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The Pilot Study of a Dual-Media Filter Consisting of Mortar and Modified Zeolite for Removing Heavy Metals from Expressway Stormwater Runoff

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Abstract: Stormwater runoff from expressways generally has high concentrations of heavy metals. However, the heavy metal removal of conventional sand filters is low, so a better alternative is required. In this study, several inexpensive materials were tested for their heavy metal adsorption, and the performances of the selected materials were evaluated via field tests. The results of laboratory experiments showed that the Cu adsorption capacity followed the order of Na-zeolite > zeolite > biochar > granular ferric hydroxide > sand ≥ orchid stone. The performance of a pilot-scale dual-media filter filled with Na-zeolite and mortar granules was monitored for four rainfall events at an expressway site, and was compared to that of a sand filter. Both filters showed similar event mean concentration (EMC) removal for BOD, COD, TOC, and T-N, without a notable decrease in hydraulic conductivity. However, the removal of T-P, Cu, Zn, Cr, Ni, and Fe by the dual-media filter was 37.6–74.8%, 59.1–90.1%, 84.9–99.7%, 100.0%, 100.0%, and 78.7–94.4%, respectively, which was up to 4.5 times of that of the sand filter. In addition, it was stable regardless of the influent EMCs. Overall, we showed that the dual-media filter is excellent in heavy metal removal from stormwater runoff, with negligible clogging.

Keywords: field test; heavy metals; Na-zeolite; mortar; stormwater runoff

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1. Introduction

Expressway stormwater runoff has a high flow rate and contains significant amounts of non-point source (NPS) pollutants due to its high imperviousness [1]. Expressways are related to a lot of sources of heavy metals in stormwater runoff. These include exhausts, which are rich in Fe, Mg, Ca, Al, and Zn, as well as non-exhaust sources such as the wear of roads, tires, brakes, and vehicles, which are rich in Al, Fe, Ca, Mg, Cr, Cu, Ni, Pb, and Zn [2–5]. This leads to higher concentrations of heavy metals in runoff compared to other NPSs such as farmland, forests, and residential areas [1,6,7]. The concentrations of heavy metals in highway runoff were highly variable and were 400, 4.9, 1900, 9650, 19,100, 21,060, 13,100, and 162,000 μg/L for Cd, Al, Cr, Cu, Ni, Zn, Pb, and Fe, respectively [1,6–10].

Generally, stormwater treatment systems such as granular media filters, infiltration trenches, and wetlands are designed and operated for the removal of suspended solids and the recharge of infiltration to aquifers [11–13]. It has been reported that heavy metal removal via sand filtration and infiltration facilities is highly variable, achieving less than 25–60% removal [13,14]. In addition, it has also been found that the media itself and nearby subsoils are seriously polluted with heavy metals [15]. Therefore, the threat of heavy metals in runoff to the aquatic and soil environment is substantial.

It has been suggested that adsorption using engineered adsorbents is the best alternative for heavy metal removal from stormwater runoff [16]. Therefore, a variety of

adsorbents have been tested for heavy metal removal from runoff, including zeolite, granular ferric hydroxide (GFH), manganese dioxide, concrete, activated carbon, calcite, iron filings, activated alumina, and sand [3,17–20]. However, most of them have the drawbacks of unstable and poor performances. Therefore, stormwater treatment systems using inexpensive and efficient adsorbents must be developed and verified under field conditions to achieve proper heavy metal removal from stormwater runoff.

In this study, six (6) easily available and affordable materials (sand, GFH, biochar, zeolite, a zeolite modified by Na-exchange (Na-zeolite), and orchid stone) were tested for their ability to remove heavy metals. Sand is the most widely used filter media for stormwater runoff filtration, so it was used as a reference [13]. Metals can be adsorbed onto GFH through the inner-sphere complexation with the binding of Fe–O–Fe and Fe–OH [21], and has shown a good performance in terms of Cu and Zn adsorption in column experiments [18]. Biochars and modified biochars can adsorb metals via electrostatic attraction, precipitation, reduction, ion exchange, and complexation [22] and were effective in the adsorption of As, Cr, Cd, Cu, and Pb [23,24], while orchid stone is pumice rock with abundant macropores and a high hydrophilicity [25]. It has been reported that zeolite is a good adsorbent of metals through electrostatic attraction and ion exchange [26], and that it was superior to sand and iron filings in the removal of Cd, Cu, Pb, Ni, Cr, and Zn [19]. In addition, the cation adsorption capacity of zeolite can be significantly improved via Na-exchange, i.e., Na-zeolite, where the doped Na serves as an efficient exchange site with other cations [26–29]. K-exchanged zeolite was also considered, but it was excluded in this study, because the K would strongly compete with the heavy metal ions, resulting in a reduced adsorption [30]. In addition, the bonding of Na is weaker, so it can be replaced by cations other than K, Ca, and Mg. Sodium has a higher mobility, weaker bonding, and stronger ability to replace disabled cations than potassium, calcium, and magnesium cations [26].

The most effective adsorbent was chosen to be the Na-zeolite via the laboratory experiments in this study. It should be noted that zeolites have a better metal adsorption capacity and can be prepared in a laboratory via hydrothermal methods [31] or from agricultural waste, i.e., rice straw [32]. However, different from the adsorbents used in water and wastewater treatment, it is regarded that cheap and readily available adsorbent materials are better for stormwater treatment devices. This is because they are rarely regenerated, have difficulty in maintenance, the treated water quality is not strongly regulated [33], and the heavy metal concentrations in runoffs are lower than those in industrial discharges [1,6,7,10]. In addition, the materials in stormwater treatment systems must have a sufficient hydraulic conductivity and storage volume to receive a variable flow of runoff, long-term stability, and ease of installation [34].

In addition to the selected Na-zeolite, mortar granules were also used in the pilot-scale filtration facility for field tests. As reported in previous studies [35,36], they can act as a supplier of alkalinity, which helps to quickly remove dissolved metals in runoff by promoting their precipitation. Therefore, we anticipated that the use of a mortar layer would decrease the heavy metal loading onto the Na-Zeolite adsorbent layer. To the best of our knowledge, this is the first pilot-scale field study using a filter comprising mortar and Na-zeolite, though research has been conducted for a swale [37], coagulation and filtration systems [38], wetlands [39–42], bioretention [43], a biochar and woodchip bioreactor [44], a filter of granular activated carbon, anthracite, and sand [45], and a sand filter [46].

2. Experimental Materials and Methods

2.1. Media

Sand, which is commonly used in water purification plants and non-point pollution reduction facilities, was purchased from Jumunjin Silica Sand Co., Ltd. (Gangneung, Republic of Korea), and the GFH was a product of Wasserchemie (Mozartstraße, Germany). Biochar, natural zeolite (3–5 mm), and orchid stone (2–4 mm) were obtained from Vermont Biochar (West Danville, VT, USA), Lexem Co., Ltd. (Pohang, Republic of Korea), and

HyugaSoil Co., Ltd. (Miyajaki, Japan), respectively. All of these were used as received. The Na-zeolite was prepared via the impregnation of 5 g of zeolite into 1 L of a 1 M NaCl aqueous solution for 48 h. The mixture was separated, washed with DDW three (3) times, and then dried at 105–110 °C for 24 h [47] (Figure 1).

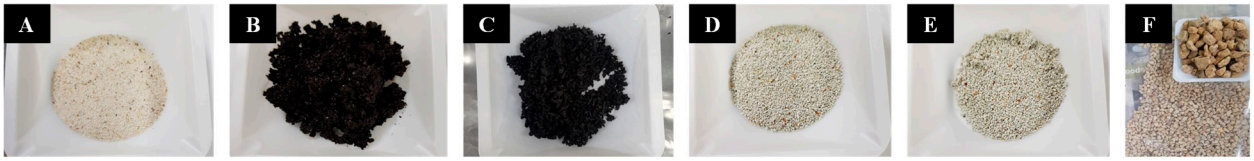

Figure 1. (**A**) Sand, (**B**) GFH, (**C**) biochar, (**D**) zeolite, (**E**) Na- zeolite, and (**F**) orchid stone.

The mortar media were prepared by mixing sand, cement, and water at a weight ratio of 5:0.5:1, followed by curing the mixture for 7 days indoors. The irregular mortar granules were crushed, and those with sizes of 3–5 mm were separated by sieving (Figure 2).

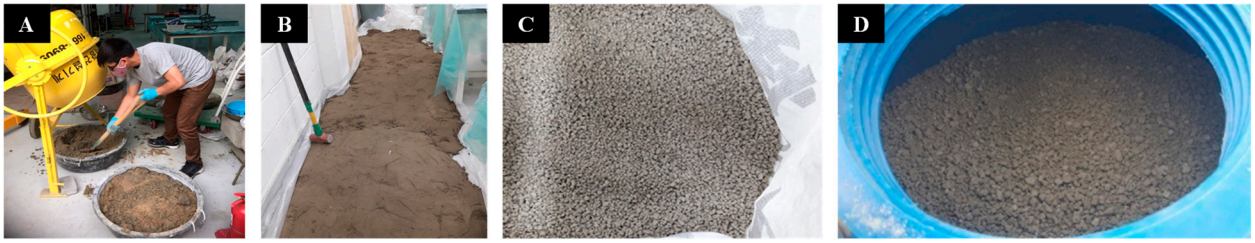

Figure 2. Preparation of mortar: (**A**) mixing, (**B**) curing, (**C**) crushed mortar granule, and (**D**) the mortar layer in the pilot plant.

2.2. Selection of Adsorbent

The sand, GFH, biochar, zeolite, orchid stone, and Na-zeolite were tested for their heavy metal adsorption to select the best medium through equilibrium adsorption capacity and adsorption rate measurements. Copper (Cu) was selected as a representative heavy metal in stormwater runoff, because Cu is commonly found at high concentrations in stormwater runoff [1,6–10] and can be an indicator that the runoff is affected by anthropogenic sources [48].

2.2.1. Equilibrium Adsorption Capacity of Filter Media

Cu solutions of various concentrations were prepared using distilled, deionized water (DDW) and $CuSO_4$, and the pH was adjusted to 5.0 using 0.1 N HCl and 0.1 N NaOH. Then, 40 mL of aqueous solution and 0.1 g of the adsorbent were mixed and stirred at 150 rpm for 16 h using a horizontal stirrer at room temperature (20 °C) to reach adsorption equilibrium. The mixture was filtered through a 0.45 µm PVDF filter, and the Cu concentration of the filtrate was analyzed using inductively coupled plasma atomic emission spectroscopy (ICP, OPTIMA 5300 DV, Perkim-Elmer, Waltham, MA, USA). The results were analyzed using Langmuir and Freundlich adsorption isotherm models (Equations (1) and (2)) [49,50].

$$q_e = q_{max} \frac{K_L C_e}{1 + K_L C_e} \quad (1)$$

$$q_e = K_F {C_e}^{1/n} \quad (2)$$

Here, q_e is the equilibrium adsorption amount (mg/g) and C_e is the equilibrium adsorbate concentration (mg/L). q_{max} and K_L are the maximum adsorption amount of the Langmuir isotherm (mg/g) and the Langmuir adsorption constant, respectively. K_F ((mg/g) $(L/g)^{1/n}$) and $1/n$ are the Freundlich model constants.

2.2.2. Adsorption Kinetics

To evaluate the adsorption rate, 0.1–10 g of the adsorbent was introduced into 50 mL of a Cu aqueous solution, and the mixture was continuously stirred at room temperature at 150 rpm. Samples were taken at predetermined times and filtered through a 0.45 µm PVDF filter. The Cu concentration in the filtrate was analyzed using the ICP. The adsorption amount and adsorption rate constants were calculated using the pseudo-first- and pseudo-second-order models (Equations (3) and (4)) [50].

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \tag{3}$$

$$\frac{dq_t}{dt} = k_1(q_e - q_t)^2. \tag{4}$$

where q_t (mg/g) is the adsorption amount at time t (min), q_e (mg/g) is the equilibrium adsorption amount, and k_1 (min^{-1}) and k_2 (L/mg·min) are the pseudo-first-order and pseudo-second-order adsorption rate constants, respectively.

2.3. Field Tests

2.3.1. Pilot Scale Plant

A couple of cylindrical fiber-reinforced plastic tanks of Φ840 × H925 mm were used as a pilot-scale filtration plant (Figure 3A,B). A tank was filled with 350, 50, 250, and 100 mm of mortar, gravel, NaCl-zeolite, and gravel (5–10 mm) from the top to bottom, respectively (mortar/NaCl-zeolite filter). Another tank was filled with 600 mm of sand (3–5 mm) and 100 mm of gravel (5–10 mm) at the bottom to support the sand layer (sand filter). The total depth of the media in both tanks was determined according to the recommendations of the Ministry of Environment for device-type granular filtration [51]. Stormwater was collected in a stormwater runoff drain at an expressway in the Republic of Korea (Figure 3C). A collection tank was installed in the drain and the runoff was distributed to the filters using PVC pipes.

2.3.2. Monitoring

Monitoring was performed for a total of four (4) rainfall events on 3 September, 5 October, 18 October, and 26 October 2018, with the antecedent dry season days of 4, 3, 7, and 3 days, respectively, and daily precipitation of 17.7, 27.2, 3.8, and 7.2 mm, respectively. Samples for a water quality analysis of the influents and effluents of both filters were collected at 5 min intervals for the first 15 min after the start of the runoff, at 15 min intervals from 15 to 60 min, and then at 30–60 min intervals thereafter, until the end of the runoff. The flowrate was also measured for each sample collection via the frequency method to calculate the event mean concentration (EMC) and the loading of contaminants. The EMC was used to evaluate the overall performance, because it is event-specific and the time variations of the concentration of each pollutant were considered [52]. The pH was measured on-site (LabQuest® 2, Vernier, Beaverton, OR, USA) and the samples were transported to the laboratory for immediate analysis. For each sample, the BOD, COD, TOC, TN, TP, SS, Cu, Zn, Pb, Cr, Ni, Fe, and conductivity were measured, following the Standard Methods for Water Pollution [53]. The EMC was calculated according to Equation (5) [54].

$$\text{EMC (mg/L)} = \frac{\text{Discharged mass during storm event}}{\text{Discharged volume}} = \frac{\int_0^T C(t)\cdot q_{run}(t)\,dt}{\int_0^T q_{run}(t)\,dt} \tag{5}$$

where $C(t)$ and q_{run} (t) are the pollutant concentration (mg/L) and runoff flowrate (L/h) at the rainfall duration t (h).

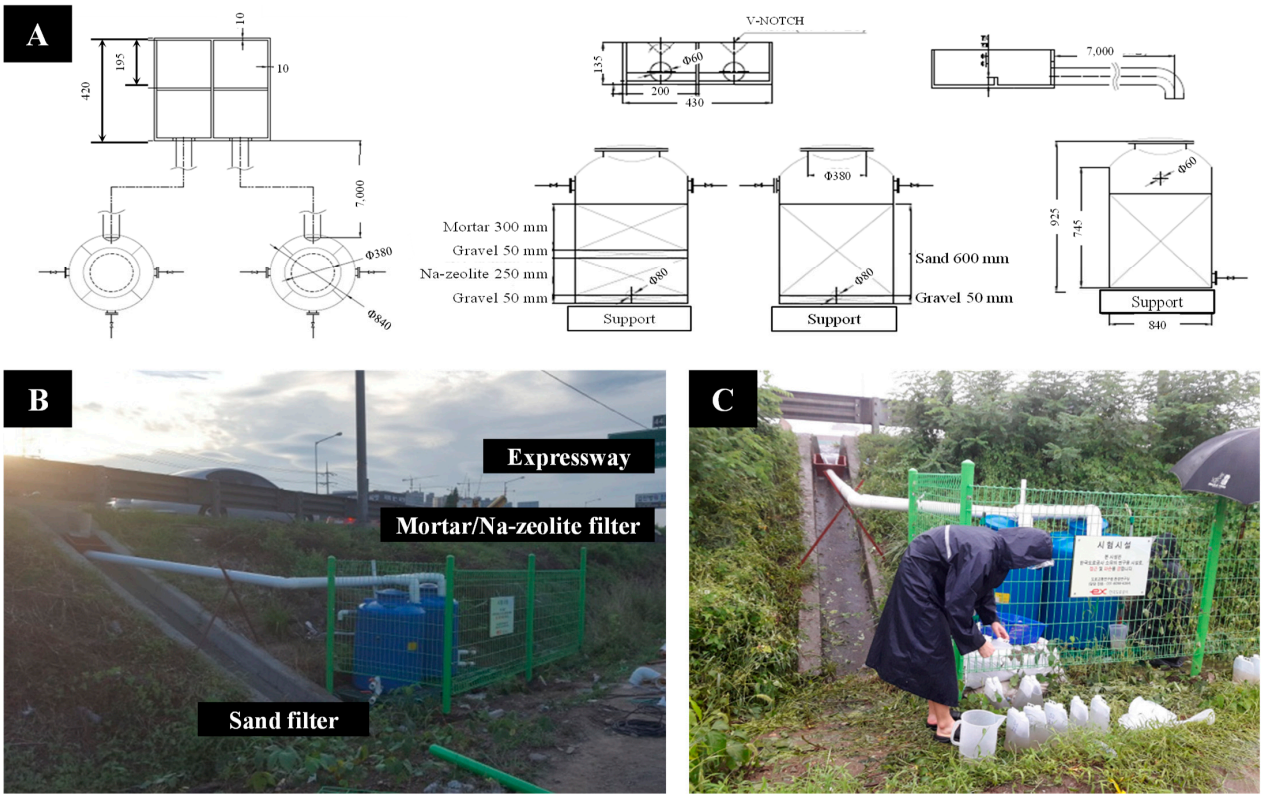

Figure 3. (**A**) Basic drawing of pilot plant, (**B**) installed pilot plant, and (**C**) the monitoring of the influents and effluents of the pilot plant.

2.3.3. In Situ Evaluation of Hydraulic Conductivity

Hydraulic conductivity is one of the most important characteristics of a stormwater treatment filter. A decrease in hydraulic conductivity, i.e., clogging of the media layers, may significantly limit the performance via decreasing the treatment capacity. This can be caused by the accumulation of solids, the development of biofilm, and the deposition of chemical precipitates such as metal (hydr)oxides and calcium carbonate [55,56]. Clogging by metal (hydr)oxides can be dominant in filtration systems treating runoff from expressways with high metal contents, as in this study [57]. Therefore, the hydraulic conductivity was measured for both filters.

Piezometers were installed at the upper inlet and lower outlet of the sand filter and the mortar/NaCl-zeolite filter. The hydraulic conductivity coefficient (k, m/h) was calculated following Equation (6), with the measured difference of the water level between the inlet and outlet and the measured outflow.

$$Q = k\frac{h}{L}At \quad (6)$$

where Q is the outlet flow rate (m^3/h) at time t (h), h is the water head difference between the inlet and the outlet (cm), L is the depth of the filter layer (m), and A is the cross-sectional area of the filter layer (m^2).

3. Results and Discussion

3.1. Cu Adsorption on Adsorbents

3.1.1. Equilibrium Adsorption

The results of the Cu adsorption isotherm using the six (6) materials are given in Figure S1, and Table 1 shows the parameters of the adsorption isotherm models for each adsorbent. The results showed better fits to the Langmuir isotherm than the Freundlich isotherm for sand, zeolite, and Na-zeolite, indicating a homogeneous and monolayer

adsorption of Cu onto them. On the other hand, the Freundlich isotherm provided a better fit for GFH, biochar, and orchid stone, suggesting that the Cu adsorption onto them was more reversible and heterogeneous than that onto the others [49].

Table 1. Cu adsorption isotherm constants of each adsorbent.

Parameters		Sand	GFH	Biochar	Zeolite	Na-Zeolite	Orchid Stone
Langmuir	$q_{max,L}$ (mg/g)	1.29	3.54	8.44	14.85	17.75	1.14
	K_L (L/mg)	0.017	2.727	0.061	1.034	0.002	0.067
	r^2	0.978	0.942	0.961	0.985	0.997	0.917
Freundlich	$1/n$	0.332	0.166	0.199	0.199	0.175	0.185
	K_F (mg/g·(mg/L)^n)	0.157	1.872	2.576	6.285	8.023	0.378
	r^2	0.958	0.963	0.997	0.931	0.957	0.981

The Cu adsorption capacity calculated by the Langmuir isotherm ($q_{max,L}$) was in the order of Na-zeolite > zeolite > biochar > GFH > sand $\geq$ orchid stone, while K_L, which was inversely proportional to the affinity of the adsorption reaction, was in the order of GFH > zeolite > orchid stone $\geq$ biochar > sand > Na-zeolite, representing the best affinity of Na-zeolite to the Cu adsorption.

3.1.2. Adsorption Kinetics

The results of the Cu adsorption kinetics using the six (6) materials are given in Figure S2, and Table 2 shows the constants of the adsorption rate models. For most of the adsorbents, the Cu adsorption rate was better suited to the pseudo-second-order kinetic model than to the pseudo-first-order kinetic model. This means that the Cu adsorption of these adsorbents was more affected by the amount of Cu on the surface of the adsorbents than the concentration of Cu in the aqueous phase [50]. The pseudo-second-order adsorption rate constant (k_2) followed the order of sand >> zeolite > orchid stone $\geq$ biochar $\geq$ GFH >> Na-zeolite, while the equilibrium adsorption amount (q_e) was in the order of Na-zeolite > zeolite > biochar > GFH >> orchid stone > sand.

Table 2. Cu adsorption rate constant of each adsorbent.

Parameters		Sand	GFH	Biochar	Zeolite	Na-Zeolite	Orchid Stone
Pseudo first order	k_1 (min^{-1})	0.193	0.025	0.069	0.187	0.067	0.022
	q_e (mg/g)	1.179	5.785	6.576	14.092	16.530	1.114
	r^2	0.957	0.972	0.944	0.985	0.972	0.976
Pseudo second order	k_2 (g/mg·min)	3.617	0.011	0.013	0.021	0.005	0.015
	q_e (mg/g)	1.260	6.234	7.346	14.894	18.510	1.412
	r^2	0.986	0.996	0.985	0.998	0.996	0.970

Based on the results in Figures S1 and S2, as well as in Tables 1 and 2, Na-zeolite was selected as the medium in the pilot-scale field tests because it had the highest Cu adsorption capacity.

3.2. Field Test

3.2.1. Rainfall Monitoring

The flowrates of the effluents from the sand filter and the mortar/Na-zeolite filter are given in Figure S3. As shown, the flowrates from the filters were similar for all the measurements for all four (4) events, indicating that the influents were evenly distributed to both filters, so the performances of the filters can be compared directly.

The BOD (biochemical oxygen demand), COD (chemical oxygen demand), TOC (total organic carbon), T-N (total nitrogen), T-P (total phosphorus), SS (suspended solids), electric conductivity, Cu, Zn, Cr, Ni, and Fe at each rainfall event are presented in Figures S4–S16. The concentrations of those in the influents were highly variable, but these variations were decreased for the effluents, indicating a reduction in the pollutants in the filters. For the BOD, COD, TOC, T-N, and SS, the profiles of the effluents from the mortar/Na-zeolite filter were similar to those of the sand filter (Figures S4–S7 and S9), suggesting a similar removal of them. However, the time courses of the T-P, conductivity, and metals were significantly different (Figures S8 and S10–S16). The concentrations of them were relatively less variable and the concentrations were lower for the mortar/Na-zeolite filter than for the sand filter, except for the metals which existed mostly at very low concentrations in the influents, i.e., Pb, Cr, and Ni. This suggests that the removal of them was more efficient and stable in the mortar/Na-zeolite filter than in the sand filter.

The EMCs of the influents and effluents, as well as the EMCs' removal, are given in Tables 3–5 and Figures 4 and 5. The EMCs of the BOD, COD, TOC, T-N, T-P, SS, Cu, Zn, Ni, and Fe in the influents were also highly variable, which were 17.8–67.8, 54.2–246.0, 7.9–15.1, 5.4–7.5, 0.31–0.45, 44.1–252.1, 0.041–0.127, 0.119–0.921, 0.006–0.015, ≤0.007, and 0.8–5.5 mg/L, respectively, while the conductivity was in the range of 649–1288 µS/cm.

Table 3. The EMCs of the influents of the pilot plant (mg/L, except for the conductivity (µS/cm)), at each event.

Rainfall Events	BOD	COD	TOC	T-N	T-P	SS	Cu	Zn	Cr	Ni	Fe	Total Metals *	Conductivity
1	28.8	73.9	15.1	5.7	0.45	235.1	0.041	0.129	0.008	0.005	2.361	2.544	854
2	17.8	54.2	10.2	5.4	0.31	44.1	0.065	0.119	0.015	0.004	0.800	1.032	679
3	28.3	81.1	7.9	7.0	0.43	61.0	0.091	0.174	0.006	0.000	1.278	1.548	1288
4	67.8	246.0	15.1	7.5	0.31	252.1	0.127	0.921	0.011	0.007	5.450	6.521	649

Note: * The sum of the concentrations of Cu, Zn, Cr, Ni, and Fe.

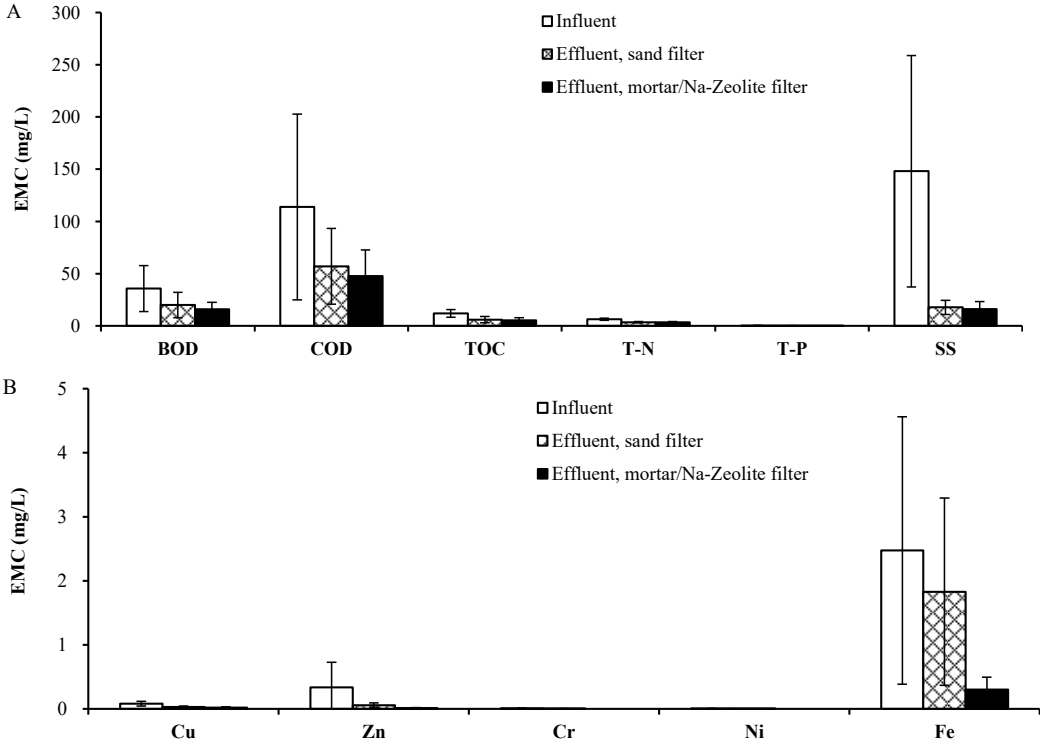

Figure 4. EMCs of the influents and the effluents of the sand filter and mortar/Na-zeolite filter: (**A**) BOD, COD, TOC, T-N, T-P, and SS, and (**B**) metals.

Table 4. The EMCs of the effluents (mg/L, except for the conductivity (μS/cm)), and the EMC removal (%), for the sand filter in each event.

Rainfall Events		BOD	COD	TOC	T-N	T-P	SS	Cu	Zn	Cr	Ni	Fe	Total Metals *	Conductivity
1	Effluent	18.9	47.5	10.0	4.1	0.12	27.3	0.019	0.105	0.000	0.000	1.665	1.789	277
	Removal	34.4	35.7	33.6	29.1	73.5	88.4	54.1	18.7	100.0	93.1	29.5	29.7	67.6
2	Effluent	10.8	30.9	4.2	3.7	0.31	15.3	0.024	0.066	0.000	0.000	0.480	0.571	324
	Removal	39.6	42.9	59.0	32.4	0.00	65.3	62.4	44.2	100.0	100.0	39.9	44.6	52.4
3	Effluent	12.8	39.4	3.4	2.7	0.30	11.6	0.053	0.027	0.003	0.000	1.276	1.359	270
	Removal	54.9	51.4	56.4	61.8	30.7	81.0	41.2	84.4	54.4	100.0	0.1	12.2	79.1
4	Effluent	37.6	110.5	6.4	3.9	0.23	17.3	0.025	0.038	0.003	0.001	3.893	3.963	303
	Removal	44.6	55.1	57.7	48.0	24.7	93.2	80.3	95.8	70.8	83.9	28.6	39.2	53.4

Note: * The sum of the concentrations of Cu, Zn, Cr, Ni, and Fe.

Table 5. The EMCs of the effluents (mg/L, except for the conductivity (μS/cm)), and the EMC removal (%), for the mortar/Na-zeolite filter at each event.

Rainfall Events		BOD	COD	TOC	T-N	T-P	SS	Cu	Zn	Cr	Ni	Fe	Total Metals *	Conductivity
1	Effluent	17.2	38.0	8.9	4.2	0.11	26.7	0.015	0.020	0.000	0.000	0.565	0.600	112
	Removal	39.7	50.0	40.4	24.1	74.8	88.5	63.5	84.9	100.0	100.0	78.7	78.8	86.5
2	Effluent	9.3	29.6	3.2	2.9	0.20	14.1	0.013	0.015	0.000	0.000	0.113	0.141	81
	Removal	47.2	45.2	68.5	44.5	37.6	66.9	80.4	87.2	100.0	100.0	85.5	86.1	87.7
3	Effluent	12.2	38.6	3.6	2.5	0.11	12.7	0.037	0.005	0.000	0.000	0.227	0.270	122
	Removal	58.4	53.6	53.2	64.5	74.6	79.2	59.1	97.0	100.0	100.0	82.7	83.0	90.3
4	Effluent	24.6	84.7	5.5	3.9	0.10	11.6	0.013	0.003	0.000	0.000	0.305	0.320	119
	Removal	64.2	65.7	63.6	49.0	66.9	95.4	90.1	99.7	100.0	100.0	94.4	95.1	80.9

Note: * The sum of the concentrations of Cu, Zn, Cr, Ni, and Fe.

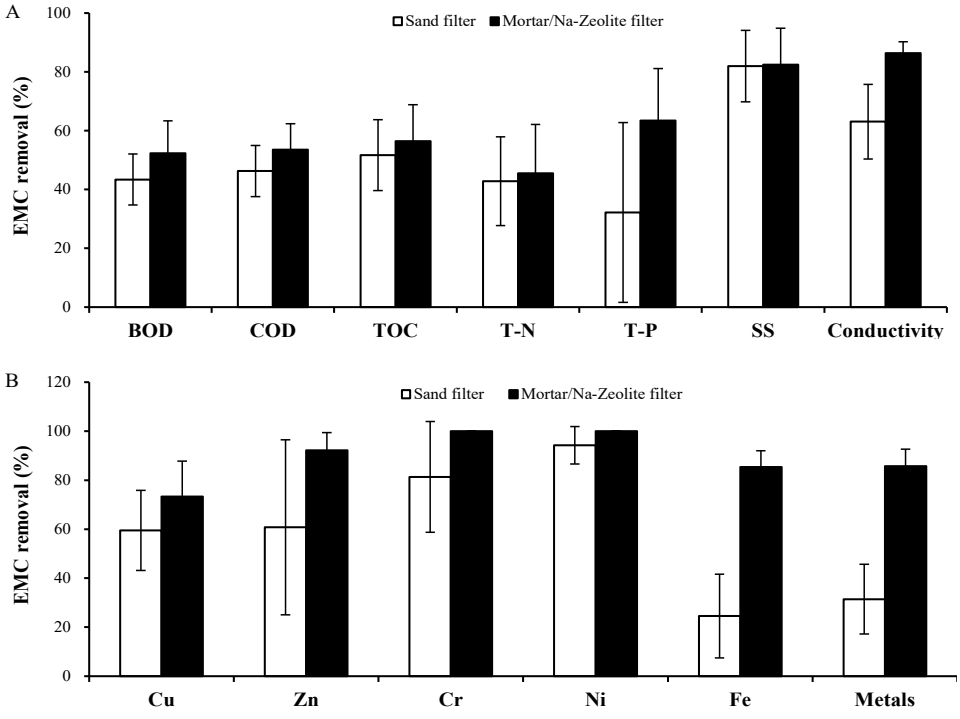

Figure 5. EMC removal by the sand filter and mortar/Na-zeolite filter: (**A**) BOD, COD, TOC, T-N, T-P, and SS, and (**B**) metals.

The EMC removal of the mortar/Na-zeolite filter was in the range of 24.1–95.4% for the BOD, COD, TOC, T-N, and SS, which was 0.8–1.4 times of that of the sand filter, while it was 37.6–100.0% for the T-P, Cu, Zn, Cr, Ni, Fe, and the total heavy metals, i.e., the sum of Cu, Zn, Cr, Ni, and Fe, which was up to 6.8 times more than that in the sand filter. On the other hand, there was a better removal of conductivity in the mortar/Na-zeolite filter (80.9–90.3%) than in the sand filter (52.4–79.1%), which suggests a higher removal of the ionic species, other than the heavy metals.

Meanwhile, these results suggest that the mortar/Na-zeolite filter and the pilot plant could be used for the removal of other metallic cations such as Cs, possibly via more modification of the zeolite [31,32,58], because zeolite is the best known ion exchanger for Cs [58].

The EMC removal of the mortar/Na-zeolite filter was slightly better than the sand filter for the BOD, COD, TOC, and T-N, while that of the SS was not notably different. The removal of T-P in the mortar/Na-zeolite filter was significantly superior to that of the sand filter. Zeolites, either modified or pristine, have a high specific surface area and well-organized pores, which provide more adsorption sites for phosphate via inner-sphere surface complexation and electrostatic attraction [59]. The removal of heavy metals was substantially higher in the mortar/Na-zeolite filter than in the sand filter. An enhancement of the removal was more significant for iron, which existed at higher concentrations in the influent, i.e., Fe (0.800–5.450 mg/L), while it was less significant for Cu, Zn, Cr, and Zn, which had concentrations far less than that of Fe (ND-0.921 mg/L).

The removal of the heavy metals in the mortar/Na-zeolite filter was comparable or superior to that in previous reports based on the results of pilot- or real-scale experiments using real stormwater runoff (Table S1). In particular, it is notable that the excellent removal of Fe, which generally exists at high concentrations in stormwater runoff [1,7], by the filter has been verified in the field, suggesting a great potential for it to be used readily. There are a number of studies reporting good performances in heavy metal removal in laboratory-scale experiments using various materials [3,17–20]. However, there are a limited number of publications of studies using real stormwater runoff; therefore, the use of the materials must be verified more.

It should be noted that the removal of heavy metals in the sand filter decreased as the concentration of the metal in the influent increased, probably because of the low adsorption capacity of the sand. However, the mortar/Na-zeolite filter was not notably affected by the influent concentration, suggesting a stable performance of the mortar/Na-zeolite filter in heavy metal removal (Figure 6).

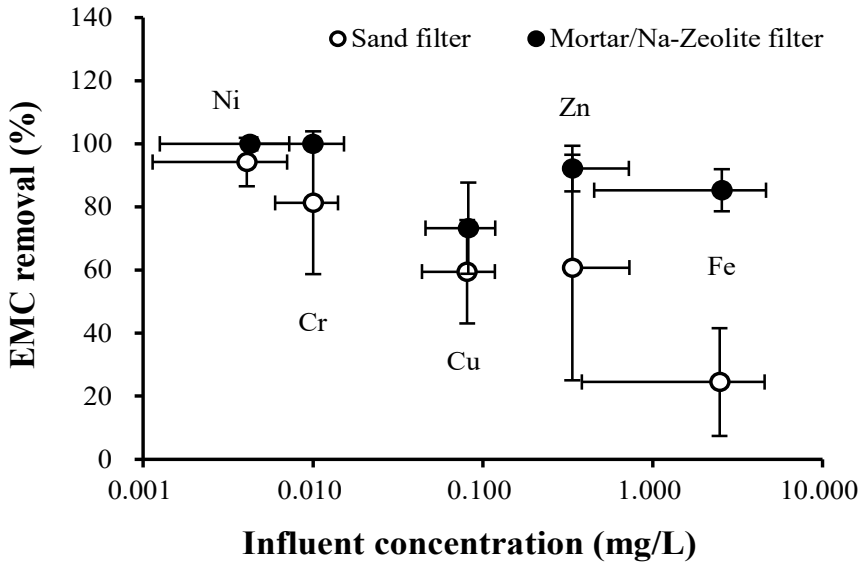

Figure 6. Correlation between heavy metal inflow EMC and removal rate.

The EMC removal of conductivity in the mortar/Na-zeolite filter was significantly higher than that in the sand filter (Tables 4 and 5), indicating that the removal of charged ions other than those analyzed in this study was also promoted by the mortar and Na-zeolite.

3.2.2. In Situ Hydraulic Conductivity Evaluation

Figure 7 shows the change in the hydraulic conductivity of the sand filter and the mortar/Na-zeolite filter for each rainfall event, and Table 6 provides the statistical analysis results for the hydraulic conductivity of each filter in the four (4) rainfall events.

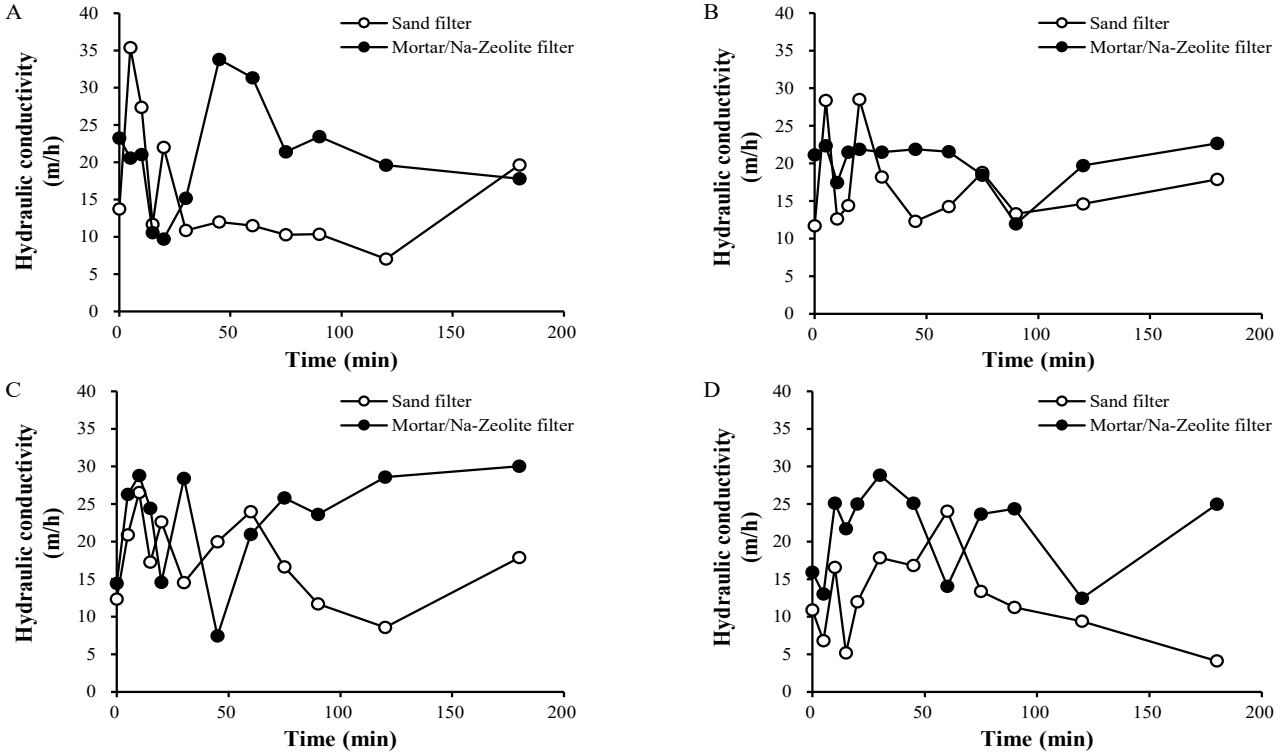

Figure 7. Changes in the hydraulic conductivity coefficient in the sand filter and the mortar/Na-zeolite filter in rainfall events: (**A**) 1, (**B**) 2, (**C**) 3, and (**D**) 4.

Table 6. Hydraulic conductivity of the sand and the mortar/Na-zeolite filters (m/h).

	Sand Filter	Mortar/Na-Zeolite Filter
Average	15.6	21.2
Median	14.3	21.6
Standard Deviation	6.6	5.8
Minimum	4.1	7.4
Maximum	35.4	33.8

The hydraulic conductivity of the sand filter and the mortar/Na-zeolite filter ranged from 4.1 to 35.4 and from 7.4 to 33.8 m/h, respectively, depending on the influent flow rate. Considering that the maximum flow rate was more than 30 m/h in the four (4) rainfall events, no serious blockages occurred in the sand filter or the mortar/Na-zeolite filter. The mortar/Na-zeolite filter consistently showed slightly higher values of hydraulic conductivity than the sand filter. Therefore, it seems reasonable that the possible formation of (hydr)oxides from the metals in the influents [57] in the mortar/Na-zeolite filter did not notably clog the filter.

4. Conclusions

Several adsorbents that are inexpensive and readily obtained were investigated as candidates for the efficient removal of heavy metals from expressway stormwater runoff. The equilibrium Cu adsorption capacity, calculated using a Langmuir isotherm, was in the order of Na-zeolite (17.75 mg/g) > zeolite >> biochar > GFH. Sand, which is commonly used as the filter material in stormwater filters, GFH, and orchid stone showed low Cu adsorptions of 1.14–1.29 mg/g.

In the field evaluation, the heavy metal removal of a dual-media filter, consisting of layers of Na-zeolite (which showed the highest Cu adsorption) and mortar granules, was investigated using pilot-scale plants, installed under the embankment section of an expressway, for four (4) rainfall events. The BOD, COD, TOC, T-N, and SS of the influents were 7.6–108.2, 23.0–318.0, 4.4–20.4, 1.0–12.4, and 6.0–361.0 mg/L, respectively. The average EMC removal percentages of them in the mortar/Na-zeolite filter were slightly higher than those in the sand filter. However, the mortar/Na-zeolite filter was superior to the sand filter for the removal of T-P and heavy metals. The concentrations of T-P, Cu, Zn, Cr, Ni, and Fe in the influents were up to 0.69, 0.280, 1.975, 0.184, 0.056, and 8.9 mg/L, respectively, and the average EMC removal of them in the mortar/Na-zeolite filter was in the range of 63.5–100.0%, which was 1.0–3.5 times that in the sand filter. In addition, the removal of the heavy metals was excellent in the mortar/Na-zeolite filter regardless of the concentration of the metals, while that in the sand filter decreased with increasing concentrations. Clogging of the filters was not significant, as evidenced by the hydraulic conductivity, which was measured in situ.

The results in this study strongly suggest that the mortar/Na-zeolite filter can be an excellent alternative for heavy metal removal from stormwater runoff, with less maintenance due to negligible clogging.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15203560/s1, Figure S1: Cu adsorption isotherm of (A) sand, (B) GFH, (C) biochar, (D) zeolite, (E) Na-zeolite, and (F) orchid stone; Figure S2: Cu adsorption kinetics of (A) sand, (B) GFH, (C) biochar, (D) zeolite, (E) Na-zeolite, and (F) orchid stone; Figure S3: The flowrate in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S4: The BOD in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S5: The COD in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S6: The TOC in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S7: The T-N in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S8: The T-P in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S9: The SS in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S10: The conductivity in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S11: The Cu in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S12: The Pb in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S13: The Zn in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S14: The Cr in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S15: The Ni in the event (A) 1, (B) 2, (C) 3, and (D) 4; Figure S16: The Fe in the event (A) 1, (B) 2, (C) 3, and (D) 4; Table S1: The removal of the EMCs of heavy metals in stormwater runoffs in lterature and this study. References [60–65] are cited in the Supplementary Materials.

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