Comparative study on nutrient removal of agricultural non-point source pollution for three filter media filling schemes in eco-soil reactors

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ABSTRACT

Nutrients (nitrogen and phosphorus) from agricultural non-point source (NPS) pollution have been increasingly recognized as a major contributor to the deterioration of water quality in recent years. The purpose of this article is to investigate the discrepancies in interception of nutrients in agricultural NPS pollution for eco-soil reactors using different filling schemes. Parallel eco-soil reactors of laboratory scale were created and filled with filter media, such as grit, zeolite, limestone, and gravel. Three filling schemes were adopted: increasing-sized filling (I-filling), decreasing-sized filling (D-filling), and blend-sized filling (B-filling). The systems were intermittent operations via simulated rainstorm runoff. The nutrient removal efficiency, biomass accumulation and vertical dissolved oxygen (DO) distribution were defined to assess the performance of eco-soil. The results showed that B-filling reactor presented an ideal DO for partial nitrification–denitrification across the eco-soil, and B-filling was the most stable in the change of bio-film accumulation trends with depth in the three fillings. Simultaneous and highest removals of NH_4^+-N (57.74–70.52%), total nitrogen (43.69–54.50%), and total phosphorus (42.50–55.00%) were obtained in the B-filling, demonstrating the efficiency of the blend filling schemes of eco-soil for oxygen transfer and biomass accumulation to cope with agricultural NPS pollution.

Key words | agricultural non-point source pollution, bio-film storage, eco-soil, filter filling schemes, nutrient removal, vertical dissolved oxygen distribution

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INTRODUCTION

With the development of agriculture and the increasing use of chemical fertilizers and pesticides rich in nutrients (nitrogen and phosphorus) in China, agricultural non-point source (NPS) pollution has become a primary threat in watershed environments (Ouyang *et al.* 2014; Shen *et al.* 2014). The management of agricultural NPS pollution has become a hot-spot in the area of water pollution control because of the characteristics of wide range, difficult control and complex uncertainties (Huang *et al.* 2014; Ronlyn 2014; Shen *et al.* 2014). Storm runoff as the carrier transporting pollutants is a major cause of agricultural NPS pollution (Guo *et al.* 2014), and picks up and carries natural and anthropogenic pollutants, finally depositing them into lakes, rivers and wetland systems. A flooding situation may arise in the first flush of storm-water (Sharma 2008) which will contaminate water bodies with pollutants like nutrients. Nitrogen and phosphorus are known to be lethal water pollutants in water bodies including lakes, rivers and reservoirs. The enrichment of aquatic environments with N and P can have an undesirable influence on their trophic state, and will lead to toxic algal blooms (Sanford & Pope 2013), oxygen depletion and loss of biodiversity (Shen *et al.* 2012). In addition, it also decreases the quality of water used for drinking, agriculture and other purposes (Hong *et al.* 2012). Therefore, nutrient removal is the key to dealing with agricultural NPS pollution. In China, the control and

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management of agricultural NPS pollution will be one of the most serious issues for water environmental protection in the next several decades (Shen *et al.* 2012).

Constructed wetlands have been one of the most efficient methods to control agricultural NPS pollution (Diaz *et al.* 2012; Zhang *et al.* 2012). The benefits of a constructed wetland are its high efficiency without the input of fossil energy, and thereby operational cost-effectiveness and easy handling. Nutrients can be removed by employing constructed wetlands, through a complex interconnected system of plants, media and biomass population (Fountoulakis *et al.* 2009). However, each technology has its own advantages and disadvantages. Constructed wetlands cannot cope with the agricultural NPS pollution effectively caused by rainstorm runoff in a short time and large flux during the rainy season (Tanveer & Sun 2012), due to the long reaction cycle. Eco-soil processing systems consisting of a mineral medium layered filling, are similar to the vertical flow constructed wetlands. The mineral medium and micro-organisms in the system provide treatment of wastewater on the basis of soil infiltration treatment technology. However, the difference is that this system changes the soil to mineral media, to provide greater voids for hydraulic conductivity and to attach more microbes, so that the efficiency of decontamination compared with the traditional soil infiltration system is improved. In eco-soil processing systems, the transformation and removal of nutrients are accomplished by interaction with penetration, adsorption, sedimentation, and biological nutrient removal processes. The runoff pollution would enter into the water bodies via the eco-soil system, which is located in the transition zone between farmland and water bodies around the waterway. As a green treatment technology, eco-soil has the unique advantage of producing higher effluent quality without using effective arable land and frequent maintenance. In addition, there is a certain aesthetic value.

Research has found that an eco-soil system has a better removal effect for nutrients, but different packing schemes with different media influence the decontamination ability of the system at the early stage of the experiment. This experiment was designed to investigate the possibility of utilization and discrepancies in the interception of nutrients in agricultural NSP pollution for three filter media filling schemes in eco-soil under simulated storm runoff. The objectives of this study are: (1) to test the overall and inter-laminar efficiency of eco-soil performances in terms of dissolved oxygen (DO) transfer, bio-film storage, and nutrient removal; and (2) to examine the treatment stability simulated by different storm runoff durations and loads. Thus, the outcomes will provide a useful reference for media packing strategy in eco-soil design.

MATERIALS AND METHODS

Experimental setup

Three identical eco-soil reactors were manufactured and placed on the campus of Jiangsu University, Zhenjiang. A diagram of the reactor and experimental setups are shown in Figure 1. The square filter column of each reactor was manufactured from opaque PVC sheets that were 0.4 m in width and 1.0 m in height. The filter column is vertically divided into four layers. The height and volume of each layer is 0.2 m and 0.032 m^3, respectively. Four kinds of materials were employed as the filter medium in the system, with the advantages being high microorganism loading and better detergency (retaining capacity of pollutants in the wastewater) as shown in other studies of constructed wetlands (Zhang *et al.* 2012). They are grit, zeolite, limestone, and gravel, and their chemical and physical properties are shown in Table 1. Three filter filling schemes were adopted: (A) increasing-sized filling (I-filling): the filter media was filled in a vertically increasing particle size manner from top to bottom; (B) decreasing-sized filling (D-filling): the opposite of I-filling; (C) blend-sized filling (B-filling): the filter media filling material was blended. The plant did not show significant effects on the removal of nutrients during a short period (1 or 2 hours), which was found in a previous study. Moreover, this experiment was carried out in autumn which is not a planting season. Therefore, the experimental setups were unplanted. For each layer, a PVC perforated pipe ($\varphi = 2.0$ cm) was installed in the center of the column for sample collection. Before the experiment, the reactors were shaken several times for filter media compaction. Flow distribution and harvesting devices were installed on the top and bottom of the reactor, respectively.

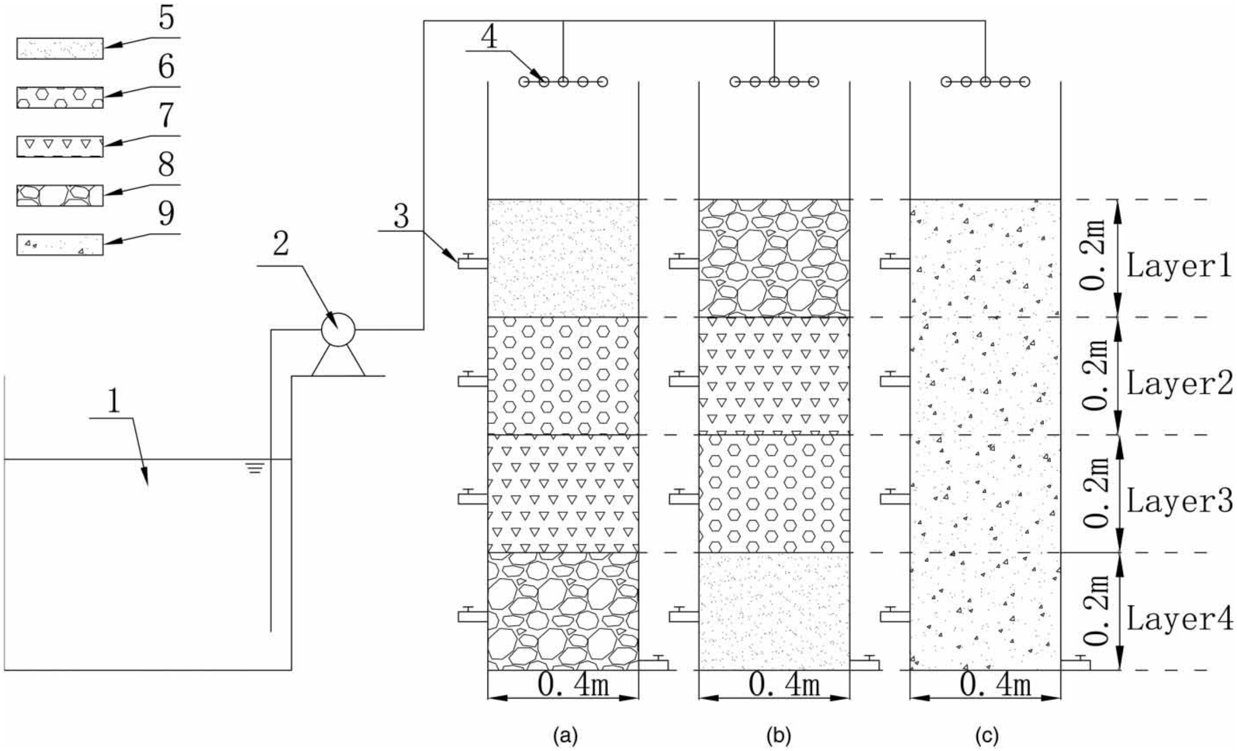

Figure 1 | Schematic description of eco-soil reactor with different media filling schemes. (a) I-filling; (b) D-filling; (c) B-filling. 1, tank; 2, peristaltic pump; 3, sample connection; 4, water distributor; 5, grit; 6, zeolite; 7, limestone; 8, gravel; 9, blend.

Table 1 | The chemical and physical properties of filter media

Parameter	Grit	Zeolite	Limestone	Gravel
Size (mm)	1–2	3–5	7–10	8–15
Porosity (%)	23	64	47	34
Al (mg/g)	96.3	85.3	30.8	68.3
Fe (mg/g)	13.0	20.1	8.9	34.4
Ca (mg/g)	27.1	28.6	260.3	80.2
Mg (mg/g)	9.8	14.0	20.9	5.2

Experimental flow and operations

Artificial wastewater was prepared and stored in a feed tank, which was pumped into the reactor to keep a constant water head. For the two loads, major nutrients were supplied by adding KNO_3 and K_2HPO_4, and organic carbon sources were supplied by adding glucose to the tap water. Intermittent operations were carried out via simulated rainstorm runoff, each operation lasting for 20 minutes and 50 minutes, because agricultural NPS pollution mainly occurs at the beginning of the rainfall (Guo *et al.* 2014). According to the literature, the hydraulic loading of 0.85 $m^3/(m^2 \cdot d)$ was used throughout the experimental period. Two weeks before the start of the experiment, the reaction device was stabilized to cultivate the growth of microorganisms. Organic loading rate is an important factor causing growth of microorganisms of eco-soil. The chemical oxygen demand (COD) and suspended solids (SS) concentration of agricultural NPS pollution water in Zhenjiang reached 40–110 mg/L and 90–150 mg/L, respectively, according to our irregular water quality detection. Nevertheless, in order to shorten the time required for maturity of microorganisms and to assess the bio-film storage in the laboratory accurately, higher organic matter concentrations of 150–400 mg/L and low levels of SS, relatively, were adopted in the wastewater used in this study. The water quality of the synthetic wastewater is listed in Table 2. The average air temperature was around 12–25 °C. A bed resting time of 7 days and flushing with wastewater (keeping the filter medium moist, to maintain the growth of

Table 2 | Average flow water quality of low and high load (mg/L)

Operations	COD	NH_4^+-N	NO_3^--N	TN	TP	DO	pH
Load 1	184 ± 11.41	26.60 ± 1.72	7.60 ± 2.11	36.12 ± 1.43	0.91 ± 0.34	3.42 ± 1.03	7.56 ± 0.32
Load 2	244 ± 19.82	48.00 ± 2.34	12.35 ± 1.64	64.44 ± 2.23	1.60 ± 0.24	3.26 ± 1.21	7.42 ± 0.22

microorganisms) was used for recovery of the system between the two operational periods of simulated rainstorm runoff.

Wastewater sampling and analysis

Samples collected from flow, porous water in each layer, and effluent were analyzed in the laboratory immediately. Ammonia (NH_4^+-N), nitrate (NO_3^--N), total nitrogen (TN), and total phosphorus (TP) concentrations were measured according to Standard Methods (State Environmental Protection Administration of China 2002). DO and pH were tested using a portable DO/pH meter (Multi3420, WTW).

Determination of bio-film storage

The biomass accumulation rate (BAR) in each layer of eco-soil was used to assess the bio-film storage. BAR is defined as the decrease in the media water storage volume over time due to bio-film accumulation, written as $BAR = (V_{0i} - V_{ti})/V_0$, where V_{0i} is the initial void in layer i evaluated according to effluent wastewater volume. V_{ti} refers to the water storage volume after t days of operation (Song et al. 2015). The flow of the experiment was artificial nutrient wastewater, and the decrease in porosity of the filter column caused by SS can be ignored.

RESULTS AND DISCUSSION

DO distribution in each layer

The void fraction and average DO concentration in each layer were measured before and after the reaction of simulated rainstorm runoff, respectively. The bar of the vertical DO level changes and void fraction are illustrated in Figure 2. Oxygen is a crucial environmental parameter that controls nitrification and denitrification. The unsaturated media of eco-soil promotes higher atmospheric oxygen diffusion inside the medium pores, which can boost nitrification. The DO concentration shows a decreasing trend for all three filling schemes from top to bottom. The DO concentration ranged from 4.4 to 0.4 mg/L, 5.7 to 1.9 mg/L, and 5.2 to 1.6 mg/L for the I-filling, D-filling, and B-filling reactors, respectively. The DO was primarily affected by the porous transfer efficiency. The larger size of filter medium in the upper layer of the D-filling and

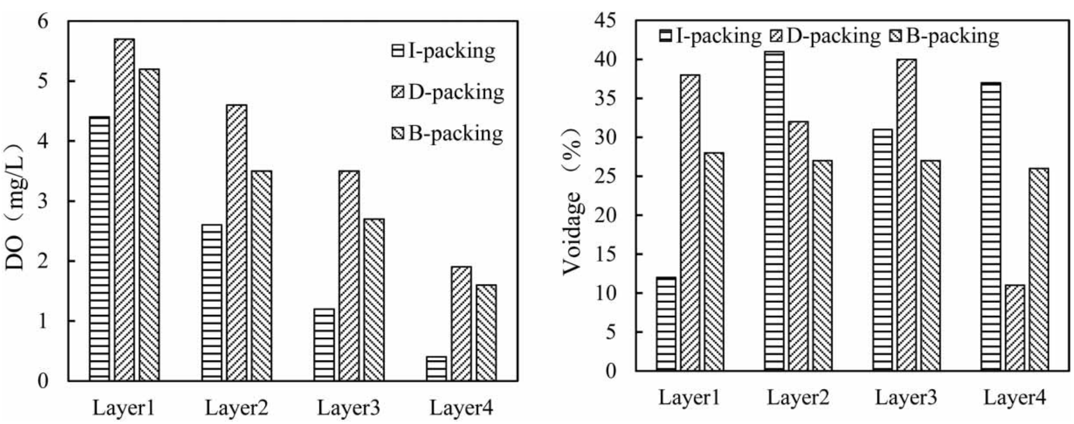

Figure 2 | DO distribution and void fraction in each layer of eco-soil for the three filling schemes.

B-filling were advantageous for atmospheric oxygen transfer, which indicated atmospheric re-oxygenation is the predominant source for eco-soil reactors (Wu *et al.* 2011). The decrease rate of DO reaching 0.67 mg/L/10 cm in the I-filling reactor was higher than the value of 0.63 mg/L/10 cm for the D-filling reactor and 0.6 mg/L/10 cm for the B-filling reactor. It is generally accepted that DO concentrations above 2.0 mg/L are essential for nitrification to occur and below 0.5 mg/L for denitrification (Bertino 2010; Zhang *et al.* 2011; Song *et al.* 2015). The DO concentration of the I-filling reactor was below 0.5 mg/L in layer 4, which indicated a relatively ideal aerobic-to-anoxic condition that was beneficial for nitrification and denitrification processes, while the D-filling reactor provided relatively higher DO levels in all layers. For the B-filling reactor, the aerobic–anoxic conditions coexist in the medium of partial space, because of the mixed loading of different size media. However, the difference is that the aerobic–anoxic condition occurred in the bottom layer of the I-filling reactor and all layers of the B-filling reactor.

Bio-film storage

The changes of BAR in each layer of eco-soil reactors are illustrated in Figure 3. In all layers, the BAR gradually increased during the entire operation. The bio-film accumulation of different layers in the three reactors obviously differed. The BAR in layer 1 for the B-filling and I-filling reactors increased to a maximum of 17.2% and 13.2% after 42 days of operation, respectively. The biomass accumulation was 8.8–12.8% higher than in the D-filling reactor, probably in the form of nitrifier or other microbes. Conversely, this increase also occurs in layer 4 in the D-filling reactor, probably in the form of denitrifying bacteria. In layers 2 and 3, the BAR and its trend of gradual increase of the three fillings was similar during the entire operation. This difference indicates a rapid reduction in the infiltration rate and effective porosity due to biomass growth. The bio-film accumulation positively correlated with the depth for the D-filling reactor and negatively correlated with the depth for I-filling and B-filling reactors. However, B-filling

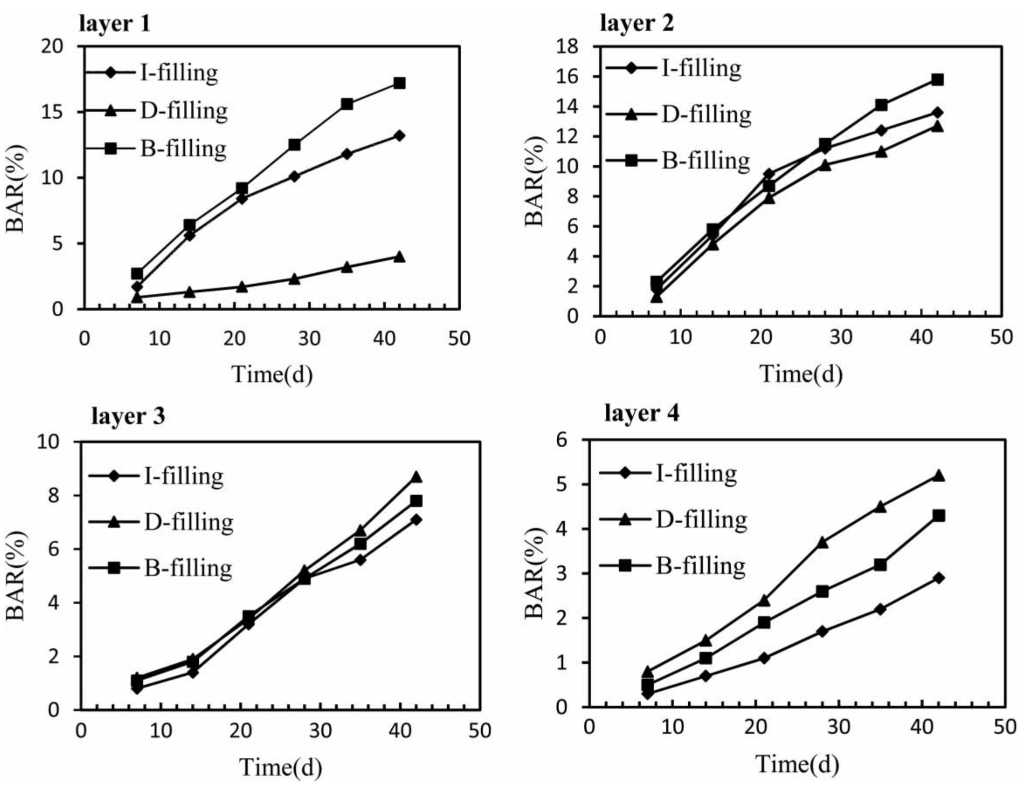

Figure 3 | BAR in each layer of eco-soil reactor for sequential influent operation.

was the most stable of changes of bio-film accumulation trends in the three fillings. In addition, the DO in all the layers of the D-filling and B-filling reactors remains at a relatively high level despite bio-film accumulation. This finding points to atmospheric re-oxygenation still serving as a major DO source, moreover, which correlated with nitrification–denitrification and the phosphorus removal of phosphorus accumulating bacteria.

Nutrient removal efficiency

Table 3 summarizes the mean pollutant removal efficiency of the eco-soil reactors concerning different filling schemes and nutrient loads. Generally, the removal of nutrients was more effective in the B-filling reactor than the I-packing and D-filling reactors. The removal efficiency of nutrients for all filling schemes has a tendency to weaken with the growth of the simulated storm-rain runoff duration under different loads. The nitrogen removal rate generally was not as good as TP for all filling strategies.

For low load 1 operation, the B-filling reactor showed an efficient ammonia oxidation and adsorption by media of 64.47–57.74% over D-filling of 58.53–50.49% and I-filling of 53.68%–46.505, reaching a concentration of 9.45–11.24 mg/L in the effluent. The increased oxygen transport capacity in the upper layer due to the larger media size provides the aerobic conditions which make ammonia easily converted to nitrate by nitrobacteria (Wu *et al.* 2011; Song *et al.* 2015). A lower effluent nitrate concentration and larger TN removal rate (49.25–43.69%) demonstrate that denitrification in the B-filling reactor is more efficient than in the I-filling and D-filling reactors. Accordingly, the B-filling reactor could enhance the TN removal rates via the partial nitrification–denitrification process in all layers. Notable differences in the interception efficiency of TP were detected among the three filling schemes. The TP removal rate was 54.95–48.35%, and the content decreased sharply to 0.47–0.41 mg/L for the B-filling reactor, while the TP removal rates were 49.45–41.76% and 42.86–37.36% for the I-filling and D-filling reactors, respectively. TP removal via the adsorption and precipitation reaction in eco-soil is governed by metallic elements (Ca, Al, Fe, and Mg) of specific media and the content of TP in the water. The cause of this difference in TP removal rates may be contact with different contact times with special medium. Contact time was longer in B-filling than I-filling and D-filling due to the even distribution of medium, while there was layered distribution in the latter.

TP removal is generally lower for high load operation. Instead, the NH_4^+-N and TN removal rate of all packing

Table 3 | Mean pollutant removal performances of the eco-soil reactors concerning different filling schemes and nutrient loads, mg/L

			I-filling reactors		D-filling reactors		B-filling reactors	
Operation	**Duration**	**Parameter**	**Effluent**	**Removal**	**Effluent**	**Removal**	**Effluent**	**Removal**
Load 1	20 minutes	NH_4^+-N	12.32 ± 1.25	53.68%	11.03 ± 2.21	58.53%	9.45 ± 2.34	64.47%
		NO_3^--N	5.46 ± 1.16	–	9.28 ± 1.53	–	7.81 ± 2.16	–
		TN	19.24 ± 3.27	46.73%	23.48 ± 3.31	34.99%	18.33 ± 2.45	49.25%
		TP	0.46 ± 0.17	49.45%	0.52 ± 0.13	42.86%	0.41 ± 0.12	54.95%
	50 minutes	NH_4^+-N	14.23 ± 2.31	46.50%	13.17 ± 2.45	50.49%	11.24 ± 1.19	57.74%
		NO_3^--N	6.03 ± 1.23	–	10.42 ± 2.53	–	7.35 ± 2.48	–
		TN	21.14 ± 1.32	41.47%	25.67 ± 3.21	28.93%	20.34 ± 2.65	43.69%
		TP	0.53 ± 0.14	41.76%	0.57 ± 0.26	37.36%	0.47 ± 0.10	48.35%
Load 2	20 minutes	NH_4^+-N	17.42 ± 2.21	63.71%	15.28 ± 3.42	68.17%	14.15 ± 2.36	70.52%
		NO_3^--N	10.16 ± 1.41	–	14.75 ± 3.27	–	13.21 ± 2.48	–
		TN	29.06 ± 2.04	54.90%	32.51 ± 3.11	49.55%	29.32 ± 3.10	54.50%
		TP	0.87 ± 0.14	45.63%	0.94 ± 0.27	41.25%	0.72 ± 0.26	55.00%
	50 minutes	NH_4^+-N	19.22 ± 3.17	59.96%	18.02 ± 2.34	62.46%	16.71 ± 1.94	65.19%
		NO_3^--N	11.06 ± 1.75	–	13.58 ± 3.24	–	13.06 ± 2.12	–
		TN	32.12 ± 3.21	50.16%	34.77 ± 3.25	46.04%	31.39 ± 3.52	51.29%
		TP	1.03 ± 0.22	35.63%	1.10 ± 0.31	31.25%	0.92 ± 0.27	42.50%

'–'means the removal rate is negative.

reactors increased approximately 10% compared to operation at low load. This difference might be caused by the significant DO consumption for high organic loads, which enlarges the anoxic areas and thus enhances denitrification when sufficient carbon is available (Zhang *et al.* 2011; Song *et al.* 2015). However, the reason for lower TP removal is that the power on medium adsorption of pollutants can be attenuated in the face of a high pollution load to a certain extent.

Nutrient removal mechanisms in each layer of reactors

The nutrient removal in different layers of eco-soil reactors is presented in Figure 4. The NH_4^+-N removal was higher in layers 2 and 3 of the I-filling and D-filling reactors, while it was achieved in all layers of the B-filling reactor. The rapid consumption of NH_4^+-N can only occur when the surrounding oxygen of the bio-film is sufficient in the filter and the filling layer of the filter has strong adsorption ability for polar contaminants. Despite high re-oxygenation in the D-filling reactor, the small specific surface area of the media in the top layers reduced the interaction between the nutrients and the bio-film due to the large particle size (Song *et al.* 2015). The TN removal rate was 14% on average among layers, and the highest removal was achieved in the bottom layer of the I-filling reactor. The D-filling reactor was less effective for TN removal. The TN removal was higher in the B-filling reactor compared to the I-filling reactor; significant differences were not detected among layers. Considering the fact that oxygen is more preferred in denitrification, the DO concentration should be maintained at <0.3–0.5 mg/L, to accomplish the denitrification process (Bertino 2010). As such, the process of bio-film growth with lower DO content and anoxic zones (Lee *et al.* 2009) had been observed in the bottom of the reactor (in Figure 3). These results indicate partial nitrification–denitrification across the eco-soil systems of the B-filling reactor, which could be attributed to the employment of blend filling, favorable for the growth of ammonium

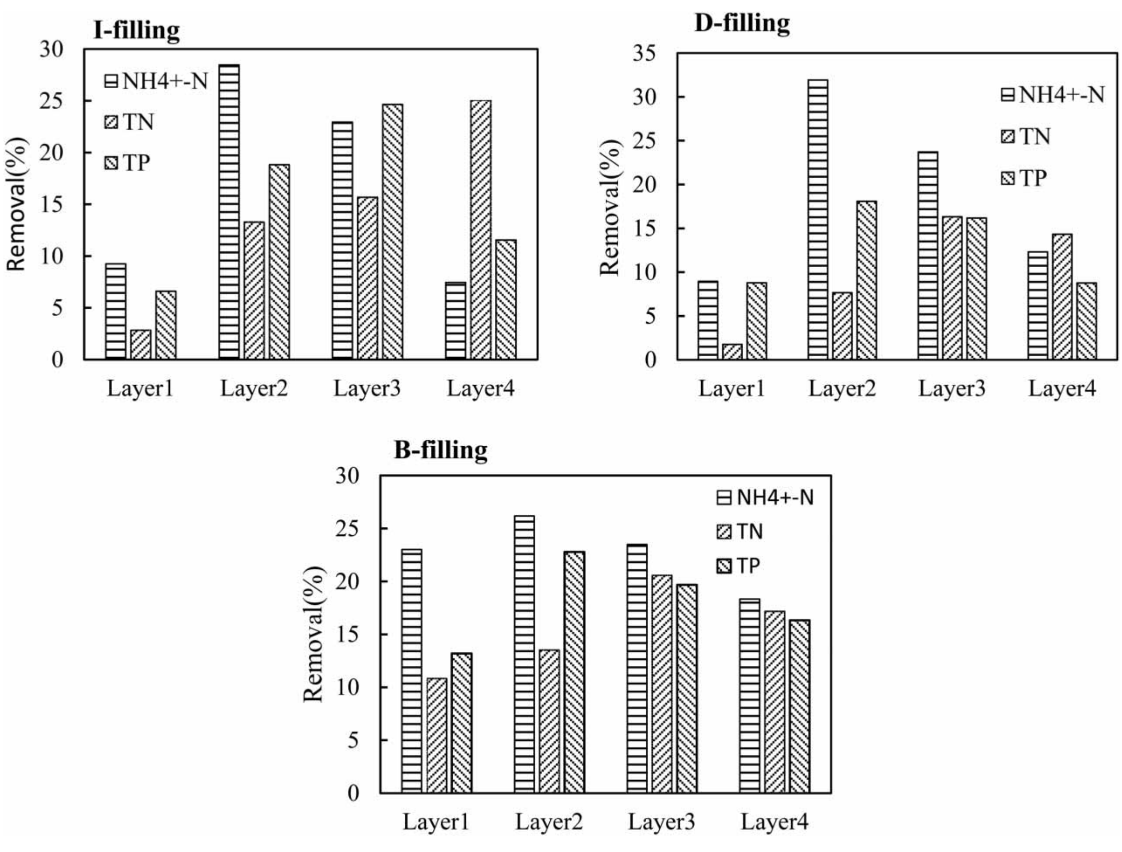

Figure 4 | Inter-laminar nutrient removal efficiency during load 1.

oxidizer bacteria and nitrate oxidizers (Zhang *et al.* 2011) by enhanced concurrent aerobic–anoxic conditions.

TP removal was not significantly different in the I-filling reactor compared to the D-filling reactor, with significant removal in the layer of limestone and zeolite. TP removal was generally not as effective as it was through each layer of medium in the B-filling reactor. The rapid consumption of TP can occur via the adsorption and precipitation reaction between phosphate and filter medium rich in metallic elements (Ca, Al, Fe, and Mg). These results indicate that TP removal could be attributed to the employment of limestone media enhanced wastewater alkalinity, favorable for the precipitation reaction between calcium and phosphate.

CONCLUSIONS

Laboratory scale model eco-soil reactors were designed to explore the interception of nutrients in agricultural NPS pollution, and have shown that the B-filling reactor provides efficient TN and TP removal. The I-filling reactor presents an aerobic-to-anoxic transition area for efficient TN removal, but TP removal was less effective compared to the B-filling reactor. The D-filling reactor does not appear to be efficient for either TN or TP removal. The B-filling reactor can maintain relatively stable intercept ability of nutrients during the growth of the simulated rainstorm runoff load and duration, and is suitable for treatment of agricultural NPS pollution caused by rainstorm runoff. If properly operated, this type of filter filling structure offers several advantages for pollutant removal, such as increased bio-film accumulation and prolonged contact time of pollutants with the medium. However, in order to evaluate the microorganism growth of systems accurately, the influence of SS on the decontamination effect of the system has been ignored in this study. The influence of SS on the system in practical application will be reflected in a further study.

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