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Greening the Grey Infrastructure: Green Adsorbent Media for Catch Basin Inserts to Remove

Stormwater Pollutants

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Abstract

Stormwater pollution is a major cause of water quality impairment. Much of the existing grey stormwater infrastructures provide little or no treatment, especially for dissolved pollutants. Due to the capital cost of installing new infrastructure, retrofitting the existing grey infrastructures is a promising alternative to reduce stormwater pollution. In this study, aluminum-based drinking water treatment residuals (WTR), a byproduct from drinking water treatment, was combined with other common materials (sand and carbon material) to formulate an adsorbent media for use in catch basin inserts to remove total petroleum hydrocarbons (TPH), and dissolved Cu, Pb, and Zn from stormwater runoff. Hydraulic and treatment performance of the adsorbent media were optimized in laboratory column experiments. A dual-layer media, WTR-amended sand with a mass percentage of 5% WTR relative to sand over carbon material with the depth ratio of 1:3, was selected as optimal. The breakthrough curves correlated well with the Yan model. During the field study, influent and effluent samples were collected from two catch basins from eight storm events and analyzed for turbidity, pH, TPH, and dissolved Cu, Pb, and Zn. The median removal efficiencies of dissolved Cu, dissolved Pb, dissolved Zn, and TPH during the field study were 27.4%, 36.3%, 69.3%, and 45.6%, respectively. The removal of particulate-bound pollutants was indicated by the reduction of turbidity with the median removal efficiencies of 81.2%. A slight increase in pH was observed. There was no water ponding during the study. Our results show that this low-tech, low-cost adsorbent media is effective in reducing metal and organic pollutants in stormwater.

Keywords

Catch basin insert; Drinking-water treatment residuals; adsorbent media; Green technology; stormwater runoff treatment

Abbreviations

1. Introduction

Metal contamination in stormwater runoff is a major concern due to its ubiquity, toxicity, and chemical resistance (Davis et al., 2001). Cu, Pb, and Zn are commonly found heavy metals in relatively high concentrations in stormwater (Kayhanian et al., 2012; Mangani et al., 2005; Maniquiz-Redillas & Kim, 2016). In addition to heavy metals, hydrocarbons are also typically found in stormwater runoff, resulting mainly from road transport activities (Göbel et al., 2007; Kayhanian et al., 2012; LeFevre et al., 2014). The low density of many of the petroleum fractions can pose major short-term problems in aquatic ecosystems, particularly for wildlife and fish (Todd et al., 1999). Stormwater characteristics are highly variable (Göbel et al., 2007). Types of pollutants in stormwater vary widely, similar pollutants result from similar land use. However, concentrations may vary and are affected by rainfall conditions (Maniquiz-Redillas et al., 2013).

Stormwater pollutants generally occur in particulate and/or dissolved phases. Generally, stormwater pollutants are reported to be present mainly in the particulate phase. However, this is true only downstream of the storm sewer system, not upstream (Bressy et al., 2012). The particulate phase has been the focus of stormwater treatment, while the dissolved phase is generally overlooked. Due to their high bioavailability, dissolved pollutants can quickly impact receiving water bodies and their biota (LeFevre et al., 2014).

Many conventional stormwater best management practices (BMPs), grey infrastructures including catch basins, are capable of capturing coarse suspended solids. However, their capability to remove dissolved pollutants is limited (LeFevre et al., 2014). Grey infrastructures are necessary for mitigating the peak flow. Enhancing the performance of existing grey infrastructures to address the dissolved pollutants would be a promising approach to address

stormwater-related pollution of receiving waters. Moreover, using the existing catch basins would be a cost-effective way to mitigate stormwater pollution.

A catch basin insert (CBI) is a device installed in an existing catch basin to remove pollutants from stormwater runoff. The removal mechanism principally involves screening, sedimentation, and adsorption as the water passes through the CBI (Edwards et al., 2004). The effectiveness of CBIs in capturing gross pollutants, i.e. particulate pollutants, has been previously demonstrated (Alam et al., 2018b, 2017). In terms of dissolved pollutants, the performance of CBIs can be enhanced by adding an adsorbent media capable of retaining dissolved metals, and organic pollutants. The advantages of CBIs include their low land requirement and the ease of installation and retrofitting them (Kostarelos and Khan, 2007). However, one of the challenges of implementing catch basin inserts is the cost of adsorbent media used in the catch basin insert.

Aluminum-based drinking water treatment residuals (WTR) is a type of drinking water treatment byproduct generated in drinking water treatment facilities that use aluminum salts, such as alum, aluminum chloride, and polyaluminum chloride, as coagulants (Nair and Ahammed, 2014). More than 2 million tons of WTRs are produced annually in the US alone (Prakash and SenGupta, 2003), while it has been reported that 18,000; 34,000; and 182,000 tons of dry solids are produced annually in Ireland, the Netherlands, and the UK, respectively (Babatunde and Zhao, 2007). WTRs are usually landfilled, but with landfill spaces becoming limited, and increasing disposal cost, reuse of WTRs for other applications is an attractive option. Many studies have demonstrated the effectiveness of drinking water treatment residuals for the removal of a wide range of environmental pollutants: aluminum (Roychowdhury et al., 2019), arsenic (Makris et al., 2006a; Roychowdhury et al., 2019), cadmium (Silvetti et al., 2015), copper (Castaldi et al., 2015), chromium (Zhou and Haynes, 2011), fluoride (Sujana et al., 1998), iron (Roychowdhury et al., 2019), lead (Castaldi et al., 2015; Roychowdhury et al., 2019; Zhou and Haynes, 2011), manganese (Roychowdhury et al., 2019), mercury (Hovsepyan and Bonzongo, 2009), phosphorus (Alam et al., 2020; Babatunde et al., 2008; Ippolito et al., 2003; Lee et al., 2015; Makris et al., 2010, 2004; O'Neill and Davis, 2011; Soleimanifar et al., 2016), perchlorate (Makris et al., 2006b), selenium (Ippolito et al., 2009), sulphate (Roychowdhury et al., 2019), tetracyclines (Punamiya et al., 2013), and zinc (Roychowdhury et al., 2019; Silvetti et al., 2015). Due to their easy availability, low cost of procurement (can be obtained free of charge from drinking water treatment plants), non-hazardous nature, and high metal and oxyanion sorption potential, use of WTRs as an adsorbent media for use in catch basin inserts to retain pollutants from road runoff can be a sustainable approach, which benefits the environment by improving the water quality and reducing the human and ecological risk of exposure to heavy metals, and lowering the demand for landfill space for the disposal of WTRs.

The objective of this study was to develop a novel "green" adsorbent media to remove pollutants from stormwater runoff, that is inexpensive and can be retrofitted into existing grey infrastructures such as the CBIs. An adsorbent media consisting of WTR, sand, and carbon material (CM) was developed and optimized based on hydraulic and treatment performance in laboratory column experiments. Then, the optimal adsorbent media was evaluated under field conditions for 4 months to demonstrate its capacity to improve pH, remove total petroleum hydrocarbons (TPH), turbidity, and dissolved metals Cu, Pb, and Zn from stormwater runoff.

2. Materials and Methods

2.1. Materials

 All chemicals used in this study were reagent grade or above. Deionized water was used for preparing all solutions and dilutions for treatment performance evaluation. Stock solutions of Cu, Pb, and Zn (5000 mg/L) were prepared using their respective nitrate salts: $Cu(NO₃)₂•2.5H₂O$ (Acros organic, NJ), Pb(NO₃)₂ (Analytical Scientific, TX), and Zn(NO3)2•6H2O (Fisher Scientific, NJ). Stock solutions were diluted to produce synthetic stormwater runoff. The gasoline and diesel fuels used in this experiment were commercially available and were obtained from gas stations. Three sorbents, namely WTR, CM, and sand were used in the development of optimal adsorbent media. WTR was collected from New Jersey American Water (NJAW) Water Treatment Plant in Delran, NJ. It was air-dried and ground to pass a 2 mm sieve. The sand (fine sand, Sakrete® Natural Play Sand) was purchased from a commercial outfit; CM (granular non-activated carbon with an approximate particle size of 2 - 3 mm) was obtained in bulk from the Hongtai Water Treatment Plant in China.

2.2. Hydraulic Performance

 WTR is relatively impermeable. Its hydraulic conductivity was approximately 10- 10 m/s (Balkaya, 2015). Therefore, improving permeability is necessary for using WTR as an adsorbent media component. One simple and economical approach is mixing the WTR with coarse materials to improve permeability. Two designs of adsorbent media with the primary difference in terms of layer (LY) were investigated. The two designs are (1) mono-layer (mixed), and (2) dual-layer (bedded) media. Five adsorbent media types from these two designs were compared:

Mono-layer (mixed)

- Sand amended with WTR:
- CM amended with WTR;
- CM amended with sand and WTR:

Dual-layer (bedded)

- 2.5 cm of sand amended with WTR over a 7.5 cm layer of CM (depth ratio of 1:3);
- 5 cm of sand amended with WTR over a 5 cm layer of CM (depth ratio of 1:1)

Sand and CM were selected since they are commonly available coarse materials and have been long used in water treatment. In addition, CM also has an intrinsic property to remove hydrocarbons (Bansal and Goyal, 2005) which are common pollutants in stormwater runoff. Conventionally, dual-layer media are used as a progressive sieve where larger particles are trapped within the coarser materials on the top layer and smaller particles are trapped within the finer materials at the bottom layer (Zouboulis et al., 2007). This conventional design is beneficial for maximizing the solid holding capacity of the adsorbent media and is widely used to treat water with a high concentration of suspended solids (Bablon et al., 1988). In contrast, the focus of this study was to improve the hydraulic conductivity and enhance dissolved pollutant removal performance of the adsorbent media. Therefore, the coarse layer (CM) was placed at the bottom of the finer layer, WTR-amended-sand. In addition to enhancing hydraulic conductivity, having WTR-amended-sand as a top layer can help to lengthen the life cycle of the CM (Li et al., 2018).

For each adsorbent media type, WTR was mixed with the base media at four mass percentage of WTR relative to based media (0%, 5%, 10%, 20%). Hydraulic performance of the adsorbent media combinations was evaluated in clear polyvinyl chloride (PVC) columns with an inner diameter of 2.54 cm and a length of 30 cm equipped with a 2.54-cm PVC male adapter fitting at both ends of the PVC columns. At the bottom end of each column, a 2.54-cm PVC center-drilled dome cap equipped with a 3.18 mm x 3.18 mm adapter and 3.18 mm tubing was installed as an outlet (Figure 1). To retain the adsorbent media within the column and mimic conditions of actual CBI, a circular layer of the CBI material, non-woven polypropylene geotextile (1200FF, NDS Inc., California), and glass wool were installed at the bottom of each column. The total depth of all adsorption beds, irrespective of mono and dual-layer media was 10 cm. During column packing, adsorbent media was uniformly spread across the cross-section every 1 cm and evenly compacted. This step was repeated until the designed depth was reached.

Initially, clean water was introduced gradually from the bottom of the columns, and the columns were gently tapped to eliminate air bubbles in the packed bed. Then, clean water was supplied continuously from the top of the column at a rate that allowed it to maintain a 10 cm water level above the bed. The effluent volume was monitored continuously at 60 seconds interval. The saturated flow condition was recorded when three consecutive readings were constant. The total duration to achieve the saturated flow condition was approximately 15 minutes. The experiment was done in duplicate columns for each treatment.

Subsequently, the hydraulic conductivity was calculated from the following equation (ASTM, 2007):

$$
k = \frac{QL}{Ath} \tag{1}
$$

where k = hydraulic conductivity (cm/sec); Q = volume of flow (mL); L = length of media along the path of flow (cm); $A = cross-sectional area of specimen (cm²)$; t = interval of time over which the flow Q occured (sec); $h =$ difference in hydraulic head across the media (cm).

Columns were designed from PVC pipers, with a center-drilled dome cap equipped with a 3.18 mm x 3.18 mm adapter. The columns were filled with 10 cm of various adsorbent combinations. Non-woven polypropylene geotextile and glass wool were used to plug the columns.

2.3. Treatment Performance

 Based on the results of the hydraulic performance study, a dual-layer media [2.5 cm WTR-amended sand over 7.5 cm layer of CM (2-LY)] was selected for testing the metal removal performance of the sorbent media. The performance of the dual-layer media was compared to that of the mono-layer media [WTR-amended sand (1-LY)]. The sand-based mono-layer media was selected for comparison since it is the simplest and the conventional form of sorbent media. Both 1-LY and 2-LY were evaluated under four mass percentages of WTR relative to the base media (0%, 5%, 10%, 20%) where the mass percentage of 0% was used as a control. Synthetic stormwater runoff with an exaggerated concentration of Cu, Pb, and Zn (6.36, 8.16, and 11.70 mg/L, respectively) was prepared by diluting stock solutions of each metal with deionized water. These high concentrations were a 100-fold exaggeration of the field conditions that were measured previously by our research group (data not shown).

During the experiment, the synthetic stormwater was supplied at a flow rate of 8 ml/min continuously, using a peristaltic pump (Ismatec Reglo Digital, Cole-Parmer, IL). The synthetic stormwater was supplied from the top of the columns to mimic the actual flow direction in catch basins. Representative samples of the effluent were collected every 5 minutes for the first 20 minutes. Then, the remaining samples were collected at a geometric progression. The collected samples were filtered through a 0.45-µm nylon syringe filter and analyzed using an inductively coupled plasma–optical emission spectrometer (ICP-OES, 5100 Agilent Technologies, CA) for concentrations of Cu, Pb, and Zn. The experiment was performed in duplicate columns, and representative samples were collected in duplicates at each sampling time.

The removal performance of each adsorbent material was evaluated based on the mass of the pollutants removed at the final bed volume of the respective pollutants. The cumulative mass of metal removed, q (mg) was calculated as follows:

$$
q = \frac{Q}{10^6} \int_{t=0}^{t=t_{total}} (C_0 - C_t) dt
$$
 (2)

where Q is the volumetric flow rate (mL/min), t_{total} is the total flow time (min), C_0 and C_t are the influent and effluent concentration $(\mu g/L)$. The integration of equation 2 for the calculation of the removed pollutant was performed using the trapezoidal rule. Subsequently, the percentage of the cumulative mass of metal removed was determined by dividing q with the cumulative mass of metal inlet.

To determine the metal removal contribution of the WTR, the removal contribution of sand and CM was first calculated. The specific removal contribution of sand was determined from 1-LY control which was calculated by dividing the mass of the pollutants removed at the final bed volume by the mass of sand. Briefly, the mass of the pollutants removed contributed by sand was determined by multiplying the mass of sand by its specific removal contribution and subtracted from the total mass of the metal removed. Similarly, the specific metal removal contribution of CM was determined from 2-LY control. After subtracting the mass removal attributable to sand, the remaining mass of the metal removed was considered as contributed by CM. Subsequently, the specific removal contribution of CM was calculated by dividing the remaining removed mass of the pollutants by the mass of CM. The specific metal removal contribution of WTR was determined from all three mass percentages of WTR (5%, 10%, 20%) from both 1-LY and 2-LY, after subtracting the removal contribution of sand and CM based on their corresponding masses and their calculated specific metal removal capacities. The specific

metal removal contribution of WTR was the average of the removal contribution determined from all six treatments with the presence of WTR (1-LY 5%, 1-LY 10%, 1-LY 20%, 2-LY 5%, 2-LY 10%, 2-LY 20%).

2.4. Modeling of fixed-bed adsorption

Prediction of the adsorption process by the newly developed adsorbent media is fundamental for designing real-life applications. Modeling of fixed-bed adsorption can provide a prediction of adsorption behavior based on the data obtained from the previous fixed-bed adsorption experiment. The fixed-bed adsorption data were fitted with two models, namely the Yan model (Yan et al., 2001), and the Callery model (Callery et al., 2016). The Yan model is also known as a modified dose-response model. It is an empirical model that was developed to predict metal removal with less error compared to other conventional models such as the Thomas or the Bohart–Adams models (Yan et al., 2001). The Callery model was developed based on the form of the Freundlich isotherm to describe the relationship between treated volume and effluent concentration. Callery model showed a high degree of accuracy to predict the performance of WTR fixed-bed adsorption experiments (Callery et al., 2016).

Yan model (Modified dose-response model)

$$
\frac{C_t}{C_0} = 1 - \frac{1}{1 + \left(\frac{V_t}{B}\right)^A}
$$
(3)

$$
\ln\left(\frac{C_t}{C_0 - C_t}\right) = A \log(V_t) - A \log(B)
$$
\n(4)

Callery model

$$
\frac{C_t}{C_0} = 1 - \frac{AMV_t^{\frac{1}{B}-1}}{BC_0}
$$
\n⁽⁵⁾

$$
\ln(C_0 - C_e) = \left(\frac{1}{B} - 1\right) \ln(V_t) + \ln\left(\frac{AM}{B}\right)
$$
 (6)

where V_t is the treated volume of synthetic stormwater runoff (L), A and B in equation 3, and 4 are Yan model constants which indicate the slope of the regression function and the cumulative volume that produces a half-maximum response, respectively. For the Callery model (equations 5 and 6) A is a constant; and B is a dimensionless constant of system heterogeneity, similar to n in the Freundlich equation (Callery et al., 2016). M is the total mass of the adsorbent media. All model parameters were obtained by conducting linear regressions based on their linear forms (equations 4 and 6). The correlation of determination (R^2) was calculated accordingly along with root mean sum-of-squares error (RMSE). They are used in this study as metrics for the comparison of the goodness of model fit to experimental data.

$$
RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left(\left(\frac{C_t}{C_0} \right)_{cal} - \left(\frac{C_t}{C_0} \right)_{exp} \right)^2}
$$
(7)

2.5. Field study

2.5.1. Study Area

To investigate the performance of the selected adsorbent media under real-life conditions, a field study was performed in a parking lot owned by the Township of Brick Municipal Building, Brick, NJ, USA. Stormwater runoff in this area discharges to Forge Pond which is a

drinking water source in the community. Two catch basins located near high traffic areas having appropriate dimensions for CBI installation were selected for the field study (Figure 2).

Figure 2. Aerial photo of the field site, Township of Brick Municipal Building

Circles indicate selected catch basins for the field study (yellow is inlet A and orange is inlet B); polygons indicate drainage area of the inlet A and inlet B, which were approximately 1,400 m² and 750 m^2 , respectively.

2.5.2 Description of the Apparatus

An apparatus was designed specifically for this study to house CBI adsorbent media, and to collect representative influent and treated runoff samples (influent and effluent) (Figure 3).

The apparatus was built using zinc-plated punched angle steel (Everbilt, GA) and 0.953 cm x 2.54 cm zinc-plated hex bolts and nuts (Everbilt, GA). An adjustable frame model catch basin insert (UltraTech International, Inc, FL) was hung at the center of the structure. The catch basin inserts were loaded with the optimal adsorbent media which was scaled up to 7.62 cm of WTRamended sand over 22.86 cm of CM by maintaining the same mixing and bed depth ratios. On top of the CBI, an impervious tray made of an acrylic sheet with an opening at the center equipped with solid gutter covers on all edges was installed to direct runoff water to the adsorbent media. A 500-ml HDPE cup was installed under the opening hole of the impervious tray to collect influent before draining into the adsorbent media. A 3,786 ml (1 gal) HDPE bucket was installed right underneath the CBI to collect the effluent. Two clear PVC tubes with an internal diameter of 0.795 cm were installed in the apparatus. One was for the influent container while the other one was for the effluent container. One end of each tube was permanently attached to the bottom of the containers. The other end was attached to the structure in an area that was accessible from above the grate and covered to prevent stormwater runoff entering into the tubes. There were two high-flow bypass openings above the top of the adsorbent media to prevent water backing onto the surface.

Figure 3 (a) diagram and (b) photo in the field site of the catch basin insert apparatus.

2.5.3. Sample Collection and Analysis

To collect samples, the tubes were pulled over the grate, rinsed thoroughly with DI water, and connected to a hand pump (Ace, IL). Before collection, water in the influent and effluent containers were pumped out to discard any residues from the earlier storm event to ensure that the samples were fresh and representative of the current storm event. After that, fresh influent and effluent were allowed to accumulate in the influent and effluent containers respectively. The initial volume of the fresh samples collected was discarded to flush the hand pump and tubes before collecting the samples for analysis. In a given storm event, 250 ml of influent and effluent were collected from each catch basin at the earliest possible time. A grab sample of influent and effluent from each inlet was collected during each storm event. Although composite sample or multiple grab samples may be preferable for evaluating the overall mass removal during a storm event, collecting a grab sample of influent and effluent is also useful for comparing (near)

simultaneous influent and effluent concentrations which were subsequently used to calculate instantaneous removal. By comparing the results from multiple storm events, the overall effectiveness of the media in the field was evaluated.

The samples were analyzed in the field for pH using a pH/ Conductivity meter (PC700, Oakton, IL) and turbidity using a portable turbidimeter (TB200, Orbeco, FL). For metal analysis, representative samples of influent and effluent were prepared in triplicates by passing through 0.45-µm nylon syringe filters and subsequently analyzed for dissolved concentrations of Cu, Pb, and Zn using an inductively coupled plasma – optical emission spectrometer (ICP-OES, 5100 Agilent Technologies, CA).

For TPH analysis, the samples were extracted according to the Texas Natural Resource Conservation Commission Method 1005 (TNRCC, 2001). Subsequently, the extract was analyzed using gas chromatography according to Reddy & Quinn (1999) with the inlet temperature of 300 °C; the oven was programmed to start at 70 °C for 1.5 minutes, ramped to 300 °C at 15 °C per minute and then held for 10 minutes. A Hewlett-Packard 5890 Series II Gas Chromatograph equipped with a Hewlett-Packard 5971 Mass Selective Detector (GC-MSD) was used. Helium was used as carrier gas. After splitless injection $(1 \mu L)$, compounds were separated by a ZB-5MS fused silica capillary column (30 m length, 0.25 mm i.d., and 0.25 µm film thickness). All analyses were conducted in duplicates. For quantification, calibration standards were prepared with commercial unleaded gasoline and diesel #2 (1:1) in n-pentane. The solvent delay was 3 minutes. Data were collected with Hewlett-Packard Chemstation software. The TPH concentration was determined by integrating the extracted-ion-current profile (EICP) of m/z 57 (C4H⁹ +), which is a major ion in aliphatic compounds (Reddy and Quinn, 1999) from 3 to 24 minutes. A retention time marker that contained n-hexane (C6), n-dodecane (C12), n-octacosane

(C28), n-pentatriacontane (C35) in n-pentane (Restek, PA) was used to determine the time for integration. The starting time of this time window was determined based on the earliest time that MSD can operate without deterring the MSD from detecting high intensity of ions from the solvent, while 24 minutes was the time that the heaviest compound, n-pentatriacontane (C35), was detected.

 All sampling containers were cleaned with laboratory-grade detergent, soaked in 10% nitric acid solution overnight, soaked in DI water, and rinsed with DI water. Every time before collecting samples, the tubes were rinsed thoroughly with DI water. Then, the pump was flushed with a large amount of DI water (~500 mL) to avoid cross-contamination. The pump and tubing were flushed with the actual samples before collection. All discarded water after rinsing and flushing was directed to other areas that did not flow into the inlets to avoid any crosscontamination or unintended dilution. Certified reference solutions were analyzed along with the samples. Low concentrations in all samples (<12 μ g/L for Cu, < 2.5 μ g/L for Pb and <40 μ g/L for Zn) is evidence that no contamination of samples occurred.

3. Results and Discussion

3.1. Hydraulic Performance

The results of the hydraulic performance experiment are presented in Figure 4. For statistical analysis, the Tukey-Kramer HSD test was performed, using JMP Pro 12.2.0. Within the same mass ratio, the difference in the hydraulic conductivity (K) of each base media was statistically significant ($p \le 0.05$) for almost all pairs of base media, except between sand-based and sand-CM-based media with the WTR mixing ratio of 5%, 10%, and 20%. Although the highest K was found in CM-based mono-layer media, sand-CM-based mono-layer media had

much lower K, comparable to the sand-based mono-layer media. This may be because the void spaces between CM were occupied by sand.

The K of all five types of adsorbent media decreased linearly with the proportion of WTR in the adsorbent media. This decrease indicated relatively high impermeability of WTR compared to sand and CM. K of CM-based mono-layer decreased at a higher rate with increasing WTR compared to other media.

From a practical perspective, it is beneficial for an adsorbent media to have a high hydraulic conductivity as this provides greater treatment capacity for stormwater runoff. However, a high flow rate may contribute to the loss of WTR particles, which may negatively affect stormwater treatment. Therefore, for the dual-layer media, 2.5 cm of WTR-amended sand over 7.5 cm of CM was considered the most suitable option for further studies.

Figure 4. Hydraulic conductivity of the five different adsorbent media at four mass percentage of WTR relative to the base media.

3.2. Treatment Performance during optimization

Higher removal of Cu, Pb, and Zn was observed as the WTR content increased (Figure 5), which was due to increasing sorption sites from Al hydroxide. Similar trends occurred in both configurations of the adsorbent media but were more pronounced in the case of 1-LY since WTR was amended across the entire adsorption bed for the 1-LY while only the top quarter of the adsorption bed was amended in the case of 2-LY (Table 1).

At WTR percentage of 10% or more, the removal performance of the 1-LY media was higher than the 5% WTR in the 2-LY media for all metals. Again, this is likely due to WTR's presence in the entire bed the 1-LY media. The mass composition of the WTR, sand, and CM in the 1-LY and 2-LY media are shown in Table 1.

Mass percentage of WTR relative to sand	Mass(g)					
	1-LY media		2-LY media			
	10 cm		Top 2.5 cm		Bottom 7.5 cm	
	WTR	Sand	WTR	Sand	CM	
0% (Control)	0.00	80.00	0.00	20.00	30.00	
5%	3.81	76.19	0.95	19.05	30.00	
10%	7.27	72.73	1.82	18.18	30.00	
20%	13.33	66.67	3.33	16.67	30.00	

Table 1. Mass composition of WTR, sand, and CM in the 1-LY and 2-LY media.

 (a) (b)

 (e) (f)

Figure 5 Percentage of the cumulative pollutant removed during sorbent optimization. (a) and (b) Cu, (c) and (d) Pb, (e) and (f) Zn; while (a), (c), (e) are sand-based mono-layer media, 1-LY, and (b), (d), (f) are 2.5 cm of WTR-amended sand over 7.5 cm of CM dual-layer media, 2- LY, at mass percentage of WTR relative to sand of 0% (control), 5%, 10%, and 20%.

The Cu, Pb, Zn removal contribution of sand, CM, and WTR are shown in Table 2. The order of metal removal contribution of the adsorbent materials was WTR > CM > sand. The metal removal contribution of WTR was at least three-fold higher than that of CM, and at least a magnitude higher than that of sand. The results emphasized the efficient removal performance of WTR, the recycled material, for use as adsorbent material.

Pollutant	Removal contribution (mg/g)				
	WTR	CМ	Sand		
Cu	7.93	1.18	0.32		
Pb	7.26	2.40	0.62		
Zn	1.80	0.49	0.17		

Table 2. Metal removal contribution of WTR, sand, and CM.

Sand has limited heavy metal removal capability primarily due to its low surface area and few sorption sites (Genç-Fuhrman et al., 2007). The removal of heavy metals by carbon was reported to be the result of a combination of mechanisms such as adsorption, precipitation, hydrogen bonding, and the physical removal of insoluble metal complexes (Ricordel et al., 2001). Electrostatic attractive or repulsive interactions between the metal ionic species in the solution and the carbon-oxygen surface groups on the carbon surface was found to be mainly responsible for the adsorption of cations. In contrast, WTR has an irreversible removal mechanism, mainly through inner-sphere complexation resulting in high sorption capacity and low desorption (Castaldi et al., 2015; Makris et al., 2007; Soleimanifar, 2018).

By comparing the removal of different metals by both types of adsorbent media under all mixing ratios, the adsorption affinity order of the three heavy metals was $Pb > Cu > Zn$. Such adsorption affinity order was consistent with other media such as dairy manure compost (Zhang, 2011), municipal solid waste composts (Paradelo and Barral, 2012), soils treated with sewage sludge supernatant (Gao et al., 1997), pristine rice husks (Alexander et al., 2017), bentonite (Bereket et al., 1997). The removal of Zn decreased substantially after 25 BV, while such a decrease collectively started after 200 BV for Cu and Pb. One factor was likely because of the higher influent concentration of Zn. Low Zn removal could also have occurred because Zn is a

transition metal with low polarizability (Axe and Trivedi, 2002) and relatively low electronegativity compared to those of Cu and Pb.

Higher removal performance is preferable from the treatment perspective. However, from a practical perspective, the main role of catch basins is to drain out the stormwater runoff in the area, which must not be compromised. Therefore, from both treatment and practical perspectives, the adsorbent media which can provide substantial removal performance and, at the same time, has high hydraulic conductivity is considered an optimal option. Based on the obtained result from both hydraulic and removal performance, 2.5 cm of WTR-amended sand at 5% WTR over 7.5 cm of CM dual-layer media was selected for the subsequent field study.

3.3. Modeling of fixed-bed adsorption in dual-layer media

The purpose of this modeling exercise was to identify an appropriate model capable of predicting the effluent concentration under the continuous flow conditions, which can be useful for scaling up. The Callery and Yan models were fitted to the breakthrough curves of the optimal dual-layer at 5% WTR for Cu, Pb, and Zn adsorption as shown in Figure 6. Model parameters determined from linear regression analysis are shown in Table 3. Based on the correlation of determination (R^2) value, the Yan model showed a better fit for all three metals compared to the Callery model. Yan model fitted well with Zn breakthrough curve $(R^2 = 0.946)$, followed by Pb $(R^2 = 0.920)$, and Cu ($R^2 = 0.860$). RMSE values are also in the agreement with the R^2 in terms of the overall goodness of fit.

From the comparison, the two models accurately predicted effluent concentration in different concentration range. To further evaluate the goodness-of-fit of the two models, RMSE values of both models were calculated into two regions of the breakthrough curves: 1) 0.0 < C_t/C_0 < 0.5 and 2) 0.5 < C_t/C_0 < 1.0. For the initial part of the breakthrough curves (0.0 < C_t/C_0 < 0.5), the Yan model had lower RMSE values compared to the Callery model showing better predict the adsorption behavior for all three metals. In the final part of the breakthrough curves $(0.5 \lt C_1/C_0 \lt 1.0)$, the Callery model had lower RMSE values for Cu and Zn, while the Yan model was lower for Pb (Table 3). Based on the results, Yan model had a better prediction of the adsorption behavior of Cu, Pb, and Zn in the media, especially the initial part of the breakthrough curves (0.0 $\leq C_1/C_0 \leq 0.5$), while Callery model could be a good option for predicting the final part of the breakthrough curves ($0.5 \le C_l/C_0 \le 1.0$).

(c)

Figure 6. Breakthrough curves modeling of 2.5 cm of WTR-amended sand at 5% WTR over 7.5 cm of CM dual-layer media for (a) Cu, (b) Pb, and (c) Zn adsorption.

(a)

3.4. Field study

The field study was conducted for 4 months, from August 2017 to November 2017. Samples were collected from 8 storm events on 29 August, 2 September, 6 September, 19 September, 9 October, 24 October, 29 October, and 7 November 2017. The sampling time after the start of each storm event were 4, 6, 5, 1, 2, 5, 5, and 1, respectively. All samples were collected within 1 – 6 hours after the continuous rainfall start which was the recommended range for collecting a single grab sample for oil and grease measurement (Khan et al., 2006). During the study period, no water ponding was observed. Catch basins were able to direct stormwater runoff as normal. Each apparatus had two high-flow bypass openings above the top of the adsorbent media to prevent water backing onto the surface.

3.4.1. Overall Treatment Performance

To visualize the treatment performance, the pollutant concentrations, pH, and turbidity of influents and effluents from all storm events from both locations are presented as box plots in Figure 7. Ranges of the targeted roadway stormwater characteristics in urban areas from other studies are shown in Table 4. Although the concentrations of dissolved metals in this study were slightly lower than the reported ranges, the order of concentrations of the three metals was consistent with the literature which was $Zn > Cu > Pb$. Out of 16 cases (eight storm events and two inlets), the influent concentrations were very low (< 3 µg/L) in all cases for Pb, and 10 cases for Cu. Turbidity was slightly lower than the reported range. The TPH concentrations and pH were well in the reported ranges. In Figure 7, % change was calculated using the following equation:

$$
\% Change = \frac{Median\ effluent\ conc. -Median\ influent\ conc.}{Median\ influent\ conc.} \times 100\tag{8}
$$

Due to the high variation and presence of outliers, the median concentration was used instead of the mean that resulted in overestimated values.

Table 4. Influent Properties

 a Huber et al. (2016)

^b USEPA (2004); criteria are based on total metal concentrations (dissolved and particulate) and hardness of 100 mg/L as CaCO₃.

Figure 7. Box plots for (a) dissolved Cu, (b) dissolved Pb, (c) dissolved Zn, (d) TPH, (e) turbidity, and (f) pH of influent and effluent from both locations during eight storm events; % Change for each parameter was calculated from the median values as shown in equation 8.

3.4.1.1. Metal Removal

For dissolved Cu, reductions were observed for 10 of 16 cases, including all six cases in which influent concentration was $>3 \mu g/L$. The inconsistent removal of dissolved Pb was also observed. Although the influent concentrations of dissolved Zn were relatively higher than dissolved Cu and dissolved Pb, the negative removal, i.e. an increase pollutant concentration as it passes through the adsorbent media, was also observed, particularly at the influent concentration less than 10 µg/L. However, the removal of dissolved Zn was more prevalent (13 of 16 cases) compared to the other two metals.

Overall, a reduction in pollutant levels was observed over the course of the study. Negative removal was observed in a few cases, particularly when the influent concentrations were low (Figure 8). The inconsistency of removal at lower concentrations has been reported in stormwater practices previously (Borne et al., 2013; Thompson et al., 2020). When influent concentrations are so low, it is difficult to achieve further reduction and reliable documentation thereof. BMPs generally have a threshold at which further reduction of concentration may not be achieved. This is referred to as the irreducible pollutant concentration. It is possible to observe negative removal when influent concentration is equal or less than the irreducible concentration (Schueler, 1996). In addition, such low concentrations are well below chronic toxicity thresholds as shown in Table 4. According to the paired t-test ($p \le 0.05$), effluent concentrations of Zn were significantly lower than that of the influent concentrations during the study period. The median removal efficiency of Cu, Pb, and Zn were 27.4%, 36.3%, and 69.3%, respectively.

(c)

Figure 8. (a) dissolved Cu, (b) dissolved Pb, and (c) dissolved Zn of influent and effluent samples from both locations during eight storm events.

3.4.1.2. Hydrocarbon Removal

The removal of TPH was observed for all storm events. TPH concentrations were in the range of 0.4–10.4 mg/L (Figure 7). The median removal efficiency of TPH was 45.6%. The effluent concentrations of TPH were significantly lower ($p \le 0.05$) than those of the influent concentrations based on the paired t-test. The result confirms the capability of CM to remove organic compounds, especially hydrocarbons (Bansal and Goyal, 2005). With the presence of CM, the flow rate through the adsorbent media increased and TPH removal capability was enhanced.

3.4.1.3 Turbidity and pH

The reduction of turbidity was observed for all storm events. The turbidity significantly decreased ($p \le 0.05$) with the median removal efficiencies of 81.2%, indicating the effective removal of the particulate-bound pollutants. The catch basin inserts used in this study was made of geotextile, a proven material used commonly for mitigating particulate pollutants (Alam et al., 2018a). The reduction of turbidity could substantially have been resulted from the use of geotextile. The variation in pH was lower than for other indicators. The influent pH was found to be in the range of pH 5.8–6.7, which is in the acidic range. According to the paired t-test, the increase in pH is statistically significant ($p \le 0.05$). For pH, the median increase of 5.9% was found. The increase of pH is favorable for dissolved metal removal by WTR (Soleimanifar, 2018).

4 Conclusions

A new "green" adsorbent media consisting of WTR, sand, and CM was developed and optimized based on hydraulic and treatment performance in laboratory column experiments, followed by a field study. The hydraulic conductivity of the adsorbent media was affected by design of the filter media (mixed or bedded), type of base media, mixing ratio of Al-WTR, and the bed depth of each layer (for the dual-layer media). The metal removal performance of the adsorbent media increased with an increasing percentage of WTR in the adsorbent media. Metal removal efficacy of WTR was higher than that of sand and CM. Based on the results from both hydraulic and removal performance studies, WTR-amended sand with a mass percentage of 5% WTR relative to sand over carbon material with a depth ratio of 1:3, exhibiting high hydraulic conductivity and substantial removal performance, was selected as the optimal media. The Yan model correlated well with the adsorption behavior of heavy metals by the optimal media. After the laboratory optimization, a 4-month field study was conducted. The removal efficiencies of dissolved Cu, dissolved Pb, dissolved Zn, and TPH from stormwater based on their median concentrations during the field study were 27.4%, 36.3%, 69.3%, and 45.6%, respectively, while the pH increased slightly. The removal of particulate-bound pollutants was indicated by the reduction in turbidity with the median removal efficiency of 81.2%. Reductions in dissolved Zn, TPH, and turbidity, and increase in pH were statistically significant, while reductions in dissolved Cu and Pb were not significant, likely due to their low dissolved concentrations in stormwater runoff. No water ponding was observed during the field study period, demonstrating that the media was able to accommodate high flow rates without flooding. With the wide availability of the WTR, and the ease of preparation by mixing it with common materials like CM and sand, communities can retrofit their catch basins to remove stormwater pollutants easily. As the result, grey infrastructures such as catch basins could become greener with the enhanced stormwater pollutant removal performance from the recycled and common materials inexpensively. The results represent field study data conducted at one site. Further investigations at various geographical regions with varitions in traffic flow (e.g. motorway, main road, service road) would be beneficial to derive site-specific information on the performance of the adsorbent media. In addition, other methods for increasing permeability, such as granulation, could be employed to eliminate the tradeoff between hydraulic conductivity and treatment performance of the adsorbent media.

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