

Article

Modelling Nutrients and Organics Removal by Biological Slow Filtration in Micro-Polluted Water Source Treatment

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Abstract: The biological slow filtration (BSF) system as a simple and efficient environmental technology has been widely applied in treatment of ‘micro-polluted’ water. At present, many related studies have focused on the removal efficiency of biological indicators (such as bacteria and viruses). However, there is less research on the removal performance of nutrients and organics in the BSF system. In this paper, we employed a lab-scale biological slow filter to study the removal efficiency and degradation mechanism of nutrients and organics. We proved through adsorption of filter layer at the early running stage and biodegradation at the later stage, the BSF system could achieve effective removal of $\text{NH}_3\text{-N}$, TN, TP, COD_{Mn} and turbidity and the corresponding removal rates are 83.65%, 42.45%, 42.94%, 60.41% and 83.55%, respectively. Furthermore, we also explored the influence of four main factors (filtration rate, filter depth, hydraulic head and temperature) and their interactions on removal rates of nutrients and organics in the BSF system and obtained the optimal operating parameters as follows: filtration rate 0.1 m/h, filter depth 0.8 m, hydraulic head 0.64 m, temperature 26.06 °C. This study would provide a theoretical foundation for the actual application of biological slow filter in treatment of micro-polluted water in developing countries and offer an optimized basis for the design of operating conditions.

Keywords: biological slow filter; nutrients; organics; degradation mechanism; influencing factors

1. Introduction

In recent years, with the rapid development of industry and the acceleration of urbanization, the surface water pollution has become increasingly serious around the world [1] and generally presents a state of ‘micro-pollution’. Since the micro-polluted surface water has the characteristic of high ammonia nitrogen ($\text{NH}_3\text{-N}$) and high chemical oxygen demand (COD), traditional physical and chemical treatments (e.g., coagulation, precipitation, filtration and disinfection) in water works could not achieve effective removal of nutrients and organic compounds in micro-polluted water resources. As a result, there will be a major threat to drinking water safety for humans. Considering the above factors, the technology of biological pretreatment or advanced treatment will be necessary in order to enhance effluent quality in the drinking water treatment process.

Compared to the advanced treatment process (such as Ozonation, Membrane separation and Activated carbon adsorption), biological pretreatment technology has the advantages of lower maintenance costs and greater pollutant elimination in water source and thus has attracted wide attention. Biofilter, as an effective technology for biological pretreatment, has been widely studied and applied. Among all kinds of biofilter processes, the biological slow filtration (BSF) process is one of the most common and effective technique and has been used for the production of clean drinking water

for nearly 200 years [2]. During the operation of the biological slow filter, the raw water flows down through the filter media layer and the supporting layer at a constant low filtration rate, and then flows out by a pipeline at the bottom of the filter tank. As the pollutants in raw water trap and accumulate in the filter layer, microorganisms in the biological slow filter absorb nutrients and continuously grow, eventually to form a stable mature biofilm attached on the inner wall of the reactor and the filter column [3]. Therefore, the pollutant removal mechanisms of biological slow filter are physical adsorption of filter column and biological flocculation and degradation of the biofilm [4]. At present, many studies have proved that the BSF process could be used in the treatment of micro-polluted water and in practice, the advantages of the BSF process in comparison to other drinking water treatment techniques are its simplicity, low cost of construction and maintenance, eco-friendly treating and high removal efficiency of pollutants. Consequently, over 650,000 locally constructed BSF units were in operation worldwide by 2014 [5].

Some researchers have confirmed that biological slow filter as a drinking water treatment process could achieve effective removal of bacteria, viruses, turbidity and other biological indicators in surface water and consequently improve the safety of drinking water [6–8]. To date, there have also been some studies focused on the removal performance of nutrients (such as nitrogen and phosphorus) by the BSF process and some achievements have been made. Heather et al. [9] have revealed that simultaneous nitrification and denitrification (SND) is occurring in the biological slow filter by monitoring the transformation of nitrogen in different forms. Sukru has also confirmed that the BSF system could remove nitrates-nitrogen ($\text{NO}_3\text{-N}$) in surface water through denitrification and the removal efficiency was related to the carbon to $\text{NO}_3\text{-N}$ (C/N) ratio [10]. However, little research has explored phosphorus removal performance in the BSF system. In addition, some studies have shown that the BSF system could achieve 50–67% COD removal and 79–92% BOD_5 removal for the treatment of secondary effluents [11]. Due to the form, concentration and composition of organic matter (e.g., COD) in surface water are different compared to secondary effluent in sewage plant and investigation on the removal performance of organic compounds in surface water by the BSF system was scant. Therefore, research on the removal efficiency of phosphorus and organics in micro-polluted surface water by the BSF system should be strengthened in the future. It is worth noting that the removal performance of target pollutants in the BSF process is not only related to the concentration of influent loads, but also affected by the design parameters and operating conditions. Among them, filter depth, filter material size, hydraulic head, filtration rates, temperature and operating mode have been widely explored [6,12,13], but related studies were focused on how single factor affected pollutants removal efficiency of the BSF system and there was a lack of research on the interactions of major influencing factors. The next step should further strengthen research on how main impact factors and their interaction affect pollutants removal efficiency of the BSF system. As we know, the BSF system achieves various pollutants removal through adsorption of filter column and microbial degradation. Campos [14] and Baghat [15] have demonstrated that microorganisms abundant in the BSF system could realize biological degradation of specific pollutants by exploring the biomass distribution characteristics of the biological slow filter. However, the related research was lacking and knowledge about the degradation kinetics of filter were little known. In the future, research will be focused on the pollutants degradation mechanisms in the BSF process and the degradation kinetics of the reactor in order to establish the pollutants degradation model and further reveal the transformation mechanism of different pollutants in the reactor [16–18].

Overall, research on the BSF system used in drinking water treatment has made some significant achievements. However, most of the previous studies focused on the removal efficiency of nitrogen and biological indicators (such as bacteria and viruses); therefore, the removal performance of phosphorus and organics in the BSF system and the degradation mechanisms of the above pollutants need to be further explored. In this paper, we conducted a lab-scale experiment to assess the removal efficiency of conventional pollutants (such as nitrogen, phosphorus, organic compound and turbidity) in a micro-polluted water source through a biological slow filter (as a drinking water treatment technology). The first objective was to evaluate the pollutants removal performance of the BSF process at different

operational conditions (e.g., filter depth, filtration rate, hydraulic head and temperature) and explore the optimal operating parameters. The second objective was to investigate the degradation kinetics of various pollutants in the BSF system by the batch experiment and model fitting and further reveal the mechanisms of pollutants removal.

2. Materials and Methods

2.1. Biological Slow Filter Design

A laboratory scale biological slow filter was designed to conduct the experiments for this study. The filter was constructed from a transparent, 350 mm diameter Perspex tube and had a total height of 1.8 m. At the bottom of the filter, a base was installed in order to reinforce the construction. The details of the biological slow filter are presented in Figure 1. The construction of a biological slow filter consists of the following five parts: (1) influent area: raw water in the feed tank was pumped to the top of the filter at a constant filtration rate (0.1 m/h) and the corresponding pump speed was 60 r/min; (2) overflow pipe: the overflow pipe was placed 5 cm below the top of the filter to control the water level and avoid the overflow at the top; (3) silica sand filter layer: the filter was filled with silica sand (with an effective diameter of 0.4–0.6 mm and a uniformity coefficient (UC) of 1.6) 0.8 m in height. This silica sand column would achieve the removal of pollutants in raw water. On the outside of the reactor, four sample sites were vertically distributed with a distance to the surface of silica sand layer of 0, 0.3, 0.5, 0.7 m, respectively in order to study the pollutants removal and microbial distribution in each filter layer; (4) supporting layer: two gravel layers were filled below the silica sand layer as supporting layer (0.3 m in height) with a gravel size of 8–16 mm and 16–30 mm, respectively. The function of this layer was to support the silica sand column and avoid the loss of filter material; (5) effluent pipe: the effluent tube (1.2 m in height, diameter was 20 mm) was made of polyvinyl chloride (PVC) and was extended from the bottom of the filter to well above the surface of the silica sand layer to prevent negative pressure in the filter bed.

The operation of biological slow filter was indoor and operating temperature was kept at about 25 °C to avoid the influence of temperature changes on the growth of microorganisms in the reactor.

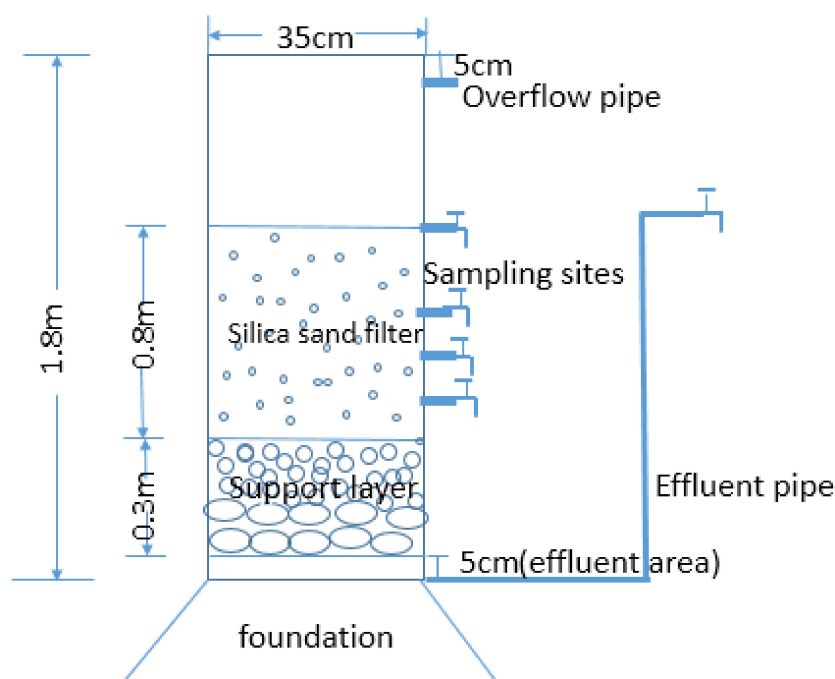


Figure 1. Design of biological slow filter.

2.2. Raw Water Characteristics

Raw water quality was designed to roughly simulate a typical micro-polluted surface water in a developing country. In this study, we employed glucose (4–6.67 g), ammonium chloride (1.528–2.292 g) and potassium dihydrogen phosphate (0.1756–0.3512 g) to simulate the concentration of organic compounds and nutrients in micro-polluted surface water. At the same time, we also added 400 mL nutrient solution of trace elements to raw water tank in order to supplement the nutritional requirements of microbial growth in filter. The municipal tap water (which we used in preparation of raw water) must be aerated with compressed air and stored at laboratory temperature for more than 16 h to remove the residual chlorine before being used. Overall, we employed 95 percent of the artificial water and mixed with 5 percent of natural Taihu Lake surface water (in Jiangsu province, china) as our experimental required raw water. Natural lake water was added to replenish required microorganisms for the filter and make filter operating closer to the natural conditions. In conclusion, we prepared 400 L raw water as our experimental influent water. In the raw water tank, we could adjust pH through hydrochloric acid or sodium bicarbonate and increase the dissolved oxygen (DO) by aeration. The characteristics of raw water are presented in Table 1.

Table 1. Raw water characteristics.

Parameter	Range	Average
Temperature(°C)	20–27	24
pH	7.4–8.13	7.82
DO (mg/L)	5.5–7.5	6.5
Turbidity (NTU)	2–7	4.5
Ammonia nitrogen (mg/L)	1–1.5	1.2
Total nitrogen (mg/L)	2–6	4
Total phosphorous (mg/L)	0.1–0.2	0.15
COD _{Mn} (mg/L)	6–10	8
Trace elements	-	-

Nutrient solution of trace elements (1 L) includes of 1 g FeSO₄·7H₂O, 0.12 g ZnSO₄·7H₂O, 0.12 g MnSO₄·H₂O, 0.03 g CuSO₄·5H₂O, 0.15 g NaB₄O₇·10H₂O, 0.06 g NaMoO₄·2H₂O, 0.18 g KI, 2.5g EDTA.

2.3. Experimental Design and Modeling

2.3.1. Factorial Experiment Design of Biological Slow Filter

In order to explore the effect of operating parameters on the pollutants removal performance of the BSF system, we selected four factors that have the greatest influence on the operation performance of the biological slow filter and designed a four-factor three-block experiment by employing response surface methodology with a Box-Behnken design in Design Expert software. Four independent variables were filtration rate, filter depth, hydraulic head and temperature respectively and the response parameters were removal efficiency of turbidity, ammonia nitrogen (NH₃-N), total nitrogen (TN), total phosphorus (TP) and permanganate index (COD_{Mn}) respectively. The details of factorial experiment design are presented in Table 2. We followed the different experimental groups which were formed by Design Expert software to conduct the experiments, through the methods of mathematical statistics and regression model, the parameters which have appreciable impact on removal performance of various pollutants in BSF system were analyzed and interactions of the above factors were also explored. In terms of the interactions of some factors with significant ($p < 0.05$), response surface plot and the corresponding contour plot could be obtained, in order to analyze the mechanism of interactions. Finally, the optimal operating parameters were obtained through optimization of the Design Expert software, which will provide reference value for the design of BSF system in practice.

Table 2. The factorial experiment design.

Parameters	Level 1	Level 2	Level 3
Filtration rate (m/h)	0.1	0.35	0.6
Filter depth (m)	0.3	0.55	0.8
Hydraulic head (m)	0.2	0.5	0.8
Temperature (°C)	8	16.5	25

Annotation: (1) adjust the filtration rate through the pump; (2) removing or adding the filter layer to change the filter depth; (3) when the rate of inflow was constant, the hydraulic head would be controlled by adjusting the outlet valve; (4) operating temperature could be controlled by indoor air conditioning.

2.3.2. Investigation on the Pollutants Removal Mechanisms by Batch Experiment and Modeling

In order to further explore the removal pathways of pollutants in the biological slow filter and to reveal their degradation mechanisms, we conducted the batch experiment; the specific design was as follows. At first, we took a certain amount of biofilm-attached silica sand filtering medium from different filter layers in a stable operating BSF system and mixed together (as A group). Meanwhile, we took the same amount of unused silica sand and washed it with deionized water (as B group). Then, we took six 500 mL conical flasks and added the same weight of silica sand in each container. Next, we poured the aerated raw water (as shown in Table 1) into the above containers and covered with gauze, respectively (each experimental group had two control groups). We put the above six conical flasks into Gas bath thermostatic oscillator and controlled the operating conditions (25 °C, 100 rpm). It is important to keep away from light to avoid the photodegradation of pollutants in raw water while the instrument is running. During the operating period of batch experiment, we took supernatant (50 mL) from six containers at 10 min, 30 min, 1.5 h, 3 h, 6 h, 12 h, 15 h, 24 h and 36 h, respectively and detected the removal rate of $\text{NH}_3\text{-N}$, TN, TP and COD_{Mn} at different times.

According to the monitoring data, we could make the curves between the quantity of adsorption and operating time (Q_t -t) or between pollutant concentrations and operating time (c-t). Then we used the common kinetics models of adsorption and biodegradation (as depicted in Table 3) to fit in order to reveal the pollutants removal mechanisms in BSF system.

Table 3. The kinetics models of adsorption and biodegradation. (a). The kinetics models of adsorption; (b). The kinetics models of biodegradation.

(a)	The Kinetics Model of Adsorption	Description of Equation
	“Lagergren” model	$Q_t = Q_e(1 - e^{-kt})$
	Corrected “Elovich” model	$Q_t = a + b \ln t$
	Power function model	$Q_t = a \times t^b$
	Parabolic diffusion model	$Q_t = a + kt^{0.5}$
(b)	The Kinetics Model of Biodegradation	Description of Equation
	Zero-order kinetics model	$C_t = C_0 - kt$
	First-order kinetics model	$C_t = C_0 \times e^{-kt}$
	Second-order kinetics model	$C_t = C_0 / (1 + C_0 kt)$

Descriptions of parameters: (1) Q_t : the quantity of adsorption at time t; (2) Q_e : the quantity of equilibrated adsorption; (3) t: operating time; (4) C_t : the concentration of pollutant at time t; (5) C_0 : the initial concentration of pollutant; (6) k, a, b were constants.

2.4. Parameter Measurements and Analytical Methods

In this study, we routinely monitored nutrients (such as total nitrogen (TN), ammonia nitrogen ($\text{NH}_3\text{-N}$), nitrites-nitrogen ($\text{NO}_2\text{-N}$), nitrates-nitrogen ($\text{NO}_3\text{-N}$), total phosphorus (TP)) by using UV Spectrophotometer (Spectrumlab 752s, Lengguang Technology, Shanghai, China) and organic compounds (e.g., COD_{Mn}) by using acid burette (50 mL) in influent and effluent of BSF system according to the procedures set forth in Standard Methods for the Examination of Water and

Wastewater [19]. We took samples (about 150 mL) every other day to analyze various pollutants removal efficiency of BSF system. At the same time, we also monitored the operating parameters (such as pH, DO and turbidity) in influent and effluent. The pH value was tested using acidimeter (Rex PHB-4, Shanghai INESA Scientific Instrument Company, Shanghai, China) and the concentration of DO was measured online with dissolved oxygen instrument (Rex JPB-607A, Shanghai INESA Scientific Instrument Company, Shanghai, China). For turbidity, we used a turbidimeter (Rex WGZ-2000, Shanghai INESA Scientific Instrument Company, Shanghai, China) to measure this parameter in order to reflect the distribution of particles in water samples of influent and effluent.

It is worth noting that the data would be processed using the software of EXCEL (EXCEL2013, Microsoft Office, Washington, USA) and the results were presented in the form of graphs by using ORIGIN software (ORIGIN2015, Origin Lab Company, Guangzhou, China). For batch experiment, we adopted the common adsorption and biodegradation kinetics model in biochemical filed to fit the results and obtained the optimal fitting model, then drawing and analyzing kinetic parameters by using the software of MATLAB (MATLAB2012, Math Works Company, Natick, MA, USA) in order to reveal the pollutants removal mechanisms of the BSF system.

3. Results and Discussions

3.1. Pollutants Removal Efficiencies of BSF System

3.1.1. Start-Up Operation of BSF System

In this study, we adopted the way of natural hanging membrane [20] to achieve the start-up operation of biological slow filter under specific conditions (filtration rate was 0.1 m/h and operating temperature was about 25 °C). In the start-up phase, we took samples every other day to analyze the changes in pollutants concentrations in influent and effluent and observed the formation of biofilm on the surface of filter layer at the same time. After about 33 days, we could observe a tawny schmutzdecke on the surface of silica sand column and the inner wall of the reactor. Furthermore, the removal rates of various pollutants were stable, which indicated that the start-up period of the BSF system was accomplished.

The removal performance of various pollutants in the BSF system was depicted in Figure 2. As shown in Figure 2, the change trend of different pollutants removal rates had obvious differences. In this study, the influent turbidity ranged from 4 to 6 NTU. With running time increasing, the removal rate of turbidity showed a trend of fluctuated rise. After 27 days of operating, removal efficiency of turbidity tended to be stable (about 83.55%) and effluent turbidity always remained below 1 NTU. The existing research has shown that turbidity is mainly removed by adsorption and retention of filter layer in the BSF system and 89% turbidity removal on average [6]. This study also confirmed that the BSF system could effectively remove turbidity in micro-polluted raw water and removal efficiency of turbidity was about 83.55%.

BSF system could also achieve the effective removal of ammonia nitrogen ($\text{NH}_3\text{-N}$), in this study, we selected the concentration of influent $\text{NH}_3\text{-N}$ was 1.2–1.5 mg/L and monitored the removal rate of $\text{NH}_3\text{-N}$ at different times. According to the results, we found that the removal rate of $\text{NH}_3\text{-N}$ remained at about 80% in the first 11 days, but at the 13th day, $\text{NH}_3\text{-N}$ removal efficiency decreased to 46.62%. After that, the removal rate of $\text{NH}_3\text{-N}$ showed a trend of fluctuated rise (as described in Figure 2). This phenomenon may be explained by the fact that the removal of $\text{NH}_3\text{-N}$ in the BSF system depended on the adsorption of the filter layer in the initial stage and because of the adsorption saturation of the filter column and the continuous growth of microorganisms in the BSF system, the removal of $\text{NH}_3\text{-N}$ was mainly accomplished by biodegradation from the 14th day. Finally, after 23 days of operation, the change of $\text{NH}_3\text{-N}$ removal rate tended to be stable and the average removal efficiency was about 83.65%. Biodegradation of $\text{NH}_3\text{-N}$ in the BSF system mainly depended on nitrification and denitrification; both Kennedy [21] and Nakhla [13] have confirmed the process of simultaneous nitrification-denitrification (SND) in the biological slow filter. During the start-up period of reactor,

we monitored the concentration of different forms of nitrogen in influent and effluent and detected that in the first 11 days of operation, the accumulation of nitrites-nitrogen ($\text{NO}_2\text{-N}$) in the BSF system gradually increased (characterized as the concentration of $\text{NO}_2\text{-N}$ in effluent water being higher than that in influent water) and then $\text{NO}_2\text{-N}$ was further oxidized to nitrates-nitrogen ($\text{NO}_3\text{-N}$) through nitrification, the amount of $\text{NO}_3\text{-N}$ in BSF system was gradually increasing. However, from the 23rd day, the accumulation of $\text{NO}_3\text{-N}$ decreased gradually and began to be negative, combined with the change trend of total nitrogen (TN) removal rate, we found that the denitrification rate tended to be stable when the BSF system operated for 33 days and the corresponding removal rate of TN was about 42.45% on average. The transformation of nitrogen in different forms and removal efficiency of TN are shown in Figure 3.

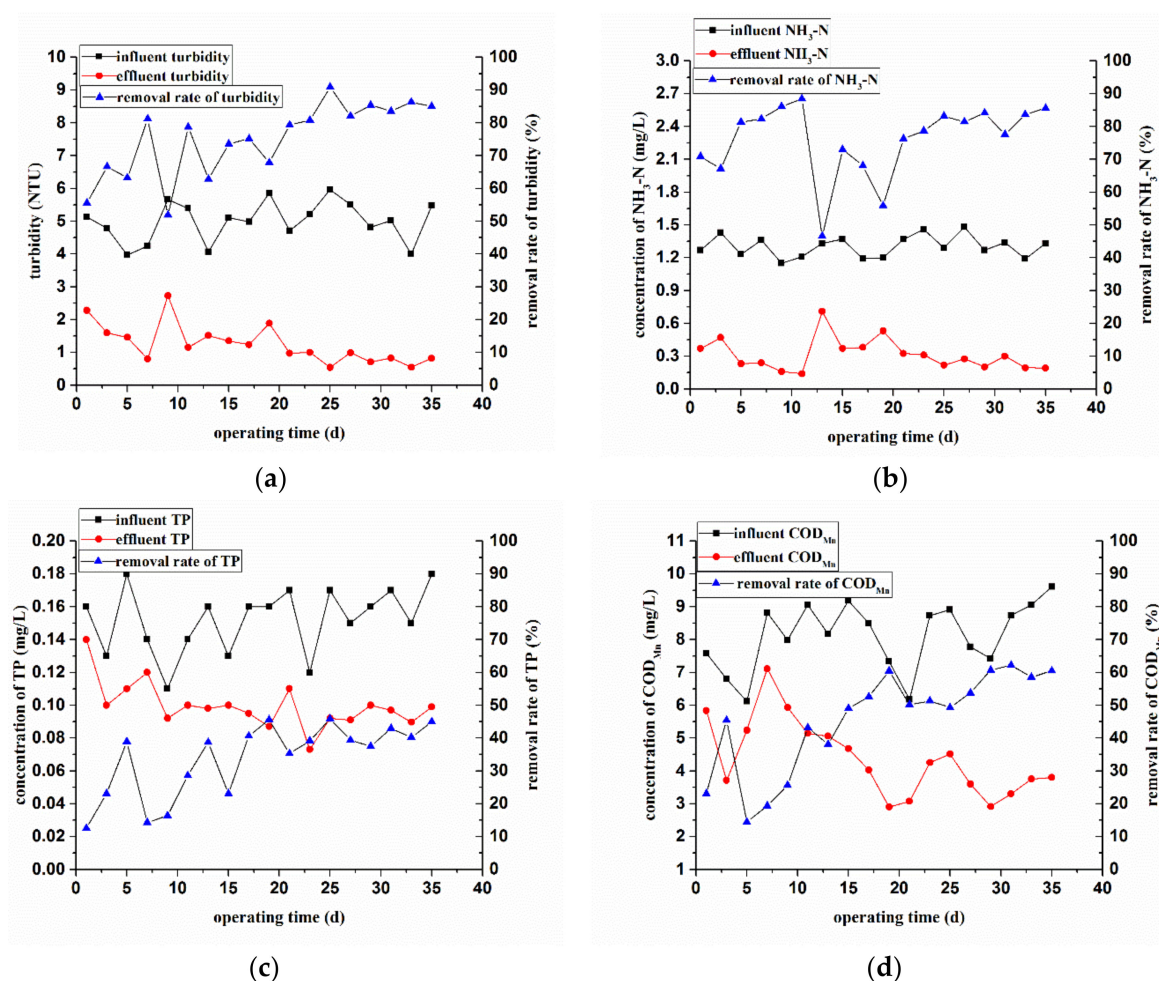


Figure 2. Removal efficiency of various pollutants in biological slow filtration system ((a) turbidity, (b) $\text{NH}_3\text{-N}$, (c) TP, (d) COD_{Mn}).

With the aggravation of water eutrophication, the amount of phosphorus in the micro-polluted water source has exceeded the standard of drinking water and its influence cannot be ignored. At present, there have been few studies on the removal performance of total phosphorus (TP) in micro-polluted raw water by the BSF system. In this study, we chose the concentration of influent TP to be 0.11–0.18 mg/L and researched the removal efficiency of TP in biological slow filter (as described in Figure 2). As can be seen from Figure 2, the change trend of TP removal rate was similar to that of $\text{NH}_3\text{-N}$. In the first five days, the removal rate of TP increased rapidly to 38.89% due to the adsorption of filter layer, but at the 7th day, TP removal efficiency decreased sharply to 14.29%. After that, the removal rate of TP presented a trend of fluctuated rise because of biodegradation. Finally, after 31 days

of operation, the change of TP removal rate tended to be stable and the average removal efficiency was about 42.94%. The amount of organic matter in micro-polluted water source also cannot be ignored. Once the concentration of organics exceeds the standard of drinking water, it will be harmful to human drinking water. In this study, we investigated the removal efficiency of organics (expressed as COD_{Mn}) in the BSF system. As shown in Figure 2, because of the adsorption of the filter layer, the BSF system could achieve 45.44% COD_{Mn} removal in the first three days and then the removal rate of COD_{Mn} decreased sharply (down to 14.38%) in the next two days. After that, the removal rate of COD_{Mn} presented a trend of fluctuated rise and COD_{Mn} removal mainly depended on biodegradation. After 19 days of operation, the removal rate of COD_{Mn} gradually stabilized and the value was about 60.41% on average. Farooq and Al-Yousef have demonstrated that BSF system achieved 50–67% COD removal for the treatment of secondary effluents [11]. That is to say, the BSF system could effectively remove organics in water, either secondary effluent or micro-polluted surface water.

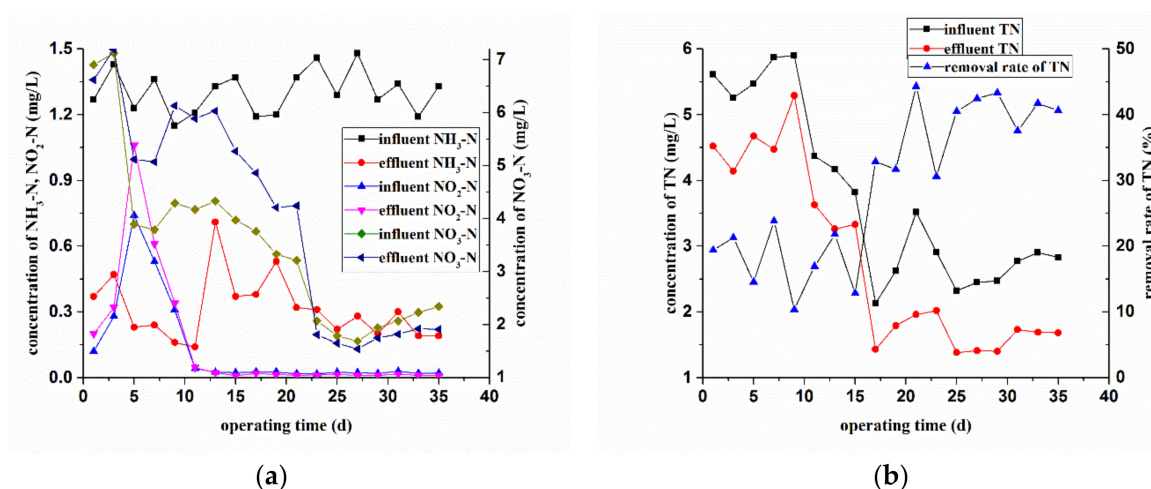


Figure 3. Transformation of nitrogen in different forms (a) and TN (total nitrogen) removal performance (b).

3.1.2. The Stable Operation of BSF System

After 33 days of start-up operation, a tawny biofilm was formed on the surface of the filter layer and the inner wall of the biological slow filter and the removal rates of various pollutants were stable fluctuated which indicated BSF system was entering the stable operating stage. The removal rates of turbidity, $\text{NH}_3\text{-N}$, TN, TP and COD_{Mn} were 83.55%, 83.65%, 42.45%, 42.94% and 60.41%, respectively.

3.2. Investigation on the Main Influencing Factors and Optimization of BSF System

In this study, we selected four parameters (including filtration rate, filter depth, hydraulic head and temperature) and designed a four-factor three-block experiment (the factorial experiment design is shown in Table 2) by employing response surface methodology with a Box-Behnken design in order to explore the influence of a single factor and its interactions on removal performance of various pollutants (including $\text{NH}_3\text{-N}$, TN, TP, COD_{Mn} and turbidity) in the BSF system. We followed the different experimental groups which were formed by Design Expert software to conduct the experiments and the specific experiment is presented in Table 4.

Table 4. A Box-Behnken experiment design (BBD) with Response surface methodology.

Run	Variables				Response				
	Filtration Rate (m/h)	Filter Depth (m)	Hydraulic Head (m)	Temperature (°C)	TN Removal Rate (%)	NH ₃ -N Removal Rate (%)	TP Removal Rate (%)	COD _{Mn} Removal Rate (%)	Turbidity Removal Rate (%)
1	0.35	0.3	0.5	8	15.66	28.51	5.17	36.12	55.17
2	0.35	0.55	0.5	16.5	35.35	72.4	50.7	48.67	74.03
3	0.1	0.3	0.5	16.5	22.8	60.46	27.4	40.17	64
4	0.35	0.8	0.8	16.5	40.45	80.3	38.5	44.43	70.81
5	0.6	0.55	0.5	25	15.95	52.4	8.33	37.37	53.48
6	0.35	0.8	0.5	8	28.61	31.43	15.01	49.04	74.82
7	0.6	0.55	0.5	8	13.16	29	13.2	23.14	46.11
8	0.35	0.55	0.5	16.5	34.62	65.41	51.03	49.37	68.21
9	0.35	0.55	0.8	8	17.42	47.84	27.36	40.01	73.11
10	0.35	0.3	0.5	25	18.12	30.12	6.88	45.14	57.48
11	0.35	0.55	0.5	16.5	39.17	75.87	24.9	42.89	79.14
12	0.35	0.55	0.8	25	31.75	72.13	33.01	48.14	75.61
13	0.35	0.8	0.5	25	38.95	79.5	36.3	53.24	80.26
14	0.35	0.3	0.2	16.5	14.71	30.4	6.1	41.67	56.66
15	0.6	0.8	0.5	16.5	17.68	56.78	27.31	41.16	64.57
16	0.1	0.55	0.5	8	29.65	67.9	34.07	29.01	69.21
17	0.35	0.55	0.5	16.5	35.35	72.38	24.9	45.55	73.98
18	0.35	0.55	0.5	16.5	36.24	73.81	25.12	45.09	77.12
19	0.6	0.55	0.2	16.5	11.37	50.64	7.28	46.53	47.17
20	0.35	0.3	0.8	16.5	10.03	28.4	4.01	38.21	60.11
21	0.6	0.3	0.5	16.5	1.02	22.89	3.01	29.24	40.07
22	0.35	0.55	0.2	25	30.56	68.8	53.8	50.61	68.18
23	0.1	0.8	0.5	16.5	42.41	85.3	45	60.45	85.71
24	0.1	0.55	0.2	16.5	27.96	65.89	35.19	52.14	72.31
25	0.6	0.55	0.8	16.5	11.79	51.1	4.87	34.12	40.08
26	0.1	0.55	0.8	16.5	36.21	74.61	38.01	50.82	70.14
27	0.1	0.55	0.5	25	38.55	77.65	35.43	58.31	78.29
28	0.35	0.8	0.2	16.5	22.52	58.91	36.31	49.97	76.21
29	0.35	0.55	0.2	8	23.41	49.34	27.61	47.04	71.14

After the different experimental groups were completed and the corresponding response values were obtained, through the methods of mathematical statistics and regression analysis, the parameters which have appreciable impact on removal performance of various pollutants in the BSF system were analyzed and interactions of the above factors were also explored. Table S1 (in the Supplementary Materials) reflects the influence of various factors and their interactions on the removal rate of different pollutants. The significance of each factor and interactions between them were checked with the help of a Fisher test (if ' $p > F$ ' value is below 0.05, then the parameter is significant). As illustrated in Table S1, three parameters (filtration rate, filter depth and temperature) all had significant influence on the removal rate of NH₃-N, TN and COD_{Mn} in BSF system. Among them, interactions between filter depth and temperature had significant influence on NH₃-N removal and between filter depth and hydraulic head had significant influence on TN removal. For the removal rate of TP and turbidity, two factors (including filtration rate and filter depth) had significant influence and there was no interaction with significant effect. At present, there is little research focused on the influence of operating parameters on the removal performance of nutrients (nitrogen and phosphorus) and organics (COD) in BSF system [13,22,23]. Among them, Nakhla has studied how filter depth and filtration rate affect nitrogen removal and demonstrated that nitrification efficiency and TN removal efficiency correlated well with filtration rate [13] and its conclusion was consistent with this study. Liu et al. [22] found that filter depth had an influence on the removal rate of COD_{Mn}, NH₃-N and turbidity in micro-polluted water and put forward the BSF system must be filled with 0.5 m filter layer at least to ensure the removal efficiency of various pollutants. In this study, we also confirmed that filter depth had a significant influence on the removal rate of nitrogen, phosphorus, turbidity and organics, with the filter depth increasing, all pollutants removal rates improved accordingly [24]. In addition, we found that with the filtration rate (or hydraulic head) increasing, the removal rate of each pollutant decreased correspondingly. Temperature is correlated well with pollutant removal performance in bioreactor

by affecting the activity of microorganisms [25]. In this research, 25 °C was suitable for the above pollutants effective removal, except turbidity.

We selected parameters and their interactions that have significant influence on the removal rate of various pollutants to fit the regression model (a second-order polynomial model) and the specific fitting details are shown in Table 5.

Table 5. Regression model of various pollutants removal rate.

Responses	Unit	Regression Equation	R ²	C.V%
NH ₃ -N removal rate	%	$69.81 - 14.08A + 15.95B + 2.53C + 10.55D + 11.62BD - 15.41B^2 - 4.74C^2 - 10.21D^2$	0.9180	11.32
TN removal rate	%	$36.15 - 10.55A + 9.02B + 1.43C + 3.83D + 5.65BC - 7.87A^2 - 7.32B^2 - 6.66C^2 - 3.71D^2$	0.9612	10.51
TP removal rate	%	$29.11 - 12.59A + 12.15B - 8.19B^2$	0.6116	39.91
COD _{Mn} removal rate	%	$44.06 - 6.61A + 5.64B + 5.70D$	0.6401	12.26
Turbidity removal rate	%	$71.70 - 12.35A + 9.91B - 0.15C - 9.65A^2 - 3.35C^2$	0.8793	7.07

Annotation: A = Filtration rate, B = Filter depth, C = Hydraulic head, D = Temperature.

Therefore, we could evaluate the corresponding pollutant removal efficiency according to the regression model of each pollutant removal rate when the operating parameters changed.

Because the interactions between filter depth and temperature had significant influence on NH₃-N removal performance and between filter depth and hydraulic head also had significant influence on removal efficiency of TN, we drew the surface response plots and corresponding contour plots (Figure 4) to help understand the nature of the response at different factor levels [26]. As can be seen from Figure 4, with the filter depth and operating temperature increasing at the same time, NH₃-N removal efficiency was significantly improved. The reason might be explained as follows: physical adsorption ability of filter layer would be improved with the filter depth increasing, so a certain amount of NH₃-N could achieve effective removal through adsorption and interception of filter column. In addition, a suitable operating temperature was beneficial to the improvement of microbial activity (about 25 °C) and thus was good for NH₃-N removal by biodegradation. Due to the increase of hydraulic head, the total nitrogen load in BSF system enhanced accordingly. Therefore, it would not be conducive to the TN removal when the hydraulic head raised. However, as the filter depth increased, the amount of TN through adsorption and retention of filter column improved correspondingly. As a consequence, the increase of filter depth could make up for the influence of hydraulic head augment on TN removal rate to a certain extent and thus ensure the stability of TN removal efficiency.

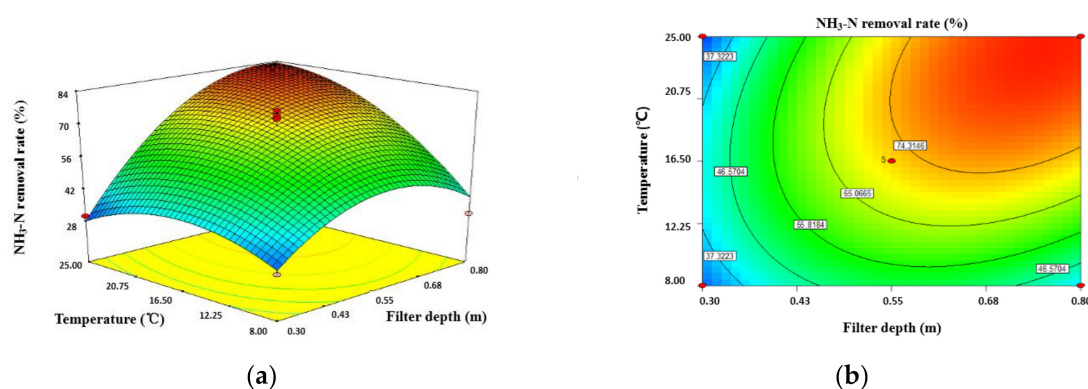


Figure 4. Cont.

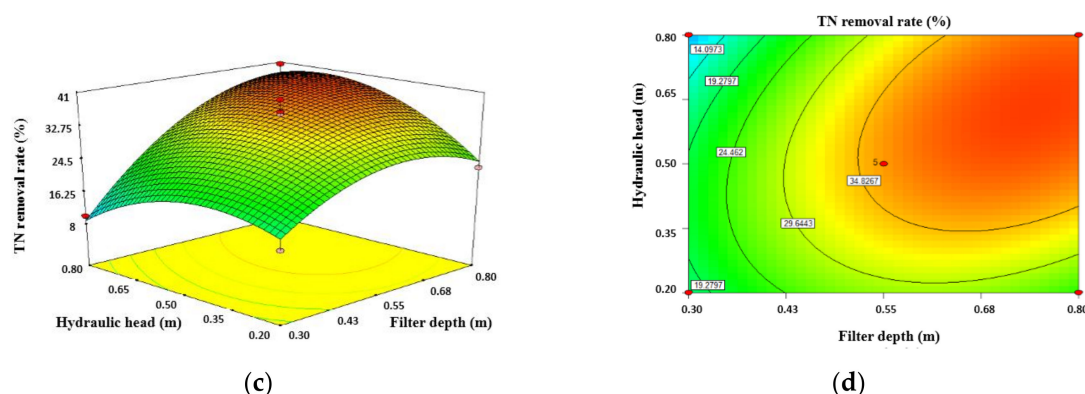


Figure 4. The surface response plots and corresponding contour plots for (a,b) $\text{NH}_3\text{-N}$ removal rate and (c,d) TN removal rate.

Response surface could be analyzed to obtain the maximum or minimum responses and the corresponding optimum conditions. In this study, the best solution of operating parameters is given in Table 6. Optimum operating conditions could provide a reference value for the BSF system design in practice.

Table 6. Optimum operating parameters of BSF system.

Variables	Filtration rate (m/h)	0.1
	Filter depth (m)	0.8
	Hydraulic head (m)	0.64
	Temperature ($^{\circ}\text{C}$)	26.06
Response	TN removal rate (%)	48.49
	$\text{NH}_3\text{-N}$ removal rate (%)	92.04
	TP removal rate (%)	45.60
	COD_{Mn} removal rate (%)	63.38
	Turbidity removal rate (%)	82.95

3.3. Study on Pollutants Removal Mechanisms and Model Fitting

In this study, we conducted the batch experiment and selected typical adsorption and biodegradation kinetic model to fit in order to reveal the removal mechanisms of nutrients and organics in the BSF system. The change trend of each pollutant adsorption amount (Q_t) or concentration with time increasing in batch experiment is shown in Figure 5. As can be found in Q_t -t chart of each pollutant, in the initial 3 h, the adsorption amount of $\text{NH}_3\text{-N}$, TP and COD_{Mn} of filter material increased rapidly, and the maximum adsorption ratio of each pollutant reached 90%, 86.45% and 77.46% respectively. After that, the adsorption rate of above pollutants slowed down gradually and finally reached the dynamic equilibrium of adsorption and the corresponding amount of equilibrium adsorption was 0.013, 0.00239 and 0.0352 mg/g respectively. The common adsorption kinetics model was chosen to fit the adsorption process of the above pollutants in the BSF system, and the details are shown in Tables 7 and 8.

According to the change trend of each pollutant concentration over time, we could discover that concentration of $\text{NH}_3\text{-N}$, TP and COD_{Mn} reduced gradually in the first 3 h due to the adsorption of filter material. After that, $\text{NH}_3\text{-N}$ concentration was still decreasing and the reduction rate did not slow down and tend to be balanced, which demonstrated that biodegradation had gradually taken advantage and realized further removal of $\text{NH}_3\text{-N}$ after 3 h. TP and COD_{Mn} concentration were also gradually reducing like the change trend of $\text{NH}_3\text{-N}$ concentration and the reason might be inferred that biodegradation of polyphosphate accumulating bacteria (PAOs) and aerobic heterotrophic bacteria could achieve further removal of TP and COD_{Mn} . When the batch experiment was running for about

15 h, the decreasing trend of TP and COD_{Mn} concentration was slowing down; this phenomenon might be explained by the fact that DO concentration in experimental containers was continually consumed and thus the activity of PAOs and aerobic heterotrophic bacteria reduced correspondingly. Therefore, it was unfavorable to the biodegradation of TP and COD_{Mn} . Xiang et al. [27] and Zhang et al. [28] have shown that the concentration of DO has an effect on the removal of TP and COD in the bioreactors and demonstrated that the lower concentration of DO would inhibit the activity of microorganisms to a certain extent, which was not conducive to biodegradation of TP and COD. The actual operation of the BSF system should maintain a certain concentration of DO to ensure the removal efficiency of specific pollutants. The fitting results of biodegradation models of nutrients and organics are shown in Tables 7 and 8.

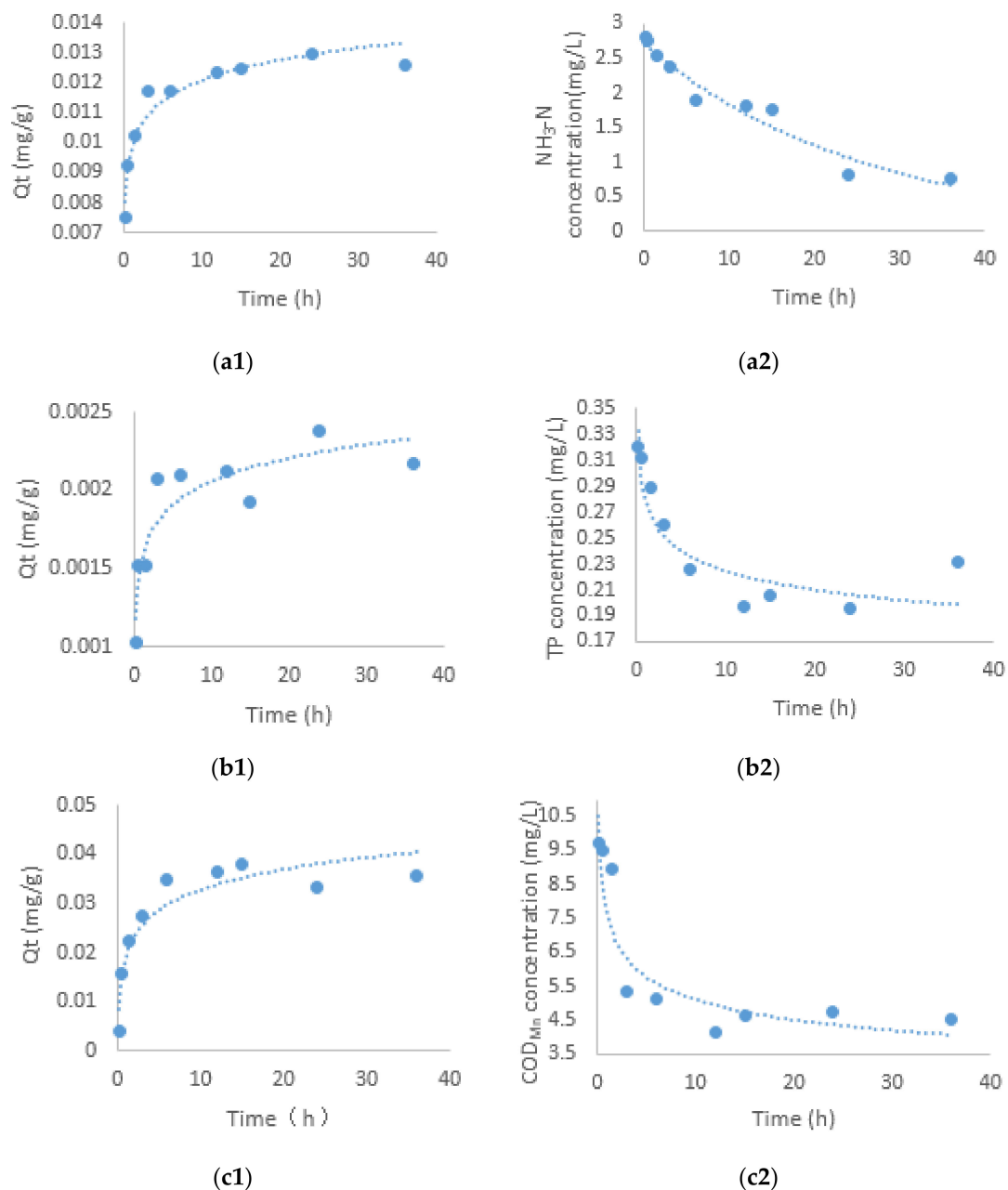


Figure 5. The adsorption amount or concentration change trend of various pollutants (a1,a2: $\text{NH}_3\text{-N}$, b1,b2: TP, c1,c2: COD_{Mn}) in batch experiment.

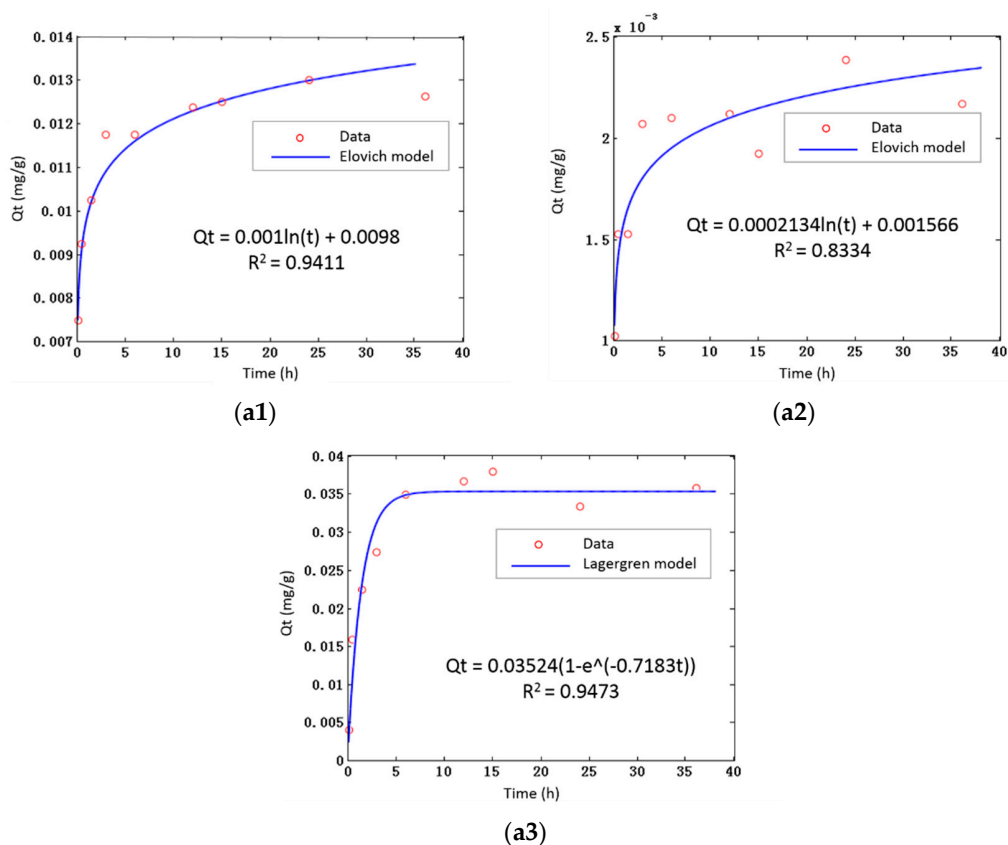
Table 7. Fitting results of adsorption kinetic model, Q_t : adsorption quantity (mg/g), t : time (h).

Index	Corrected "Elovich" Model	R^2	Lagergren Model	R^2	Power Function Model	R^2	Parabolic Diffusion Model	R^2
NH ₃ -N	$Q_t = 0.001\ln t + 0.0098$	0.9411	$Q_t = 0.01192(1 - e^{-4.608t})$	0.6921	$Q_t = 0.0096t^{0.0959}$	0.9166	$Q_t = 0.009001 + 0.0008032t^{0.5}$	0.7108
TP	$Q_t = 0.0002\ln t + 0.0016$	0.8334	$Q_t = 0.002022(1 - e^{-3.377t})$	0.6916	$Q_t = 0.0015t^{0.1311}$	0.8208	$Q_t = 0.00139 + 0.000174t^{0.5}$	0.6263
COD _{Mn}	$Q_t = 0.006\ln t + 0.0191$	0.8974	$Q_t = 0.03524(1 - e^{-0.7183t})$	0.9473	$Q_t = 0.0144t^{0.3462}$	0.7716	$Q_t = 0.01458 + 0.004707t^{0.5}$	0.6323

Table 8. Fitting results of biodegradation kinetic model, C: concentration of NH₃-N, TP and COD_{Mn} (mg/L).

Index	Zero-Order Kinetics Model	R^2	First-Order Kinetics Model	R^2	Second-Order Kinetics Model	R^2
NH ₃ -N	$C = -0.0593t + 2.5901$	0.909	$C = 2.7055e^{-0.039t}$	0.9331	$C = 2.828/(1 + 0.06394t)$	0.9434
TP	$C = -0.0026t + 0.2767$	0.4271	$C = 0.273e^{-0.01t}$	0.4188	$C = 0.2897/(1 + 0.0185t)$	0.5552
COD _{Mn}	$C = -0.128t + 7.7003$	0.4496	$C = 7.359e^{-0.019t}$	0.4687	$C = 9.018/(1 + 0.06538t)$	0.6792

As can be seen in Tables 7 and 8, the adsorption kinetics of NH₃-N and TP could be well fitted by Elovich model, while the adsorption process of COD_{Mn} could be better fitted by Lagergren model. Based on the biodegradation process of each pollutant, the second order kinetic model achieved better fitting of the biodegradation process of NH₃-N while the fitting results of TP and COD_{Mn} were not ideal. Considering the concentration change trend of TP and COD_{Mn} (as shown in Figure 5) simultaneously, we discovered that both biodegradation process of TP and COD_{Mn} would be better fitted by Power function model. The optimal fitting results of adsorption and the biodegradation kinetic model for NH₃-N, TP and COD_{Mn} are shown in Figure 6.

**Figure 6.** Cont.

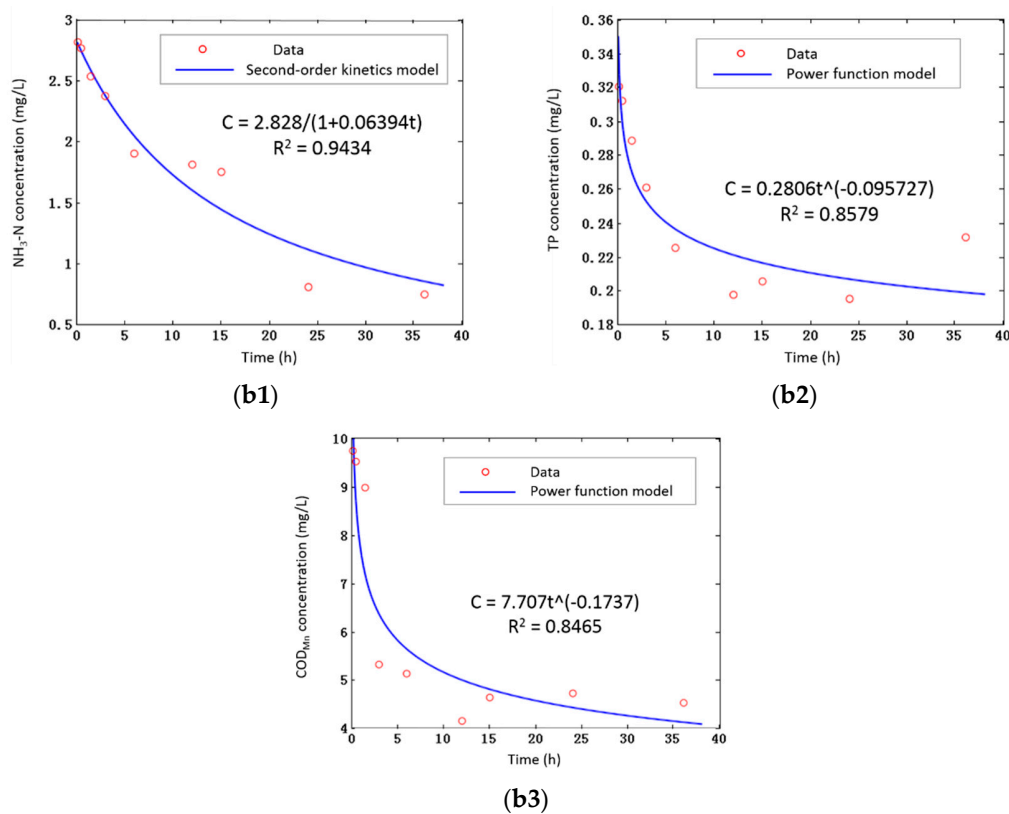


Figure 6. The optimal fitting results of adsorption and biodegradation kinetic model for (a1,b1) NH₃-N, (a2,b2) TP and (a3,b3) COD_{Mn}.

All in all, removal mechanisms of nutrients and organics in the BSF system could be expressed as follows: the adsorption of filter layer was dominant at the early running stage, and then the adsorption capacity of filter layer diminished, and biodegradation enhanced gradually at the same time, thus ensuring the removal efficiency of various pollutants in different periods. This study further explored the removal mechanisms of nutrients and organics in the BSF system and selected the optimal kinetic model to fit the adsorption and biodegradation process of above pollutants, it would be beneficial to evaluate removal performance of nutrients and organics in the BSF system at different operating stages in practice.

3.4. Investigation on the Removal Performance of Pollutants in Different Filter Layers

A certain volume of water sample was taken from the sample sites of each filter layer in the stable operating BSF system to analyze the removal rules of various pollutants in different filter layers (as shown in Figure 7). As can be seen in Figure 7, with the filter layer increasing, the concentration of turbidity, NH₃-N, TP and COD_{Mn} all had a reducing trend and the corresponding decline rates of concentration were gradually slowing down. In the range of 0–0.3 m filter layer, the removal rates of the above pollutants were accounted for 75.06%, 79.77%, 67.06% and 60.42% respectively of the corresponding total removal rate, which indicated that the surface filter layer (0–0.3 m) contributed a lot to the above pollutants removal and when the depth of filter layer increased to 0.5 m, turbidity, NH₃-N, TP and COD_{Mn} could be all basically eliminated. Analysis of this phenomenon might be related to the distribution difference of DO concentration and microbial communities in different filter layers [29]. The surface filter layer with abundant DO would be beneficial for the aerobic growth and thus enhanced aerobic biodegradation of NH₃-N, TP and COD_{Mn}, while with the filter layer increasing, the amount of DO was continually consumed and the mass transfer resistance was increasing correspondingly,

the DO concentration in the bottom filter layer was low and activity of aerobe reduced accordingly. Therefore, the removal performance of $\text{NH}_3\text{-N}$, TP and COD_{Mn} in this filter layer was not efficient.

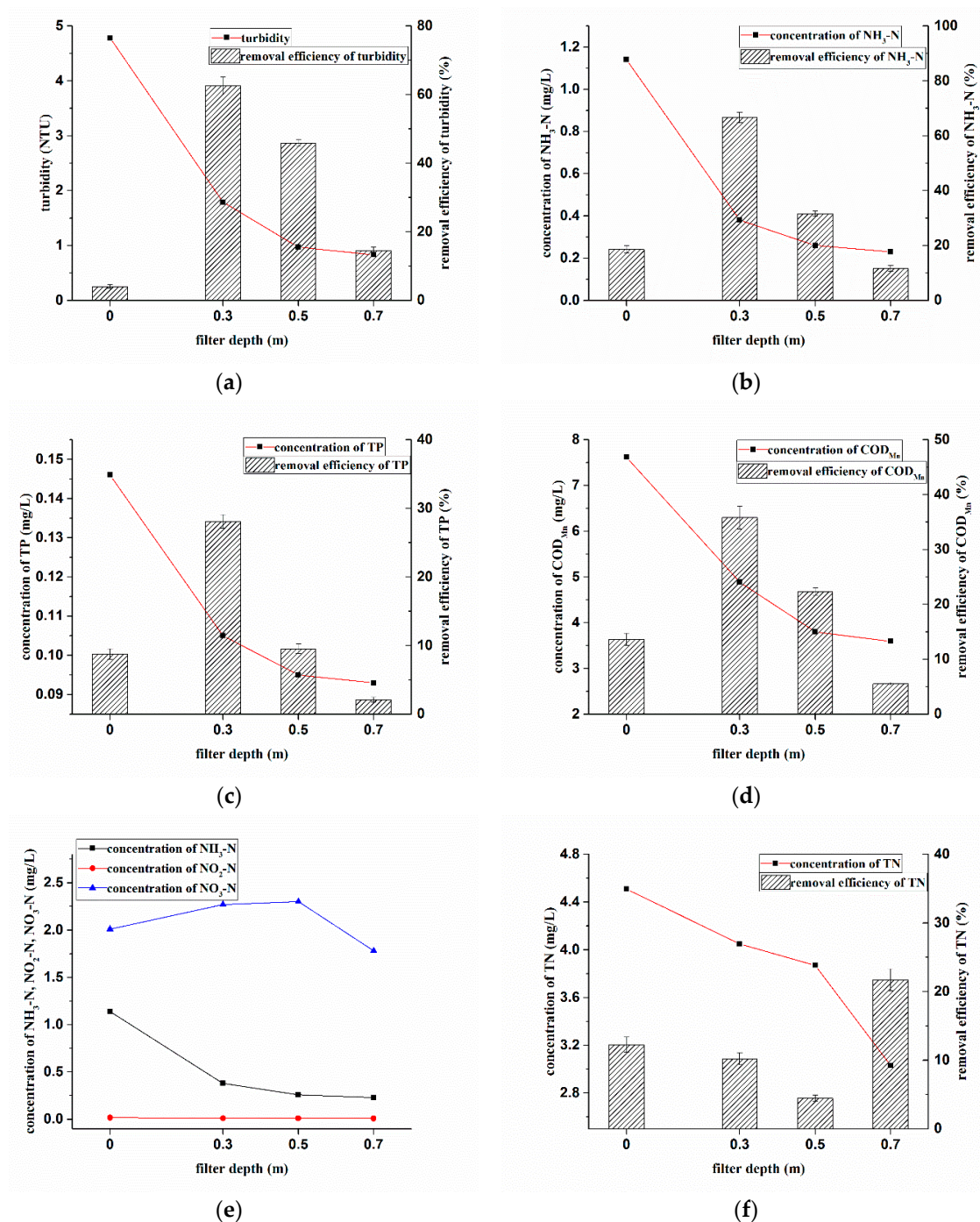


Figure 7. The removal rules of various pollutants ((a) turbidity, (b) $\text{NH}_3\text{-N}$, (c) TP, (d) COD_{Mn} , (e) three forms of nitrogen, (f) TN) along different filter layers.

By observing the transformation rules of different forms of nitrogen (including $\text{NH}_3\text{-N}$, $\text{NO}_2\text{-N}$ and $\text{NO}_3\text{-N}$) along filter layers (in Figure 7) and the distribution of DO in different filter layers, we could conclude that nitrification was dominated in the surface filter layer (ranged from 0 to 0.3 m), which showed that there was a large accumulation of $\text{NO}_3\text{-N}$ correspondingly with $\text{NH}_3\text{-N}$ concentration decreased at the same time and the DO concentration remained within the range of 5–6.5 mg/L.

Furthermore, in the middle filter layer (ranged from 0.3–0.5 m), nitrification and denitrification both existed, which characterized by the accumulation rate of $\text{NO}_3\text{-N}$ gradually slowed down with $\text{NH}_3\text{-N}$ concentration decreasing and DO concentration sustained 2–3 mg/L in this filter layer, but in the bottom filter layer (ranged from 0.5–0.7 m), denitrification was dominated and characterized by the accumulation of $\text{NO}_3\text{-N}$ became negative and TN concentration in this filter layer reduced sharply; in addition, the DO concentration of the bottom filter layer was about 0.5 mg/L and this anaerobic environment was benefit to the activity of denitrifying bacteria. In conclusion, each filter layer had different transformation laws of nitrogen and all closely related to the distribution of DO.

This study confirmed that the removal performance of various contaminants in different filter layers were distinct. The surface filter layer was advantageous to the interception and aerobic biodegradation of pollutants. However, the bottom filter layer was suitable for anaerobic biodegradation of specific pollutants. This phenomenon was closely related to the distribution difference of DO concentration and microbial community in different filter layers. The next study should be focused on the specific composition of microbial community in each filter layer and further investigate the relationship between removal efficiency of various pollutants and microbial composition in the BSF system.

4. Conclusions

This paper is based on a lab-scale biological slow filter to study the removal performance and degradation mechanism of the typical pollutants (including nutrients and organics) in micro-polluted water source and therefore provide a theoretical foundation for the actual application of biological slow filter in treatment of micro-polluted water in developing countries. In addition, this research also explored the effect of main influence factors and their interactions on pollutants removal efficiency in the BSF system and obtained the optimal scheme of operating parameters so as to provide an optimized basis for the actual operation of the BSF system. The primary conclusions are summarized as follows:

- (1) Ammonia nitrogen ($\text{NH}_3\text{-N}$), total nitrogen (TN), total phosphorus (TP), permanganate index (COD_{Mn}) and turbidity all have good removal performance in a stable operating biological slow filter and the corresponding removal rates are 83.65%, 42.45%, 42.94%, 60.41% and 83.55%, respectively.
- (2) Using the response surface methodology to investigate the influence of four main parameters (including filtration rate, filter depth, hydraulic head and temperature) on various pollutants removal rate in the BSF system and we can discover that the change of the above operating parameters would have different degrees of influence on the pollutants removal efficiency. Furthermore, the interactions between filter depth and temperature have a significant influence on $\text{NH}_3\text{-N}$ removal performance; the interactions between filter depth and hydraulic head have a significant influence on TN removal performance.
- (3) The optimal operating parameters are: filtration rate 0.1 m/h, filter depth 0.8 m, hydraulic head 0.64 m, temperature 26.06 °C. And the corresponding removal rates of $\text{NH}_3\text{-N}$, TN, TP, COD_{Mn} and turbidity are 92.04%, 48.49%, 45.60%, 63.38% and 82.95%, respectively.
- (4) We proved that the removal pathway of nutrients and organics in the BSF system by batch experiment and kinetic model fitting, and found that the adsorption of the filter layer was dominant at the early running stage, and then the adsorption capacity of the filter layer diminished and biodegradation enhanced gradually at the same time, thus ensuring the removal efficiency of the above pollutants in different periods.
- (5) By studying the removal rules of various pollutants in different filter layers, we could find that turbidity, $\text{NH}_3\text{-N}$, TP and COD_{Mn} are mainly removed in the surface filter layer, while TN is basically eliminated in the bottom filter layer. According to the transformation regularity of nitrogen in different forms along filter layers, we can conclude that nitrification is dominated

in the surface filter layer while denitrification is dominated in the bottom filter layer. This phenomenon is closely related to the distributional difference of DO in each filter layer.

Supplementary Materials: The following is available online at <http://www.mdpi.com/2227-9717/6/8/128/s1>, Table S1: Influence of various factors and their interactions on the removal rate of different pollutants.

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