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Field Application of Spent Lime Water Treatment Residual for the Removal of Phosphorus and Other Pollutants in Urban Stormwater Runoff

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Abstract: The threat of anthropogenic eutrophication and harmful algal blooms in lakes requires the development of innovative stormwater best management practices (BMPs) to reduce the external loading of phosphorus (P). This paper presents the findings of a 5-year study of a full-scale P removal structure constructed in Minnesota, USA with spent lime drinking water treatment residual (DWTR), a by-product of water softening at a local water treatment plant. Influent and effluent water samples were collected by auto-samplers during 43 storm events during the growing season. Samples were analyzed for P constituents, heavy metals, total suspended solids (TSS), and pH. Toxicity of the effluent was assessed using *Ceriodaphnia dubia*. Flow-weighted removal effectiveness was calculated for each storm event. Overall, the spent lime DWTR reduced total P loading by 70.9%, dissolved reactive P by 78.5%, dissolved P by 74.7%, and TSS by 58.5%. A significant reduction in heavy metals was also observed. Toxicity tests indicated the aquatic toxicity of the effluent treated with spent lime DWTR was not different from untreated stormwater. This study provided long-term real-world data that demonstrated that a full-scale P removal structure with spent lime DWTR significantly reduced P and other pollutants in stormwater discharging to an urban lake. Therefore, spent lime DWTR, which is currently treated as a waste product, is a promising filter material for stormwater treatment.

Keywords: eutrophication; lake restoration; lime sludge; nutrients; pollution; water quality



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1. Introduction

Anthropogenic eutrophication of freshwater lakes has led to deteriorated water quality and the proliferation of harmful cyanobacterial blooms [1]. Reduction of external nutrient loading is a fundamental strategy to reverse the effects of anthropogenic eutrophication and restore freshwater lakes [2]. Phosphorus (P) is widely recognized as the key nutrient to target in external load reduction strategies [3,4]. P can accumulate in a watershed via atmospheric deposition, fertilizer, and sewage [5] and then be conveyed to lakes by stormwater and agricultural runoff [6]. Runoff from urban areas also contains elevated concentrations of metals [7]. In response, several stormwater best management practices (BMPs) have been developed to mitigate the flux of P as well as other pollutants from stormwater to receiving water bodies.

Stormwater BMPs encompass a wide spectrum of solutions for improving water quality and/or reducing runoff, relying on the mechanisms of filtration, infiltration, sedimentation and chemical reaction—often in conjunction with one another [8]. Structural stormwater BMPs (e.g., wetlands and detention ponds) and non-structural stormwater BMPs (e.g., land use changes and animal waste management policies) have been used

to reduce external P loading [9]. Among these options, media filters are one of the most effective BMPs for reducing total suspended solids (TSS) and particulate P but are less effective at removing dissolved P [10]. However, dissolved P cannot be ignored because it can remain a substantial long-term source of P to eutrophic water bodies due to historical P accumulation in the watershed that produces legacy P fluxes [11]. Dissolved P is also a greater threat to lake water quality because it is generally richer in bioavailable inorganic P that is directly available for uptake [12]. Thus, adsorbent materials have been incorporated into filtration media to enhance the chemical precipitation and adsorption mechanisms that more effectively target dissolved P [13]. The incorporation of mineral adsorbents targeting P into filtration media generates a “P removal structure” [14].

P removal structures are filters and weirs that incorporate P sorbing materials, which target dissolved P in non-point source runoff and wastewater by forcing flow through the P sorbing material [15]. P removal structures can be placed at stormwater “hot spots” to intercept and filter nutrient-rich or otherwise contaminated stormwater [16]. Recycled waste materials are particularly attractive as a cost-effective way to achieve this purpose [17].

Several recycled filtration media have been identified for use in P removal structures, including steel slag, coir, and drinking water treatment residuals (DWTRs) [18,19]. DWTRs are a byproduct of water treatment production [20] that have many promising environmental applications [21]. They have been incorporated into biofiltration media [22] and constructed wetlands [23] to remove P from incoming stormwater. DWTRs typically contains high amounts of amorphous aluminum (Al) and iron (Fe) minerals formed from Al- and Fe-based coagulants [24]. However, some DWTRs are rich in calcium (Ca), which is also an effective P sorbent [25].

Spent lime DWTR, which is the sludge byproduct of water softening at a water treatment plant (WTP), is a promising P sorbent due to its high concentration of Ca and magnesium (Mg) [26] and has been previously studied in the laboratory as a P removal filter material [27]. Similar lime-based materials have been used in other stormwater treatment approaches, including sequential biofiltration systems [28], a riparian buffer [29], lime-sand filters [30], and as a substrate in rain gardens [26]. Furthermore, other Ca-rich materials have been studied for use in P removal structures, including steel slag [31], acid mine drainage residue [32], burnt lime [30], and limestone [33]. Thus, Ca-rich spent lime DWTR is a feasible and practical choice as a filter material for a P removal structure.

Nevertheless, the long-term effectiveness of spent lime DWTR in removing P from stormwater in the field remains unknown. Laboratory studies are insufficient to determine the long-term performance of full-scale P removal structures. P retaining filters that showed promising results in the laboratory have been found to be orders of magnitude less effective when scaled up [34]. Instead, studies of field applications are necessary to ascertain true P removal performance under in situ conditions [18].

Therefore, the objective of this study was to determine the long-term effectiveness of a full-scale P removal structure constructed from spent lime DWTR for removing P from stormwater discharge to a eutrophic lake. The study also measured the removal of other pollutants, including TSS and metals, and assessed the toxicity of the treated effluent.

2. Materials and Methods

2.1. Site Description

The study site was adjacent to Wakefield Lake, a 9.1-ha lake in Maplewood, Minnesota, USA, with an average depth of 1.2 m and maximum depth of 2.7 m. The total watershed of the lake is 3.8 km², consisting primarily of low-density residential and commercial land use. Wakefield Lake discharges to another lake, as part of a chain of lakes within the Ramsey-Washington Metro Watershed District. The lake was designated by the state of Minnesota as impaired in 2002 due to excess nutrients and frequent algal blooms (2002–2011 growing season average total P = 106 µg/L, chlorophyll-a = 29 µg/L). The P removal structure was constructed to treat stormwater inflow at one of the three stormwater outfalls located at the lake. This outfall has a watershed of 1.15 km², including a large commercial area with

impervious surfaces, and is estimated to contribute the highest concentration of total P among all the lake's stormwater outfalls.

2.2. Characterization of Spent Lime Drinking Water Treatment Residual

The spent lime DWTR was provided by the Saint Paul Regional Water Services drinking WTP in Saint Paul, Minnesota, USA. The DWTR was produced from the single-stage water softening process using lime and collected as a sludge in the solids contact clarifier. During water treatment, the lime was combined with a small dose of aluminum sulfate (alum) to facilitate coagulation. However, the primary mode of flocculation was via precipitation induced by lime softening. As per the plant's normal operation, the sludge was dewatered via filter press and collected into a truck for transport. The dewatered filter cake was delivered directly to the study site and used without further modification. Total moisture was determined via oven drying at 105 °C. Total dry weight concentrations of Al, arsenic (As), barium (Ba), boron (B), cadmium (Cd), Ca, chromium (Cr), copper (Cu), iron (Fe), lead (Pb), Mg, manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), potassium (K), selenium (Se), silver (Ag), sodium (Na), and zinc (Zn) were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES) (EPA 200.7) following acid digestion (EPA 3050). Nitrogen components, including ammonia (NH₃-N), total Kjeldahl nitrogen (TKN), and nitrate (NO₃-N) were determined using the Kjeldahl method with mercury as a catalyst following acid digestion. Total P (TP) was determined using colorimetry (EPA 365.1) following acid digestion. Cyanide was determined via extraction (EPA 9013A).

2.3. Design and Operation of Treatment System

The spent lime P removal structure was constructed at the site of an existing stormwater outfall into Wakefield Lake. The existing outfall was disconnected from the lake approximately 3 m from the lake shoreline. A 28-m³ trench was excavated with a longer dimension parallel to the lake shoreline and perpendicular to flow (Figure 1). A wooden divider was installed to split flow into two streams and distribute it to the edges of the treatment cell. A riser pipe with twelve 2.5-cm perforations at its bottom was installed in the middle of the treatment cell and connected to a 6-inch diameter drainpipe that discharged to the lake. The top of the riser was open at the top (0.76 m above base) to allow overflow of standing water in the treatment cell during high intensity flows. The cell was filled with 20 m³ of spent lime DWTR, which had a surface area of 36.6 m² at the surface. Thus, during a storm event, the stormwater split laterally and infiltrated into the DWTR. The infiltrating stormwater reached the perforations of the riser and discharged to the lake via the drainpipe. As stormwater saturated the DWTR and began ponding on top, the head on the saturated media increased the outflow. If the head increased to the height of the open riser, this surface stormwater mixed with infiltrated flow in the drainpipe and discharged directly to the outfall.

Dimensions of a treatment cell were designed to facilitate maximum flow and treat as much water as possible but also allow for enough contact time for Ca to react with P. Based on preliminary testing in column tests, a contact time of 10 min or less was considered acceptable for targeted P removal. Therefore, the cell was designed to achieve around 5 to 10 min of contact time. Dissolved P removal from stormwater using Ca-rich materials generally requires 0.5 to 9 min of contact time [35].

Water level in the cell was measured at 10-min intervals during Phases 1 and 2 (2012 to 2014) using a Level Troll 500 water level data logger within a piezometric well installed 1 m from the outflow riser pipe.

Maintenance efforts were conducted each year in the spring prior to monitoring. To reduce compaction of the top layer of lime due to foot traffic and crumbling of the lime material with weathering, the top 0.3 m was mixed manually with a shovel.

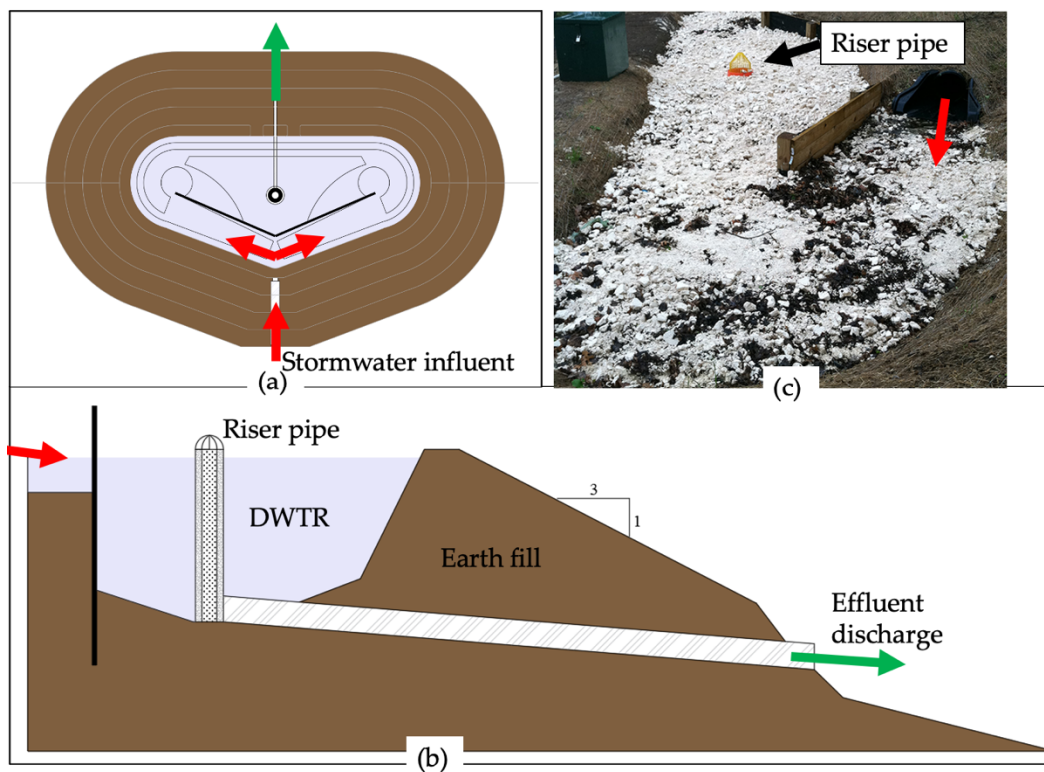


Figure 1. The P removal structure with spent lime DWTR, as depicted in (a) plan view, (b) cross section, and (c) actual photograph after construction.

2.4. Flow Monitoring and Water Quality Analysis

Stormwater inflow and outflow were measured with an area-velocity sensor that used a pressure transducer and Doppler sensor (ISCO 750 Area Velocity Flow Module, Teledyne ISCO, Lincoln, NE, USA). The sensor continuously measured inflow and outflow velocities and recorded values every 10 min during the growing season (generally from April/May to October/November) for the years 2012–2013 (Phase 1), 2014 (Phase 2), and 2015–2016 (Phase 3). Water samples were collected at the inlet and outlet of the spent lime P removal structure using ISCO 6700 and ISCO 6712 auto-samplers. The auto-samplers were programmed to collect flow-paced samples based on input from the area-velocity sensors. The auto-samplers collected discrete draws for every 3300 L of flow that was calculated to pass the sensor at a rate equivalent to a 1-L sample (3 discrete draws) per 10,000 L of flow. The auto-sampler had a capacity for eight 1-L bottles, meaning a maximum of 24 draws could be collected during a storm event. Following a storm event, the 1-L sample bottles were collected and combined into a composite sample for analysis, creating a single flow-paced sample for each the influent and effluent, representing the overall storm event water chemistry. A storm event was defined in the analysis as the duration between the first and last sample collected by the auto-samplers.

During Phase 1 of the study, water samples were analyzed for TP, total dissolved P (TDP), dissolved reactive P (DRP), TSS, chloride, and metals (Ca, Fe, Al, Cu, Pb, and Zn). During Phase 2 of the study, water samples were analyzed for pH. For this phase only, each 1-L sample was analyzed separately, rather than compositing samples from the rain event. During Phase 3 of the study, composite water samples were analyzed for TP, DRP, TSS, and chloride.

P was analyzed using colorimetry (EPA 365.1/Standard Methods 4500-P), with DRP filtered using a 0.45- μm membrane filter prior to analysis and TDP filtered and digested with persulfate prior to analysis. Metals were determined with ICP-AES (EPA 200.7). TSS was determined with filtration and drying (Standard Methods 2540 D). pH was measured with

a calibrated pH electrode (YSI 600 XLM V2). Chloride was determined by amperometric titration (Standard Methods 4500-Cl E).

2.5. Contact Time

Contact time is an important design parameter for P removal structures because it affects treatment efficiency and the chemical properties of treated stormwater [18,36]. The contact time (t_c) between stormwater and spent lime DWTR at a given 10-min time interval (i) was calculated using (Equation (1)):

$$t_{c,i} = \frac{\eta V_i}{q_i}, \quad (1)$$

where V is the volume occupied by the stormwater in the P removal structure at a given 10-min interval (m^3), η is the porosity of the spent lime, and q is the outflow at a given 10-min interval (m^3/min). The volume V was dependent on the depth of saturation, described by a polynomial equation derived from the relationship between depth and cross-sectional surface area in the construction drawings. The water depth was measured by the piezometric well. The porosity η was estimated to be 0.45 based on a porosity measurement of an in situ core collected from the spent lime. The contact time was determined at each 10-min interval where depth and q were measured. A flow-weighted average contact time was then determined for each storm event that had the necessary data available.

2.6. Toxicological Analysis

The chronic toxicity of the treated discharge was assessed using a single concentration screening test with static renewal and the sensitive daphnid *Ceriodaphnia dubia* [37]. The daphnids were obtained from in-house cultures, which were verified for quality control using sodium chloride as a reference toxicant in the same months as the tests, in accordance with EPA methods [37]. The daphnids were maintained within treated effluent (100% concentration), which was sampled as described above, for a period of 7 days. Reproduction was the primary endpoint monitored, with survival also being recorded. Untreated stormwater influent (100% concentration) and a moderately hard laboratory control water were used for comparison. The tests were conducted using stormwater from two different storm events in May and June 2012. Hardness (EPA Method 130.1), alkalinity (EPA Method 310.1), and specific conductance (EPA Method 120.1) were measured for each sample at the beginning of the test. pH was recorded before and after each water renewal. Differences in the reproduction outcomes were assessed using the non-parametric Kruskal–Wallis test with Bonferroni correction. Between group differences were assessed using Dunn's post hoc comparison test. A statistical significance (α) of 0.05 was used.

2.7. Pollutant Removal Assessment

Composite samples of the inflow and outflow to the P removal structure were collected by the auto-sampler at fixed volume intervals, thereby creating flow-weighted averages of each storm event. The percentage of pollutant removed for individual storm events was computed as the percent reduction from the inflow to the outflow concentrations. If a parameter was not detected in a sample, the values were set equal to half the detection limit for the purpose of pollutant removal analysis.

In addition, the total mass of P removed by the structure was estimated by (Equation (2)):

$$\text{Total P removed} = Q (\bar{C}_{in} - \bar{C}_{out}), \quad (2)$$

where Q is the total flow monitored and \bar{C}_{in} and \bar{C}_{out} were the flow-weighted influent and effluent TP concentrations during all sampled storm events, respectively.

Discrete flow volume was calculated for each 10-min time interval, and total flow volume was determined by the sum of discrete flow volumes over the corresponding period. Because the samples were flow-paced, the total mass of a pollutant in the influent

and effluent was equivalent to the product of the total flow during the storm event and the concentration. For analysis, storm event flow and cumulative flow were computed as the average of the inflow and the outflow to account for variability in the flow measured by the two area-velocity meters.

Differences in the influent and effluent pollutant concentrations were statistically assessed using the Wilcoxon signed-rank test using a significance value (α) of 0.05. This non-parametric statistical test does not require normally distributed data and can robustly handle outliers.

3. Results

3.1. Characterization of Spent Lime DWTR

The spent lime DWTR was primarily composed of Ca (333 mg/g), Mg (35.7 mg/g), Al (5.4 mg/g), and Fe (5.1 mg/g) (Table 1). Although bicarbonate was not provided, it was assumed to be the primary anion. The concentrations of all 10 regulated metals were below the pollution concentration limits specified by the U.S. EPA's Part 503 regulations on land application of biosolids, meaning the spent lime DWTR was equivalent to biosolids of exceptional quality and would be acceptable for land application [38]. While the spent lime was dewatered with filter press at the WTP, the initial moisture content was 59.3%. Therefore, additional dewatering likely occurred via evaporation following placement in the P removal structure.

Table 1. Physiochemical characterization of spent lime DWTR used in the P removal structure.

Parameter	Concentration (mg/kg Dry Weight)	Parameter	Concentration (mg/kg Dry Weight)
% Moisture	59.3	Magnesium	35,700
Aluminum	5350	Manganese	115
Ammonia as N	74.4	Mercury	<0.046
Arsenic	<10.9	Molybdenum	<8.2
Barium	122	Nickel	116
Boron	<81.6	Nitrate as N	<7.4
Cadmium	<1.6	Total Phosphorus	117
Calcium	333,000	Potassium	<1350
Chloride	113	Selenium	<8.2
Chromium	184	Silver	<5.4
Copper	15.3	Sodium	<533
Cyanide	<1.3	Sulfate	<98.9
Iron	5080	TKN	831
Lead	<10.9	Zinc	<10.9

3.2. Storm Event Hydrology and Contact Time

Flow data were recorded for a total duration of 753.5 days during the 5 calendar years of 2012 to 2016, representing 41.3% of the total time. Flow was not monitored during the non-growing season (58.7% of time) when freezing temperatures prevented monitoring. However, overall rainfall and high intensity rainfall are more common during the growing season. A total of 43 storm events from 2012 to 2016 were used in the data analysis following the exclusion of four storm events due to incomplete data. The total recorded flow during the 43 storm events that were included in the analysis was 1823 m³, which was 33.7% of all recorded flow over the 5-year period (Supplementary Materials Table S1).

The median storm event flow was 32.6 m³, ranging from 7.2 to 98.6 m³ (Figure 2a). The inter-quartile range of peak flow rates recorded during storm events was between 6.7 and 25.7 L/s, but one storm recorded a peak 10-min discrete flow rate of 55.2 L/s (Figure 2b). Generally, most storm events were less than 8 h (Figure 2c), but in a few cases, storm event durations were 30 to 50 h. However, most of these long-duration storms were comprised of two flow peaks separated by little to no flow.

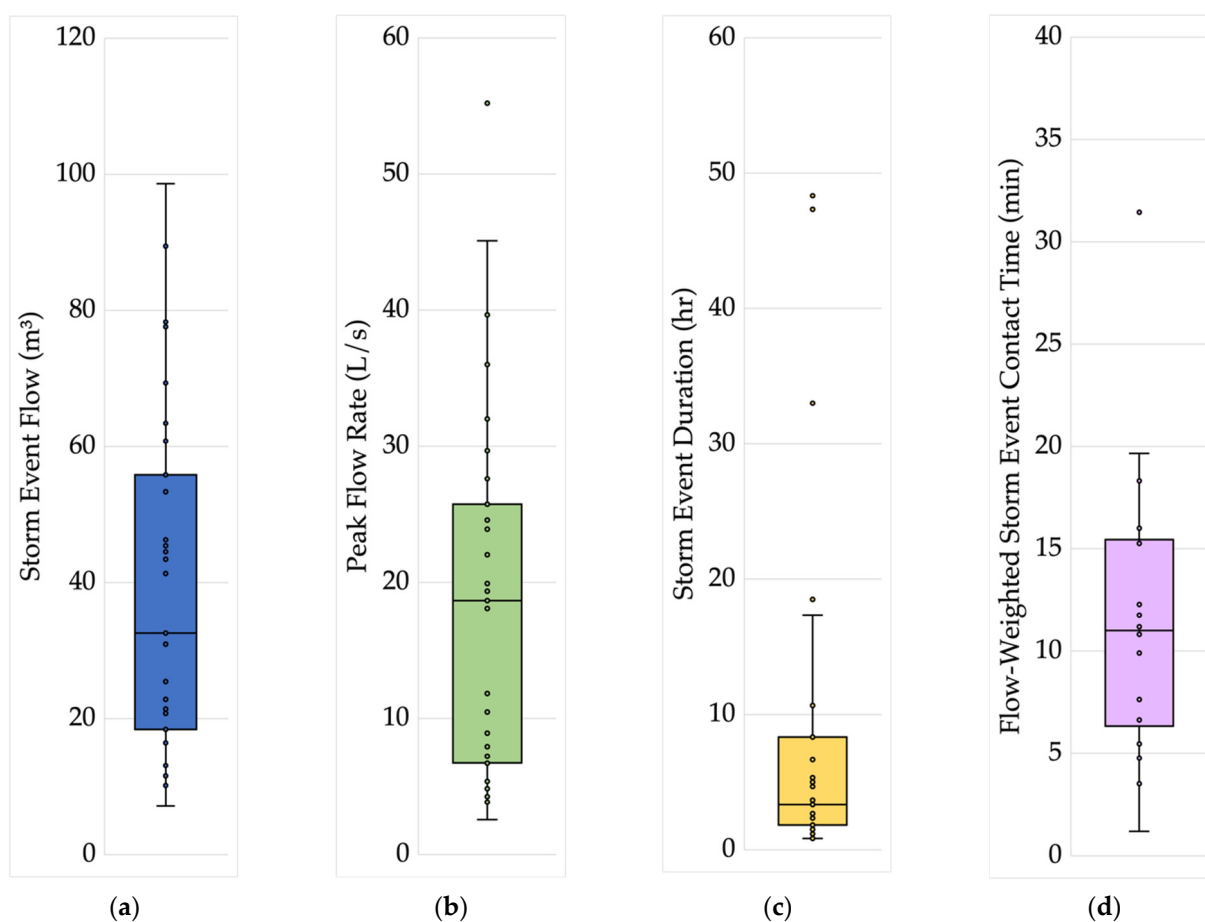


Figure 2. Distribution of hydrologic statistics from all storm events that were sampled and included in the analysis ($n = 43$), including (a) the total flow from each discrete storm event, (b) peak 10-min recorded flow rate during each storm event, and (c) duration of the storm event defined by the first and last sample collected. (d) The distribution of flow-weighted contact time for storm events in which the piezometer was installed to monitor water levels ($n = 18$).

A flow-weighted contact time was calculated for each storm event in which the piezometer was recording the saturation depth inside the spent lime DWTR. Storm event contact times were between 1.2 and 19.7 min with one outlier at 31.4 min (Figure 2d). Three out of 18 storm events (17%) had contact times less than 5 min.

3.3. Removal of Phosphorus and Suspended Solids

The flow-weighted event mean concentrations of TP in the influent stormwater were generally around 0.3 to 0.5 mg/L in individual storm events but were as high as 2.7 mg/L during storm event 40 (Figure 3a). The overall flow-weighted average of all 35 storm events for which TP was analyzed was 0.36 mg/L (Table 2). The effluent discharge from the P removal structure had lower TP concentration for every sampled storm event. The overall average TP concentration in effluent was 0.11 mg/L, corresponding to a statistically significant 70.9% reduction in TP.

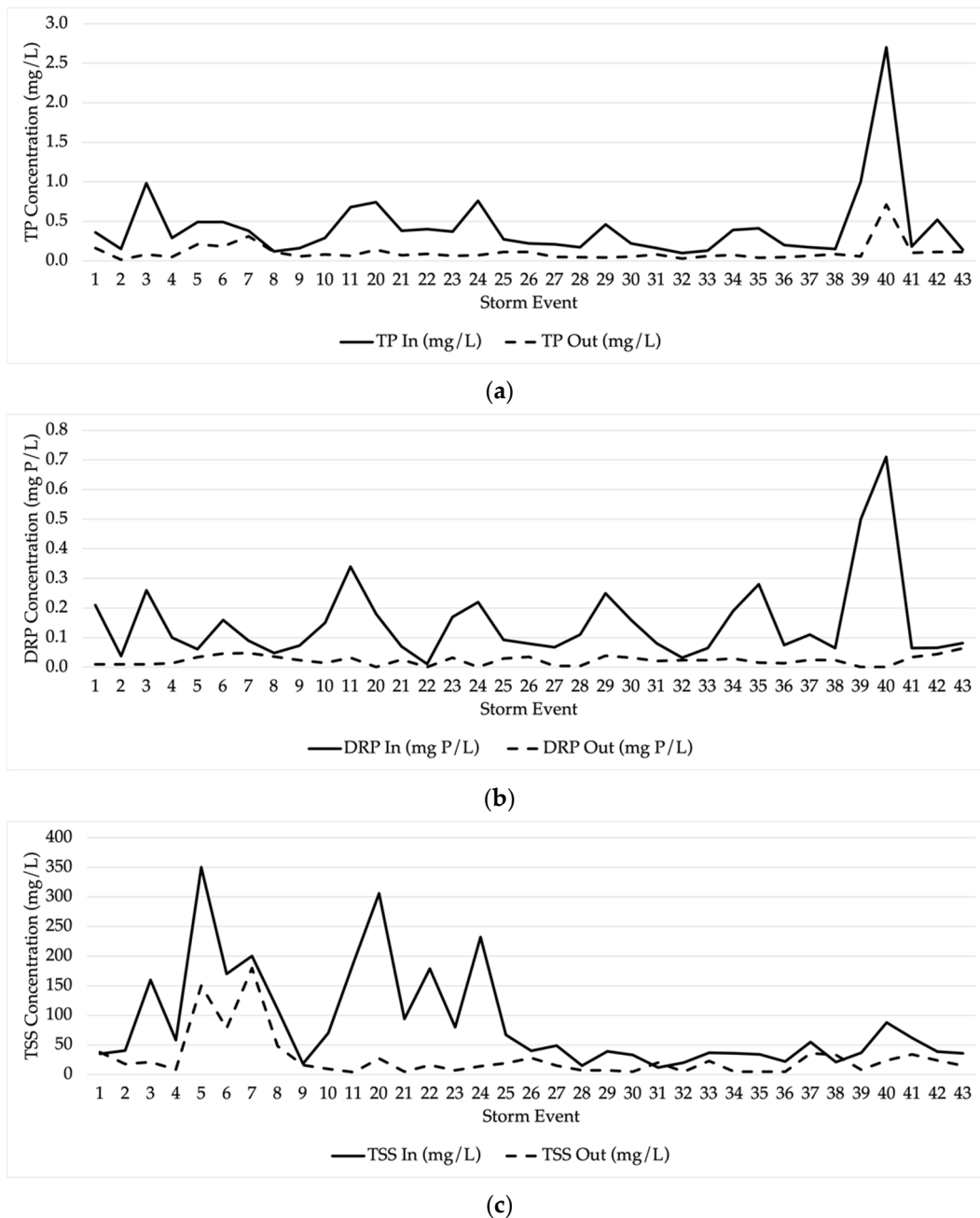


Figure 3. Flow-weighted average influent (solid line) and effluent (dotted line) concentrations of (a) total P, (b) total reactive P, and (c) TSS for 35 sampled storm events.

DRP comprised 34% of the TP loading (Table 2). The influent event mean concentrations of DRP followed a similar trend to that of TP (Figure 3b). Like TP, the effluent DRP concentrations were lower than the influent concentrations for all storm events, resulting in a 78.1% overall reduction in DRP. TDP was analyzed in a smaller subset of 10 storm events, but the results indicated that dissolved P was effectively removed as well, with a statistically significant reduction of 74.7% (Table 2). Dissolved P comprised 41% of TP.

Event mean influent concentrations of TSS fluctuated between 12 and 350 mg/L (Figure 3c). Effluent from the P removal structure reduced TSS concentrations in 33 out of 35 storm events (94%) with an overall reduction of 58.8%, which was statistically significant.

Table 2. Flow-weighted characterization of water quality and percent removal for nutrients, solids, and metals from influent stormwater and effluent discharge.

Parameter	Influent Flow-Weighted Avg. (Range) ^{1,2}	Effluent Flow-Weighted Avg. (Range) ^{1,2}	Flow-Weighted % Removal (Range) ^{1,3}	<i>p</i> -Value ⁴	Sampled Storm Events	Sampled Flow (m ³)
Total P (mg/L)	0.36 (0.096–2.7)	0.11 (0.14–0.71)	70.9% (8.3–94.5)	≤0.001 ***	35	1369
Dissolved Reactive P (mg/L)	0.12 (0.011–0.71)	0.026 (0.001–0.064)	78.5% (22.0–99.9)	≤0.001 ***	35	1369
Total Dissolved P (mg/L)	0.15 (0.14–0.34)	0.038 (0.014–0.091)	74.7% (–5.8–95.6)	0.008 **	10	452
TSS (mg/L)	92.4 (12–350)	38.0 (4.0–180)	58.8% (–66.7–97.9)	≤0.001 ***	35	1369
Chloride (mg/L)	4.1 (0.42–29)	5.9 (1.6–58.1)	–45.6% (–733–42.7)	0.018 *	33	1312
Aluminum (mg/L)	3.2 (0.33–8.5)	1.6 (0.11–4.0)	50.6% (4.0–91)	0.014 *	11	530
Calcium (mg/L)	11 (3.6–16)	8.2 (2.6–18)	28.1% (–12–65)	0.13	11	530
Iron (mg/L)	3.7 (0.45–9.1)	1.7 (0.12–4.3)	54.9% (13–93)	<0.001 ***	11	530
Copper (µg/L)	23 (8.7–29)	14 (2.5–24)	38.3% (–101–88)	n/a	4	213
Lead (µg/L)	9.7 (3.1–12)	2.4 (0.50–3.8)	75.5% (–13–94)	n/a	4	213
Zinc (µg/L)	108 (57–140)	28 (6.6–69)	74.3% (–22–94)	n/a	4	213
pH ⁵	6.09 (5.19–7.08)	8.02 (7.15–9.52)	—	—	8 [†]	454

Notes: ¹ Overall average of all sampled storm events weighted by storm event flow. Range is the range of discrete values for each storm event. ² In some cases, the minimum value in the range represents a parameter not detected, in which case a value of one half the detection limit was used. ³ A negative % removal indicates an increase. ⁴ *p*-value from testing null hypothesis that the difference between the influent and effluent concentrations was zero using a two-tailed Wilcoxon signed-rank test. * *p* < 0.05; ** *p* < 0.01; *** *p* < 0.001. n/a = not applicable due to insufficient sample size. ⁵ Statistics for pH were computed after converting pH to the activity of the hydrogen ions. [†] = For pH, individual samples were analyzed, rather than composites. Therefore, each storm event had up to 24 samples for each the influent and effluent.

Throughout the 35 storm events during which TP was sampled, 1369 m³ of stormwater was estimated to flow through the P removal structure (Table 2). Based on the flow-weighted mean concentration of 0.36 mg/L, the P loading during sampled storm events was 0.49 kg. Assuming equal TP concentration in the flow that was observed but not sampled, the total P loading was 1.98 kg, or 22.9 mg P/ha/day. Because stormwater flow was not monitored during the non-growing season, the actual cumulative P loading on the P removal structure over the 5-year monitoring period was higher.

3.4. Factors Potentially Associated with Phosphorus Removal

The spent lime DWTR effectively reduced all forms of P from incoming stormwater. However, removal effectiveness varied across different storm events. Contact time, influent P concentration, and P saturation were all factors that may have potentially affected P removal effectiveness. First, it appeared that TP removal was negatively associated with the peak flow rate through the P removal structure (Figure 4a). When the peak flow rate in a storm event did not exceed 20 L/s, the event mean P removal was 60% or higher. On the other hand, if a storm event included a peak flow rate greater than 30 L/S, the event mean P removal was 60% or lower. TP removal may also have been associated with the influent TP concentration (Figure 4b). The spent lime DWTR was generally less effective at reducing P in stormwater that had a lower concentration of TP.

A trend in P removal effectiveness does not appear from the cumulative flow through the P removal structure (and, therefore, time) (Figure 4c). If a P sorbent has become

saturated with P, the removal effectiveness will decline. The cumulative TP removal reached 70% after the first 2000 m³ of flow and maintained that same average through the duration of the study, suggesting the spent lime DWTR still had sufficient P sorption capacity (Figure 5). Based on an estimate of the dry weight mass of the spent lime DWTR added to the structure, it was estimated that after 5 years the amount of TP removed was equivalent to around 200 mg P/kg dry weight.

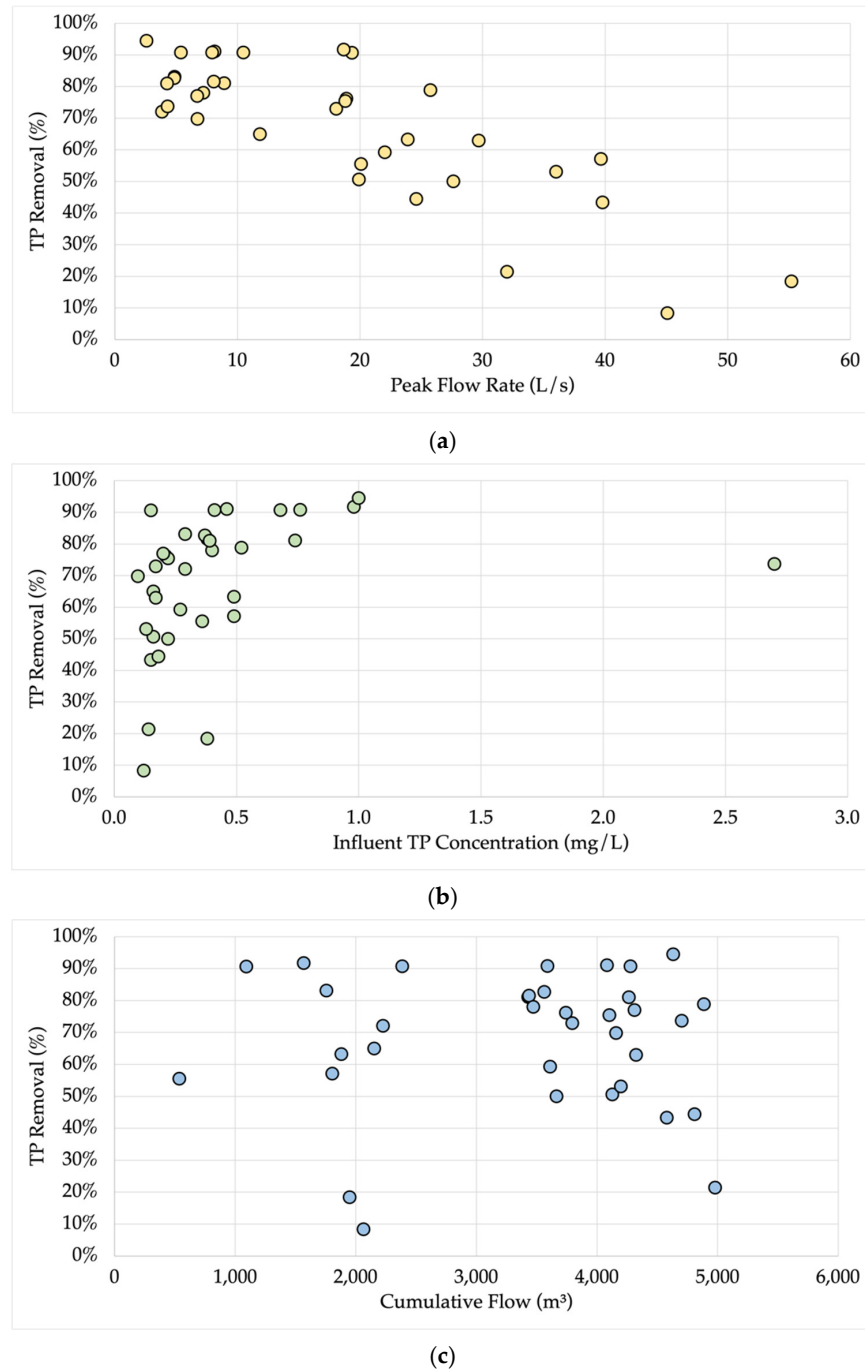


Figure 4. Event mean TP removal (%) plotted against (a) peak flow rate observed during storm event, (b) event mean TP concentration in stormwater, and (c) cumulative total of stormwater that flowed through the spent lime DWTR.

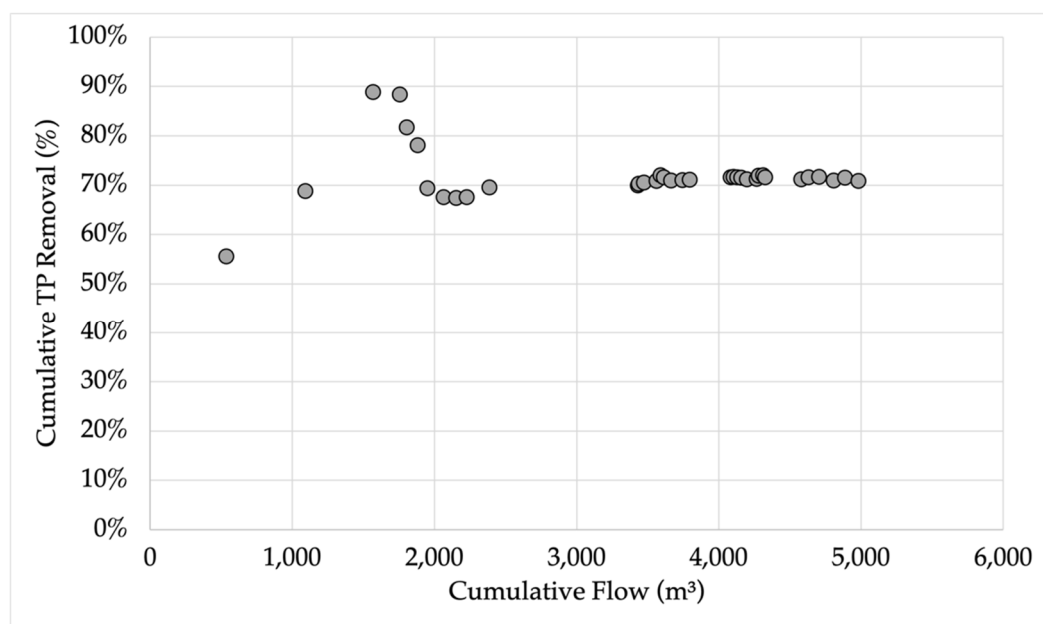


Figure 5. Flow-weighted TP removal calculated cumulatively for each storm event as a function of the cumulative flow that passed through the spent lime DWTR.

3.5. Metals Removal

Overall, the P removal structure produced a net reduction in the concentration of the six metals that were analyzed (Figure 6). The reductions in Al and Fe were statistically significant, but the reduction in Ca was not significant at the 0.05 significance level (Table 2). Due to the insufficient sample size, the significance of reductions in Cu, Pb, and Zn could not be tested. However, the reductions in Pb and Zn (75.5% and 74.3%, respectively) were comparable to those of P constituents.

3.6. Aquatic Toxicity

Survival was 90% or higher in all three groups (laboratory control, untreated stormwater, and treated effluent) during both toxicity tests conducted in May and June of 2012 (Table 3). The means of neonates (i.e., reproductive offspring) per female were generally higher in the treated (24.8–25.9) and untreated stormwater (20.3–26.9) groups compared to the laboratory control (17.7–18.7). Statistical testing of the full set of results with the Kruskal–Wallis test indicated that there was a significant difference in the reproductive outcomes among the groups ($\chi^2(2) = 18.74, p < 0.001$). The post hoc Dunn's test using a Bonferroni corrected α of 0.017 indicated that the influent and effluent were different from the laboratory control but that the reproductive outcomes in the influent and effluent groups were not significantly different. Therefore, there were no differences in the survival, reproductive output, and inferred toxicity between the stormwater previously discharged directly to the lake and the stormwater discharge treated with the spent lime DWTR.

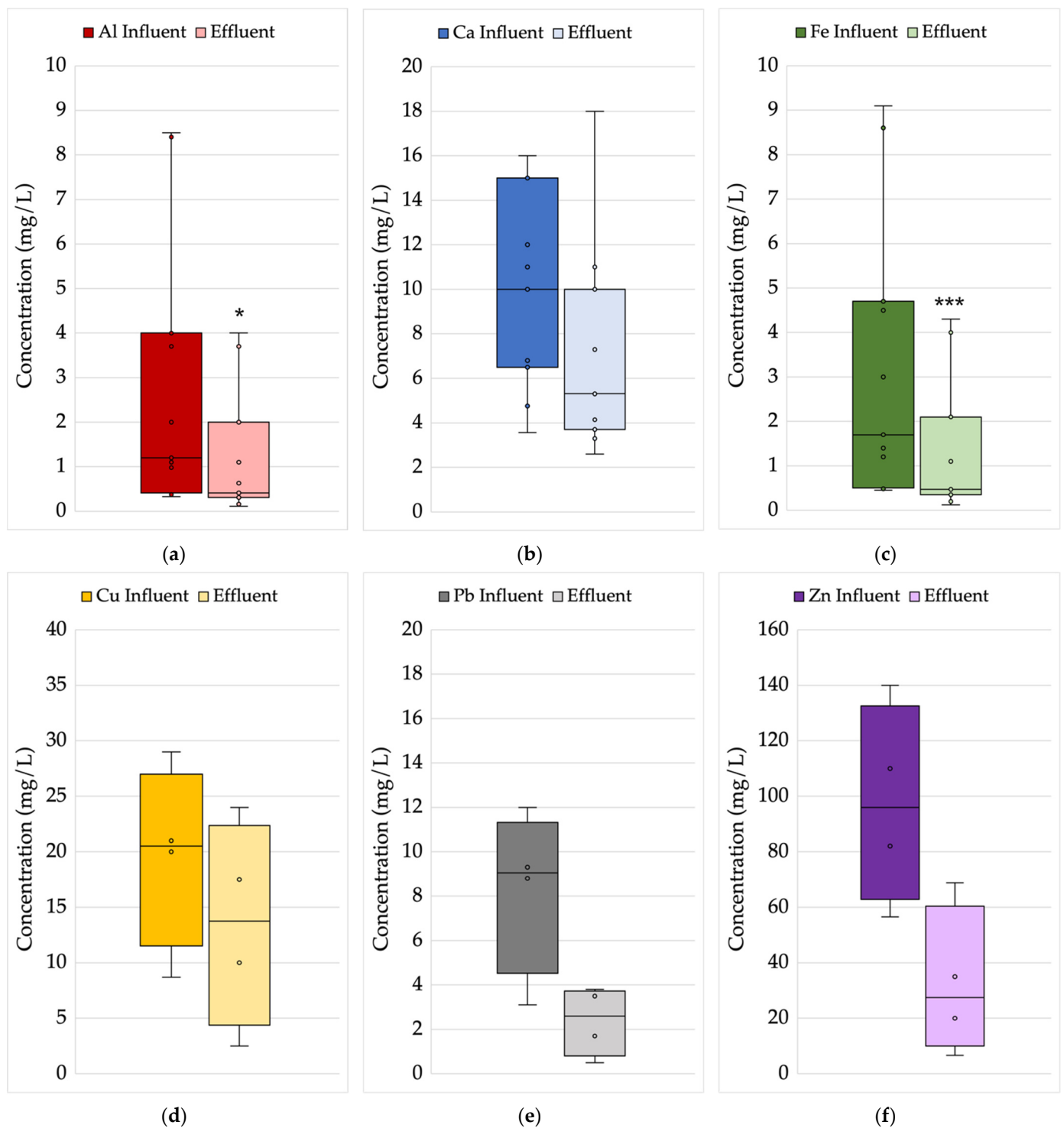


Figure 6. Distribution of event mean concentrations in the influent stormwater and effluent discharge for the metals (a) aluminum, (b) calcium, (c) iron, (d) copper, (e) lead, and (f) zinc. Asterisks represent statistically significant differences in the post-treatment effluent compared to the pre-treatment influent (* = $p < 0.05$; *** = $p < 0.001$). If no asterisks are shown, either no significant differences were detected, or the sample size was too small to test for differences.

Table 3. Results of seven-day laboratory tests to assess chronic toxicity conducted with *Ceriodaphnia dubia* and stormwater collected at the inlet (untreated) and the outlet (treated) of the spent lime treatment cell on two dates.

Sampling Date	Water Source	% Survival ¹	Avg. Young (Range) ²	pH Range	Hardness (mg/L as CaCO ₃)	Alkalinity (mg/L as CaCO ₃)	Specific Conductance (µS/cm)
21 May 2012	Lab Control	90	18.7 (7–22)	6.64–8.20	104	88	227
	Stormwater In	100	20.3 (15–25)	6.14–7.41	20	8	95
	Effluent Out	100	25.9 (21–29)	6.66–8.02	128	92	576
19 June 2012	Lab Control	100	17.7 (7–27)	6.92–8.26	112	84	222
	Stormwater In	100	26.9 (22–29)	6.37–7.45	8	24	42
	Effluent Out	90	24.8 (13–30)	6.97–7.87	116	108	469

Notes: ¹ Total survival of the adults at the completion of the seven-day testing period. ² Young is the total number of *Ceriodaphnia dubia* neonates produced per female adult during the seven-day testing period.

A comparison of the water quality between the stormwater influent and treated discharge showed that the pH, hardness, and alkalinity increased following treatment with the spent lime DWTR. Results from the storm monitoring also showed that the spent lime DWTR increased the pH in the slightly acidic stormwater from an average of 6.09 to an alkaline 8.04. (Table 2).

4. Discussion

Addressing external P loading to freshwater lakes is a critical strategy to combat the global threat of increasing harmful cyanobacterial blooms [39] and eutrophication in general. Municipalities and local authorities around the world need cost-effective and ecologically friendly materials to combat external P loading [40]. While many stormwater BMPs are effective at reducing particulate forms of P and point sources of P, the buildup of P in a watershed creates a more difficult problem to solve. Once a watershed has accumulated P, it will continue to be a source of lower concentrations of P for an extended period, generating what is known as “legacy P” [41]. Most stormwater BMPs are ineffective at non-point sources of legacy dissolved P [10,42]. The results from this study showed that spent lime DWTR generated from water softening offers a valuable, low-cost material capable of protecting water bodies from the threat of legacy and other sources of P in stormwater runoff. The results add to the growing number of proposed uses for DWTRs for the purpose of lake restoration, including as a bed for constructed wetlands [43], as a sorbent for flow-through treatment of eutrophic water [44], and as a P sorbing ballast in “flock and lock” treatment [45]. This study also provided valuable long-term data from a full-scale field application, allowing for the determination of true P removal performance under in-situ conditions, rather than laboratory data, which can overestimate performance by orders of magnitude [34].

4.1. Phosphorus Removal Performance

The P removal structure with spent lime DWTR was able to cumulatively remove 70.9% of TP. This finding was consistent with removal rates of P removal structures with other forms of high P sorption capacity filter media. For example, P removal structures with arc furnace steel slag removed around 50% of TP [15], while those with industrial spent lime removed around 60% of TP [18,30]. TP removal efficiency of the spent lime DWTR was higher than the median removal percentage of all reported media filters (45%) [10].

The spent lime DWTR used in this study was also consistently effective at removing TP for the duration of the study. After 5 years of operation, the structure was estimated to remove at least 1.41 kg of P from stormwater during the growing season alone, which equates to around 200 mg P/kg dry weight DWTR. This estimate did not account for P flux during the non-growing season, meaning the real amount of P captured within the spent lime DWTR was higher. Other P sorbing materials in flow-through devices have effectively

removed TP while sorbing up to 2000 to 3000 mg P/kg material [18,30,46]. Other DWTRs, which were primarily Al-based, have had even higher P sorption capacities, with laboratory studies reporting values as high as 10,000 or 20,000 mg P/kg [24,47,48]. One laboratory study using spent lime DWTR reported a P sorption capacity of 55,000 mg P/kg [49]. However, the results of such short-term laboratory experiments should be used with caution to predict the lifespan of a P removal structure. A review of stormwater BMPs has shown that most studies overestimate the long-term performance of the BMP due to assumptions based on short-term experiments [50]. Nevertheless, the long-term real-world data from this study demonstrated that this P removal structure with spent lime DWTR may be expected to maintain the high percentage of P reduction for several more years before the P sorbing material needs to be replaced. Nevertheless, the accumulation of P should be monitored regularly to prevent breakthrough [30].

A key finding in this study was that the spent lime DWTR was also able to cumulatively remove 78.5% of DRP and 74.7% of TDP. Dissolved P is particularly difficult for most stormwater BMPs to remove. Overall, retention ponds have the best performance for reducing dissolved P (median of 50%), but this is generally for high influent concentrations [10]. Media filters, which are commonly sand filters, are most effective at overall P removal but are less effective at achieving low levels of P in the effluent [51]. Efficient and consistent removal of dissolved P is generally only possible if chemical binding methods (e.g., P binding minerals) are used in conjunction with sedimentation [52,53]. Thus, the data suggest that binding of P to mineral surfaces was an important mechanism, which is further discussed below.

A limitation to the P removal performance was detected during high intensity rainfall events. When the peak flow rate of a storm exceeded 30 L/s, TP removal for that event was less than 60%. This reduction in P removal effectiveness was likely due to ponding on the structure's surface that forced untreated overflow to discharge directly via the standpipe. Reductions in P removal due to high flow rates have been documented in other P removal structures [16] and maintaining sufficient treatment during high flow rates remains one of the biggest design challenges [32].

4.2. Pollutant Removal Mechanisms

Stormwater BMPs that rely on filtration as the primary treatment mechanism, such as sand filters, are effective at removing TSS and particulate P, as was observed in this study. However, sorptive removal mechanisms are often necessary in conjunction with filtration to achieve sufficient removal of soluble pollutants. Filtration media have been enhanced with iron filings [54], Al-based DWTRs [55], and industrial byproducts [27] in order to enhance soluble pollutant removal. These P sorption materials can enhance P removal through a variety of mechanisms. It has been proposed that lime- and Ca-based filters rely primarily on chemical precipitation to remove P [26,30]. However, the findings of this study suggested that chemical precipitation (i.e., surface precipitation) may have only played a minor role as a P removal mechanism. First, the observed effluent pH ranges were generally around 8 and often less than 8. Precipitation of P with lime generally requires pH greater than 8 and peaks as pH exceeds 11 [30]. Secondly, precipitation of phosphates in soils dominates at higher P concentrations (around 10–15 mg P/L), whereas at lower concentrations adsorption to calcium carbonate minerals dominates [56]. Concentrations of P in the stormwater of this study were over a full order of magnitude lower than that threshold. Finally, P removal due to precipitation is much more sensitive to contact time, especially at lower concentrations [27]. While the flow-weighted contact times in most storm events were 5–15 min (with 10 min being considered a long contact time), P removal performance was relatively consistent throughout the full range of contact times. Thus, while surface precipitation likely occurred, it did not appear to dominate as a P removal mechanism.

Further enhancing the possibility for adsorptive ligand exchange, the spent lime DWTR in this study was also generated with a small dose of alum, likely introducing

Al hydroxides. The high concentrations of amorphous Al in Al-based DWTRs facilitate effective and selective P adsorption mechanisms primarily through ligand exchange on mineral surfaces [57,58]. Because the spent lime DWTR was effective at removing TSS, dissolved P, and metals, it is likely that filtration, precipitation, and adsorption were key pollutant removal mechanisms working in conjunction with one another [59].

Removal of Pb was likely driven by the adsorption of Pb on to calcite surfaces by moving into available Ca sites [60]. In addition, Pb is very insoluble in the presence of high carbonate concentrations [61], likely improving the removal of this metal from stormwater. Cu may have been removed due to adsorption or precipitation on to calcite surfaces at high pH [62] or associated with trapped organic matter in the filter media [63]. However, because the dissolved component of metals was not monitored, the contributions of filtration and precipitation mechanisms cannot be differentiated.

4.3. Design Considerations

P removal structures distribute P-rich flow over a bed and force flow through a P sorbing material with a contact time long enough to treat the water even in high flow conditions. Therefore, there are several important design variables, outside of storm sizing, that need to be considered with respect to P removal. First, the hydraulic conductivity of the material must be sufficient to achieve the desired goal. P sorbing materials tend to have lower hydraulic conductivity than sand [64]. Hydraulic conductivity may be further reduced as precipitants and amorphous minerals reduce the effective porosity, potentially causing clogging [65]. Thus, the physiochemical properties of the filter media are key to performance in stormwater BMPs, with the ideal media having coarse particle sizes, high hydraulic conductivity, and high particle surface area [36].

A second design consideration is the need to perform regular maintenance. One of the three components that define a properly designed P removal structure is the ability to retain and replace the P sorbing material [15]. Once the lifetime (i.e., P sorption capacity) of the P sorbing material has been reached, it is necessary to replace it. However, other intermittent maintenance is necessary too. Because the spent lime DWTR was somewhat plastic, the pore space could be reformed by breaking apart and repacking the spent lime cake. The annual maintenance that was performed likely helped maintain a consistent hydraulic conductivity and also exposed new surfaces for P sorption. Such regular maintenance can be used to remove trash and leaves and to inspect for the development of preferential pathways. Ponding can quickly lead to preferential flow through filtration media and reduce P removal effectiveness [66]. Furthermore, if surface flow becomes dominant over infiltration, the BMP can fail to reduce soluble P [23].

A third key design consideration is the fact that most P removal structures are required to operate under variable flow and P loading conditions (i.e., low to very high). This fact is important because many laboratory studies of P sorption use high P concentrations and long contact times, which are not representative of field conditions [36].

Overall, there is no “one size fits all” approach to reducing external P loading to lakes. Site-specific characteristics, such as the proportion of dissolved P in runoff, are important considerations in the design and construction of P removal structures [18]. To facilitate these design decisions, software has been designed to aid in the selection and design of P removal structures [67].

5. Conclusions

The use of spent lime DWTR is an attractive option as a filter media in a structure that removes P and pollutants from stormwater runoff. Data from a 5-year, real-world field application showed that spent lime DWTR, used without modification, was able to significantly reduce all forms of P, including dissolved and reactive P. In addition, the P removal structure reduced concentrations of particulates and heavy metals. It is likely that the pollutants were removed through a combination of filtration, precipitation, and adsorption, as has been shown in previous laboratory studies with similar materials.

Finally, tests with the daphnid *Ceriodaphnia dubia* indicated that the aquatic toxicity of the effluent was no different than untreated stormwater, and the increases in the effluent pH were within acceptable ranges. The ability of the spent lime DWTR to achieve significant pollutant reductions with short contact times and across a full range of storm event flow rates demonstrated that it is an effective, efficient, and low-cost stormwater BMP for protecting receiving water bodies.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/w14132135/s1>, Table S1: full dataset of monitored storm events.

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