1 **Treatment of artificial secondary effluent for effective**
2 **nitrogen removal using a combination of corncob carbon**
3 **source and bamboo charcoal filter**

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ABSTRACT

Nitrogen from secondary effluents has endangered the aquatic environment and human health. A new experimental setup combining a corncob carbon source and bamboo charcoal filter (BCF) was used to effectively purify artificial sewage treatment plant tail water that had high nitrate and nitrogen content, and a low carbon to nitrogen (C/N) ratio. The nitrogen concentration in the reactor's effluent was below the limit of 15 mg/L when the influent COD concentration was 20 mg/L–60 mg/L, the influent NO_3^- -N concentration was 15 mg/L–30 mg/L, and the hydraulic retention time (HRT) was 1 h–4 h. The reactor's maximum volumetric load of total nitrogen was 0.585 kg/(m³·d). The COD and NO_3^- -N balances of the reactor indicated that 4.77 g corncob was required to remove 1 g NO_3^- -N and 2 g corncob was required to release 1g COD.

Keywords: denitrification; carbon sources; corncob; bamboo charcoal; materials balance

1. Introduction

Restriction on nitrogen and phosphorus effluent concentrations is critical for prevention of eutrophication in lakes (Abell et al., 2010). Regulations regarding nutrient concentrations in wastewater have been enforced in China (Paerl et al., 2011). However, several conventional sewage treatment plants need to be extended or upgraded to satisfy more stringent nutrient discharge standards (Jeong et al., 2006). This is a difficult task as it requires additional land, money, and time (Jokela et al., 2002).

Although frequent fluctuations in water quality can cause an excess of carbon sources in the effluent water, organic matter content is normally low in sewage in China. Consequently, carbon source required for denitrification during secondary processing is insufficient resulting in high nitrate and nitrogen levels, and a low carbon to nitrogen (C/N) ratio in the sewage tail water (Chiu et al., 2007; Guo et al., 2010; Her and Huang, 1995; Ryu et al., 2008). Commonly, carbon sources can be divided into two broad categories: traditional and new. Traditional carbon sources contain low molecular organic matter and sugars (Karanasios et al., 2016). Liquid organic matter, such as glucose, methanol, ethanol, and acetic acid, are preferred as they are easily used by denitrifying bacteria (Bu et al., 2015; Lim et al., 2006). Sugars such as glucose and sucrose are already in use as additional carbon sources. New carbon sources are solid sources that contain natural plant cellulose materials and biodegradable polymers (Ines et al., 1998; Luo et al., 2014; Luo et al., 2016; Volokita et al., 1996a; Volokita et al., 1996b).

In recent years, intensive research has been conducted on solid carbon sources primarily consisting of cellulose. Fibre carbon sources can be easily acquired at a low cost and mainly include cotton (Volokita et al., 1996a), paper (Volokita et al., 1996b),

straw (Ines et al., 1998), and reed rod. Corncobs are an attractive external carbon source for denitrification (Wang et al., 2013; Xu et al., 2009; Yang et al., 2015). Additionally, in view of high nitrate and nitrogen levels, and low C/N in the tail water of sewage treatment plants, effective denitrification needs new adsorption materials to serve as fillers in biological filters (Jia et al., 2013; Ryu et al., 2009). Bamboo charcoal can be used as a filler for adsorption packing to enable microbial degradation (Chen et al., 2012). A combination of corncob carbon source and bamboo charcoal filter (BCF) may be a feasible solution.

This study aims to utilize a cheap agricultural waste as the solid carbon source and bamboo charcoal as fillers of denitrifying biological filter to purify sewage treatment plant tail water effectively. In view of high nitrate and nitrogen content, and a low C/N, the optimization parameters for continuous operation at different water loads, hydraulic retention time, height of packing reactor, and running effect of the denitrifying biological filter were investigated to ensure effluent compliance with the GB18918-2002 1A discharge standard (MOEP, 2002). It expands the application of new, cheap, and harmless materials and provides a theoretical and design basis for the denitrification process in sewage treatment plants.

2. Materials and methods

2.1 Experimental devices

The experimental device setup is displayed in Fig. 1. The experimental devices contained a solid carbon source reactor and a denitrification filter that were made of polymethyl methacrylate. The carbon source reactor had an inner diameter of 80 mm and a height of 320 mm with an effective volume of 1206 L. Simulated municipal wastewater was pumped from the bottom to the top through a porous retainer plate and then passed through the solid carbon source using a peristaltic pump. The

denitrification filter was a fixed bed reactor filled with bamboo charcoal. The particle size of bamboo charcoal was reduced to 2 mm–4 mm using a special crusher after soaking, cleaning, and passing through a 6 mm–10 mm mesh. The effluent flowed from top to bottom into the denitrification filter with an inner diameter of 80 mm and a height of 650 mm with an effective volume of 0.785 L. The total effective volume was about 2 L when the fill height was 400 mm. The perforation aperture of the porous retainer plate was 1.5 mm. There were four sampling openings every 80 mm above the porous retainer plate. Backwash using water and compressed air was designed to prevent filter clogging.

[Fig. 1 was inserted here]

2.2 Operating conditions

Operating conditions of the experimental devices are shown in Table 1. Different influential concentrations, fluxes, and hydraulic retention times were maintained at different times.

2.3 Experiment analysis

Sampling was arranged every three days to ensure all operating conditions were tested. The glucose, ammonium acetate, potassium nitrate, and potassium dihydrogen phosphate were used to add COD, $\text{NH}_3\text{-N}$, $\text{NO}_3^-\text{-N}$, and TP, respectively. Three water samples were collected each time in the influent, corncob effluent and BCF effluent. The corncob removal rate represented the removal rate between the influent and corncob effluent. The total removal rate represented the removal rate between the influent and BCF effluent. The levels of COD and $\text{NH}_3\text{-N}$ were analysed according to standard methods (Zhou et al., 2015a). $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ levels were determined using ion chromatography (ICS-900, Thermo, USA).

[Table 1 was inserted here]

3. Results and discussion

3.1 Solid carbon source

After the completion of bamboo charcoal hanging membrane, corncob was added to the experimental reactor. The effect of different quantities of solid carbon source on denitrification was compared, as shown in Fig. 2a. The removal rate of NO_3^- -N was found to become higher as the quantity of corncob increased. With an addition of 60 g, 140 g, and 200 g of corncob, the removal rate of NO_3^- -N was 38.1%, 51.0%, and 67.1%, respectively. The total removal rates of the whole treatment system were 49.1%, 83.2%, and 87.4%, respectively, for these additions. The corncob served as a solid carbon source as well as a carrier of the denitrifying bacteria. The visible natural microbial membrane on the corncob facilitated partial removal of NO_3^- -N. There were micro-holes, mid-sized holes, and large holes in the bamboo charcoal, enabling penetration of denitrifying bacteria into the internal pores. Bamboo charcoal, an environmentally friendly and inexpensive biosorbent (Zhou et al., 2015b), is a strong adsorbent of ammonium. Other research has shown bamboo charcoal to have higher COD removal efficiency than commercial activated carbon (Hata et al., 2016). Therefore, the adsorption capacity and the denitrification ability of the BCF played a noticeable role in the adsorption and degradation of nitrogen.

[Fig. 2 was inserted here]

Effects of three different corncob concentrations on effluent COD levels are shown in Fig. 2b. Initially, the total concentration of effluent COD was high in all three cases. However, it later fell to below 40 mg/L in conformance with the GB18918-2002 1A discharge standard. Although effluent COD levels were lesser when 200 g corncob was used in comparison to the levels when 140 g corncob was used, excess corncob with more denitrifying bacteria reduced the carbon source in the

denitrification filter resulting in poor denitrification efficiency. Meanwhile, more corncobs mean more material cost. Therefore, 140 g was the best quantity of corncob considering the material cost and denitrification efficiency.

3.2 Performance of the denitrification filter

Hydraulic retention time (HRT) was one of the most important parameters in corncob-bamboo charcoal filter (C-BCF) reactor. Fig. 3a shows the different HRT that were studied using 140 g corncob as carbon source. Theoretically, the nitrate removal rate increased with the extension of HRT. The research showed that low HRT had no significant effect on NO_3^- -N removal rate, which was contrary to the effect at high HRT (Suhr et al., 2013). However, the experiment results showed that nitrate removal rate changed little between HRT of 2 h and 4 h. It indicated that the C-BCF reactor had strong nitrate nitrogen impact resistance and offers better application prospect. The COD concentrations were observed under three HRT conditions. As shown in Fig. 3b, the change in COD could be roughly divided into two stages: rapid decline phase and stable phase. In the initial stages, the COD of corncob effluent was high and was subsequently reduced by the denitrifying bacteria of the BCF reactor. As the soluble substances and cellulose in the corncob were used by the microorganisms, substances poorly soluble or hard to decompose such as lignin. There was no significant difference in COD removal rates between HRT of 1 h and 4 h. When running time of the setup was sufficient (ten days), the reactor could keep the effluent COD concentration stable at 10 mg/L–20 mg/L. HRT had little influence on ammonia levels.

[Fig. 3 was inserted here]

Therefore, the HRT should be 1 h–2 h when the influent COD concentration is 40 mg/L and NO_3^- -N concentration is 20 mg/L. The highest removal rate of NO_3^- -N was

above 80% under HRT of 2 h.

In order to simulate the influence of COD fluctuation of urban sewage treatment plant effluent, different influent COD concentrations of 20 mg/L, 40 mg/L, and 60 mg/L were designed. The HRT was set at 2 h and the initial concentration of NO_3^- -N was 20 mg/L. The NO_3^- -N variation is presented in Fig. 4a. The degree of denitrification increased with increase in COD and C/N (Hidaka et al., 2003). The total removal rate of NO_3^- -N increased from 71.8% to 83.2% as influent COD increased from 20 mg/L to 40 mg/L. However, it fell to 77.1% when the COD concentration was 60 mg/L. Simultaneously, the effluent NH_3 -N showed a similar trend (Fig. 4b). When COD was 40 mg/L, the highest corncob removal rate and total removal rate were 39.9% and 58.0%, respectively. Moreover, efficient denitrification was achieved when the influent COD was between 20 mg/L–60 mg/L and influent NO_3^- -N and the HRT were 20 mg/L and 2 h, respectively. The NO_3^- -N removal rate was 70%–85% and NO_3^- -N concentration was below 6 mg/L. The highest NO_3^- -N removal rate was 83.2% when the influent COD was 40 mg/L. The effluent COD (10 mg/L–40 mg/L) did not exceed the GB18918-2002 1A discharge standard (MOEP, 2002). Although the NH_3 -N removal rate was low at 8.9% when the influent COD concentration was 20 mg/L, average effluent NH_3 -N was 3.6 mg/L while the average effluent NO_3^- -N was 5.6 mg/L. The sum of two was still less than 15 mg/L, which qualified for the GB18918-2002 1A discharge standard (MOEP, 2002).

[Fig. 4 was inserted here]

The influence of different influent NO_3^- -N levels on the effluent levels is presented in Fig. 5a. When influent NO_3^- -N levels were 15 mg/L and 20 mg/L, there was little change in the corncob effluent levels (51.4% and 51.0%, respectively). Additionally, the denitrification rate was 6.4 mg/(L·h) and 8.5 mg/(L·h), respectively.

When influent NO_3^- -N was 30 mg/L, the denitrification and the NO_3^- -N removal rates were 9.7 mg/(L·h) and 38.7%, respectively. The results reveal the nitrogen removal efficiency reduced greatly when the NO_3^- -N concentration was greater than 20 mg/L. However, unlike the nitrogen removal efficiency, the denitrification rate changed significantly when NO_3^- -N was below 20 mg/L. The denitrification rate rose as the influent NO_3^- -N increased until its growth rate dropped at a certain point. This is probably because the denitrification rate follows the zero order reaction when the organic carbon source is adequate.

The pressure of decomposing bacteria increased as the influent NO_3^- -N concentration increased. The decomposition product of corncob was unsaturated under insufficient carbon conditions and resulted in the restrained growth of the bacteria. The NO_3^- -N concentration of the BCF effluent was similar to the corncob effluent. When influent NO_3^- -N concentration was 15 mg/L, 20 mg/L, and 30 mg/L, the total removal rates were 81.7%, 83.2%, and 60.2%, respectively and the total denitrification rates were 6.1 mg/(L·h), 8.3 mg/(L·h) and 9.1 mg/(L·h), respectively. This was consistent with an on-site denitrification factorial experiment that changed the soluble COD/ NO_3^- -N ratio from 4 to 12, and HRT from 50 min to 170 min (Suhr, 2013).

[Fig. 5 was inserted here]

The effect of different influent NO_3^- -N concentrations is shown in Fig. 5b. The effluent COD increased with influent NO_3^- -N concentrations. When the NO_3^- -N concentration was 30 mg/L, the effluent COD concentration was slightly high, but was still lower than 50 mg/L, in accordance with the GB18918-2002 1A discharge standard (MOEP, 2002). The soluble microbial products in the BCF may cause its effluent COD concentration to rise in the first two days.

The influence of NO_3^- -N on NH_3 -N concentrations was different (Fig. 5c). The removal rates of NH_3 -N were 38.3%, 58.0%, and 39.9% when the NO_3^- -N concentration was 15 mg/L, 20 mg/L, and 30 mg/L, respectively. Denitrifying bacteria recorded good performance under 20 mg/L of NO_3^- -N as suitable nutrition improved the microbial metabolism. Strong assimilation was conducive to the removal of NH_3 -N.

The dissolved oxygen (DO) of the influent was in the range of 1.31 mg/L to 3.77 mg/L, while its levels in the effluent was in the range of 2.87 mg/L to 3.52 mg/L. The DO of corn cob effluent was below 1 mg/L. DO levels in the BCF increased first and then decreased. This may be attributed to low C/N, less denitrifying bacteria, and predominance of aerobic bacteria in the back part of the BCF. The optimum DO for denitrifying bacterial growth should be below 0.5 mg/L. However, during diffusion through the biological membranes in water, oxygen was probably consumed by the outer aerobic microorganisms. Inner biological layers were still under anaerobic conditions, which protected the internal denitrifying bacteria. Consequently, trial denitrification could proceed smoothly.

Therefore, NO_3^- -N was controlled in the range of 15 mg/L–30 mg/L under 2 h HRT, when influent COD was 40 mg/L. Under this condition, the removal rate was in the range of 60.2%–83.2% and the denitrification rate was in the range of 6.1 mg/(L·h)–9.3 mg/(L·h). Although the average effluent NO_3^- -N was a bit high (12.01 mg/L) under 30 mg/L NO_3^- -N, it could be reduced to 6.2 mg/L by changing the HRT to 4 h. The effluent COD was below 50 mg/L in all cases and the effluent NH_3 -N was in the range of 1.6 mg/L–2.5 mg/L.

3.3 Nitrogen variation in BCF

Five samplings from top to bottom, at every 80 mm along the direction of the

water in the BCF were performed. Nitrogen compounds' variations were observed (Fig. 6a) under different operating conditions. The accumulation of NO_2^- -N was obvious. Other researchers also found this phenomenon in denitrification experiments (Her and Huang, 1995; Pujol and Tarallo, 2000; Rogalla and Bourbigot, 1990). NO_2^- -N was the intermediate product in the process of denitrification, as a result of insufficient carbon source (Her and Huang, 1995). Little accumulation of NO_2^- -N under operating conditions V and VIII occurred, as C/N was high. The NO_2^- -N concentrations were below 1 mg/L in all cases and the effluent had no toxicity. The removal rate of NH_3 -N was nearly the same. This showed that adsorption of NH_3 -N was at a saturated state. Adsorption is not an effective mechanism for the removal of NH_3 -N. Fig. 6 illustrates that NO_3^- -N concentration changed significantly along with the height of BCF.

The bamboo charcoal adsorbed NO_3^- -N and enhanced its delivery to the biological film. The degradation of denitrification bacteria maintained the concentration gradient of the bamboo charcoal substrate. Thus, the BCF executed intensified adsorption and degradation of NO_3^- -N. Critical factors were the quantity of denitrification bacteria and the C/N. Generally, C/N greater than three facilitated the growth of denitrifying bacteria, which was advantageous to the removal of NO_3^- -N. Organic matter in the corncob effluent is adsorbed by bamboo charcoal under poor nutritional conditions.

[Fig. 6 was inserted here]

The compounds contributing to total nitrogen (TN) included NO_3^- -N, NH_3 -N, and NO_2^- -N. The ideal height of the bamboo charcoal filling in the BCF can be decided based on the total nitrogen compounds' removal efficiency. Adjusting the height of BCF could allow increase in the volumetric load and improve the denitrification efficiency. Table 2 indicates that when the total nitrogen volume load was the highest

at 0.585 kg/(m³•d), the TN removal efficiency was 55.14% and the concentration of TN compounds was 10.94 mg/L. This efficient performance was consistent with experiments of denitrification using methanol in tertiary filtration, which were more expensive (Koch and Siegrist, 1997). All effluent concentrations of TN compounds were lower than the GB18918-2002 1A discharge standard limit of 15 mg/L (MOEP, 2002).

[Table 2 was inserted here]

3.4 Materials balance

Carbon source balance was calculated using the average denitrification rate and average COD concentration under all operating conditions. The carbon sources released by corncob were partly used by denitrification bacteria. Some part of the carbon sources was also consumed by DO.

The accumulation of NO₂⁻-N was ignored because of its low levels. Similarly, the consumption of carbon and nitrogen for cell synthesis was also ignored. The amount of carbon source for denitrification, C_N, was calculated as:

$$C_N = 2.86[\text{NO}_3^- - \text{N}] + \text{DO} \quad (\text{Soranno et al., 1997}). \quad \text{Equation (1)}$$

The corncob released COD consists of two parts: C_N and the difference in value of corncob COD (ΔC). The balance of corncob COD and NO₃⁻-N is displayed in Table 3.

[Table 3 was inserted here]

After the reactor was run for 57 d, the corncob was weighed by removing the film using ultrasonic equipment and then dried at 50 °C. The corncob weighed 48 g, which accounted for 34.3% of the original 140 g. Removal per gram of NO₃⁻-N needed 4.77 g corncob.

The balance of BCF COD and NO₃⁻-N was calculated using the same method. The

actual monitoring value of the water was less than the COD consumption in the BCF. The difference in the value was 25.66 g. It was probably because the corncob produced large amounts of organic matter in the preliminary stage, which was adsorbed by the bamboo charcoal. Desorption of the organic matter supplied a part of the carbon source (Jeong et al., 2006). This expounded the hypothesis that bamboo charcoal was not only an ideal filter material for denitrification, but also played a role in the storage and supply of the organic matter. Thus, bamboo charcoal can be used as a filler for adsorption packing to enable microbial degradation. The mechanism works through the concentration gradient as well as extracellular enzymes (Chen et al., 2011). Organic matter and nitrates are absorbed by bamboo charcoal and microorganisms are fixed in the filter. **Microbes are mainly concentrated on the surface of the bamboo charcoal particles and in the apertures where the organic matter and nitrate will be consumed.** Consequently, the founded concentration gradient improves the diffusion of pollutants into the bamboo charcoal. Additionally, biodegradation is conducive to the regeneration of bamboo charcoal. Extracellular enzymes generated by the secretion and disintegration of cells can directly enter the holes and interact with the inner organic matter and nitrates (Beaulieu et al., 2014; Gomez et al., 2002). Liberation from the original adsorption surface and the bacterial decomposition on the surface of bamboo charcoal creates a synergy of the adsorption and degradation processes.

4. Conclusions

The tail water of an artificial sewage treatment plant was effectively purified by a corncob carbon source and bamboo charcoal filter. The reactor's effluent nitrogen concentration was below the limit of 15 mg/L when influent COD concentration was 20 mg/L–60 mg/L, influent NO_3^- -N concentration was 15 mg/L–30 mg/L, and HRT

was 1 h–4 h. The reactor's maximum volume loading of total nitrogen was 0.585 kg/(m³·d). The COD and NO₃⁻-N balances of the reactor indicate that 4.77 g corncob is required to remove 1 g NO₃⁻-N and 2 g corncob is required to release 1 g COD. The new combined corncob carbon source and bamboo charcoal filter technique appears to be a feasible approach to treat secondary effluents effectively. The results prove that this process is compact, stable, convenient to install, and cost effective to build and operate as a tertiary treatment to remove nitrogen effectively.

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Figure Captions

Fig.1. Scheme of the pilot-scale combination of corncob carbon source and bamboo charcoal filter.

Fig.2. The concentration of (a) NO_3^- -N and (b) COD in corncob effluent and BCF effluent and their removal rate under different dosages of corncob.

Fig.3. The concentration of (a) NO_3^- -N, (b) COD and (c) NH_3 -N in corncob effluent and BCF effluent and their removal rate under different hydraulic retention times.

Fig.4. The influences of different COD influent loads on the concentration of (a) NO_3^- -N and (b) NH_3 -N in corncob effluent and BCF effluent and their removal rate.

Fig.5. The influences of different NO_3^- -N influent loads on the concentration of (a) NO_3^- -N, (b) COD and (c) NH_3 -N.

Fig.6. Effect of height of BCF on nitrogen compounds concentration and total nitrogen removal rate.

438 Table 1 Operation parameters of different stages

Operational condition	Starting and ending time (d)	Influent concentration (mg/L)				Flux (L/h)	Hydraulic retention time (h)
		COD	NO ₃ -N	NH ₃ -N	TP		
I	1-8	40	20	4	1	1	2
II	9-13	40	20	4	1	2	1
III	14-18	40	20	4	1	0.5	4
IV	19-23	20	20	4	1	1	2
V	24-28	60	20	4	1	1	2
VI	29-57	40	20	4	1	1	2
VII	61-65	40	30	4	1	1	2
VIII	66-70	40	15	4	1	1	2
IX	71-74	40	30	4	1	0.5	4

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441 Table 2 Effect of BCF height on nitrogen removal under conditions I - V & VII-VIII

operating condition	height of TN removal		effluent TN	TN volume load
	bamboo charcoal (mm)	efficiency (%)	(NO ₃ ⁻ -N+NH ₃ -N+NO ₂ ⁻ -N) (mg/L)	(NO ₃ ⁻ -N+NH ₃ -N) [kg/(m ³ .d)]
I	160	75.86	6.62	0.431
II	400	55.14	10.94	0.585
III	320	71.51	6.80	0.156
IV	240	59.08	9.95	0.347
V	240	71.86	6.62	0.335
VII	400	56.69	14.73	0.408
VIII	240	74.06	4.90	0.269

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444 Table 3 Balance sheet of corncob COD and NO₃⁻-N

Operated conditions	$\Delta C(g)$	Quality of corncob removal NO ₃ ⁻ -N (g)	COD released by corncob (g)	$\Delta C'$ (g)	Quality of removal NO ₃ ⁻ -N in BCF (g)	the theory of COD consumption in BCF (g)
I	3.46	2.10	9.84	6.47	2.64	6.97
II	-2.06	1.62	3.05	4.49	0.67	1.20
III	-1.07	0.35	0.05	0.84	4.92	13.89
IV	-0.50	1.14	3.01	-1.88	1.15	2.94
V	-3.51	1.49	1.00	0.41	0.77	1.85
VI	-4.03	3.36	6.92	13.44	8.60	22.58
Total	-7.71	10.06	23.87	23.77	18.76	49.44

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Fig.1

Fig.2

Fig.3

Fig.4

Fig.5

Fig.6