



Comparison of filter media materials for heavy metal removal from urban stormwater runoff using biofiltration systems



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ABSTRACT

The filter media in biofiltration systems play an important role in removing potentially harmful pollutants from urban stormwater runoff. This study compares the heavy metal removal potential (Cu, Zn, Cd, Pb) of five materials (potting soil, compost, coconut coir, sludge and a commercial mix) using laboratory columns. Total/dissolved organic carbon (TOC/DOC) was also analysed because some of the test materials had high carbon content which affects heavy metal uptake/release. Potting soil and the commercial mix offered the best metal uptake when dosed with low (Cu: 44.78 µg/L, Zn: 436.4 µg/L, Cd: 1.82 µg/L, Pb: 51.32 µg/L) and high concentrations of heavy metals (Cu: 241 µg/L, Zn: 1127 µg/L, Cd: 4.57 µg/L, Pb: 90.25 µg/L). Compost and sludge also had high removal efficiencies (>90%). Heavy metal leaching from these materials was negligible. A one-month dry period between dosing experiments did not affect metal removal efficiencies. TOC concentrations from all materials increased after the dry period. Heavy metal removal was not affected by filter media depth (600 mm vs. 300 mm). Heavy metals tended to accumulate at the upper 5 cm of the filter media although potting soil showed bottom-enriched concentrations. We recommend using potting soil as the principal media mixed with compost or sludge since these materials perform well and are readily available. The use of renewable materials commonly found in Singapore supports a sustainable approach to urban water management.

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1. Introduction

Urban stormwater runoff consists of a broad range of pollutants that have a significant negative impact on receiving waters (Laurenson et al., 2013). Increasingly stringent regulations on stormwater discharge led to the growth of stormwater best management practices (BMPs) in many developed countries. BMPs are also known as Low Impact Development (LID), Water Sensitive Urban Design (WSUD), Sustainable Urban Drainage Systems (SUDS), Innovative Stormwater Management and ABC Waters design in different parts of the world. These systems are built to 1) reduce hydrologic and water quality disturbance to urban and downstream waterways; 2) make use of stormwater as an alternate water resource (Roy-Poirier et al., 2010; Barbosa et al., 2012).

Biofiltration systems are increasingly popular. They adopt natural processes such as filtration, adsorption, vegetation uptake and

biotransformation to slow down stormwater runoff rates, reduce the runoff volumes and retain pollutants before their discharge into receiving waters. The general design includes a topsoil filter layer that supports vegetation and promotes mechanical and chemical processes of pollutant removal. Hence, its composition plays an important role in pollutant removal. A gravel layer at the bottom improves drainage (Davis et al., 2001). A transition layer with intermediate grain sizes is sometimes placed in-between to limit the migration of filter layer materials to the gravel layer.

The filter media typically includes a soil-based material (sandy loam being the usual guideline material), organic matter, or other materials (e.g. vermiculite, perlite) that are added in various proportions to provide sufficient hydraulic conductivity for water drainage, pollutant removal and support for vegetation growth (Davis et al., 2009). A mulch layer is usually added on top to retain moisture and heavy metals (Davis et al., 2001, 2003; Muthanna et al., 2007; Hatt et al., 2008).

Biological waste material (e.g. coconut, water treatment sludge, tea leaves, rice husk etc) can also be used in biofiltration systems as they have the potential to remove heavy metals via ion exchange,

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surface adsorption and complexation (Sud et al., 2008; Gadd, 2009). Using them as filter media material supports a waste minimisation strategy that reflects sustainable urban stormwater management (Barbosa et al., 2012). Published work found that coconut coir, compost and sludge are able to achieve high metal removal rates (Seelsaen et al., 2007; Gadd, 2009; Bhatnager et al., 2010; Chiang et al., 2012). Coconut coir and compost contain humic substances, cellulose, lignin and carboxyl groups that have a high tendency to bind metals especially between pH range 6–8 via surface complexation and ion exchange processes (Quek et al., 1998; Pinõ et al., 2006; Hasany and Ahmad, 2006). The drinking water treatment process produces sludge that is disposed at landfills. This material has a high heavy metal removal potential due to its large surface area to volume ratio, highly reactive surface and potential reuse via regeneration (Ippolito et al., 2011; Chiang et al., 2012).

Singapore is a highly urbanized island with limited land and water resources. Recent interest in BMPs arose from the need to reduce water quality impacts of stormwater runoff for environmental protection and also for water harvesting. Urban runoff is now being treated and re-used as drinking water. Heavy metals are important pollutants in Singapore's environment because they are toxic at low concentrations and accumulate in living organisms. They originate mainly from vehicles, roads and industrial activities dispersed across the island nation (Joshi and Balasubramanian, 2010; Yuen et al., 2012). Biofiltration systems are suitable because they have the potential to improve urban water quality, can be designed as small systems, provide an aesthetics effect and increase biodiversity in a highly urbanized setting (Kazemi et al., 2011). However, the limited availability of sand in the country means that other types of materials must be considered as alternatives for the filter media.

Published work based on laboratory and field studies show that the filter media materials may contribute heavy metals to the environment through leaching (Dietz and Clausen, 2006; Hatt et al., 2008). Ideally, the materials should contain negligible amounts of heavy metals. The depth of the filter media may also affect its performance. Higher metal uptake may occur in longer columns because there is more material for adsorption/ion-exchange and a longer contact time for these processes to take place (Sousa et al., 2010; Acheampong et al., 2012). However, longer columns may experience higher leaching rates from the mobilization of heavy metals within the media (Davis et al., 2003; Feng et al., 2012).

The objective of this study is to examine the heavy metal removal potential of five materials commonly found in Singapore to test their suitability as filter media material for biofiltration systems. The study uses laboratory columns and soil as a control for comparison with other materials. While published column studies on BMPs usually use soil as the main component with varying proportions of compost or other materials, we filled the columns with 100% treatments of each material to test their heavy metal removal potential. The study examined the leaching potential of the materials, investigated the effect of metal concentration/depth of filter media on metal removal, and quantified the metal accumulation in these materials. The heavy metals studied were Copper (Cu), Zinc (Zn), Cadmium (Cd) and Lead (Pb), because these are the predominant metals found in Singapore urban runoff (Joshi and Balasubramanian, 2010). Total/dissolved organic carbon (TOC/DOC) is included in sample analysis because some of these materials have high carbon content which provides an energy source for microbial processes that remove nutrient or heavy metals as well as plant growth. However, DOC leaching out of the media may also increase the mobility of heavy metals out of the columns (e.g. McLaughlan and Al-Mashaqbeh, 2009; Blecket et al., 2011).

2. Methods and materials

2.1. Materials tested

The filter media materials we tested were chosen using the criteria that they were commonly used and easily available to landscaping contractors in Singapore: potting soil (control), coconut coir, compost, sludge and a commercial mix. The potting soil is classified as loamy sand and has low organic matter content compared with compost and coconut coir (Table 1). The coconut coir fibre consists of coconut husk and hairy fibre chopped into different sizes, producing a highly porous and heterogeneous mix. The compost is horticultural waste made up of leaves and bark material found within Singapore. The sludge is a grey-coloured cake obtained from a local drinking water treatment plant which uses aluminium sulphate for the coagulation process. The sludge had already been de-watered in a filter press at the treatment plant. It was further dried in our laboratory and hammered into relatively uniform sand-sized particles to obtain a texture similar to potting soil. The commercial mix was added as a comparison with the other materials. The commercial material is a patented mix that consists of light granular particles (EnviroMix[®]). It is often used as a media for green roofs in Singapore. The estimated cost and lifetime of each media is given in Table 1.

Table 1
Properties of the materials tested.

	Potting soil	Coconut coir ^a	Compost	Commercial	Sludge ^b
pH	6.44	4.76	6.79	7.17	6.46
LOI (%)	6.6	96.2	75.7	2.6	38.0
Sand (%)	75.3	–	29.3	15.1	90.5
Silt (%)	18.3	–	57.7	5.9	9.2
Clay (%)	2.9	–	0.36	0.12	0.3
Cu (mg/kg) ^c	5.6	115.0	24.7	137.9	19.1
Zn (mg/kg) ^c	35.2	10.9	89.7	153.7	71.5
Cd (mg/kg) ^c	0.13	0.013	0.38	0.70	0.086
Pb (mg/kg) ^c	9.0	0.21	17.8	46.3	9.8
Expected lifespan (year) ^d	>1	0.5–1	0.5–1	Often longer than vegetation lifespan	Unknown
Estimated cost (USD/kg) ^e	0.12	0.4–0.79	0.16	0.79	–

^a It was not possible to use the hydrometer method to determine particle size for Coconut coir as it floats on the surface and does not sink at all within 24 h.

^b Sludge contains iron, aluminium hydroxides, sediment and humic substances removed from raw water as well as coagulating agents added to water, such as activated carbon and polymers (Makris and O'Connor, 2007). The pH of our Sludge is slightly acidic in comparison to other sludge studied, which is slightly alkaline (pH = 8.1, Chiang et al., 2012). The Sludge from the drinking water treatment plants in Singapore is aluminium-based (aluminium sulphate being the coagulant used). The pH falls within the expected range of 6.5 ± 0.3 for this type of sludge according to ASCE characterization (Ippolito et al., 2011). Aluminium toxicity should not be an issue at this pH value.

^c Metal extraction was conducted based on the USEPA 3051A method. Note that this is just an extraction method which does not involve complete digestion of the materials tested but provides a comparison for metals retained in the media at two different depths of each media.

^d Approximate estimate based on information from landscape contractors. Lifespan is expected to vary depending on plant grown, whether the material is used indoors or outdoors. Generally, coconut coir will disintegrate relatively quickly in a tropical environment. Compost is quickly taken up by plants in the tropical environment. There is no information about the lifespan of Sludge as this is a relatively new material used as filter media.

^e Estimated cost is based on information from various suppliers of the materials in Singapore. Currently, the higher price of coconut coir may be because this material is imported into Singapore whereas the other materials (e.g. potting soil, compost) are locally available. There is no price information for Sludge because we obtained it free from the water treatment plant. Conversion rate from Singapore dollar (SGD) to US Dollar is approximately 1SGD = 0.79 USD.

2.2. Column setup and experimental procedures

Ten PVC columns (inner and outer diameters of 103 mm and 110 mm respectively) were used to allow free flow of water by gravity, similar to that found in field conditions. The material in each column was packed to the desired depth of 300 mm and 600 mm by pouring 5 L of ultrapure deionised water (18.2 MΩcm) through the column (uniform rate of 400 ml/min). The 300 mm depth is the minimal requirement for vegetation growth and 600 mm is the recommended filter media depth for bioretention systems in Singapore, based on Australian guidelines (PUB, 2011). The setup consists of a long column and short column placed next to each other for each material tested.

The laboratory experiments were divided into three sets. For the first set, the columns were flushed five times with ultrapure deionised water to determine the leaching potential of metals and TOC from the filter media. The second set involved five dosings of low metal concentration synthetic runoff. In set three, the treatments were dosed five times with a high metal concentration synthetic runoff. A one month no-dosing period was observed between each set of experiments to let water drain from the columns. The potting soil showed signs of saturation during the last two dosings of each experimental run. Outflow from the column became increasingly slow.

2.3. Synthetic runoff preparation

Low and high metal concentration synthetic runoff containing Cu, Zn, Cd and Pb (experiment sets 2 and 3) were prepared by dissolving salts of the four metals (copper sulphate, zinc chloride, cadmium chloride and lead chloride) in ultrapure deionized water. We consider the dissolved metal fraction, as it is the most bioavailable and common form in urban runoff (Sansalone and Buchberger, 1997; Sun and Davis, 2007). The target low and high concentrations of each metal falls within the range typically found in Singapore urban runoff (Joshi and Balasubramanian, 2010): Cu (44.8 µg/L, 241 µg/L), Zn (436.4 µg/L, 1127 µg/L), Cd (1.82 µg/L, 4.57 µg/L) and Pb (51.3 µg/L, 90.3 µg/L). At the end of the high concentration dosings (Experiment 3), each column had been dosed with 2.29 mg of Cu, 15.73 mg of Zn, 0.06 mg of Cd and 1.30 mg of Pb.

2.4. Synthetic runoff dosing

Two litres of synthetic runoff was manually poured onto a column once every two days (uniform rate of 400 ml/min). Outflow from the column was collected 24 h later, as 90% of the dosed water exits the column within this time period. Synthetic runoff was applied again the following day. Column outflow was collected and tested for pH and TOC. For metal analysis, the outflow was filtered through 0.45-µm Millipore PTFE filter paper and acidified with 2% ultrapure nitric acid. These samples were stored at 4 °C before analysis. All glassware and plastic containers used for sample collection and analysis were acid-washed with dilute nitric acid and rinsed with ultrapure deionized water.

2.5. Collection of filter media material

Samples from the top 5 cm and bottom 5 cm of a column were collected at the end of the dosing experiments. These samples, together with the original materials, were air-dried, sieved and ground. Metal concentrations were determined by microwave acid digestion with 65% nitric acid (USEPA Method 3051A). The samples were also tested for pH, organic matter (via loss-on-ignition, LOI) and particle size (via hydrometer).

2.6. Elemental analysis

Metal concentrations in water samples and digested filter media samples were analysed using an ICP-MS (Agilent Technologies, 7700 Series). Five calibration standards were used for the analysis together with two certified reference materials (Environment Canada Certified Reference Material TMDA 64.2 and TM23.4). The detection limits for the four metals are: Cu: 0.167 ± 0.0077 µg/L, Zn: 0.155 ± 0.072 µg/L, Cd: 0.0011 ± 0.0006 µg/L, Pb: 0.0092 ± 0.0050 µg/L. The digested solid samples were also tested using the same calibration standards and two certified reference materials (NIST Standard Reference Material 2710a and 2711a). TOC/DOC was tested using a Shimadzu Total Organic Carbon Analyser (TOC-L).

2.7. Data analysis

The ability of each material to remove the four metals in synthetic runoff (removal efficiency, based on concentration) was calculated as:

$$\text{Removal efficiency (\%)} = \frac{(C_r - C_s)}{C_r} * 100\% \quad (1)$$

where C_r is the metal concentration in synthetic runoff and C_s is the metal concentration in outflow from the column (µg/L).

Another index, the 600/300 ratio, shows the magnitude of depth-related differences in metal outflow concentrations from the longer (600 mm) and shorter (300 mm) columns:

$$600/300 \text{ ratio} = \frac{C_{600}}{C_{300}} \quad (2)$$

where C_{600} is the mean outflow metal concentration (less inflow concentration) from the 600 mm column (µg/L); C_{300} is the mean outflow metal concentration (less inflow concentration) from the 300 mm column (µg/L).

The dosed/original ratio gives the magnitude of metal accumulation in the material after 15 dosings of water:

$$\text{Dosed/original ratio} = \frac{C_{\text{dosed}}}{C_{\text{original}}} \quad (3)$$

where C_{dosed} is the metal concentration in the materials after 15 dosings and C_{original} is the metal concentration in the original material prior to dosings (mg/kg).

To identify the magnitude of differences in metal accumulation between the top and bottom layers of the column material, we determined a top/bottom ratio, which is calculated as:

$$\text{Top/bottom ratio} = \frac{C_{\text{top}}}{C_{\text{bottom}}} \quad (4)$$

where C_{top} is the metal concentration in materials sampled from the top 5 cm and C_{bottom} is the metal concentration in materials sampled from the bottom 5 cm (mg/kg).

3. Results

3.1. Performance of filter media

All materials tested leached heavy metals when dosed with ultrapure deionised water. The mean metal concentrations at the end of the deionised water dosings were very low and can be considered as background trace metal levels; Cu (4.5 µg/L), Zn

(1.8 µg/L), Cd (0.02 µg/L) and Pb (0.11 µg/L) (Fig. 1). These values comply with the World Health Organization (WHO, 2011) drinking water limit for Cu (2000 µg/L), Zn (no guideline), Cd (3 µg/L) and Pb (10 µg/L).

The commercial mix leached the least Cu (2.31 µg/L), Zn (0.59 µg/L) and Pb (0.03 µg/L) (Table 2). Potting soil was ranked second best after the commercial mix. Compost leached out the highest concentrations of Cu (9.91 µg/L), Zn (14.97 µg/L) and Pb (0.33 µg/L); sludge, the highest for Cd (0.031 µg/L) (Table 2).

Subsequent dosings of the columns with synthetic runoff showed that filtering by potting soil, compost, commercial mix and sludge are able to achieve background metal concentration levels in outflow water (Fig. 1). Coconut coir managed to remove Pb, but outflow concentrations exceeded the WHO limit—especially for the high concentration dosings. At the end of the 3 experimental sets, the columns were still achieving trace metal concentrations similar to those observed during the deionised water flushing experiment (Set 1) with the exception of coconut coir (for Pb, Fig. 1).

TOC was negligible in all water dosed onto the columns (0.27 ± 0.19 mg/L). Outflow from the columns had initially high TOC concentration values but these gradually decreased with each set of experimental dosings (Fig. 2). Potting soil and the commercial mix had lowest TOC outflow concentrations (below 10 mg/L) whereas sludge, compost and coconut had highest concentrations (Table 2). By the tenth run, potting soil and the commercial mix began to show signs of organic carbon exhaustion due to the repeated column flushing. Compost, coconut coir and sludge were still leaching organic carbon (TOC approximately 10 mg/L) at the end of the 15 dosings (Fig. 2).

The occurrence of one-month dry periods between the experimental runs allowed us to briefly examine the effect of dry periods on TOC leaching and heavy metal removal. Blecken et al. (2009) observed that dry periods reduced heavy metal uptake due to a multitude of factors including mobilization, leaching of accumulated metals and flushing of metal-organic matter complexes upon wetting (Yin et al., 2002; Hatt et al., 2007a).

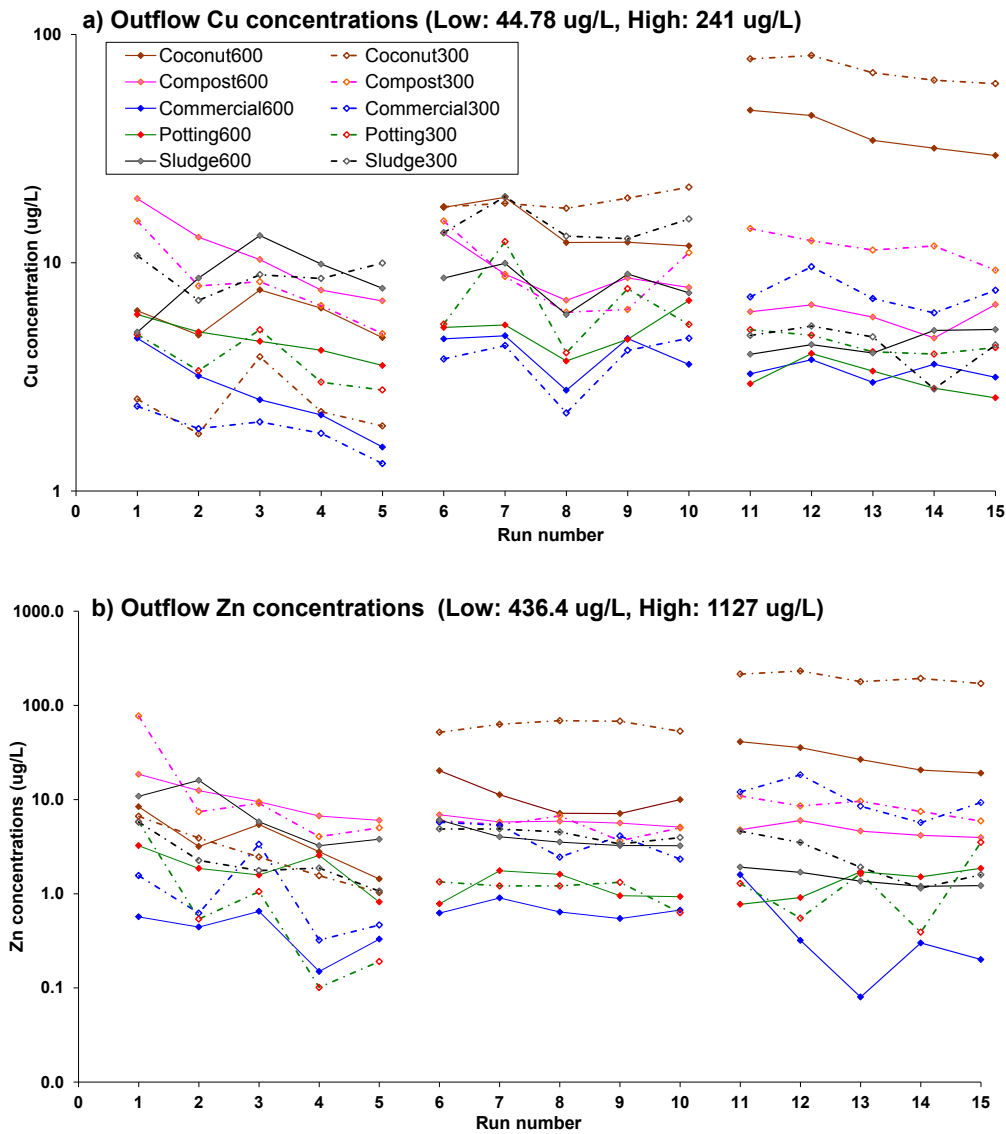


Fig. 1. Outflow metal concentrations for the deionised water flushings (Runs 1–5), low concentration dosings (Runs 6–10) and high concentration dosings (Runs 11–15) for the long (600 mm) and short (300 mm) columns. Insert graph for Cd and Pb show the outflow concentration values for Coconut coir, which are usually above those of the other materials. The low and high concentrations of synthetic runoff for each metal are also included.

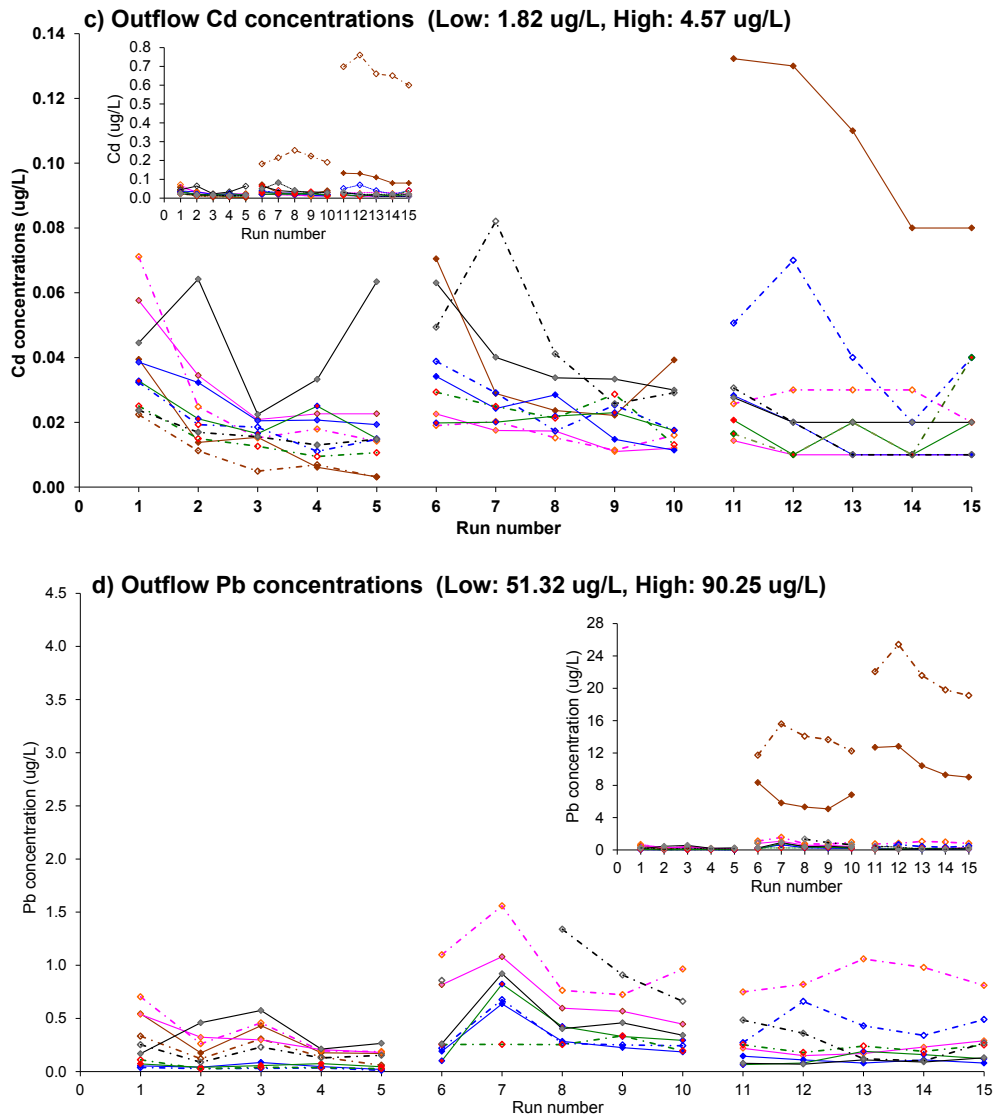


Fig. 1. (continued).

Outflow TOC concentrations were elevated at the start of an experimental run after a 1-month dry period, especially coconut coir (Fig. 2). Soil studies have shown that drying causes soil aggregate disruption and cracking, releasing organic matter that is usually protected in soil aggregates upon wetting (Denef et al., 2001). Coconut coir experiences the greatest amount of cracking due to its porous nature. It is hypothesized that subsequent wetting may cause expansion and release of organic carbon from this material.

The impact of a 1-month dry period between the low and high concentration dosing experiments did not seem to have an effect on metal removal efficiency in this study, a finding similar to that reported by Hatt et al. (2007a). The removal efficiencies for the low concentration dosings were already very high (above 90%) for Zn, Cd and Pb for all materials except coconut coir. Removal efficiencies improved during the high concentration dosings, especially for Cu; removal efficiency increased from 66.4% to 97.8% for sludge (Table 2). These trends suggest that while the materials were already performing well during the low concentration dosings, they were still able to maintain high removal performance for the high concentration dosing experiments, even for Cu removal. The removal efficiencies are also comparable to published findings

using soil-based media (Sun and Davies, 2007; Blecken et al., 2009, 2011; Feng et al., 2012).

3.2. Effect of filter media on heavy metal uptake/removal

Coconut coir performed comparatively worse in metal uptake. Although Zn and Cd uptake are high (removal efficiencies over 90%) and comparable with other studies using soil-based materials, the removal efficiencies were the lowest among the materials tested (Cu: 54% and 74%, Pb: 79% and 82% for low and high concentration dosings, respectively) (Table 2).

Of the four metals, Cu removal was noticeably lower for all materials. Research on coconut-based materials also reported poorer adsorption of Cu when compared to other metals (Quek et al., 1998; Sousa et al., 2010; Blecken et al., 2011; Anirudhan and Sreekumari, 2011). This was attributed to the smaller ionic radius of Cu which resulted in a poorer affinity for metal ions on active adsorption sites (Anirudhan and Sreekumari, 2011).

Overall, the experimental results show that potting soil and the commercial mix provided the best removal efficiency for the metal dosing experiments even though the dosing rate was very high (400 ml/min) (Table 2). Potting soil gave best metal uptake for high

Table 2Outflow concentrations (mean \pm standard deviation) and removal efficiency (RE) from the 10 columns for the deionised water flushing, low and high dose experiments.

		Potting soil		Coconut coir		Compost		Commercial mix		Sludge	
		$\mu\text{g/L}^a$	RE (%) ^c	$\mu\text{g/L}^a$	RE (%) ^c	$\mu\text{g/L}^a$	RE (%) ^c	$\mu\text{g/L}^a$	RE (%) ^c	$\mu\text{g/L}^a$	RE (%) ^c
TOC ^b	Flushing	11.9 \pm 14.4	na	10.7 \pm 6.3	na	74.0 \pm 78.5	na	6.1 \pm 5.6	na	131.1 \pm 88.7	na
	Low-dose	4.2 \pm 2.2	na	18.0 \pm 12.8	na	30.3 \pm 14.8	na	2.5 \pm 1.3	na	46.1 \pm 21.7	na
	High-dose	2.3 \pm 1.0	na	13.7 \pm 10.1	na	20.2 \pm 8.3	na	1.76 \pm 0.98	na	14.6 \pm 7.0	na
Cu	Flushing	4.2 \pm 1.0	na	4.2 \pm 2.1	na	9.9 \pm 4.5	na	2.3 \pm 0.99	na	8.9 \pm 2.2	na
	Low-dose	6.1 \pm 2.5	(82.7 \pm 6.1)	16.7 \pm 3.4	(54.2 \pm 9.8)	9.3 \pm 3.1	(73.6 \pm 8.6)	4.0 \pm 0.9	(87.4 \pm 6.1)	11.5 \pm 4.1	(66.4 \pm 13.1)
	High-dose	3.8 \pm 0.9	(98.2 \pm 0.4)	53.7 \pm 19.1	(73.8 \pm 9.0)	8.9 \pm 3.4	(95.8 \pm 1.5)	5.4 \pm 2.3	(97.4 \pm 1.1)	4.5 \pm 0.7	(97.8 \pm 0.4)
Zn	Flushing	1.5 \pm 1.8	na	3.2 \pm 2.4	na	15.0 \pm 20.9	na	0.59 \pm 0.92	na	5.5 \pm 5.2	na
	Low-dose	1.2 \pm 0.4	(99.7 \pm 0.08)	36.0 \pm 27.0	(92.1 \pm 6.0)	5.6 \pm 0.9	(98.8 \pm 0.2)	2.3 \pm 2.1	(99.5 \pm 0.4)	4.2 \pm 0.9	(99.1 \pm 0.2)
	High-dose	1.4 \pm 0.9	(99.9 \pm 0.08)	113.3 \pm 91.1	(90.5 \pm 7.5)	6.6 \pm 2.4	(99.5 \pm 0.2)	5.6 \pm 6.3	(99.5 \pm 0.5)	2.1 \pm 1.1	(99.8 \pm 0.09)
Cd	Flushing	0.017 \pm 0.008	na	0.010 \pm 0.010	na	0.028 \pm 0.018	na	0.021 \pm 0.009	na	0.031 \pm 0.020	na
	Low-dose	0.02 \pm 0.005	(98.6 \pm 0.3)	0.13 \pm 0.1	(92.1 \pm 6.1)	0.02 \pm 0.004	(99.0 \pm 0.2)	0.02 \pm 0.009	(98.5 \pm 0.5)	0.04 \pm 0.02	(97.3 \pm 1.1)
	High-dose	0.02 \pm 0.009	(99.7 \pm 0.2)	0.4 \pm 0.3	(91.3 \pm 6.8)	0.02 \pm 0.009	(99.6 \pm 0.2)	0.03 \pm 0.02	(99.3 \pm 0.5)	0.02 \pm 0.007	(99.6 \pm 0.2)
Pb	Flushing	0.05 \pm 0.02	na	0.24 \pm 0.16	na	0.33 \pm 0.18	na	0.03 \pm 0.02	na	0.25 \pm 0.15	na
	Low-dose	0.3 \pm 0.2	(99.3 \pm 0.4)	9.9 \pm 4.0	(79.4 \pm 8.6)	0.9 \pm 0.3	(98.2 \pm 0.7)	0.3 \pm 0.2	(99.3 \pm 0.4)	1.1 \pm 1.2	(97.7 \pm 2.7)
	High-dose	0.2 \pm 0.07	(99.8 \pm 0.08)	16.2 \pm 6.0	(81.9 \pm 6.8)	0.6 \pm 0.4	(99.4 \pm 0.4)	0.27 \pm 0.2	(99.7 \pm 0.2)	0.2 \pm 0.1	(99.8 \pm 0.2)

Mean pH and TOC of the ultrapure deionised water used for the flushing, low and high dose experiments is 6.91 \pm 0.67 and 0.27 \pm 0.19 mg/L.^a TOC concentration is in mg/L whereas all metal concentrations are reported in $\mu\text{g/L}$.^b Only TOC concentrations are reported here. For some reason, DOC concentrations from our samples were \geq than the TOC concentrations for the same sample. This was attributed to contamination from the laboratory processing, particularly during sample filtration despite care taken to prevent contamination. Since particulate organic carbon in many types of water samples is very low (<5%), TOC is often equal to DOC within analytical precision (Mopper and Qian, 2006). Our column effluent samples were generally quite free of particulates with the exception of the potting soil (300 mm) column. Therefore, TOC is taken to represent, to some extent, the DOC concentration of our column outflow samples.^c Metal removal efficiencies for soil-based filter media reported by other authors (Hsieh and Davis, 2005; Sun and Davis, 2007; Blecken et al., 2009, 2011; Feng et al., 2012): Cu: 24–93%; Zn: 94–99%; Cd: 89.6–99%; Pb: 46–99%.

concentration dosings. Sludge did comparatively well for the high metal dosings (ranked second). Coconut coir consistently provided the worst metal uptake of all metals for all experimental runs. High removal efficiencies were achieved despite a short contact time with the material; 90% of inflow water exited the columns within 24 h. Typically, most metal uptake occurs within the first 10–20 min from the start of an experiment (Jang et al., 2005; Quek et al., 1998).

3.3. Effect of filter media depth on heavy metal leaching and uptake/removal

The 600/300 ratio compares depth-related differences in outflow metal concentrations between the long and short columns

(Equation (2), Table 3). Statistical analysis of the outflow concentrations between long and short columns were conducted using the Analysis of Variance (ANOVA) test ($\alpha = 0.05$, Fig. 1, Table 3).

For the deionised water dosings, the longer columns generally had higher outflow concentrations of all metals (the 600/300 ratio values are greater than 1), reflecting the greater amount of material available for leaching and longer contact time between material and synthetic runoff (Sousa et al., 2010, Table 3). Outflow concentrations were not always proportional to the column length. The greatest depth differences were observed for the longer column filled with sludge. This column leached out more Zn (4.84 times), Cd (2.78 times) and Pb (2.08 times) than its shorter counterpart. Outflow from the longer coconut coir column had Cu concentrations 2.4 fold higher than that of its shorter counterpart (Table 3).

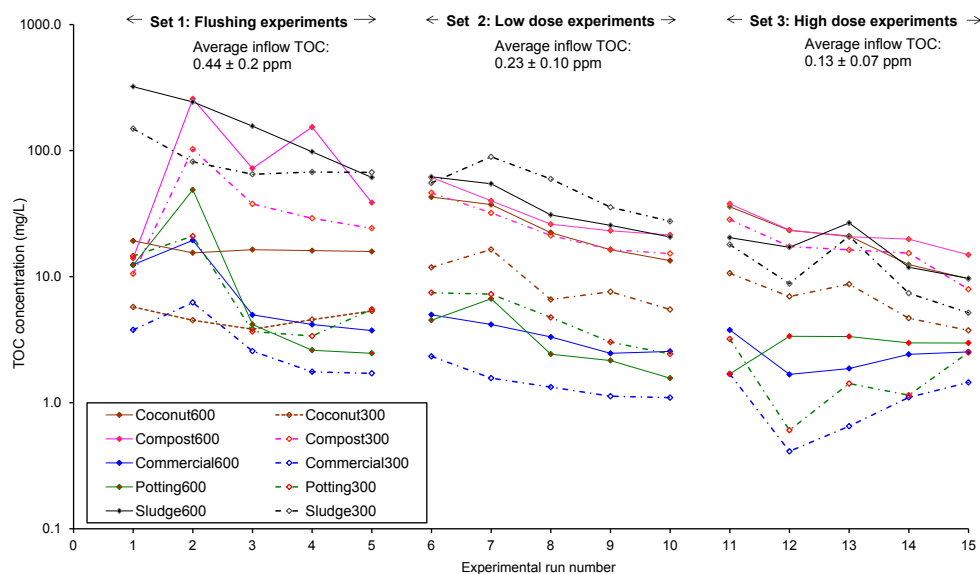


Fig. 2. Outflow TOC concentrations (less influent concentrations) for the deionised water dosings (Runs 1–5), low concentration dosings (Runs 6–10), high concentration dosings (Runs 11–15) for the long (600 mm) and short (300 mm) columns.

Table 3
The 600/300 ratio for deionised water flushings, low and high concentration dosings based on average outflow concentrations (less inflow concentration) from the 600 mm and 300 mm columns.

600/300 ratio ^a		Potting soil	Coconut coir	Compost	Commercial mix	Sludge
Cu	Flushing	1.22	2.42 ^b	1.33	1.53	0.99
	Low-dose	1.07	1.50 ^b	1.01	0.89	1.41 ^b
	High-dose	1.01	1.24 ^b	0.99	1.03	1.00
Zn	Flushing	1.08	1.66	0.53	0.22	4.84
	Low-dose	1.00	1.13 ^b	1.00	1.00	1.01
	High-dose	1.00	1.17 ^b	1.00	1.01	1.00
Cd	Flushing	1.70	1.60	1.10	1.51	2.78 ^b
	Low-dose	0.99	1.13 ^b	1.01	1.01	1.02
	High-dose	0.99	1.14 ^b	1.01	1.01	0.99
Pb	Flushing	1.30	1.59	0.85	2.12	2.08
	Low-dose	1.00	1.22 ^b	1.00	1.00	1.08
	High-dose	1.00	1.16 ^b	0.99	1.01	1.00

^a Values above 1 indicate that the outflow concentrations from the 600 mm columns are higher than the shorter 300 mm columns and vice versa.

^b Difference between the average outflow-inflow concentration of each material for the 600 mm and 300 mm columns are statistically significant at $\alpha = 0.05$ (ANOVA).

Despite the 600/300 ratios showing higher outflow metal concentrations for most materials, only Coconut coir (Cu) and Sludge (Cd) columns showed depth-related differences that were statistically significant (Table 3). Further, the longer columns sometimes leached less than its shorter counterpart although the depth-related differences were not statistically significant (e.g. longer compost column for Zn and Pb, Table 3). These exceptions were probably a reflection of material heterogeneity for each pair of columns.

When the same columns were dosed with synthetic runoff, both short and long columns demonstrated almost similar outflow metal concentrations; values of the 600/300 ratios were close to 1 (Table 3). Exceptions are seen for the longer coconut coir (all metals) and Sludge (for Cu only) columns where the differences in outflow metal concentrations between the long and short columns were statistically significant (Table 3). The longer coconut coir column had higher metal outflow concentrations for both low and high concentration dosings for all metals (Table 3). The difference in Cu concentrations between the long and short Sludge column is statistically significant only for the low concentration dosings (600/300 ratio = 1.41, Table 3).

3.4. Location of metal uptake within the column

The materials were generally enriched in heavy metals at the end of the dosing experiments especially the upper layers; potting soil (Cu, Zn) and coconut coir (Zn, Cd, Pb), compost (all metals), Sludge (Cd) (Table 4). The main mechanism for metal enrichment of all four metals on these materials is by ion-exchange and complexation with the negatively charged surface sites of materials tested. These processes are enhanced in the presence of organic matter (e.g. coconut coir, compost and sludge) which contains polar functional groups that dissociate and take part in metal uptake through surface complexation and exchange of metal cations (Jang et al., 2005; Hasany and Ahmad, 2006). These processes are also enhanced in materials with a high specific surface area (e.g. sludge). In addition, the alkaline or near-alkaline pH of most materials, except coconut coir, favors metal binding due to a decrease in surface potential and proton competition (Yin et al., 2002, Table 1). There were some exceptions where metal concentrations at the end of 15 dosings were similar to or lower than concentrations at the beginning of the experiment; Cd

Table 4
Ratio of dosed/original heavy metal concentrations and top/bottom heavy metal concentrations for the four metals.

		Potting soil						Coconut coir						Compost						Commercial mix						Sludge					
		600 mm		300 mm		600 mm		300 mm		600 mm		300 mm		600 mm		300 mm		600 mm		300 mm		600 mm		300 mm		600 mm		300 mm			
		Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom				
Cu	Dosed/original ^a	4.20	12.14	4.70	4.06	0.38	0.12	2.12	0.95	2.27	0.98	1.27	1.29	1.04	1.02	1.33	0.98	1.37	1.04	1.04	1.02	1.04	1.02	1.04	1.02	1.04	1.02	1.04			
	Top/bottom ^b	0.35	–	1.16	–	3.11	–	2.23	–	2.30	–	0.99	–	1.02	–	1.36	–	1.31	–	1.02	–	1.02	–	1.02	–	1.02	–	1.31			
Zn	Dosed/original ^a	2.2	3.4	2.4	2.1	17.9	1.5	3.6	1.0	2.7	1.0	1.06	0.93	0.76	0.87	1.7	1.0	1.7	1.1	0.88	–	0.88	–	0.88	–	1.72	–	1.58			
	Top/bottom ^b	0.65	–	1.11	–	11.7	–	3.66	–	2.70	–	1.14	–	1.14	–	1.72	–	1.02	–	0.96	–	0.96	–	0.96	–	2.8	–	3.0			
Cd	Dosed/original ^a	1.3	0.9	1.2	0.7	50.7	4.3	2.9	0.7	2.9	0.6	1.03	0.96	0.96	0.97	2.8	0.7	3.0	0.7	1.07	–	1.07	–	1.07	–	3.82	–	4.56			
	Top/bottom ^b	1.42	–	1.67	–	11.9	–	4.38	–	4.75	–	1.07	–	1.07	–	3.82	–	3.0	–	0.99	–	0.99	–	0.99	–	1.4	–	1.1			
Pb	Dosed/original ^a	2.0	2.9	1.9	2.2	76.3	7.4	5.8	0.8	1.7	0.7	1.06	1.01	0.97	1.00	1.4	1.0	1.5	1.1	1.06	–	1.06	–	1.06	–	1.47	–	1.30			
	Top/bottom ^b	0.69	–	0.85	–	10.3	–	7.7	–	2.31	–	1.05	–	1.05	–	1.47	–	1.30	–	0.97	–	0.97	–	0.97	–	1.47	–	1.30			

^a Values above 1 indicate that the metal concentration in the materials sampled at the end of 15 dosings is higher than the original pre-dosed material and vice versa (i.e. metal enrichment on the materials occur after dosings).
^b Values above 1 indicate that the materials sampled from the top 5 cm of the column have a higher metal concentration than the bottom 5 cm and vice versa (i.e. metal enrichment occurs in the top 5 cm of the column).

(bottom layers of compost, potting soil, commercial mix and sludge), Zn (Commercial mix) and Cu (coconut coir) (Table 4).

The dosed/original ratio of Cu for coconut coir is exceptionally low (ranging from 0.11 to 0.38) for both columns suggesting that Cu leaching occurred over the course of the experiments (Table 4). Copper has the strongest affinity for dissolved organic matter (DOM) through the formation of Cu-DOM complexes. The mobility of DOM determines Cu leaching (Yin et al., 2002; Toribio and Romanya, 2006). The coconut coir material had the highest LOI (96.2%) amongst the five materials and ranked third in TOC effluent concentrations. Only compost and sludge had higher TOC, but the effluent from the coconut coir columns was darkest in colour indicating outflow rich in humic substances. These substances contain polar functional groups that dissociate encourage heavy metal uptake through surface complexation and exchange of metal cations (Jang et al., 2005; Hasany and Ahmad, 2006). The abundance of humic substances leaching out of the coconut coir columns mobilises Cu to a greater extent than other materials such as compost and sludge, resulting in the lower Cu removal efficiency for coconut coir.

The top/bottom ratio shows that not all the materials tested exhibited the typical top heavy metal accumulation reported by many researchers (e.g. Davis et al., 2001, 2003; Dietz and Clausen, 2006; Muthanna et al., 2007; Sun and Davis, 2007; Blecken et al., 2009; Hatt et al., 2008) (Table 4). The depth–concentration profile of potting soil showed higher heavy metal concentrations in the bottom layers of the longer column; Cu (0.35), Zn (0.65) and Pb (0.69, Table 4). During the course of the experiments, outflow from both potting soil columns became increasingly slow. Compaction of the material led to lower percolation rates. Accumulation of water at the base of the column resulted in a longer contact time between solution and media. Increased metal uptake via adsorption or ion-exchange caused the bottom layer to have higher metal concentrations (Hasany and Ahmad, 2006; Sousa et al., 2010).

The Commercial mix exhibited quite an even distribution of metal concentration between the top and bottom layers; Zn (0.88), Cd (0.99) and Pb (0.97) for the shorter column and for Cu (0.99, longer column) (Table 4). The porous and homogeneous nature of this material allows water to flow through the column quickly, possibly resulting in a more even distribution of metal uptake throughout the column.

The long and short columns filled with coconut coir, compost and sludge all exhibited the typical top heavy accumulation of heavy metals; values of the top/bottom ratio were greater than 1 (Table 4). This may explain why depth of material packed in a column (300 mm or 600 mm) does not play an important role in determining heavy metal outflow concentration from these materials since most of the metals dosed onto the columns are removed by the upper layers (see Table 4). Subsequent layers of materials play an insignificant role in metal removal.

4. Discussion

4.1. Design and maintenance considerations

Generally, all the materials are suitable for use as filter media materials on their own when compared to soil-based materials tested in this study (potting soil) and elsewhere (Table 2). All leached minimal heavy metals and gave very high removal efficiencies (>90%) for the four metals, with the exception of coconut coir. The poor performance of coconut coir may be attributed to its porous and heterogeneous nature. The pore spaces were unevenly sized and may be connected in the column to form preferential flow pathways. This loose structure allowed rapid movement of water through the column, minimising contact with the coconut material

and lowering metal removal performance (especially for Cu and Pb). All materials tested also had fast drainage rates, even potting soil, although it showing signs of decreased outflow rates with increased dosings. The high metal uptake and fast drainage rates make them suitable as filter media material in Singapore.

Part of the reason for the good performance of the materials could be because the pH of the deionized water used in making the synthetic runoff (6.91) fell within the optimal pH range (6–8) for metal sorption to occur (Davis et al., 2001; Pinõ et al., 2006; Acheampong et al., 2012; Anirudhan and Sreeumari, 2011). The mean pH of Singapore's stormwater falls within this range (Chui, 1997). The mean pH of the column outflow ranges between 6.80 and 7.83 for the five materials tested. Some buffering is observed for coconut coir. The pH of coconut coir material is acidic (4.76) but average pH of the effluent was only slightly acidic (6.80 ± 0.56 , Tables 1 and 2).

A variety of factors will complicate the application of our laboratory results to field situations. Urban runoff contains fine sediment (usually less than $6 \mu\text{m}$ in diameter) that may clog up the filter media, decreasing hydraulic conductivity over time (Siriwardene et al., 2007; Hatt et al., 2008; Le Coustumer et al., 2012). The extent of clogging is a function of sediment concentration and hydraulic characteristics of the flow. Plant roots, especially those with thick roots, and soil organisms may counteract such changes in hydraulic conductivity through the creation of macropores (Le Coustumer et al., 2012). The fine sediments are also enriched with heavy metals above natural levels (e.g. Cu, Zn enriched over 10 fold, Yuen et al., 2012). Changes in temperature, pH and DOC of the biofiltration system from the decomposition of organic matter and acids from root systems may re-mobilise metals previously sorbed onto fine sediments (Laurenson et al., 2013). The production of dissolved organic carbon affects the mobility of metals such as Cu, discussed earlier, as well as encourages nutrient leaching from the material (McLaughlan and Al-Mashaqbeh, 2009).

For design considerations, potting soil and the commercial mix are the best choices for heavy metal removal, even though compost and sludge also perform very well. The compaction of potting soil with repeated dosings presents a challenge to water drainage, especially during wet monsoon conditions when rainfall occurs frequently. During such conditions, it is very likely that ponding or overflow occurs from the biofiltration system resulting in minimal or no heavy metal treatment. Heavy metal accumulation in the bottom layers of the potting soil material also creates difficulties for maintenance and may result in heavy metal leaching to the groundwater over time. The commercial mix does not have the problems faced by potting soil and may be suitable for most applications in Singapore. Its main disadvantages are that it must be supplied through a dealer and it costs more than six times the price of potting soil.

Compost, coconut coir and sludge possess properties that may alleviate the problems associated with potting soil. Their porous structure ensures a relatively high hydraulic conductivity on a longer timescale. None of the columns showed signs of decreasing outflow rates throughout the 15 dosings. Their high organic carbon content supplements the low levels found in potting soil (LOI = 6.6%, Table 1) for nitrogen removal via denitrification although care must be taken not to add too much to prevent excessive leaching of organic carbon and nutrients (especially nitrogen) out of the biofiltration systems (Hatt et al., 2007b). Hatt et al. (2007b) found that adding organic materials such as compost and mulch to their filter media helped Zn uptake but led to nutrient leaching out of their system. In this respect, careful consideration has to be given to the relative proportions of compost and sludge that is added to potting soil. Although compost and sludge leach metals, these concentrations are negligible and below

WHO drinking water limits. Coconut coir also has oxygen-filled pores that support microbial populations that aid in heavy metal removal (Park et al., 2011).

To obtain optimal heavy metal removal efficiency and maintain adequate drainage rates using easily available materials, we recommend using potting soil as the main component of the filter media and mixing in compost or sludge. The top 5 cm of the potting soil filter media can be mixed with coconut coir which acts as the equivalent of the mulch layer found in many bioretention systems. This layer performs the major role of heavy metal uptake via adsorption (e.g. Davis et al., 2001; Dietz and Clausen, 2006; Sun and Davis, 2007; Muthanna et al., 2007). The porous structure of coconut coir enhances hydraulic conductivity and also lowers the chance of surface layer clogging by fine sediments found in urban runoff. The poor removal of Cu and Pb from coconut coir material at the surface may be resolved by adsorption on the potting soil/compost/sludge mix found below the top 5 cm depth of this filter media.

Hatt et al. (2007b) suggested a shorter filter media depth given that the majority of metal uptake occurs on the surface. In Singapore's case, we recommend using thick filters for maximum metal removal, optimal vegetation growth and treating the large urban runoff volumes generated in Singapore, since our results suggests that depth generally does not have an effect on metal removal (Table 3). Since the metals are enriched mainly in the upper 5 cm of the media, maintenance of the biofiltration system by removing the upper layers will have a smaller impact on a system with a deeper filter media layer than a shallower one.

Finally, maintenance of the filter media material is crucial for the continued functioning of the biofiltration system which can last up to 15–20 years (Davis et al., 2001). From our results, if potting soil alone is used, then maintenance involves the removal of the entire media since the bottom layers are metal-enriched. The inclusion of compost and sludge, to encourage metal accumulation of heavy metals, only requires the removal of the upper 5–10 cm of the filter media material. This removes the layer that is most metal-enriched and most clogged by fine sediments (e.g. Hatt et al., 2008). If coconut coir is used on the top layer, then replacement with new coconut material will be necessary as it shrinks in volume over time (Bhatnager et al., 2010). Hatt et al. (2008) suggests scraping the top layer of the filter media once every two years, but this frequency may be increased if more easily biodegradable material such as coconut coir or compost is used. Replacement of the upper layers of the filter media not only extends the life of the system but is also less disruptive and more economical. The heavy metal-saturated material that is removed may then be re-generated using acids (e.g. HNO₃, HCL, EDTA) where the hydrogen ion in the acids displaces the metals bonded to the media (Anirudhan and Sreekumari, 2011).

5. Concluding remarks

The present study examined the metal removal capability of five materials available in Singapore as potential filter media material for the construction of biofiltration systems. Three of the materials were compost, coconut coir and sludge. These materials have the potential to be recycled as filter media materials, therefore encouraging the sustainable use of waste materials. Heavy metal leaching was minimal and TOC was gradually exhausted over time during the 3 experimental sets. The one month dry period between experiments seem to only affect TOC but not heavy metal removal. Depth-related differences were negligible and heavy metal enrichment occurred on the upper layers of the materials except for the potting soil and commercial mix.

From these results, we find that the materials tested can all be used individually as filter media material, with the exception of

coconut coir which may be best used as an additive to the other materials. Despite the high dosing rates and variable metal concentrations in the synthetic runoff, they performed equally well when compared with other soil-based materials reported in published literature. The control, potting soil, performs the best but problems with compaction and bottom metal accumulation trends require the addition of other materials such as compost/sludge/coconut coir to enhance its performance. Using materials high in organic carbon in the filter media has the possibility of Cu leaching and poor Pb uptake seen in the case of the coconut coir material. Future work in this area includes examining the relative proportions of compost/sludge/coconut coir as additives, because this will vary on a case-to-case basis depending on target pollutant treatment, as well as the long-term performance of these materials to aid in biofiltration systems design that are suitable for Singapore's climate and urban runoff conditions.

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References

- Acheampong, M.A., Pakshirajan, K., Annachhatre, A.P., Lens, P.N.L., 2012. Removal of Cu (II) by biosorption onto coconut shell in fixed-bed column systems. *J. Ind. Eng. Chem.* <http://dx.doi.org/10.1016/j.jiec.2012.10.029>.
- Anirudhan, T.S., Sreekumari, S.S., 2011. Adsorptive removal of heavy metal ions from industrial effluents using activated carbon derived from waste coconut buttons. *J. Environ. Sci.* 23, 1989–1998.
- Barbosa, A.E., Fernandes, J.N., David, L.M., 2012. Key issues in sustainable urban stormwater management. *Water Res.* 46, 6787–6798.
- Bhatnager, A., Vilar, V.J.P., Botelho, C.M.S., Boaventura, R.A.R., 2010. Coconut-based biosorbents for water treatment- a review of the recent literature. *Adv. Colloid Interfac.* 100, 1–15.
- Blecken, G.-T., Marsalek, J., Viklander, M., 2011. Laboratory study of stormwater biofiltration in low temperatures: total and dissolved metal removals and fates. *Water Air Soil Poll.* 219, 303–317.
- Blecken, G.-T., Zinger, Y., Deletic, A., Fletcher, T.D., Viklander, M., 2009. Influence of intermittent wetting and drying conditions on heavy metal removal by stormwater biofilters. *Water Res.* 43, 4590–4598.
- Chiang, Y.W., Ghyselbrecht, K., Santos, R.M., Martens, J.A., Swenne, R., Cappuyns, V., Meesschaert, B., 2012. Adsorption of multi-heavy metals onto water treatment residuals: sorption capacities and applications. *Chem. Eng. J.* 200–202, 405–415.
- Chui, P.C., 1997. Characteristics of stormwater quality from two urban watersheds in Singapore. *Environ. Monit. Assess.* 44, 173–181.
- Davis, A.P., Hunt, W.F., Traver, R.G., Clar, M., 2009. Bioretention technology: overview of current practice and future needs. *J. Environ. Eng. ASCE* 135, 109–117.
- Davis, A.P., Shokouhian, M., Sharma, H., Minami, C., 2001. Laboratory study of biological retention for urban stormwater management. *Water Environ. Res.* 73, 5–14.
- Davis, A.P., Shokouhian, M., Sharma, H., Minami, C., Winogradoff, D., 2003. Water quality improvement through bioretention: lead, copper and zinc removal. *Water Environ. Res.* 75, 73–81.
- Denef, K., Six, J., Bossuyt, H., Frey, S.D., Elliott, E.T., Merckx, R., Paustian, K., 2001. Influence of dry-wet cycles on the interrelationship between aggregate, particulate organic matter, and microbial community dynamics. *Soil Biol. Biochem.* 33, 1599–1611.
- Dietz, M.E., Clausen, J.C., 2006. Saturation to improve pollutant retention in a rain garden. *Environ. Sci. Technol.* 40, 1335–1340.
- Feng, W., Hatt, B.E., McCarthy, D.T., Fletcher, T.D., Deletic, A., 2012. Biofilters for stormwater harvesting: understanding the treatment performance of key metals that pose a risk for water use. *Environ. Sci. Technol.* 46, 5100–5108.
- Gadd, G.M., 2009. Biosorption: critical review of scientific rationale, environmental importance and significance for pollution treatment. *J. Chem. Technol. Biotechnol.* 84, 13–28.
- Hasany, S.M., Ahmad, R., 2006. The potential of cost-effective coconut husk for the removal of toxic metal ions for environmental protection. *J. Environ. Manage.* 81, 286–295.
- Hatt, B.E., Fletcher, T.D., Deletic, A., 2007a. Hydraulic and pollutant removal performance of stormwater filters under variable wetting and drying regimes. *Water Sci. Technol.* 56, 11–19.

- Hatt, B.E., Deletic, A., Fletcher, T.D., 2007b. Stormwater reuse: designing biofiltration systems for reliable treatment. *Water Sci. Technol.* 55, 201–209.
- Hatt, B.E., Fletcher, T.D., Deletic, A., 2008. Hydraulic and pollutant removal performance of fine media stormwater filtration systems. *Environ. Sci. Technol.* 42, 2535–2541.
- Hsieh, C.H., Davis, A.P., 2005. Evaluation and optimization of bioretention media for treatment of urban storm water runoff. *J. Environ. Eng. ASCE* 131, 1521–1531.
- Ippolito, J.A., Barbarick, K.A., Elliott, H.A., 2011. Drinking water treatment residuals: a review of recent uses. *J. Environ. Qual.* 40, 1–12.
- Jang, A., Seo, Y., Bishop, P.L., 2005. The removal of heavy metals in urban runoff by sorption on mulch. *Environ. Pollut.* 133, 117–127.
- Joshi, U.M., Balasubramanian, R., 2010. Characteristics and environmental mobility of trace elements in urban runoff. *Chemosphere* 80, 310–318.
- Kazemi, F., Beecham, S., Gibbs, J., 2011. Streetscape biodiversity and the role of bioretention swales in an Australian urban environment. *Lands. Urban Plan.* 101, 139–148.
- Laurenson, G., Laurenson, S., Bolan, N., Beecham, S., Clark, I., 2013. The role of bioretention systems in the treatment of stormwater. In: Sparks, D.L. (Ed.), *Advances in Agronomy*, vol. 120. Elsevier Inc, PA., pp. 223–274.
- Le Coustumer, S., Fletcher, T.D., Deletic, A., Barraud, S., Poelsma, P., 2012. The influence of design parameters on clogging of stormwater biofilters: a large-scale column study. *Water Res.* 46, 6743–6752.
- Makris, K.C., O'Connor, G.A., 2007. Beneficial utilization of drinking-water treatment residuals as contaminant-mitigating agents. In: Sarkar, D., Datta, R., Hannigan, R. (Eds.), *Developments in Environmental Science Volume 5 Concepts and Applications in Environmental Geochemistry*. Elsevier Ltd., Amsterdam, pp. 609–635.
- McLaughlan, R.G., Al-Mashaqbeh, O., 2009. Effect of media type and particle size on dissolved organic carbon release from woody filtration media. *Bioresour. Technol.* 100, 1020–1023.
- Mopper, K., Qian, J., 2006. Water analysis: organic carbon determinations. In: Meyers, R.A. (Ed.), *Encyclopaedia of Analytical Chemistry*. John Wiley & Sons, Ltd., New York, pp. 1–9.
- Muthanna, T.M., Viklander, M., Gjesdahl, N., Thorolfsson, S.T., 2007. Heavy metal removal in cold climate bioretention. *Water Air Soil Poll.* 183, 391–402.
- Park, J.H., Lamb, D., Paneerselvama, P., Choppala, G., Bolan, N., Chung, J.-W., 2011. Role of organic amendments on enhanced bioremediation of heavy metal(loid) contaminated soils. *J. Hazard Mater.* 185, 549–574.
- Pinõ, G.H., de Mesquita, L.M.S., Torem, M.L., Pinto, G.A.S., 2006. Biosorption of cadmium by green coconut shell powder. *Min. Eng.* 19, 380–387.
- Public Utilities Board (PUB), 2011. *Engineering Procedures for ABC Waters Design Features*. PUB Singapore, Available: <http://www.pub.gov.sg/abcwaters/abcwatersdesignguidelines/Pages/ABCDesignGuidelines.aspx>.
- Quek, S.Y., Al-Duri, B., Wase, A.J., Forster, C.F., 1998. Coir as a biosorbent of copper and lead. *Process Saf. Environ.* 76, 50–54.
- Roy-Poirier, A., Champagne, P., Fillion, Y., 2010. Review of bioretention system research and design: past, present and future. *J. Environ. Eng. ASCE* 136, 878–889.
- Sansalone, J.J., Buchberger, S.G., 1997. Partitioning and first flush of metals in urban roadway storm water. *J. Environ. Eng. ASCE* 123, 134–143.
- Seelsaen, N., McLaughlan, R., Moore, S., Stuetz, R.M., 2007. Influence of compost characteristics on heavy metal sorption from synthetic stormwater. *Water Sci. Technol.* 55, 219–226.
- Siriwardene, N.R., Deletic, A., Fletcher, T.D., 2007. Clogging of stormwater gravel infiltration systems and filters: insights from a laboratory study. *Water Res.* 41, 1433–1440.
- Sousa, F.W., Oliveira, A.G., Ribeiro, J.P., Rosa, M.F., Keukeleire, D., Nascimento, R.F., 2010. Green coconut shells applied as adsorbent for removal of toxic metal ions using fixed-bed column technology. *J. Environ. Manage.* 91, 1634–1640.
- Sud, D., Mahajan, G., Kaur, M.P., 2008. Agriculture waste material as potential adsorbent for sequestering heavy metal ions from aqueous solutions – a review. *Bioresour. Technol.* 99, 6017–6027.
- Sun, X., Davis, A.P., 2007. Heavy metal fates in laboratory bioretention systems. *Chemosphere* 66, 1601–1609.
- Toribio, M., Romanyà, J., 2006. Leaching of heavy metals (Cu, Ni and Zn) and organic matter after sewage sludge application to Mediterranean forest soils. *Sci. Total Environ.* 363, 11–21.
- World Health Organisation (WHO), 2011. *Guidelines for Drinking-water Quality*, fourth ed. WHO, Geneva, Switzerland.
- Yin, Y., Impellitteri, C.A., You, S.-J., Allen, H.E., 2002. The importance of organic matter distribution and extract soil solution ratio on the desorption of heavy metals from soils. *Sci. Total Environ.* 287, 107–119.
- Yuen, J.Q., Olin, P.H., Lim, H.S., Benner, S.G., Sutherland, R.A., Ziegler, A.D., 2012. Accumulation of potentially toxic elements in road deposited sediments in residential and light industrial neighbourhoods of Singapore. *J. Environ. Manage.* 101, 151–163.