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A promising approach of reject water treatment using a tidal flow constructed wetland system employing alum sludge as main substrate

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Abstract

This study examined a novel reuse of the alum sludge, an inescapable by-product of the production of drinking water when aluminium salt is added as a coagulant, as the main medium in a laboratory-scale multi-stage constructed wetland (CW) system for reject water treatment. Such reject water is a main concern in municipal wastewater treatment plant (MWWTP) for increasing the organic and nutrient loading. A "tidal flow" strategy was employed to enhance the wetland aeration to stimulate organic pollutants and N oxidation while the "step feed" operation was adopted to supply the necessary amount of carbon source for denitrification. The results reveal that alum sludge acted as P adsorbent can secure the P removal. Meanwhile, high removals of ammoniacal-nitrogen and organic matters can also be obtained due to the active bacteria attached growth on the alum sludge surface. The results show that average removal efficiencies of $65.4 \pm 12.3\%$ for COD, $67.8 \pm 9.2\%$ for BOD₅, $33.6 \pm 17.0\%$ for N and $99.5 \pm 0.49\%$ for P can be achieved over a period of 190 days. This indicates that novel reuse of alum sludge as medium in CW system can provide a promising approach for reject water treatment. Therefore, it will significantly reduce the amount of pollutants feedback through reject water recycling in a MWWTP.

Keywords

Alum sludge; constructed wetland; nutrient control; reject water; reuse; wastewater treatment plant

INTRODUCTION

In municipal wastewater treatment plant (MWWTP) that employs biological nutrients removal (BNR) process for phosphorus (P) and nitrogen (N) removal, approximately 80-90% of P is removed in which the P in wastewater is transferred into the cell of the phosphorus accumulation organisms (PAOs) and approximately 60-75% of N is removed while 25-40% of N is transferred into biomass of activated sludge. Meanwhile, approximately 50% of organic matters (OM) is transferred into carbon dioxide and/or water and 50% of it is converted into biomass (Tchobanoglous et al., 2003). The removal of the excess activated sludge from the BNR process can therefore lead to the ultimate removal of P, N and OM from municipal wastewater. However, during the ensuing sludge treatment process including thickening and anaerobic digestion, P, N and OM are released from bio-cell into the aqueous phase in the form of supernatant, which is termed as reject water. Such waste stream of reject water is normally recycled back to the main wastewater process for treatment due to the low quality. This will accordingly contribute from 10-80% of P load, 20- 30% of N load and 3-5% of COD load to the BNR process (Pitman, 1999; Loosdrecht and Salem, 2006; Dosta et al., 2007). As a result, P and N removal efficiency can be considerably affected. Although BNR process has been widely used for decades in municipal wastewater treatment, many facilities still struggle to meet water quality goals. Among the factors affecting P and N removal performance, the amount of P and N feedback through reject water recycling is considered as one of the most critical determinant factor to ensure the nutrient removal efficiency of BNR process. Therefore, it is necessary to remove of P and N from reject water and this can significantly reduce the P and N load to the main stream of the MWWTP.

Recently, several biological treatment processes have been reported for N removal from reject water. These include SHARON process, ANAMMOX process, BABE process (Loosdrecht and Salem, 2006), sequencing batch reactor (SBR) and membrane bioreactor (MBR) (Dosta et al., 2007) and biofilter (Hwang et al., 2000). Meanwhile, chemical aids and/or physical–chemical processes have also been used for this purpose. However, the extra operation costs, extra management of the treatment process, potential negative effect on the pH of the reject water and increase in sludge production may not favour their use. Therefore, it is still necessary to explore suitable and cost effective approach and low-cost materials for reject water treatment to reduce the N, P and OM loading in MWWTP. Compared with N feedback through reject recycling, P feedback may have significant effects on nutrient removal due to the fact that P cannot be converted into gaseous compounds or other non-polluting forms. To remove P from the recycled reject water, industrial by-products, particularly those that contain high levels of alum and/or iron, have been considered as a cost-effective alternative to chemical aids. Recent studies have showed that alum sludge, an inescapable by-product of the production of drinking water when aluminium salt is added as a coagulant, possesses a latent P adsorption capacity and excellent P removal performance (Dayton and Basta, 2005; Makris et al., 2005; DeWolfe, 2006; Babatunde et al., 2007; Razali et al., 2007). In a previous study, the uses of air-dried alum sludge (solid form) and thickened alum sludge (liquid form) to reduce P-feedback through reject water recycling in a MWWTP were suggested. One strategy was the use of the thickened alum sludge for co-conditioning and dewatering with the anaerobically digested sludge in MWWTP. The other strategy involves the use of dewatered alum sludge cakes in a fixed bed for P immobilization in reject water (Yang et al., 2009). However, from a holistic point of view, the process for reducing nutrient feedback and accumulation in MWWTP should duly consider N, P and OM removal, rather than P itself from the recycled reject water.

To this end, constructed wetland (CW) is therefore considered to treat the reject water since it has been increasingly applied worldwide as a low-cost technology for various wastewater treatment including nutrient removal (Prochaska and Zouboulis, 2006; Vymazal, 2007; Sim et al., 2008). More significantly, a novel CW system employing dewatered alum sludge as main substrate to serve as P adsorbent and biofilm carrier has been recently developed (Zhao et al., 2010). This provided a good showcase of using “waste” for wastewater treatment and thus contributed to sustainable development. The aims of this study were: (1) to investigate the applicability of the alum sludge-based CW system for high N, P-containing reject water treatment; (2) to assess N, P and OM removal and system performance as well as the design parameters for the possible large application of reject water treatment in MWWTP.

MATERIAL AND METHODS

Alum sludge collection and preparation The experimental alum sludge, with moisture content of 83.0%, was collected from the mechanical dewatering unit of a water treatment plant located in South Xi’an city, P.R. China, where poly-aluminium chloride (PAC) was used as principal coagulant for treating a reservoir water. Thereafter, the alum sludge was air-dried at room temperature for 14 days. The air-dried alum sludge with moisture content of 26.3% was then ground and sieved into different particle sizes ranging from 2 to 50mm as the main substrate to be used in the laboratory scale CW system. Characterization of the alum sludge was conducted and the results are shown later in relevant section.

Reject water collection and pretreatment The reject water studied was collected periodically from the holding tank of NO.4 MWWTP in Xi’an city, P.R. China. The MWWTP employs an anaerobic/anoxic/aerobic (A²/O) process for municipal wastewater treatment. The reject water is the mixture of the supernatant from the thickening tank (the primary sludge and the excess activated

sludge thickening unit) and the supernatant from the centrifuge (sludge dewatering unit). The SS in the raw reject water is in the range of 120.1 to 1,350.5 mg/l with average of 514.6 mg/l. It was quite obvious that this level of SS will lead to clogging of the CW very rapidly. Therefore, the SS must be removed from the reject water to ensure a successful operation of the CW system. Accordingly, a pre-treatment process using sand bed filtration (sand diameter ranged 0.8-1.3 mm and the d_{50} of 1.0 mm) is adopted with flow rate of 5 m³/m² h. The pre-treated reject water was stored at room temperature and then used as the influent for CW. Table 1 summarizes the characteristics of pretreated reject water used in this study.

Table 1 Characteristics of the reject waster after sand filter pretreatment

Parameters	Average \pm SD, n=26	Minimum	Maximum
PO ₄ ³⁻ -P (mg/l)	57.3 \pm 9.4	43.7	75.5
Ammoniacal-nitrogen, NH ₄ ⁺ -N (mg/l)	72.5 \pm 16.9	45.8	101.3
COD (mg/l)	296.7 \pm 125.5	144.5	563.5
BOD ₅ (mg/l)	148.1 \pm 52.5	75.3	235.6
pH	7.2 \pm 0.4	6.7	8.2
Temperature ^b (°C)	18.0 \pm 2.7	13.2	23.8

Laboratory scale CW set-up and operation A lab-scale four-stage vertical subsurface flow constructed wetland (VSFCW) system was constructed using Pyrex tubes (100cm high with an internal diameter of 9.4cm) packed firstly with gravel to 10 cm at the bottom to act as supporting layer followed by 55 cm in depth of prepared alum sludge with particle size ranged from 50 to 32mm, 32 to 16mm, 16 to 8mm and 8 to 4mm for 1st, 2nd, 3rd and 4th stage, respectively. *Typha angustifolia* was planed to the top of each stage. Figure 1 shows schematic description of the system. The characteristics and configuration of the alum sludge based multi-stage VSFCW is summarized in Table 2.

A “tidal flow” plus “step-feed” strategy was adopted for the operation of the CW system. Tidal flow strategy allows the enhancement of aeration efficiency and “step-feed” operation provides the necessary amount of carbon source for denitrification. Tidal flow was generated by peristaltic pumps that were controlled by pre-set timers. In each stage, the ‘tide’ took place once every 6 hrs and given 4 cycles per day. Step-feed model was set as 80% of inflow was pumped into the 1st stage while 20% was pumped into the 3rd stage, as shown in Figure 1. Each cycle allows 1.76 L of reject water to pass through, giving a hydraulic loading rate (HLR) of 1.0 m³/m² d for 1st and 2nd stages 2.2 L of reject water was introduced to 3rd and 4th stages, thus giving a HLR of 1.26 m³/m² d.

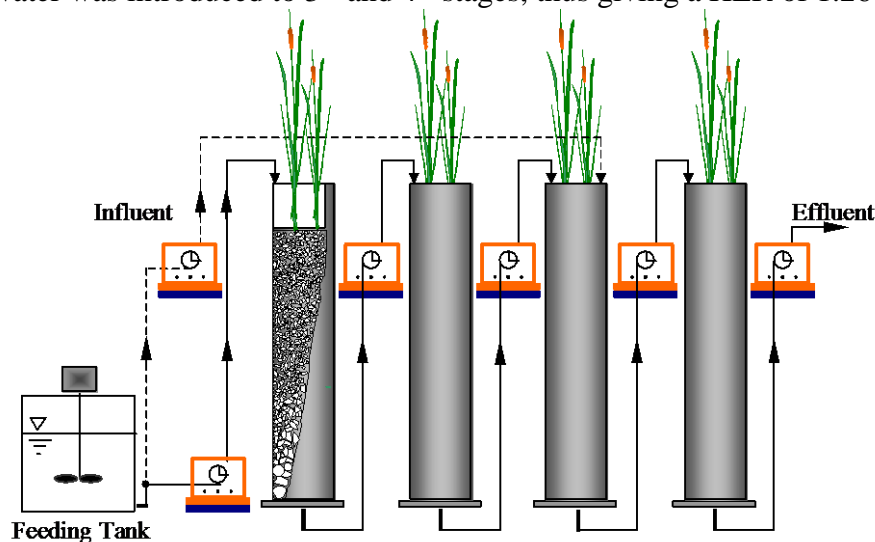


Figure 1 Schematic description of the alum sludge based multi-stage VSFCW system

Table 2 Characteristics and configuration of the multi-stage VSFCW system

Parameters	Constructed wetland system			
	1 st Stage	2 nd Stage	3 rd Stage	4 th Stage
Bed depth (cm)	65	65	65	65
Alum sludge layer (cm)	55	55	55	55
Supporting layer (Gravel) (cm)	10	10	10	10
Amount of alum sludge (kg)	3.0	3.1	3.2	3.2
Particle size of alum sludge (mm)	50-32	32-16	16-8	8-4
Contact time (hours)	5	5	5	5
Drain time (aeration time) (hour)	1	1	1	1
HLR of individual stage (m ³ /m ² d)	1.0	1.0	1.26	1.26

Analysis The air-dried alum sludge, which was further oven dried at 103 ± 2 °C, was used for examining its physical and chemical properties during which the oven-dried samples were digested if necessary using the standard method of nitric acid digestion (3030 E, APHA, 1992). The chemical composition of the alum sludge was determined using Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES) (IRIS Advantage, Thermo Elemental, Franklin, Massachusetts, USA), ion chromatography (DX-120, Dionex, Sunnyvale, California, USA) and Total Organic Carbon (TOC)-V_{CSH} (TOC-V_{CSH}, Shimadzu, Tokyo, Japan).

During the CW system operation period, samples of influent and effluent from each stage were collected periodically and analysed for BOD₅ and COD. The samples were also filtered using 0.45 μ m filter membrane for N and P measurement. The filtered samples were analyzed for P concentration using ICP-MS (X series) (for P concentration less than 0.05 mg-P/l) and the stannous chloride method (for P concentration higher than 0.05 mg-P/l) (4500-P D, APHA, 1992). COD, BOD₅ and Ammoniacal-nitrogen were analysed using standard methods of 5210 B, 5220 B and 4500-NH₃ C, respectively (APHA, 1992).

RESULTS

Physical and chemical properties of alum sludge The principal physical and chemical properties of the alum sludge used in this study are shown in Table 3. Specific elemental metal component of aluminium, calcium and iron content (expressed as oxidized metal) in the alum sludge was 231.7, 12.9 and 5.1 mg/g-sludge, respectively. The other principal chemical components were silicon oxide (expressed as SiO₂), humic substances (expressed as TOC) and orthophosphate (expressed as phosphorus), with mass content of 23.6, 21.5 and 0.71 mg/g-sludge, respectively. Clearly, aluminium is the dominant component in the alum sludge.

Table 3 The physical and chemical properties of the alum sludge

Properties	Unit	Average \pm SD, n=3
Water content of raw alum sludge	%	83.0 \pm 2.1
Water content of air-dried alum sludge	%	26.3 \pm 1.3
Aluminium (as Al ₂ O ₃)	mg/g-sludge	231.7 \pm 4.2
Calcium (as CaO)	mg/g-sludge	12.9 \pm 1.7
Iron (as Fe ₂ O ₃)	mg/g-sludge	5.1 \pm 1.0
Silicon Oxide (as SiO ₂)	mg/g-sludge	23.6 \pm 3.9
Humic substances (as TOC)	mg/g-sludge	21.5 \pm 6.9
PO ₄ ³⁻ (as phosphorous)	mg/g-sludge	0.7 \pm 0.3

Characteristics of P removal Figure 2 presents the P removal performance of the CW system. The phosphate concentration in the reject water ranged from 43.7 to 75.5 mg-P/l. The phosphate in the effluent at the operation period (up to 190 days) was in the range of 0.002 to 0.8 mg-P/l. The very low P concentration achieved indicates the excellent P removal ability of the alum sludge. As shown in Figure 2, the overall P removal efficiency was in the range from 99.9 to 98.7% while the average P removal was 99.5%. This indicated the potential benefit of P reduction in reject water by using the alum sludge-based CW system, which could eliminate the P loading in wastewater treatment process when the reject water is recycled.

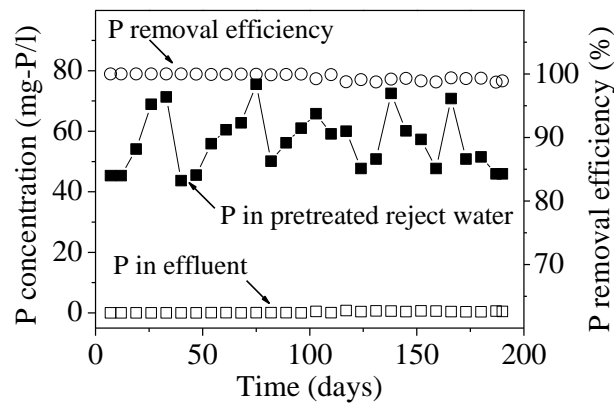


Figure 2 Characteristics of P removal during the operation period

Figure 3 shows the contribution of individual stage to overall removal of P during the operation period. The amount of P removal in mass in 1st, 2nd, 3rd and the 4th stage was 11.0, 3.8, 3.3 and 0.5 mg-P/g-sludge, respectively, while the overall P removal contributed by each stage was ranged from 19.1 to 80.0%, 0.003 to 54.6%, 11.0 to 21.8% and 0 to 9.9% respectively. This demonstrated that the 1st stage plays a key role in P immobilization. However, it should be noted that the P removal contributed by particularly individual stage was highly dependent on the amount of P in the effluent of the previous stage.

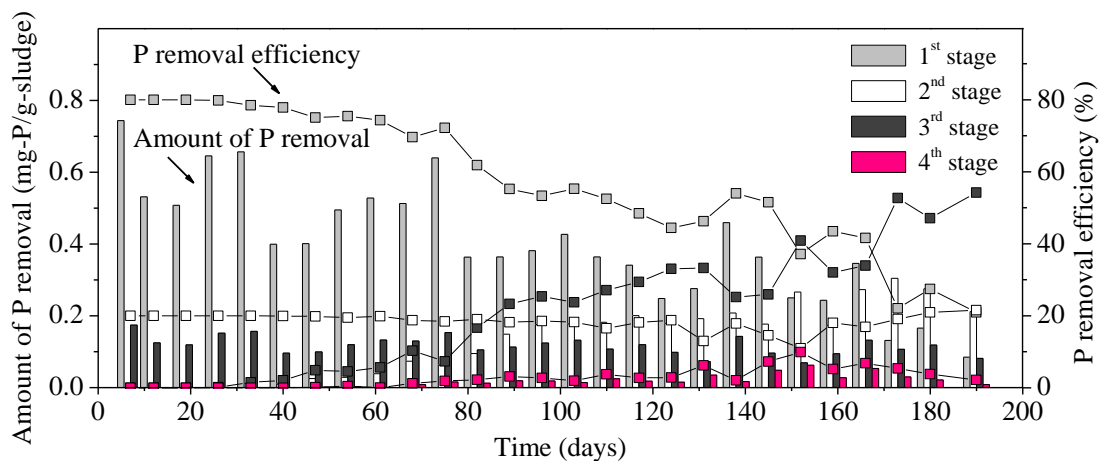


Figure 3 Contribution of individual stage to overall removal of P during the operation period

Characteristics of ammoniacal-nitrogen removal Figure 4 shows the ammoniacal-nitrogen removal performance of the CW system. The ammoniacal-nitrogen concentration in the reject water ranged from 45.9 to 95.9 mg-N/l while the ammoniacal-nitrogen in the effluent was in the range of 14.0 to 82.2 mg-N/l. This gives $33.6 \pm 17.0\%$ of ammoniacal-nitrogen removal from reject water.

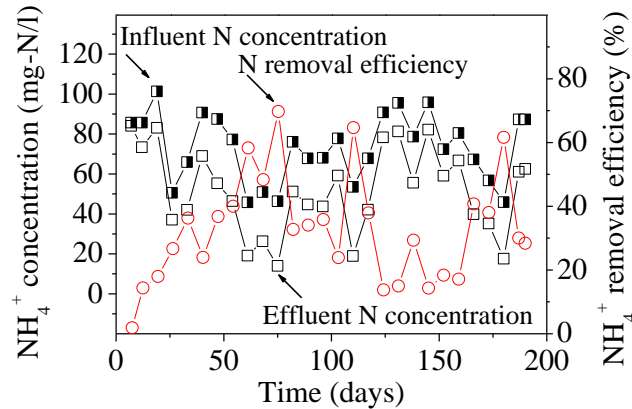


Figure 4 Characteristics of $\text{NH}_4^+\text{-N}$ removal during the operation period

Figure 5 shows the contribution of individual stage to overall removal of ammoniacal-nitrogen during the operation period. The average amount of ammoniacal-nitrogen removal (expressed as removal loading) in 1st, 2nd, 3rd and the 4th stage was in turn 3.1, 3.6, 5.8 and 3.1 $\text{g-N/m}^2 \text{ d}$ while the overall ammoniacal-nitrogen removal contributed by each stage was ranged from 1.2 to 15.1%, 2.11 to 18.0%, 2.7 to 28.1% and 2.9 to 16.1% respectively.

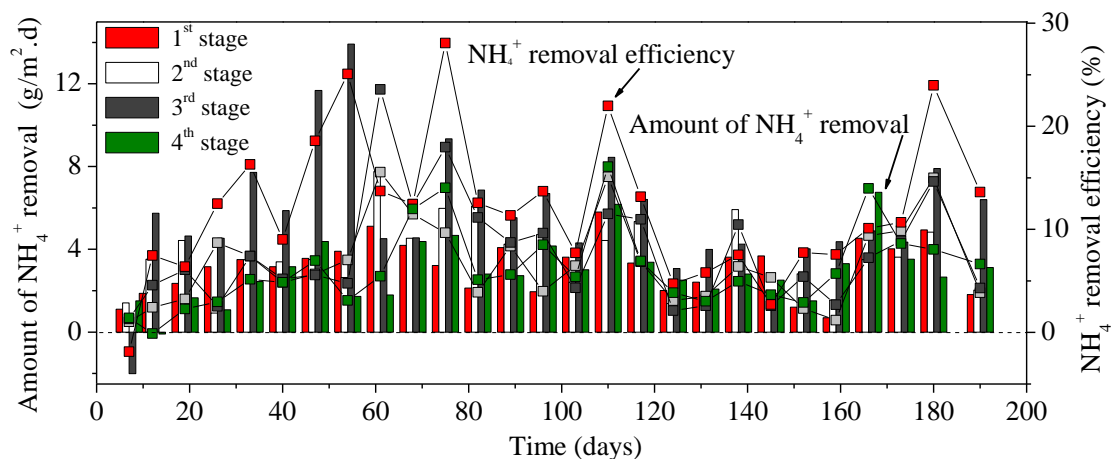


Figure 5 Contribution of individual stage to overall removal of $\text{NH}_4^+\text{-N}$ during the operation period

Characteristics of organic matter removal Figure 6 presents the organic matter ((expressed as COD and BOD_5) removal performance of the CW system. The COD concentration in the reject water ranged from 144.5 to 563.5 mg/l . The COD in the effluent of the system ranged from 57.3 to 323.5 mg/l . As a result, the average COD removal efficiency was 65.4% (see Figure 6 (a)). The BOD_5 concentration in the reject water ranged from 75.3 to 235.6 mg/l while the BOD_5 in the effluent of the alum sludge based multi-stage VSFCW system ranged from 22.0 to 101.9 mg/l . The average BOD_5 removal efficiency of 67.8% was thus achieved (see Figure 6 (b)).

Figure 7 shows the contribution of individual stage to overall removal of COD and BOD_5 during the operation period. The average amount of COD removal (expressed as removal loading) in 1st, 2nd, 3rd and the 4th stage was in turn 36.6, 20.1, 36.3 and 16.7 $\text{g/m}^2 \text{ d}$, respectively. The overall COD removal contributed by each stage was ranged from -5.0 to 33.8%, 0.6 to 17.2%, -0.7 to 30.4% and -0.4 to 17.3% respectively (see Figure 7 (a)). The average amount of BOD_5 removal (expressed as removal loading) in 1st, 2nd, 3rd and the 4th stage was 21.0, 10.2, 12.9 and 10.5 $\text{g/m}^2 \text{ d}$, respectively. The overall BOD_5 removal contributed by each stage was ranged from 0.4 to 44.0%, -2.0 to 26.8%, -5.3 to 29.9% and -4.6 to 33.5% respectively (see Figure 7 (b)).

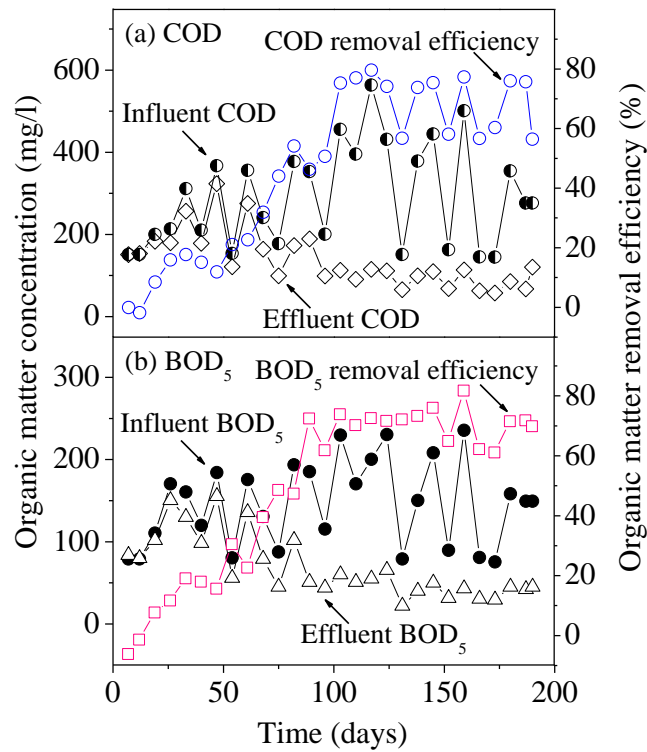


Figure 6 Characteristics of organic matter removal during the operation period

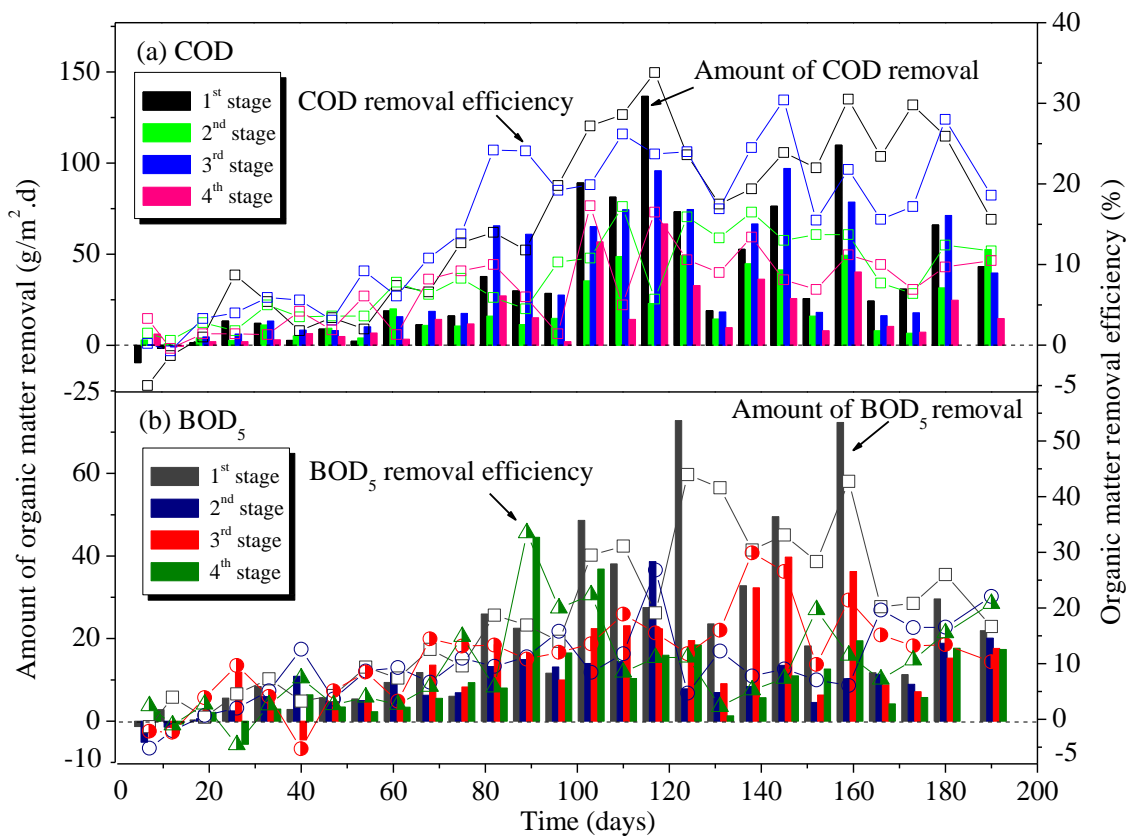


Figure 7 Contribution of individual stage to overall removal of organic matter during the operation period

DISCUSSION

As has been expected, examination of the alum sludge (see Table 3) indicated that it contains several metal ions that relate to P-metal adsorption and/or precipitation. Particularly, aluminium ion is the dominant component in the alum sludge, and it can potentially contribute to adsorption and chemical precipitation of P onto the alum sludge. Although the variety in experimental methodologies and conditions of each study may give different results, the reported P adsorption capacity of alum sludge ranges from 1.1 to 150.0 mg-P/g-sludge (Kim et al., 2003; Dayton and Basta, 2005; Novak and Watts, 2005; DeWolfe, 2006). Most recently, a long-term operation of a lab-scale CW employing an Irish alum sludge as wetland media has shown a strong affinity of phosphorus immobilization and estimated lifetime of the CW regarding the P adsorption saturation is 9-40 years (Zhao et al., 2009). Although the estimation of the lifetime of the alum sludge-based CW in this study was out of the scope of this paper, the effluent P concentration in this study was lower as 0.002 mg-P/l. The cumulative P adsorbed onto the alum sludge at 190 days of operation is 4.55 mg-P/g-sludge. Correspondingly, reduction of 99.5% of the P loading in reject water was achieved. Thus, it is reasonable to believe that the alum sludge-based CW under current investigation is reliable and can be operated for longer to achieve a secured P removal.

As shown in Figure 5 and Figure 7, the alum sludge-based CW system also presents high N, COD and BOD₅ removal rate under a very high HLR adopted in this study. This could be attributed to the biological degradation of the OM with attached growth bacteria on alum sludge. In addition, enhanced oxygen supply by using the tidal flow operation strategy is another factor to promote the OM oxidation. The theoretical oxygen supply rate of the CW system using tidal flow operation strategy is 137.2 g/m² d (Babatunde, 2007). However, the oxygen transfer capacity of the tidal flow CW system still need to be studied to reveal the ammoniacal-nitrogen and organic matter removal pathway and mechanisms in such system. It should be pointed out that there were negative COD and/or BOD₅ removal efficiencies being recorded during the operating time of 0 to 12 days (see Figure 6 and 7). This was a sign of the desorption/release of OM organic matters from the alum sludge as a result of the competitive adsorption between phosphate and humic substances for adsorption site on the surface of alum sludge. This has been previously investigated and reported (Yang et al., 2006).

Overall, the current study has demonstrated the reuse of a Chinese alum sludge as the main wetland substrate for reject water treatment in MWWTP. This is by for the first approach to propose such novel use of CW for reject water treatment. The suggested reject water treatment process can be sand filter followed by a multi-stage alum sludge-based CW system.

CONCLUSIONS

The removals of COD, BOD₅, N and P from reject water of a MWWTP using a novel alum sludge based multi-stage vertical subsurface flow constructed wetland system were investigated. The results shown that significant reduction up to 99.5% of P-feedback through the reject water recycling to the main wastewater treatment stream can be achieved using such alum sludge-based constructed wetland. Under high HLR of 1.0 and 1.26 m³/m² d with step feed and tidal flow operation strategy, the ammoniacal-nitrogen and organic matter removal perform well in the treatment system. The application of alum sludge-based constructed wetland for reject water treatment is a promising approach, representing a win-win scenario of using “waste” for wastewater treatment. The proposed overall treatment system is sand filter for removing SS augmented with a multi-stage alum sludge-based CW system.

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