

# A two-stage soil infiltration system incorporated with heterotrophic denitrification (TSISHD) for urban runoff treatment

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## ABSTRACT

To enhance denitrification performance of soil infiltration, a soil infiltration system incorporated with heterotrophic denitrification (TSISHD) for urban runoff treatment was developed. Sawdust and grass powders were added in the anaerobic stage (ANS) to provide organic carbon sources for the denitrification process, and the reduction environment was improved by iron addition in the ANS. Aerobic respiration and nitrification primarily occurred in the upper aerobic stage (AES), which removed 86.68% of the chemical oxygen demand (COD) and 91.80% of the  $\text{NH}_4^+\text{-N}$ . Moreover, heterotrophic denitrification occurred in the bottom ANS when added sawdust and grass powders were used as a carbon source. Overall, the TSISHD showed remarkable removal efficiencies of 88.29%, 82.50%, 92.05%, and 78.10% for COD,  $\text{NO}_3^-\text{-N}$ ,  $\text{NH}_4^+\text{-N}$ , and total phosphorus, respectively, and the corresponding effluent concentrations met the national quality standard of China for class V surface water. The removal efficiencies were significantly higher than those of the previous soil infiltration systems without inoculated microbes. The developed system has the potential to treat urban runoff.

**Key words** | heterotrophic denitrification, pollutants, soil infiltration, urban runoff

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## INTRODUCTION

The quantity of impervious surfaces such as building roofs, roads, driveways, and parking lots increases during land development and urbanization. These surfaces prevent stormwater from following its natural route into and through the soil. The quality of the runoff also declines as more pollutants from the impervious surfaces are carried directly into storm drainage systems and eventually into neighboring streams, resulting in eutrophication (Zhang *et al.* 2008; Fowler *et al.* 2009; Tomar & Suthar 2011; Hou *et al.* 2013). However, there is currently great demand to use collected rainwater for drinking and other purposes in areas subject to water shortage.

To date, conventional methods for the treatment of urban runoff have included wetlands and ponds, as well as physical separation techniques such as filtration and screening devices. However, the application of wetlands and ponds is not feasible in many urban areas because they

require large spaces for installation. Accordingly, there is an increasing need to develop ecologically viable and safe small-scale urban runoff treatment technologies for onsite treatment of urban runoff. Infiltration systems have the potential to replenish groundwater by enabling urban runoff to return to aquifers (Murakami *et al.* 2008). In addition, urban runoff seepage water from such systems can be reused (Tota-Maharaj & Scholz 2010; Hou *et al.* 2013). Multilayer infiltration systems comprising natural materials effectively remove most pollutants, including organic matter (chemical oxygen demand ( $\text{COD}_{\text{Cr}}$ )), total nitrogen (TN), ammonia-nitrogen ( $\text{NH}_4^+\text{-N}$ ), and total phosphorus (TP) with reductions of 68.67%, 23.98%, 82.66%, and 92.11% of  $\text{COD}_{\text{Cr}}$ , TN,  $\text{NH}_4^+\text{-N}$ , and TP, respectively (Hou *et al.* 2013). Effluent that meets critical values following treatment can then be used for firefighting, irrigation, washing cars and machinery, or as toilet or cooling water.

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However, systems that use natural soil as filter media are subject to clogging (Poza-Morales *et al.* 2013).

The multi-soil-layering (MSL) system, which fills pores around the soil using large particle materials such as sand and zeolite to create a filter layer, was developed to maintain relatively high removal efficiency and avoid clogging (Watsuki *et al.* 1993). When compared with traditional soil infiltration methods, the MSL has a remarkably longer service life and removal efficiencies, with reductions of 80%, 75%, and 95% of TP, TN, and biochemical oxygen demand (BOD), respectively, being observed during treatment of restaurant wastewater (Luanmanee *et al.* 2001). However, the nitrate removal efficiency is relatively low in the MSL, primarily due to the inhibition of denitrification by aeration and lack of electron donors caused by the low C/N (Masunaga & Sato 2007). It has been reported that artificial aeration could improve nitrogen removal efficiency (Butterworth *et al.* 2013); however, excessive aeration leads to decreases in both denitrification efficiency and overall nitrogen removal efficiency (Luanmanee *et al.* 2002). When compared with continuous aeration, intermittent aeration reduced operational costs while creating different micro-environments for effective nitrogen removal (Dong *et al.* 2012). Kong *et al.* (2014) developed a soil infiltration column with an upper aerobic stage (AES) and a lower anaerobic stage (ANS) incorporated with sulfur-utilizing autotrophic denitrifiers for domestic wastewater treatment and found that denitrification performance of soil infiltration was enhanced.

Although there have been some attempts to combine the AES and ANS to establish a two-stage reactor (Chinalia *et al.* 2012; Kong *et al.* 2014), few studies have investigated

incorporation of denitrification with soil-based nitrification for treatment of urban runoff to date. Ordinarily, the event mean concentrations of COD, TN, and TP from runoff of urban catchment mixed with road, roof, plaza, open space, parking lots are slightly lower than the value of the corresponding pollutant parameters of local sewage (Zhang *et al.* 2012). It is more probable to treat urban runoff successfully by soil infiltration systems with inoculated microbes. Therefore, in this study, laboratory-scale two-stage soil infiltration systems incorporated with heterotrophic denitrification (TSISHDs) were developed by integrating an AES with an ANS, which allowed polluted urban runoff to flow directly into the heterotrophic denitrification stage without pumping, reducing space and energy costs. To evaluate the performance of organic carbon addition and artificial aeration modes in the TSISHDs, the removals of COD,  $\text{NO}_3^-$ -N,  $\text{NO}_2^-$ -N, and TP in both the aerobic and ANS were investigated using synthetic urban runoff, and the denitrification mechanism was analyzed in the TSISHD. In addition, the water quality of the TSISHD effluent was compared with the national quality standard of China (Environmental Quality Standard of Surface Water in China, GB 3838-2002).

## MATERIALS AND METHODS

### Urban runoff characterization

Table 1 lists the primary categories of urban runoff pollutants, pollutants associated with each category, typical concentrations, likely runoff sources, and potential effects. A

**Table 1** | Summary of urban runoff pollutants relevant to this study

Category	Parameters	Possible sources	Possible effects	Solution concentration (mg L <sup>-1</sup> )
Organic enrichment	COD <sub>Cr</sub>	Urban runoff; agricultural runoff; landfills; septic tanks; atmospheric deposition; soil erosion	Dissolved oxygen depletion; odors; toxicity toward fish and other aquatic organisms	250
Nutrients	TN	Urban runoff; agricultural runoff; fertilizers; landfills; septic tanks; atmospheric	Surface water: algal overgrowth and blooms; ammonia toxicity. Ground water: nitrate toxicity	25
	$\text{NH}_4^+$ -N	deposition; soil erosion		20
	$\text{NO}_3^-$ -N			2.25
	TP			1.46

**Table 2** | Water quality standards for various pollutants in China

Parameters/Standard value	Environmental quality standard of surface water in China (2002)				
	I	II	III	IV	V
pH/ ≤	6–9	6–9	6–9	6–9	6–9
COD/ ≤	15	15	20	30	40
TP/ ≤	0.02	0.1	0.2	0.3	0.4
NH <sub>4</sub> <sup>+</sup> -N/ ≤	0.15	0.5	1.0	1.5	2.0

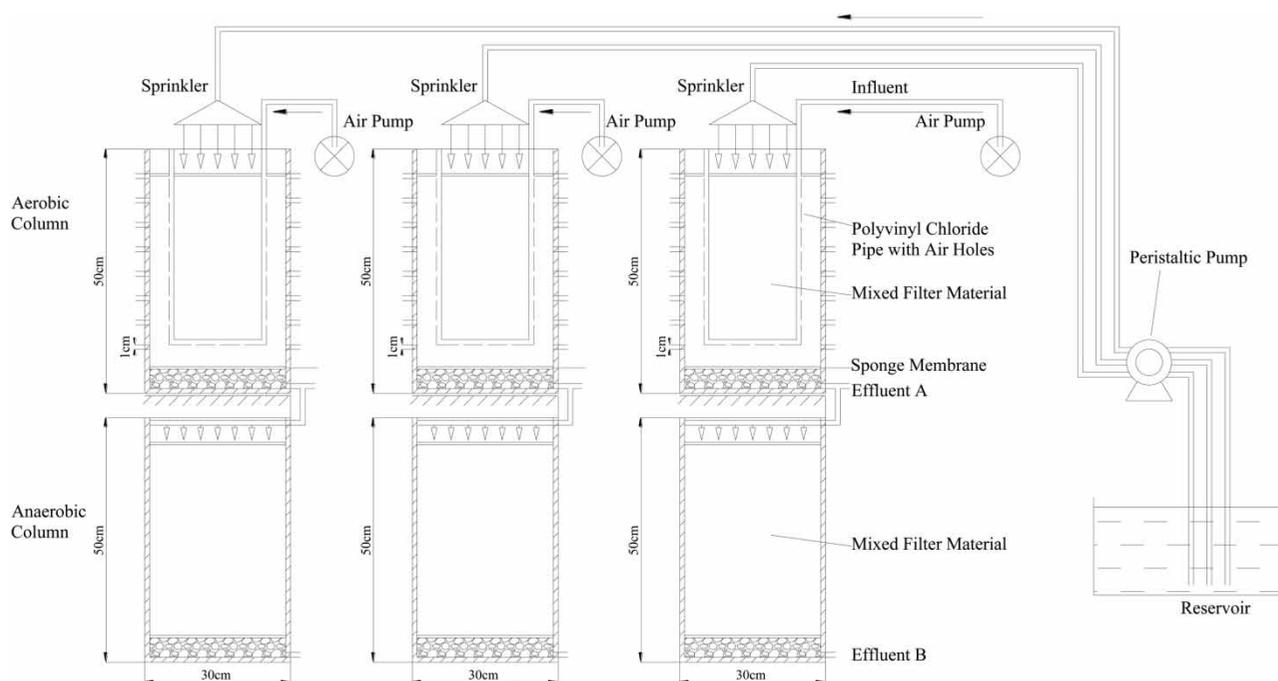
runoff supply system was prepared in a water tank containing synthetic urban runoff with specific concentrations of solution as previously described (Li *et al.* 2007; Zhang *et al.* 2012; Hou & Zhang 2014) (Table 1). The runoff contained COD<sub>Cr</sub>, TP, TN, and NH<sub>4</sub><sup>+</sup>-N from roof and road surfaces, with average concentrations exceeding the 40 mg/L, 0.4 mg/L, 2.0 mg/L, and 2.0 mg/L limits set by the class V surface water quality standard (GB3838-2002) of the Ministry of Environmental Protection of the People's Republic of China, respectively (Table 2). The runoff influent was obtained as follows: pharmaceutical influent was prepared in the laboratory, after which 28.8 L of influent solution containing 6.75 g of dextrose (C<sub>6</sub>H<sub>12</sub>O<sub>6</sub>), 2.2012 g of ammonium

chloride (NH<sub>4</sub>Cl), 0.8743 g of sodium nitrite (NaNO<sub>2</sub>), and 0.1845 g of potassium dihydrogen phosphate (AR, Sino-pharm Chemical Reagent Co., Shanghai, China) in tap water (16–25 °C) was added to each column for every experimental cycle. The concentrations of COD, NH<sub>4</sub><sup>+</sup>-N, NO<sub>2</sub><sup>-</sup>-N, and TP in the synthetic runoff water were 156.00–344.00 mg L<sup>-1</sup>, 2.25–7.19 mg L<sup>-1</sup>, 5.98–6.44 mg L<sup>-1</sup>, and 0.74–3.56 mg L<sup>-1</sup>, respectively. The pH in the synthetic runoff remained constant over the course of the experiment and was slightly alkaline for all treatments.

### Experimental apparatus

As shown in Figure 1, the TSISHD system mainly consisted of three replicate reactors, three aeration pumps, and a synthetic water reservoir to avoid the uncertainty. Each system was divided into two sections, an aerobic column and an anaerobic column, to improve the nitrogen removal efficiency.

Two polyvinyl chloride reactors with dimensions of 30 cm (L) × 20 cm (W) × 50 cm (H) were separated into an AES for heterotrophic nitrification and an ANS for heterotrophic denitrification. The two reactor columns were

**Figure 1** | The laboratory-scale TSISHD reactors.

connected with sealed pipes. The AES column was independent of the ANS column to ensure the growth of nitrifying and denitrifying bacteria, as well as to avoid the mutual effects of the AES and ANS.

The AES, which contained quartz sands (grain diameters 0.5–1 and 2–4 mm), volcanic materials (2–4 mm), zeolites (2–4 mm), and activated carbon (2–4 mm), all from Shengquan Water Clean Materials Co., Beijing, China, had an oxygen-enriched environment formed by both surface natural re-oxygenation and internal mechanical aeration with an air pump (At-A7500, Atman, Chuangxing Co., Zhongshan, China). The aeration flow rate was 1 L min<sup>-1</sup> and the aeration time was 8 h every day. A spongy porous membrane (30 × 20 × 1 cm) was placed over the surface of the AES to uniformly distribute the influent. A polyvinyl chloride pipe (length 120 cm, diameter 1 cm) with holes (diameter 5 mm) drilled every 2 cm was bent into a 'U' shape, and then buried at a depth of 5 cm from the sidewall and bottom of the AES to ensure sufficient aeration. The holes were set facing down to prevent clogging by the filter materials.

The ANS contained quartz sands (grain diameters 0.5–1 and 2–4 mm), volcanic materials (2–4 mm), zeolites (2–4 mm), and activated carbon (2–4 mm) similar to those in the AES, and more densely packed filter materials (Table 3). Specifically, there was less quartz sand of larger sizes and volcanic materials added in the ANS than in the AES to create a relatively lower porosity relative to the AES. Wood chips and sawdust were obtained locally and crushed into particles (diameter 2–4 mm) by a small high-speed pulverizer (ZN-04, Kingslh Co., China) after washing and drying.

## Experimental procedure

Activated sludge was collected from a secondary sedimentation tank (Gaobeidian Wastewater Plant, Beijing, China)

to obtain nitrifying bacteria, while another sample of activated sludge was acquired from an anaerobic tank (Yanshan Beer Plant, Beijing, China) for denitrifying bacteria. The filter materials were adequately mixed in a 25-L plastic bucket using a shovel after washing. The mixture for ANS was then placed into one ANS reactor, after which 500 mL of the acclimated solution was inoculated into the filter materials. The mixture of filter materials was then compacted firmly with the shovel to reduce the porosity and create an anaerobic environment. Next, 1,000 mL of cultured nitrifying bacteria solution was poured into a bucket containing filter materials and then stirred thoroughly with a shovel, after which the mixture was poured into one AES reactor.

Synthetic water was stored in a 28.6-L plastic tank as a reservoir and then pumped onto the surface sponge via three sprinklers at a flow rate of 20 mL min<sup>-1</sup> using a peristaltic pump (Lead 15–24, Longer Precision Pump Co., Baoding, China), after which it was allowed to flow through the AES and ANS by gravity until being discharged from the bottom outlet. The flow rate during experiments was fixed at 20 mL min<sup>-1</sup>, roughly corresponding to one moderate rain, similar to the occurrence in Beijing City. To reflect the discontinuities of urban runoff, the hydraulic retention time (HRT) was designed to be 8 h. The TSISHD was operated for 66 days at room temperature (20–25 °C). Samples were collected daily from the Influent, Effluent A, and Effluent B, and COD, NO<sub>3</sub><sup>-</sup>-N, NH<sub>4</sub><sup>+</sup>-N, and TP for each sample were analyzed. Data from the three replicate reactors were collected, respectively, and their average values were displayed.

## Analytical methodology

Analytical reagents were used to prepare standards and stock solution (Sinopharm Chemical Reagent Co., China).

**Table 3** | Components of filter materials

Stage	Quartz sand (S) (%, v/v)	Quartz sand (L) (%, v/v)	Volcanic material (%, v/v)	Zeolite (%, v/v)	Activated carbon (%, v/v)	Scrap iron filings (%, v/v)	Sawdust and grass powder (%, v/v)
AES	30	35	20	10	5	–	–
ANS	50	15	10	10	5	5	5

The COD was digested with potassium dichromate using a 5B-1F Speed Digester (Lianhua Tech Co., Beijing, China) at 150 °C for 10 min and then determined by ultraviolet spectrophotometry at 610 nm.  $\text{NH}_4^+\text{-N}$  was measured by Nessler's reagent colorimetry based on the absorbance at 420 nm. TP was measured by molybdate colorimetry based on the absorbance at 700 nm. All spectrophotometric analyses were conducted using a Hach DR6000 spectrophotometer (Loveland, CO, USA).  $\text{NO}_3\text{-N}$  and  $\text{NO}_2\text{-N}$  were analyzed using a Dionex ICS-2500 ion chromatograph (Dionex Corporation, Sunnyvale, CA, USA). Water temperature and pH were monitored using a FE30 Benchtop Meter (Mettler Toledo Co., Toledo, OH, USA).

After cleaning and drying, the sawdust and grass powder samples were ground into powder of the same particle size and analyzed using a 2400CHNS/O Perkin Elmer Series II Analyzer at Peking University Analysis and Testing Center. The results showed that the mean C, N, and H percentages in sawdust were 46.27%, 0.085%, and 6.22%, respectively, and the average C, N, and H percentages in grass powder were 42.87%, 0.60%, and 5.94%, respectively.

## RESULTS AND DISCUSSION

### COD

COD is an important indicator of organic load of urban runoff. The COD load in effluent from TSISHDs was significantly lower than the initial levels. As shown in Figure 2, TSISHD showed effective removal ability for COD.

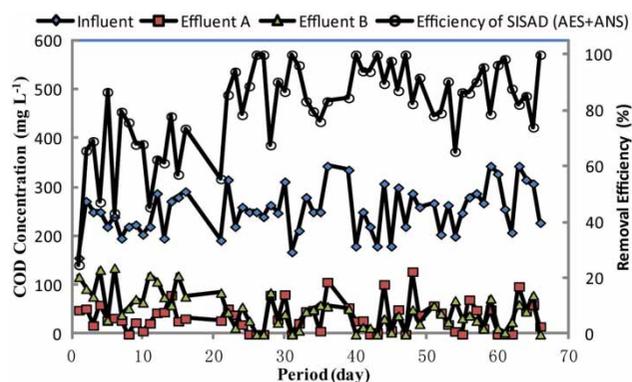


Figure 2 | COD concentration and removal efficiency.

Specifically, the COD concentration decreased to 40.00 mg  $\text{L}^{-1}$  on day 23, giving a removal efficiency of 81.82%, after which the average COD removal efficiency in Effluent A from the AES was 87.33%. These findings indicated that bio-film formation for AES was completed on day 23. Conversely, although most COD was consumed in the AES, the COD concentration in Effluent B continued to decrease. Similar to Effluent A, the COD concentration in Effluent B decreased to 117.33 mg  $\text{L}^{-1}$  on day 1, giving a removal efficiency of 24.79%, while it was reduced to 13.33 mg  $\text{L}^{-1}$  on day 23, after which the COD concentration was kept almost constant. These results showed that ANS also had the capacity to remove residual COD from AES, as indicated by the average COD concentration in Effluent B being 31.90 mg  $\text{L}^{-1}$ . Therefore, TSISHD exhibited high performance for removal of COD, with an average removal efficiency of 88.29%, which was better than the efficiency of 68.67% reported for multi-layer infiltration systems (Hou et al. 2013).

The COD values from Effluent B changed greatly throughout the operation process, and were greater than that of Effluent A during the beginning of the experiment. This is because the filter in the ANS column contained sawdust and grass powder, which dissolved organics continuously. The average C percentage of sawdust and grass powder reached 46.27% and 42.87%, respectively, both of which led to increased dissolution of organic matter in the ANS column. As shown in Figure 2, the total COD removal efficiency was more than 75% and the average removal rate reached 88.29% from day 23. In the earlier stage, the grass powder could dissolve a great deal of organic matter in a short period, ensuring that anaerobic microbes quickly adapted to the environment. The organic matter dissolved from sawdust decreased and the dissolution became stable during the late ANS, which enabled normal microbial growth and stability of the system. Accordingly, the COD of Effluent B was much greater than that of Effluent A during the beginning of the experiment, while in the stable stage the COD value in Effluent B was greatly reduced, with most samples containing less than the 40 mg  $\text{L}^{-1}$  set by the national quality standard of China (Environmental Quality Standard of Surface Water in China, GB 3838-2002) for class V surface water (Table 2).

The COD removal observed in the developed system occurred via several pathways: (1) COD was consumed as nutrient during growth of aerobes in AES, which is similar

to the results of a study conducted by Sato *et al.* (2005); (2) COD was used as an organic electron for heterotrophic denitrification in ANS; and (3) the activated carbon granules in the mixture adsorbed a portion of COD (Zhao *et al.* 2013) in both AES and ANS, especially during the initial period of the experiment.

### NH<sub>4</sub><sup>+</sup>-N

As shown in Figure 3, AES showed good ammonium removal capacity, with an average NH<sub>4</sub><sup>+</sup>-N removal efficiency of 91.80% being observed in Effluent A. This was mainly due to ammonium adsorption by zeolite in the filter materials. Pell & Nyberg (1989) reported that better surface transfer of oxygen via soil aeration benefited the growth of nitrifying bacteria, and that more than 80% of the NH<sub>4</sub><sup>+</sup>-N could be removed from the soil surface under hydraulic loading of 6.7 cm d<sup>-1</sup>.

Conversely, the concentration of NH<sub>4</sub><sup>+</sup>-N in Effluent B from ANS generally remained below 0.5 mg L<sup>-1</sup>, which met the national quality standard of China (Environmental Quality Standard of Surface Water in China, GB 3838-2002) for class II surface water (Table 2). The average NH<sub>4</sub><sup>+</sup>-N concentration in Effluent B was 0.36 mg L<sup>-1</sup> with an average removal efficiency of 92.05%, indicating that the residual NH<sub>4</sub><sup>+</sup>-N from the AES was used in the ANS. The decrease of NH<sub>4</sub><sup>+</sup>-N in the ANS was attributed to adsorption by the filter materials.

Both AES and ANS showed removal performance on NH<sub>4</sub><sup>+</sup>-N, even on the first day, with 81.73% and 89.75% removal efficiencies, respectively. Nevertheless, the nitrate

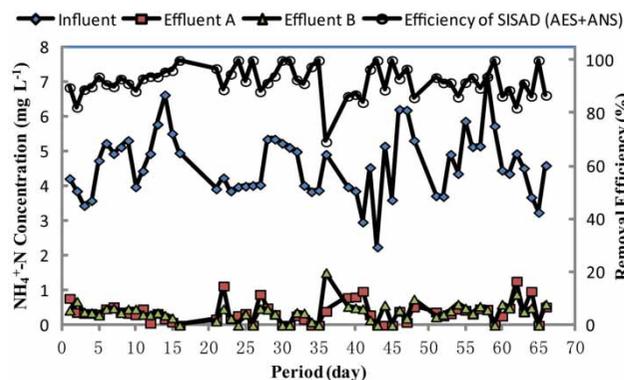


Figure 3 | NH<sub>4</sub><sup>+</sup>-N concentration and removal efficiency.

concentration in Effluent A (Figure 4) was less than 15 mg L<sup>-1</sup> before day 13, and reached 51 mg L<sup>-1</sup> on day 21. In other words, the NO<sub>3</sub><sup>-</sup>-N generation rate was unable to keep up with the NH<sub>4</sub><sup>+</sup>-N removal rate. Therefore, the NH<sub>4</sub><sup>+</sup>-N removal during the initial period (until day 21) mainly resulted from adsorption by the mixed filter materials. Zeolite has been shown to have an excellent adsorbing effect on NH<sub>4</sub><sup>+</sup>-N, and such adsorption follows the Freundlich model (Zhao *et al.* 2013). In this study, a series of batch tests were also conducted to estimate the adsorption capacities for different materials. The results suggested that zeolite had a 10.13 mg g<sup>-1</sup> saturated adsorption capacity for 48 h, and the best performance among sand, volcanic material, and activated carbon (data not shown). As shown in Table 3, zeolite accounted for 10% (v/v) of the mixed filter materials filled in the reactor, weighing 10,500 g. The average influent NH<sub>4</sub><sup>+</sup>-N concentration was 4.61 mg L<sup>-1</sup>, and 89.75% NH<sub>4</sub><sup>+</sup>-N removal in the reactors on day 1 was hypothesized as the adsorption efficiency, 39.71 mg NH<sub>4</sub><sup>+</sup>-N could be adsorbed by the

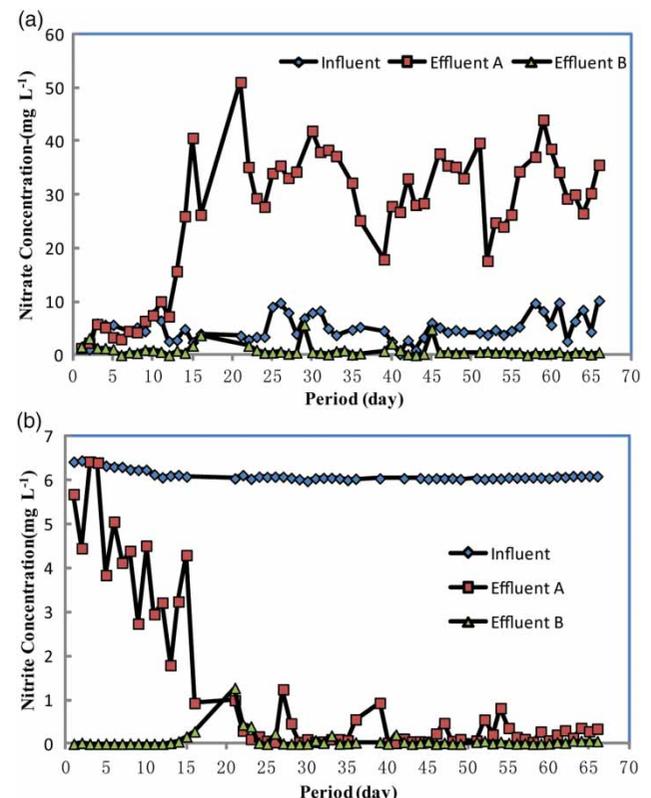


Figure 4 | NO<sub>3</sub><sup>-</sup>-N (a) and NO<sub>2</sub><sup>-</sup>-N (b) concentration.

zeolite in one set of reactor (AES + ANS) every day (9.6 L synthetic runoff water) for every system. However, 10,500 g zeolite could adsorb 17,728 mg  $\text{NH}_4^+\text{-N}$  calculated using an HRT of 8 h, then divided by  $39.71 \text{ mg d}^{-1}$ , calculating 446 days as the theoretical saturated adsorption period for  $\text{NH}_4^+\text{-N}$ . It exceeded the 21 days mentioned above, after which the ammonia removal was considered resulting from microbial activities. In addition, zeolite might never achieve a practical saturated adsorption for  $\text{NH}_4^+\text{-N}$  owing to the oxidation of  $\text{NH}_4^+\text{-N}$ . Thus, the  $\text{NH}_4^+\text{-N}$  removal was due to both adsorption and nitrification.

After day 14, nitrifying bacteria gradually formed a bio-film, at which time the conversion from ammonium to nitrate by nitrifying bacteria gradually became the dominant mechanism of removal instead of adsorption. Overall, the average  $\text{NH}_4^+\text{-N}$  removal efficiency of the entire system was 92.05%, and the highest removal efficiency was 100%, indicating that the TSISHD had high  $\text{NH}_4^+\text{-N}$  removal and that artificial aeration could enhance ammonia removal in the soil infiltration system.

### $\text{NO}_3^-$ -N and $\text{NO}_2^-$ -N

Nitrate is an important indicator of water pollution, and its high concentration in freshwater bodies leads to eutrophication. In this study, treatment of effluent with the experimental TSISHD had a significant impact on nitrate concentration. As shown in Figure 4, the concentration of  $\text{NO}_3^-$ -N in Effluent A was  $1.25 \text{ mg L}^{-1}$  on day 1, then increased rapidly. After 15 days, the  $\text{NO}_3^-$ -N concentration in Effluent A had increased to  $40.61 \text{ mg L}^{-1}$ , and it continued to increase to the highest concentration of  $51.04 \text{ mg L}^{-1}$  until day 21.

The  $\text{NO}_2^-$ -N concentration in Effluent A was  $6.68 \text{ mg L}^{-1}$  on day 1, after which it decreased to  $0.93 \text{ mg L}^{-1}$  on day 16 with small fluctuations. The concentration then decreased to between 0.02 and  $1.24 \text{ mg L}^{-1}$ , giving an average of  $0.25 \text{ mg L}^{-1}$ .

As discussed above, the average  $\text{NH}_4^+\text{-N}$  removal efficiency in the AES was 91.80%, and it was believed that all of the  $\text{NH}_4^+\text{-N}$  was converted into  $\text{NO}_3^-$ -N. Based on these findings, a theoretical average  $\text{NO}_3^-$ -N concentration of  $4.61 \times (62/18) = 15.88 \text{ mg L}^{-1}$  could be obtained from AES. However, the average  $\text{NO}_3^-$ -N concentration detected

in Effluent A was  $26.77 \text{ mg L}^{-1}$ . This discrepancy was probably due to the influent nitrate not participating in the reaction, but being directly discharged with the effluent.

The  $\text{NO}_3^-$ -N concentration in Effluent B remained in the range of  $0.00\text{--}5.67 \text{ mg L}^{-1}$ , with an average concentration of  $0.88 \text{ mg L}^{-1}$ , giving a removal efficiency of 82.50%. These findings demonstrated that ANS had high denitrification performance.

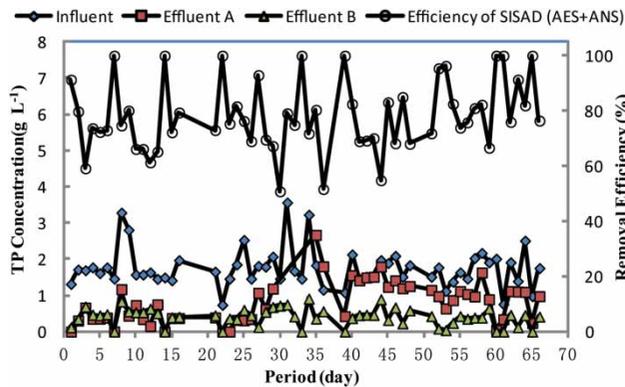
Rapid degradation of organic matter in the AES column led to a lack of carbon available to the denitrifying bacteria, and the aerobic environment in the AES column was not conducive to the growth and reproduction of denitrifying bacteria; therefore,  $\text{NO}_3^-$ -N accumulated in the column. Conversely, although there were occasional fluctuations, the  $\text{NO}_3^-$ -N removal rates in the ANS column were high, with an average removal rate of 82.5%. The removal efficacy of the TSISHDs was similar to those observed in constructed wetlands by other researchers (Prabu & Udayasoorian 2007; Calheiros *et al.* 2009; Tomar & Suthar 2011); however, the systems developed herein require much less space. In general, denitrification processes involved the initial  $\text{NO}_3^-$ -N reduction to  $\text{NO}_2^-$ -N, followed by further reduction to nitric oxide (NO), nitrous oxide ( $\text{N}_2\text{O}$ ), and finally, molecular nitrogen ( $\text{N}_2$ ) (Delwiche & Bryan 1976). The higher removal efficacy in this study was mainly because sawdust and grass powder within the anaerobic column, which contained an average of 46.27% and 42.87% C, respectively, continually dissolved, providing sufficient carbon for the growth of denitrifying bacteria.

In addition,  $\text{NO}_2^-$ -N added to the influent and generated in the AES was mostly removed by the ANS, with an average  $\text{NO}_2^-$ -N concentration of  $0.09 \text{ mg L}^{-1}$  and an average removal efficiency of 98.58% in the ANS from day 17.

### TP

As shown in Figure 5, the average TP concentration of the AES was  $0.80 \text{ mg L}^{-1}$ , giving a removal efficiency of 60.31%. The removal was mainly due to adsorption and consumption by micro-organisms.

The average TP concentration in Effluent B from the ANS was  $0.40 \text{ mg L}^{-1}$ , meeting the national quality standard of China (GB 3838-2002) for class V surface water (Table 2). In addition, the total removal efficiency of the reactor was



**Figure 5** | TP concentration and removal efficiency.

78.10%, which was better than the efficiency of 60% reported for bioretention systems (Ergas *et al.* 2010). It is believed that the volcanic materials in the mixed filters played a significant role in adsorbing phosphorus. Yang *et al.* (2013) reported that volcanic materials, such as Kunama mud, exhibited significant phosphate adsorption capacity. Furthermore, phosphate was necessary for denitrification by denitrifying bacteria because phosphorus is used by the organisms to produce ADP sulfurylase (Bruser *et al.* 2000); consequently, phosphate could be removed.

As shown in Figure 5, the ANS column had greater TP removal than the AES column. This was because the ANS column contained iron, which can induce phosphate precipitation, enabling increased TP removal from the influent water. Luanmanee *et al.* (2002) obtained a similar conclusion in a study of a MSL system for domestic wastewater treatment, indicating a good ability to remove phosphorus that was mainly related to mixed media diversity and different filter materials having the ability to absorb phosphorus. In addition, iron and phosphate in the ANS column reacted with water to generate phosphate that could not be dissolved, which resulted in the interception of phosphorus.

## CONCLUSIONS

A two-stage soil infiltration system incorporated with heterotrophic denitrification (TSISHD) was constructed for treating synthetic urban runoff. Sawdust and grass powders were added in the ANS to provide organic carbon, which

was needed for the denitrification process, while the reduction environment was improved by the addition of iron in the ANS. The results revealed that the mixed-media filled system solved the clogging problem that occurs in traditional soil infiltration systems. The system also showed good performance for treating synthetic urban runoff, and the corresponding effluent concentrations met the standards set by the national quality standard of China (Environmental Quality Standard of Surface Water in China, GB 3838-2002) for class V surface water. The removal efficiencies were significantly higher than those of the previous soil infiltration systems without inoculated microbes. The majority of COD and  $\text{NH}_4^+\text{-N}$  were removed from the AES by aerobic respiration and nitrification, respectively. Phosphorus was primarily removed by adsorption, while nitrate and nitrite were reduced by heterotrophic denitrification in the ANS. Further investigations will be carried out in aspects of the design optimization by response surface methodology to adapt to different urban runoff regimes.

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## REFERENCES

- Bruser, T., Selmer, T. & Dahl, C. 2000 'ADP sulfurylase' from *Thiobacillus denitrificans* is an adenylsulfate: phosphate adenyltransferase and belongs to a new family of nucleotidyltransferases. *J. Biol. Chem.* **275**, 1691–1698.
- Butterworth, E., Dotro, G., Jones, M., Richards, A., Onunkwo, P., Narroway, Y. & Jefferson, B. 2013 Effect of artificial aeration on tertiary nitrification in a full scale subsurface horizontal flow constructed wetland. *Ecol. Eng.* **54**, 236–244.
- Calheiros, C. S. C., Rangel, A. O. S. S. & Castro, P. M. L. 2009 Treatment of industrial wastewater with two-stage constructed wetlands planted with *Typha latifolia* and *Phragmites australis*. *Bioresour. Technol.* **100**, 3205–3213.

- Chinalia, F. A., Garbossa, H. P., Rodriguez, J. A., Lapa, K. R. & Foresti, E. 2012 A two-stage aerobic/anaerobic denitrifying horizontal bioreactor designed for treating ammonium and H<sub>2</sub>S simultaneously. *Appl. Biochem. Biotechnol.* **168**, 1643–1654.
- Delwiche, C. C. & Bryan, C. C. 1976 Denitrification. *Annu. Rev. Microbiol.* **30**, 241–262.
- Dong, H., Qiang, Z., Li, T., Jin, H. & Chen, W. 2012 Effect of artificial aeration on the performance of vertical-flow constructed wetland treating heavily polluted river water. *J. Environ. Sci. (China)* **24**, 596–601.
- Ergas, S. J., Sengupta, S., Siegel, R., Pandit, A., Yao, Y. F. & Yuan, X. 2010 Performance of nitrogen-removing bioretention systems for control of agricultural runoff. *J. Environ. Eng.* **136**, 1105–1112.
- Fowler, D., Pilegaard, K., Sutton, M. A., Ambus, P., Raivonen, M., Duyzer, J., Simpson, D., Fagerli, H., Fuzzi, S., Schjoerring, J. K., Granier, C., Neftel, A., Isaksen, I. S. A., Laj, P., Maione, M., Monks, P. S., Burkhardt, J., Daemmgen, U., Neiryneck, J., Personne, E., Wichink-Kruit, R., Butter-Bahl, K., Flechard, C., Tuovinen, J. P., Coyle, M., Gerosa, G., Loubet, B., Altimir, N., Gruenhage, L., Ammann, C., Cieslik, S., Paoletti, E., Mikkelsen, T. N., Ro-Poulsen, H., Cellier, P., Cape, J. N., Horvath, L., Loreto, F., Niinemets, U. & Palmer, P. I. 2009 Atmospheric composition change: ecosystems–atmosphere interactions. *Atmos. Environ.* **43**, 5193–5267.
- Hou, L. Z. & Zhang, X. 2014 Comparing urban runoff quality of a felt roof and an asphalt road in Beijing. *Hydrol. Res.* **45** (2), 282–291.
- Hou, L. Z., Liu, F., Feng, C. P. & Wan, L. 2013 Efficiencies of multilayer infiltration systems for the removal of urban runoff pollutants. *Water Sci. Technol.* **67** (8), 1851–1858.
- Kong, Z., Feng, C. P., Chen, N., Tong, S., Zhang, B. G., Hao, C. B. & Chen, K. 2014 A soil infiltration system incorporated with sulfur-utilizing autotrophic denitrification (SISSAD) for domestic wastewater treatment. *Bioresour. Technol.* **159**, 272–279.
- Li, L., Yin, C., He, Q. & Kong, L. 2007 Contribution of pollution load of storm runoff in urban areas of Hanyang, Wuhan City on the receiving water. *China Environ. Sci.* **27** (3), 312–316 (in Chinese).
- Luanmanee, S., Attanandana, T. & Masunaga, T. 2001 The efficiency of a multi-soil-layering system on domestic wastewater treatment during the ninth and tenth years of operation. *Ecol. Eng.* **18** (2), 185–199.
- Luanmanee, S., Boonsook, P., Attanandana, T. & Wakatsuki, T. 2002 Effect of organic components and aeration regimes on the efficiency of a multi-soil-layering system for domestic wastewater treatment. *Soil Sci. Plant. Nutr.* **48** (2), 125–134.
- Masunaga, T. & Sato, K. 2007 Characteristics of wastewater treatment using a multi-soil-layering system in relation to wastewater contamination levels and hydraulic loading rates. *Soil Sci. Plant Nutr.* **53** (2), 215–223.
- Murakami, M., Sato, N., Aneqawa, A., Nakada, N., Harada, A., Komatsu, T., Takada, H., Tanaka, H., Ono, Y. & Furumai, H. 2008 Multiple evaluations of the removal of pollutants in road runoff by soil infiltration. *Water Res.* **42**, 2745–2755.
- Pell, M. & Nyberg, F. 1989 Infiltration of wastewater in a newly started pilot sand-filter system: reduction of organic matter and phosphorus. *J. Environ. Qual.* **18** (4), 451–475.
- Pozo-Morales, L., Franco, M., Garvi, D. & Lebrato, J. 2013 Influence of the stone organization to avoid clogging in horizontal subsurface-flow treatment wetlands. *Ecol. Eng.* **54**, 136–144.
- Prabu, P. C. & Udayasoorian, C. U. 2007 Treatment of pulp and paper mill effluent using constructed wetland. *EJEAFChe* **6** (1), 1689–1701.
- Sato, K., Masunaga, T. & Wakatsuki, T. 2005 Characterization of treatment processes and mechanisms of COD, phosphorus and nitrogen removal in a multi-soil-layering system. *Soil. Sci. Plant. Nutr.* **51** (2), 213–221.
- Tota-Maharaj, K. & Scholz, M. 2010 Efficiency of permeable pavement systems for the removal of urban runoff pollutants under varying environmental conditions. *Environ. Prog. Sustain.* **29** (3), 358–369.
- Tomar, P. & Suthar, S. 2011 Urban wastewater treatment using vermi-biofiltration system. *Desalination* **282**, 95–103.
- Wakatsuki, T., Esumi, H. & Omura, S. 1993 High-performance and N-removable and P-removable on-site domestic wastewater treatment system by multi-soil layering method. *Water Sci. Technol.* **27** (1), 31–40.
- Yang, S. J., Ding, D. H., Zhao, Y. X., Huang, W. L., Zhang, Z. Y., Lei, Z. F. & Yang, Y. N. 2013 Investigation of phosphate adsorption from aqueous solution using Kanuma mud: behaviors and mechanisms. *J. Environ. Chem. Eng.* **1**, 355–362.
- Zhang, W., Zhang, S. C., Wan, C., Yue, D. P., Ye, Y. B. & Wang, X. J. 2008 Source diagnostics of polycyclic aromatic hydrocarbons in urban road runoff, dust, rain and canopy throughfall. *Environ. Pollut.* **153**, 594–601.
- Zhang, W., Che, W., Liu, D. K., Gan, Y. P. & Lv, F. F. 2012 Characterization of runoff from various urban catchments at different spatial scales in Beijing, China. *Water Sci. Technol.* **66** (1), 21–27.
- Zhao, C., Zheng, Z. Y., Zhang, J., Wen, D. H. & Tang, X. Y. 2013 Adsorption characteristics of ammonium exchange by zeolite and the optimal application in the tertiary treatment of coking wastewater using response surface methodology. *Water Sci. Technol.* **67** (3), 619–627.

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