Evaluation of modified permeable pavement systems with coal gangue to remove typical runoff pollutants under simulated rainfall

Xiaoran Zhang, Yiran Tian, Junfeng Liu, Yinrui Wang, Ziyang Zhang and Haiyan Li

ABSTRACT

Coal gangue (CG) as mineral waste was properly treated and applied as the filter media in permeable pavement systems due to its good sorption ability and mechanical strength. Batch experiments show the maximum adsorption capacity of calcined CG to phosphorus could reach 2.63 mg/g. To evaluate the removal effect of typical runoff pollutants including chemical oxygen demand (COD), total nitrogen (TN), total phosphorus (TP) and ammonia (NH_4^+-N), the gravel and sand in a traditional pavement system were replaced by CG respectively. The leaching behavior of the four pollutants in CG modified systems is limited and comparable with traditional system, indicating pretreated CG as filler would not cause environmental risk. CG-based pavement systems improved the removal efficiency of the four pollutants especially for TP. The removal mechanisms including interception, adsorption and microorganism degradation. The removal rates of COD, NH_4^+-N and TN by CG modified and traditional systems decreased with rainfall duration, while it is not obviously changed with rainfall recurrence period and drying period. Overall, the permeable pavement with CG layers that replaced both sand and gravel layers show best removal efficiency of all pollutants investigated especially for TP (>95%). This study provides a new way for CG utilization and gives the reference for the process design of permeable pavement.

Key words | coal gangue, drying period, permeable pavement, rainfall intensity, removal, runoff

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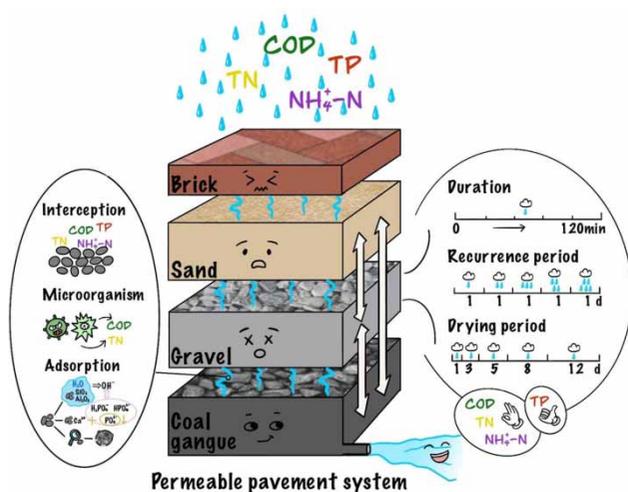Nanjing 211500,
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HIGHLIGHTS

- CG based pavements improved the removal of the pollutants especially for TP (>95%).
- CG replaced sand and gravel layers show best removal efficiency for all pollutants.
- The removal of COD, TN and NH_4^+-N decreases as the rainfall duration increases.
- Rainfall recurrence and drying period effect on pollutants removal is not obvious.
- Interception, adsorption and microorganism degradation could be removal mechanisms

GRAPHICAL ABSTRACT

INTRODUCTION

With rapid urbanization, the area of impermeable surface in the city keeps increasing, which causes an increase of stormwater runoff volume and peak flow rate. In addition, stormwater runoff could contain various pollutants, and the typical pollutants includes chemical oxygen demand (COD), total nitrogen (TN), total phosphorus (TP) and ammonia (NH_4^+-N) (Ostrom & Davis 2019). These pollutants are discharged into rivers, lakes and other water bodies, and not only seriously deteriorate the runoff water quality, but also threaten human health and the ecological system. Low Impact Development (LID) aims to manage rainwater at source and it employs natural processes for urban stormwater management. Construction of the 'Sponge City' in China was proposed in 2012 and follows a similar concept. The main aim is to transform current urban areas into a 'Sponge City' by upgrading the existing urban drainage infrastructure and utilizing natural systems. During the construction, LID measures such as permeable pavements, bioretention and rain gardens were widely applied and the water retention, infiltration and drainage can be improved (Wang *et al.* 2019).

As the cornerstone for LID and sustainable site design, permeable pavements are implemented to reduce urban runoff peak and improve stormwater runoff quality (Weiss *et al.* 2017). According to normal practices, the common permeable pavements are made of infiltration fillers, roadbed and drain pipes. The infiltration fillers are the key parts of the system that includes the surface layer (e.g. permeable

bricks, permeable asphalt or permeable concrete), a basement layer (e.g. graded gravels or hollow concrete, coarse sand or medium sand). When the stormwater runoff flows through the pavement systems, its quantity will be reduced by evaporation and infiltration. At the same time, pollutants in the runoff will also be removed by permeation, interception, filtration and other mechanisms to some extent (Li *et al.* 2017). The International Stormwater Best Management Practice (BMP) database shows that total suspended solids (TSS) and certain total metals could be effectively removed by permeable pavements. However, the removal efficiency for dissolved pollutants such as total nitrogen (TN), total phosphorus (TP), ammonia nitrogen (NH_4^+-N), and chemical oxygen demand (COD) is limited (Ostrom & Davis 2019). Niu *et al.* (2016) evaluated the runoff pollution reduction performance by common permeable systems made of permeable bricks, coarse sand and gravel. Their results indicated that the average removal rates of TP and COD were 71.2% and 24.1% for permeable brick, 54.8% and 9.0% for a 5-cm coarse sand layer, and 72.2% and 26.1% for a 20-cm gravel layer, respectively (Niu *et al.* 2016). Ammonia nitrogen and TN cannot be steadily removed by that system. Since the filler of the permeable pavement system plays an important role in dissolved pollutant removal, efforts were paid to developing the filler of the system. Related research indicated that the surface layer of permeable pavements is more capable of removing pollutants than other layers (Liu *et al.* 2019). This is mainly due

to the initial material retention effect. However, the basement layer is thicker than the surface layer, which allows sufficient time for pollutants to pass through, thus the basement layer filler has greater potential to remove pollutants. Ostrom & Davis (2019) proposed an enhanced stormwater treatment media comprised of expanded shale aggregate, aluminum-based water treatment residual and a psyllium-based binder. A 5-cm layer of media was installed as the basement layer in permeable pavement. The concentration of dissolved phosphorus from effluent of the modified system fell from 0.22 mg/L to 0.05 mg/L (Ostrom & Davis 2019). On the other hand, due to the large space of the basement layer, the quantity of the filling materials is large and their cost should be reduced due to economic reasons. Therefore, to improve the removal efficiency of runoff pollutants in permeable pavement systems, besides improvement of the surface layer, it is also vital to optimize the basement layer with energy efficient and cost-effective materials.

Coal gangue (CG) is generated during coal mining activities. About 15–20% of CG by weight of the total coal output is discharged during its production. CG has become a primary environmental concern around the world (Zhang & Ling 2020). It was reported that 175 million tons of coal waste have been stocked in dumps for decades in Europe. In Spain, 2.2 million tons of coal mining by-products were produced in 2009 (Frías *et al.* 2012). CG in China has accumulated more than 50 billion tons since 2017, and is expected to grow rapidly continually. If they were not properly treated, CG not only occupies a large amount of land, but also causes potential pollution to the surrounding atmosphere, soil and other environments. Therefore, the treatment and comprehensive utilization of CG has gradually attracted people's attention. CG is known for its high ash yield, low heating value, and low carbon content (Zhou *et al.* 2014), with large specific surface area and a certain amount of metal oxide (Jablonska & Siedlecka 2015; Ding *et al.* 2017; Wu *et al.* 2017; Zhou *et al.* 2019). There are numbers of studies reported that CG has great sorption capacity towards pollutants such as phosphorus and ammonium. Ding *et al.* (2017) used newly discharged CG and spontaneous combustion CG as experimental raw materials to perform adsorption experiments towards phosphate solutions and reported the main sorption mechanisms are ion exchange (phosphate and hydroxide). The maximum adsorption capacity of spontaneous combustion CG (7.08 mg/g) towards phosphorus is much larger than newly discharged CG (2.50 mg/g). The main reason is that a high temperature pretreatment process can increase the metal oxide (i.e. SiO_2 and Al_2O_3) content on the surface of CG,

which could enhance the ion exchange capacity between phosphate and CG, thus increasing the adsorption capacity (Ding *et al.* 2017). Zhang *et al.* (2013) found that CG has a certain removal effect on ammonium. When the dosage is 10 g/L and the temperature is 25 °C, the adsorption capacity of CG increased as the concentration of ammonium solution increased. The adsorption capacity could reach 6.0 mg/g at high concentration (Zhang *et al.* 2013). Wu *et al.* (2017) suggested that CG has the potential to be used as landfill liner materials due to its good adsorption performance and low leaching amount of pollutants (Wu *et al.* 2017). Some researchers also suggest that CG can be used as a pavement foundation material due to its hard texture (Xu *et al.* 2016; Zhu *et al.* 2017). Xu *et al.* (2016) use CG as the sole raw material to prepare construction brick. The synthesized brick sintered at 1,200 °C possesses the optimal properties, with water absorption and compressive strength values of 3.65% and 45.61 MPa, respectively. The leaching toxicity of sintered bricks prepared under the optimum condition were below the China environmental quality standards for surface water (class I) prescribed in GB 3838-2002 (Xu *et al.* 2016), which indicated that CG burned at a high temperature could be safely used. In addition, as an industrial solid residue, CG costs much lower than normal filling materials such as gravel and sand. The price of CG varies with its carbon and sulfur content. The CG used in pavement systems requires extremely low carbon and sulfur content, which only cost 50 yuan (5.75 pounds) per ton, while gravel and sand cost 400 yuan (46 pounds) and 600 yuan (69 pounds) per ton, respectively. It is also cheaper than other proposed adsorbents such as activated carbon and zeolite.

According to the above advantage, properly treated CG may be applied as the advanced filling material in a permeable pavement system. Therefore, the present study aimed at evaluation of the performance of modified permeable pavement systems with CG especially in the basement layer to remove typical dissolved runoff pollutants (i.e. COD, NH_4^+-N, TN and TP) under simulated rainfall. Our objectives were (i) to investigate the adsorption behavior of these typical runoff pollutants on CG through adsorption experiments and model fitting; (ii) to measure the leaching and removal effect of the above pollutants by CG-based permeable pavement systems with different structures as compared with a traditional system by cell set-up; (iii) to further study the influence of rainfall duration, recurrence and drying period on the removal behavior towards the above pollutants by the CG-based permeable pavement systems as well as the traditional systems. This study could

not only enhance the removal ability for runoff pollutants of pavement systems, but also provide a new way to utilize the important solid waste CG.

MATERIALS AND METHODS

Preparation of sorbents and filling materials

CG was collected from Datong region, Shanxi province, China. The received CG was first calcinated at 900 °C for 6 h in an incinerator, followed by grinding the particles using a crusher and sifting with sieves to a particle size of 0.075 mm, 0.5–1.0 mm and 5.0–20.0 mm. CG a with particle size of 0.075 mm was used for adsorption batch experiments. CG with a particle size of 0.5–1.0 mm and 5.0–20.0 mm, river sand with particle size of 0.5–1.0 mm, and common gravel for construction with particle size of 5.0–20.0 mm were used as filling materials for box experiments. The permeable cement bricks for the surface layer of pavements were bought from Aidaoaihe (Beijing) Technology Co., Ltd

Sorption of phosphate on CG based on batch experiment

In order to prove the adsorption characteristics of CG for phosphorus, we conducted batch experiments. It should be noted that, in actual applications, the size of CG particles (ranges from 0.5 to 20 mm) as filler are much larger than as sorbents (0.075 mm) in batch sorption experiments. The adsorption capacity should be smaller than that conducted in the batch experiments due to the decrease in specific surface area. However, the results of the batch of experiments are sufficient to provide a theoretical basis for the application of CG as filler in real cases.

Sorption kinetics and isotherms of phosphate on CG were measured by batch experiments. CG used for all batch experiments was 0.075 mm. For the kinetic experiment, 12 g CG was added into a 1,000 mL glass beaker which contained 600 mL phosphate solution (KH_2PO_4) with a concentration of 20 mg/L. The mixture was then put on a magnetic stirrer and the samples were collected at appropriate time intervals (0–150 h). For the isotherm experiment, 0.8 g CG was added into a 50 mL glass centrifuge tube that contained 40 mL phosphate solution with a concentration range between 0.5 and 100 mg/L. The mixture was then shaken in a shaker at a constant temperature (25 °C) for 150 h. The concentration of

phosphate in the supernatant was analyzed. The amount of phosphate adsorbed, q (mg/g), was then calculated using the following equation (Equation (1)).

$$q = \frac{(C_0 - C_e)V}{m} \tag{1}$$

where C_0 (mg/L) is the initial concentration of phosphate, V (L) is the volume of the solution and m (g) is the weight of CG.

The data of adsorption kinetic of phosphate were analyzed with pseudo first-order and pseudo second-order kinetic model. The general form of the two models can be described by Equations (2) or (3):

$$q_t = q_e(1 - e^{-K_1 t}) \tag{2}$$

$$q_t = \frac{K_2 q_e t}{1 + K_2 q_e t} \tag{3}$$

where q_t (mg/g) is the amount of solute adsorbed on unit CG at time t (min), q_e (mg/g) is the amount of adsorbate adsorbed per gram at equilibrium, K_1 and K_2 are the rate constant of the pseudo first-order and pseudo second-order model, respectively.

The data of adsorption isotherm of phosphate were fitted with Langmuir and Freundlich isotherm model. The general form of the two models can be described by Equations (4) and (5):

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \tag{4}$$

$$q_e = K_F \times C_e^{\frac{1}{n}} \tag{5}$$

where q_e (mg/g) is the amount of adsorbate adsorbed per gram at equilibrium; q_m (mg/g) is the saturated adsorption capacity; C_e (mg/L) is the adsorbate equilibrium concentration; K_L and K_F are the Langmuir adsorption constant and Freundlich isotherm coefficient respectively, and n is a constant representing the adsorption intensity.

Set-up of modified permeable pavement cells

Four PVC box-type permeable pavement cells (40 cm in length, 20 cm in width and 65 cm in height) were set to evaluate the removal efficiency of typical pollutants. The traditional set up named as 'Control' was designed according to the typical permeable pavement design (CJJ/T 188-2012). The surface layer of the cell was paved by permeable

cement bricks of 20 cm length × 10 cm wide × 5 cm height. The basement consisted of three layers. River sand (particle size 0.5–1.0 mm) with a height of 3 cm was used for the first layer. Gravel (particle size 5.0–20.0 mm) with the height of 35 cm was used for the second layer. River sand (particle size 0.5–1.0 mm) with a height of 5 cm was placed under the gravel layer as the third layer. At the bottom of the cell, perforated polyvinyl chloride (PVC) pipes of 2-cm diameter were used to drain the infiltrated water out of the pavement cell. Synthetic stormwater consisting of dissolved pollutants was pumped from the water tank through a pipe to the rainfall simulator by peristaltic pump, which was designed to provide control of the rainfall volume, intensity, and duration. Synthetic stormwater was evenly introduced into the system through spray nozzles at a set rainfall intensity that was controlled by peristaltic pump. For the basement layer of the three CG modified boxes, the two sand layers, the gravel layer and all three layers were replaced by CG with the corresponding particle size, respectively. They were named CG-S, CG-G, and CG-SG respectively. The size of the materials was fixed at 0.5–1.0 mm, 5.0–20.0 mm and 0.5–1.0 mm for the first, second and third layers, respectively. The different grades' selection was evaluated to make sure the permeability (78 mm/h) of each cell was similar. Figure 1 shows the designs of the permeable pavement cells.
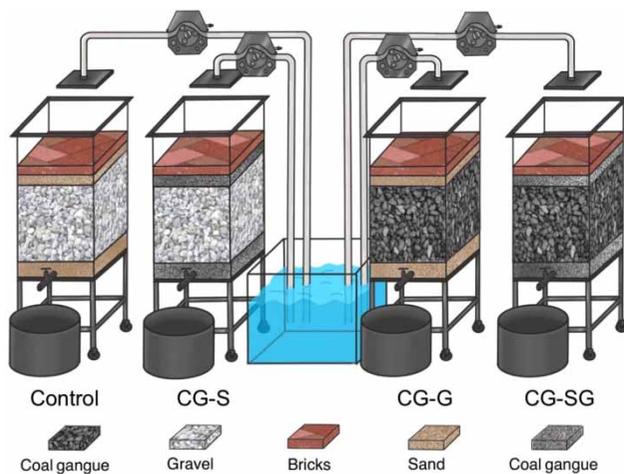

Figure 1 | Schematic diagrams of the four permeable pavement systems' set up. For the filling materials in the basement layer, the size of the materials of each sub-layer was 0.5–1.0 mm, 5.0–20.0 mm and 0.5–1.0 mm with a height of 30 cm, 350 and 50 for the first, second and third sublayer, respectively. Control (as traditional set up) was successively filled with sand, gravel, sand. CG-S was successively filled with CG, gravel, CG. CG-G was successively filled with sand, CG, sand. CG-SG was all filled with CG.

Rainfall simulation and runoff calculation

Leaching experiment

To make sure of the safety of CG as a permeable pavement filler, a leaching experiment was carried out in advance. Four pavement cells were leached by tap water for two hours every day for 9 days until the pollutants' concentration of effluent stabilised (Zhong *et al.* 2018). The rainfall recurrence interval was set as 1 a, with a rainfall intensity of 215.31 L/s·hm^{-2}. Effluent samples were collected and the concentrations of the target pollutants including COD, TN, TP and NH$_4^+$-N were measured. COD was measured according to the Fast digestion-spectrophotometric method (HJ/T 399-2007). TN was measured according to Alkaline potassium persulfate digestion UV spectrophotometric method (HJ 636-2012). TP was measured according to Ammonium molybdate spectrophotometric method (GB 11893-89). NH$_4^+$-N was measured according to Nessler's reagent spectrophotometry (HJ 535-2009). Then the data were compared with China National Standard for Environmental quality standards for surface water (GB3838-2002).

Runoff simulation experiment

For the runoff simulation experiment, COD, NH$_4^+$-N, TN and TP were chosen as the target pollutants because they are the main pollutants in the runoff of the urban area. Raw water (influent water) was prepared based on the representative concentration of surface runoff. COD, NH$_4^+$-N and TP were prepared with glucose, NH$_4$Cl and KH$_2$PO$_4$ respectively, and their corresponding concentrations were 350.0 mg/L, 5.0 mg/L and 1.5 mg/L, respectively. The prepared TN is composed of 5.0 mg/L NH$_4^+$-N (made of NH$_4$Cl) and 6.0 mg/L NO$_3^-$-N (made of KNO$_3$), thus the TN concentration is 11.0 mg/L, summed up of the above two. The solution pH was controlled at 7.0 with HCl and NaOH. The concentration of runoff pollutant is selected based on relevant studies (Zhong *et al.* 2018). The concentration selected is typical in urban road in Beijing. In this study, constant pollutant concentration was adopted mainly to control variables, so as to better study the influence of rainfall conditions on pollutant removal efficiency. The removal efficiency of target pollutants by the four permeable pavement cells was measured under simulated rainfall. The operating conditions, such as rainfall duration, rainfall recurrence period and rainfall drying period were also examined on the influence of the removal efficiency. The removal rate for different forms of pollutants was

calculated by Equation (6):

$$\eta = \frac{C_0 - C_t}{C_0} \times 100\% \tag{6}$$

where η (%) is the removal rate; C_0 (mg/L) is the pollutants concentration in the run off before getting into the pavement columns; and C_t (mg/L) is the pollutant concentration in the outflow.

To investigate the effect of rainfall duration on runoff pollutant removal, the effluent of each device started sampling immediately the water flowed out from the bottom of the device, then was sampled at 5 min, 10 min, 15 min, 20 min, 25 min, 30 min, 40 min, 50 min, 60 min, 75, 90 and 120 min. Rainfall recurrence interval was set as 1 a, with the rainfall intensity of 215.31 L/s·hm^{-2}. Samples were analyzed within 24 h after sampling. During this process, the variation of effluent pollutant concentration with rainfall duration was observed.

To study the influence of rainfall recurrence period on the removal efficiency of pollutants, five different recurrence periods (i.e. 0.5 a, 1 a, 2 a, 3 a and 5 a) were taken to investigate the pollutants' removal rate from different permeable pavement systems. Based on the empirical model of rainfall intensity in Beijing Equation (7) (DBпT 969-2013), five rainfall intensities under different recurrence intervals (P) in years were simulated. Samples were collected and detected to investigate the effect of rainfall intensity on the pollutants' removal rate from different permeable pavement systems.

$$q = \frac{2001 \times (1 + 0.811 \times \lg P)}{(t + 8)^{0.711}} \tag{7}$$

where q (L/s·hm^{-2}) is the rainfall intensity; P (a) is the rainfall recurrence interval, and t (min) is the rainfall duration. Note that the rainfall duration consists of the confluence time of runoff (usually 5–15 min) and the flow duration of rainwater in the pipe. The present study focuses on the removal behavior of runoff pollutants by the filler in a permeable pavement system; the effect of pipe drainage is not considered. Therefore, the rainfall duration was set as 15 min, and thus q was between 163 L/s·hm^{-2} and 337 L/s·hm^{-2} while P ranged from 0.5 a to 5 a (rainfall recurrence period involved in this study). In the simulation experiment of rainfall drying period, five different drying periods (i.e., 1 d, 3 d, 5 d, 8 d and 12 d) were taken to investigate the removal behavior of pollutants from different permeable pavement systems.

RESULT AND DISCUSSION

Characterization of CG

The chemical composition of CG was analyzed by X-ray fluorescence (XRF) techniques, and the results are shown in Table 1. The results showed that the main chemical component of CG is Al_2O_3 and SiO_2, which accounts for 50.26% and 33.50%, respectively. The specific surface area measured by the Brunauer-Emmett-Teller method is 16.66 m^2/g when the grain size is 0.075 mm. The surface morphology of CG was analyzed by scanning electron microscopy (SEM), and the results are displayed in Figure 2. As can be seen from Figure 2, the surface of calcined CG was rough, which may provide many more adsorption sites, thereby increasing the adsorption capacity towards pollutants.
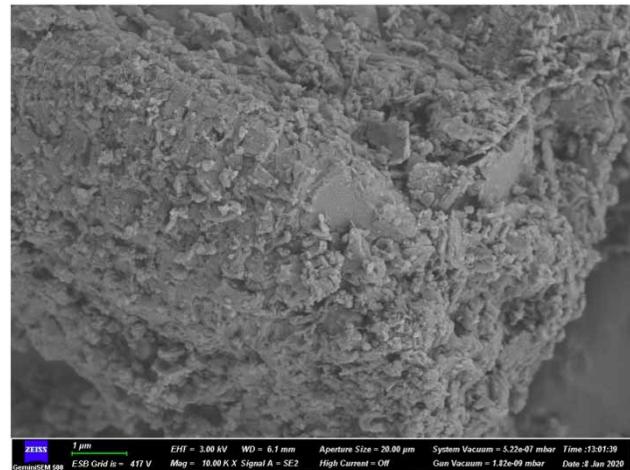

Figure 2 | The surface morphology of CG measured by scanning electron microscopy (SEM).

Table 1 | Chemical compositions of calcinated CG

Compositions	Al$_2$O$_3$	SiO$_2$	Fe$_2$O$_3$	TiO$_2$	CaO	SO$_3$	K$_2$O	MgO	MnO	ZnO	CuO	Total
Content (wt%)	50.26	33.50	5.08	4.17	1.56	0.49	0.41	0.05	0.03	0.03	0.02	95.60

Adsorption kinetics and isotherms of phosphorus on CG

In order to provide the basis of design criteria for the modified pavement media, both adsorption kinetics and isotherms were studied especially for phosphorus. In our pretest, adsorption was also measured for COD, NH_4^+-N and TN. However, the removal of COD and TN by CG in the batch experiment is very limited, which indicated that there is not any adsorption progress occurring on COD and TN to CG. CG adsorbed NH_4^+-N better than the above two, but it is still not significant. In the present study, the CG show little effect towards TN. However, some researchers found CG has a certain effect on treating ammonium wastewater (Zhang *et al.* 2013). The main reason could be due to the differences in production area, coal seams and pretreatment methods of CG, causing different sorption characteristics towards ammonium. Therefore, we focused on investigation of the static sorption behavior of phosphorus on CG. Figure 3 shows the sorption kinetics and isotherm of phosphorus on CG as well as their model fitting curves, and the deduced fitting parameters are shown in Table 2. For sorption kinetics, in the first 30 h, over 80% of the phosphorus was removed by CG. After this fast-initial sorption step, the phosphorus uptake rate increased slowly with time, and no significant change was observed after 75 h. The phosphate concentration after adsorption by CG decreased from 20 mg/L to

1.98 mg/L. The calculated removal efficiency is over 90.11%. The fitting results indicated that the pseudo-second-order kinetic model fitted well ($r^2 = 0.95$) with the adsorption data rather than the pseudo-first-order kinetic model ($r^2 = 0.92$). This implied that the adsorption behavior of phosphorus by CG was mainly by chemisorption. These findings are in good agreement with the results reported by Ding *et al.* (2017), who claimed that the chemical adsorption process of phosphorus from calcined CG is due to the coordination between the oxides of Si, Al, Fe and H_2O to form a hydroxylated surface. The hydroxyl groups on the surface undergo anion exchange reaction with $H_2PO_4^-$, HPO_4^{2-}; and PO_4^{3-}; as a consequence, phosphorus is removed (Ding *et al.* 2017). From the XRF results (Table 1), the CG used in this research is rich in Al, Si and Fe, as shown in the content of Al_2O_3, SiO_2 and Fe_2O_3 accounting for 50.62%, 33.50% and 5.08%, respectively.

For the isotherms, the removal rate was higher for the low initial phosphorus concentration, and lower for high initial phosphorus concentration. As the initial phosphorus solution concentration increased from 0.5 mg/L to 100 mg/L, the removal rate dropped from 92.33% to 13.78%. Sorption increases with the increase of phosphorus loads in a low phosphorus concentration range, and tends to reach a constant value at a high phosphorus concentration range. At the high phosphorus
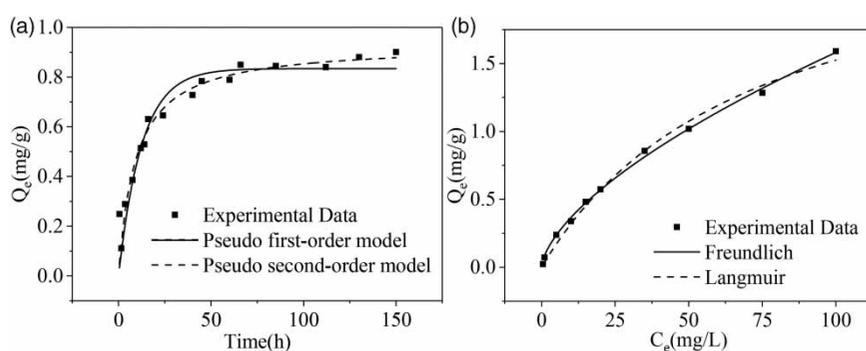

Figure 3 | (a) Adsorption kinetics of phosphate on CG fitted by pseudo-first-order and pseudo-second-order models (solid-liquid ratio is 20 g/L, phosphorus initial concentration is 20 mg/L and constant temperature is 25 °C); (b) adsorption isotherm of phosphate on CG fitted by Langmuir and Freundlich models (solid-liquid ratio is 20 g/L, equilibration time is 150 h and constant temperature is 25 °C).

Table 2 | Parameters of adsorption kinetic and isotherm of CG towards phosphorus

Sorption kinetics				Sorption isotherm			
Pseudo-first-order	K_1	q_e (mg/g)	r^2	Langmuir	K_L (L/g)	q_m (mg/g)	r^2
	0.080	0.834	0.920		0.014	2.625	0.992
Pseudo-second-order	K_2	q_e (mg/g)	r^2	Freundlich	K_F (mg/g)	n	r^2
	0.116	0.932	0.947		0.085	1.575	0.998

concentration region, the adsorption capacity is over 1.50 mg/g, suggesting that CG has the potential to be used as an adsorbent for phosphorus removal. Both the Freundlich model and Langmuir model fitted very well with the data. The Langmuir model shows slightly larger r^2 than Freundlich model (0.998 and 0.992, respectively). The maximum adsorption capacity deduced from the Langmuir model could reach 2.63 mg/g. This implied that the behavior of phosphorus adsorption by CG was characterized by a multilayer sorption. The Freundlich isotherm coefficient (K_F) and n value were 0.085 and 1.575, respectively. The n value greater than 1 represented favorable adsorption conditions. This conclusion is consistent with the study of Ding *et al.* (2017), who claimed that the Langmuir model fitted better than the Freundlich model to describe adsorption of phosphate by spontaneous combustion of CG. The adsorption process occurs on a homogenous surface by monolayer adsorption, and the maximum phosphate adsorption capacity is reported to be 7.08 mg/g (as P) (Ding *et al.* 2017). The maximum adsorption capacity measured in the present study (2.63 mg/g) is smaller than the value reported by Ding *et al.*, which is probably due to the difference in factors such as calcination temperature and the source of CG. The SiO_2 and Al_2O_3 in CG could be effective for the sorption of phosphate. The XRF result in this research showed that the content of Al_2O_3 and SiO_2 accounts for 50.26% and 33.50% respectively (Table 1). Besides, the porosity of CG could provide sorption sites towards phosphate due to the surface area. From the SEM result (Figure 2), the surface of CG was rough, and the BET result showed that its specific surface area was 16.66 m^2/g, which could provide more adsorption sites, thereby increasing the adsorption capacity. Zhang *et al.* (2012) indicated that the phosphate concentration after adsorption by CG from practical waste water decreased from 0.625 to 0.121 mg/L at pH 7, meeting Class III of the China National Standard for Environmental quality standards for surface water. They indicated that CG can be used as an adsorbent to remove phosphate from wastewater (Zhang *et al.* 2012). In our study, the modified pavement systems with CG exhibited an excellent adsorption affinity for phosphorus, which could be feasibly applied for phosphorus removal from runoff.

Leaching experiments for modified pavement systems

CG not properly treated might cause pollution since there are pollutants such as sulphides, chlorides and aromatics remaining (He *et al.* 2018; Peng *et al.* 2018; Wang *et al.* 2018). In our study, CG was preheated at 900 °C for 6 h and during the heating process, which is helpful for removal of pollutants especially for aromatics. That also make the CG safe to be recycled. In order to investigate the leaching behavior of pollutants (COD, TN, TP and NH_4^+-N) in CG relative to gravel and sand, leaching experiments were performed. Figure 4 shows the leaching process of pollutants (COD, TN, TP and NH_4^+-N) in four permeable pavement cells. Four pavement cells were leached by tap water for two hours every day for 9 days. The rainfall recurrence interval was set as 1a. As can be seen, the concentration of four kinds of pollutants in effluent fluctuated around zero, showing that the COD, NH_4^+-N, TN and TP concentrations difference between influent and effluent of the four pavement systems ranged from −8.00 to 22.00 mg/L, −0.03 to 0.09 mg/L, −0.42 to 1.22 mg/L and −0.05 to 0.04 mg/L, respectively. There is no obvious difference between different pavement systems. The stabilized effluent pollutant concentration meets Class II of the China National Standard for Environmental quality standards for surface water. This phenomenon indicates that, compared to the traditional permeable pavement system (Control), the leaching of pollutants from CG-modified permeable pavement system is also very limited. Researchers have tried to use several kinds of materials as layers in the LID devices. You *et al.* (2019) explored the leaching behavior of pollutants from shells, fly ash, basic oxygen furnace slag, bottom ash and refining slag to investigate their feasibility as bioretention fillers. The results showed that the phosphate concentration in leachate for fly ash was relatively higher (i.e. around 50 mg/L on the first day and then over 70 mg/L on the tenth day). For bottom ash, basic oxygen furnace slag and shells, the release amount of phosphate were all below 10 mg/L, which could be used for the bioretention fillers (You *et al.* 2019). Zhong *et al.* (2018) filled the bioretention system with sawdust and dredged mud in a certain proportion, and the leaching test showed that the release amount of COD, TN and TP is at around 600 mg/L, 60 mg/L and 2 mg/L at the beginning then stable at around 50 mg/L, zero and zero, respectively (Zhong *et al.* 2018). The concentration of pollutants leached from this research is much lower than the above study, which also makes sure the safe application of CG into permeable pavement systems. Although CG may cause pollution in some cases, if properly treated, the pollutant leaching risk will be effectively reduced.
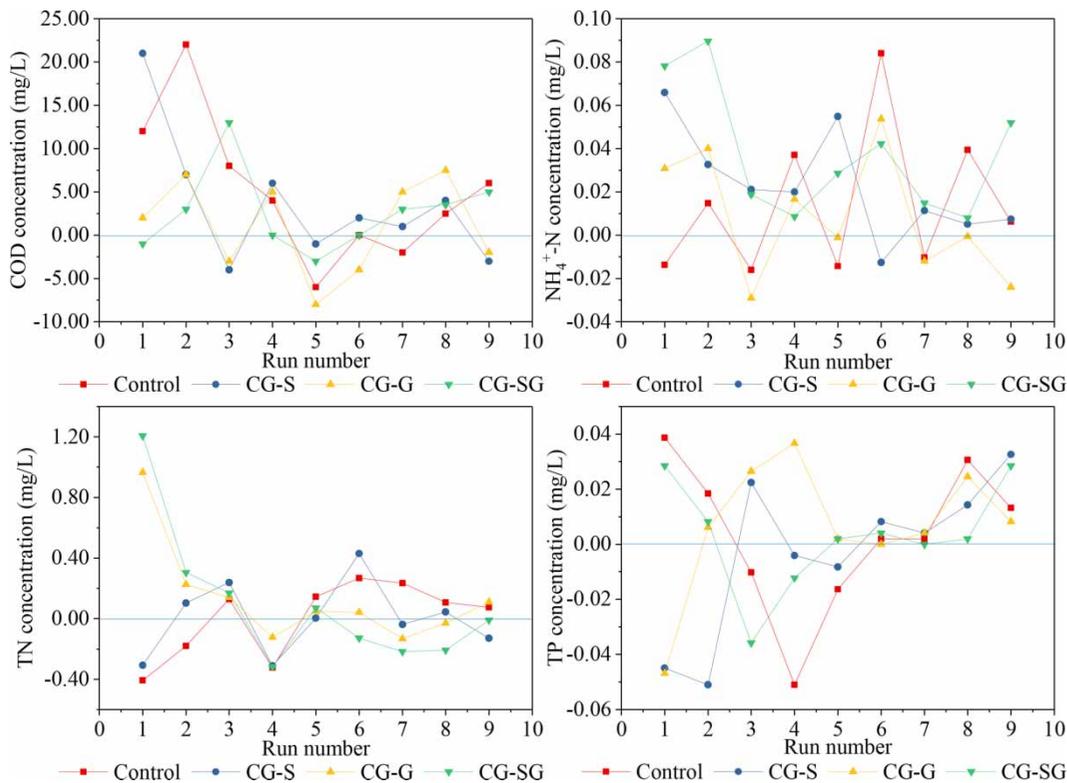
Figure 4 │ Leaching process of stormwater runoff pollutants (COD, TN, TP, NH_4^+-N) in permeable pavement cells.

Impact of different rainfall factors on removal of runoff pollutants

Impact of rainfall duration on removal of runoff pollutants

To compare the effect of rainfall duration on the pollutant removal efficiency of four pavement systems, the effluent pollutant concentrations were measured under different rainfall durations. Figure 5 shows the variation of runoff pollutant concentration with rainfall duration in four kinds of permeable pavement cells. For the majority of pollutants, with the increase of rainfall duration from 0 to 120 min, the effluent concentration increased rapidly at the beginning, then stabilized gradually. The system replayed by CG showed different extents of improvement on removal of different pollutants. It is obvious that the TP removal effect of the three pavement devices filled with CG is significantly improved. The device that replaces the gravel layer with CG has a certain improving effect on the removal of nitrogen pollutants (TN and NH_4^+-N). There is no significant difference in the removal of COD by the four pavement devices, except for CG-S being slightly better. On the whole, the pollutant removal effect of

the four permeable pavement systems is in the order of CG-G > CG-SG > CG-S > Control.

Figure 5(a) shows the variation of effluent COD concentration of the four pavement systems with rainfall duration. The four permeable pavement simulation experimental cells all show good removal efficiency for COD in the early stage of rainfall. At the moment of initial water discharge, the effluent COD concentrations for the four devices were 3–8 mg/L, and the removal rate was 97.83%–99.18%. The effluent concentration rose rapidly in the first 30 minutes and slowed down in 30–75 minutes, then the removal rates remained almost stable after 75 min. For rainfall duration from 0 to 120 min, the COD removal rate decreased from 97.69%–99.19% to 1.91%–29.35%. The pretest for adsorption of COD on to CG as well as sand and gravel is limited, showing that the adsorption could not occur during the rushing process. The removal of COD by permeable pavement systems mainly depends on interception by different layers. Generally, the interception capacity of the traditional pavement system is very limited. The stronger interception effect of the modified pavement system with CG on COD than the traditional system (Control) might be due to its larger specific surface area (16.66 m²/g) and porous structure, but the interception effect is still limited under 2 h duration. Data show that

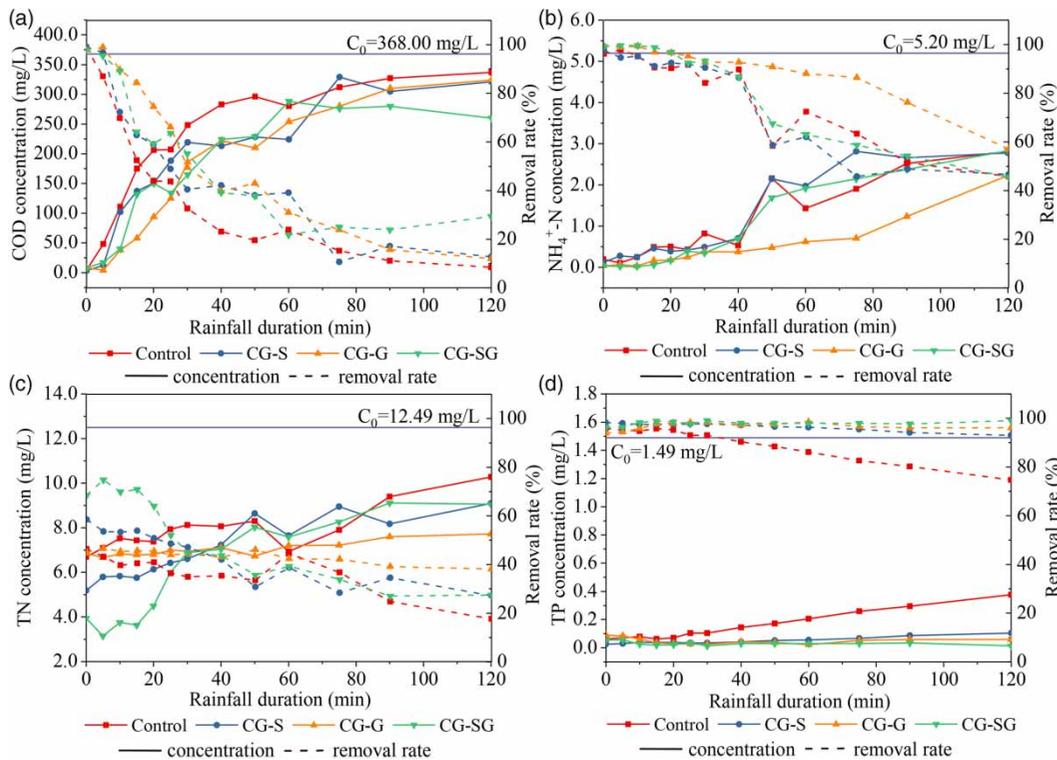**Figure 5** | The variation of runoff pollutant (COD, TN, TP and NH_4^+-N) concentration with rainfall duration in four kinds of permeable pavement cells. C_0 is the influent concentration of pollutants. (a) COD (b) NH_4^+-N (c) TN (d) TP.

COD concentration in the effluent of the traditional pavement system (Control) was 337 mg/L at 120 min (with a removal rate of 10%), indicating that breakthrough nearly occurred. COD concentration in the effluent of the devices CG-S and CG-G was 321 mg/L and 324 mg/L at 120 min (with a removal rate of 12 and 14%) respectively. For CG-SG, the effluent concentration of COD at 120 min is lower than the other three boxes (260 mg/L), corresponding with a removal rate of 25%. On the whole, the removal rates of COD after 2 h rainfall duration of the four permeable pavement systems follow the order of CG-SG > CG-G ≈ CG-S> Control. Even though the removal of COD by the four systems is not significant, the best removal efficiency for COD (%) was observed in CG-SG, showing that all layers replaced by CG may have better interception capacity for COD. Niu *et al.* (2016) indicated that the average removal rates of COD were 24.1% for the surface permeable brick layer, 9.0% for the 5-cm bedding layer and 26.1% for the 20-cm gravel layer under 2 h rainfall duration (Niu *et al.* 2016). Its COD removal rate is slightly higher than this study, but still not satisfactory. Besides, China National Standard for Environmental quality standards for surface water shows that the Class V of surface water COD concentration should be lower than 40 mg/L. The

effluent COD concentration of permeable pavement will pose risks to the environmental quality of groundwater and surface water. Therefore, efforts should be paid to improve the COD removal efficiency by permeable pavements in order to reduce organic load in runoff.

Figure 5(b) shows the variation of effluent NH_4^+-N concentration of the four pavement systems with rainfall duration. Between the rainfall durations of 0–2 h, the NH_4^+-N removal effect of CG-G is always larger than Control, CG-S and CG-SG. The effluent NH_4^+-N concentration of the four systems steadily increased with rainfall duration; the initial effluent concentrations were 0.03–0.19 mg/L and the removal rates were 96.27%–99.35%. For Control, CG-S and CG-SG, the effluent NH_4^+-N concentration grew slowly from 0 to 40 min, then rose rapidly from 40 to 90 min, and finally the removal rates remained almost stable after 75 min. For device CG-G, the effluent concentration rose slowly from 0 to 80 min, then rose rapidly from 80 to 120 min. The effluent NH_4^+-N concentrations were 2.82 mg/L, 2.78 mg/L and 2.83 mg/L, corresponding to removal rates of 45.81%, 46.62% and 45.52% for Control, CG-S and CG-SG, respectively. For CG-G, the concentration was 2.23 mg/L at rainfall duration of 2 h with a removal rate of 57.06%. It is obvious

that CG-G has a better removal effect towards NH_4^+-N than the other three devices. Related research shows that NH_4^+-N could be removed by cation exchange with kaolin (Liu *et al.* 2019). The main chemical composition of kaolin is SiO_2 and Al_2O_3, while CG is rich in SiO_2 (33.50%) and Al_2O_3 (50.26%) in this study (Table 1). Although both gravel and CG contain a certain amount of kaolin, the content of kaolin in CG is much greater than in gravel. Thus, CG shows better removal capacity towards NH_4^+-N than gravel by cation exchange. This may be the reason why the removal rate of NH_4^+-N by CG-G and CG-SG is slightly larger than the other devices with a gravel layer.

Figure 5(c) shows the variation in TN effluent concentration of the four pavement systems with rainfall duration. Between rainfall durations from 0 to 2 h, the four permeable pavement systems show large differences in the removal of TN. The TN effluent concentration of the four systems follows the order of CG-G (6.86 mg/L) ≈ Control (6.69 mg/L) > CG-S (5.18 mg/L) > CG-SG (3.94 mg/L), with a removal rate of 45.11%, 46.41%, 58.53% and 68.48% respectively. The TN effluent concentration of CG-SG rose rapidly from rainfall duration 0 to 90 min, and then rose to 9.05 mg/L at 120 min (with a final removal rate of 27.51%). The TN effluent concentration of Control and CG-S continued to rise slowly through the whole process to 10.28 mg/L and 9.09 mg/L (with a final removal rate of 17.70% and 27.22% respectively) at 120 min. For CG-G, the effluent concentration rose to 7.72 mg/L at 120 min (with a removal rate of 38.18%), showing the best TN removal effect among the four devices. TN removal has remained as a technical challenge in permeable pavement systems, mainly due to its high solubility and poor absorbability. The removal of TN mainly takes place through the nitrification-denitrification processes via microorganisms (Norton *et al.* 2017). The gap of permeable pavement fillers could provide an aerobic environment for ammonifying and nitrifying bacteria to promote ammonization and nitrification, respectively. The obtained NO_3^--N from the nitrification reaction was fixed by the assimilation of microorganisms, and then converted into N_2 by denitrification in the deep anaerobic zone and discharged from the pavement system (Chen *et al.* 2013). However, because of the runoff with nutrients in the permeable pavement system with a faster flow rate and a shorter residence time in the filler layer, it is not conducive to the growth and function of microorganisms. Therefore, the pavement system is not effective in removing nitrogen pollutants. On the other hand, in the long-term operating system saturated with runoff that could continuously provide nutrients for the

growth of microorganisms; as a consequence, the removal of TN could be promoted.

Figure 5(d) shows the variation of TP effluent concentration of the four pavement systems with rainfall duration. It could be seen that the three CG modified pavement systems show significant larger removal efficiency for phosphorus than the traditional system. Overall, the TP removal effect of the four permeable pavement systems follows the order of CG-SG > CG-G > CG-S > Control. The removal of TP is mainly due to absorption as was demonstrated in the adsorption experiment. The traditional pavement fillers were sand and gravels, which were limited in phosphorus absorption. Especially for CG-SG, because it was filled with more CG than the other two CG modified systems, it possessed a larger sorption capacity. The initial effluent phosphorus concentration in the traditional pavement system is low, and it continues to rise with the increase of rainfall duration until 0.38 mg/L (corresponded to a final removal rate of 74.7%) at 120 min. The effluent phosphorus concentration of the other three devices remains low over time. The effluent phosphorus concentration of CG-S and CG-G after 2 h rainfall duration stabilized at 0.10 mg/L and 0.06 mg/L (with a final removal rate of 93 and 96%), respectively. Both effluent concentrations meet Class II of China National Standard for Environmental quality standards for surface water. The TP effluent concentration of CG-SG stabilized at 0.01 mg/L (with a removal rate of 99%), which meets Class I of China National Standard for Environmental quality standards for surface water. The removal of phosphorus in the traditional pavement system (Control) is mainly due to the adsorption that occurs between phosphorus and both gravel and sand. The composition of gravel contains Ca^{2+}, which interacts with PO_4^{3-} in runoff to generate $Ca_3(PO_4)_2$ as precipitate, causing the reduction of TP (Wang *et al.* 2016). With the extension of rainfall duration, the phosphorus effluent concentration of Control keeps rising. The mineral phase composition of CG is mainly quartz, montmorillonite, kaolinite and illite. The metal oxides, such as SiO_2 and Al_2O_3, due to the ion exchange process of their hydrates and phosphate, have a certain ability to adsorb phosphate. The free Ca^{2+} in CG will also chemically react with the water-soluble PO_4^{3-}, resulting in precipitation (Ding *et al.* 2017). As discussed in the previous section, the SiO_2 and Al_2O_3 contained in CG could enhance the removal of phosphate. Phosphate was absorbed by the CG and retained in the pavement system. This could be the main reason for the continuous low concentration of phosphorus in the effluent of the CG modified pavement systems. The effluent concentration of TP in the traditional pavements increased with rainfall duration, while the CG modified system did not show a similar

trend. To some extent, this can be explained by phosphorus being adsorbed stably and phosphorus precipitation not being easily washed out by the current in the CG modified system. In addition, due to the high adsorption capacities and the large amount of CG fille in the pavement system, it is not necessary to worry about the saturation of CG towards phosphorus from urban runoff. In normal cases, it will take hundreds of years to reach the adsorption saturation.

Impact of rainfall recurrence period on removal of runoff pollutants

To compare the effect of rainfall recurrence period on the pollutant removal by four pavement systems, the removal rates under different rainfall recurrence periods (0.5 a, 1 a, 2 a, 3 a and 5 a) were measured. Figure 6 shows the effects of different rainfall recurrence periods (rainfall intensity) on the pollutant (COD, NH_4^+-N, TN and TP) removal efficiency of four permeable pavement systems. For the traditional pavement system (Control), the removal rate of COD decreased with the increase in rainfall recurrence period, from 55.00% with a semi-annual rainfall intensity to 9.73% with a rainfall intensity

of once every three years (Figure 6(a)). The COD removal rate picked up to 16.91% when it came to a rainfall intensity of once every five years. As expected, the increase of rainfall intensity of leads to faster water flow in the system. As mentioned above, the reduction of COD mainly depends on the interception of the filler layer in the system. Therefore, when the scouring speed becomes faster, the interception effect becomes worse. In addition, the pollutants accumulated in the systems are flushed out with the water flow. This may explain the removal efficiency of COD decreasing with increasing rainfall intensity. The removal rate of TP also decreased with the increase in rainfall intensity, from 92.92% to 64.20%. This may be due to the adsorption efficiency of traditional gravel, sand or other fillers for TP being limited and unstable; it also is affected by the intensity of the water scour. The removal rates of nitrogen pollutants (TN and NH_4^+-N) were not considerably different among the different rainfall intensities, perhaps owing to their different removal mechanisms. Liu *et al.* (2019) studied the influence of rainfall intensity on removal of pollutants (NH_4^+-N, NO_X-N, TP and TSS) in permeable pavement. They indicated that NH_4^+-N, NO_X-N and TP concentrations decrease with the increase of rainfall intensity through the surface pavement
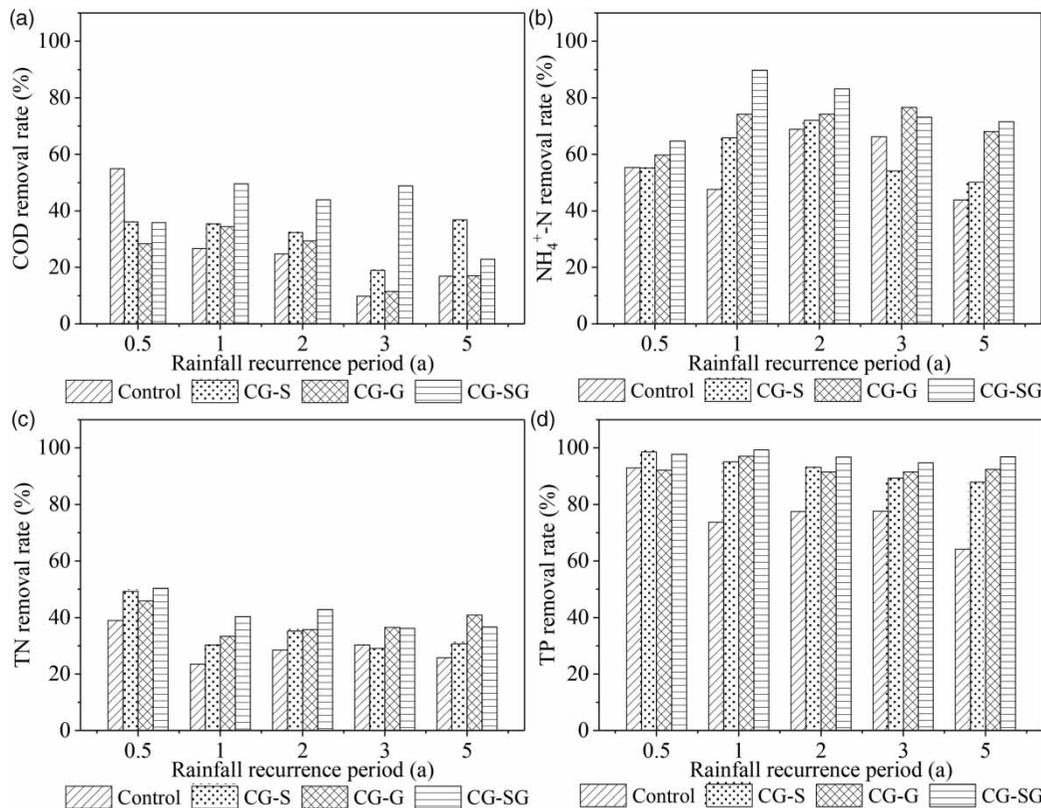

Figure 6 | The effects of different rainfall recurrence periods (rainfall intensity) on pollutant (COD, TN, TP and NH_4^+-N) removal efficiency of four permeable pavement systems. (a) COD, (b) NH_4^+-N, (c) TN and (d) TP.

layer (Liu *et al.* 2019). Those findings are in good agreement with the results in this research.

For the other three CG modified pavement systems, the changes in the four pollutants with rain intensity are not obvious. The removal rates of these three systems for TP continued to be high, basically above 90%. As shown in Figure 6, under the five simulated rainfalls with different rainfall intensities, CG-G and CG-SG had significantly higher removal rates towards the four pollutants than the traditional device except for the removal of COD with the semi-annual rainfall intensity. Most of the pollutants' removal effect by CG-SG is better compared to the CG-G. This indicates that using CG as a permeable pavement filler shows a better removal effect for pollutants regardless of rainfall recurrence. Ostrom & Davis (2019) used the aluminum-based materials for the permeable pavement filler, finding that their removal rate towards dissolved phosphate ranged from 48% to 98%, and it decreased as rainfall intensity increased (Ostrom & Davis 2019). For the CG modified permeable pavement in this research, the removal rates for TP continued to be high, and did not decrease with the increase in rainfall intensity. That might be due to the large saturation adsorption capacity of CG for phosphate. The phosphate in the runoff is quickly adsorbed as it flows through the CG filler. In the short term, the CG in the device will not reach adsorption saturation. Therefore, the TP removal rate continues to be high.

According to the experimental data, the changes of removal rate with rainfall intensity is not obvious. This may be mainly because of the mixture of several types of filler in the basement layer of the pavement system causing non-exclusive factors affecting the removal efficiency of pollutants. Liu *et al.* (2019) studied the removal of nitrogen and phosphate by permeable brick surface material. They found the removal rates of NH_4^+-N, NO_X-N and TP linearly decreased with the increase in rainfall intensity in this surface layer (Liu *et al.* 2019). Besides the homogeneity of the materials, another possibility is that there is a certain level of media layer under the water outlet, water saturates the media layer following each storm and remains there until the start of the next storm. As new influent is applied, water retained from the previous storm is pushed out of the outlet. Contact time for this 'old' water with the media is a function of the time between storms rather than the rainfall intensity of the current storm simulation. Similarly, Ostrom & Davis (2019) observed the difference between a permeable pavement device with internal water storage zone and one without. Their result showed that the removal rate of dissolved phosphate from the device without an internal water storage zone has a good linear relationship with rainfall intensity ($r^2 = 0.769$). However, the linear relationship of the device with an internal water storage zone is not good (Ostrom & Davis 2019). Our result is also speculated to be the cause of 'old' water.

Impact of rainfall drying period on removal rate of runoff pollutants

To compare the effect of rainfall drying period on the pollutant removal of four pavement systems, the removal rates were measured under different rainfall drying periods. Figure 7 shows the effects of different rainfall drying periods on the pollutant (COD, NH_4^+-N, TN and TP) removal efficiency of four permeable pavement systems. With the increase in rainfall drying period, the removal rates of the four pollutants by the four permeable pavement systems did not change obviously. CG modified devices are generally more efficient than the traditional device regardless of the rainfall drying period. Among the four devices, CG-SG shows the best removal efficiency for removal of the four pollutants. Under rainfall periods from 1 to 12 days, its removal rates for COD, NH_4^+-N, TN and TP can reach 32.56–51.16%, 86.70–93.45%, 26.91–46.05% and 95.53–99.32%, respectively. While the removal rates of COD, NH_4^+-N, TN and TP by the traditional system is much lower (24.63–32.25%, 47.62–68.91%, 21.22–32.04% and 64.62–77.88%, respectively). The improved removal rates of the modified systems are mainly due to the interception and adsorption of CG. The better removal effect of TN by the system modified with CG might be mainly due to the action of microorganisms. In the case of certain-term operation of the device (1–5 days of drying period), a certain number of microorganisms may have grown inside the devices. Dan *et al.* (2018) reported that CG contains substantial amounts of carbonaceous shale, organic matter and trace elements, which could provide a proper living environment for the growth of microorganisms (Dan *et al.* 2018). The better growth of microorganisms in CG than sand and gravel could be helpful in microbiological denitrification.

CONCLUSION

In this study, the performance of CG on removal of typical dissolved runoff pollutants was systematically evaluated by bath experiment and as filler in modified permeable pavement systems. The main conclusions are drawn as follows:

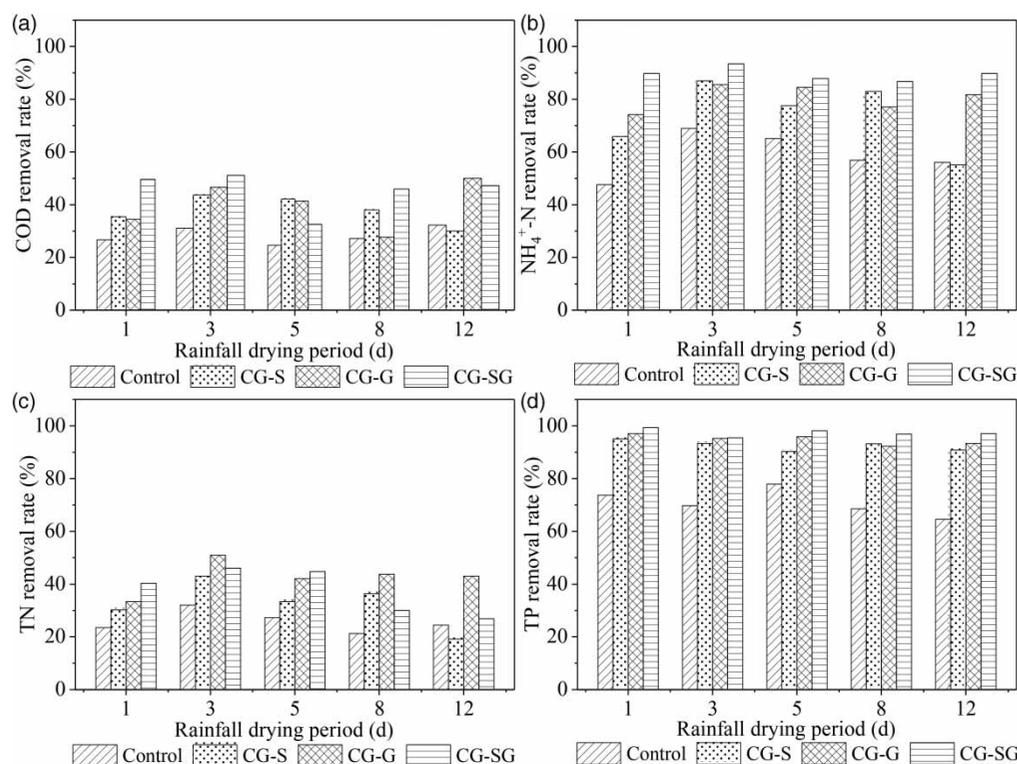(i)   Batch experiment shows that CG have good adsorption performance especially for TP. The maximum sorption

Figure 7 │ The effects of different rainfall drying period on pollutants (COD, TN, TP and NH_4^+-N) removal efficiency of four permeable pavement systems. (a) COD, (b) NH_4^+-N, (c) TN and (d) TP.

capacity could reach 2.63 mg/g and the mechanism is mainly chemisorption and multilayer sorption. This indicates that CG has the potential to be used as an adsorbent.

(ii) The leaching behavior of COD, TN, TP and NH_4^+-N in the modified CG is limited and comparable with the traditional permeable pavement system, indicating properly treated CG as filler would not cause environmental risk.

(iii) With the increase in rainfall duration, the removal rates of COD, NH_4^+-N and TN by traditional permeable pavement system and CG-based permeable pavement systems decreased. CG-based permeable pavement systems show the best efficiency for TP removal (>93.02%). The removal mechanisms include the physical interception and chemical adsorption of fillers and degradation by microorganisms attached to the filler.

(iv) For traditional permeable pavement and three CG modified pavements, both the influence of the rainfall recurrence period and rainfall drying period on removal of runoff pollutants is not obvious. Regardless of the recurrence period of 0.5–5 years or the drying period of 1–12 days, the removal of pollutants by CG-SG is better, and the removal rate of TP is always above 95.53%.

(v) In general, CG replaced both the sand and gravel layers with corresponding particle size; (CG-SG) show the best efficiency for removing runoff pollutants. The removal effect of the CG modified pavement system towards TP is superior. It should be noted that even though it has a certain removal effect on COD, NH_4^+-N and TN, but the effect is not obvious. Further research is necessary to be carried out to explore the proper ways to improve the removal of the other three pollutants.

This research provides a new way for the resource utilization of CG and the performance improvement method of permeable pavement system.

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CONFLICTS OF INTEREST

The authors have declared no conflict of interest.

DATA AVAILABILITY STATEMENT

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

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