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Comparison of urban stormwater plant biofilter designs for nutrient and metal removal in South Africa

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ABSTRACT

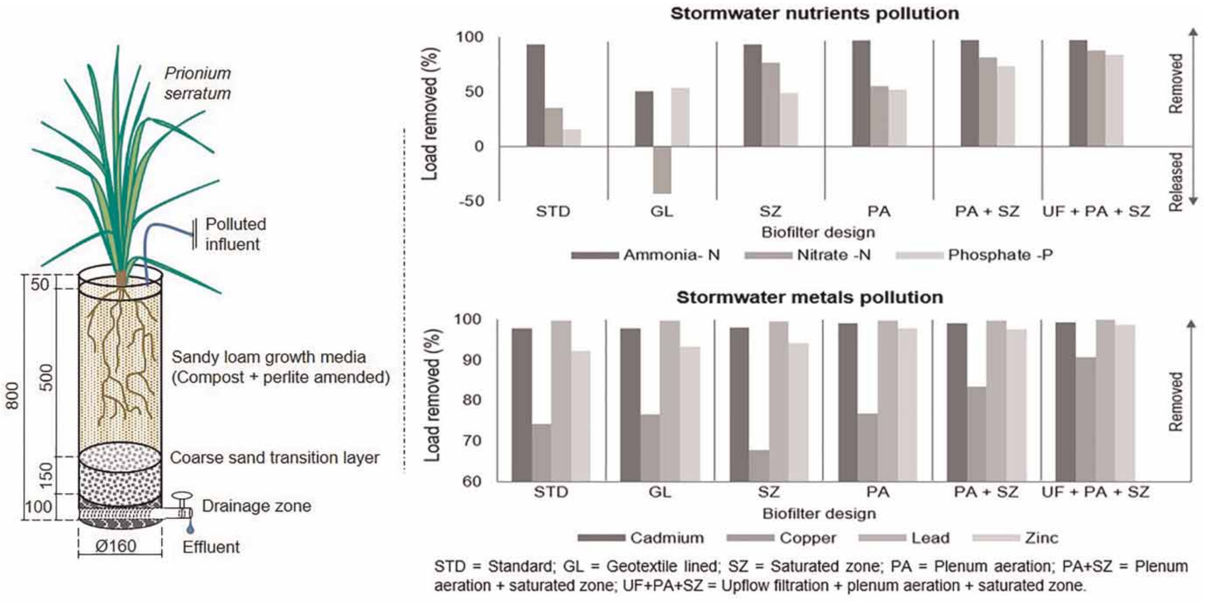
This paper presents a comparison of six plant biofilter designs for urban stormwater quality improvement and reports on their performances. Thirty-six columns were populated with the endemic South African plant *Prionium serratum*, representing plant biofilter designs that incorporate different pollutant removal mechanisms in the biofiltration process. The experimental biofilter columns were subjected to low, typically observed and high urban nutrient and metal synthetic stormwater pollution for five months. Significant loads of NH_3-N and dissolved Cd, Pb and Zn were removed, whereas removal of NO_3^--N, PO_4^{3-}-P and dissolved Cu was more variable. The most efficient design was found to include standard plant biofiltration techniques with upflow filtration, plenum aeration and a saturated zone supporting anaerobic microbial activity. It was found that the most efficient design removed on average 96% of urban stormwater nutrient and metal loads.

Key words: metals removal, nutrients removal, plant biofilter design, urban stormwater runoff pollution

HIGHLIGHTS

- Assessment of various plant biofilter designs (air plenum, different media, geotextile inclusion, saturated zone inclusion).
- Inclusion of endemic South African plant *Prionium serratum* and nationally available materials.
- Significant NH_3-N and dissolved Cd, Pb and Zn removal.
- Varied NO_3^--N, PO_4^{3-}-P and dissolved Cu removal.

GRAPHICAL ABSTRACT



INTRODUCTION

Rapid urbanisation has led to a significant increase in runoff volumes, peak flows, sedimentation and nutrient and metal pollution to urban stormwater, resulting in urban watercourse degradation (Line & White 2007; Al-Ameri *et al.* 2018). Due to the absorption capacity of water, many substances are collected by stormwater runoff, making urban stormwater one of the primary pollution vectors to natural water systems (Abbasi & Abbasi 2011). To mitigate these impacts, improved urban stormwater management strategies such as water sensitive urban design (WSUD) and low impact development (LID), are increasingly preferred in South Africa (Armitage *et al.* 2014). The WSUD approach promotes the systematic integration of green infrastructure (GI) and nature-based solutions into urban spaces as key components of climate change adaptation and runoff mitigation strategies (Dumitru & Wendling 2021). Within GI, sustainable yet effective plant biofiltration systems provide both water quality and quantity benefits to the dense and confined urban area (Payne *et al.* 2015). Plant biofiltration makes use of vegetation and soil infiltration to attenuate runoff flows (Hatt *et al.* 2009); and improve water quality (Shrestha *et al.* 2018) through particulate discharge, filtration, sorption, plant and microbial uptake and evapotranspiration (Wadzuk *et al.* 2015; Wang *et al.* 2018a).

Both laboratory and field plant biofiltration studies investigating nutrients have demonstrated effective ammonia (NH_3) removal (Davis *et al.* 2001). However, more variable removal efficiencies have been reported for nitrate (NO_3^-) (Hatt *et al.* 2009) and orthophosphate (PO_4^{3-}) (Hsieh *et al.* 2007). In some studies high NO_3^- (Bratieres *et al.* 2008) and PO_4^{3-} (Dietz & Clausen 2006) leaching was reported, resulting in poor overall nutrient removal. Leaching of NO_3^--N, the primary obstacle for effective nutrient removal in GI initiatives, is due to inadequate anaerobic denitrification (Zinger *et al.* 2013). For PO_4^{3-}-P, leaching into discharged stormwater due to weathering and mineralisation processes have been found to increase in the presence of organic matter (Henderson *et al.* 2007), further exacerbated with compost amendments (Mullane *et al.* 2015). Enhanced nutrient removal by vegetation occurs either directly through plant uptake, root retention and soil maintenance or indirectly through enhanced microbial activity in the root zone (Bratieres *et al.* 2008).

In stormwater runoff, dissolved metals contribute a large proportion of pollutants with those of most interest, viz. cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn) present naturally or caused by anthropogenic activities (Hocaoglu-Ozyigit & Genc 2020). In biofilters, the primary metal removal pathway is sorption to the upper layer of the growth media until breakthrough or infiltration failure occurs as a result of clogging (Le Coustumer *et al.* 2012). Varying metal removal has been demonstrated, particularly in anaerobic conditions, which have been found to decrease redox potential (Dietz & Clausen 2006), while Cu, Pb and Zn are further influenced by organic matter (Warren & Haack 2001; Al-Ameri *et al.* 2018) and may leach (Blecken *et al.* 2010; Lenth & Dugapolski

2011). The contribution by vegetation, though to a lesser extent when compared with unvegetated mechanisms (Blecken *et al.* 2009), is the extraction of micronutrients Cu and Zn as well as non-essential, potentially toxic Cd and Pb (Tangahu *et al.* 2011).

In biofilters stocked with vegetation, pollutants are removed through physical, chemical or biological processes, enhancing water quality (Wang *et al.* 2018b) and providing a permanent removal mechanism via harvesting (Kumar *et al.* 2017); this highlights the importance of vegetation for urban stormwater quality improvement (Bratieres *et al.* 2008). The processes and pathways affecting nutrient and metal removal are complex and vary between biofilter designs and operational conditions, making it difficult to consistently maintain overall treatment performance, particularly for dissolved pollutants (Hunt *et al.* 2008).

Inadequate dissolved nutrient and metal treatment negates biofiltration initiatives as pollutants are most bioavailable in these forms (Maniquiz-Redillas & Kim 2016). Therefore, removing particulate pollutants only will not adequately reduce the negative impacts on water systems (Winston *et al.* 2017). In recent years adaptations in filter depth (Bratieres *et al.* 2008), filter media (Reddy *et al.* 2014), carbon sources (Blecken *et al.* 2009), plant species (Payne *et al.* 2018), stepped retention (Wang *et al.* 2017), intermittent dry-wet cycles (Hatt *et al.* 2007) and anaerobic saturation (Zinger *et al.* 2013) have been made to optimise biofiltration. Such adaptations demonstrated improved performance of certain biofilter elements, however, no obvious differences and some unsatisfactory findings were also observed (Wang *et al.* 2018b). For example, biofilters integrating a saturated zone for enhanced denitrification (the preferred method for optimisation) have in some cases shown no improvement (Dietz & Clausen 2006) and in others leached pollutants (Zinger *et al.* 2013).

Despite increased popularity for GI application (Kim & Song 2019), design of scientifically based plant biofiltration systems for overall pollutant removal has not evolved greatly since integrating a saturated zone as the primary removal mechanism of NO_3^-. Therefore, to advance the knowledge on plant biofilter design for stormwater runoff quality improvement in South Africa, the performance of novel plenum aeration and upflow filtration in combination with zone saturation was investigated in this study additional to more common biofilter designs. Here a proven efficient South African endemic phytoremediator, viz. *Prionium serratum* (See Jacklin *et al.* 2021a) was included in the designs.

This experiment forms part of a larger study on South African urban stormwater management, its underutilization of WSUD, potential advancements, and development of a conceptual urban stormwater biofilter model.

METHODS

Engineered plant biofilter design

Thirty-six columns populated with *P. serratum* were constructed from Ø160 mm PVC piping, for the introduction of various engineered media and removal pathways to each of the six plant biofilter designs (see Table 1 below), in duplicate and for three different pollutant dosing strengths (low, typically observed and high).

All columns received unpolluted municipal tap water for nine months to allow the establishment of *P. serratum* prior to synthetic stormwater dosing and testing. Similar to the methods adopted by Hatt *et al.* (2007), this period of municipal supply allowed for the discharge of labile nutrients out of the plant biofilter columns to achieve lower and greater consistency of nutrient concentrations within the effluent from the experimental biofilters. Due to potential water sensitivity in the areas where biofilters may be required, effort was made to limit water wastage as much as possible during the nine-month establishment period, resulting in low effluent volumes.

Table 1 | Overview of the six biofilter designs under study

Design identifier	Biofilter description (All designs integrated a standard growth media layer with *Prionium serratum*)	Additional to soil media (All growth media integrated sandy loam amended with compost+perlite)
STD	Standard stormwater plant biofilter	None
GL	Geotextile lined	None
PA	Plenum aeration	Zeolite
SZ	Saturated zone	Vermiculite+attapulgite
PA+SZ	Plenum aeration+saturated zone	Zeolite+vermiculite+attapulgite
UF+PA+SZ	Upflow filtration+plenum aeration+saturated zone	Zeolite+vermiculite+attapulgite

In comparing effluent values taken directly after biofilter construction (mean concentration: NH_3 -*N*=1.54 mg/L, NO_3^--*N*=4.81 mg/L and PO_4^{3-}-*P*=5.97 mg/L; mean load: NH_3-*N*=0.15 mg, NO_3^--*N*=0.48 mg and PO_4^{3-}-*P*=0.59 mg) with values recorded in the ninth month of unpolluted tap water dosing (mean concentration: NH_3-*N*=0.039 mg/L, NO_3^--*N*=1.67 mg/L and PO_4^{3-}-*P*=1.75 mg/L; mean load: NH_3-*N*=0.0039 mg, NO_3^--*N*=0.16 mg and PO_4^{3-}-*P*=0.17 mg), effluent concentrations and loads reached stable and low average levels, indicating a limit to leaching potential from the media after the nine month flushing period. This mobilisation and subsequent discharge of labile nutrients was accomplished with significantly less water required for flushing than utilised in Hatt *et al.* (2007), benefitting water scarce initiatives. Following this period, a twice-weekly 1.94 L synthetic stormwater dosing, as well as effluent sampling continued for 4 months.

Standard design features

The following design features were standard within all the biofilters.

A drainage outlet for each biofilter was covered by a 100 mm depth gravel drainage zone and a 150 mm coarse sand transition layer. This was topped with 500 mm depth growth media (with varying designs in between these layers – see section below) to support root development and plant growth as recommended by Payne *et al.* (2015). The inner column walls of the Ø160 mm PVC piping were abraded to prevent preferential surface flow (excluding the geotextile layer (GL) biofilter) and sealed at the base.

The growth media consisted of sandy loam with a tested typical infiltration rate of 145 ± 17 mm/hr. This was amended with 5% compost (solid phase organic matter) as a carbon source and 5% perlite for its natural drainage and water absorption capabilities (Payne *et al.* 2015; Prodanovic *et al.* 2018). This combination of materials was chosen to maintain influent water diffusion through the growth media and alleviate transplantation stress (Le Coustumer *et al.* 2012).

The selection of *P. serratum* as the plant species used in this research was derived by a desktop study, which resulted in a phyto-guide (Jacklin *et al.* 2021b), which relies on existing recommendations and knowledge of removal processes of South African phytoremediators, and previous research into South African plant species for application to stormwater runoff treatment biofilters (Jacklin *et al.* 2021c). Additionally, plant physiological properties such as growth rate, lifespan, tolerance and hardiness as well as morphological traits such as above- and below-ground biomass were considered. This species was deemed appropriate due to the plant's rapid growth rate and biomass production and its vegetative contribution to water quality improvement, which was deemed sufficient over the planned experimental period. For South African GI initiatives where the use of *P. serratum* would be unsuitable, due to climatic or habitat conditions, the use of potential alternatives should be promoted (see Jacklin *et al.* 2021a).

Growth and filter media design variations

The six different plant biofilter designs are illustrated in Figure 1. Design variations were informed by research reported in published literature as indicated. The standard (STD) biofilter represents a typical design for plant biofiltration systems, which rely primarily on plant uptake and media sorption processes for pollutant removal (Bratieres *et al.* 2008). The GL biofilter replaces inner column wall abrasion with nonwoven geotextile fabric for possible additive microbial establishment (see Valentis & Lesavre 1990). In this design, geotextile was inserted along the length of the PVC pipe inner walls, instead of laterally across the biofilter to avoid clogging (see Palmeira *et al.* 2008); it therefore increase the potential load treated. Restricting clogging minimises influent loss to surface runoff (Le Coustumer *et al.* 2012). For the saturated zone (SZ) plant biofilter, an anaerobic SZ was included, which consisted of a 1:1 mixture of vermiculite (0.5–1.4 mm) and attapulgite (0.5–1.4 mm) media at a depth of 200 mm within the experimental column for enhanced nutrient and metal removal (see Wang *et al.* 2018b), while the growth media and vegetation above supplied carbon for denitrification (see Payne *et al.* 2015). Similar to the GL plant biofilter, the plenum aeration (PA) biofilter integrated zeolite (0.8–1.4 mm) based on an experimental concept devised by Smith (2015) for the removal of pollutants from household wastewater. Two Ø40 mm horizontal air plena were constructed between three zeolite media layers below the growth media for enhanced passive aeration, increasing oxygen ingress to support nitrification. The PA+SZ plant biofilter design combined nitrification and denitrification process within a single plant biofilter column by supplying atmospheric -oxygen for nitrification and -carbon dioxide (carbon source) for denitrification. Finally, the upflow+PA+SZ plant biofilter design was included for investigation. The influent drained into a Ø30 mm orifice and was driven vertically through the upflow filter by the difference in water head at the rate of experimental

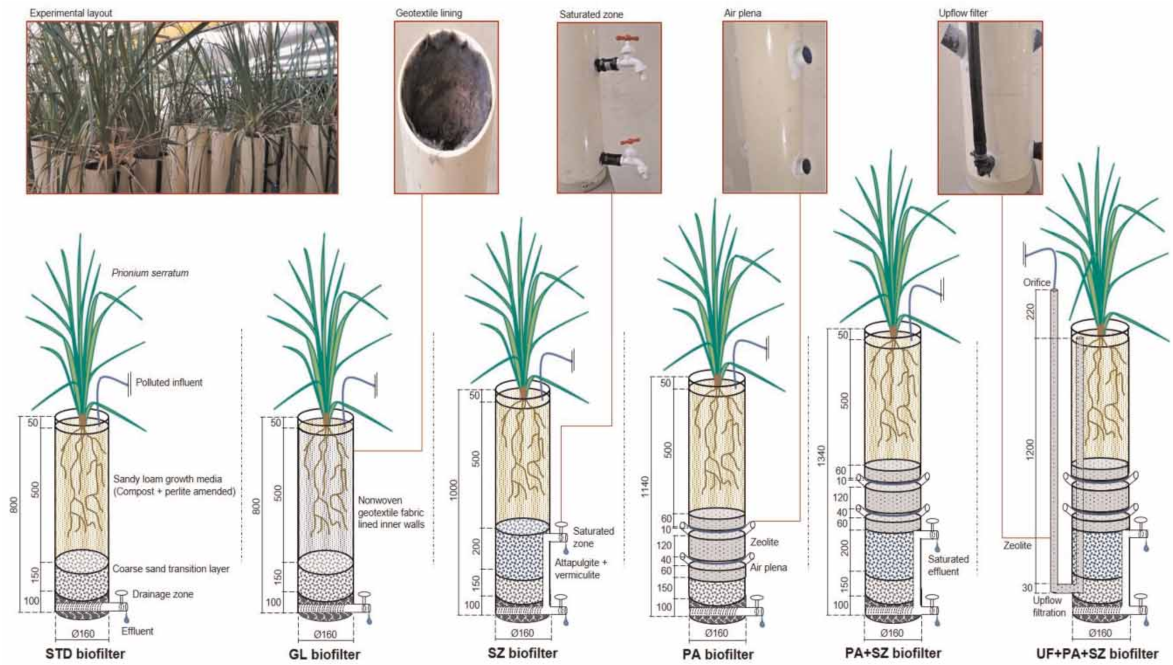


Figure 1 | Schematic of the six plant biofilter designs, with photographic panels illustrating significant aspects of the experiment. *Note*: STD=Standard; GL=Geotextile lined; SZ=Saturated zone; PA=Plenum aeration; PA+SZ=Plenum aeration+saturated zone; UF+PA+SZ=Upflow filtration+plenum aeration+saturated zone.

influent between the influent orifice and the surface of the growth media (Cucarella & Renman 2009). Integrating upflow filtration as a pretreatment may reduce the maintenance frequency required to combat top layer clogging and media breakthrough (the point where sorption sites on the media are exhausted and pollutants leach), decreasing the cost of rehabilitation and ensuring long-term functionality (Blecken *et al.* 2017).

Synthetic stormwater dosing and effluent testing

Synthetic stormwater (Table 2) was prepared to reflect published low, typically observed and high urban stormwater nutrient and metal concentrations, as well as similar stormwater biofilter investigations (see Taylor *et al.* 2005; Göbel *et al.* 2007). Dissolved metals were selected as the majority of Cd, Cu, Pb and Zn species are typically found in this form and are most mobile within biofilters (Sun & Davis 2007).

Irrigation was done by submersible pumps in separate dosing tanks for each pollution treatment via an automated system equipped with pump agitators to ensure uniform dispersion. Influent concentrations were monitored throughout and synthetic stormwater was replaced with a freshly mixed batch at 10-day intervals.

Five effluent sampling rounds were initiated after a month of synthetic stormwater dosing at 20-day intervals. This 30-day dosing period prior to effluent collection ensured discharge of non-polluted tap water (used for establishing *P. serratum*) from the columns. This period further provided time for the polluted influent to saturate the anaerobic zones in the SZ, PA+SZ and UF+PA+SZ biofilter columns, as well as filling the upflow filtration chamber. Due to potential water loss to evapotranspiration as well as drying of the media between irrigation events, percentage pollutant load removal (L_{rem}) was used and considered the influent ($C_{inf}V_{inf}$) and effluent ($C_{eff}V_{eff}$) loads of each design for every sampling round (see Equation (1)). The overall percentage pollutant load removed for each design was then estimated as the mean of the 5 sampling rounds.

$$L_{rem} = \frac{C_{inf}\,V_{inf} - C_{eff}\,V_{eff}}{C_{inf}\,V_{inf}} \times \frac{100}{1} \qquad (1)$$

where L_{rem}=pollutant load removed (%), C_{inf}=influent concentration (mg/L), V_{inf}=total influent volume for sampling round (L), C_{eff}=effluent concentration (mg/L), V_{eff}=effluent volume of sampling round (L).

Various water quality parameters were analysed in the Stellenbosch University Water Quality Laboratory for each plant biofilter design and influent pollution strength. These included NH_3-N, NO_3^--N, PO_4^{3-}-P and total

Table 2 | Influent synthetic stormwater concentrations and overall performance of the engineered plant biofilters

Pollution strength	Prepared influent synthetic stormwater concentrations – C_{inf} (mg/L)						
	NH_3-N	NO_3^--N	PO_4^{3-}-P	Cd	Cu	Pb	Zn
Low	0.20	0.50	0.22	0.002	0.02	0.08	0.15
Typically observed	0.40	0.95	0.35	0.0045	0.045	0.15	0.30
High	2.5	13.5	1.5	0.032	6.8	2.8	35
Source chemical	NH_4Cl	KNO_3	K_2HPO_4	$CdCl_2$	$CuSO_4$	$PbCl_2$	$ZnCl_2$

	Measured mean effluent concentrations and mean loads removed													
	NH_3-N		NO_3^--N		PO_4^{3-}-P		Cd		Cu		Pb		Zn	
Biofilter design type	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)	$\bar{x}C_{eff}$ (mg/L)	$\bar{x}L_{rem}$ (%)
Low pollution strength														
STD	0.065	89	1.24 +	23	0.60 +	15	0.00019	96	0.026 +	59	0.0016	99	0.05	89
GL	0.18	50	1.33 +	−44	0.23 +	43	0.00014	96	0.015	59	0.00058	99	0.027	90
SZ	0.18	89	1.11 +	74	1.10 +	42	0.00051	97	0.07 +	59	0.0052	99	0.11	91
PA	0.065	96	2.26 +	47	1.10 +	41	0.00023	98	0.05 +	70	0.0026	99	0.04	96
PA+SZ	0.069	96	1.03 +	76	0.64 +	66	0.00021	98	0.04 +	76	0.0028	99	0.04	96
UF+PA+SZ	0.032	98	0.63 +	85	0.35 +	81	0.00013	99	0.023 +	86	0.00055	99	0.02	98
Typically observed pollution strength														
STD	0.037	97	1.60 +	47	0.96 +	15	0.00023	98	0.016	88	0.00094	99	0.052	94
GL	0.30	59	2.49 +	−42	0.23 +	63	0.00005	99	0.0052	93	0.00052	99	0.019	96
SZ	0.11	96	1.74 +	78	1.33 +	56	0.00045	98	0.092 +	76	0.0054	99	0.098	96
PA	0.062	98	3.02 +	63	1.14 +	62	0.00024	99	0.067 +	82	0.0035	99	0.032	98
PA+SZ	0.055	98	1.15 +	85	0.58 +	80	0.00032	99	0.037	90	0.0014	99	0.05	98
UF+PA+SZ	0.034	99	0.79	90	0.42 +	86	0.00024	99	0.022	94	0.00099	99	0.038	98
High pollution strength														
STD	0.12	98	9.94	77	2.46 +	49	0.00022	99	0.023	99	0.0025	99	0.033	99
GL	1.11	75	14.36 +	42	1.02	63	0.0001	99	0.0098	99	0.0013	99	0.018	99
SZ	0.30	98	7.85	93	1.44	88	0.0005	99	0.12	99	0.009	99	0.077	99
PA	0.096	99	12.39	89	1.79 +	86	0.00026	99	0.097	99	0.005	99	0.039	99
PA+SZ	0.06	99	7.52	93	1.54 +	88	0.00024	99	0.045	99	0.002	99	0.059	99
UF+PA+SZ	0.04	99	5.50	95	0.74	94	0.00025	99	0.041	99	0.002	99	0.036	99

Note effluent concentration>influent concentration assigned a +.

C_{inf}, influent concentration; $\bar{x}C_{eff}$, mean effluent concentration; $\bar{x}L_{rem}$, mean percent load removed.

suspended solids (TSS), all measured with the HACH DR3900 spectrophotometer, by implementing TNTplus™ methods 10205, 10206, 10209 and photometric method 8006 respectively. Temperature, pH, electrical conductivity (EC), dissolved oxygen (DO) and total dissolved solids (TDS) were measured with the HACH HQ440d benchtop multi-parameter meter. Dissolved Cd, Cu, Pb and Zn were analysed at the Stellenbosch University Central Analytical Facility: ICP-MS division on an Agilent 8800 QQQ ICP-MS instrument, with polyatomic interferences removed by a 4th generation Octopole Reaction System (Agilent 2015). Prior to nutrient and metal analyses, samples were filtered with 0.45 μm syringe filters.

Data analysis

Statistical analyses were conducted using R statistical software and statistically significant differences were accepted at an unadjusted p-value$\leq$0.05 to maintain the power of the test (Feise 2002). Since data was typically non-normally distributed, as established by the Lilliefors test, the Kruskal-Wallis H-test was used to ascertain the significance of difference in influent concentrations, as well as between the effluent concentrations and percent removals of the six engineered designs for each pollution strength and parameter. With significance, the Wilcoxon Rank Sum paired test was used to compare pollutant influent and effluent concentrations, in addition to comparing effluent concentrations and percentage removal between designs to assess plant biofilter treatment performance. With this information, the Fisher Least Significant Difference (LSD) test was used to detect the differences in percentage removal between designs for each pollutant parameter.

RESULTS AND DISCUSSION

Engineered plant biofilter water quality results, as well as the low, typically observed and high influent stormwater concentrations are listed in Table 2. Prior to determining percentage removals, results of the Kruskal-Wallis H-test confirmed that influent concentrations, monitored throughout the study, were not statistically significantly different (p>0.05) for each pollution strength between sampling rounds, allowing comparisons of effluent quality among the biofilter designs (Winston *et al.* 2017).

Effluent concentration quality

As shown in Table 2 the mean effluent concentrations and percentage load removals by the biofilter designs varied across pollutant type and were influenced by influent strengths. The plant biofilters had lower mean effluent concentrations compared to those of the influent in 92 (73%) of the 126 treatments, with substantially decreased NH_3-N and dissolved Cd, Pb and Zn, which were consistently lower in all engineered designs for all pollution strengths. Furthermore, on four and six occasions for Cd and Pb respectively, the effluent concentrations were below laboratory detection limits (i.e. Cd<0.0000045 mg/L and Pb<0.000047 mg/L).

In contrast, removal performance was more variable for PO_4^{3-}-P and NO_3^--N, as well as for dissolved Cu. Here, mean effluent PO_4^{3-}-P concentrations were higher in all designs exposed to low and typically observed pollution dosage, as well as from the STD, PA and PA+SZ biofilters exposed to high pollution dosage. For NO_3^--N, mean effluent concentrations were higher than the influent in all designs exposed to low and typically observed pollution, except for the UF+PA+SZ biofilter, which was lower than the corresponding typically observed influent concentration. In addition, when exposed to high NO_3^--N pollution, only the GL biofilter was observed to generate leachate. For dissolved Cu under low strength pollution only, the GL biofilter had a reduced mean effluent concentration, while under typically observed pollution higher mean effluent concentrations were produced by the SZ and PA columns with the remaining designs showing effluent concentrations that were lower than the influent concentrations.

Statistical analysis using the Wilcoxon Rank Sum test for comparing influent and effluent concentrations for each pollutant parameter over the three pollution strengths showed significant differences (p$\leq$0.05) in almost all the treatments. The plant biofilters where statistically significant differences were not found between influent and effluent concentrations were: GL in low PO_4^{3-}-P (p=0.63) and high NO_3^--N (p=0.45) pollution; SZ and PA in typically observed Cu (p=0.17 and p=0.44) and high PO_4^{3-}-P (p=0.74 and p=0.26) pollution; PA in high NO_3^--N (p=0.82) pollution; PA+SZ in high PO_4^{3-}-P (p=0.85) pollution; and UF+PA+SZ in low dissolved Cu (p=0.54) and typically observed PO_4^{3-}-P (p=0.17) pollution. In evaluating effluent quality, the results showed reduced pollutant concentrations in the majority of the treatments, suggesting biofilter efficiency, with the exception of NO_3^--N, PO_4^{3-}-P and dissolved Cu. This notion which considers concentration only, however, is inadequate in assessing water-quality treatment, as it does not account for the change in volume (Li & Davis 2009).

Pollutant load removal performance

The mean nutrient and metal load removal percentages over all sampling rounds are reported in Table 2 with further illustrations on the performances between designs provided in the supplementary material (Figure S1). As mentioned, determining plant biofilter performance by solely evaluating effluent concentrations can be misleading, since a biofilter treating high influent concentrations will always appear more efficient than one treating low influents, even if both are achieving the same effluent quality (Strecker *et al.* 2002). This dependence is of importance in this study, as the function also applies to pollutant loads (Lampe *et al.* 2005). From the reported values, percentage load removals were found to be influenced by influent strength, as opposed to effluent concentrations, further demonstrated by the Wilcoxon Rank Sum test comparing percent removal between the low, typically observed and high influent strengths for each pollutant. For all dissolved metals, removal between the three influent strengths were significantly different ($p \leq 0.05$). Similarly, percentage removal of high influent nutrients was significantly different to the other pollution strengths. Between low and typically observed nutrient pollution, however, this was not the case, recording no significant differences ($p > 0.05$) in percent removal.

Substantial percentage removal in dissolved metals was recorded for Cd (>96%), Pb (>99%) and Zn (>89%) across the dosing strengths, while the generally more variable removal of nutrients NO_3^--N and PO_4^{3-}-P, as well as dissolved Cu ranged from −44 to 99%, 15 to 94% and 59 to 99% respectively. Notably, the mean percent removal of NH_3-N was high (>89%) across all biofilter designs, except GL, which ranged from 49 to 75%. Furthermore, the common issue of NO_3^--N leaching, the main obstacle to effective nutrient removal in biofilters (Zinger *et al.* 2013), was found in only the GL biofilter. Overall the most efficient engineered designs (based on mean percentage loads removed across the pollution strengths) were the UF+PA+SZ, PA+SZ and SZ biofilters, removing on average 96, 93 and 88% of urban pollutants respectively.

Nutrients

Effective NH_3-N load removal was found in almost all engineered designs, across the pollution strengths, suggesting an enhancement of the nitrification process. The best-performing design was the UF+PA+SZ biofilter, reporting efficient (>98%) mean NH_3-N removal. In contrast, the GL biofilter was least efficient, ranging from 49 to 75%, which may have been due to its inability to retain water, decreasing the hydraulic retention time, thereby decreasing water-quality improvement (Hunt *et al.* 2012). In addition to some surface volatilisation, removal primarily occurs in the biofilter rhizosphere under aerobic conditions via biological processes that convert NH_3-N to NO_3^--N for plant uptake (Payne *et al.* 2014). In the presence of organic matter, applicable to this study due to growth media amendment with compost, NH_3-N can be rapidly nitrified (Hunt *et al.* 2015). From the results, although efficiency was recorded in all biofilter columns, NH_3-N removal was consistently enhanced in designs incorporating air-plena, suggesting successful microbial nitrification in aerobic conditions.

Percent NO_3^--N removal varied notably between designs and pollution strengths, however, in almost all designs the influent load was reduced, with the exception of the worst-performing GL biofilter, which was observed to leach NO_3^--N for low (−44%) and typically observed (−42%) pollution dosages. Similar to the case of NH_3-N, the best performing design was the UF+PA+SZ biofilter, reporting efficient (>85%) mean NO_3^--N removal. Removal processes for NO_3^--N rely on denitrification and biotic assimilation within the biofilter, without which NO_3^--N is released to the water column and leaching is observed (Henderson *et al.* 2007).

In the presence of plants and a carbon source, NO_3^--N removal is enhanced via the promotion of microbial nitrification and denitrification, as well as direct uptake, emphasizing the importance of vegetation and compost (in the appropriate solid phase) in plant biofilters (Muerdter *et al.* 2018). It is believed that biotic assimilation, which include NO_3^--N uptake by plants, bacteria, fungi and other microbes, is the major removal pathway in plant biofilters (Payne *et al.* 2014), with denitrification accounting for <15% of treatment (Fowdar *et al.* 2018). This assumption is, however, not applicable to all biofilters, when influent NO_3^--N nears 10 mg/L as revealed by Barron *et al.* (2019), the influence of denitrification has shown to increase in proportion to a decrease in plant assimilation. In the current study, with all columns similarly incorporating *P. serratum*, variations in NO_3^--N load removal between biofilter designs suggest a shift in reliance from plant assimilation to denitrification. In addition, the notion that this shift from plant assimilation to denitrification is wholly reliant on high influent concentration is uncertain from the findings of our study, as varying percentage NO_3^--N removal between designs under low, typically observed and high concentrations were reported. Therefore, engineering biofilter design (SZ, PA, UF or a combination of these features), other than plant species selection, may significantly influence treatment performance in varying NO_3^--N environments. From the results in this study, designs incorporating a SZ increased NO_3^--N removal, inferring enhanced denitrification and plant uptake in anaerobic conditions.

For mean percent PO_4^{3-}-P removal, the influent load was reduced by all biofilter designs. Similar to the other nutrient pollutants, the best-performing design was the UF+PA+SZ biofilter, efficiently (>81%) removing the greatest pollutant loads. The worst-performing biofilter design was STD, recording mean PO_4^{3-}-P removal ranging from 15 to 49%. The main removal pathway for PO_4^{3-}-P within plant biofilters is unvegetated media filtration and sorption (Fowdar *et al.* 2017), with plants contributing through direct uptake and storage between roots, as well as the provision of oxygen for additive media to root sorption (Barron *et al.* 2019). Due to the presence of the high above- and below-ground biomass *P. serratum* species in the plant biofilters, enhanced PO_4^{3-}-P removal can be explained by its extensive root system and presence of root hairs (Bratieres *et al.* 2008). In addition, amending biofilter growth media with compost has been found to increase PO_4^{3-}-P in effluent runoff, decreasing percent removal (Lenth & Dugapolski 2011). Somewhat surprisingly, in this study the rapidly infiltrating GL biofilter outperformed SZ, PA and PA+SZ, suggesting possible filtration and sorption to the geotextile fabric.

Metals

As shown in Table 2 and further illustrated in Figure S1, dissolved metals were efficiently removed. The addition of carbon (from compost) and perlite to the biofilter growth media, with a higher capacity of removal than soil only, absorb heavy metals like Cd, Cu, Pb and Zn (Bratieres *et al.* 2008). Similar to what was found in the case of nutrient load removal, the best performing biofilter design for dissolved metal removal was the UF+PA+SZ filter. The mean load removal percentages achieved by the various engineered designs were high across the different influent pollution strengths for Cd (>96%), Pb (>99%) and Zn (>89%). In addition, for high influent dissolved Cu, percent removal was also high (>99%). These negligible removal variations between the biofilter designs for dissolved Cd, Pb and Zn were of little practical importance given the consistently high removal, which supports previous findings (Barron *et al.* 2019). For Cu, however, removal was less efficient and more variable between designs, with the SZ biofilter recording the lowest percent removal, ranging from 59 to 99%. As the influent strength decreased, variation in percent Cu removal increased, ranging from 59 to 86% and 76 to 94% for low and typically observed pollution dosing levels respectively, which is consistent with similar published investigations (see Davis *et al.* 2001; Lenth & Dugapolski 2011). Variation may be due to the formation of Cu-organic matter complexes within the biofilter growth media as Cu has a strong affinity to organic matter (Ponizovsky *et al.* 2006). This process is significantly influenced by the form of organic matter, while solid organic matter adsorbtion of Cu is a main removal pathway in biofilters (Temminghoff *et al.* 1997). Dissolved organic matter tends to mobilise Cu and in some instances can result in leaching lasting several years (Mullane *et al.* 2015). In this study, the growth media amended with compost consisted of solid phase organic matter. Therefore, it is believed that Cu immobilisation likely occurred. The SZ biofilter consistently had the least efficient removal values for Cu and Pb, and in some instances Cd and Zn. These findings suggest a reduction in redox potential, an important mechanism for dissolved metal removal in stormwater biofiltration, within the anaerobic SZ, which corresponds with previous studies (see Dietz & Clausen 2006).

High metal extraction may be provided by metal hyperaccumulating plants; however, their use in biofiltration is relatively untested (Kratky *et al.* 2017). While *P. serratum* is not identified as a hyperaccumulator species, it has been found to offer water purification services (Rebelo 2018) and enhance treatment efficiency in laboratory investigations (Jacklin *et al.* 2020). The plant benefits from a large above-ground biomass and extensive root system, appropriate traits for metal removal in biofilters (Sun & Davis 2007). Micronutrients Cu and Zn, as well as non-essential toxic Cd and Pb uptake by plants, though relatively small compared with unvegetated mechanisms such as filtration and adsorption in the upper layer of the growth media (Blecken *et al.* 2009), provide a permanent pollutant removal pathway via harvesting as metal accumulation occurs over time (Kumar *et al.* 2017).

Comparison of the 6 different designs

As mentioned previously, water-quality treatment cannot be examined without considering hydrologic improvements, as reducing effluent volume from biofilters is an important mechanism in reducing total pollutant loads. In many successful GI initiatives, the primary reason effluent load is reduced from the influent load is as a result of modifying hydrology and water balance (Hunt *et al.* 2012) and through decreasing immediate discharge by increasing evapotranspiration (Davis *et al.* 2009). Although the use of percentage load removal (which considers variation in through flow volumes) is a better reflection than effluent concentration of biofilter efficiency, it is not a representation of the extent of pollutants entering and leaving a system.

A significant amount of influent volume was retained by the engineered biofilters and did not drain from the system, comparable with a study by Hunt *et al.* (2006), reporting more than 50% reduction in effluent volumes. In our study the influent and effluent volume ratios over the sampling periods (20 days), ranged from 0.54 to 0.12 for the six engineered designs, almost identical to the findings of Li & Davis (2008), reporting ratios ranging from 0.60 to less than 0.10 in field biofiltration systems. We postulate that a combination of the plenum-aerated zones, the dry periods between dosing events, as well as the water uptake potential of *Prionium serratum* (transpiration and water within plant tissue) and its extensive moisture-retaining root system reduced discharge.

The variability in effluent concentrations and percent removals between experimental biofilters alludes to differences in removal pathways within the designs. For the optimisation of plant biofiltration, design must seek to combine appropriate removal mechanisms for overall urban stormwater pollutant treatment. Therefore, we assessed the performance of each biofilter design with the aim of ascertaining possible pollutant removal pathways. Differences in influent loads and ranges of removed nutrients and metals loads between designs are shown in Figures 2 and 3 respectively. The differences in removal efficiency between the biofilter designs for each pollutant parameter was analysed by use of the Wilcoxon Rank Sum test, with statistically significant differences represented by the outcomes of the Fisher's LSD test.

The STD design

The STD biofilter design mean influent to effluent volume ratio of 0.31 relied primarily on plant uptake and media sorption for pollutant removal. The growth media, amended with perlite and compost (solid phase organic matter) as a carbon source for denitrification was replicated for all designs. The STD biofilter, supporting aerobic nitrification, performed well for NH_3-N load removal compared with the other designs (Figure 2).

For NO_3^--N, the STD biofilter was not as efficient, resulting in significantly lower loads removed compared with all other designs, except for the GL biofilter, as a result of the lack in anaerobic sites required for successful denitrification. For the removal of PO_4^{3-}-P, the STD biofilter was significantly less efficient than all other designs, a common finding in new and establishing standard design biofilters (see Blecken *et al.* 2009). Removal of PO_4^{3-}-P by the STD biofilter, which recorded leaching in lower-strength pollution (low and typically observed pollution), improved with time. This finding may be as a result of the influence of plant growth and solid state organic matter (see Lenth & Dugapolski 2011; Fowdar *et al.* 2017). While biofilters can benefit from solid state organic matter for removing certain pollutants, like dissolved metals (Bratieres *et al.* 2008), the breakdown reaction of organic matter can result in the release of PO_4^{3-}-P (Hsieh *et al.* 2007). As expected, all dissolved metals were efficiently removed, as rapid metal removal occurs via sorption and/or filtration in the upper layer of the growth media. This was similarly found in other published literature (see Davis *et al.* 2001). The removal of dissolved metal loads were comparable with other designs, as a result of the similar vegetated growth media layer between designs.

The GL design

The GL biofilter design had a mean influent to effluent volume ratio of 0.54 and introduced nonwoven geotextile fabric to support increased microbial activity and infiltration (Valentis & Lesavre 1990). During dosing events, some effluent discharge was detected within 5 minutes of initiating irrigation under saturated media conditions, which decreased to mere seconds in high volume irrigation under dry media conditions. This suggests rapid preferential flow through the geotextile fabric instead of the growth media, decreasing hydraulic retention time. Therefore, it is recommended that this design not be employed.

Separation of pollutants from the influent flow requires time for various removal mechanisms to be effective and any design that decreases hydraulic retention time is expected to decrease water-quality treatment (Hunt *et al.* 2012). This may explain the significantly less NH_3-N and NO_3^--N loads removed compared with the other designs, as insufficient retention time hampered the nitrification and denitrification processes. In contrast, flow down the inner walls of the columns through the fabric minimised interactions within the growth media, thereby preventing the breakdown reaction of compost and restricting PO_4^{3-}-P mobilisation and ultimate deposition. Efficient dissolved metal load reduction was observed, with the GL biofilter the only design recording below laboratory detectable limits in some cases (two measurements for Cd and four for Pb in typically observed

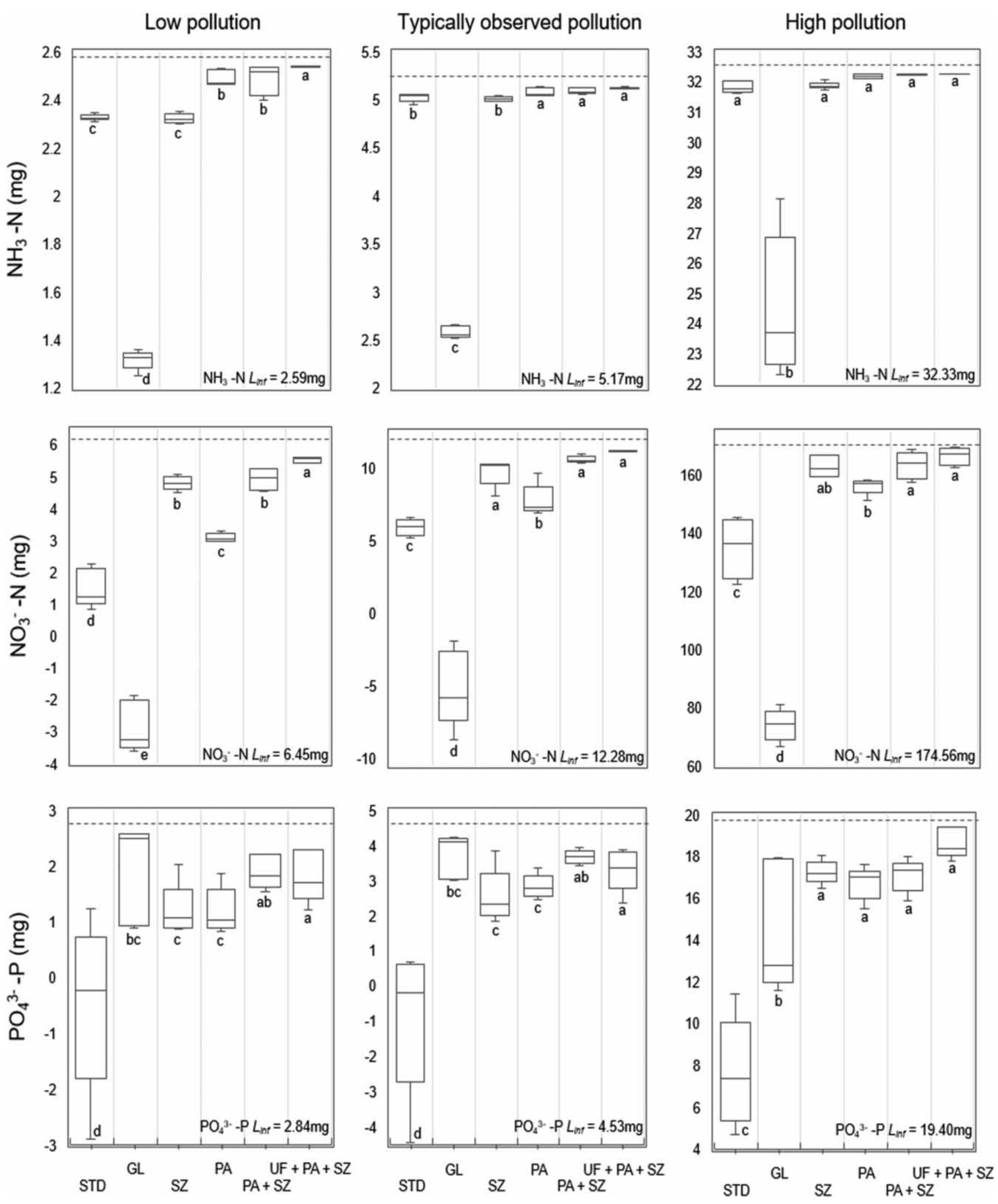


Figure 2 | Boxplots of nutrient loads removed by the different biofilter designs over the sampling rounds. Note the different scales of vertical axes; L_{inf}=influent pollutant load, illustrated by the dotted lines. Designs with the same letter did not show statistically significant differences (Fisher's LSD test).

and high pollution dosage levels). This may have been due to microbial community establishment on the fabric or direct sorption enhancing metals removal, which warrants further investigation in future research.

The SZ design

The SZ biofilter design had a mean influent to effluent volume ratio of 0.21 with an anaerobic SZ consisting of inexpensive vermiculite and attapulgite media with increased surface area, adsorption and cation exchange capacities. These media have been found to offer great potential as alternatives to conventional sand for the removal of stormwater pollutants in the saturated zone (Tichapondwa & Van Biljon 2019). Carbon came from the growth media and root exudates, which likely supported denitrification (Payne *et al.* 2015).

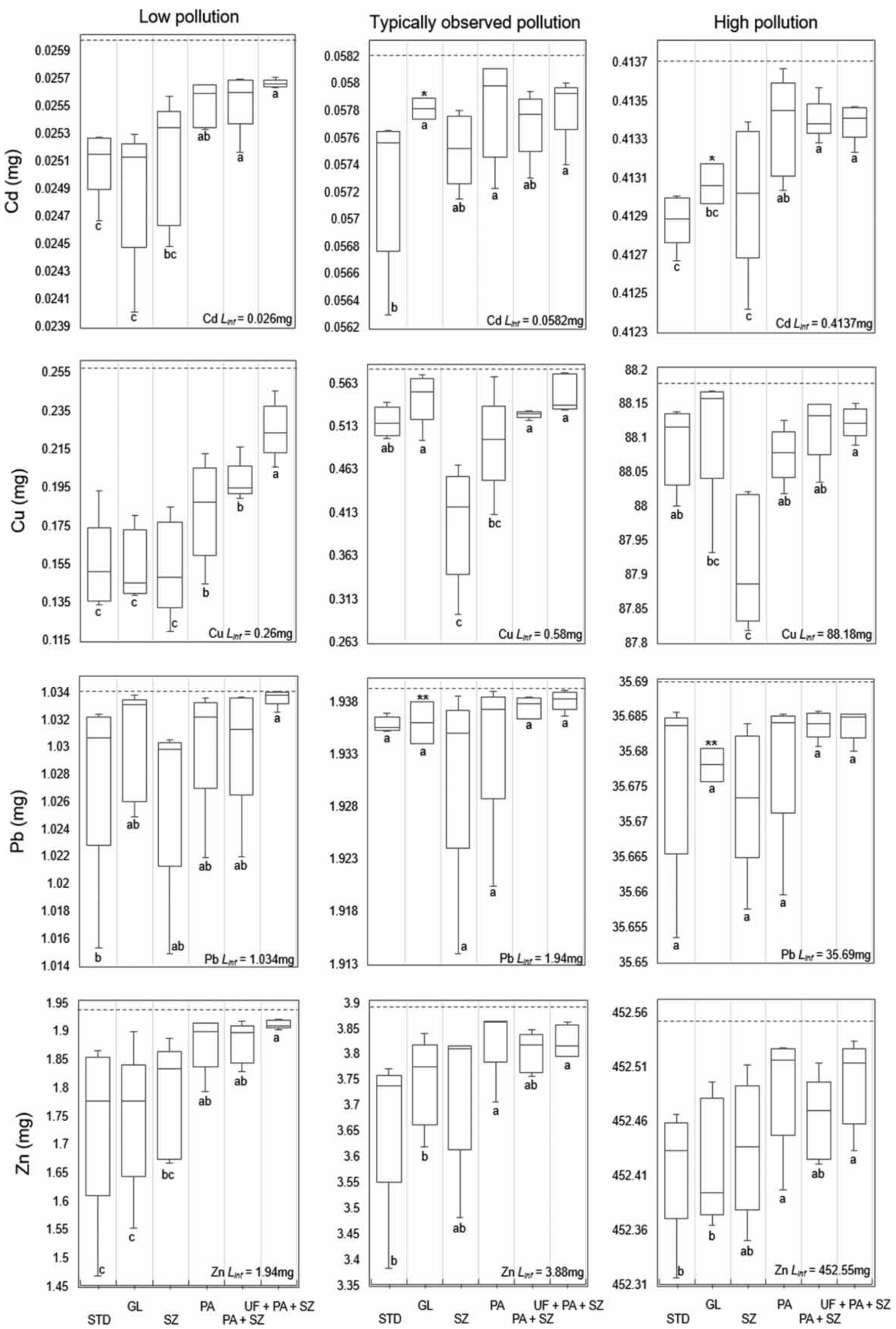


Figure 3 | Boxplots of metal loads removed by the different biofilter designs over the sampling rounds. Note the different scales of vertical axes; L_{inf}=influent pollutant load, illustrated by the dotted lines; *=below detectable limit. Designs with the same letter did not show statistically significant differences (Fisher's LSD test).

In the presence of a saturated zone the removal efficiency of NO_3^--N increased significantly compared with other designs, while the differences in removal of NH_3-N and PO_4^{3-}-P were not significant (except for the GL and STD biofilters respectively, as discussed earlier). The slightly lower (but not statistically significantly different) PO_4^{3-}-P removal recorded by the SZ biofilter when compared with designs aimed at increasing aerobic conditions is in contrast with the findings by Palmer *et al.* (2013), who reported significant effluent PO_4^{3-}-P increase under anaerobic conditions. For the removal of dissolved metals, although the differences for Cd, Pb and Zn between biofilter designs were not statistically significant and of no practical importance, significantly lower Cu loads were removed compared with the other designs. This is similar to previous findings reporting a reduction in redox potential under anaerobic conditions, with the filter acting as a source of pollutants rather than a sink (Dietz & Clausen 2006). A reduction in redox potential hinders metal speciation, which decreases metal adsorption to media, which is the primary metal removal pathway in biofilters (Muerdter *et al.* 2018).

The PA design

The PA biofilter design had a mean influent to effluent volume ratio of 0.12 and contained two horizontal air plena between zeolite media layers to enhance passive aeration for the nitrification process by increasing the surface area available for oxygen ingress (Smith 2015). In the case of high influent pollution dosing levels the standard biofilter growth media becomes saturated and may clog, requiring a correspondingly high oxygen demand by the media for successful nitrification (Blecken *et al.* 2009). Offering higher cation exchange capacities, sorption and acid catalysis properties than soil (Mondal *et al.* 2021), zeolite media decreases clogging and increases nutrient and metal removal from polluted water while supporting microbial growth and oxygenating biological regeneration (Markou *et al.* 2014).

In assessing load removal between designs, enhanced NH_3-N removal by the PA biofilter suggests successful growth media oxygenation. A lack in anaerobic conditions for denitrification may be the cause of significantly lower loads of NO_3^--N removed when compared with designs containing a saturated zone. In contrast, no statistically significant differences were recorded in assessing PO_4^{3-}-P load removal between this and other designs, possibly as a result of filtering and sorption to the zeolite media. For the removal of dissolved metals loads, though not significantly different, removal efficiency may have benefitted from zeolite's high cation exchange capacity, favouring adsorption.

The PA±SZ design

The PA+SZ biofilter design had a mean influent to effluent volume ratio of 0.12 and combined pollutant removal pathways for support of enhanced nitrification and denitrification.

In comparing the performance of the PA+SZ biofilter with the other designs (excluding UF+PA+SZ) for nitrogen removal, statistically significantly reduced loads of both NH_3-N and NO_3^--N inferred successful microbial nitrification (in the growth media) and denitrification (in the saturated zone) as well as direct plant uptake. Similar to the other nutrients, reduced yet not statistically significantly different PO_4^{3-}-P loads were recorded between this and other designs not benefitting from a combination of removal pathways. This type of efficiency is predominantly attributed with extending biofilter depth (Davis *et al.* 2006), and/or the addition of engineered filter media increasing the possibility of sorption. Similar to other biofilter designs, the majority of dissolved metals were removed and was presumed to occur in the upper layer of the growth media.

The UF±PA±saturated zone SZ design

The UF+PA+SZ biofilter design (UF+PA+SZ) had a mean influent to effluent volume ratio of 0.12 and combined efficient removal pathways for improved urban stormwater pollutant removal. The zeolite upflow filter propelled polluted influent against gravity through engineered filter media (Sanz *et al.* 1996), providing additional removal mechanisms for particles, nutrients and dissolved metals (Winston *et al.* 2017).

In comparing loads removed between designs the UF+PA+SZ biofilter consistently outperformed all other designs across each pollutant parameter, though not always statistically significantly different. The upflow filter relies primarily on filtration and adsorption to the zeolite media, which may be prone to clogging and exhaustion as a mechanism for removal, possibly resulting in treatment failure long term. In this event refinements to design for optimised urban stormwater treatment may be meaningless and will require periodic maintenance to ensure long-term functionality (Blecken *et al.* 2017). Therefore, positioning the upflow filter as a separate entity on the outside rather than inside of the biofilter will facilitate media replacement and/or regeneration, a possible improvement to design. This also promotes the possible use of interchangeable cartridges in the event of clogging or exhaustion, simplifying the media maintenance process and increasing design feasibility (Winston *et al.* 2017).

CONCLUSIONS

This study demonstrated the pollutant removal performances of six different plant biofilter designs exposed to low, typically observed and high influent nutrients and metals dosing levels for urban stormwater runoff quality improvement. In addition to the endemic South African species *P. serratum*, the designs incorporated various engineered media using nationally available materials (perlite, vermiculite, zeolite and attapulgite) as well as biofilter column modifications (geotextile lining, saturated zone, plenum aeration and upflow filtration) for performance testing. The most efficient design combined standard plant biofiltration techniques with UF, PF and a SZ for anaerobic microbial activity support, removing on average 96% of urban stormwater nutrients and metals loads. In all plant biofilter designs significant differences in loads of NH_3-N and dissolved Cd, Pb and Zn were removed from the synthetic stormwater, whereas removal of NO_3^--N, PO_4^{3-}-P and dissolved Cu was more variable.

Although appropriate removal pathways were suggested for their affinity to specific urban stormwater pollutants, some unsatisfactory findings were observed, which may jeopardize the performance of intricately designed biofilters. For example, the rapid preferential flow of the GL biofilter resulting in poor hydraulic retention time, decreasing water-quality treatment, thus we recommend that this design not be considered. In addition, retrofitting of a saturated layer into plant biofilters for enhanced denitrification resulted in unsatisfactory metal treatment. Coupled with conflicting reports regarding the effect of redox potential on metal solubility (Rieuwerts *et al.* 1998), retrofitting a saturated layer must consider the target contaminant of concern since no clear benefit was observed for the treatment of all pollutants.

The findings of this study confirm the notion that plant biofilters as applied to treatment of stormwater to improve quality treatment must include consideration of hydrologic improvements, as modifications in the water balance may be a contributing factor for a reduced pollutant load. Recognising the benefits of the various removal pathways to plant biofilters, it is possible to design a system for site- and pollutant-specific water treatment by addressing appropriate hydrologic and water-quality targets respectively. In urban stormwater biofilters the plant uptake, media sorption, microbial activity and infiltration benefits offered to water-quality treatment by these types of systems are valid and practically applicable.

This study focused on nutrient and metal pollutants under consistent dosages, and future work should investigate other pollutants under variable and intermittent dosages. In addition, the influence of conversion from tap water to synthetic stormwater on plant and associated microbial activity must be assessed, with future work advised to measure the physiological state (redox potential) within the biofilters at short intervals (i.e. twice-weekly) following conversion. Due to the degradatory influence of total suspended solids, as a result of conventional stormwater management techniques, it is imperative that the biofilters are subjected to influent suspended solids over an extended period of time to determine the conditions under which failure occurs, warranting future research. Furthermore, future work could investigate the feasibility of interchangeable upflow filtration cartridges in the event of media clogging or exhaustion, simplifying the maintenance process. Finally, risks associated with polluted-laden media disposal during replacing or replenishing must be assessed due to the disposed media's potential to act as a pollution source.

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DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

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