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# Novel two-stage vertical flow biofilter system for efficient treatment of decentralized domestic wastewater

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ABSTRACT

A novel two-stage system consisting of a trickling filter and a multi-soil-layering (MSL) bioreactor was developed for enhanced treatment of domestic wastewater from decentralized sources. The trickling filter was packed with coarse zeolite segregated by 3 thin iron scraps layers, referred to as the Iron modified Zeolite Trickling Filter (IZTF). The effective particle size of the zeolite ranged 3–5 mm, and the zeolite to iron scraps ratio was 95:5 by dry weight. The MSL was a layered filter system consisted of Zeolite permeable layers (ZPL) and Soil mixture block (SMB) layers in alternated form. Simulated septic tank effluent was fed into the apparatus through a perforated pipe on top of the trickling filter at a loading rate of 440, 640 and 920 $L\,m^{-2}\,d^{-1}$, respectively, and dropped into the MSL. Total removal rates of $COD_{Cr}$, TP, Ammonium, and TN ranged 90.3%–95.2%, 92.0%–94.0%, 85.1%–86.9%, and 58.9%–63.8% with mean final effluent concentrations of 12, 0.28, 5.66, and 21.0 $mg\,L^{-1}$, respectively at mean hydraulic loading rate of 920 $L\,m^{-2}\,d^{-1}$ and under relatively stable conditions. The system operated without any sign of oxygen shortness even at the maximum loading rate of 250 $g\,COD_{Cr}\,m^{-2}\,d^{-1}$ and 40 $g\,NH_4^{+}\text{-}N\,m^{-2}\,d^{-1}$ without artificial aeration, and clogging did not occur during the study owing to better water dispersion of the MSL, which indicated that the hybrid system could operate well for COD removal and nitrification with little management and labor demand. A comparative study also showed that the IZTF with identical total packing height and iron proportions performed better than the single MSL in TP removal, and the aerobic condition as well as the partial high Fe:P ratio was hypothesized to be the dominant factor for that.



## 1. Introduction

In developing countries like China, because of the lack of management of the sewage in vast rural areas, rural water pollution sources are becoming more and more prominent (Chen et al., 2006; Wang et al., 2011). Since rural wastewater changes greatly in quality, quantity, and spatial distribution, the decentralized treatment, characterized in disposing on-site and reusing on-site, proves a much better choice as it avoids plumbing works for water collection (Massoud et al., 2009; Pant et al., 2013; Wang et al., 2011). In order to be economically affordable, environmentally sustainable, and socially acceptable (ElMekawy et al., 2013; Massoud et al., 2009), the technologies should be low-cost, energy saving, low-maintenance, effective, and stable (Aiyuk et al., 2004; Heistad et al., 2006).

Typically, decentralized on-site wastewater management for an individual home consists of a septic tank for pretreatment and an effluent dispersal system (Brix and Arias, 2005; Dawes and Goonetilleke, 2003; Leverenz et al., 2010). For nitrogen and phosphorus removal, the treatments mainly include Constructed Wetlands (CW), Intermittent Sand Filters (ISF), and Land Treatment Systems (LTS) (Healy et al., 2007; Massoud et al., 2009; Tanner et al., 2012). However, following population growth and urbanization, the cost and availability of land is becoming a restrictive factor that demands efficiency in terms of facility area (Aiyuk et al., 2004; Parkinson and Tayler, 2003; Sevda et al., 2013). The Vertical Flow CW (VFCW) promises a good alternative that satisfies the aforementioned requirements well (Brix and Arias, 2005). It was reported the most common design in France was the two-stage VFCW, which accepted raw sewage intermittently in the first stage

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mainly with coarse gravel and fulfilled the nitrification in the second stage with much finer media. Such systems, if well designed, can achieve an outlet level of 60 mg $L^{-1}$ in COD, 15 mg $L^{-1}$ in SS and 8 mg $L^{-1}$ in TKN with an area of 2–2.5 $m^2$ $PE^{-1}$ (person equivalent) (Molle et al., 2005), less than official guidelines of 3.2 $m^2$ $PE^{-1}$ by the Danish Ministry of Environment for the on-site treatment of domestic sewage (Brix and Arias, 2005).

Zeolite is aluminosilicate mineral with the features of high ion exchange capacity, advanced porous structure (molecular sieve) and low density and has good ion exchange selectivity for the $NH_4^+$ (Gottardi, 1978). It is reported that zeolite usage is helpful to the nitrifiers in the filter and enhances the ammonium removal of the filter, which promotes the regeneration of the $NH_4^+$ saturated zeolite in return (Lahav and Green, 1998; Yidong et al., 2012). Thus it would be promising if zeolite should be used as the filter media for efficient control of nitrogen in domestic wastewater treatment.

Many studies on multi-soil-layering system (MSL), a novel soil based technology that tries to make use of the soil to improve the treatment efficiency of the VFCW, have been conducted (Attanandana et al., 2000; Luanmanee et al., 2001, 2002; Masunaga et al., 2007; Sato et al., 2011; Wakatsuki et al., 1993). In the MSL, soil is made into brick-like blocks and packed alternately, with the margins filled with permeable granular zeolite. The system generally consists of several Soil block layers alternated with Zeolite permeable layers to a total height of about 1 meter or more. Some other materials such as the wood chips and granular iron are often mixed into the soil before making the blocks (Soil mixture block, SMB) to enhance the nitrogen and phosphorus removal. Wastewater is fed into the apparatus from the top and drained through the porous pipes equipped at the bottom. Generally, the MSL system performs quite well in COD, TP, and ammonium removal under relatively higher hydraulic loading rates, but intermittent aeration is needed to avoid clogging and ensure stable removal efficiency of the COD and the TP for long period operation (Attanandana et al., 2000; Luanmanee et al., 2002). TN removal often drops with artificial aeration due to the drain of the nitrate but it would rise again when aeration is no longer provided (Luanmanee et al., 2001, 2002). In former studies, ammonium removal rate would drop after long periods of operation without aeration, which reduces the TN removal as well (Luanmanee et al., 2001; Masunaga et al., 2007). Thus MSL system would call for some extra cost and excessive management in terms of the artificial aeration, which would restrict its implementation in decentralized rural sewage treatment.

In this study, a novel hybrid two-stage vertical flow system that targeted the removal of pollutants such as $COD_{Cr}$, nitrogen and phosphorus simultaneously in sewage from non-centralized sources was developed. A coarse zeolite trickling filter (ZTF) with low packing height was utilized to adsorb and transform the ammonium as well as to trap and degrade the organic matters to some extent before a MSL bioreactor was used for further treatment to ensure the quality of the effluent. It was assumed that iron was easier to ionize under aerobic conditions and the precipitate would be intercepted or adsorbed more efficiently in soil, so iron scraps were added into the ZTF in addition to the SMBs to promote phosphorus removal. We were quite concerned about the stability and maintenance requirements of this treatment plant and tried to omit artificial aeration and decrease the probability of clogging. Pollutant removal mechanisms would be analyzed according to the results.

## 2. Materials and methods

### 2.1. Experimental apparatus and materials

The schematic showing of the experimental apparatus is presented in Fig. 1. Lidless acrylic boxes with apertured bottoms (the total aperture area rate was 28%) were packed with different media to simulate the biofilters and they all measured 32 cm in width, 16 cm in thickness and 60 cm in height. Wastewater was pumped into the boxes using a submerged pump controlled by a time switch (DH48S-S) and dispersed into the filter media through a perforated pipe.

The structure and detailed dimensions of the IZTF and MSL were shown in Fig. 1b, and the material compositions of their segments as well as the material properties were listed in Table 1.

The natural zeolite was produced in Jinyun, Zhejiang province of China with the effective particle size of 3–5 mm which was a little larger than that in many earlier studies where the particle size was 1–3 mm (Luanmanee et al., 2001; Masunaga et al., 2007; Wakatsuki et al., 1993). The SMBs were formed using the mixture of clayey soil, sawdust and iron scraps with the ratio of approximately 75%, 10% and 15%, respectively, by dry weight (Luanmanee et al., 2001, 2002). The soil was got from the Yuelu Mountain in Changsha, China and was half air dried and crushed before sifting through the meshed screen with the mesh size of 2 mm. The iron scraps were lathe iron cutting scraps got from the metal technology practice base of Hunan University and was washed in boiled 5% NaOH solution for 30 min to remove the adsorbed oil and activated in the diluted HCl solution for about 15 min before using.

### 2.2. Wastewater for treatment and operational conditions

Wastewater used for the tests was prepared using glucose, starch, $NH_4Cl$, etc. to simulate the typical rural sewage pretreated using the septic tank. The simulated wastewater was prepared by dissolving glucose 30 g, starch 10 g, $NH_4Cl$ 20 g, $KH_2PO_4$ 0.8 g, $K_2HPO_4$ 1.6 g, $NaHCO_3$ 10–12 g, $MgSO_4$ 2 g, $CaCl_2$ 2 g, $MnSO_4$ 0.3 g and peptone 0.5 g into every 100 L tap water. The simulated water quality is listed in Table 2.

The apparatus mainly operated throughout three phases.

Comparisons between the ZTF and the MSL with the same packing height were conducted after the start up of the apparatus in phase 1. To seed the filters, simulated wastewater was mixed with condensed activated sludge got from the aeration tank of the Guozhen wastewater treatment plant in Changsha, China with the volume ratio of 2:1 and agitated using a circulating pump before fed into the apparatus. The feeding of activated sludge remained for a whole week and only simulated wastewater was fed thereafter. The Hydraulic Loading Rate (HLR) was kept at approximately

**Table 1**
Properties of the experimental materials and their proportions to form different filter segments.

| | Zeolite | Soil | Sawdust | Iron scraps |
|---|---|---|---|---|
| Bulk density (packing density) (g $cm^{-3}$) | 0.79 | 1.31 | 0.12 | 0.73 |
| Granularity (mm) | 3–5 | Light Red soil (<2 mm) | 1–3 | 1–5 |
| *Mass percent by dry weight* (%) | | | | |
| ZTF | 100 | 0 | 0 | 0 |
| IZTF | 95 | 0 | 0 | 5 |
| SMB | 0 | 75 | 10 | 15 |

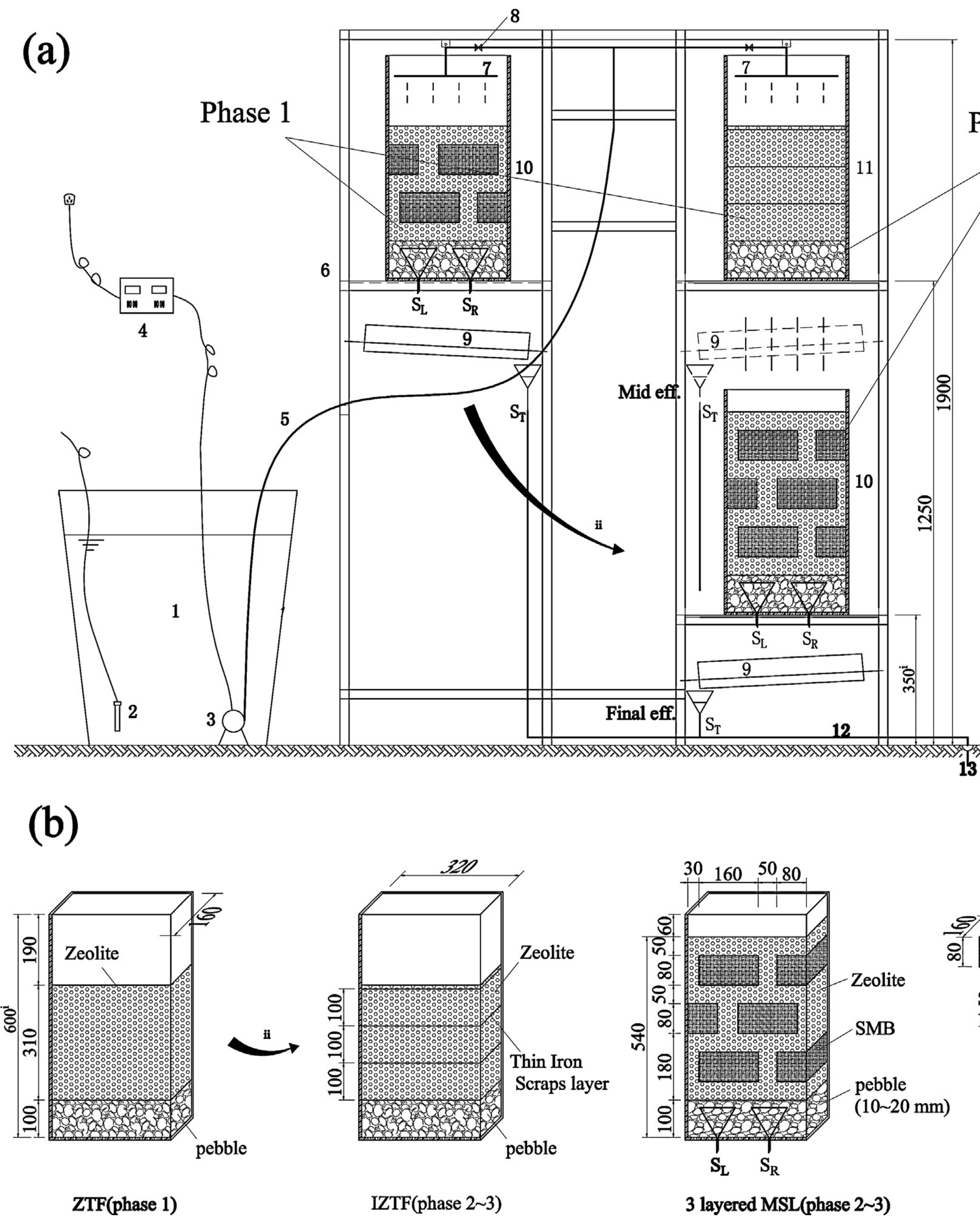


**Fig. 1.** Schematic showing of the experimental apparatus: (a) the apparatus, where: 1 raw water tank; 2 electrical heating bar;3 Submerged pump;4 Time switch; 5 Influent pipe; 6 Supporting iron stand; 7 perforated distribution pipe; 8 valve; 9 demountable V tank; 10 MSL module; 11 IZTF module; 12 Drainage pipe; 13 Floor drain. S* Sampling points; (b) structure of the IZTF and MSL. Notes: (i) The dimension unit is mm. (ii) The arrows indicate the modifications of the apparatus from phase 1 to phase 2.

$240\,L\,m^{-2}\,d^{-1}$ during the startup and was raised to about $440\,L\,m^{-2}\,d^{-1}$ later during phase 1.

In phase 2, the ZTF and the MSL both got some modification before the MSL was moved right under the ZTF to form the two-stage IZTF + MSL system for long term operation study (Fig. 1a,b), as it turned out in phase 1 that the 2 layered MSL performed worse in COD and ammonium removal than the ZTF and TP removal was also far from satisfactory. The ZTF was rebuilt by paving a thin layer of iron scraps and zeolite mixture (3:1 by weight and to avoid iron agglomeration) every 10 cm thick zeolite layer, expecting for more phosphorus removal, which would be refered to as "Iron modified Zeolite Trickling Filter (IZTF)" later. While the 3 layered MSL was reconstructed by building another soil mixture block layer and zeolite permeable layer in the original MSL, increasing its packing

**Table 2**
Simulated wastewater quality (determined in the second experimental phase).

| Items | $COD_{Cr}$ ($mg\,L^{-1}$) ($n = 13$) | TN ($mg\,L^{-1}$) ($n = 14$) | $NH_3$-N ($mg\,L^{-1}$) ($n = 14$) | TP ($mg\,L^{-1}$) ($n = 12$) | pH ($n = 8$) |
|---|---|---|---|---|---|
| Values | 214 ± 61[a] | 58.6 ± 4.92 | 42.8 ± 2.76 | 4.72 ± 0.97 | 6.50 ± 0.29 |

[a] Mean value ± standard deviation.

**Table 3**
The detected water production rate of different sampling points during different experimental phases.

| | | MSL | | | (I)ZTF |
|---|---|---|---|---|---|
| | | $S_L$[b] ($mL\,h^{-1}$) | $S_R$ ($mL\,h^{-1}$) | $S_T$ ($mL\,min^{-1}$) | $S_T$ ($mL\,min^{-1}$) |
| Phase 1 | A | 64.4 ± 25.9[a] | 161.9 ± 34.9 | 249.9 ± 48.5 | 238.6 ± 52.3 |
| | B | 156.2 ± 11.4 | 311.9 ± 119.8 | 443.8 ± 95.1 | 439.7 ± 89.2 |
| Phase 2 | A | 180.4 ± 52.7 | 200.1 ± 17.3 | 440.7 ± 25.4 | N.D.[c] |
| | B | 315.5 ± 67.6 | 350.0 ± 68.4 | 631.4 ± 34.8 | N.D. |
| | C | 325.7 ± 4.5 | 409.3 ± 110.6 | 917.0 ± 89.0 | N.D. |

[a] Mean value ± standard deviation.
[b] $S_L$, $S_R$ represent partial samples taken under the SMB and under the slot between SMBs, respectively. $S_T$ is full samples taken under lower ends of the V tanks.
[c] N.D. means not detected or omitted since replaceable data existed.

height to about 54 cm. "New" zeolite was mixed with "old" zeolite before using to speed up the microorganism growth in the latter phase.

In this phase, mean HLR was set at 440, 640, and 920 $L\,m^{-2}\,d^{-1}$ in steps, and the HLRs was not changed until the effluent water quality became stable again. Water samples were continuously taken and analyzed as scheduled except during the winter holiday when the apparatus operated under a very low hydraulic loading rate for three weeks. The loading rate of 640 $L\,m^{-2}\,d^{-1}$ was recovered after that and kept for another 3 weeks (according to the results gained at the beginning of this phase) to ensure the recovery of the apparatus before continuing the sampling and analyzing.

Performance of the apparatus under hydraulic shock load was studied in phase 3. The HLR of the apparatus was set at about 1600 $L\,m^{-2}\,d^{-1}$ for 3 h from 16:00 to 19:00 in the afternoon and was kept at 440 $L\,m^{-2}\,d^{-1}$ for the rest of the day everyday. According to the results gained in phase 2, the apparatus operated under that condition for three weeks before the sampling and analyzing. Samples were taken at 18:30 and 21, 23, 07, 15 o'clock, respectively, as a whole cycle.

The time interval between water feedings was 12 min and the HLR was changed by adjusting the water feeding time using the time switch. HLR was measured through dividing the volume of the effluent by the sampling time under stable running conditions and was usually recorded with other effluent parameters at the same time. The summarized HLR detection results were listed in Table 3. The perforated influent pipe was often taken down and cleaned up to ensure a relatively constant influent rate throughout the whole study. The ambient temperature and water temperature were recorded continuously during the whole research. At the beginning of phase 2 (about day 1 in the figures), a thermostatic electrical heating bar was put into the influent tank to keep the influent temperature at about 16 °C from then on as the ambient temperature dropped a lot at that time and was banned after the winter holiday when the temperature rebounded.

### 2.3. Water sampling and analytical methods

To inspect the mechanism of the MSL in pollutant removal, two funnels were buried into the pebble layer at the bottom of the MSL module right under the SMB and the gap between two SMBs, respectively as wastewater may went through different numbers of SMB layers before getting collected, and the samples were named "$S_L$" and "$S_R$", respectively. Mid effluent samples of the former IZTF stage and final effluent samples of the latter MSL stage were taken at the lower ends of the V tanks which were named "Mid eff." and "Final eff.", respectively (Fig. 1a,b).

Chemical analysis of every batch water sample was accomplished in two days, and samples were kept in the refrigerator at about 4 °C when time not allowed. The analysis methods were: $COD_{Cr}$ using the potassium dichromate method; SS by the gravimetric method; TN by the potassium persulfate oxidation-Ultraviolet spectrophotometry; $NO_3^-$-N by the ultraviolet spectrophotometry; TP by the Potassium persulfate digestion-colorimetric Method; and water pH using a composite pH electrode, all according to standard methods (APHA, 1998) except for $NH_4^+$-N by the nessler's reagent colorimetric method according to Chinese national standard methods (EPA of China, 2002).

## 3. Results and discussion

### 3.1. Performance of the hybrid IZTF + MSL system under relatively constant conditions

In this part, mainly the series of data gained during the second phase was inspected, when the hybrid IZTF + MSL apparatus was assembled and fed with wastewater and operated through a nearly four-month phase continuously under relatively constant environmental conditions except for the increased loading rate step by step. Results from the phase 1 were also introduced in the discussion.

#### 3.1.1. Oxygen demand pollutant removal

Data of the upper IZTF stage effluent and the lower MSL stage effluent were both plotted which were marked with the suffixes of "mid" and "final", respectively (Fig. 2).

Despite the good biodegradability of the organic matters in the simulated wastewater, this result indicated that the trickling filter structure was not short for oxygen supplicant for the removal

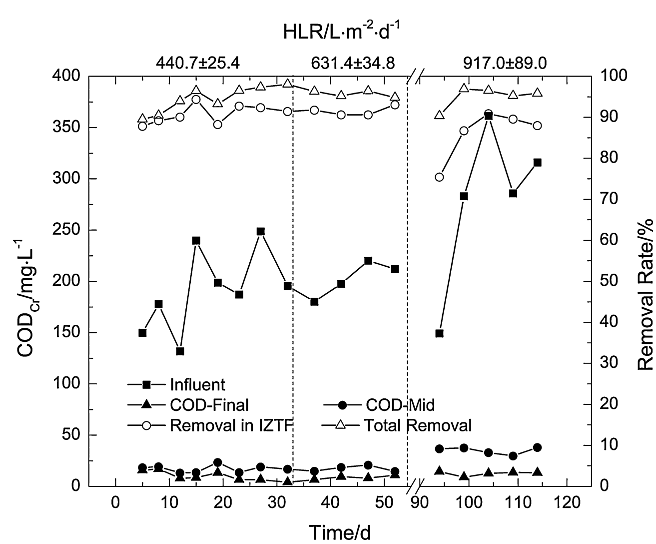


**Fig. 2.** $COD_{Cr}$ and its removal rate in the hybrid apparatus during experimental phase 2.

**Table 4**
Comparisons among two-layered MSL, ZTF and IZTF with the same low packing height of 41 cm as single reactors under direct raw wastewater feeding.

| | | Influent (phase 1, $n = 4$) | MSL eff. (phase 1) | ZTF eff. (phase 1) | IZTF (phase 2) | |
|---|---|---|---|---|---|---|
| | | | $S_T$ ($n = 8$) | $S_T$ ($n = 8$) | Influent ($n = 6$) | $S_T$ ($n = 6$) |
| HLR ≈ 240 L m$^{-2}$ d$^{-1}$ | $COD_{Cr}$ | 265 ± 17[a] | 19 ± 8(92.8) | 14 ± 4(94.7)[b] | – | |
| | Ammonium | 41.1 ± 3.52 | 6.05 ± 1.51(85.3) | 3.94 ± 0.78(90.4) | – | |
| | Nitrate | – | 13.9 ± 1.00 | 16.4 ± 0.48 | – | |
| | Total nitrogen | 60.0 ± 3.59 | 21.9 ± 1.91(63.5) | 21.9 ± 3.28(63.5) | – | |
| | Total phosphorus | 5.32 ± 0.46 | 2.2 ± 0.26(58.6) | 3.98 ± 0.42(25.2) | – | |
| HLR ≈ 440 L m$^{-2}$ d$^{-1}$ | $COD_{Cr}$ | 210 ± 15 | 44 ± 13(79.0) | 16 ± 5(92.4) | 188 ± 36 | 17 ± 5(91.0) |
| | Ammonium | 40.6 ± 3.36 | 17.5 ± 1.13(56.9) | 7.49 ± 0.40(81.6) | 43.7 ± 2.39 | 7.66 ± 0.65(82.5) |
| | Nitrate | – | 5.69 ± 1.48 | 9.38 ± 2.60 | – | 8.05 ± 2.90 |
| | Total nitrogen | 57.7 ± 3.69 | 27.5 ± 4.34(52.3) | 20 ± 2.90(65.3) | 62.2 ± 5.19 | 18.7 ± 3.15(69.9) |
| | Total phosphorus | 4.61 ± 0.66 | 2.43 ± 0.26(47.3) | 3.85 ± 0.73(16.5) | 5.04 ± 0.32 | 0.82 ± 0.04(84.0) |

[a] Mean value ± standard deviation; the units are mg L$^{-1}$.
[b] The values in the round brackets represent the mean removal rate (%) of the corresponding pollutants.

of the COD with the general concentration of a typical septic tank effluent even under the HLR of 1000 L m$^{-2}$ d$^{-1}$, corresponding to the maximum organic loading rate (OLR) of approximately 250 g $COD_{Cr}$ m$^{-2}$ d$^{-1}$. Furthermore, the comparative study in phase 1 also showed that COD and ammonium removal as well as effluent nitrate concentration of the (I)ZTF group was noticeably greater than the MSL group (Table 4), indicating that the zeolite trickling filter was more efficient in COD digestion and ammonium nitrification which might owing to the coarse filling character that enhanced the oxygen recovery in the bioreactor. From the hydraulic aspect, the results (Table 3) indicated that the MSL influent dispersion was evener when upper IZTF effluent acted as MSL influent than when wastewater was directly dispersed into the MSL through the porous pipe. Thus, the contact of wastewater and SMBs increased (Sato et al., 2005), enhancing the removal of organic matters as they would be adsorbed physically and chemically and subsequently decomposed by microorganisms in soil (Luanmanee et al., 2002; Sato et al., 2011) which has high purification capacity due to its advanced porous, water-air, and chemical features (Chen et al., 2009; Luanmanee et al., 2001). As a result, the final effluent of the hybrid IZTF + MSL system contained COD in extremely low concentration of less than 20 mg L$^{-1}$, disregarded of the varied organic loading rates tested.

### 3.1.2. Phosphorus removal

The hybrid IZTF + MSL apparatus exhibited excellent performance in TP removal throughout the whole study. The effluent TP was always stable at a concentration of lower than 0.5 mg L$^{-1}$ with a constant mean removal rate of 94.8% despite the fluctuation of the influent TP concentration (Fig. 3a).

Comparison of the performances between the single MSL system and the IZTF with the same gross content of iron scraps (about 5% by dry weight) under the same HLR of about 440 L m$^{-2}$ d$^{-1}$ in phase 1 showed that the iron scraps paved in the aerobic zeolite permeable layer had much more remarkable effect in phosphorus fixing than mixed in the SMBs (Table 4). This might because iron transforms into $Fe^{3+}$ more easily in the aerobic permeable zeolite layer and the hydrolysis of ferric ion is slower than the ionic sedimentation reaction to form ferric phosphate at slower iron oxygenation rates according to Svanks's (1971) study which also found the optimized pH for phosphate removal by ferric ion was higher than 6.5 and the removal rate could reach 98% in oxygen saturated solution. The precipitate would either be adsorbed or got intercepted into the filter media especially when passing through the SMBs. Moreover, the structure of the IZTF provided a high Fe:P ratio at partial zones, which enhanced the phosphorus removal (Fytianos et al., 1998).

TP removal rate was also plotted against influent HLR (Fig. 3b), the results showed that the TP removal rate of the IZTF was negatively linear correlated with the HLR with the correlation coefficient of −0.8342, which indicated that the phosphorus removal was mainly due to the chemical precipitation which was a process mainly restricted by the contact time between orthophosphate and ferric iron when influent TP concentration was relatively high. The data of the MSL part showed little sign of that relationship as its

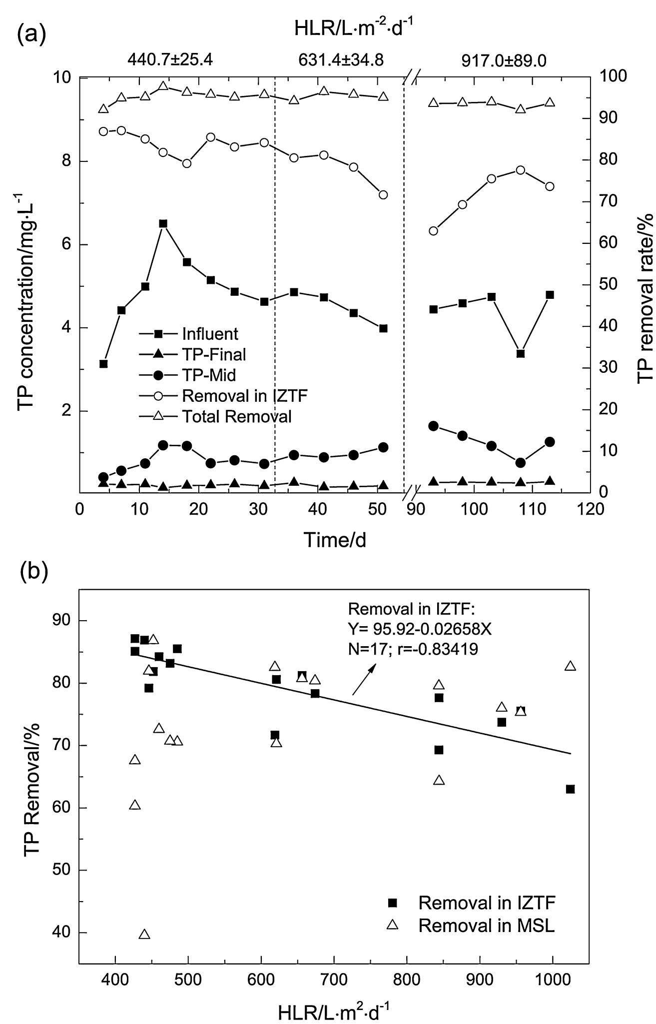


**Fig. 3.** Diagram of TP removal in phase 2: (a) TP concentration and removal rates; (b) TP removal rate versus influent hydraulic loading rate.

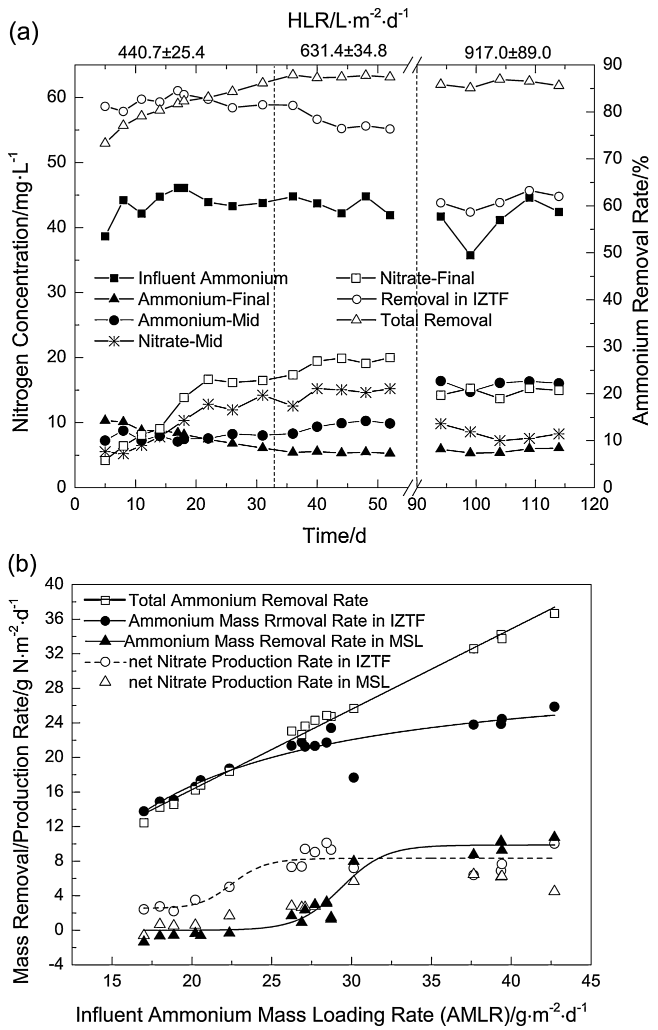


**Fig. 4.** Performance of the hybrid apparatus on ammonium removal and transformation: (a) the concentrations and removal rates with time; (b) ammonium mass removal rate and nitrate production rate versus influent ammonium loading rate.

influent TP was less than or around 1 mg $L^{-1}$ and the iron scraps only existed in the SMBs and the restricted process may changed to the iron ion production and diffusion processes. However, Mean TP removals in the MSL under direct inflow (in phase 1) and the MSL under the IZTF (based on the mid IZTF effluent) were 47.3% and 67.8%, respectively (under the same mean HLR of 440 L $m^{-2}$ $d^{-1}$), which might also because the influent dispersion of the latter MSL was evener, thereby enhanced the contact of wastewater with the iron scraps.

### 3.1.3. *Ammonium removal and nitrification*

Fig. 4 showed the performance of the apparatus in ammonium removal and nitrate production during the whole experimental phase. The Ammonium Mass Removal Rates (AMRR) of different parts of the hybrid system against the influent Ammonium Mass Loading Rate (AMLR) was plotted in Fig. 4b.

The results showed that the final effluent ammonium kept at a low level of about 5 mg $L^{-1}$, with the removal rate of about 85% under the experimental AMLR of 15–45 g $m^{-2}$ $d^{-1}$, which showed remarkable stability. But ammonium concentration of the mid effluent from the IZTF stage showed visible increments when influent HLR increased. Effluent nitrate concentration increased steadily with time at the beginning of phase 2, indicating continuous growth of nitrifiers in the biofilter. The increase continued for about 22 days before it finally became relatively stable.

Judging from the AMRR (Fig. 4b), the ammonium removal and the Nitrate Production Rate (NPR) of the IZTF stage showed similar trend: they increased significantly with influent AMLR and became stable as the loading rate reached about 28 g $NH_4^+$-N $m^{-2}$ $d^{-1}$. The final AMRR and NPR were approximately 24 g $NH_4^+$-N $m^{-2}$ $d^{-1}$ and 8 g $NO_3^-$-N $m^{-2}$ $d^{-1}$, respectively. That result indicated that within the study scope and period and under the relatively stable environmental conditions, the nitrification rate of the IZTF would reach a relatively stable level in the changes of the loading, which might because of the limited gross nitrifier biomass on the filler surface.

The nitrification rate in IZTF did not show obvious drop even when influent OLR increased to 250 g COD $m^{-2}$ $d^{-1}$, which was quite different with the results of previous studies on trickling filters packed with inert filler. For example, it has been recently shown that the increase in $BOD_5$ load had adverse influence on the nitrification capacity of the nitrifying trickling filter (Van den Akker et al., 2011) and effluent ammonia showed more remarkable fluctuation under a longer organic shock load (Hu et al., 2011). Several reasons may account for that: firstly, the zeolite as the filter media was much more capable to adsorb the ammonium than traditional materials owing to its specific cation exchange function for the ammonium (Gottardi, 1978), providing abundant substrate for the nitrifiers regardless of the fluctuation of influent (Lahav and Green, 1998); secondly, the relatively large particle size of the media allowed the existence of the air layer around the biofilm; thirdly, the extremely low HLR (<1 $m^3$ $m^{-2}$ $d^{-1}$), the intermittent inflow and fast drainage in the IZTF promoted the air flow and oxygen diffusion to the biofilm (Lahav et al., 2001). All of that benefited the nitrifiers and made the apparatus a promising nitrifying bioreactor (Yidong et al., 2012).

The AMRR of the IZTF was constantly higher than the mid effluent NPR, indicating the remarkable existence of adsorption and simultaneous denitrification in the stage. Since high HLR provided more carbon resources, reduced the HRT and consumed most DO, the remarkable denitrification might be mainly attributed to the activities of polyhydroxyalkanoates (PHA) storage bacteria according to Krasnits et al.'s (2013) research which indicated that entrapped particulate organic matter contributed to the reducing power for denitrification in biofilm reactors.

As for the MSL system, the AMRR in the MSL increased dramatically with the increment of its influent AMLR, which ensured the final effluent ammonium concentration to stay at a low level. The net NPR of the MSL showed a similar trend to the IZTF at the beginning but slightly decreased at high influent loading rates.

However, when the AMLR was very low, the former IZTF effluent was quite low in the concentration of ammonium but would be rich in nitrate when prosperous nitrifiers community had been built up in the IZTF. As a result, the nitrifiers in the latter MSL were starved of ammonium and would grow quite slowly and exhibit low level of nitrifying rate. This would be more probable during the start up of the apparatus, too, which could be proved by the data gained during the beginning of the second phase when the net NPR was very low (Fig. 4b).

Furthermore, the former IZTF effluent ammonium concentration overpassed the latter MSL effluent at the beginning of phase 2 (Fig. 4a), indicating the remarkable ammonium adsorption equilibrium effect of the zeolite filler. The MSL influent in phase 1 when raw wastewater was its influent was much higher in ammonium concentration than at the beginning of phase 2 when the MSL was moved under the IZTF, thus, the adsorped ammonium in MSL tended to get desorbed and went back to the water at the beginning of phase 2 due to the movement of the adsorption equilibrium (Sarioglu, 2005) and raised the effluent ammonium concentration.

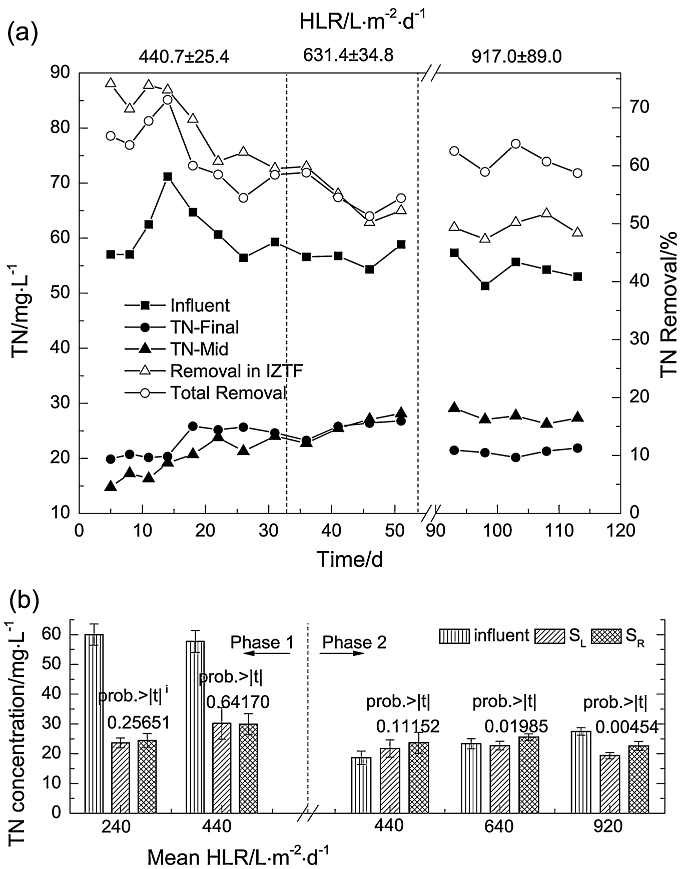


**Fig. 5.** TN removal results: (a) TN concentrations and the removal efficiency in phase 2; (b) TN concentrations of partial samples from the MSL in phase 1 and phase 2. Note: (i) the decimals above the bars were calculated probabilities of not to reject the null hypothesis according to the two-population $t$ test for the means of $S_L$ and $S_R$ where the null hypothesis was $\overline{S_L} - \overline{S_R} = 0$, and the alternative hypothesis was $\overline{S_L} - \overline{S_R} \neq 0$.

### 3.1.4. Denitrification and TN removal

$NO_3^-$, as a negative ion, was less readily adsorbed by zeolite or the soil colloidal particles compared to $NH_4^+$. Thus finding out effective ways to enhance the denitrification process for more nitrate removal was critical in promoting TN removal in infiltration systems (Kellogg et al., 2010; Leverenz et al., 2010; Pant and Adholeya, 2009). TN of water samples determined during the study is shown in Fig. 5a.

Although TN removal of mid IZTF continued decreasing with the influent nitrogen loading rate, total TN removal rate increased as the HLR increased to 920 L m$^{-2}$ d$^{-1}$. When influent wastewater loading rate was low, which meant low nitrogen and organic loading, the adsorbed ammonium tended to be nitrified into nitrate and drained very fast to the lower MSL system but could not be denitrified in time because of the lack of carbon sources, thus caused the effluent nitrate to increase significantly and the TN removal to be deteriorated. When influent loading rate increased to a higher level, as discussed in part 3.1.3, the nitrification rate stopped increasing as the gross nitrifiers were limited, but the denitrification rate would increase as the OLR increased especially in the lower MSL system, considering the extra ammonium from the upper ZTF would be adsorbed by the Zeolite permeable layer filters, the TN of the MSL effluent would decrease as a whole. That was consistent with the TN data observed in this study.

TN concentrations of partial samples from the MSL in phase 1 and phase 2 were shown in Fig. 5b. The result showed that in phase 2, TN concentration of the partial samples on the left ($S_L$) was significantly lower than that of the partial samples on the right ($S_R$) at the significance level ($\alpha$) of 0.05, and the differences tended to widen as the loading rate increased, indicating that the SMBs packed in the MSL significantly enhanced the denitrification of the nitrate since wastewater had to flow through one more SMB layer (compared with the $S_R$) before collected as the $S_L$. In fact, the denitrification effect was greater than displayed in the figure taking the adsorption feature of the zeolite filler into consideration. Data gained from phase 1 when $S_L$ and $S_R$ water all passed through only one SMB layer did not show significant difference ($\alpha = 0.05$), which also confirmed that conclusion.

However, the MSL as a whole did not perform well in denitrification as the maximum TN removal rate only reached 8 g N m$^{-2}$ d$^{-1}$ (including the ammonium adsorption) and the final effluent nitrate remained at relatively high concentration (about 20 mg L$^{-1}$ at medium OLR), which might be attributed to a scarcity of the carbon source, the coarse filter media that caused the HRT to be too short (Healy et al., 2006) and the competition from the aerobes since DO was not short (Rivett et al., 2008). So improvements can still be made to enhance the independent capability of the MSL in denitrification. It was reported that there were two factors that ensured the long-term denitrification: the continued supply of C to denitrifiers and the maintenance of adequate saturated hydraulic conductivity (Schipper et al., 2010). So, alternatives for more effective denitrification of the system may include: adding effective solid carbon sources into the SMBs, decreasing the particle size of the filter media in the MSL to hold more water in the SMBs (Sato et al.,

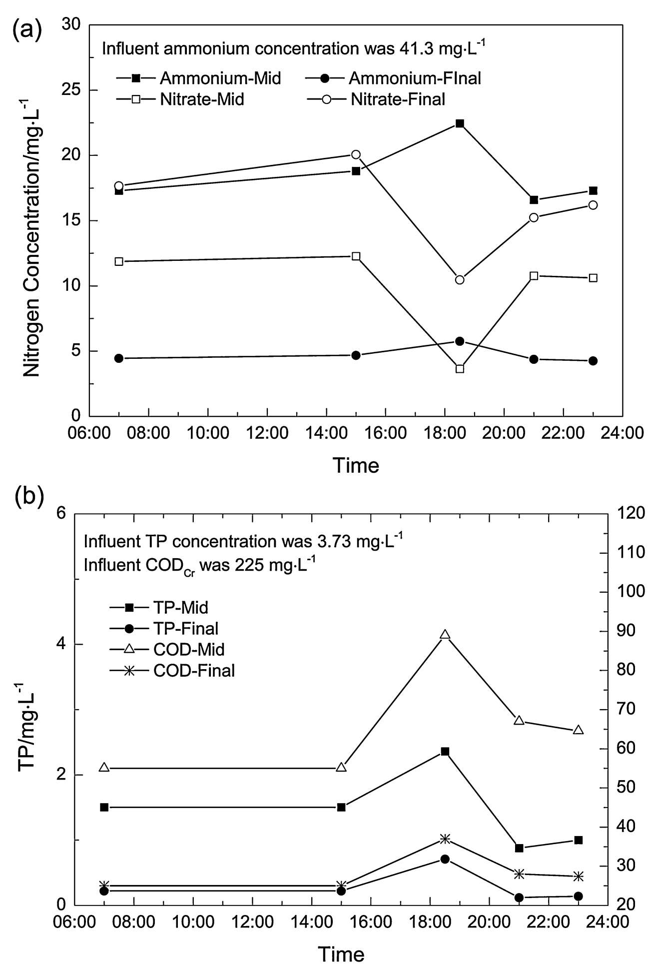


**Fig. 6.** The 24-h performance of the apparatus under shock loadings.

2005) and changing the final effluent style from draining mode into submerged or semi-submerged flow mode.

### 3.2. Effect of shock loadings and the treatment stability

Fig. 6 showed the performance of the apparatus in phase 3 under shock loading for 3 h in a 24-h cycle.

The results showed that the final MSL effluent TP and COD kept below 0.8 mg $L^{-1}$ and 40 mg $L^{-1}$, respectively, despite the sudden increase of the influent loading rate, though the mid IZTF effluent showed more remarkable increase. As for the ammonium, the result was the same except that the mid IZTF effluent ammonium concentration fluctuated much greater. As for the nitrate, the concentration showed a sudden drop when the HLR was adjusted to 1600 L $m^{-2}$ $d^{-1}$ but that went up again as the HLR was set back again to about 440 L $m^{-2}$ $d^{-1}$. The nitrate increased slowly as time went on and that was considered to be the combined effects of both the nitrification and denitrification in the biofilter.

It was found in phase 3 that the former IZTF effluent ammonium concentration nearly doubled that in phase 2 when the HLR were similar, which may because the adsorbed ammonium tended to get desorbed back into the effluent after the shock loading. But we were still confident in the two-stage apparatus of controlling the effluent ammonium concentration because the MSL as the second stage showed good performance in nitrification and ammonium adsorption. Before the loading rate increased, the ammonium concentration of the IZTF effluent kept low, leaving the zeolite in the MSL system abundant in adsorption sites. When the shock load occurred, the ammonium concentration of the IZTF effluent increased to some extend separately, but could be trapped in the lower MSL system, ensuring that the final effluent ammonium concentration be not too high. However, the TN removal rate was only about 50%, indicating that the denitrification rate was always limited in the apparatus.

Through out the study, no ponding, clogging or channeling happened during the operation of the apparatus. The distribution of wastewater to the MSL was much more homogeneous than directly pouring. The majority of the pollutants were consumed by the upper IZTF stage which was packed with coarse filler, which assured the steady performance of the lower MSL stage. The structure showed great stability in performance even when shock loadings occurred.

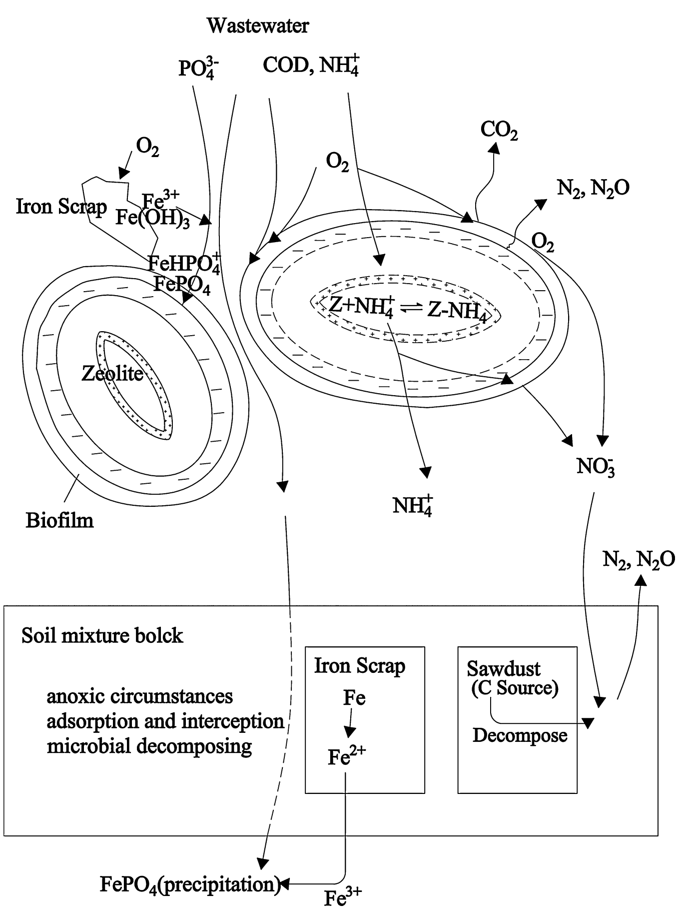


**Fig. 7.** Diagrammatic mechanism of the pollutant removal processes.

### 3.3. Schematic pollutant removal mechanisms

According to the results and discussion above, the supposed removal mechanism of various pollutants might be described as in Fig. 7 (Luanmanee et al., 2001).

## 4. Conclusions

A novel two-stage system consisting of a trickling filter packed with coarse zeolite and a multi-soil-layering bioreactor was developed aiming at decentralized wastewater treatment. The highlights of the results and conclusions include:

(1) Thin iron scraps layers paved in the trickling filter enhanced the TP removal remarkably due to the aerobic conditions, the partial high Fe:P ratio and the evener hydraulic conditions of the structure, and chemical precipitation was found to be the major reason for TP removal rather than the microorganism activities. The effluent TP kept lower than 0.5 mg $L^{-1}$ with a consistent average removal rate of 94.8%.
(2) Trickling filter packed with coarse zeolite and the intermittent wastewater feeding ensured that the oxygen supplement was not short, making the facility a good nitrifying reactor. The technology was librated from artificial aeration and the effluent ammonium kept at a low level of approximately 5 mg $L^{-1}$.
(3) The MSL did not show a satisfactory denitrification efficiency, which might be attributed to a scarcity of the carbon source and the coarse filter media that caused the HRT to be too short. Low loading rate often resulted in a significant increase in effluent nitrate due to the movement of the ammonium adsorption equilibrium and the robust nitrifying process. The best TN removal rate reached 61.5% with the corresponding mass removal rate of 30.8 g N $m^{-2}$ $d^{-1}$ under the HLR of approximately 920 L $m^{-2}$ $d^{-1}$.
(4) Coarse filter media in the first stage inhibits clogging of the latter infiltration stage, which reduced the management demand again and made the facility appeal to the decentralized sewage treatment. The two-stage structure ensured the stability of the final effluent quality even under hydraulic shock loadings.

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