions and contributes to a daily cyclical pattern of atmospheric $PM_{2.5}$, days where the 'random' variation in $PM_{2.5}$ exceeded that of the maximum 'cyclical' variation in $PM_{2.5}$ concentration (see Pettit et al. 2020) were defined as bushfire days and excluded from analysis, as these days were not representative of Sydney's normal air quality. Data from weekdays were used for analyses, with data from weekends excluded due to differences in traffic volumes and the presence of the 'ozone weekend effect', which commonly leads to higher concentrations of O_3 on weekends in urbanised areas due to alterations in the local atmospheric VOC to NO_x ratio (Gao and Niemeier 2007; Pont an Fontan 2001; Wolff et al. 2013).

3. Results

The Eastern Distributor had an average daily (6:00 am to 6:00 pm) traffic count of 33,267 cars and 1,175 trucks in the adjacent northbound lanes over the course of sampling at this site (Transurban 2020). The section of the Hills Motorway adjacent to the biofilter arrays had an average bidirectional daily traffic (6:00 am to 6:00 pm) count of 70,985 cars and 4,691 trucks over the course of sampling at this site (Transurban 2020).

At each site, ambient concentrations of all pollutants were associated with traffic density, as expected. At the Eastern Distributor the average daily ambient $PM_{2.5}$ concentration at each 15-minute interval was significantly correlated with the passing volume of cars (r = 0.372, p = 0.012, n = 48) and trucks (r = 0.625, p = 0.000, n = 48), while the daily ambient $PM_{2.5}$ concentration at each 15-minute interval was significantly correlated with the volume of passing trucks at the Hills Motorway (r = 0.550, p = 0.000, n = 48). At the Eastern Distributor, pollutant concentrations were generally higher and exhibited greater fluctuations throughout each day, due to greater variations in traffic volume (Figures 2-3).

The average concentrations of the three pollutants detected in the effluent of all biofiltration treatments were lower than the ambient pollutant concentrations, thus all treatments had positive SPREs for all pollutants (Figures 2-3), indicating that filtration of $PM_{2.5}$, NO_2 and O_3 from the ambient air at two different roadside environments was achieved.

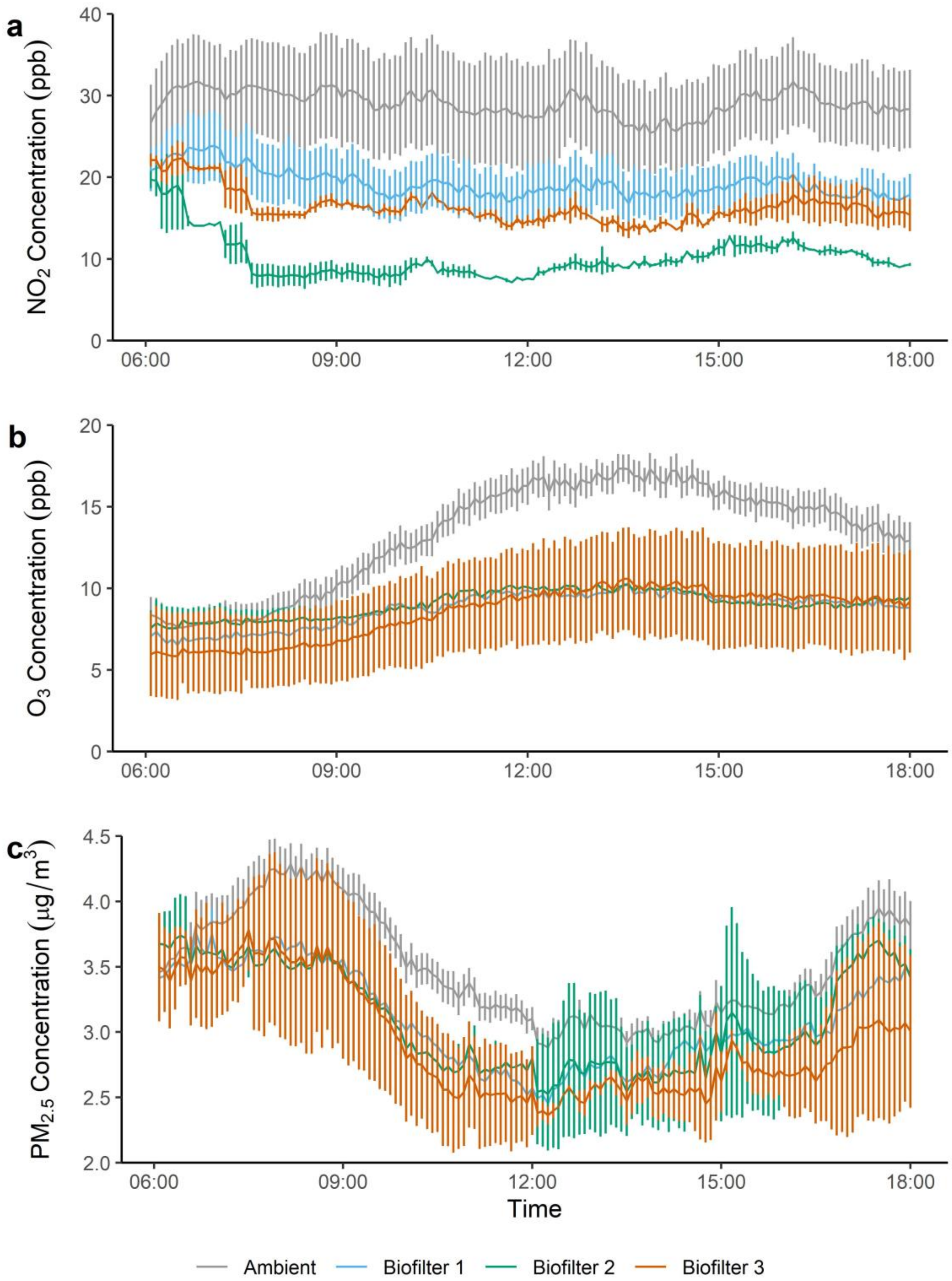

Figure 2. The average ambient and filtered effluent concentrations of air pollutants at the Eastern Distributor for each time point across the trial period of June 2019 to November 2019 (means ± SEMs). a = NO_2; b = O_3; c = $PM_{2.5}$. Biofilter 1: fans with 186.70 m^3/h flow rate at 0 Pa static pressure, Biofilter 2: fans with 186.70 m^3/h flow

rate at 0 Pa static pressure + granular activated carbon cassettes, Biofilter 3: fans with flow rate of 269.3 m^3/h at 0 Pa static pressure.

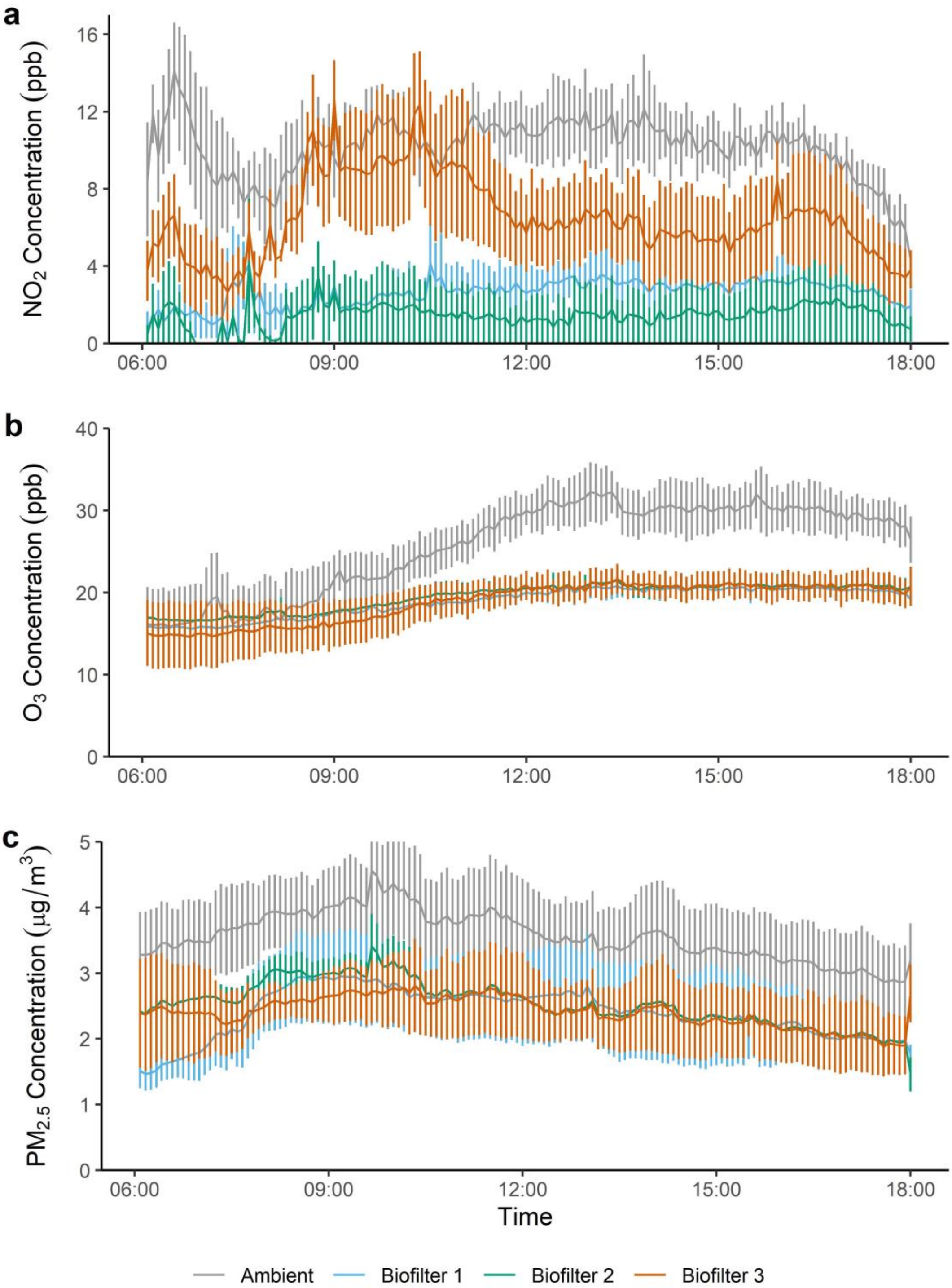

Figure 3. The average ambient and filtered effluent concentrations of air pollutants at the Hills Motorway for each time point across the trial period of November 2019

to May 2020 (means ± SEMs). a = NO_2; b = O_3; c = $PM_{2.5}$. Biofilter 1: fans with 186.70 m^3/h flow rate at 0 Pa static pressure, Biofilter 2: fans with 186.70 m^3/h flow rate at 0 Pa static pressure + granular activated carbon cassettes, Biofilter 3: fans with flow rate of 269.3 m^3/h at 0 Pa static pressure.

The average airflow through each of the plenums using 120 mm fans was 169.02 ± 4.37 m^3/h. The average airflow through plenums containing GAC was 169.01 ± 11.17 while the average airflow of plenums with 140 mm fans was 178.41 ± 22.68 m3/h.

The SPREs were taken as a function of airflow rate to calculate the CADR of each treatment (Figure 4). The plenums with larger fans and thus the highest volumetric flow rates achieved the highest CADRs for ozone and $PM_{2.5}$, while the biofilter incorporating GAC produced the largest CADR for NO_2. The CADRs of all of the pollutants however, were not statistically different amongst the biofilter treatments or sites (two-way ANOVA for each pollutant; in all cases $p > 0.05$ for both factors; Supplementary Table 1).

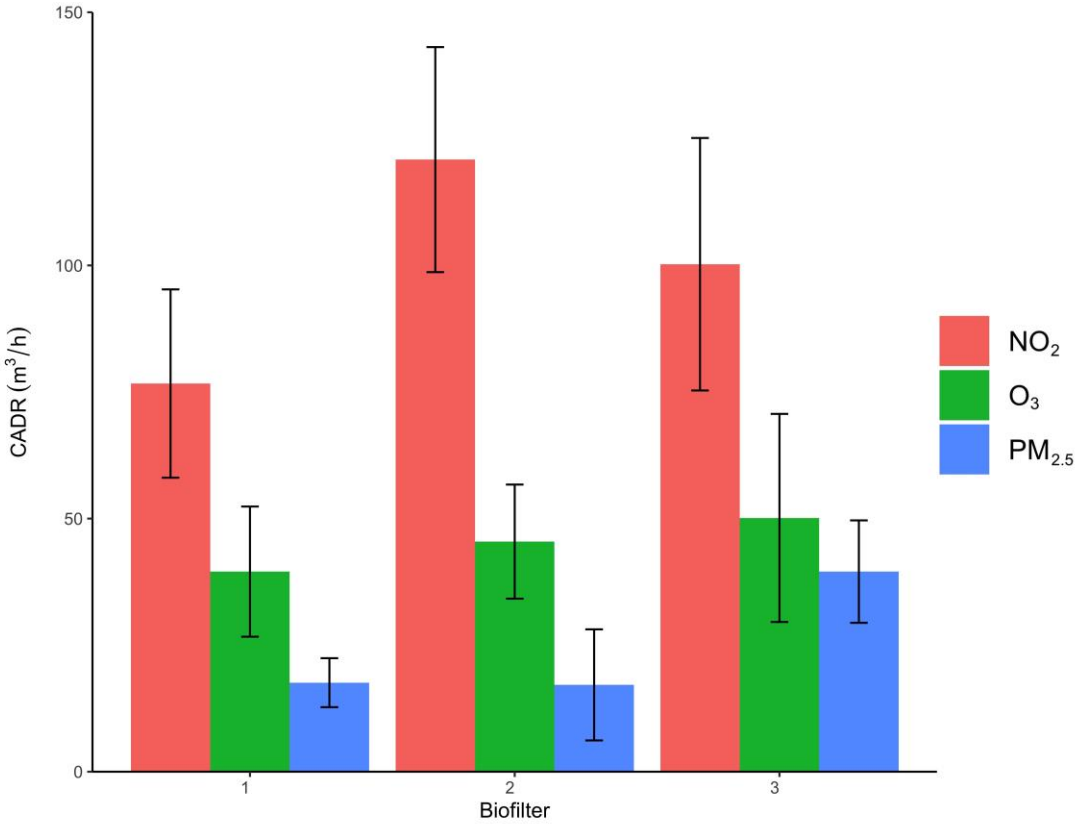

Figure 4. The average clean air delivery rates (CADRs) for 1 m^2 biofilter plenums across treatments, consolidating data from both sites (means ± SEMs). Biofilter 1: fans with 186.70 m^3/h flow rate at 0 Pa static pressure, Biofilter 2: fans with 186.70 m^3/h flow rate at 0 Pa static pressure + GAC cassettes, Biofilter 3: fans with flow rate of 269.3 m^3/h at 0 Pa static pressure (n = 14, 5 and 5 independent plenums for Biofilters 1, 2 and 3 respectively).

A series of Pearson's correlations assessing the association between ambient concentrations of the three pollutants and the SPREs of each treatment showed that almost all treatments exhibited statistically significant positive relationships between removal efficiency and pollutant concentration (Table 2). As the ambient concentration of all pollutants increased, the SPRE of all treatments increased as well. This trend was particularly strong for O_3 across all biofilter treatments.

Table 2. Pearson's correlation matrix of associations between SPRE and ambient pollutant concentration. n = 144 observations for each correlation. * indicates statistical significance whereby p = <0.05. Pearson's r values are shown.

Treatment	**Ambient NO_2 concentration**	**Ambient O_3 concentration**	**Ambient $PM_{2.5}$ concentration**
Plenum SPRE	0.166*	0.980*	0.203*
GAC SPRE	0.141	0.976*	0.572*
140 mm Fan SPRE	0.165*	0.946*	0.167*

4. Discussion

Mitigating air pollution resulting from traffic emissions is becoming increasingly problematic in urban regions, particularly so in built-up areas, where population exposure to urban air pollution is likely to increase in the next decade as urban development disproportionately occurs along main road sites (Paton-Walsh et al. 2019). Most current air pollution mitigation strategies aim to reduce source emissions, with varying effectiveness on ambient air quality (Carslaw et al. 2016; Zhang and Gu 2013), but there are no methods currently employed on a medium to large scale for the active reduction of roadside pollution *in situ*. This work represents the first field assessment of a novel botanical biofiltration system for the mitigation of NO_2, O_3 and $PM_{2.5}$ from traffic emissions. In all cases, the concentrations of these pollutants were considerably reduced by the biofilter treatments, so that the concentrations of all pollutants were lower in the effluent air stream than in the ambient air.

4.1 NO_2 filtration

The concentration of NO_2 in the effluent air was considerably lower than ambient, irrespective of the ambient NO_2 concentrations, with average SPREs across all sampling periods ranging from 57.81-75.63%, depending on the treatment. While there were clear differences in the ambient concentration profile of the pollutants between the two sites, the average daily temporal pattern of NO_2 was consistent within sites, with neither site showing clear fluctuations in NO_2 concentration related to traffic volume or sunlight intensity.

When standardised by substrate volume, the NO_2 CADRs recorded in this study are substantially higher (by ~20-30%) than those detected under elevated NO_2 concentrations in Pettit et al. (2019b), most likely due to the use of different systems and pollutant inlet concentrations between the studies. The volumetric airflow rate has been a critical parameter for determining the optimal CADR of biofilters (Guieysse et al. 2008). This has most commonly been explored through the removal of VOCs, whereby larger airflow rates lead to reduced SPREs but often increased CADRs by increasing the volume of air that is processed (e.g. Wang and Zhang 2011). In this case however, the different airflow rates provided by different fans did not lead to significant differences in the NO_2 CADR amongst the treatments, and it is likely that greater variation in volumetric flow rates will be required to produce significant differences in CADRs. Additionally, the use of GAC did not significantly increase the NO_2 SPRE, in contrast to previous studies where activated carbon has been used successfully to filter NO_2 from contaminated air streams (Yoo et al. 2015). Nonetheless, the GAC augmented biofiltration treatment used in this experiment did not considerably reduce the airflow rate (i.e. volumetric airflow rates where very similar to that of the plenums without GAC cassettes), and thus did not compromise the CADRs. The use of different activated carbon-based adjunct filter designs (modifications to GAC type and volume) requires further exploration to thoroughly determine whether effects similar to that observed in laboratory studies (Yoo et al. 2015) can be achieved.

As this work did not measure the ambient or filtered concentrations of VOCs, this remains an important consideration for future research. Previous work conducted in laboratory scale experiments (Pettit et al. 2019c; Treesubsuntorn and Thiravetyan 2018) and indoor trials (Darlington et al. 2001; Pettit et al. 2019a; Wang and Zhang 2011) has highlighted that botanical biofilters are efficient at filtering a range of different VOCs, however it remains unknown how such systems can filter specific VOC mixtures and concentrations associated with traffic emissions. Furthermore, it is important to monitor any possible VOC emissions emitted by the biological components of the system as there is potential for VOCs to react with NO_2 to lead to the formation of O_3 (Atkinson 2000).

4. 2 O_3 filtration

The ambient concentration of O_3 generally increased through the day at both sites – as is commonly observed in urban areas (Pancholi et al. 2018; Warmiński and Bęś 2018). Although the concentration of NO_2 was higher at the Eastern Distributor site than the Hills Motorway, the concentration of O_3 was higher at the Hills

Motorway than the Eastern Distributor, which may reflect the seasonal differences in sampling periods between the two sites (Warmiński and Bęś 2018). In all cases, the concentration of O_3 in the effluent air stream generally started out equal to the 6 am ambient O_3 concentrations, and remained at this level, while the ambient concentration rose throughout the day. Although it is possible that there is a threshold concentration of O_3 that cannot be filtered with the system tested here, the different concentrations of O_3 at each site, in both the ambient and effluent air streams of all biofilter treatments suggests such possible effects may be concentration dependent. Both NO_2 and O_3 are photo-chemically sensitive under sunlight conditions (Atkinson 2000). As the plenum intercepted sunlight, it is difficult to determine what effect the plenum alone may have had on these pollutants, however the contribution of any possible effects on the NO_2 or O_3 concentrations resulting from shading are likely to be minimal due to the short residence time of effluent gas within the plenums (~2 s).

Although the botanical biofiltration of NO_2 and O_3 has been observed in laboratory studies using spiked pollutant concentrations (Pettit et al. 2019b), this work represents the first instance whereby the continuous removal of traffic sourced pollutants by botanical biofiltration has been recorded. The *in situ* measurements from this study provide a more accurate estimate of the air cleaning potential of botanical biofiltration than scaled up estimates from laboratory studies, and reflect their likely performance for their intended purpose.

The fate of the filtered pollutants, and their ramifications for the biofiltration system, remains unclear. Previous work has noted the potential production of nitric acid within the growth substrate, as NO_2 combines with irrigation water to produce nitric acid and NO (Zheng et al. 2016). Alternatively, the co-biofiltration of O_3 and NO_2 may affect a form of pH control due to the generation of alkaline products from

O_3 biofiltration (Maldonado-Diaz and Arriaga 2015). Although it was not the intention to assess filtration products within the media in this study, any changes in substrate pH were insufficient to visibly affect plant health or influence system performance.

4. 3 $PM_{2.5}$ filtration

The average $PM_{2.5}$ CADRs through the botanical biofilters were lower than those of the gaseous pollutants. Irga et al.'s (2017) laboratory study used a spiked dose of particles from combusting diesel fuel, and observed greater botanical biofilter $PM_{2.5}$ SPREs than this study (~48%). It is unknown whether the chemical composition and size distribution of particles differ between these studies, and it is possible that variation in these characteristics may have led to these discrepancies, as larger particles are removed with greater efficiency (Pettit et al. 2017). Nonetheless, the SPREs presented in the current work reflect the removal of particle compositions encountered in roadside environments. Although there were no significant differences in the $PM_{2.5}$ CADR amongst the treatments using different airflow rates in this study, Irga et al. (2017) found that the rate constant of $PM_{2.5}$ concentration decay increased with volumetric flow rate through the filter until a threshold airflow rate was reached. It is possible similar effects were not observed in this experiment due to the relatively small differences in airflow rates amongst the treatments. The current findings also show that the $PM_{2.5}$ SPRE will vary throughout the day, as the concentration of $PM_{2.5}$ in the effluent airstreams closely mirrored the fluctuating pattern of the $PM_{2.5}$ inlet concentration at both sites.

4.4 Incorporation into urban design and future developments

The results from this study demonstrate proof of concept for *in situ* botanical biofiltration, and suggest that botanical biofilters may be an effective solution to help mitigate air pollution exposure. With the tested biofilter systems, however, the pollutant reduction effects are unlikely to impact the ambient air quality outside of the zone immediately adjacent to the biofilter array. The implementation of larger arrays in targeted locations will thus be required to have such an effect, and while the relationship between CADR and wall size is clear, the relationship between wall size and ambient air quality effect remains untested at this stage. There is considerable potential to implement large green walls, since such infrastructure consumes relatively little space at street level. In the case of the current experiment, the size of the green walls could be considerably increased by extending their height; in this regard, the green wall would consume the same ground footprint yet have a larger area and filtration capacity.

Careful site selection will likely be needed to obtain effective biofiltration, and thus realize the greatest benefits in ambient air quality enhancement. While the ambient pollution profile may influence filtration efficiency, the urban geometry and airflow characteristics of the site will affect both the dispersion of air pollution emissions (Di Sabatino et al. 2013) and the dispersion of filtered air. Environments where the dispersion of air pollution emissions is limited, such as car parks and traffic tunnels, promote the accumulation of air pollution, and thus the use of botanical biofilters may be of considerable value in such locations. Additionally, botanical biofilters may find value in environments where other forms of greening, such as trees, cannot be used. Nonetheless, the positive association between removal efficiency and ambient pollution concentration detected in the current research suggests that botanical biofilters are most effective in those environments where they are most needed. Although positive associations between SPRE and ambient concentrations were detected across the range of ambient pollution concentrations observed in this study, previous work testing SPREs at higher pollution concentrations has shown inverse relationships between these variables (Pettit et al. 2020), and further work is still required to understand the complex relationship between biofilter pollutant removal efficiency across the range of relevant ambient concentrations.

It is clear that different forms of urban greening are associated with different effects on ambient air pollution concentrations. Passive green walls have been recommended as a suitable green infrastructure for reducing PM concentrations through the deposition of PM onto plant foliage, without affecting the air exchange between the street canyon and air above it (Abhijith et al. 2017; Litschke and Kuttler 2008). Furthermore, passive walls are able to alter the flow and dispersion patterns of air pollutants, so that pedestrian pollutant exposure may be reduced in open road conditions (Abhijith et al. 2017). The air quality reductions detected in our study were simply the result of biofiltration, and future work, with the use of modified and larger active botanical biofilters, is needed to determine the effect of these combined mechanisms on ambient pollutant concentrations. While the behaviour of air pollution in the atmosphere is commonly modelled, the concept of modelling the dispersion and behaviour of ‘clean air’ is a novel concept and thus *de novo* research is necessary to truly assess biofilter effects on ambient air quality.

5. Conclusion

This work has demonstrated the potential for botanical biofilters to filter traffic associated air pollutants – NO_2, O_3 and $PM_{2.5}$ – from roadside environments. Clean air delivery rates of up to 121 m^3 /h, 50 m^3 /h and 40 m^3 /h per m^2 of active green wall biofilter were achieved for the three pollutants respectively, with pollutant removal efficiency positively correlated with their ambient concentrations. On the basis of this research, several infrastructure-scale systems are planned for installation in critical locations around Australia. Future work will thus aim to assess the influence of these systems on the general ambient air quality conditions experienced by populations residing proximal to the biofilters.

Funding

This work was supported by Transurban Ltd and Jobs for NSW [F19 7192]. T. Pettit and R. Fleck are supported by Australian Government Research Training Program Scholarships. P.J. Irga is supported by the University of Technology Sydney (UTS) Chancellor's Postdoctoral Research Fellowship scheme, facilitated through The Centre for Green Technology.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

Abhijith, K.V., Kumar, P., Gallagher, J., McNabola, A., Baldauf, R., Pilla, F., Broderick, B., Di Sabatino, S. and Pulvirenti, B., 2017. Air pollution abatement performances of green infrastructure in open road and built-up street canyon environments–A review. *Atmospheric Environment*, *162*, pp.71-86.

Abhijith, K.V. and Kumar, P., 2019. Field investigations for evaluating green infrastructure effects on air quality in open-road conditions. *Atmospheric Environment*, *201*, pp.132-147.

Aeroqual Limited, 2019. Aeroqual AQY 1. Accessed from https://www.aeroqual.com/wp-content/uploads/AQY1-Spec-Sheet-1.pdf on 6/4/2020.

Atkinson, R., 2000. Atmospheric chemistry of VOCs and NOx. *Atmospheric Environment*, *34*(12-14), pp.2063-2101.

Attal, E., Côté, N., Shimizu, T. and Dubus, B., 2017. Optimization of acoustic absorption by green walls made of foliage and substrate. *The Journal of the Acoustical Society of America*, *141*(5), pp.3691-3691.

Australian Bureau of Statistics, 2019. 3218.0 - Regional Population Growth, Australia, 2017-18. Accessed on 3/4/2020.
https://web.archive.org/web/20190518102422/http://www.abs.gov.au/ausstats/abs@.nsf/Latestproducts/3218.0Main%20Features12017-18?opendocument&tabname=Summary&prodno=3218.0&issue=2017-18&num=&view=

Carslaw, D. C., Murrells, T. P., Andersson, J. & Keenan, M., 2016. Have vehicle emissions of primary NO2 peaked? *Faraday Discussions 189***,** pp.439-454.

Cowie, C.T., Garden, F., Jegasothy, E., Knibbs, L.D., Hanigan, I., Morley, D., Hansell, A., Hoek, G. and Marks, G.B., 2019. Comparison of model estimates from an intra-city land use regression model with a national satellite-LUR and a regional Bayesian Maximum Entropy model, in estimating NO2 for a birth cohort in Sydney, Australia. *Environmental Research*, *174*, pp.24-34.

Crawford, J., Cohen, D.D., Griffiths, A.D., Chambers, S.D., Williams, A.G. and Stelcer, E., 2017. Impact of atmospheric flow conditions on fine aerosols in Sydney, Australia. *Aerosol and Air Quality Research*, *17*(7), pp.1746-1759.

Darlington, A.B., Dat, J.F. and Dixon, M.A., 2001. The biofiltration of indoor air: air flux and temperature influences the removal of toluene, ethylbenzene, and xylene. *Environmental Science & Technology*, *35*(1), pp.240-246.

Di Sabatino, S., Buccolieri, R. and Salizzoni, P., 2013. Recent advancements in numerical modelling of flow and dispersion in urban areas: a short review. *International Journal of Environment and Pollution 7*, *52*(3-4), pp.172-191.

European Environmental Agency, 2011. Air Quality in Europe Cities and Towns. No 12/2011. European Environmental Agency, Copenhagen.

Gallagher, J., Baldauf, R., Fuller, C.H., Kumar, P., Gill, L.W. and McNabola, A., 2015. Passive methods for improving air quality in the built environment: a review of porous and solid barriers. *Atmospheric Environment*, *120*, pp.61-70.

Gao, H.O. and Niemeier, D.A., 2007. The impact of rush hour traffic and mix on the ozone weekend effect in southern California. *Transportation Research Part D: Transport and Environment*, *12*(2), pp.83-98.

Gromke, C., Buccolieri, R., Di Sabatino, S. and Ruck, B., 2008. Dispersion study in a street canyon with tree planting by means of wind tunnel and numerical investigations–evaluation of CFD data with experimental data. *Atmospheric Environment*, *42*(37), pp.8640-8650.

Guieysse, B., Hort, C., Platel, V., Munoz, R., Ondarts, M. and Revah, S., 2008. Biological treatment of indoor air for VOC removal: Potential and challenges. *Biotechnology Advances*, *26*(5), pp.398-410.

Horoshenkov, V.K., Khan, A., Benkreira, H., Mandon, A. and Rohr, R., 2011. Acoustic properties of green walls with and without vegetation. *The Journal of the Acoustical Society of America*, *130*(4), pp.2317-2317.

Ibrahim, I.Z., Chong, W.T., Yusoff, S., Wang, C.T., Xiang, X. and Muzammil, W.K., 2019. Evaluation of common indoor air pollutant reduction by a botanical indoor air biofilter system. *Indoor and Built Environment*, p.1420326X19882080.

Irga, P.J., Paull, N.J., Abdo, P. and Torpy, F.R., 2017. An assessment of the atmospheric particle removal efficiency of an in-room botanical biofilter system. *Building and Environment*, *115*, pp.281-290.

Irga, P.J., Pettit, T.J. and Torpy, F.R., 2018. The phytoremediation of indoor air pollution: a review on the technology development from the potted plant through to functional green wall biofilters. *Reviews in Environmental Science and Biotechnology*, *17*(2), pp.395-415.

Jeanjean, A.P., Buccolieri, R., Eddy, J., Monks, P.S. and Leigh, R.J., 2017. Air quality affected by trees in real street canyons: The case of Marylebone neighbourhood in central London. *Urban Forestry & Urban Greening*, *22*, pp.41-53.

Karner, A.A., Eisinger, D.S. and Niemeier, D.A., 2010. Near-roadway air quality: synthesizing the findings from real-world data. *Environmental Science & Technology*, *44*(14), pp.5334-5344.

Litschke, T. and Kuttler, W., 2008. On the reduction of urban particle concentration by vegetation–a review. *Meteorologische Zeitschrift*, *17*(3), pp.229-240.

Maldonado-Diaz, G. and Arriaga, S., 2015. Biofiltration of high formaldehyde loads with ozone additions in long-term operation. *Applied Microbiology and Biotechnology*, *99*(1), pp.43-53.

Manso, M. and Castro-Gomes, J., 2015. Green wall systems: A review of their characteristics. *Renewable and Sustainable Energy Reviews*, *41*, pp.863-871.

NSW Environmental Protection Authority (EPA), 2012. Tracking Sources of Air Pollution in NSW Communities: Air emissions inventory for the Greater Metropolitan Region of NSW. https://www.epa.nsw.gov.au/-/media/epa/corporate-site/resources/air/120404aeibrochure.pdf?la=en&hash=47C46BD763F1580267FC4BD92133FEFC0935C807 accessed on 18/6/2020.

NSW Health, 2014. TP03: Health Effects of Traffic-Related Air Pollution. NSW Government, Advisory Committee on Tunnel Air Quality https://www.chiefscientist.nsw.gov.au/__data/assets/pdf_file/0004/52987/Road-Tunnels_TP03_Health_effects_of_traffic_related_air_pollution.pdf

Nowak, D.J., Crane, D.E. and Stevens, J.C., 2006. Air pollution removal by urban trees and shrubs in the United States. *Urban Forestry & Urban Greening*, *4*(3-4), pp.115-123.

Pascal, M., Corso, M., Chanel, O., Declercq, C., Badaloni, C., Cesaroni, G., Henschel, S., Meister, K., Haluza, D., Martin-Olmedo, P. and Medina, S., 2013. Assessing the public health impacts of urban air pollution in 25 European cities: results of the Aphekom project. *Science of the Total Environment*, *449*, pp.390-400.

Pancholi, P., Kumar, A., Bikundia, D.S. and Chourasiya, S., 2018. An observation of seasonal and diurnal behavior of O3–NOx relationships and local/regional oxidant

(OX= O3+ NO2) levels at a semi-arid urban site of western India. *Sustainable Environment Research*, *28*(2), pp.79-89.

Pasquier, A. and André, M., 2017. Considering criteria related to spatial variabilities for the assessment of air pollution from traffic. *Transportation Research Procedia*, *25*, pp.3354-3369.

Paton-Walsh, C., Rayner, P., Simmons, J., Fiddes, S.L., Schofield, R., Bridgman, H., Beaupark, S., Broome, R., Chambers, S.D., Chang, L.T.C. and Cope, M., 2019. A Clean Air Plan for Sydney: An Overview of the Special Issue on Air Quality in New South Wales. *Atmosphere*, *10*(12), p.774.

Petrova, S.T., 2020. Efficiency of *Pinus nigra* JF Arnold in removing pollutants from urban environment (Plovdiv, Bulgaria). *Environmental Science and Pollution Research*, *27*(31), pp.39490-39506.

Pettit, T., Irga, P.J., Abdo, P. and Torpy, F.R., 2017. Do the plants in functional green walls contribute to their ability to filter particulate matter?. *Building and Environment*, *125*, pp.299-307.

Pettit, T., Irga, P.J. and Torpy, F.R., 2018a. Towards practical indoor air phytoremediation: A review. *Chemosphere*, *208*, pp.960-974.

Pettit, T., Irga, P.J. and Torpy, F.R., 2018b. Functional green wall development for increasing air pollutant phytoremediation: Substrate development with coconut coir and activated carbon. *Journal of Hazardous Materials*, *360*, pp.594-603.

Pettit, T., Irga, P.J. and Torpy, F.R., 2019a. The in situ pilot-scale phytoremediation of airborne VOCs and particulate matter with an active green wall. *Air Quality, Atmosphere & Health*, *12*(1), pp.33-44.

Pettit, T., Irga, P.J., Surawski, N.C. and Torpy, F.R., 2019b. An Assessment of the Suitability of Active Green Walls for NO2 Reduction in Green Buildings Using a Closed-Loop Flow Reactor. *Atmosphere*, *10*(12), p.801.

Pettit, T., Bettes, M., Chapman, A.R., Hoch, L.M., James, N.D., Irga, P.J., and Torpy, F.R, 2019c. The botanical biofiltration of VOCs with active airflow: is removal efficiency related to chemical properties?. *Atmospheric Environment*, *214*, p.116839.

Pettit, T., Irga, P.J. and Torpy, F.R., 2020. The botanical biofiltration of elevated air pollution concentrations associated the Black Summer wildfire natural disaster. *Journal of Hazardous Materials Letters*, 1, p.100003.

Pont, V. and Fontan, J., 2001. Comparison between weekend and weekday ozone concentration in large cities in France. *Atmospheric Environment*, *35*(8), pp.1527-1535.

Roads and Maritime Services, Transport for NSW, NSW Government. 2018. The Eastern Distributor. https://www.rms.nsw.gov.au/documents/about/environment/air/ebook/files/assets/downloads/page0006.pdf accessed on 17/6/2020.

Salmond, J.A., Williams, D.E., Laing, G., Kingham, S., Dirks, K., Longley, I. and Henshaw, G.S., 2013. The influence of vegetation on the horizontal and vertical distribution of pollutants in a street canyon. *Science of the Total Environment*, *443*, pp.287-298.

Tong, Z., Baldauf, R.W., Isakov, V., Deshmukh, P. and Zhang, K.M., 2016. Roadside vegetation barrier designs to mitigate near-road air pollution impacts. *Science of the Total Environment*, *541*, pp.920-927.

Transurban, 2020. NSW Toll Road Data. https://nswtollroaddata.com/data-download/ accessed on 18/5/2020.

Treesubsuntorn, C. and Thiravetyan, P., 2018. Botanical biofilter for indoor toluene removal and reduction of carbon dioxide emission under low light intensity by using mixed C3 and CAM plants. *Journal of Cleaner Production*, *194,* pp.94-100.

Venetsanos, A.G., Bartzis, J.G., Andronopoulos, S. and Vlachogiannis, D., 2001. Vehicle effects on street canyon air pollution pattern. *WIT Transactions on Ecology and the Environment*, *47*, pp.1-10.

Venkatram, A. and Schulte, N., 2018. *Urban Transportation and Air Pollution*. Elsevier: Amsterdam, Netherlands.

Vos, P.E., Maiheu, B., Vankerkom, J. and Janssen, S., 2013. Improving local air quality in cities: to tree or not to tree?. *Environmental Pollution*, *183*, pp.113-122.

Wang, Z. and Zhang, J.S., 2011. Characterization and performance evaluation of a full-scale activated carbon-based dynamic botanical air filtration system for improving indoor air quality. *Building and Environment*, *46*(3), pp.758-768.

Warmiński, K. and Bęś, A., 2018. Atmospheric Factors Affecting a Decrease in the Night-Time Concentrations of Tropospheric Ozone in a Low-Polluted Urban Area. *Water, Air, & Soil Pollution*, *229*(11), p.350.

Wolff, G.T., Kahlbaum, D.F. and Heuss, J.M., 2013. The vanishing ozone weekday/weekend effect. *Journal of the Air & Waste Management Association*, *63*(3), pp.292-299.

World Health Organisation, 2019. Healthy Environment for Healthier Populations: Why Do They Matter and What Can We Do? Geneva https://www.who.int/publications-detail/healthy-environments-for-healthier-populations-why-do-they-matter-and-what-can-we-do

Yoo, J.Y., Park, C.J., Kim, K.Y., Son, Y.S., Kang, C.M., Wolfson, J.M., Jung, I.H., Lee, S.J. and Koutrakis, P., 2015. Development of an activated carbon filter to remove NO2 and HONO in indoor air. *Journal of Hazardous Materials*, *289*, pp.184-189.

Zhang, Y., Mo, J., Li, Y., Sundell, J., Wargocki, P., Zhang, J., Little, J.C., Corsi, R., Deng, Q., Leung, M.H. and Fang, L., 2011. Can commonly-used fan-driven air cleaning technologies improve indoor air quality? A literature review. *Atmospheric Environment*, *45*(26), pp.4329-4343.

Zhang, Y. & Gu, Z., 2013. Air quality by urban design. *Nat Geoscience,* 6, pp.506-506.

Zheng, M., Li, C., Liu, S., Gui, M. and Ni, J., 2016. Potential application of aerobic denitrifying bacterium Pseudomonas aeruginosa PCN-2 in nitrogen oxides (NOx) removal from flue gas. *Journal of Hazardous Materials, 318,* pp.571-578.

Supplementary Table 1: Results comparing the CADRs amongst the three biofilter treatments and the two sites. A two factor ANOVA was used for each air pollutant.

Pollutant	Source	df	*F*	*p*
NO_2	Site	1	3.597	0.076
	Treatment	2	0.541	0.593
	Site x treatment	2	0.235	0.793
O_3	Site	1	2.248	0.151
	Treatment	2	0.507	0.611
	Site x treatment	2	0.435	0.654

$PM_{2.5}$	Site	1	0.107	0.747
	Treatment	2	1.885	0.181
	Site x treatment	2	0.526	0.6