Influence of fillers on the removal of rainwater runoff pollutants by a permeable brick system with a frame structure base
Zizeng Lin, Hai Yang and Huiming Chen

ABSTRACT
To fully investigate the effectiveness of fillers in the removal of pollutants from rainwater, gravel, zeolite, slag, volcanic rock and iron filings with a 3–5 cm particle size were applied to construct a brick paving system with a frame structure for the removal of pollutants. Total suspended solids (TSS), chemical oxygen demand (COD), ammonia nitrogen (NH3-N), total nitrogen (TN), total phosphorus (TP) and heavy metals (Cu, Zn, and Pb) in the influent and effluent were measured, and the effectiveness and mechanism of pollutant removal were further investigated. The results showed that the permeable brick system effectively reduced TSS, TP, Zn, Cu and Pb and was relatively ineffective in reducing NH3-N, TN and COD. The removal results obtained using different materials show that (1) physical interception is the main reason for TSS and TP removal, (2) the adsorption and ion exchange properties of zeolite enable it to highly absorb ammonia nitrogen, (3) iron filings can effectively reduce NO3-N, and (4) adding fillers rich in iron oxide, such as volcanic rock or slag, can contribute to COD adsorption. The study provides a feasible technical path for improving the practicability of permeable pavement.

Key words | fillers, frame structure, pollutant removal rate, runoff

INTRODUCTION
In recent years, permeable pavements have become one of the most frequently used low-impact development techniques (Niu et al. 2016). This infiltration-based technology allows rainwater to eventually infiltrate into the natural soil or discharge into a drainage system with a certain amount of pollutant removal (Chu & Fwa 2019), and the materials comprising the various layers are closely related to the ability of the paving system to remove contaminants. First, the characteristics of permeable pavement surface materials are related to the pollutant removal efficiency (Pratt et al. 1995; Zhang et al. 2018). For example, research found that the average total suspended solids (TSS) removal efficiency of six common surface materials (porous asphalt, porous concrete, cement brick, ceramic brick, sand base brick, and shale brick) was nearly 90.0% and the efficiency of shale brick in removing chemical oxygen demand (COD), NO3-N, and total nitrogen (TN) was 88.2%, 35.1%, and 17.5%, respectively (Li et al. 2017). Second, the influence of the base material on the effluent water quality is greater than that of the surface materials since the base layer is the underlying structure of the system and provides the last effluent treatment step (Sañudo-Fontaneda et al. 2014).

Currently, permeable brick paving systems mainly use gravel or cement concrete blocks as the base layer. These materials have some problems, such as the low bearing capacity of crushed stone; furthermore, the construction of cement concrete blocks is very inconvenient because of their heavy weight. Research has shown that the base layer accounts for approximately 2/3 of permeable pavement in volume but the contribution rate of the base layer to runoff pollutant removal is only 10–25% (Sañudo-Fontaneda et al. 2014). This poor mitigation effectiveness indicates that the base layer is not being fully utilized. Therefore, there is a need for improvement of the base layer, which is mainly because the base layer has an important influence on the removal rate of pollutants.

To increase the pollution-removal contribution by the base layer and further improve effluent quality, a frame
base layer with the Chinese character ‘well’ was introduced into the permeable brick paving system, and different fillers could be used inside the well to improve the permeable brick paving system. There are three main advantages to adopting this strategy. One is to reduce the use of cement concrete and its negative impact on water quality; the second is that the flow rate can be adjusted by changing the diameter of the filler; and the third is the removal of contaminants by the filtration, retention and adsorption characteristics of the fillers.

In this study, a permeable brick paving system was constructed using ceramic permeable brick as the surface layer and a ‘well’-shaped frame as the base layer. Gravel, zeolite, volcanic rock, slag and iron filings with the same particle size were added to the base layer. The role and mechanism of the fillers in the removal of contaminants were analysed by changing the type of filler. The objectives and novelties of this research are to explore the feasibility of introducing a well structure and to research the removal effectiveness and mechanism of specific fillers on characteristic pollutants.

MATERIALS AND METHODS

Experimental setup

The test device for the permeable brick paving system is shown in Figure 1. The dimensions of the device were 0.8 m × 0.8 m × 0.5 m. A total of 16 permeable bricks measuring 20 cm × 20 cm × 5.0 cm were placed in close proximity. One 10-mm-diameter outlet pipe was set at the bottom of the permeable brick system. An overflow pipe with a diameter of 10 mm was set at the top of the device.

Materials

Ceramic permeable brick

The size of the ceramic permeable brick was 20 cm × 20 cm × 5.5 cm (Figure S1, Supplement). The main properties of the ceramic brick, such as the splitting tensile strength, permeability coefficient, frost resistance, slip resistance and porosity, all meet the requirements of the Chinese national standard for permeable paving bricks and permeable paving flags (Table 1).

Table 1 | The main properties of the ceramic permeable brick

<table>
<thead>
<tr>
<th>Index</th>
<th>Splitting tensile strength (MPa)</th>
<th>Permeability coefficient (cm/s)</th>
<th>Frost resistance (%)</th>
<th>BPN</th>
<th>Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GB/T25993 – 2010</td>
<td>Average value ≥4.5</td>
<td>Minimum value ≥3.4</td>
<td>≥2.0 × 10⁻²</td>
<td>20</td>
<td>≥65</td>
</tr>
<tr>
<td>Ceramic brick</td>
<td>5.3</td>
<td>4.7</td>
<td>3.1 × 10⁻²</td>
<td>4</td>
<td>89</td>
</tr>
</tbody>
</table>

BPN, British Pendulum Number.
Base layer

The base layer employed a well-shaped frame structure with a symmetrical shape measuring 40 cm × 40 cm × 25 cm and a space ratio of approximately 40%, which contained the filler (Figure S2, Supplement).

Fillers

The fillers of gravel, zeolite, slag, volcanic rock and iron fillings with 3–5 cm particle size were purchased from a Nanjing local building market (Figure S3, Supplement). Scanning electron microscope (SEM) micrographs of the fillers are shown in Figure 2 at 300 times magnification. From Figure 2, we can see a shiny surface with silicate crystals in the gravel; a slightly curved surface with lumps in the zeolite; a well-developed porous structure with large pores in the volcanic rock; a well-developed porous structure with different pore sizes in the slag, where small pores are attributed to the evaporation of organic matter due to the combustion of coal; and a flat surface without any pores in the iron fillings.

The energy-dispersive X-ray spectroscopy (EDS) analysis results are enumerated in Table 2. The elements of calcium, magnesium and aluminium in the fillers mainly exist in the form of oxides, and iron exists as elemental iron.

The specific surface area and pore structure of the fillers are enumerated in Table 3. The specific surface area of the gravel was small, which meant there was little chance of surface adsorption. The micropore volume of the iron fillings was not determined, which meant poor adsorption of pollutants because micropores play an important role in the adsorption process.

Structural form

Five structural forms were constructed using the five fillers, named configurations 1–5, and the detailed structure is shown in Table 4.

Figure 2 | SEM photographs of fillers at 300 times magnification.
Simulated rainfall

The test was based on a 2-hour rainfall duration and 20-year repetition period in Nanjing as the experimental rainfall conditions. The concentrations of various pollutants in the synthetic rainwater are shown in Table 5. In the experiment, the average concentration was selected to determine the removal effect of pollutants.

Test methods

According to the Chinese National Standard Methods (SEPA of China 2002), TSS was determined by the gravimetric method (GB 11901-89), COD was determined by the fast digestion–spectrophotometric method (HJ/T 399-2007), the concentrations of NH₃-N, TN, and TP were determined by the spectrophotometric method, and the concentrations of Zn, Cu and Pb were determined by atomic absorption spectrophotometry (TAS-990, China).

The specific surface area was measured by a surface area and porosimetry analyser (V-Sorb 2800, China), and the main elemental composition was determined by an energy dispersive spectrometer (FEI Quanta 200, The Netherlands).

RESULTS AND DISCUSSION

Removal of TSS

The TSS concentrations and removal rates of permeable brick pavements with different fillers are shown in Figure 3. The removal rate of TSS in all permeable brick pavements showed a trend of ‘rising-stable’ with increasing rainfall duration. The main reason for this tendency may be physical interception inside the permeable brick, which plays a major role in the removal of TSS in permeable brick systems. The TSS was mainly filtered by the voids and pores inside the permeable bricks at the beginning of the rainfall, and the internal voids in the permeable bricks were gradually occupied by suspended particles during the rainfall process, which was helpful in retaining more suspended particles, thereby increasing the TSS removal rate and reducing the TSS concentration. Soon after, a dynamic

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**Table 2 | EDS analysis results of fillers**

<table>
<thead>
<tr>
<th>Element</th>
<th>Gravel</th>
<th>Zeolite</th>
<th>Volcanic rock</th>
<th>Coal slag</th>
<th>Iron filings</th>
</tr>
</thead>
<tbody>
<tr>
<td>O</td>
<td>55.85</td>
<td>48.55</td>
<td>40.62</td>
<td>42.70</td>
<td>23.12</td>
</tr>
<tr>
<td>Mg</td>
<td>0.05</td>
<td>0.92</td>
<td>4.01</td>
<td>0.85</td>
<td>0</td>
</tr>
<tr>
<td>Al</td>
<td>6.11</td>
<td>7.14</td>
<td>7.94</td>
<td>13.93</td>
<td>0.71</td>
</tr>
<tr>
<td>Si</td>
<td>35.41</td>
<td>38.33</td>
<td>26.52</td>
<td>25.40</td>
<td>2.51</td>
</tr>
<tr>
<td>K</td>
<td>0.29</td>
<td>1.45</td>
<td>0.65</td>
<td>1.65</td>
<td>0</td>
</tr>
<tr>
<td>Ca</td>
<td>0.27</td>
<td>2.39</td>
<td>7.55</td>
<td>4.25</td>
<td>0.19</td>
</tr>
<tr>
<td>Mn</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0.60</td>
<td>0.48</td>
</tr>
<tr>
<td>Fe</td>
<td>1.80</td>
<td>1.22</td>
<td>9.43</td>
<td>7.42</td>
<td>70.79</td>
</tr>
<tr>
<td>Na</td>
<td>0.25</td>
<td>0</td>
<td>2.37</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>C</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>3.2</td>
<td>2.2</td>
</tr>
<tr>
<td>Totals</td>
<td>100.00</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100.00</td>
</tr>
</tbody>
</table>

**Table 3 | Specific surface area and pore structure of the fillers**

<table>
<thead>
<tr>
<th>Item</th>
<th>Specific surface area (m²/g)</th>
<th>Micropore volume (×10⁻⁴ cm³/g)</th>
<th>Average pore size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gravel</td>
<td>0.56</td>
<td>1.68</td>
<td>20.56</td>
</tr>
<tr>
<td>Zeolite</td>
<td>14.37</td>
<td>1.97</td>
<td>19.07</td>
</tr>
<tr>
<td>Volcanic rock</td>
<td>8.61</td>
<td>3.05</td>
<td>18.04</td>
</tr>
<tr>
<td>Coal slag</td>
<td>4.15</td>
<td>3.24</td>
<td>30.85</td>
</tr>
<tr>
<td>Iron filings</td>
<td>10.47</td>
<td>0</td>
<td>17.81</td>
</tr>
</tbody>
</table>

**Table 4 | Detailed structure of the configurations**

<table>
<thead>
<tr>
<th>Item</th>
<th>Surface</th>
<th>Levelling</th>
<th>Base</th>
<th>Cushion</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ceramic</td>
<td>Coarse sand with diameter 0.5–1.0 mm</td>
<td>Gravel</td>
<td>Gravel with diameter 15–20 mm</td>
</tr>
<tr>
<td>2</td>
<td>Permeable brick</td>
<td></td>
<td>Zeolite</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td>Volcanic rock</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td>Slag</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td>Iron filings</td>
<td></td>
</tr>
</tbody>
</table>

**Table 5 | Concentration of runoff rainwater pollutants**

<table>
<thead>
<tr>
<th>Items</th>
<th>Minimum value (mg/L)</th>
<th>Average value (mg/L)</th>
<th>Maximum value (mg/L)</th>
<th>Reagent</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSS</td>
<td>185</td>
<td>370</td>
<td>810</td>
<td>Kaolin</td>
</tr>
<tr>
<td>TP</td>
<td>0.9</td>
<td>1.71</td>
<td>2.85</td>
<td>Na₂HPO₄</td>
</tr>
<tr>
<td>TN</td>
<td>3.7</td>
<td>11.4</td>
<td>9.7</td>
<td>NaNO₃, NH₄Cl</td>
</tr>
<tr>
<td>COD</td>
<td>5.7</td>
<td>250</td>
<td>15.9</td>
<td>Glucose</td>
</tr>
<tr>
<td>Ammonia</td>
<td>125</td>
<td>7.41</td>
<td>450</td>
<td>NH₄Cl</td>
</tr>
<tr>
<td>Cu</td>
<td>0.2</td>
<td>0.4</td>
<td>0.8</td>
<td>CuSO₄</td>
</tr>
<tr>
<td>Zn</td>
<td>0.3</td>
<td>0.6</td>
<td>1.2</td>
<td>ZnSO₄</td>
</tr>
<tr>
<td>Pb</td>
<td>0.25</td>
<td>0.5</td>
<td>0.7</td>
<td>Pb(Cl)₂</td>
</tr>
</tbody>
</table>
stable equilibrium between retention and the hydraulic shearing force was achieved, and the TSS in the effluent remained stable.

The base layer formed by the internal fillers in the frame structure also had an interception and adsorption effect on TSS (Jiang et al. 2015; Niu et al. 2016; Kamali et al. 2017), which was the main reason for the improvement in the removal rate of the paving systems. The TSS effluent concentration from the No. 1, No. 2, No. 3, No. 4 and No. 5 paving systems decreased to 61.14 mg/L, 44.21 mg/L, 9.69 mg/L, 18.59 mg/L and 23.43 mg/L, respectively. All five kinds of fillers effectively removed TSS in the runoff, and the TSS removal effectiveness ranking was volcanic rock > iron filings > slag > zeolite > gravel.

The TSS removal rate was the highest for the paving system filled with volcanic rock. Compared with the other fillers, the surface of the volcanic rock was rougher with a natural honeycomb texture, and voids and gaps between the particles were not easily formed during accumulation, which is more conducive to the interception of suspended particles.

Similar to the volcanic rock, the removal rate of slag was also above 90%. This result was due to two reasons: the surface of the slag was rough and porous, which meant effective adsorption, and slag usually contains Fe and Al ions, which play a certain role in flocculation, so the suspended solids were destabilized and aggregated, thereby improving the TSS removal effectiveness (Eren & Acar 2007).

The paving system with iron filings was also effective in removing TSS, which may be because some iron filings were converted to Fe$^{2+}$ or Fe$^{3+}$ by electrochemical, oxidation and displacement reactions (Cheng et al. 2007; Reddy et al. 2014; Statham et al. 2015). Various basic salts containing iron ions are good colloid flocculants with strong adsorption–flocculation activity, and they can adsorb or flocculate suspended particles in water and make them settle, purifying the water.

The effectiveness of the zeolite pavement system was greater than that of the gravel pavement system. The specific surface area of the zeolite was larger, which was beneficial to the adsorption retention of suspended solids on the surface. However, the removal efficiency of TSS by the zeolite was not much greater than that of the gravel, which indicated that the removal of TSS by adsorption was limited and that interception by the fillers was the main reason.

### Removal of TP

The concentration and removal rate of TP in permeable brick paving systems with different fillers are shown in Figure 4. The change trend in TP concentration was almost the same as that of TSS. Eck et al. found that TP was easily adsorbed on suspended particles and removed with TSS by filtration by permeable brick, which is consistent with the conclusions of this research (Cates et al. 2009; Eck et al. 2012).

The TP effluent concentrations of the No. 1, No. 2, No. 3, No. 4 and No. 5 paving systems were 0.520 mg/L, 0.280 mg/L, 0.171 mg/L, 0.146 mg/L and 0.220 mg/L, respectively, and the removal efficiency of TP from high to low was slag > volcanic rock > iron filings > zeolite > gravel. The TP concentration of the effluent in the paving system filled with gravel was higher than the 0.4 mg/L limit for class V of the Chinese Environmental Quality Standards for Surface Water.
Standard for Surface Water (GB 3838-2002), which posed a risk of polluting the urban rainwater environment.

The removal rate of the slag pavement system was the highest. Slag contains certain metal elements, such as Ca, Fe, Al and Mg, that can precipitate with phosphate in water. Studies have shown that the phosphorus removal capacity of materials is positively correlated with metal elements such as Ca, Mg, Al and Fe (Drizo et al. 1999). On the other hand, slag can facilitate the rapid penetration of phosphate into its interior because of its larger micropore size, which is also conducive to precipitation.

The volcanic rocks were rich in pores and contained a large number of elements, such as Al, Ca, Fe and Mg, that can react with phosphate, and their effectiveness was not much different from that of the slag.

Similar to the TSS removal mechanism, Fe$^{2+}$ and Fe$^{3+}$ released by electrochemical, oxidation and displacement reactions with the iron filings precipitated with phosphate, forming iron salt precipitation, which realized the removal of phosphorus pollutants. The reaction formulas are $3\text{Fe}^{2+} + 2\text{PO}_4^{3-} = \text{Fe}_3(\text{PO}_4)_2$ and $2\text{Fe}^{3+} + 2\text{PO}_4^{3-} = \text{Fe}_2(\text{PO}_4)_3$.

The effectiveness of the zeolite paving system was higher than that of the gravel system, which may be due to the high adsorption capacity (Johansson 1999) and the total content of matrix elements (Ca, Mg, Al, and Fe) of zeolite being higher than those of gravel. These two aspects determined the higher phosphorus removal performance of the zeolite.

Three paving systems, the slag, volcanic rock and iron filings, all showed higher TP removal efficiency than that of the gravel, and the average removal rate exceeded 90%. This result showed that the chemical precipitation of phosphorus in the three fillers may be auxiliary to the removal of TP in rainwater.

**Removal of NH$_3$-N**

The concentration and removal rate of NH$_3$-N in permeable brick paving with different fillers are shown in Figure 5. The NH$_3$-N removal efficiency was similar in the five paving systems, and the removal effectiveness in the early rain period was better than that in the later period of rainfall.

The low NH$_3$-N removal rate by the ceramic permeable brick occurred because the dissolved ammonia ions were difficult for the permeable bricks to filter and retain, and the removal of NH$_3$-N was mainly due to adsorption and ion exchange of ammonia nitrogen by the fillers in the permeable brick paving systems (Welker et al. 2013; Jiang et al. 2015; Niu et al. 2016).

The NH$_3$-N effluent concentrations in the No. 1, No. 2, No. 3, No. 4 and No. 5 paving systems were 5.616 mg/L, 4.428 mg/L, 4.818 mg/L, 5.340 mg/L and 5.038 mg/L, respectively, and the order of NH$_3$-N removal efficiency from high to low was zeolite > volcanic rock > iron filings > slag > gravel.

The zeolite added to the frame base was more effective than the other fillers in removing NH$_3$-N. The removal of NH$_3$-N by zeolite occurs via two mechanisms. First, zeolite has a high specific surface area and can adsorb ammonia ions by dispersive force, resulting in greater adsorption of NH$_3$-N. Second, the silicate skeleton of zeolite has a negative charge, attracting NH$_3$-N in rainwater runoff (Shirvani et al. 2006; Wang & Peng 2010). Therefore, some of the NH$_3$-N in rainwater runoff can be effectively removed.

The volcanic rock pavement system was also effective, mainly because of its high porosity, which can provide more adsorption sites for NH$_3$-N.

The effectiveness of the iron filings was slightly lower than that of the volcanic rock, and the overall removal rate was better. This result was because iron filings produce flocculating agents with strong adsorption properties by electrochemical, oxidation and displacement reactions. On the other hand, the iron filings contained a certain proportion of carbon that had a large number of acidic or basic groups, such as carbonyl and phenolic hydroxyl groups, and these acidic or basic groups have a certain adsorption capacity for NH$_3$-N (Keum & Li 2004).

The effectiveness of the slag was better than that of the gravel. On the one hand, the specific surface area of the slag was higher than that of the gravel, which meant a good adsorption capacity. On the other hand, the slag contained
a certain amount of unburned coal, which has properties similar to those of activated carbon, and these substances also have a certain adsorption capacity (Eren & Acar 2007; Acevedo et al. 2015).

Removal of TN

The concentration and removal rate of TN in permeable brick paving with different fillers are shown in Figure 6. The trend of TN concentration change was similar to that of NH$_3$-N. This similarity occurred because in our research, TN was composed of NH$_3$-N and NO$_3$-N, and the TN effluent concentration was mainly affected by NH$_3$-N since the anion NO$_3^-$ was not easily adsorbed by the fillers (Davis et al. 2013).

The TN effluent concentrations of the No. 1, No. 2, No. 3, No. 4 and No. 5 paving systems were 9.338 mg/L, 8.334 mg/L, 7.991 mg/L, 8.605 mg/L and 7.529 mg/L, respectively, and the order of removal efficiency from high to low was iron fillings > zeolite > volcanic rock > slag > gravel.

The iron fillings added to the frame had the highest TN removal effectiveness of all the fillers, which was mainly due to the large amount of Fe in the iron fillings, which can effectively reduce NO$_3$-N, and the adsorption reaction formula is $5\text{Fe} + 2\text{NO}_3^- + 6\text{H}_2\text{O} = 5\text{Fe}^{2+} + 12\text{OH}^- + \text{N}_2$ (Zhang et al. 2010). This reaction provided a feasible technical path for improving the removal efficiency of TN, that is, optimizing the composition of iron fillings and zeolite to remove nitrate nitrogen and ammonia nitrogen, respectively. The TN effluent concentration of the other four paving systems was mainly affected by NH$_3$-N, so the order of removal efficiency of TN was consistent with that of NH$_3$-N.

Removal of COD

The concentration and removal rate of TN in permeable brick paving with different fillers are shown in Figure 7. The COD removal efficiency for the five paving systems showed a trend of ‘falling-stable’ with increasing rainfall duration. The COD removal mechanism mainly included adsorption and interception (Jiang et al. 2015; Niu et al. 2016), and the fillers in the frames of the paving systems mainly relied on adsorption to remove the pollutants.

The COD effluent concentrations of the No. 1, No. 2, No. 3, No. 4 and No. 5 paving systems were 162.458 mg/L, 129.583 mg/L, 95.988 mg/L, 101.120 mg/L and 140.184 mg/L, respectively, and the order of removal efficiency from high to low was volcanic rock > slag > zeolite > iron fillings > gravel.

The COD removal rate of the volcanic rock and the slag was obviously better than those of the zeolite, iron fillings and gravel, mainly due to the high content of Fe$_2$O$_3$ in volcanic rock and slag, which has good adsorption for COD (Koupai et al. 2016). Because of its large specific surface area, zeolite also had good removal effectiveness, which was mainly attributed to its good adsorption capacity. The reason that the iron fillings were superior to the gravel in the paving systems may be consistent with the removal of NH$_3$-N, and this superiority was mainly attributed to the formation of flocculants that could convert dissolved COD into solid matter, which was more likely to be retained; furthermore, a proportion of the carbon contained in the iron fillings also had a certain adsorption capacity for COD.

The removal rates of the volcanic rocks and gravel differed by 25.58% at the same particle size, indicating that
the adsorption properties of the materials played a key role in COD removal and adding fillers rich in Fe₂O₃ can help remove dissolved organic matter.

**Removal of heavy metals**

The concentrations and removal rates of Zn, Cu, and Pb in permeable brick paving systems with different fillers are shown in Figure 8. It can be seen that the removal rates of Zn, Cu and Pb in all permeable brick pavement systems showed a trend of ‘rising-stable’ with increasing rainfall duration. The main reason for this tendency may be that heavy metals are usually removed along with TSS filtration by permeable brick (Walker & Hurl 2002; Jiang et al. 2015).

The removal efficiency of Zn from high to low was volcanic rock > slag > zeolite > iron filings > gravel. The difference in the zinc removal rate was mainly related to the TSS removal and heavy metal adsorption capacity of the fillers (Li et al. 2012; Stagge et al. 2012; Jiang et al. 2015).

The removal efficiency of Cu from high to low was iron filings > volcanic rock > zeolite > slag > gravel. The removal rate of Cu in the paving system filled with iron filings was the best because zero-valent iron can reduce Cu²⁺ to copper, and the reaction formula is as follows: Fe⁰ + Cu²⁺ → Fe²⁺ + Cu⁰. On the other hand, the formation of iron-based flocculants through a series of reactions (Cheng et al. 2007; Reddy et al. 2014; Statham et al. 2015) yielded a strong adsorption capacity, and a certain proportion of the carbon (Acevedo et al. 2015) inside the iron filings also had a certain adsorption capacity for Cu.

The removal efficiency of Pb from high to low was iron filings > volcanic rock > zeolite > slag > gravel. The removal rate of Pb in the paving system filled with gravel was the worst, and the Pb concentration of the effluent was higher than the 0.1 mg/L limit for the class V Environmental Quality Standard for Surface Water, which posed a risk of polluting the urban rainwater environment.

The effectiveness of the removal of Zn, Cu and Pb by the systems filled with gravel, zeolite, volcanic rock, slag and iron fillings was as follows: Zn > Pb > Cu, Pb > Zn > Cu, Zn > Cu > Pb, Zn > Pb > Cu, Zn > Pb > Cu, and Pb > Cu > Zn, respectively. This result may be due to the different binding potential of heavy metals to the suspended solids in the runoff and the fillers in the frame (Pitt et al. 1995; Stagge et al. 2012; Jiang et al. 2015).

**Environmental benefit of the frame structure**

The environmental benefit of the frame structure to the paving system is positive. First, the efficiency of the fillers inside the frame structure was beneficial in removing pollutants from rainwater. Second, the frame structure is conducive to the presence of oxygen and is helpful for the reproduction and survival of denitrifying bacteria, polyphosphate bacteria and other microorganisms, which is
beneficial to the degradation of pollutants such as N and P. Third, a paving system using a frame structure will not have a reduced bearing capacity, and the use of a frame structure has no effect on the life cycle of the paving system.

CONCLUSIONS

(1) A permeable brick paving system was effective in removing TSS, TP, Zn, Cu, and Pb. However, the TP and Pb concentrations of the effluent in the paving system filled with gravel were higher than the class V limit of the Chinese environmental standard, which posed a risk of polluting the urban rainwater environment.

(2) The removal rates of NH₃-N and TN in all the pavement systems were not ideal. Zeolite had a high adsorption effect on NH₃-N, and iron filings could effectively reduce NO₃-N. The effectiveness of removing TN in rainwater can be further improved by optimizing the composition of the iron filings and zeolite to remove nitrate nitrogen and ammonia nitrogen, respectively.

(3) Permeable brick paving systems with different fillers had a large difference in COD removal, and the removal rates of the volcanic rocks and the gravel differed by 25.58%, indicating that the adsorption properties of the materials played a key role in COD removal. Adding fillers rich in Fe₂O₃ can help remove dissolved organic matter.

(4) The efficiency of the fillers inside the frame structure was beneficial in removing pollutants from rainwater, and the environmental benefit of the frame structure to the paving system was positive. The study provides a feasible technical path for improving the practicability of permeable pavement.

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SUPPLEMENTARY MATERIAL

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