


Stormwater runoff pollution control performance of permeable concrete pavement and constructed wetland combined system: toward on-site reuse

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ABSTRACT

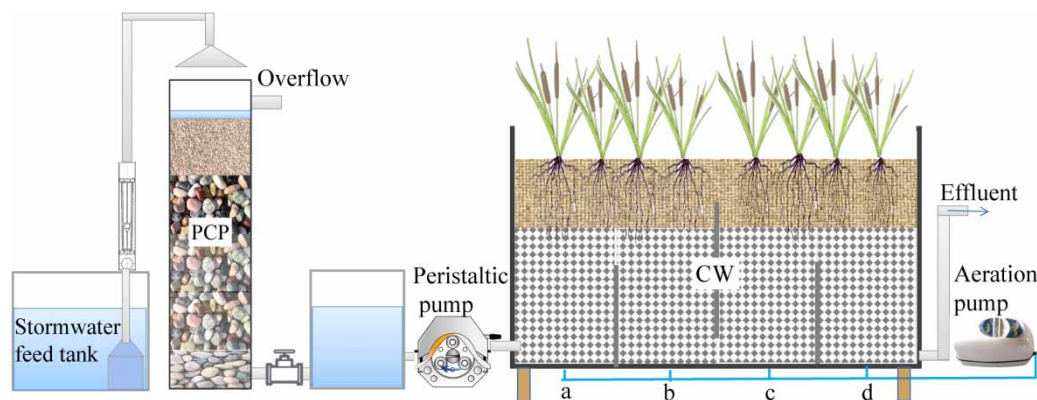
Urban waterlogging and the deterioration of receiving water quality caused by stormwater runoff have become increasingly significant problems. Based on the concept of combining grey and green infrastructure, a combined permeable concrete pavement (PCP) and constructed wetland (CW) system has been developed to treat stormwater runoff and enable on-site reuse. The results showed that the removal rate of suspended solids (SS) by PCP ranged from 96.61 to 99.20%; however, the chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) concentrations in the effluent did not meet the standards required for rainwater reuse. For the combined PCP-CW system, the removal rates of COD, TN and TP by the CW were 48.45–75.12%, 47.26–53.05%, and 59.04–75.28%, respectively, under different hydraulic loading (HL) rates; thus, the effluent TN concentrations did not consistently meet the reuse standards. Further optimization of aeration in different parts of the CW revealed that aeration in the middle and front sections of the wetland had the most significant effect on pollutant removal, under which the TN concentrations in the effluent met the standard required for reuse. The effluent from the combined PCP-CW system was able to fully meet the stormwater reuse standards under these optimized conditions, and the reuse of urban stormwater runoff can therefore be realized.

Key words: constructed wetland, permeable concrete pavement, runoff pollution, stormwater reuse

HIGHLIGHTS

- PCP-CW combined system based on the concept of combining grey-green infrastructure has been developed.
- Runoff pollution control performances of PCP and PCP-CW were evaluated.
- Micro-aeration can improve the pollution control performance of the PCP-CW system.
- The effluent from PCP-CW can meet the rainwater reuse standard under optimized conditions.
- A feasible way for on-site stormwater reuse is provided.

GRAPHICAL ABSTRACT



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1. INTRODUCTION

Rapid urbanization has increased the impervious area of cities, leading to urban waterlogging and threatening people's lives and property; meanwhile, the pollutants in rainfall runoff further aggravate the urban water environment pollution, leading to the 'black and odorous water' phenomenon observed in river water bodies (Barbosa *et al.* 2012; Zhu *et al.* 2019). To tackle these issues, the application of concepts such as low-impact development and water-sensitive urban design, and their associated technologies, has been proposed internationally (Baek *et al.* 2015; Nguyen *et al.* 2019). In addition, the concept of 'sponge cities' has been proposed in China (Jia *et al.* 2022). These measures aim to dissipate urban stormwater runoff and mitigate the negative hydrological impacts of urbanization. However, it should be noted that stormwater harvesting is also an effective way to alleviate urban water scarcity, urban waterlogging and runoff pollution.

Urban road runoff monitoring has been carried out since the 1970s in many countries. Shinya *et al.* (2003) explored the relationship between road runoff quality and traffic intensity, storm intensity, and drought duration. Research on road runoff pollution in China began relatively recently, but in the past few decades, a succession of experiments has been carried out in cities such as Beijing and Xi'an (Ren *et al.* 2008). Due to the high traffic intensity in cities in China, the concentration of pollutants in urban road runoff is relatively high. Wang *et al.* (2022) summarized the urban road runoff water quality in cities in China, and typical concentration ranges for total suspended solids (TSS), chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP), were reported as follows: TSS: 77–1,347.9 mg/L; COD: 31.4–488.1 mg/L; TN: 0.81–8.46 mg/L; TP: 0.139–1.930 mg/L.

Pervious concrete pavement (PCP), a form of grey infrastructure, is one of the main measures in urban stormwater runoff control. In addition to reducing urban rainfall runoff, it also can be effective as a method of water quality treatment. The main PCP-based runoff pollutant removal mechanisms are filtration, adsorption, precipitation and biodegradation (Braswell *et al.* 2018). The removal rates of suspended solids (SS), COD, TN, and TP by PCP have been reported as 89.3–97.9%, 32.8–39.52%, 12.1–69.0% and 51.0–84.4%, respectively, and the corresponding effluent concentrations as 8.7–46.2 mg/L, 253.6–377.6 mg/L, 0.13–17.6 mg/L and 0.02–1.05 mg/L, respectively (Drake *et al.* 2014; Roseen *et al.* 2014; Li *et al.* 2017; Zhang *et al.* 2018; Liu *et al.* 2020). In China, the Technical Code for Rainwater Control and Utilization in Buildings and Residential Areas (GB 50400-2016) requires SS and COD values of 10 and 30 mg/L, respectively, for rainwater reuse, and the Technical Specification for Rainwater Utilization Engineering (DB32/T 3813-2020) further requires TN and TP values of 5.0 and 0.2 mg/L, respectively. These research findings show that PCP can be effective in the treatment of runoff pollutants, but its runoff effluent quality values do not meet the standards required for on-site reuse. In addition, with the increasingly widespread use of PCP, a rapid decline in its permeability due to clogging is a common concern (Drake & Bradford 2013; Roseen *et al.* 2014). Recent studies have found that, when performed in a timely manner, maintenance measures such as pressure flushing can, to some extent, restore the permeability of PCP and extend its lifespan (Zhao *et al.* 2020).

Constructed wetland (CW) is an effective and increasingly widely used technology for road runoff pollution control. Alihan *et al.* (2017) monitored the pollutant removal efficiency for 42 rainfall events in two campus-based CWs. It was found that the removal rates of TSS, TN and TP by CW were 63–79%, 38–54% and 58–74%, respectively. Bang *et al.* (2019) further investigated the effect of hydraulic loading (HL) on pollutant removal by CW. Their results showed that the removal of SS and organic matter by CW was less affected by HL at values of 750–1,500 L/(m²d). Insufficient dissolved oxygen (DO) from biological metabolism is one of the main factors limiting the removal of pollutants (such as organic matter) via aerobic processes in traditional CWs (Saeed *et al.* 2020). Several studies have been carried out to increase DO by micro-aeration of CW. The results show that the removal rate of TN by aerated CW is better than that of unaerated CW (Dong *et al.* 2020). However, CW is prone to clogging after long-term operation. There are many causes of clogging in CW – SS clogging, biofilm clogging and clogging due to improper operation – the most common of which is SS. Sacco *et al.* (2021) found that stormwater has a high sediment load and low organic matter and nutrient content compared to municipal wastewater, making CW highly susceptible to clogging when treating stormwater runoff. Pre-treatment facilities such as sedimentation tanks and coarse filters are generally used to remove SS and avoid clogging in CW (Sultana *et al.* 2015; Ávila *et al.* 2016). The effective SS removal performance of PCP makes it suitable as a pre-treatment system for CWs.

As stated above, although PCP is somewhat effective as a runoff pollution treatment, rainwater reuse standards cannot be met using PCP alone. CW, a form of green infrastructure, can further enhance stormwater runoff water quality treatment, but blockages often occur. Based on the concept of combining grey and green infrastructure, this study combines PCP with CW to explore the feasibility of stormwater runoff pollution treatment and on-site reuse using a combined PCP-CW system.

This study was conducted with the following specific objectives: (1) to evaluate the removal efficiency of runoff pollutants (SS, COD, TN, TP) via PCP; (2) to determine the efficiency with which the combined PCP-CW system removes runoff pollutants; (3) to optimize the pollutant treatment efficiency of the combined PCP-CW system and analyze the mechanism through which it functions. The research findings presented here detail the efficiency and optimization of the treatment of polluted stormwater runoff using a PCP-CW system, and provide a feasible approach to the realization of on-site stormwater reuse.

2. MATERIALS AND METHODS

2.1. Configuration of the combined PCP-CW system

As shown in Figure 1, the PCP-CW test apparatus consisted of a raw stormwater feed tank, a PCP column, a baffled CW apparatus. Synthetic stormwater was stored in a 50-L feed tank. A submersible pump was used to simulate rainfall at the surface of the PCP. The intensity of rainfall was controlled by flowmeters and flow-regulating valves. The effluent from the PCP column was collected using an intermediate transfer tank and then distributed to the CW using a peristaltic pump.

2.1.1. PCP test columns

The PCP test columns were made from 200-mm diameter PVC piping with a column height of 45 cm. The test column was filled from bottom to top with 10 cm of gravel with a 10–20 mm particle size, 10 cm of gravel with a 5–10 mm particle size, and 10 cm of PCP surface layer. The inner wall of the pipe was lined with a geotextile to prevent short-cut flow at the side wall. The overflow port was placed 5 cm above the PCP surface layer, sampling ports #1–#3 were placed 3 cm above the bottom of the surface layer and each of the gravel layers, and an effluent outlet was placed at the bottom of the lower gravel layer.

2.1.2. Preparation of the PCP surface layer

2–5 mm granite aggregate was used as the aggregate and ordinary P. O 42.5 grade silicate cement was used as the binder. An aggregate/cement ratio of 3.7:1 and a cement/water ratio of 3.3:1 by weight were used, and a polycarboxylate water reducer of 0.3% by weight relative to the cement was added to improve the compatibility of the mixture. The specific steps for preparing the PCP surface layer were as follows: First, the aggregate and 50% of the required water were poured into a rotary drum mixer and mixed rapidly for 30 seconds; then, the predetermined amounts of cement, polycarboxylate water reducer, and the remaining 50% of the water were added to the mixer and mixed for another 2 min. The concrete mixture was poured into the PCP column on top of the pre-set gravel layers. After smoothing with a metal spatula, the mixture was compacted with a metal block to ensure uniform compaction. The PCP blocks were cured for 3 days at 20 °C and >95% relative humidity. The PCP blocks were then covered with plastic sheeting and held for an additional 21 days.

2.1.3. Baffled CW apparatus

A horizontal submerged CW with a length × width × height configuration of 1.2 m × 0.6 m × 0.6 m was used. Three baffles with a height of 35 cm were placed in the CW to divide the configuration into four 30-cm-wide zones (Figure 1). The CW showed an overall horizontal subsurface flow with vertical flow in each of the baffled zones. The substrate depth (i.e., the

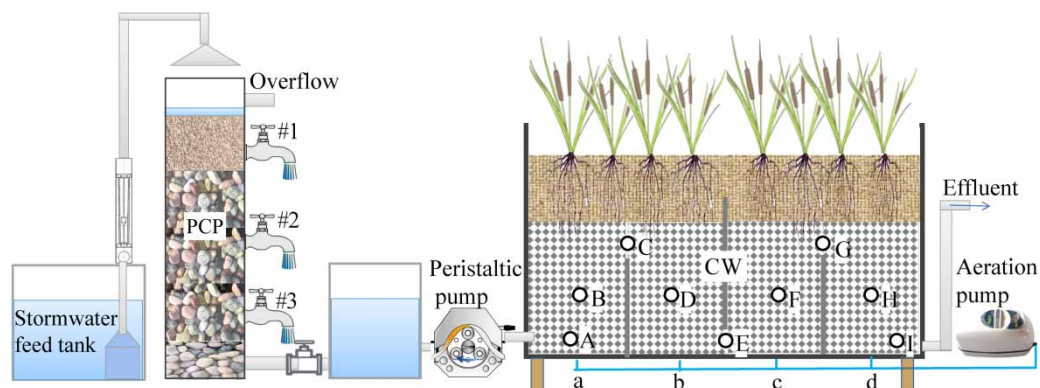


Figure 1 | Schematic diagram representing the combined PCP-CW system.

5–10 mm diameter gravel layer) was 40 cm, with a 20 cm coarse sand layer (2–3 mm diameter quartz sand) on top, to support the plants. The *Phragmites australis* plants, with a height of 0.8–1.0 m, were taken from a campus pond and were planted in the CW at a density of 40 plants/m². The CW apparatus was equipped with one inlet, nine sampling ports (labelled A–I, at 5, 15, and 30 cm from the bottom of each individual zone) and one outlet port (Figure 1). The aeration pipe was pre-installed at the bottom of the CW gravel layer, and the aeration rate was controlled and adjusted by an air pump.

2.2. Synthetic stormwater

The water quality of stormwater runoff varies widely among regions, and studies have shown that the general concentration ranges of SS, COD, TN, and TP in stormwater runoff are 31–1,020 mg/L, 18–276 mg/L, 3.32–27.13 mg/L, and 0.25–1.44 mg/L, respectively (Niu *et al.* 2016; Hu *et al.* 2020; Zheng *et al.* 2021; Rong *et al.* 2022). To investigate the effectiveness with which the combined PCP-CW system removes runoff pollutants under adverse conditions, the pollutant concentrations in the raw water used in this test are shown in Table 1.

2.3. Methodology

2.3.1. Start-up of the PCP column

The PCP experimental columns were inoculated with water from a campus lake. A continuous feed was maintained at a flow rate of 15.0 mL/min, and 5–10 g of substrate was collected from the base layer every 48 h to determine the oxygen uptake rate (OUR) of the microorganisms. A stable OUR value indicated that the microbial inoculation of the PCP device was successful.

2.3.2. Experimental scheme

2.3.2.1. Runoff pollutant removal performance of PCP. Using the city of Nanjing, China as the reference area, rainfall recurrence intervals of 2a, 5a, 10a and 20a were selected for this study; according to the local rainfall intensity formula, the corresponding rainfall intensities are $I = 0.824, 1.044, 1.209, \text{ and } 1.375$ mm/min, respectively. Based on these values, the stormwater inflow rates corresponding to the area of the PCP column can be calculated to be 1.533, 1.941, 2.250, and 2.558 L/h, respectively. 100 ml water samples were taken every 10 min at sampling ports #1–#3 to detect the changes in pollutant concentrations and to determine the corresponding removal efficiency. The simulated rainfall lasted for 120 min at each rainfall intensity, and the experiment was repeated three times for each rainfall intensity.

2.3.2.2. Runoff pollutant removal performance of the PCP-CW system. Synthetic stormwater runoff was simulated at the surface of the PCP, and the effluent from the PCP column was connected to the inlet of the CW system via a hose. The PCP-purified effluent entered the CW for further treatment, forming a combined PCP-CW system. The influent HL of the CW was set to $0.2 \text{ m}^3/(\text{m}^2\text{d})$, $0.4 \text{ m}^3/(\text{m}^2\text{d})$ and $0.8 \text{ m}^3/(\text{m}^2\text{d})$, and the corresponding hydraulic retention times (HRTs) were 19.2, 9.6, and 4.8 h, respectively. During the experiments, the room temperature was 20 ± 4 °C, and each hydraulic load was continuously fed for 14 days. Every 24 h, 100 mL of water was collected at sampling ports A–I to determine the concentrations of COD, TN, and TP.

2.3.2.3. Optimization of runoff pollution removal efficiency of the PCP-CW system. During the test, the average climate temperature was 25–30 °C, the DO in the influent was 0.73–1.12 mg/L, and the CW system was continuously operated at a hydraulic load of $0.8 \text{ m}^3/(\text{m}^2\text{d})$. Aeration was performed at different positions along the CW (a, b, c, d, a + b, a + c, a + d and b + c) to compare the influence of aeration position on the pollutant removal rate. The gas/water ratio was 5:1 and

Table 1 | Synthetic stormwater water quality

Indexes	Concentration (mg/L)	Reagent
SS	800 ± 164.22	Mud and sand
COD	200 ± 15.32	C ₆ H ₁₂ O ₆
TN	10 ± 2.88	NH ₄ Cl
TP	1 ± 0.025	KH ₂ PO ₄

the aeration rate was 1,500 mL/min. Every 5 h, 100 mL of water samples were collected from sampling ports A–I, and the concentrations of DO, COD, TN, and TP in the water samples were measured.

2.3.2.4. Treatment performance of the combined PCP-CW system for actual stormwater runoff. Actual stormwater runoff was collected from the campus stormwater drainage system during three rainfall events over the period June to September 2022. Samples were collected directly from the inspection shaft of the stormwater drainage system in several prepared 50 L plastic containers, which were washed with a 3% (V/V) nitric acid solution prior to every use. The collected stormwater was immediately taken to the laboratory and mixed in a settling tank. After allowing the stormwater to settle for 1 h to lower the concentrations of impurities and larger particles in the water and reduce the risk of clogging in the PCP and CW, the supernatant was transferred to the feed tank for the combined PCP-CW system. The PCP column was tested under a rainfall intensity of 1.209 mm/min, corresponding to a rainfall recurrence interval of 10a, while the CW system was continuously operated, without aeration, at an HL of 0.8 m³/(m²d). Water samples were collected from PCP sampling ports #1–#3 and CW sampling ports A–I, and the concentrations of SS, COD, TN, and TP were measured for the collected (i.e., actual/non-synthetic) stormwater and the corresponding water samples from all PCP and CW sampling ports.

2.4. Sample analysis

Once the water samples were collected, the pH and temperature were measured *in situ* using a pH meter (PHS-3C, China) and a mercury thermometer, respectively. DO was measured using a portable DO analyzer (Raycom DO-957, Shanghai, China). SS was quantified by filtering water samples through pre-weighed glass-fiber filters through a membrane with a pore size 0.45 µm and drying at 110 °C. COD was analyzed by the closed reflux titrimetric method. TN was determined by the alkaline potassium persulfate oxidation – persulfate method, while TP was analyzed by the potassium persulfate oxidation – ammonium molybdate spectrophotometry method.

The OUR was determined via the closed intermittent aeration method. The water sample or substrate sample was placed in a closed container and aerated until the DO was saturated, the DO measurement probe was inserted into the container, and the container was sealed. The container was placed on a magnetic stirrer. The magnetic stirrer was set at a speed of 50 rpm at a temperature of 20 ± 2 °C, and the DO concentration in the container was recorded at 1-min intervals for 10 min. The DO-t curve can be obtained, and OUR was calculated as the slope of the straight line (i.e., Equation (1)):

$$\text{OUR} = \frac{\text{DO}_1 - \text{DO}_2}{\Delta t} \quad (1)$$

where DO₁ is the concentration following aeration (mg/L), DO₂ is the concentration after standing for 10 min (mg/L), and Δt is the standing time after the aeration of water samples (10 min in this study).

3. RESULTS AND DISCUSSION

3.1. Runoff pollutant removal performance of PCP

The pollutant concentrations for samples collected from ports #1–#3 of the PCP column under different rainfall recurrence intervals are shown in Table 2. The removal rates of SS, COD, TN and TP showed a decreasing trend with increasing rainfall intensity and HL. The removal rates of COD, TN and TP by PCP were 46.36–55.86%, 25.83–35.23% and 69.16–75.32%, respectively, which do not meet the standards required for rainwater reuse. This is mainly due to the fact that the removal of COD, TN, and TP depends largely on microbial action. During the rainfall period, simulated rainfall passed through the PCP column for a short period of time – about 6–14 min in this study, depending on rainfall intensity and the permeability coefficient of the PCP – and it is difficult for microorganisms to achieve effective removal of COD, TN and TP with such a short residence time; further treatment is therefore needed to meet the standard for rainwater reuse.

The removal rate of SS by PCP showed a limited decrease of 3–8% with the increases in rainfall recurrence interval. Increases in rainfall recurrence interval and HL accelerate the rate of water flow through the pores of the PCP surface layer, resulting in a decrease in the SS removal rate; however, the SS removal rate can reach 96.61–99.20%, such that the effluent meets the standard required for rainwater reuse. Our results indicate that the surface layer accounted for 53.87–59.33% of the SS removal; generally, the smaller the pore, the more significant the interception effect. SS concentration in the effluent reaches the reuse requirement of 10 mg/L under a rainfall recurrence interval of less than 5 years.

Table 2 | Effect of PCP on pollutant removal under different rainfall recurrence intervals

Item	Influent concentration (mg/L)	Sampling point	Rainfall recurrence interval								RS ^b (mg/L)
			2a		5a		10a		20a		
			RE ^a (%)	Concentration (mg/L)	RE ^a (%)	Concentration (mg/L)	RE ^a (%)	Concentration (mg/L)	RE ^a (%)	Concentration (mg/L)	
SS	764–822 (Mean = 800)	#1	59.33 ± 1.14	325.36 ± 9.13	57.88 ± 1.34	336.96 ± 10.75	56.49 ± 1.56	348.08 ± 12.49	53.87 ± 1.44	369.04 ± 11.5	≤ 10
		#2	96.12 ± 0.8	31.04 ± 6.41	94.37 ± 1.02	45.04 ± 8.19	94.37 ± 0.9	45.04 ± 7.19	89.70 ± 1.11	82.40 ± 8.84	
		#3	99.20 ± 0.18	6.40 ± 1.44	98.75 ± 0.23	10.00 ± 1.86	97.56 ± 0.25	19.52 ± 1.96	96.61 ± 0.22	27.12 ± 1.80	
COD	187–206 (Mean = 200)	#1	40.07 ± 1.83	119.86 ± 3.66	39.97 ± 1.35	120.06 ± 2.7	36.12 ± 2.47	127.76 ± 4.94	32.98 ± 2.51	134.04 ± 5.02	≤ 30
		#2	51.56 ± 2.32	96.88 ± 4.65	49.99 ± 1.57	100.02 ± 3.13	47.58 ± 1.16	104.84 ± 2.33	41.27 ± 1.43	117.46 ± 2.85	
		#3	55.86 ± 1.36	88.28 ± 2.72	53.80 ± 0.83	92.40 ± 1.65	51.45 ± 1.43	97.10 ± 2.87	46.36 ± 1.04	107.28 ± 2.07	
TN	13–16 (Mean = 15)	#1	23.35 ± 2.73	11.50 ± 0.41	21.96 ± 2.33	11.71 ± 0.35	19.61 ± 3.33	12.06 ± 0.50	15.60 ± 2.47	12.66 ± 0.37	≤ 5.0
		#2	30.20 ± 3.07	10.47 ± 0.46	29.50 ± 3.80	10.58 ± 0.57	26.88 ± 1.93	10.97 ± 0.29	23.03 ± 1.73	11.55 ± 0.26	
		#3	35.23 ± 2.60	9.72 ± 0.39	33.67 ± 1.73	9.95 ± 0.26	30.63 ± 2.80	10.41 ± 0.42	25.83 ± 1.40	11.13 ± 0.21	
TP	0.9–1.1 (Mean = 1.0)	#1	62.13 ± 1.67	0.38 ± 0.02	60.29 ± 1.47	0.40 ± 0.01	58.01 ± 2.36	0.42 ± 0.02	55.21 ± 1.88	0.45 ± 0.02	≤ 0.2
		#2	72.58 ± 1.01	0.27 ± 0.01	69.54 ± 2.49	0.30 ± 0.02	66.86 ± 1.16	0.33 ± 0.01	65.14 ± 1.49	0.35 ± 0.01	
		#3	75.32 ± 1.86	0.25 ± 0.02	72.68 ± 1.33	0.27 ± 0.01	70.41 ± 2.20	0.30 ± 0.02	69.16 ± 1.13	0.31 ± 0.01	

Note: The effluent concentration and removal efficiency values are expressed as mean ± SD ($n = 3$); RE, removal efficiency; RS, concentration for reuse standard.

^aWater samples were taken every 10 min at sampling ports #1–#3 to measure the changes in pollutant concentrations and determine the corresponding removal efficiency.

^bThe indexes of SS and COD in rainwater reuse standard refer to GB 50400-2016, the indexes of TN and TP in rainwater reuse standard refer to DB32/T 3813-2020.

Similar to the SS removal results, the surface layer was found to account for 71.14–71.73% of the total COD removal. The abundant micropores in the PCP surface layer can provide useful adsorption sites for COD removal (Li *et al.* 2017). As stormwater runoff passes through PCP for only a short time, microorganisms play a limited role in COD removal (Niu *et al.* 2016). The effluent COD concentration did not reach the reuse requirement of 30 mg/L under different rainfall recurrence intervals, and further treatment is needed.

The overall TN removal rate was low and decreased further with increases in rainfall recurrence interval and rainfall intensity; however, under conditions of low rainfall intensity, increased HRT is conducive to the nitrification and denitrification processes involved in biological nitrogen removal (Kuruppu *et al.* 2019). In general, the effluent TN concentration ranged from 6.48 to 7.42 mg/L and did not reach the reuse requirement of 2.0 mg/L.

The contribution of the surface layer to TP removal was 79.8–82.4%, which may be attributed to the interception of granular phosphorus by the PCP surface layer; additionally, the silicate cement in the surface layer of PCP can effectively adsorb phosphate in water, resulting in reduced TP concentrations (Grzmił & Wronkowski 2006; Wu *et al.* 2021; Yu *et al.* 2021). Some studies have also shown that PCP increases the pH of stormwater, which makes it easier for orthophosphate to react with metal cations in granite gravel and cement, which are the raw materials of the permeable concrete layer, leading to phosphate precipitation (Luck *et al.* 2008; Eck *et al.* 2012; Brown & Borst 2015; Yu *et al.* 2021). TP concentration in the effluent ranged from 0.25 to 0.31 mg/L under different rainfall recurrence intervals, which does not meet the reuse standard of 0.2 mg/L; further treatment is therefore required.

3.2. Performance of CW and PCP-CW in runoff pollutant removal

As shown in Figure 2(a), the COD removal rate by the CW decreased with increases in HL. Under HL rates of 0.2–0.8 m³/ (m²·d), the COD removal rate by the CW ranged from 48.02 to 75.04%, and the corresponding effluent concentration ranged from 24.25 to 50.5 mg/L. The observed trend in the variation of COD concentrations was fairly consistent for all of the HL

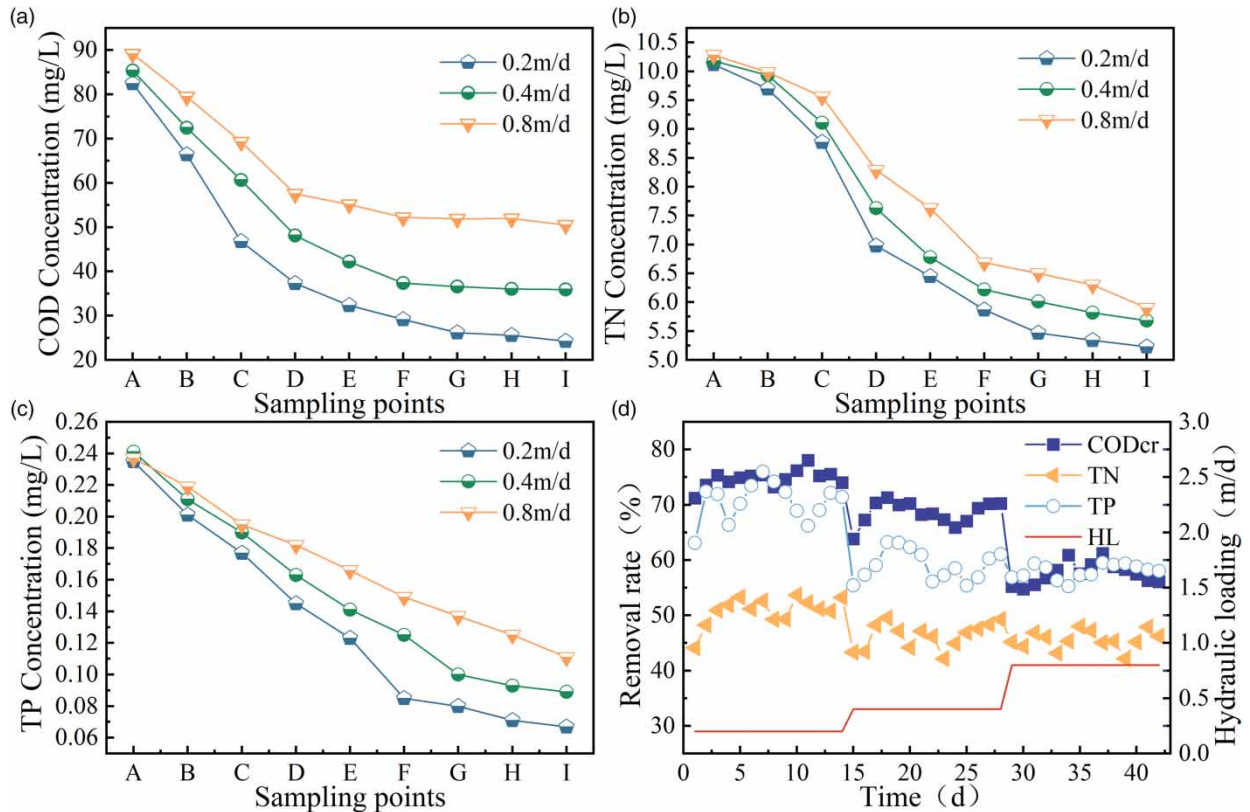


Figure 2 | Variation in pollutant concentrations and removal rates by CW under different hydraulic loadings: (a) COD concentrations; (b) TN concentrations; (c) TP concentrations; and (d) removal rates.

rates assessed in this study: as HL increased, the COD concentrations measured at sampling ports A–D in the front section of the CW decreased markedly and then stabilized, consistent with the findings reported by Akrotos & Tsihrintzis (2007), who observed in their study that the COD concentration decreased rapidly in the front-third section of the CW apparatus. This is mainly due to the rich nutrient supply and the large numbers of microorganisms present in the front section of the CW, which together result in effective COD removal (Alemu *et al.* 2018). The decrease in COD removal rate with increasing HL is mainly due to the decrease in HRT, which affects the substrate adsorption and microbial degradation time of organic matter (Ewemoje *et al.* 2015; Abou-Elela *et al.* 2017). Under the HL of $0.2 \text{ m}^3/(\text{m}^2\cdot\text{d})$, the effluent COD concentration of the combined PCP-CW system reached the reuse requirement of 30 mg/L.

It can be seen from Figure 2(b) that the TN removal rate decreased with the increase in HL. The removal of TN by the CW was mainly a result of nitrification and denitrification processes (Reddy 1983; Maltais-Landry *et al.* 2009; Alemu *et al.* 2018), the high HL shortened the HRT, resulting in a decrease in the TN removal rate (Trang *et al.* 2010; Li *et al.* 2012). The removal rate of TN by the CW was 46.97–53.03%, but the effluent TN concentration, under different HL rates, did not meet the reuse requirement of 5 mg/L. Further optimization of CW is needed to improve TN removal.

As shown in Figure 2(c), the TP removal rate for the CW ranged from 58.89 to 75.19%, depending on the HL rate. Phosphorus removal in CW systems mainly occurs via substrate adsorption, microbial assimilation and plant uptake, but plant uptake accounts for less than 10% of the phosphorus removal (Shen *et al.* 2019; Yang *et al.* 2019). High HL rates shortened the time of adsorption and microbial assimilation processes, which resulted in lower TP removal rates. The effluent TP concentration for the combined PCP-CW system achieved the reuse standard of 0.2 mg/L under several different HL rates, outperforming the TP removal reported by Carrillo *et al.* (2022). This enhanced performance may be attributed to the use of a folded flow continuous wave in this study, which enables a greater degree of contact and a more thorough interaction between phosphorus, plant roots and substrate.

In general, the overall removal rates of SS, COD, TN, and TP by the combined PCP-CW system reached 96.61–99.5%, 74.75–87.88%, 60.65–65.15%, and 88.90–93.30%, respectively. Our results indicate that the primary roles of PCP are the interception and adsorption of particulate matter, the reduction of SS and TP in stormwater runoff, and the prevention of subsequent blockage of the CW. The CW is then able to further improve the system's removal of COD, TN, and TP.

3.3. Effect of aeration on PCP-CW runoff pollutant removal and analysis of the mechanism of action

Figure 3(a) shows the variation of DO concentration along the CW after micro-aeration at different positions (a, b, c, d, a + b, a + c, a + d, b + c). The DO concentration in the CW ranged from 0.11 to 0.82 mg/L under unaerated conditions, and from 0.22 to 2.85 mg/L at different sampling ports under aeration at a rate of 1,500 mL/min.

Under the single-position aeration condition (a, b, c, d), the average effluent DO concentration at sampling port I was lower than that under the multi-position aeration condition (a + b, a + c, a + d, b + c). For single-position aeration, aeration in the front section of the CW was conducive to maintaining good aerobic conditions in the front section and reasonably anaerobic conditions in the back section, which are conducive to nitrification and denitrification processes, improving the TN removal rate—for example, the lowest effluent DO concentration at sampling port I was 0.18 mg/L under the aeration condition at position a. In this study, the baffled subsurface flow used in the CW was implemented to increase the overall flow distance of the water, to maximize the contact between the pollutants, plant roots, and substrates. In addition, the baffled structure is able to change the spatial distribution of DO in the wetland and improve the removal efficiency of organic matter, nitrogen and phosphorus.

The variation in COD concentration at different aeration positions is shown in Figure 3(b). Multi-position aeration at the front and middle sections (a + b and a + c) of the wetland was more beneficial for COD removal than single-position aeration. Under the condition of multi-position aeration at a + c, an average COD removal rate of 89.95% was achieved, meeting the reuse standard for the effluent COD concentration. The COD removal in the CW mainly occurs via microbial metabolism and substrate adsorption (Li *et al.* 2014). In this study, the change in aeration position mainly affected the microbial metabolism function; when the aeration position was a + c, the average DO concentration in the CW system was 1.87 mg/L, which was 1.36 mg/L higher than that under the same HL without aeration and is conducive to the growth of aerobic microorganisms and improved COD degradation efficiency.

The variation of TN concentration along different aeration positions is shown in Figure 3(c). Aeration at the front and middle sections (a and a + b) of the wetland, especially the joint aeration at a + b, was more conducive to TN removal.

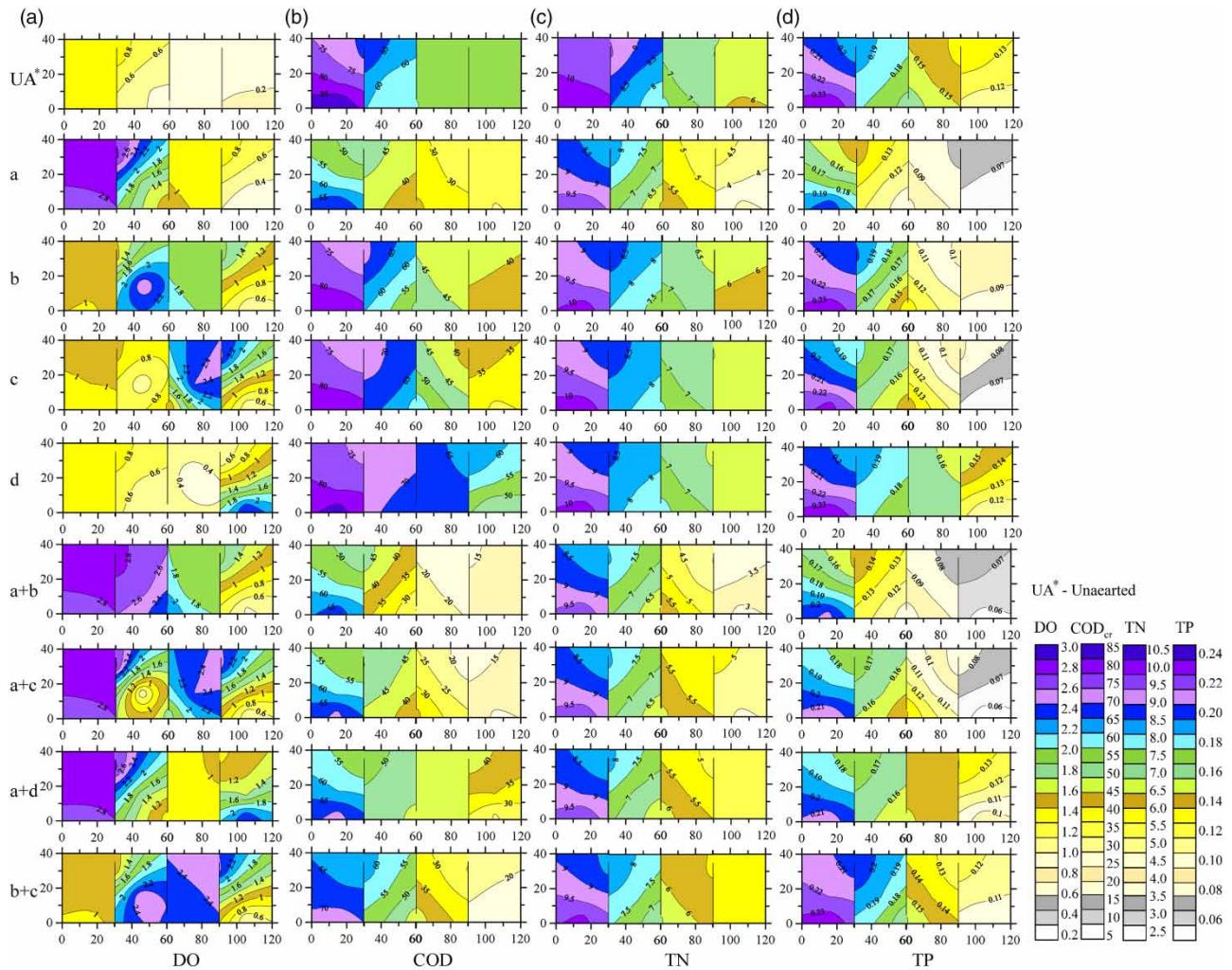


Figure 3 | Variation of pollutant concentrations in CW under different aeration position conditions: (a) DO; (b) COD; (c) TN; and (d) TP.

The average TN removal rate was 73.76%, which was 26.79% higher than the average removal rate under the same HL without aeration. The effluent concentration of TN was less than 5.0 mg/L, which meets the standard required for reuse.

Under the condition of single-position aeration at a, the maximum removal rate of TN reached 69.31%, and the removal effect was superior to that of other single-position aeration conditions. For multi-position aeration, aeration at a + b was best for TN removal. This is mainly due to the fact that joint aeration at a + b ensures a high DO concentration in the front section of the CW, promoting nitrification, and maintains a low DO concentration in the back section, promoting denitrification; this balance is crucial for the nitrogen removal process. For multi-position aeration at a + b, the average DO concentration in the front section of the CW system is 1.95 mg/L higher, at the same HL, than without aeration, which is beneficial to nitrification and nitrate accumulation, while the average DO concentration in the back section, at the same HL, was only 0.28 mg/L higher than without aeration, which had a limited effect on denitrification. In general, it was beneficial to enhance the denitrification process (Maltais-Landry *et al.* 2009). Aeration at b, c and d will increase the DO concentration in the back section, which will enhance the denitrification process and reduce the TN removal rate.

As shown in Figure 3(d), good TP removal efficiencies were achieved when aeration was applied in the front and middle sections of the wetland (a, c and a + b). The average TP removal efficiency reached 79.21%, which was 20.32% higher than that at the same HL without aeration. The TP concentration in the effluent ranged from 0.06 to 0.11 mg/L; at these values, the standard of 0.2 mg/L required for reuse is met. Considering the baffled CW structure used in this study, the DO is relatively low at the bottom of the CW and relatively high near the surface within each compartment. In addition, aeration at different

locations also creates an alternating aerobic and anaerobic environment within the CW, which may promote the uptake and release of phosphorus by microorganisms and improve TP removal efficiency (Luo *et al.* 2017). In addition, *Phragmites australis* were planted in the CW apparatus in this study, and its rich root system may promote the absorption of phosphorus by plants and improve the efficiency of TP removal (Huett *et al.* 2005).

In summary, changing the aeration position can affect the spatial distribution of DO in CWs, thereby affecting pollutant removal, especially organic matter and nitrogen removal (Wang *et al.* 2015). The multi-position aeration at the front and middle sections of the CW improved the removal of runoff pollutants in the enhanced wetland. In addition, the removal of COD and TP was enhanced due to the high nutrient availability and rapid proliferation of microorganisms. With the consumption of DO by microorganisms, the back section of the CW is in an anoxic-anaerobic state, which is beneficial to the growth of denitrifying bacteria and enhances nitrogen removal. By optimizing the location of the micro-aeration in the CW, enhanced removal of organic matter, nitrogen, and phosphorus in the wetland was achieved, and the effluent fully met the rainwater reuse standard.

3.4. Actual stormwater runoff treatment performance of the combined PCP-CW system

As shown in Figure 4, the removal rates of SS, COD, TN, and TP of actual stormwater runoff achieved by the combined PCP-CW system reached 99.29–99.58%, 79.89–83.73%, 56.25–60.48%, and 88.83–91.84%, respectively. In general, the pollutant removal efficiency for actual stormwater was close to that for synthetic stormwater for SS, COD, and TP, but the TN removal rate for actual stormwater was lower than that for synthetic stormwater. This is mainly because the TN in actual stormwater contains complex components such as particulate nitrogen and organic nitrogen, leading to a decreased removal rate.

Although there were substantial differences in the influent SS concentrations for the three rainfall events, the SS concentrations in the effluent all reached the standard required for reuse with limited fluctuations, indicating that PCP-CW effectively intercepts particulate matter. The effluent concentrations of COD, TN, and TP were 20.01–49.68 mg/L,

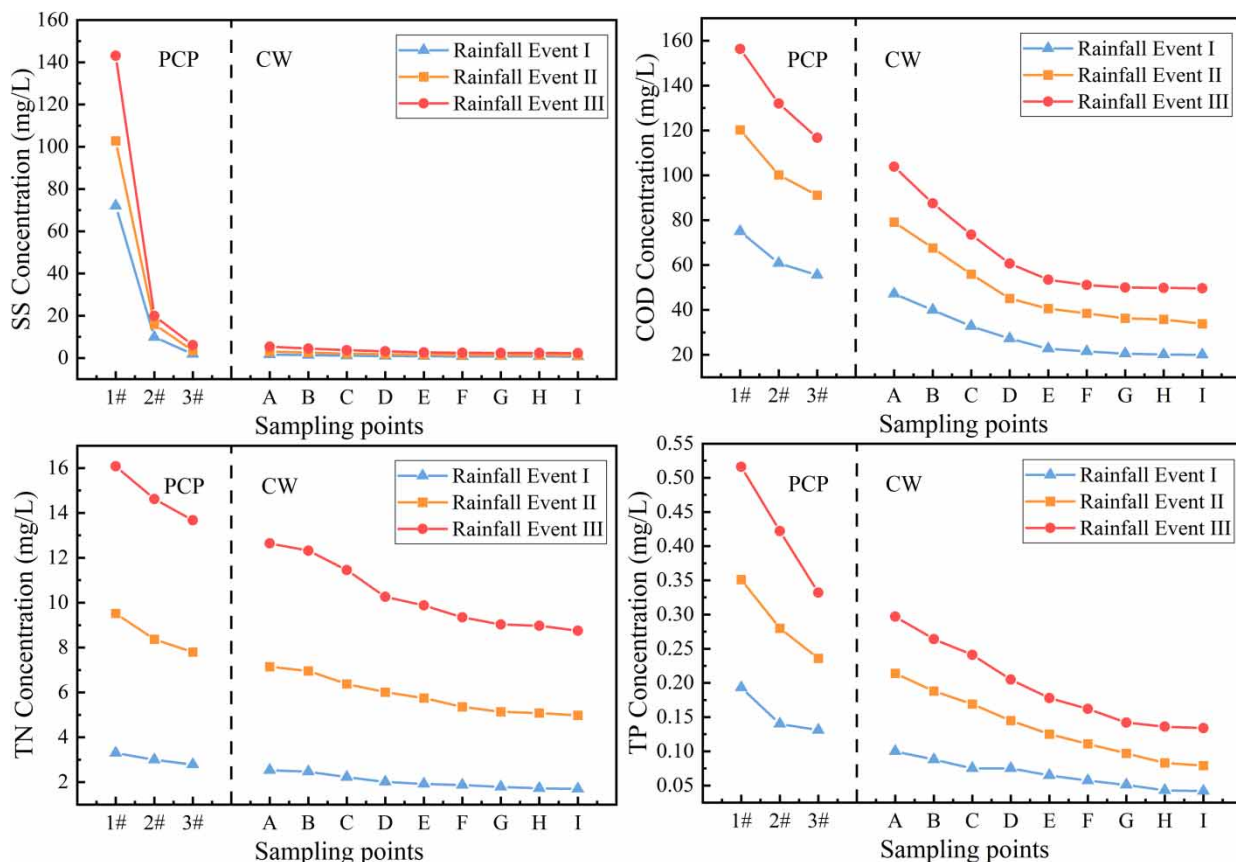


Figure 4 | Pollutant removal efficiencies of the PCP-CW system for actual stormwater treatment.

1.70–8.75 mg/L, and 0.04–0.13 mg/L, respectively; essentially, the reuse standards were met under rainfall events I and II, for which influent concentration was low, but the COD and TN concentration reuse requirements were not met under rainfall event III, for which the influent concentration was high. Therefore, the application of aeration optimization and other measures in engineering applications requires further exploration.

4. CONCLUSION

In order to realize the in situ reuse of stormwater runoff, the treatment efficiency of the combined PCP-CW system and the effects of optimization measures were studied. The main conclusions are as follows:

- (1) The pollutant removal efficiency of PCP decreased with increasing rainfall intensity. PCP achieved a removal rate of 96.61–99.20% for SS, which met the rainwater reuse standard, while the effluent COD and TN did not meet the reuse standard and required further treatment. The PCP surface layer made the most substantial contribution to the pollutant removal – the gravel particles in the surface layer and base layer adsorbed and intercepted SS and particulate organic matter.
- (2) Under different HL rates and HRT conditions, the removal rates of COD, TN and TP by the CW were 48.45–75.12%, 47.26–53.05% and 59.04–75.28%, respectively. The removal efficiency of CW decreased with increases in HL. In general, the overall removal rates of SS, COD, TN, and TP by the combined PCP-CW system reached 96.61–99.5%, 74.75–87.88%, 60.65–65.15%, and 88.90–93.30%, respectively. In addition to the adsorption and interception of SS and particulate organic matter, PCP also prevents subsequent blockage of the CW. The CW is then able to further enhance the removal of COD, TN, and TP.
- (3) By optimizing the aeration position, the spatial distribution of DO and the removal efficiency of pollutants in the CW can be improved. The results from the comparison of different aeration positions show that the optimal aeration effect was achieved in the front section of the CW, with a TN removal efficiency of 73.76%, which is 26.5% higher than the average removal rate without aeration. Aeration in the front section can increase the DO level, promote the rapid proliferation of microorganisms, improve the removal efficiency of COD and TP, and facilitate the nitrification process. With the consumption of DO by microorganisms, the back section of the CW is in an anoxic-anaerobic state, which is beneficial to the growth of denitrifying bacteria and enhances the removal of nitrogen. By optimizing the micro-aeration positions in the CW, enhanced removal of organic matter, nitrogen and phosphorus in the CW was achieved, and the effluent fully met the rainwater reuse standard. This combination of grey and green infrastructure thus provides a feasible approach to the purification of stormwater runoff and the realization of on-site stormwater reuse.

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AUTHOR'S CONTRIBUTION

J.Z. supervised the study, did data curation, did formal analysis, wrote, reviewed, and edited the article. L.S. did experimental work, wrote the original draft. M.W. did experimental work and data curation. J.H. performed the methodology and did data curation. J.T. did experimental work. All authors read and approved the final manuscript.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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