**Introduction**

Non-point source pollution has increasingly impacted water pollution in cities with the improvement of controlling point-source pollution [1]. Storm-water runoff contains excess nutrients that contribute to the process of eutrophication of surface water, resulting in the degradation of the quality of drinking water and recreational water. Monitoring results of 61 lake (reservoir) nutritional status from the Chinese environmental report of 2015, eutrophication, nutrition, and oligotrophic lakes (reservoirs) were 23%, 67%, and 10%, respectively [2]. As one of the major nutrients in storm runoff, nitrogen mainly comes from lawn fertilizers, fallen leaves, grass clippings,
fertilized soil, detergents, atmospheric deposition, and precipitation [3]. Pollutants in rainwater accumulate, scour, and are transported before they enter the receiving water [4]. Scouring involves supply (background value) and transmission limits. Nitrogen scouring performed the process of supply limitation, whereas phosphorus performed the transmission limitation. Expressing the flushing of nutrients based on particles (especially nitrogen pollutants scoured), may produce inaccurate results [5]. Therefore, appropriate measures should be taken to reduce the nitrogen load in storm-water runoff, which is discharged into the receiving water.

Total nitrogen (TN) includes particulate organic nitrogen (PON), dissolved organic nitrogen (DON), ammonia (NH$_3$-N), nitrite (NO$_2$-N), and nitrate (NO$_3$-N) [6]. Dissolved pollutants (DON, NH$_3$-N, NO$_2$-N, and NO$_3$-N) are more mobile, bioavailable, and are captured via different mechanisms than particles [7]. A bioretention system utilizes decentralized landscape, hydrology and hydraulic design, replacement or reinforcement of soils, plants to control storm runoff, and non-point source pollution. The media intercept pollutants through sedimentation and adsorption when rain runs into the bioretention system and then is transformed through conversion between pollutants and microbial degradation. Li et al. [8] found that the total nitrogen (TN) load cut rate (inflow-outflow TN load through the perforated drain pipe/inflow TN load) is 41% when the inflow TN load is 14.0 kg/(ha year), and the percentage caused by leakage loss accounted for 31.5%, whereas that retained in the bioretention facilities accounted for 9.7%. Meanwhile, 5.3 kg/ha-year of the captured PON (6.7 kg/ha-year) appears to undergo mineralization, ammonification, and nitrification to DON and NO$_3$-N between storm events. NH$_3$-N is a positive ion, which can be removed by adsorption to a negatively charged media when rain runs into the system first. The removal of NH$_3$-N by bioretention soil media (BSM) primarily relies on the nitrification of NH$_3$-N to NO$_3$-N by nitrite and nitrate bacteria, then NO$_3$ transforms to NO/NO$_2$/N$_2$ under anoxic conditions through denitrification.

A variety of internal and external conditions affect the bioretention operation results [9-10]. Bratieres et al. [11] tested plant species, filter media, filter depth, filter area, and pollutant inflow concentration for optimal design. The “optimal specifications” can remove up to 70% nitrogen and over 95% suspended solids consistently. The optimally designed biofilter is at least 2% of its catchment area and possesses a sandy loam filter planted with C. appressa or M. Ericifolia. Enhanced nitrogen removal has been observed when the bioretention system includes a submerged zone height, carbon source, and artificial packing [12-14]. A triple-layer design would be designed to ensure nitrogen removal. A top layer of organic-amended sand could capture particle organic matter and promote plant growth, and the artificial packing middle layer could help adsorb pollutants, including nitrogen, and an internal storage zone with e-donor could promote denitrification. On the whole, the key problem for nitrogen removal of bioretention design is BSM composition and structural configuration.

This study analyzed the concentration of nitrogen pollutants under intermittent and continuous operation through a simulated rainfall test. We explored the discharge of changing pollutant concentrations for three constructed bioretention devices with different media-layering characteristics. Moreover, the nitrogen removal effect was evaluated by analyzing the quantity of accumulation, leakage, and exhaustion for the bioretention facility under continuous operation, which is dominated by fast adsorption. Additionally, the main influencing factors for bioretention operation were identified, and we established a correlation model with nitrogen removal.

**Materials and Methods**

**Device Setup**

Ten bioretention tanks from 1# to 10# were designed and constructed at the outdoor test ground of Xi’an University of Technology in 2013, and each tank had different

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**Fig. 1.** Cross-section of bio-retention pilot plant. Construction of test tank includes an aquifer, mulch, planting soil layer (PSL), artificial packing layer (APL), and gravel drainage layer (GDL) from top to bottom. WDW for water distribution weir, ITW for inflow triangle weir, OTW for outflow triangle weir, OVTW for over triangle weir, and WG for waterproof geotextile. 
structural configurations. Bioretention tanks 7#, 9#, and 10# were selected for the experiments on intermittent running and continuous operation. The dimensions of each groove were 2.0 m×0.5 m×1.05 m (length×width×depth). The structural configuration was aquifer layer, planting soil, artificial fillers, and gravel drainage layer from the top downward. Artificial packing layer of test grooves were fly ash mixing sand (FS, volume ratio 1:1), blast furnace slag (BFS), and plant soil (PS). *Ligustrum quihoui* Carr and *Ophiopogon japonicas*, which are commonly used for greening roads, were planted in the bioretention devices, with a covering layer of *Platanus orientalis Linn* leaf. An XTHJ recorder (Teng Hui temperature control instrument and meter plant in Yuyao, Zhejiang Province) was installed in front of a 30° triangular weir to observe the inflow, outflow, and overflow current capacities. The cross-section of a bioretention plant is shown in Fig. 1, and a picture of the retention plant is shown in Fig. 2.

**Monitoring and Analysis Methodology**

The ratio of the surface area to the catchment area of typical bioretention facilities is 1/20–1/10 [15]. This ratio was set to 17:1 in this study, and rainfall was calculated by 2 h rainfall volume in 0.5 a (year), 2 a, and 5 a, based on the Xi’an rainstorm intensity formula [16]. High and low pollutant concentrations were designed from actual monitoring data in Xi’an city according to the studies of Yuan [17] and Dong [18]. The concentrations of the test water are shown in Table 1. This test required the simulation of the complete process of rains from the start to the end, including “grow-peak-fall.” Cen et al. [19] found a single-peak rain pattern mainly in a short period of rains, and most of the peak flow appeared in the front and at the center using extensive modeling and comparison. The effect was better observed when the peak flow or storage volume was calculated using the Chicago-pattern (also known as kiefer and Chu rain pattern). This process could meet the general requirements for accuracy, and the strong process was relatively easy to determine with a wide range of applications both locally and abroad. This test was conducted in accordance with the Chicago-pattern, and the factor of peak flow was 0.3.

The parameters for the water quality analyses were temperature, pH, electrical conductivity, dissolved oxygen (DO), TN, total dissolved nitrogen (TDN), NO$_3$-N, NH$_3$-N, and chemical oxygen demand (COD). Water temperature, pH, conductivity, and DO were used in the instrumental measurement using a HACH HQ40d two-circuit input multi-parameter numerical analysis. The water sample was filtered with a 0.45 μm filterable membrane, and the original water sample was oxidized with potassium persulfate. UV spectrophotometry was performed to determine TDN and DN separately. NO$_3$-N was determined by employing the phenol disulfonic acid spectrophotometric method. NH$_3$-N was analyzed by adopting Nessler’s reagent colorimetric method. COD was examined using HACH DRB200 digestion and UV spectrophotometry.

A single-field rainfall event mean concentration (EMC) is the quotient of the total pollutant and the total runoff volume in one rainfall event. This parameter was used to compare quite different rainfall events based on pollutant quantity. However, the change in the actual rainfall process concentration was separately identified. This process enabled the increase in the amount of separate points as far as possible to calculate the pollutant average concentration in a single rainfall event because pollutant concentration cannot correspond with current capacity, as shown by Equation (1), and concentration removal and load removal as a calculation shown by Equations (2) and (3):

$$\text{EMC} = \frac{M}{V} = \frac{1}{t} \int_{0}^{t} Q(t) C(t) \, dt = \frac{1}{t} \int_{0}^{t} Q(t) \, dt = \sum C \cdot Q \cdot \Delta t_i / \sum Q \cdot \Delta t_i$$

(1)

$$R_c = \frac{(\text{EMC}_{in} - \text{EMC}_{out})}{\text{EMC}_{in}}$$

(2)

$$R_L = \frac{(M_{in} - M_{out})}{M_{in}} \times 100\%$$

(3)

where $R_c$ is the pollutant concentration removal rate, $R_L$ is load removal rate, $M_{in/out}$ is inflow/outflow pollutant load, $\text{EMC}_{in/out}$ is the mean concentration of inflow/outflow pollutant, $c (t)$ is the mass concentration of pollutants over time, and $Q (t)$ is flow of unit time.

**Table 1. Water concentration and required reagent (mg/L).**

<table>
<thead>
<tr>
<th>Concentration</th>
<th>COD</th>
<th>NO$_3$-N</th>
<th>NH$_3$-N</th>
<th>TP</th>
<th>Cu</th>
<th>Zn</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>High</td>
<td>600</td>
<td>14</td>
<td>6</td>
<td>2.5</td>
<td>1.0</td>
<td>1.5</td>
<td>0.05</td>
</tr>
<tr>
<td>Medium</td>
<td>300</td>
<td>8</td>
<td>3</td>
<td>1.0</td>
<td>0.5</td>
<td>0.8</td>
<td>0.03</td>
</tr>
</tbody>
</table>
Experimental Scheme

Clear water was used to run the bed before the test. The routine test (including tests 1, 3, 5, and 8) involved medium pollutant concentration. Water volume was calculated under the 5a return period, seven days antecedent dry time, and non-internal submerged zone. Test 1 was the preliminary test. Test 2 established the submerged zone height of 150 mm. Antecedent dry time for tests 4 and 6 were 15 and three days, respectively. Water volume of test 7 was under the 2a return period. Test 9 had higher water concentration. The scheme of intermittent running tests is shown in Table 2.

Adsorption capacity was likely reduced in a few hours when the continuous inflow was considered to be constant under medium concentration. This test increased the hydraulic load from low to high step-by-step. This experiment was designed such that three bioretention facilities ran in low inflow volume (0.5a) for 24 h, then ran for 12 h with medium inflow volume (2a), finally at high inflow volume (5a) for 8 h with effective volume of 2,700 L for the high cistern. The drain valve opening and inflow concentration were constant in each return period. Inflow sample was collected every 2 h, whereas the outflow sample was collected after 0.5 h to determine the concentrations at different times. The media was assumed to be exhausted if continuous $C_{\text{out}} / C \geq 1$ (where $C_{\text{out}}$ is effluent concentration, and $C$ is influent concentration). The water volume schedule for a continuous run is shown in Table 3.

Results and Discussion

Determining Soil Textural Classification and Pollutant Background Value

This test collected the surface soil (10–15 cm) for soil textural classification and pollutant original content analysis, according to the Technical Specification for Soil Environmental Monitoring [20]. After natural drying, the plants, stones and other residues were eliminated from soil samples, then these samples were screened through a 2-mm soil sieve after crushing. Soil textural classification was tested through Mastersizer 2000 wet measurement using United States soil size classification standard (clay<0.002 mm, silt ≈ 0.05~0.002 mm, sand ≈ 2~0.05 mm) for soil texture classification, and then determined soil texture in accordance with United States soil textural triangle.

The results of soil particle analysis indicated that the percentage of clay, silt, and sand of plant soil in this test are 3.49%, 90.36%, and 6.15%, respectively. This means that the textural classification of the soil in these three bioretention tanks is silty soil, which is dominated by silt, and clay content is relatively small. Pollutant background value is shown in Table 4, and it demonstrates that surface soil background values of pH, TC, TOC, and TN were similar in the three bioretention facilities.
Nine rainfall events were conducted for the intermittent running test. Inflow pollutant concentrations are constant, and the sample of the inflow was set at 0, 60, and 120 min after the start of the test. The outflows were sampled once every 15 min, when the discharge pipe was allowed to leak water. The process lines of eight field tests on pollutant concentration, except for the preliminary experiment (Test 1), are shown in Fig. 3.

NO\textsubscript{3}-N and NH\textsubscript{3}-N were considered as target pollutants. Outflow pollutant concentrations of three bioretention ponds with different packing layers showed distinct trends. Outflow pollutant concentration of 7# (artificial packing layer for FS) gradually increased along with time, whereas those of 9# (artificial packing layer for BFS) and 10# (artificial packing layer for PS) fluctuated and then decreased slowly (Fig. 3). The infiltration process in the layered bioretention tank is divided into three forms, which are a high permeability layer over a lower permeability layer, a low permeability layer over a higher permeability layer, and homogeneous infiltration media. Hsieh et al. [21] noted that a greater proportion of ammonium was removed when there was a high permeability layer over a lower permeability layer. The infiltration rate order of media in this test is BFS>PS>FS.

The best load-reduction rates of nitrogen pollutants is 7# (more than 70%), for which Hsieh et al. obtained a similar conclusion.

O’Neill and Davis [22] combined 5% water treatment residual (WTR) and 3% triple-shredded hardwood bark in sand loam in a bioretention tank to improve media adsorption capacity for phosphorus. The concentrations of TP and TDP gradually decreased with inflow time under constant and variable loads, similar to the 9# and 10# bioretention tanks, but remarkably different with the 7# bioretention tank in this test. Changes in the water concentration trend in the current study were affected by three aspects. On the one hand, the surface adsorption capacity of the soil filler achieved a certain recovery with an antecedent dry period of seven days in the general test. The filler surface had faster adsorption points in initial rainfall, while the filler surface fast adsorption area was occupied gradually over time, such that water concentration gradually increased. Columns of municipal compost were irrigated to simulate 6-month, 24-hour rain storms in the Seattle-Tacoma region, and the results showed that compost could serve as a sustained source of leaching of nutrients and metals [23]. Lucke and Nichols [24] also found the negative TN removal for the performance of street-side bioretention basins after 10 years in operation. Water pollutant load was smaller in the early 20 min, when a Chicago rain pattern was chosen as the simulation precipitation test. Then increases and decreases were also observed, indicating the worst media adsorption case when the higher hydraulic load comes. Accumulated pollutants from the system may rush out during rainfall peak flows. Tang et al. [25] results showed that the rain garden had little removal capability for dissolved nitrogen and phosphorus, and they contributed it to the existence of preferential flow in soil. What’s more, the difference in packing combinations had different permeabilities, absorption capacities, etc. Thus the outflow concentration wave may be due to preferential flow.

### Analytical Results Under Continuous Operation Test

The exhaustion points of the media were examined through breakthrough curves. Bioretention lifetime was evaluated through cumulative inflow volume and pollutant loads. Breakthrough curves of soil solutes could be divided into continuous and pulse input soil solute breakthrough curves according to the inflow type of trace elements. It could also be divided into saturated and unsaturated solute
breakthrough curves according to the status of soil water content. Schedules for a continuous operational test are shown in Table 3, and the outflow concentration trends of NH$_3$-N and NO$_3$-N in three bioretention facilities are shown in Fig. 4.

NH$_3$-N in 7# bioretention was $C_{out} > C$ (1-10%) after running for 40 h and 28 h for NO$_3$-N. NH$_3$-N effluent concentration for 9# was $C_{out} > C$ (1-10%) after running for 30 h. NO$_3$-N, which accumulated in the 9# bioretention in the early time discharged with the synthetic rainwater, resulted in a higher outflow concentration when initial outflow was sampled. NH$_3$-N concentration was $C_{out} > C$ (1-10%) for 10# after 26 h. Then, water concentration rose continually, and $C_{out} > C$ (1+10%) after 30 h. NH$_3$-N adsorbed in soil particles surface was scoured out when hydraulic load increased, which led to greater outflow than inflow concentration. NO$_3$-N concentration fluctuated near $C$ (1-10%) after 14 h, which indicated that NO$_3$-N concentration reached adsorption exhaustion for planting soil more quickly than other tanks. The corresponding

![Fig. 4. Concentration change process in continuous operation. 'C' refers to influent pollutant concentration: a) NH$_3$-N at 7#, b) NO$_3$-N at 7#, c) NH$_3$-N at 9#, d) NO$_3$-N at 9#, e) NH$_3$-N at 10#, f) NO$_3$-N at 10#.](image-url)
percentages of the accumulated pollutants, NH$_3$-N and NO$_3$-N, in total inflow pollutant load of three bioretention tanks when they reached exhaustion point as follows: #7, 77.39% and 76.02%; #9, 59.93% and 61.94%; and #10, 48.9% and 36.5% – demonstrating the relatively poor performance of planting soil.

The discharge ratio of the bioretention basin was 17:1, and its surface area was 1 m$^2$. The rainfall volumes in 17 m$^2$ catchment area of #7, #9, and #10 bioretention basins were 0.52, 0.34, and 0.26 year of rainfall in Xi’an when NH$_3$-N achieved exhaustion. The corresponding water volume reduction rates of the basins were 28.82%, 31.84%, and 34.84%, respectively. Corresponding rainfall volumes for NO$_3$-N were 0.30, 0.15, and 0.15 year of rainfall in Xi’an, and water volume reduction rates were 44.28%, 34.84%, and 34.84%, respectively. O’Neill et al. [26] studied phosphorus adsorption though a 2.5 cm inner diameter mini-column, and inflow concentration of the dissolved phosphorus was 120 µg/L. Media without WTR, hardwood bark, or leaf compost were evidently exhausted. The inflow volumes of the two unamended groups (BSM, bioretention soil media and LFBSM, low-fines bioretention soil media) of bioretention were 220 and 250 mm rainfall volume of the 20:1 catchment ratio when BSM achieved exhaustion, which is probably equal to 0.23 year of the rainfall in the District of Columbia. The accumulation rainfall volume increased several tens of times when special media was combined into the bioretention soil media to achieve exhaustion. In continuous operational test, NH$_3$-N and NO$_3$-N actual mean inflow concentrations were 5.1 and 8.5 mg/L, respectively. The experimental result indicated that nitrogen pollutant can achieve adsorption exhausted points in a relatively short time under continuous operation fast adsorption, and the performance of NO$_3$-N is more evident.

### PLS Modeling Analysis

TN removal rate was the only dependent variable $y$ in the statistical analysis of TN removal for bioretention facilities. The independent variable $x$ included the inflow pollutant concentration, inflow hydraulic loading, antecedent dry time, submerged zone height, and media factor. All factors had two variables, except for antecedent

<table>
<thead>
<tr>
<th>Test number</th>
<th>GN</th>
<th>IC(mg/L)</th>
<th>IV(L)</th>
<th>ADT (day)</th>
<th>SD (mm)</th>
<th>PF(10$^{-3}$d/m)</th>
<th>MV(%)</th>
</tr>
</thead>
<tbody>
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<td>19.79</td>
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<td>532.82</td>
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<td>7</td>
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<td>264.23</td>
<td>19.90</td>
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</table>

Note: GN is groove number; IC is inflow concentration of TN; IV is inflow volume; ADT is antecedent dry time; SD is submerged depth; PF is packing factor; MV is measured values of TN.
dry time and media factor, which had three variables. Curve analysis showed a strong linear relationship between TN concentration and influencing factors. Thus, Equation (4) and the primitive variable Equation (5) in the PLS model were standardized as follows:

\[ y^* = a_1x_1 + a_2x_2 + a_3x_3 + a_4x_4 + a_5x_5 \]  

\[ \hat{y} = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4 + b_5x_5 \]  

...where \( y \) is the TN removal rate (%), \( x_1 \) is the inflow TN concentration (mg/L), \( x_2 \) is inflow hydraulic loading (L), \( x_3 \) is the antecedent dry time (day), \( x_4 \) is the submerged zone height (mm), and \( x_5 \) is the media factor (media adsorption capacity divided by the padding infiltration rate) \( (10^{-3} \text{d/m}) \).

Twenty-one groups of data from seven rainfall events were considered as modeling samples. Tests 3 and 8 were used in the model examination, and the modeling and examination samples are listed in Tables 5 and 6. The standardized variable Equation (6) is as follows:

\[ y^* = -0.079x_1 - 0.019x_2 - 0.346x_3 + 0.255x_4 + 0.647x_5 \]  

The primitive variable Equation (7) is as follows:

\[ \hat{y} = 31.056 - 0.292x_1 - 0.004x_2 - 1.340x_3 + 0.063x_4 + 0.053x_5 \]  

The difference between simulated and measure values of TN removal was analyzed using PLS (Fig. 5). The regression line for the observed values showed better fitting with deterministic coefficient \( R^2 \) of 0.622. The simulated and measured values were close (residuals within 16).

As shown in Fig. 6, the result from the PLS analysis between TN removal rate and influencing factors showed...
that media factor was the most important influencing factor for TN removal in the bioretention facilities. The order of importance in other factors is as follows: antecedent dry time, submerged depth, inflow concentration, and inflow water volume. Five important factors were selected in the study. Antecedent dry time, inflow concentration, and inflow water volume were considered as the external influencing factors for bioretention ponds. Packing factor and submerged zone height were considered as the internal influencing factors. Correlation analysis showed a positive relationship between TN removal and the media factor and submerged zone height. This result indicates that the higher the media factor and deeper the submerged zone, the greater TN removal will be. However, a negative relationship was found between TN removal and antecedent dry time, inflow concentration, and inflow hydraulic loading. TN removal rate was relatively worse with longer antecedent dry time, relatively high inflow concentration, and inflow hydraulic loading.

Conclusions

Intermittent and continuous running test results showed that the 7# bioretention tank had the highest pollutant load cut rate, and the trend of water pollutant concentration increased gradually with time. BFS as artificial packing layer (9#) followed 7#, but pure planting soil bioretention (10#) showed the worst results. Pollutant concentration performances of 9# and 10# showed fluctuating tendencies, followed by gradual reduction. When 7#, 9#, and 10# reached their exhaustion points, the percentages of pollutant loads that accumulated in the media and the total inflow pollutant load followed the order of 7#<9#<10#. The media is the most critical factor for the removal efficiency of nitrogen compared with antecedent dry time, submerged area height, inflow concentration, and inflow volume. A positive relationship was found between TN removal rate and packing factors and submerged area height. By contrast, a negative relationship was found between TN removal rate and antecedent dry time, submerged area height, inflow concentration, and inflow volume.

The microbial role in bioretention tanks should be strengthened in future studies, and the accumulated status and migration process of runoff pollutants in bioretention systems should be explored further. Pollutant migration should be studied over time using long-term test or simulation to evaluate the lifespan of bioretention.

Acknowledgements

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