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Article

Effect of potassium chlorate on the treatment of domestic sewage by achieving shortcut nitrification in constructed rapid infiltration system

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Abstract: Constructed rapid infiltration system (CRI) is a new type of sewage biofilm treatment technology, but due to its anaerobic zone lacks of the carbon sources and the condition for nitrate retention, its nitrogen removal perfomance is very poor; However, shortcut nitrification-denitrification process presents distinctive advantages, as it saves oxygen, requires less organic matter and needs less time for denitrification compared to conventional nitrogen removal method. Thus, if the shortcut nitrification-denitrification process could be applied to CRI system properly, the simpler, more economic and efficient nitrogen removal method will be obtained. But, as its reaction process shows that the first and the most important step of achieving shortcut nitrification-denitrification is to achieve shortcut nitrification. Thus, in this study, we explored the feasibility to achieve shortcut nitrification, which produces nitrite as the dominant nitrogen species in effluent, by addition of potassium chlorate ($KClO_3$) to the influent. In an experimental CRI model system, the effects on nitrogen removal, nitrate inhibition and nitrite accumulation were studied, and the advantages of achieving shortcut nitrification-denitrification were also analysed. The results showed that shortcut nitrification was successfully achieved and maintained in a CRI system by adding 5 mM $KClO_3$ to the influent at a constant pH of 8.4. Under these conditions nitrite accumulation rate was increased, while a lower concentration of 3 mM $KClO_3$ had no obvious effect. The addition of 5 mM $KClO_3$ in influent presumably allowed sufficient activity of ammonia-oxidizing bacteria (AOB) but inhibited nitrite-oxidizing bacteria (NOB) strongly enough to result in a maximum nitrite accumulation rate of up to over 80%. As a result, nitrite became the dominant nitrogen product in the effluent. Moreover, if the shortcut denitrification will be achieved in the subsequent research, it could save 60.27 mg carbon source (CH_3OH) consumption when treatment of per liter sewage in CRI system compared with full denitrification process.

Keywords: shortcut nitrification; constructed rapid infiltration system; potassium chlorate inhibition; domestic sewage

1. Introduction

Sewage treatment technology for domestic sewage and polluted surface water treatment in small towns—Constructed rapid infiltration system (CRI) is a new sewage biofilm treatment technology put forwarded by professor Zhong zuoshen[1], due to it presents the both advantages of sewage rapid infiltration land treatment system and constructed wetland system, attracted increasing attention in recent years[2]. CRI system is mainly composed of feeding tank, grille, preliminary sedimentation tank, rapid infiltration tank and outlet system, adopts the dry-wet alternating operation mode and uses natural river sand, coal gangue, natural gravel, etc., to replace natural soil as filling medium to improve hydraulic load to 1.0-1.5m/d[3], the pictures of practical engineering of CRI system are shown in Fig.1.The removal mechanism of CRI system is to use the filling medium and microorganisms grown on the filing medium to adsorp, intercep and decompose the pollutants in sewage[4]. Especially, due to CRI system has the unique structure and feeding mode, its filling medium has the aerobic, facultative and anaerobic environment to grow abundant microorganism to make efficient sewage treatment[5]. As the previous practice showed, CRI system has a significant effect on the treatment of domestic sewage in small towns[6], whose removal rates of CODcr, NH_4^+-N, suspended solid (SS) and linear alkylbenzene sulfonates (LAS) could reach to above 85%, 90%, 95% and 95%, respectively and has the advantages of less energy-intensive, more environment-friendly and has a remarkable economic benefit compared with the conventional treatment systems[7]. Although, CRI system has a good removal effect of NH_4^+-N, due to its anaerobic zone lacks of the carbon sources for denitrification and the condition for nitrate retention[8], the concentration of nitrate in effluent is so high that total nitrogen (TN) removal rate only can reach to 10-30%[9].

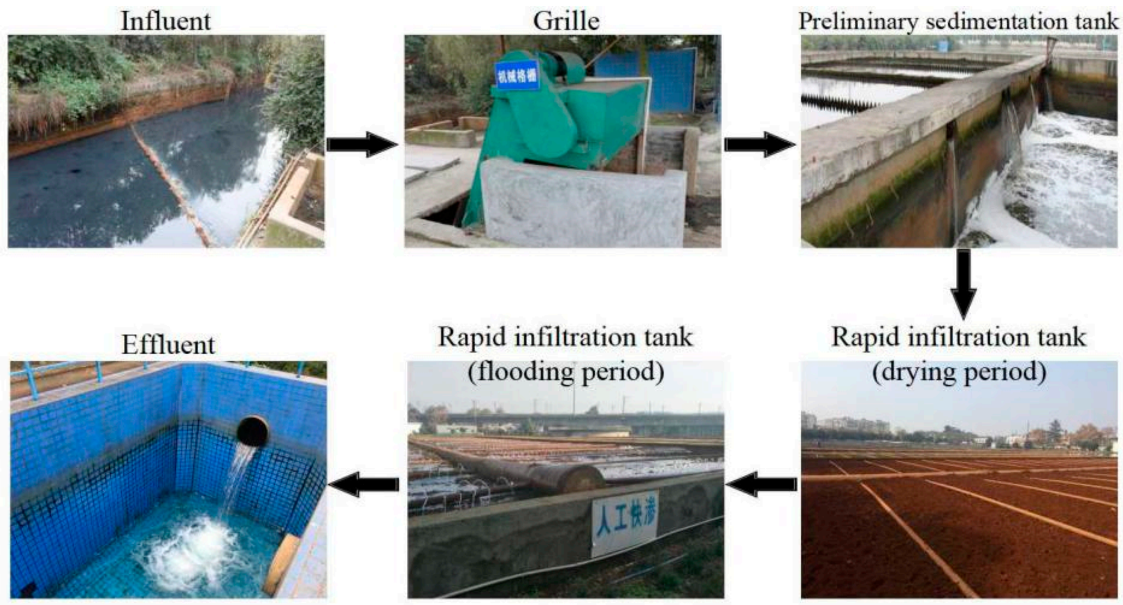

Figure1. Practical engineering of Phoniex River CRI system operated successfully for 12 years in Chengdu, China.

To enhance the nitrogen removal performance of CRI, the methods of adding extra carbon sources, optimizing the packing structure[10] and changing the water feeding patterns[1] were adopted, but those methods were all rely on the full nitrification- denitrification process which were difficult to overcome the problem of carbon source consumption and reduction of denitrifying bacteria activity during the long-term operation, and were also difficult to be popularized in the actual engineering ,due to their complex operating process.

Shortcut nitrification-denitrification is a novel biofilm nitrogen removal process which allows oxidation of ammonia to nitrite but no further oxidation to nitrate and reduces nitrite into nitrogen gas directly to achieve nitrogen removal in the system (Fig.2). Shortcut nitrification-denitrification presents distinctive advantages, as it saves oxygen and requires less organic matter compared to full nitrification-denitrification. But, as the Fig.2 shows, for shortcut nitrification-denitrification to be employed, the key point is to achieve shortcut nitrification, in other word, is to accumulate and maintain enough nitrite, which is produced by ammonium-oxidizing bacteria (AOB), and at the same time to inhibit or wash out nitrite-oxidizing bacteria (NOB), which would oxidize the produced nitrite to nitrate[11]. The conditions required to inhibit nitrite oxidization can be established with high concentrations of ammonium, a low concentration of dissolved oxygen, a high concentration of free nitrous acid and a permissive temperature. So far, shortcut nitrification has been achieved in various systems, such as aerated constructed wetland[12], a sequencing batch reactor (SBR)[13]and submerged biofilters[14], all of which resulted in high nitrite accumulation rates. The use of specific inhibitors can further improve shortcut nitrification. For example, Xu et al[13] studied the effect of hydroxylamine addition on shortcut nitrification in SBR, and Chen et al[15] used this same inhibitor in CRI; both found nitrite accumulation rates reaching more than 90%. Sukru and Erdal[14] and Cui et al[16] found that increasing salinity could further promote the accumulation of nitrite. Moreover, Ge et al[17] showed that low concentrations (4mg/L) of chlorine could improve the nitrite accumulation rate to reach 60-70%. Already in 1957, chlorate was described as a specific inhibitor of NOB: chlorate could inhibit the growth of autotrophic nitrite oxidizers at low concentration and completely inhibit nitrite oxidation at high concentrations[18]. Subsequent studies reported that addition of chlorate could result in nitrite to become the dominant product of NO_x in effluent, by allowing AOB activity while inhibiting NOB. For instance, Xu et al[11] showed that addition of chlorate to aerobic granules resulted in a 90% increase of nitrite accumulation in the effluent. Other studies showed that chlorate inhibited the oxidation of nitrite to nitrate, but it did not affect the oxidation of NH_4^+ to NO_2^- [19]; likewise, Xu et al[11] found that oxidation NH_4^+ to NO_2^- was not severely inhibited by chlorate. Such studies showed that shortcut nitrification can be achieved effectively by addition of specific inhibitors including chlorate, but the effect of adding potassium chlorate ($KClO_3$) in CRI has not yet been studied in detail.

Here, we tested whether potassium chlorate could improve performance of shortcut nitrification and removal efficiency of pollutants in a CRI system under experimental conditions and prospected the benefits of achieving shortcut denitrification.

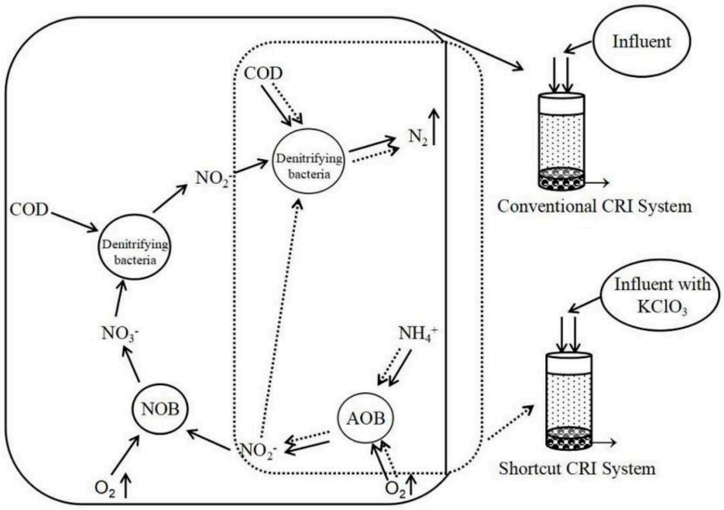

Figure 2. Comparison of full nitrification-denitrification process and shortcut nitrification-denitrification process (“→”reprents the process of full nitrification-denitrification; “⇢” reprents the process of shortcut nitrification-denitrification;)

2. Materials and methods

2.1 Experimental design

Four separate CRI columns were constructed using PVC (diameter 8 cm, height 30 cm) in the laboratory under controlled conditions. The temperature was kept constant at 34.2±1.1˚C. The filling medium of the columns consisted of two functional layers: a 5 cm deep supporting layer consisting of pebbles（5.0-10.0mm）and gravel (3.0-4.0mm) 2mm at the bottom, covered by a 20 cm deep treatment layer filled with 90% river sand (0.25 - 0.30 mm), 5% marble sand (1.0 - 2.0 mm) and 5% zeolite sand (1.5-1.7mm) on the top of the supporting layer (Fig.3). The influent sewage was circulated

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by a water pump so that it entered at the top of the column, moved through the packing medium vertically, and left by the outlet where water quality was measured.

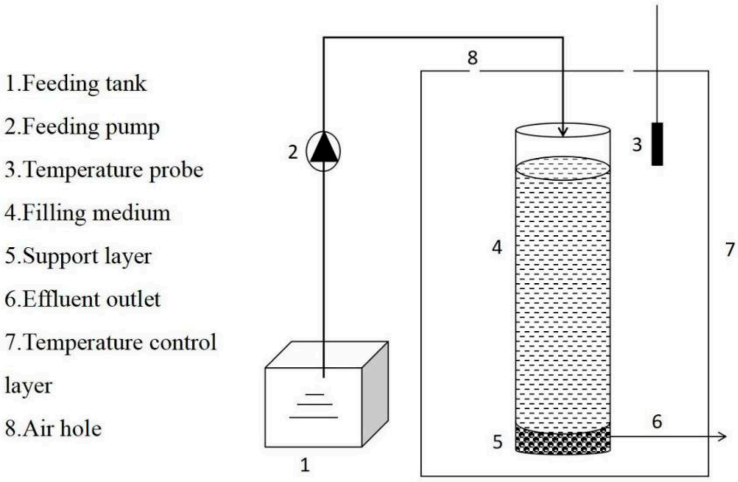

Figure 3. Experimental CRI system

2.2. Sewage and operational conditions

The influent sewage used in this study was a mixture of synthetic sewage and domestic sewage, with water quality parameters as shown in Table 1. The water was fed into the system by a dry-wet alternating operation mode as follows: water feeding was allowed twice daily with a hydraulic load of 0.6 m/d and a hydraulic retention time of 1.5 h. The system was run for 70 days of continuous operation, by which time the removal rates of ammonium nitrogen (NH_4^+-N) in the effluent reached 88%, indicating biofilms had formed successfully in the CRI and the scanning electron microscope pictures of filling medium of each columns are shown in Fig.4. In order to investigate the effect of potassium chlorate addition, the experimental columns were used as individual Models. Model 1 was the control treatment not receiving additions, and the pH of influent of Models 2-4 was adjusted to 8.4 by addition of NaOH solution. Moreover, in Model 3 $KClO_3$ was added to the influent at a final concentration of 5 mM, while in Model 4 a concentration of 3 mM $KClO_3$ was used.

Table 1. Water quality parameters of influent.

Water quality parameters	Concentration (mg/L)
Chemical Oxygen Demand (COD)	245.22±30.51
NH_4^+-N	53.93±7.31
NO_3^--N	1.15±0.92
NO_2^--N	0.14±0.12
Total Nitrogen (TN)	55.35±7.14
pH	7.3±0.2 (control), 8.4 (Models 2-4)
Temperature (°C)	34.2±1.1

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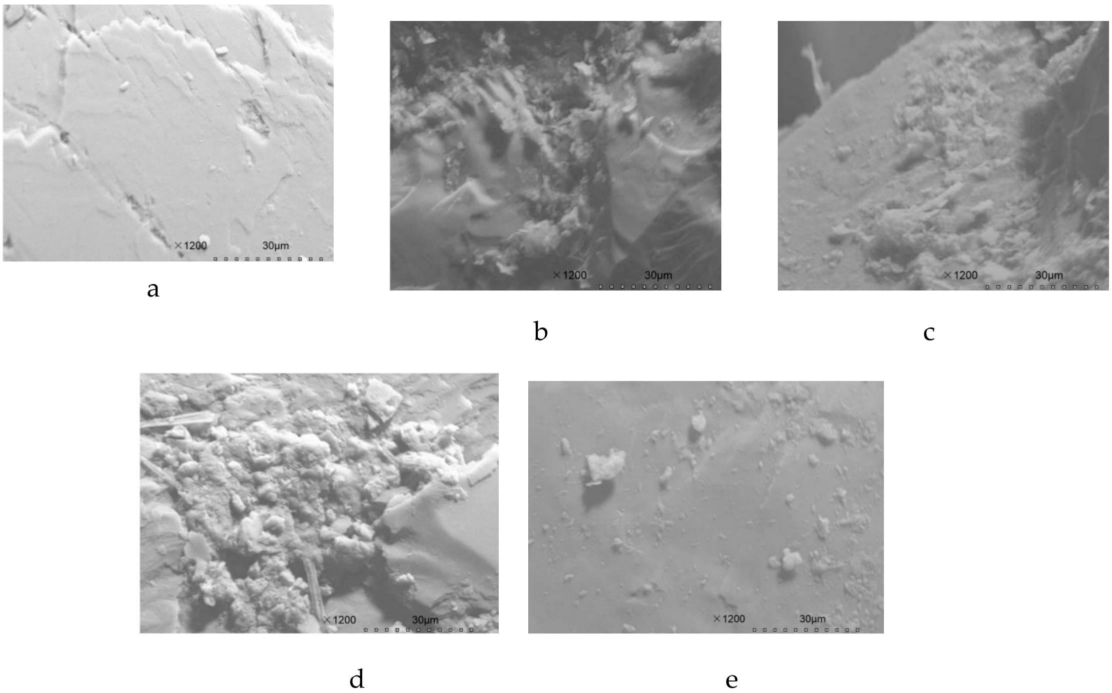

Figure 4. SEM images of filling medium. (a) blank filling medium;(b-e) filling medium (formed with biofilm) of Model 1-4

2.3. Analytical methods

Water samples from influent and effluent were collected every 2 days, filling medium samples were collected after biofilm formed successfully. Concentration of COD in the water was determined using the potassium dichromate method; concentration of nitrogen in the form of ammonium was determined by Nessler's Reagent Colorimetric Method, nitrate (NO_3^--N) by UV-spectrometry, nitrite (NO_2^--N) by molecular absorption spectrophotometry, and total nitrogen (TN) by UV-spectrometry, using standard procedures[20]. The biofilm of the filling medium was prepared by the glutaraldehyde fixation method[21] and observed by using scanning electron microscope (SEM). The nitrite accumulation rate was calculated as the ratio of $NO_2^-/(NO_2^-+NO_3^-)$[12].

3.Results and Discussion

3.1. Effect of potassium chlorate on removal efficiency of ammonium nitrogen

The removal efficiency of nitrogen in the form of ammonium in CRI system was compared between the controls (with and without pH adjustment) and after the addition of two concentrations of $KClO_3$ to the influent. Removal efficiency was calculated as the difference in concentration between influent and effluent (influent concentration minus effluent concentration) divided by the concentration in influent.

Adjustment of the influent pH to 8.4 only had a minor effect on ammonium nitrogen removal during the first 10 days (Fig. 5). There was no difference in removal efficiency between Model 4 (with addition of 3mM $KClO_3$) and Model 2 (without $KClO_3$ addition), as both reached approximately 87% removal on average (Fig. 5). However, in presence of 5mM $KClO_3$ (Model 3), the NH_4^+-N removal efficiency was reduced, though it still reached 66% on average. This is most likely the chlorate has a slight inhibition of AOB activity, as a result of which NH_4^+-N oxidation efficiency was less efficient.

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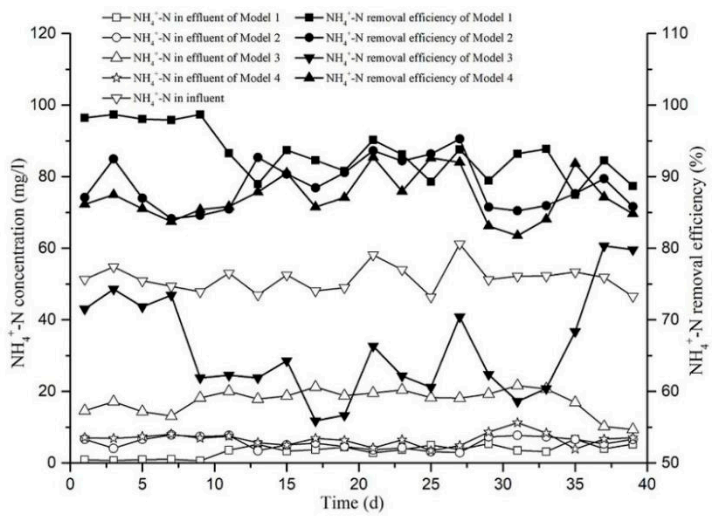


Figure 5. The ammonium-nitrogen removal efficiency (in %) and the absolute concentration of NH4+-N in the four experimental models of CRI

3.2. Effect of potassium chlorate on nitrate accumulation in CRI system

Accumulation of nitrogen in the form of nitrate was next assessed. As can be seen in Fig. 6, there was no significant difference between Model 2, resulting in a nitrate concentration of on average 36.24 mg/L, and Model 4, resulting in 34.51 mg/L. Very similar results were obtained for the control in which the pH of the influent had not been adjusted (Model 1). In contrast, Model 3 resulted in much lower nitrate concentrations of approximately 7.39 mg/L on average, which represented an 80% reduction compared to the control. As shown, the nitrate concentration in effluent of Model 3 was reduced within 48 hr after addition of 5 mM $KClO_3$ and reached a minimum of 2.92 mg/L on day 13. This result shows that addition of 5 mM $KClO_3$ in influent was able to strongly prevent the oxidation of nitrite, a condition that favors the accumulation of nitrite and is desired for shortcut nitrification achievement.

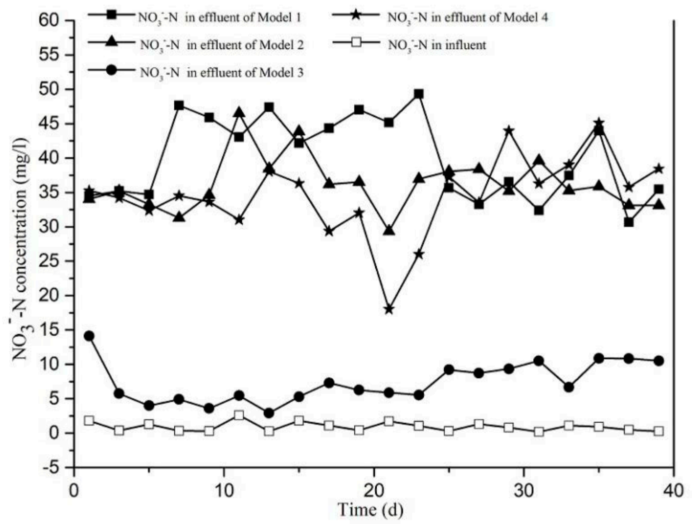


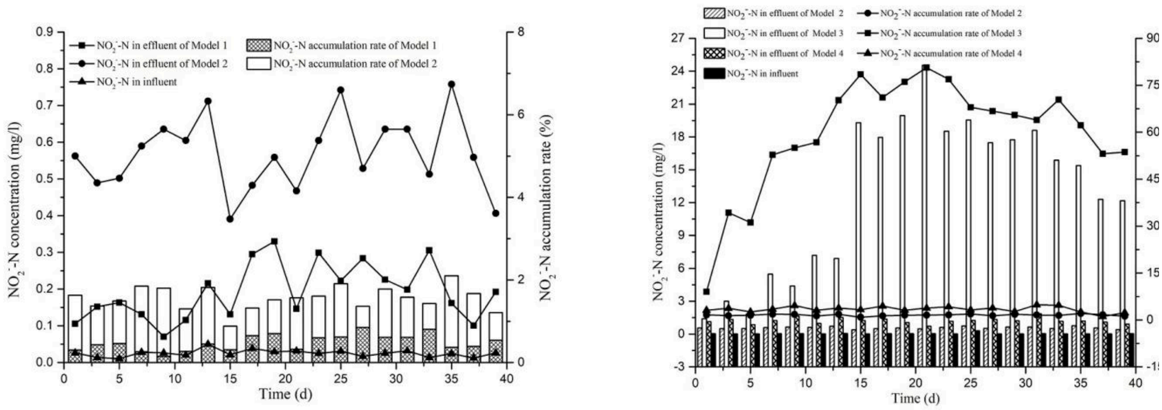
Figure 6. The nitrate-nitrogen concentration in influent and effluent in the four experimental models of CRI

3.3 Effect of potassium chlorate and pH on nitrite accumulation in CRI system

Previous studies have described that the pH of the influent is a decisive factor for inhibiting NOB activity. For instance, Banashri[19] described that nitrite accumulation can be improved at high pH. We also observed (Fig. 7a) that adjustment of the influent water pH to 8.4 slightly improved the average nitrite accumulation rate, from 0.50% in Model 1 (unadjusted pH) to 1.5% in effluent of Model 2 (pH 8.4). Nevertheless, this increase was too weak to support shortcut nitrification. Thus, a controlled pH of 8.4 is by itself insufficient to enable effective shortcut nitrification in CRI.

The average nitrite accumulation rates in our models are shown as curves in Fig. 7b. As can be seen, these rates were very low in Model 2 (1.56% on average) and Model 4 (3.43%), but much increased in Model 3, resulting in 59.80% accumulation rates on average. Thus, the addition of 5mM $KClO_3$ strongly supported accumulation of nitrite in the model CRI system. Combined with the data presented in Figures 6 and 7, it can concluded that, whereas nitrate was the dominant product in effluent of Models 1, 2 and 4, nitrite was the dominant nitrogen product of Model 3, as a result of effective nitrite oxidation inhibition.

As apparent in Fig. 7b, the nitrite accumulation rate in effluent of Model 3 increased sharply during the first 7 days (from initially 9.02% to 52.76%) and further increased to reach a plateau of up to 80% during days 15-23. The nitrite concentration peaked at day 21 at 24.54mg/L. After this, the nitrite accumulation rate slightly decreased, still maintaining 53% at day 39. This indicates that shortcut nitrification can be not only achieved but also maintained in the tested CRI system by addition of 5mM $KClO_3$ in the influent at a pH of 8.4.



a

b

Figure 7. Nitrite accumulation in effluent. Panel a: Effect of pH on nitrite accumulation in the effluent of Model 1 (unadjusted pH) and Model 2 (pH 8.4). Panel b: The nitrite accumulation rate (curves) and concentration (bars) in effluent.

3.4 Prospects for the achievement of shortcut nitrification-denitrification in CRI system

In 1975, the concept of shortcut nitrification-denitrification biological nitrogen removal process was put forwarded for the first time by Votes after he finding the phenomenon of NO_2^--N accumulation during nitrification process[22]. Then, this new nitrogen removal process has attracted increasing attention from experts and researchers. The equations of nitrification(Eq1) and shortcut nitrification (Eq2) are shown as follows:

$$NH_4^+ + 2O_2 \rightarrow NO_3^- + H_2O + 2H^+, \quad (1)$$

$$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + H_2O + 2H^+, \quad (2)$$

As we can see from Fig.6 and Fig.7, the average nitrate concentration of Model 2 and Model 3 are about 36.24mg/l and 7.39mg/l, respectively, the average nitrite concentration of Model 2 and Model 3 are about 0.57mg/l and 12.98mg/l, respectively. Therefore, according to the Eq1 and Eq2, Model 3 reduced about 21.77 mg (treatment of per liter sewage) oxygen consumption during the process of nitrification compared with Model 2. Most of the denitrifying bacteria are facultative and use organic matters as carbon sources under the anoxic condition to provide energy. Gómez et al[23] found that CH_3OH is an ideal carbon source for denitrification. Thus, if CH_3OH is chosen as carbon

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source, the equations of full denitrification (Eq3) and shortcut denitrification (Eq4) are shown as follows:

$$NO_3^- + 1.08CH_3OH + 0.24H_2CO_3 \rightarrow 0.056C_3H_7O_2N + 0.47N_2 + 1.68H_2O + HCO_3^-, \quad (3)$$

$$NO_2^- + 0.67CH_3OH + 0.53H_2CO_3 \rightarrow 0.004C_3H_7O_2N + 0.48N_2 + 1.23H_2O + HCO_3^-, \quad (4)$$

According to Eq3 and Eq4 and the data from Fig.6 and Fig.7, if the subsequent shortcut denitrification will be achieved, the dosage of CH_3OH used for Model 2 denitrification will consume 98.38 mg (treatment of per liter sewage) during operating period, but, the Model 3 only needs 38.11mg (treatment of per liter sewage) CH_3OH for denitrification and shortcut denitrification, which was only 38.73% of the consumption of Model 2. Thus, achievement of shortcut nitrification-denitrification in CRI system will not only improve denitrification rate, simplify reaction process, but also save oxygen and carbon source consumption, as well as operating costs significantly.

Conclusions

(1) The addition of 3 mM $KClO_3$ to influent at a constant pH of 8.4 is not sufficient to support the activity of AOB and inhibit that of NOB so that shortcut nitrification does not take place in CRI.

(2) Adjusting the pH of influent to 8.4 alone did not contribute much to establish shortcut nitrification in CRI.

(3) The addition of 5 mM $KClO_3$ in influent (pH=8.4) supported efficient shortcut nitrification by supporting the activity of AOB and inhibiting that of NOB, whereby the inhibition of NOB was more significant and much stronger. This resulted in nitrite to become the dominant product of NO_X in effluent, and stable, long-term shortcut nitrification was successfully achieved in the CRI system.

(4) Compared with full nitrification, shortcut nitrification saved more than 21.77 mg (treatment of per liter sewage) oxygen consumption during reaction process. In addition, according to the data of nitrate and nitrite in Fig.6 and Fig.7, the consumption of carbon source (CH_3OH) for subsequent denitrification was calculated and analyzed by using Eq3 and Eq4, the results showed that the consumption of carbon source (CH_3OH) of Model 3 was only 38.73% of the consumption of Model 2. Therefore, compared with conventional sewage treatment methods, achievement of shortcut nitrification and denitrification process in CRI system will both taking the advantages of CRI system which has the unique structure and feeding mode to construct aerobic, facultative and anaerobic environment for microorganism enriching in the filling medium , and the shortcut nitrification-denitrification process which could improve denitrification rate, save oxygen and carbon source consumption, as well as operating costs.

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Conflicts of Interest: The authors declare no conflict of interest.

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