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Nitrogen and phosphorus removal from WWTP effluent using a 2-stage continuous-backwash upflow filter system

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ABSTRACT

Several biological processes exist for simultaneous removal of nitrogen and phosphorus. These biological processes, such as activated sludge, SBR, A_2O, which involve one or more anaerobic-anoxic-aerobic reactors, are widely used globally due to their economic feasibility and environmental sustainability when compared to physical treatments. However, these processes have disadvantages, including large space requirements due to the arrangement of the reactors and relatively low phosphorus removal efficiencies. In this study, a 2-stage continuous-backwash upflow sand filter system was developed for tertiary treatment. 1st filter is operated for nitrification, and the biological denitrification occurred in 2nd filter simultaneously with the physical adsorption of phosphorus. Lab-scale batch tests were conducted to evaluate the influences of different parameters. Following this, the lab-scale continuous system was operated for about 50 d, and the nitrogen and phosphorus removal efficiencies were evaluated. By the end of the test (days 30–50), total nitrogen and total phosphorus removals of 87.4% and 92.7%, respectively, were achieved.

Keywords: Continuous-backwash upflow sand filter; Nitrogen removal; Phosphorus removal; Tertiary treatment

1. Introduction

Enrichment of nitrogen and phosphorus in surface water, especially in a confined stream or retention basin, causes eutrophication, which favors algal growth. This algal growth is more commonly known as an algal bloom. This phenomenon has harmful effects on water quality, habitats, and fish and other aquatic life through decreases in the dissolved oxygen (DO) concentration [1]. Nitrogen and phosphorus in discharged wastewater can be key inducers of eutrophication in the receiving water bodies [2].

The purpose of tertiary treatment is to provide a final treatment stage to further improve the effluent quality before it is discharged to the receiving environment (sea, river, lake, wetland, ground, etc.). More than one type of tertiary treatment process may be used at a given treatment plant to remove nutrients and toxins, and disinfect effluent. Advanced or tertiary treatment targets specific wastewater pollutants, such as nutrient (N and P) removal, and/or improved removal of other contaminants with advanced, and often expensive, technologies, such as coagulation–sedimentation and granulated activated-carbon adsorption [3].

Biological nitrogen removal is achieved through sequential nitrification under aerobic conditions and denitrification under anoxic conditions. During nitrification, ammonium (or ammonia) is oxidized to nitrite by ammonium oxidizing bacteria (AOB) and then to nitrate by nitrite oxidizing bac-

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teria (NOB). During denitrification, nitrite and/or nitrate is denitrified to nitrogen gas with organic carbon serving as the electron donor [2].

Different technologies have been applied for phosphate removal from wastewater, namely, physical treatments [4], chemical precipitation using ferric, calcium, or aluminum salts [5], biological treatments [6], and adsorption [7,8]. Adsorption is one of the most suitable technique for phosphate removal, particularly at low concentrations. Several types of phosphate adsorbents have been investigated, such as aluminum oxide and hydroxide, ferric oxide and hydroxide, inorganic sorbents, polymeric anion exchange resins, etc.

Simultaneous biological nutrient removal has been observed in full-scale system for several decades [9–11]. In the previous study, biological nitrogen removal is consistently observed, while biological phosphorus removal (BPR) occurs. These biological processes, such as activated sludge, SBR, A_2O, which involve one or more anaerobic-anoxic-aerobic reactors, are widely used globally due to their economic feasibility and environmental sustainability when compared to physical treatments. However, these processes have disadvantages, including large space requirements due to the arrangement of the reactors and relatively low phosphorus removal efficiencies. There were no previous studies on biological nitrogen removal and physical phosphorus removal using one structure.

In this study, a combined process of biological nitrogen removal and physical phosphorus removal process were developed. A 2-stage continuous-backwash upflow sand filter system was developed for simultaneous removal of nitrogen and phosphorus from sequencing batch reactor effluent. The 1st filter reactor was operated under aerobic condition, and 2nd filter reactor was operated under anoxic condition. In the 2nd filter reactor, biological removal of nitrogen and physical adsorption of phosphorus was occurred simultaneously. Biological denitrification and physical adsorption of phosphorus occurred simultaneously with the biological denitrification in the 2nd reactor. Lab-scale batch tests were conducted to evaluate the influences of different parameters. Following this, the lab-scale continuous system was operated for about 50 d, and the nitrogen and phosphorus removal efficiencies were evaluated.

2. Materials and methods

2.1. Experimental method

2.1.1. Batch-type test

To investigate the optimal parameters for the simultaneous removal of nitrogen and phosphorus, the lab-scale batch test was preceded by 1 L of reactor. The phosphorus removal test were conducted with 1 L of reactor, and 500 ml volume of silica sand were packed in the reactor. An artificial wastewater with 1.92 mg of PO_4^{3-}/L were used as raw water. To investigate the effect of $FeCl_3$ solution on the removal of phosphorus, $FeCl_3$ concentration was adjusted to 3, 5, 7, and 9 mg/L.

Effect of Mixed liquor suspended solids (MLSS) and Disolved oxygen (DO) concentration on nitrification were investigated with an artificial wastewater containing 20 mg/L of NH_4-N and 140 mg/L of HCO_3. HCO_3 were added to supply an enough alkalinity needed to the nitrification. Activated sludge from swine manure treatment facility were used as seed sludge. The test condition of MLSS were lower than 1,000 mg/L, and higher than 3,000 mg/L. The DO concentration were adjusted to be lower or higher than 3 mg/L by controlling aeration time.

In the test of biological denitrification, two reactors were operated at the temperature of 25° and 35° using water bath to compare the effect of temperature. Seed sludge were obtained from denitrification reactor of municipal wastewater treatment facility. DO concentration of each reactor was adjusted lower than 1 mg/L to maintain anoxic condition.

2.1.2. Design of continuous-backwash upflow filter

A size of upflow filter reactor is φ 200 mm × 1,200 mm H. Sand filter height was 1 m H and filtration area is 0.03 m^2. Silica sand used in the study has more than 90% of silica, and was under 2 mm of diameter. The filters were composed of layered cylinders. In the lower part of the inner cylinder, continuous backwash occurred by an air-lift pump. When the pressured air discharged, the media rise to the upper portion of the filter. As the media collided with each other, biofilms and adsorbate would detach from the surface of the medium.

2.1.3. Continuous-type test

The lab-scale continuous system is shown in Fig. 1. The 1st tank and 1st filter were operated under aerobic conditions for nitrification, and the 2nd tank and 2nd filter were operated under anoxic conditions for denitrification. The 3rd tank served to store the treated water. The feed composition is showed in Table 1. Aerobic and anoxic microbes obtained from activated sludge reactor and denitrification reactor of municipal wastewater treatment plant were seeded to the 1st and 2nd filter, respectively. The simultaneous biological denitrification and physical adsorption of phosphorus are occurred in 2nd filter. $FeCl_3$ solution for PO_4^{3-} adsorption and CH_3OH for carbon source of denitrification were added to 2nd anoxic reactor. Fig. 2 shows a mechanism of simultaneous removal of nitrogen and phosphorus.

The continuous system was operated for 50 d. The operating conditions of system are shown in Table 2. The 1st filter was operated with a 4 h hydraulic retention time (HRT), while the 2nd filter was operated with a 2 h HRT. This was done to compensate for the difference in growth rates of the nitrifiers and denitrifiers. 5 mg/L solution of $FeCl_3$was added along with influent to the 2nd filter.

2.2. Analytical method

The TN concentration was analyzed using Hach method 10071 (TNT Persulfate Digestion Method), while the TP concentration was analyzed using Hach method 8190 (ascorbic acid method with acid persulfate digestion). The NO_3^--N and NO_2^--N concentrations were analyzed in accordance with Hach method 10020 (chromotropic acid method) and Hach method 8507 (diazotization method), respectively. The ammonium concentration was analyzed using Hach method 10031 (Salicylate Method).

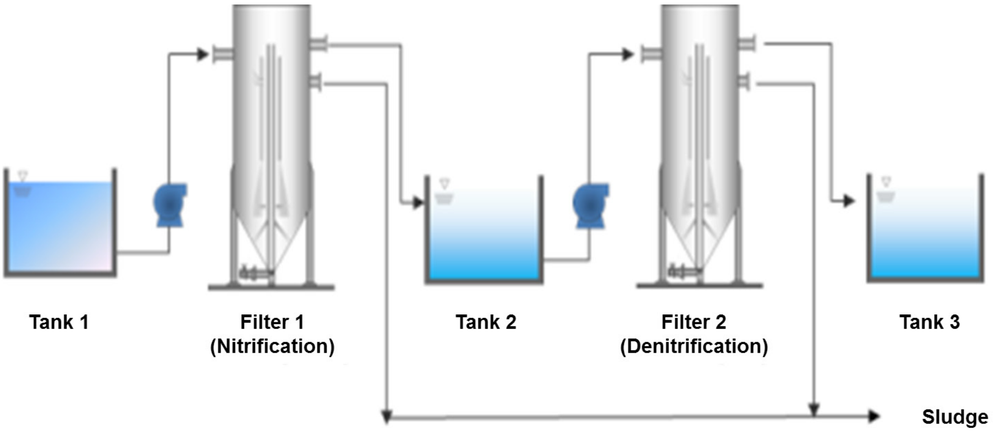


Fig. 1. Schematic diagram of the 2-stage continuous-backwash upflow sand filter system.

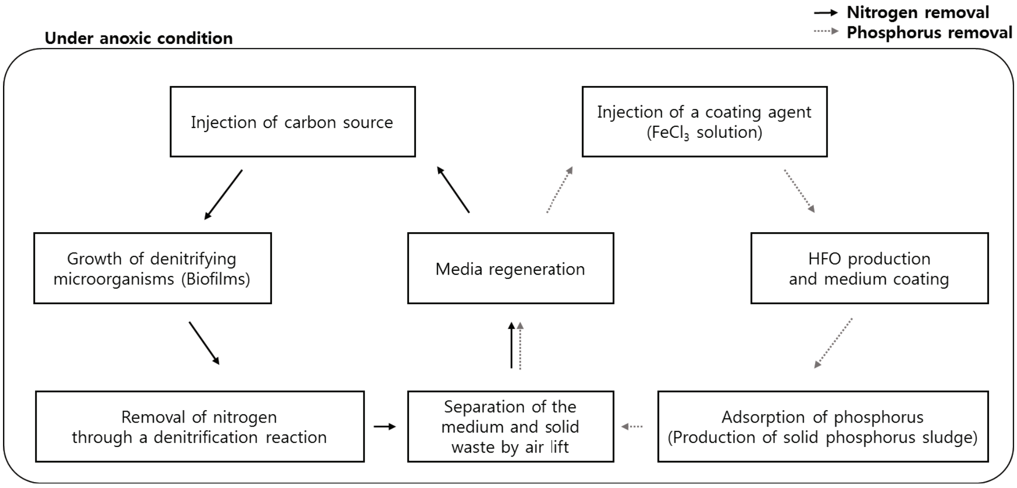


Fig. 2. A mechanism of simultaneous removal of nitrogen and phosphorus.

Table 1
Feed composition

Nutrient	Concentration
Total phosphorus	0.9±0.4
Total nitrogen	14.3±4.7
NO_3^--N	2.0±0.3
NH_4^+-N	11.3±4.7
SS	6.3±3.0
CODmn	6.7±2.0

Table 2
Operating condition

Parameters	Tank 1 + Filter 1	Tank 2 + Filter 2
Flow rate (L/h)	20	20
HRT (h)	4	2
Condition	Aerobic	Anoxic
Temp. (°C)	24.8±1.1	24.7±1.1
DO (mg/L)	3.1±0.4	0.6±0.3

3. Results and discussion

3.1. Optimization of experimental conditions in batch test

3.1.1. Effect of HFO-coated silica sand on phosphorus removal

Fig. 3 shows the phosphate removal by $FeCl_3$ concentration. Reactor with 5 ppm of $FeCl_3$ solution showed the best performance, with a phosphate removal of 56.3% after 1 h of treatment. When the $FeCl_3$ solution added to reactor, various hydrated ferric oxides (HFO) such as amFe$(OH)_3$, FepOr(OH)s·nH_2O, alpha-FeOOH is formed. The silica sand has a negative charge at pH 6.5–7.5, which is the general pH range of wastewater [12]. As ferrate ions have a positive charge, the silica sand surfaces are coated with ferrate by electrostatic attraction.

Zhou et al. studied about activated carbon filter with nano-sized HFO coating, and it possesses a higher adsorp-

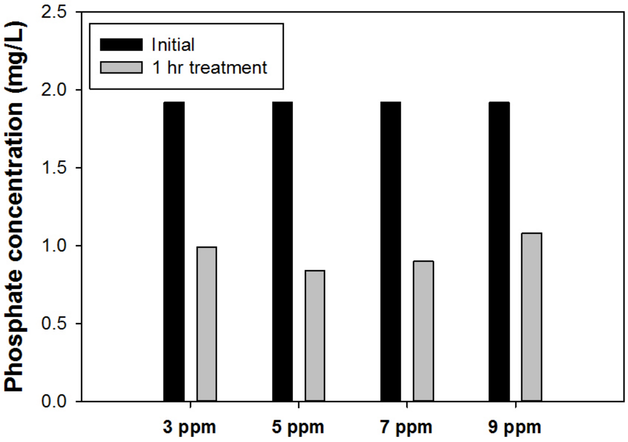


Fig. 3. Phosphate removal with HFO coated silica sand in the batch test (initial PO_4^{3-} concentration: 1.92 mg/L).

tion capacity for the phosphate than that of the hydrated ferric oxides. The ferric oxide coated on the surface of the media reacts with the phosphate in wastewater, according to following equations [Eqs. (1)–(3)] [13].

$$\equiv FeOH^0 + PO_4^{3-} + 3H^+ \leftrightarrow \equiv FeH_2PO_4 + H_2O \quad (1)$$

$$\equiv FeOH^0 + PO_4^{3-} + 2H^+ \leftrightarrow \equiv FeHPO_4^{-} + H_2O \quad (2)$$

$$\equiv FeOH^0 + PO_4^{3-} + H^+ \leftrightarrow \equiv FePO_4^{2-} + H_2O \quad (3)$$

3.1.2. Effect of DO and MLSS concentration on nitrification

To investigate the effect of DO and MLSS concentrations on nitrification, batch-type nitrification tests were conducted. There was no significant changes on DO concentration with 1000 mg MLSS/L (Figs. 3a and b). The best performance was exhibited in the condition of 3,000 mg MLSS/L and greater than 3 mg/L of DO; about 75% of the ammonium was converted to nitrate (Fig. 3d). From these figures, it can be seen that the MLSS concentration has a greater influence on nitrification than DO concentration. When the MLSS concentration was 3,000 mg/L and the DO concentration was below 3 mg/L, about 30% of the ammonium was converted to nitrate (Fig. 3c). By comparing Figs. 3c and 3d, the effect of DO concentration on nitrification was confirmed; it needs over 3 mg/L of DO concentration to nitrification.

3.1.3. Effect of operation temperature on denitrification

To investigate the effect of temperature on denitrification, two reactors were operated, one at 25°C and the other at 35°C (Fig. 5). Treatment at 25°C for 4 h resulted in 44.5% of nitrate removal, whereas treatment at 35°C for the same duration resulted in 89.3% of nitrate removal. The growth constants of nitrifying bacteria are greatly affected by temperature. The optimum temperature for the growth of nitrifying bacteria, according to the literature, is between 28°C and 36°C, although an optimum temperature of up to 42°C has been reported for nitrobacter by Painter [14].

In Figs. 4a and b, the pH increases with treatment time. In both cases, this increase was due to the removal of acidic nitrate.

3.2. Continuous system operating

3.2.1. Phosphorus removal

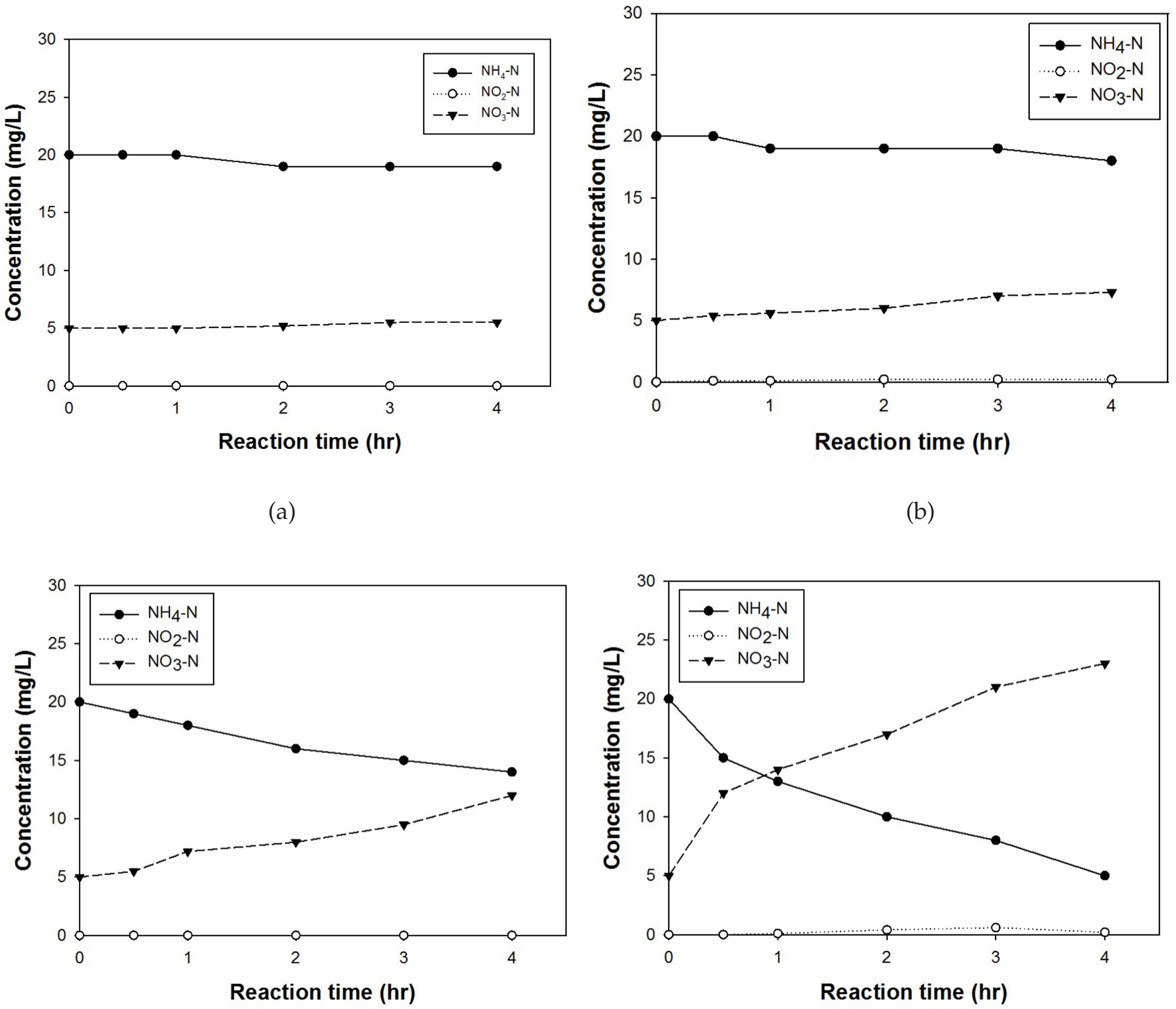
Fig. 6 shows influent and effluent TP concentrations, and the TP removal efficiency. As the result of the batch test, a 5 mg/L solution of $FeCl_3$ was injected to the influent of the 2nd filter. The average influent TP concentration was 1.4 ± 0.2 mg/L. On average, 77.8% of TP was removed during the operation. The removal efficiency of TP consistently increased, reaching over 90% after 30 d of operation, while the effluent TP concentration effluent consistently decreased. From day 40 onwards, the effluent TP concentration showed a stable and consistent trend, while the influent concentration increased. The adsorption efficiency of media increased as a result of repeated adsorption and desorption.

3.2.2. Nitrogen removal

Fig. 7 shows the influent and effluent TN concentrations and the TN removal efficiencies of filters 1 and 2. The influent TN concentration was approximately 10 mg/L for the first 30 d, after which it doubled. There was no shock due to over loading; the effluent TN concentration remained below 4 mg/L. Consequently, the TN removal efficiency increased to 87.4% after 30 d of operation, whereas only 73.8% was removed in the first 29 d. The effluent concentrations of 2nd filter were maintained under 5 mg/L of T-N. In the previous study on 2-stage biological nitrogen removal filter [15], T-N concentrations at the outlet of 2nd filter was maintained at about 1~5 mg/L for the influent concentrations of 12~36 mg/L.

Figs. 8a and b show the ammonium removal and nitrate conversion efficiencies. In the filter 1 process (Fig. 8a), around 7 mg/L of ammonium was injected consistently for the first 29 d. The average ammonium injection concentration was raised to 16 mg/L after 30 d. During the first 29 d, the effluent ammonium concentration remained consistently below 2.5 mg/L. More than 75% of ammonium was removed during this time. From day 30 to day 37, the effluent ammonium concentration increased to 10 mg/L and then stabilized at 5 mg/L after day 40. About 80% of the ammonium was removed in the last 10 d (days 40–50). As shown in Fig. 7(b), the influent nitrate concentration was about 2.5 mg/L throughout the operation. The effluent nitrate concentration was maintained at about 7 mg/L during the first 30 d. After day 30, as the influent ammonium concentration doubled, the effluent nitrate concentration increased gradually. After day 40, over 80% of ammonium was converted to nitrate.

Fig. 8c shows the denitrification efficiency of filter 2. In the first 30 d, the influent nitrate concentration was about 7 mg/L and increased to 15 mg/L after 30 d. The effluent nitrate concentration remained below 2.5 mg/L throughout whole operation. After 35 d of operation, the removal efficiency of nitrate was above 80%. It is lower than previous study on 2-stage biological filter system [15]; about 94% of nitrate were removed in 2nd filter in the study. The low



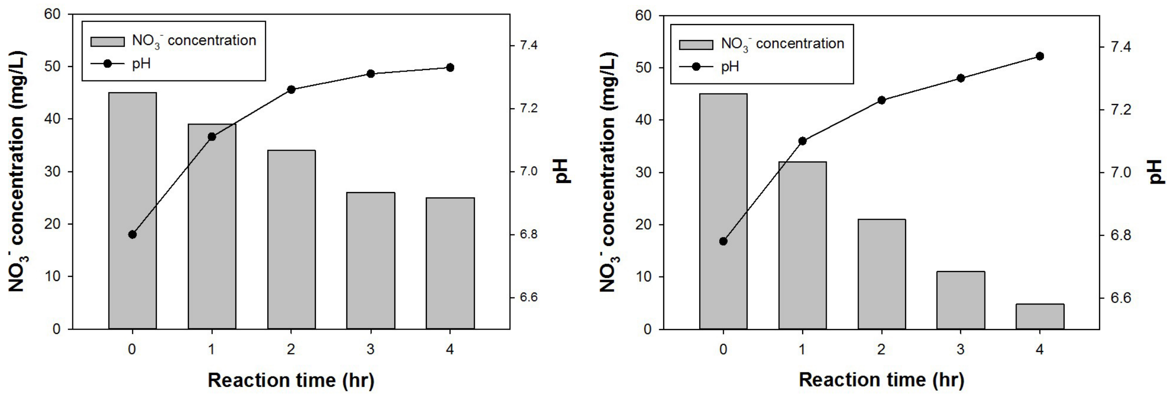
(a)

(b)

(c)

(d)

Fig. 4. Batch type nitrification test (a: MLSS 1,000 mg/L, DO under 3 mg/L, b: MLSS 1,000 mg/L, DO over 3 mg/L, c: MLSS 3,000 mg/L, DO under 3 mg/L, d: MLSS 3,000 mg/L, DO over 3 mg/L).



(a)

(b)

Fig. 5. Denitrification test in batch type reactor (a: operated in 25°C, b: operated in 35°C, initial nitrate concentration: 45 mg/L).

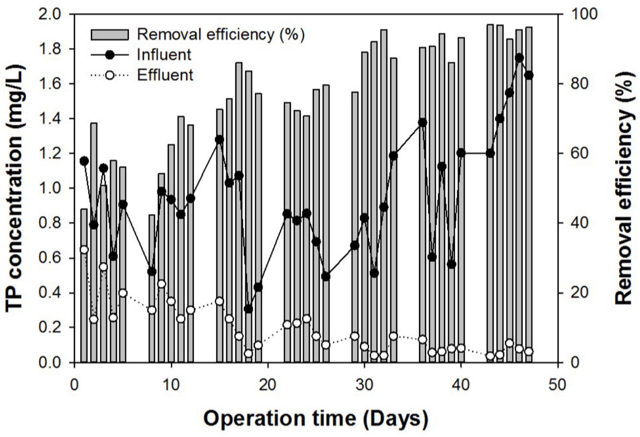


Fig. 6. Influent/effluent TP concentration and removal efficiency during the operation (filter 1 + filter 2).

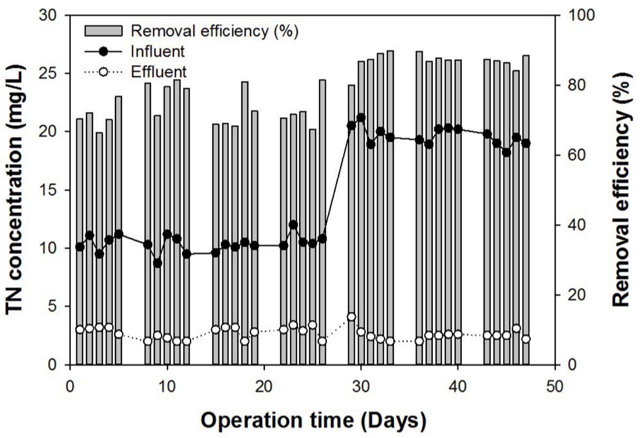
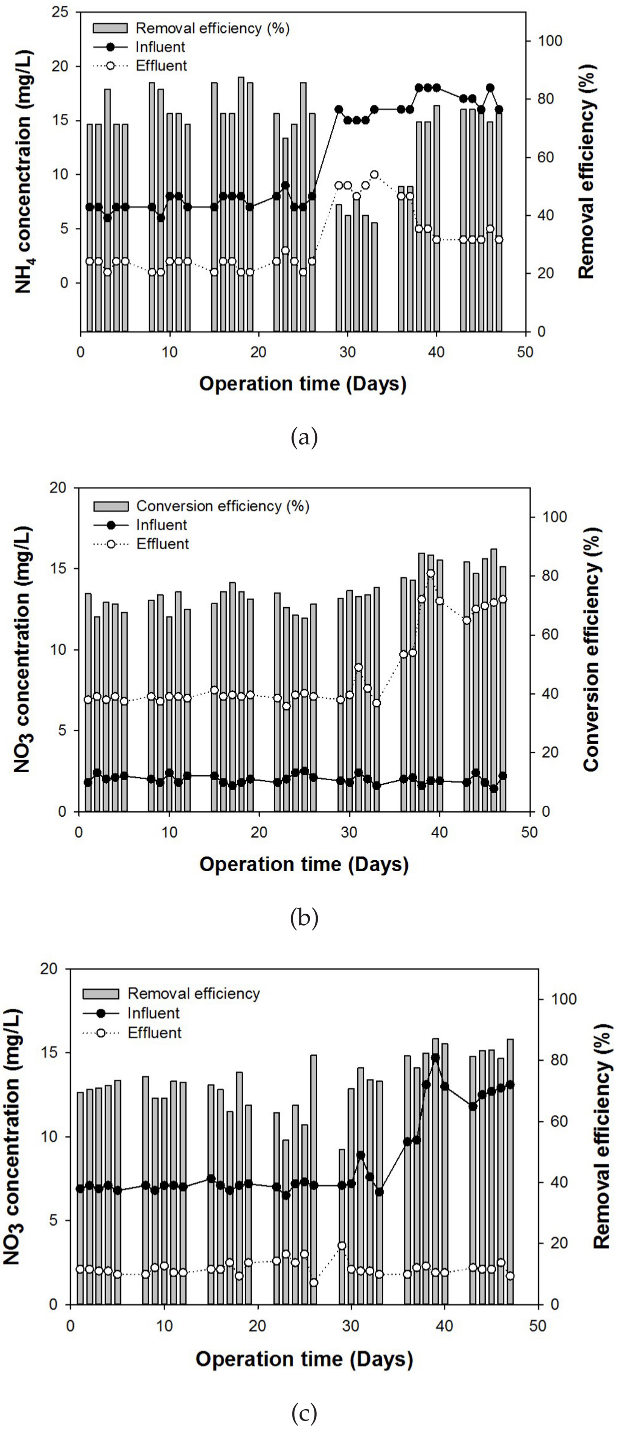


Fig. 7. Influent/effluent TN concentration and removal efficiency during the operation (filter 1 + filter 2).

denitrification efficiency was might caused by elimination of biofilm during continuous backwashing by air lift.

4. Conclusions

In this study, the nutrient removal efficiency of a 2-stage continuous-backwash upflow sand filter system was evaluated. Biological nitrification and denitrification processes and physical adsorption of phosphorus occurred simultaneously. The process was operated for 50 days, with the removal efficiency stabilizing after day 30. Finally, about 92.7% of TP and 87.4% of TN were removed in the process. Nitrogen processes were conducted in two steps: nitrification and denitrification. Over 80% of ammonium was converted to nitrate in the 1st filter, and about 80% of nitrate was removed in the 2nd filter. These results indicated that the 2-stage continuous-backwash upflow sand filter system can be applied to tertiary process for nutrient removal, but further study on full-scale WWTP plant is required to on demonstrate the advantages of this technology.



(a)

(b)

(c)

Fig. 8. Ammonium and nitrate concentration and removal efficiency during the operation (a: ammonium concentration in filter 1, b: nitrate concentration in filter 1, c: nitrate concentration in filter 2, ammonium concentration of filter 1 influent doubled after 30 d).

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