Pilot study on the treatment of low carbon and nitrogen ratio municipal sewage by AOAAO sludge-membrane coupling process with multi-point inflow

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Abstract

A new multi-point inflowA1/O2/A3/A4/O5 sludge-membrane coupling process and pilot plant were developed and designed to solve the problem of nitrogen and phosphorus removal of low C/N domestic sewage in southern China. By changing the distribution ratio of multi-point influent, the removal effect and transformation rule of organic matter, nitrogen and phosphorus in the system were studied. Results showed that when the average low C/N ratio of influent was 2.09 and the influent distribution ratio was 1:1, the average concentrations of COD, NH_4^+- N, TN and TP in the effluent were 21.31 mg/L, 0.60 mg/L, 12.76 mg/L and 0.34 mg/L, respectively, and the average removal rates are 87.3%, 98.7%, 74.1% and 88.1% respectively. When the low temperature was 12–15°C, the average removal rates were 87.3%, 98.7%, 74.1% and 88.1%, respectively. Compared with the traditional A^2O process under the same conditions, the TN removal rate was increased by 15.4%, and the TP removal rate was increased by 22.2%. This system has obvious advantages in treating wastewater with low carbon and nitrogen ratio, which solved the problem that the effluent of biological phosphorus removal from low C/N ratio domestic sewage was difficult to be lower than 0.5 mg/L.

1. Introduction

With the problem of water eutrophication becoming more and more serious, the standard of nitrogen and phosphorus emission is becoming more and more strict (Zhang et al. 2017). As a simple simultaneous nitrogen and phosphorus removal process, the traditional A^2O process is one of the most widely used biological nitrogen and phosphorus removal processes in urban wastewater treatment plants of China due to its simple structure, small control complexity and difficulty in sludge bulking (Abyar et al. 2018; Yan et al. 2016; Ji et al. 2019). However, the traditional A^2O process is a single activated sludge system, and there are a series of problems. Since denitrifying bacteria, nitrifying bacteria and phosphorus accumulating bacteria live in the same environment, the competition between denitrifying bacteria and phosphorus accumulating bacteria for carbon source and the contradiction between nitrifying bacteria and phosphorus accumulating bacteria for SRT will be caused, inevitably (Smolders et al. 2010; Natalia et al. 2018; Peng et al. 2006; Li et al., 2019). In recent years, researchers have made improvements on the basis of the traditional A^2O process, and many improved A^2O processes have been greatly explored and developed (Liu et al. 2020; Peng et al. 2020; Wang et al. 2019). However, the problem of nitrogen and phosphorus removal in domestic sewage with low C/N ratio has not been solved yet. Chang and Ouyang (2000) Found that compared with the traditional A^2O process, the improved AOAO process with multi-point influent could greatly save energy consumption and carbon source, but there were some problems such as poor resistance to impact load of NH_4^+-N. Vaiopoulou and Aivasidis (2008) In the study of multi-point influent UCT process system found that the total phosphorus of biological effluent still could not reach the National Class A discharge standard of China (GB 18918 – 2002) stably. Nan et al. (2018) Enhanced the denitrification and phosphorus removal of DPAOs in the improved A^2O-BAF double sludge system by step-feeding, so as to realize the effective utilization of carbon source in the system, however, it still could not solve the contradiction between denitrifying bacteria and phosphorus accumulating bacteria for carbon source competition, and backwashing would consume a lot of water. Peng and Ge (2011) Successfully enhanced the SND effect of the system by controlling the lower DO concentration in the aerobic stage and increasing the sludge concentration in the step-feeding process, which enhanced the utilization rate of carbon source in the system. However, the problem of reaching the standard of nitrogen and phosphorus has not been solved yet. For urban domestic sewage, especially for low C/N ratio domestic sewage, the contradiction between organic matrix competition and SRT is particularly prominent, which seriously limits the further improvement of nitrogen and phosphorus removal efficiency (Deng et al. 2016). It is urgent to optimize and upgrade the traditional A^2O process.

In this paper, a multi-point inflow pre-anoxic-oxic-anaerobic-anoxic-oxic (A1/O2/A3/A4/O5) sludge-biofilm coupling process was developed and designed. So far, this new multi-point inflow A1/O2/A3/A4/O5 sludge-biofilm coupling process has not been studied by others. In this project, On the basis of traditional A^2O, pre-anoxic reactor(A1) and sludge-biofilm coupling reactor(O2) were added to remove organic carbon sources in raw water by using nitrate in the reflow sludge, so as to reduce the restraining influence of organic matter on nitrification of sludge-biofilm coupling reactor(O2) reactor. At the same time, in view of the contradiction of SRT between nitrifying bacteria and phosphorus accumulating bacteria, suspended biological carrier was added in the O2/O5 reactor to make nitrifying bacteria adhere to the filler. Since most nitrification occurred on the biofilm, the biological phosphorus removal efficiency could be improved by reducing the SRT of the activated sludge in the system without affecting nitrification, thus solving the problem of SRT contradiction between nitrifying bacteria and phosphorus accumulating bacteria, and improving the nitrification efficiency and load impact resistance of the system. The competition between denitrifying bacteria and phosphorus accumulating bacteria for carbon source was solved by multi-point water inflow, and the utilization rate of carbon source was improved. In this paper, the effects of different multi-point influent ratio and temperature on the performance of nitrogen and phosphorus removal of the system were studied, and compared with the traditional anaerobic-anoxic-oxic (A^2O) process, which provided an effective theoretical basis for the optimization and upgrading of the existing sewage treatment plant and the treatment of low carbon and nitrogen ratio (low C/N) ratio sewage. It provides a new idea to solve the contradiction of SRT between nitrifying bacteria and phosphorus accumulating bacteria, and competition between denitrifying bacteria and phosphorus accumulating bacteria for carbon source.

2. Materials And Method

2.1 A1/O2/A3/A4/O5 pilot plant

The schematic plan diagram of the A1/O2/A3/A4/O5 sludge-membrane coupling reactor was shown in Fig. 1 (a),and the process flow diagram of pilot reactor was shown in Fig. 1 (b). The size of the reactor was 1.94 m × 0.45 m × 0.5 m, and the effective volume was 265 L. Furthermore, the device was made of carbon steel anticorrosive material, which was composed of pre-anoxic section (A1), aerobic sludge-biofilm coupling section (O2), anaerobic section (A3), anoxic section (A4), aerobic sludge-biofilm coupling section (O5) and sedimentation tank (C), with volumes of 21L, 75L, 38L, 56L and 75L respectively. The treated sewage enters the pre-anoxic section (A1) and the anaerobic section (A3) respectively through multi-point distribution of inlet water. Part of the sludge in the sedimentation tank (C) returned to the pre-anoxic section (A1), and the excess sludge was discharged. The nitrification liquor in the aerobic sludge-biofilm coupling section (O5) was returned to the anoxic section (A4). MBBR suspended filler with a filling ratio of 30% was added into the aerobic sludge-biofilm coupling section (O2) and the aerobic sludge-biofilm coupling section (O5). Keeping the total residence time and parameters of section A (anaerobic section, anoxic section) and section O unchanged, the traditional A^2O comparison device was set up, and the process flow was shown in Fig. 10 (a).

2.2 Test water and analytical methods

The actual domestic sewage from Ma Anshan Economic Development Zone, Anhui Province, China was used as the test water. It was found that the sewage was a typical urban domestic sewage with low $C/N_{(BOD5/TN)}$ and high concentration of nitrogen and phosphorus while low BOD_5 and COD. The actual influent water quality was shown in Table 1.

The Do and temperature were measured by flexihq30d portable dissolved oxygen meter. The pH was measured by pH meter. The NH_4^+-N, nitrite nitrogen, nitrate nitrogen, COD, TP, TN and SS were determined by Nessler colorimetric method, N-(1-naphthalene)-ethylenediamine spectrophotometric, thymol spectrophotometry, potassium dichromate method, ammonium molybdate spectrophotometry, alkaline potassium persulfate digestion UV spectrophotometry, and gravimetric method (APHA. 1998).

Table 1
Intake Water Quality

Water quality index	**pH**	**COD (mg/L)**	**BOD_5 (mg/L)**	**NH_4^+-N (mg/L)**	**TN (mg/L)**	**TP (mg/L)**	**C/N (BOD_5/TN)**
Range	7.0~7.9	157.3 ~ 199.5	100.1 ~ 126.9	36.5 ~ 49.9	41.8 ~ 54.4	1.8 ~ 3.8	1.7 ~ 3.7
Mean concentration.	7.4	160.6	102.1	45.1	48.9	2.9	2.09

2.3 Operating conditions

The multi-point influent A1/O2/A3/A4/O5 sludge-biofilm coupling pilot plant was operated under the temperature (12–30°C). The inoculated sludge was taken from the sludge cake of the South Ma Anshan sewage treatment plant. The MLVSS/MLSS of the seed sludge was 0.54, the sludge concentration was 36.57 g/L (measured by MLSS), and the inoculation amount of sludge was 2850 mg/L (measured by MLSS). Under normal operation of the system, the average MLSS in the system was 2032 mg/L. The previous inlet flow research experiment found that the system had the best effect on pollutant removal when the flow was 0.91 m/d. Under the operating condition of keeping the total inlet flow at 0.91 m/d, the system was investigated by adjusting different inlet flow distribution ratio ($Q_{A1\ segment}$: $Q_{A3segment}$). At the same time, the influences of different temperature ranges on the system were analyzed and recorded, and the results were compared with the traditional A^2O process. Samples were taken twice a day to measure the different pollutant concentrations of the influent, the end of pre-anoxic section (A1), the end of aerobic sludge-biofilm coupling section (O2), the end of anaerobic section (A3), the end of aerobic sludge-biofilm coupling section (O5), and the effluent. Operation parameters as follows: the total inflow was 0.91 m/d, the filling ratio of suspended fillers in the aerobic sludge-biofilm coupling section (O2/O5) was 30% and the dissolved oxygen (DO) was maintained at about 1.5-3.0 mg/L, and the DO of pre-anoxic/anaerobic/anoxic reactors (A1/A3/A4) was controlled in 0.10–0.15 mg/L. The flow of influent and reflux were controlled jointly by valve regulation and manual measurement. The sludge reflux ratio was 75% and the nitrification reflux ratio was 250%. Total HRT was 7 h and SRT was 7.5 days. As showed in Table 2, the multi-point influent operation was divided into 5 stages (Run1-Run5), and the influence of temperature on the process performance was considered. Finally, a comparative study was conducted with the traditional A^2O device under the same working condition.

Table 2 Experimental procedure and parameter for the process in steady state experiments

Run	day	Operating conditions					
		Water inflow (m/d)	Influent ratio (Q_{A1}:Q_{A3})	Average inflow load (kg/(m^3·d))			
				COD	NH_4^+-N	TN	TP
1	1-15	0.91	3:7	0.58	0.15	0.17	0.0096
2	16-30	0.91	4:6	0.54	0.16	0.17	0.0100
3	31-45	0.91	5:5	0.58	0.15	0.16	0.0100
4	46-60	0.91	6:4	0.58	0.15	0.17	0.0098
5	61-75	0.91	7:3	0.57	0.16	0.16	0.0099
6	76-83	0.91	5:5	0.58	0.15	0.16	0.0100
7	84-91	0.91	5:5	0.57	0.16	0.16	0.0099
8	92-99	0.91	5:5	0.58	0.15	0.16	0.0100
Traditional A^2O	100-140	0.91	-	0.58	0.15	0.17	0.0096

3. Results And Discussion

3.1 Removal performance of COD with different inflow ratio.

As showed in Fig. 2, it could be seen that under the fluctuation of COD in influent ranging from 157.3 mg/L to 199.5 mg/L, and different multi-point influent ratio ($Q_{A1\ segment}$: $Q_{A3\ segment}$) had basically no influence on COD removal. Under the operating conditions of Run1 to Run5, the effluent COD concentrations were 23.93 mg/L, 21.96 mg/L, 21.31 mg/L, 19.97 mg/L and 21.80 mg/L, respectively, and the removal rates were 85.77%, 86.97%, 87.34%, 88.24% and 87.03%, respectively. Under all operating conditions, the effluent COD was better than the National Class A discharge standard of China (GB 18918 – 2002).

As showed in Fig. 3, most of COD was removed in the pre-anoxic section (A1) and anaerobic section (A3), while only a small part of COD was consumed in the anoxic section (A4) and aerobic sections(O2/O5).Moreover, with the increasing of the influent ratio of pre-anoxic stage (A1), the available COD in the pre-anoxic section (A1) was also increasing. Correspondingly, with the continuous decrease of the influent ratio of the anaerobic section (A3), the available COD in the anaerobic section (A3) and the anoxic section (A4) was also decreasing. Under the conditions of Run1-Run5 with different multi-point influent ratio ($Q_{A1\ segment}$: $Q_{A3\ segment}$), the cumulative total removal of COD in the pre-anoxic/anaerobic/anoxic stage (A1/A3/A4) were 100.96 mg/L, 115.79 mg/L, 119.08 mg/L, 119.87 mg/L, 121.41 mg/L, respectively. The cumulative effective utilization ratio of COD in the total COD removal rate were 70%, 79%, 81%, 80% and 83%, respectively. However, the cumulative removal capacity of the aerobic stage (O2 / O5) was 43.27 mg/L, 30.78 mg/L, 27.93 mg/L, 29.97 mg/L, 24.86 mg/L, respectively. The cumulative removal rate was 30%, 21%, 19%, 20% and 17%, respectively. Most of the organic matter was consumed in the anaerobic and anoxic stages. It is considered that the raw water first entered the pre-anoxic (A1) and anaerobic (A3) stages, most of the COD in influent would be consumed by anaerobic phosphorus release and anoxic denitrification as electron donor, which resulted in that different proportion of multi-point influent had basically no influence on COD removal, which was basically consistent with the conclusion of Wang et al. 's study on the effect of nitrogen and phosphorus removal in multi-stage AO system through different influent methods(Abyar et al. 2018; Wang et al. 2018).

3.2 Removal performance of nitrogen with different inflow ratio

Figure 4 showed the removal efficiency of NH_4^+-N by different multi-point influent ratio. It could be seen from Fig. 4 that although the influent ammonia nitrogen concentration was high and fluctuates greatly (40.22–50.89 mg/L), and the average influent NH_4^+-N concentration was 45.14 mg/L, the effluent ammonia nitrogen concentration was relatively stable, and the effluent ammonia nitrogen concentration with 97% coverage could reach less than 5 mg/L. The average effluent ammonia nitrogen concentrations under different operating conditions of Run1 ~ Run5 were 3.94 mg/L, 0.80 mg/L, 0.60 mg/L, 1.47 mg/L and 2.95 mg/L, respectively. The removal rates were 91.31%, 98.28%, 98.69%, 96.71% and 93.75%, respectively. The system showed strong nitrification capacity. It was considered that most nitrification taken place on the aerobic (O2/O5) packing biofilm, which prolonged the SRT of nitrifying bacteria and enhanced the nitrification ability of the system. This feature could also be confirmed in the study of Falahti-Marvastand Karimi-Jashni (2015) and Artigade et al. (2005).

Compared with other operating conditions, the ammonia nitrogen concentration in the effluent of Run1 decreased first and then increased with the increasing of the influent distribution ratio. In Run1 operation condition (influent ratio is 3:7), due to the large proportion of influent water in the anoxic section (A4), the NH_4^+- N load in the aerobic section (O5) was too large, which exceeded the treatment load of the aerobic section (O5). Nitrifying bacteria could not nitrify ammonia nitrogen completely in time, resulting in the increase of ammonia nitrogen concentration in effluent. Under the conditions of Run2 and Run3 with the influent ratio of 4:6 and 5:5, the effluent ammonia nitrogen had a better performance, which indicated that the operating condition of the system was the best under the inflow distribution mode of Run2 and Run3. However, with the continuous increase of ammonia nitrogen in Run4 and Run5, it was considered that the effect of ammonia treatment in Run4 and Run4 were better than that in Run1. Moreover, the increase of the proportion of feed water in the front section was helpful to the production of denitrification alkalinity, which promoted the ammonia nitrogen degradation effect of the system. It could be seen from Fig. 4 and Fig. 5 that the multi-point influent A1/O2/A3/A4/O5 process had a good treatment effect on the removal of ammonia nitrogen in low C/N ratio domestic sewage under the appropriate influent ratio. In this study, adding pre-anoxic section (A1) and aerobic section (O2), on the one hand, increased the residence time of wastewater in the anaerobic stage, so that denitrifying phosphorus accumulating bacteria can fully reserve PHB; on the other hand, the presence of certain nitrate made a large amount of denitrifying phosphorus accumulating bacteria in anaerobic section (A3), which was conducive to the further improvement of denitrifying nitrogen and phosphorus removal process. According to the traditional A^2O theory of nitrogen and phosphorus removal, there was a competitive relationship between phosphorus accumulating bacteria and denitrifying bacteria. Reasonable distribution of organic carbon sources in raw water by step-feeding was conducive to the realization of nitrogen and phosphorus removal, which is consistent with the conclusion of this study (Zhu et al., 2007). In conclusion, the ammonia nitrogen removal rate was the highest when the influent ratio was 5:5.

Figure 5 showed the variation law of NH_4^+-N and NO_3^--N under various working conditions. It could be seen from Fig. 5 that a large amount of ammonia nitrogen was removed by nitrification in the aerobic section (O2) and the aerobic section (O5), and the ammonia nitrogen in the effluent of the system was very low, which indicated that the aerobic sludge-membrane coupling system had a strong nitrification capacity. Moreover, with the increasing of the influent ratio of the pre-anoxic section (A1), the ammonia nitrogen in the effluent of the aerobic section (O2) was also increasing. Correspondingly, with the continuous decrease of the influent ratio of the anaerobic section (A3), the ammonia nitrogen content in the effluent of the aerobic sludge membrane coupling system was very low. The concentration of ammonia nitrogen in the effluent of aerobic stage (O5) was also decreasing. The reason for the increase of ammonia nitrogen in the effluent of aerobic stage (O5) under the condition of Run5 was that the influent ratio of pre-anoxic stage (A1) is too high, which leaded to the high ammonia nitrogen in effluent of aerobic stage (O2), which leaded to the increase of ammonia nitrogen in effluent of aerobic stage (O5). For the variation law of nitrite nitrogen in each section of the reactor, because the content of nitrite nitrogen was too low in the process of system operation, so the research on nitrite nitrogen was omitted in the process of data statistics. As for the variation law of nitrate nitrogen in each section of the reactor, it could be seen from Fig. 6 that nitrate nitrogen could be effectively removed in anaerobic section (A3) and anoxic section (A4). Under the condition of Run1, due to the high proportion of influent water in the anaerobic section (A3), a large amount of nitrate nitrogen was produced in the aerobic section (O5). The nitrate nitrogen returned to the anoxic tank (A4) without sufficient carbon source for denitrification, resulting in a large amount of NO_3^--N in the effluent. Under Run2 and Run3 conditions, as the inflow ratio of anaerobic section (A3) continuously decreases, the nitrate nitrogen concentration generated by nitrification of aerobic section (O5) also decreases. The nitrate nitrogen generated by aerobic section (O2) and aerobic section (O5) reflux to anoxic section (A4) was denitrified by DPAOs, so the nitrate nitrogen content in effluent was relatively low. Then, under the operating conditions of Run4 and 5, with the further decrease of the influent ratio of the anaerobic section (A3), the carbon source in the influent of the anaerobic section (A3) was insufficient. Although the influent ratio of the pre-anoxic section (A1) was very high, most of the organic matter was consumed in the pre-anoxic (A1) and aerobic (O2) sections, so the remaining carbon source could not make up for the carbon source demand of the subsequent units, and the denitrification process was hindered and NO_3^--N could not be effectively removed, resulting in a high concentration of nitrate nitrogen in the effluent, which was consistent with Mahne et al. (1996). In conclusion, when the influent ratio was 5:5, the removal effect of NO_3^-- N was the best.

Fig. 6 showed the TN removal effect of different multi-point influent ratio. From Fig. 6, it could be found that the influent TN concentration was 43.84-57.29 mg/L, and the average influent concentration was 48.91 mg/L. Under the condition of Run1-Run5, the average TN concentration in the effluent were 17.17 mg/L, 12.81 mg/L, 12.76 mg/L, 14.76 mg/L and 17.17 mg/L, and the removal rates were 66.25%, 74.09%, 74.12%, 70.54% and 64.86%, respectively. Under the condition of Run1-Run5, with the continuous increase of influent ratio in pre- anoxic stage (A1), the TN removal rate first increased and then decreased. Fig. 7 showed the C/N ratio of influent and its consumption in Section A under various working conditions. It could be seen from Fig.7 that the average C/N (BOD_5/TN) ratio of influent water were 2.11, 2.19, 2.18, 2.17 and 2.20 respectively. The ratio of C/N (BOD_5/TN) consumption in A (A1/A3/A4 cumulative) section under each working condition (Run1 ~ Run5) were 4.42, 5.16, 3.23, 3.10 and 4.17. The C/N ratio in influent had little change, but its consumption in A segment had a great change.

It could be seen from Fig. 6 that the TN removal effect was the worst under Run1 condition. It shows that the high influent ratio of anaerobic section (A3) leads to too high influent ammonia nitrogen load, and the nitrifying bacteria could not timely transform ammonia nitrogen. In addition, from Fig. 7, you could see that compared with Run3 and Run4, the consumption of $C/N_{(BOD5/TN)}$ in A section under Run1 working conditions was higher, however, a large amount of nitrate nitrogen in the reflux nitrification solution would lead to insufficient carbon sources in the anoxic segment (A4), so the nitrate nitrogen could not be removed in time through the denitrification in the anoxic segment (A4),therefore, the effluent of the aerobic segment (O5) contained a large amount of nitrate nitrogen (see Figure 5). Moreover, a large amount of nitrate nitrogen was returned to the pre-anoxic section (A1) through sludge reflux. It could be demonstrated from Fig. 3 that there was no sufficient carbon source in the pre-anoxic section (A1) to remove a large amount of nitrate nitrogen from the return sludge, which could also be seen from Run1 in Figure 5. Under Run2 and Run3 conditions, TN removal efficiency was the best, which was consistent with the ammonia nitrogen removal effect in Fig. 4. In Run1-Run5, the nitrogen removal effect was mainly affected by the adequacy of the influent carbon source. Under Run2 condition, the $C/N_{(BOD5/\ TN)}$ consumption ratio of section A was the highest, which indicated that there was sufficient carbon source for denitrification under Run2 condition, so the TN removal rate was very high, and denitrifying phosphorus removal might exist in the anoxic section (A4), which was consistent with the high removal rate of TP under Run2 condition in Fig. 8. In addition, although the $C/N_{(BOD5/\ TN)}$ consumption ratio of section A under Run2 condition was much higher than that of Run3, the TN concentration in effluent was similar, indicating that nitrogen removal occurs in the aerobic stage. This could also be seen from the nitrogen change of Run2 in Fig. 5. It could be seen from Fig. 5 that under Run2 condition, ammonia nitrogen was significantly reduced in the aerobic sludge membrane coupling section (O5), while the nitrate nitrogen content was not significantly increased. The reason for this phenomenon might be simultaneous nitrification and denitrification in the coupling section of aerobic sludge-membrane (O5), and denitrification occurred when ammonia nitrogen was converted into nitrate nitrogen. Under Run3 condition, the $C/N_{(BOD5/TN)}$ consumption ratio of section A was as low as 3.23, which was far lower than the theoretical value of denitrification, but the total nitrogen removal rate was the best. The analysis showed that under Run3 condition, there were denitrifying phosphorus accumulating bacteria (DPAOs) in the anoxic section (A4). In the absence of carbon source, the denitrifying phosphorus accumulating bacteria (DPAOs) used nitrate nitrogen as electron acceptor to achieve nitrate nitrogen removal. Therefore, nitrate nitrogen in the anoxic section (A4) decreased significantly (see Fig. 5). The high removal rate of TP under Run3 condition in Fig. 8 also confirmed this point. When the influent ratio of pre-anoxic stage (A1) was higher than that of anaerobic stage (A3) (flow ratio was more than 5:5), TN removal rate began to decline. It could be seen from Fig. 7 that the C/N consumption ratio of section A was the lowest under Run4 condition, which indicated that denitrifying bacteria lack sufficient carbon source for denitrification, so TN removal rate was reduced. However, under the condition of Run5, the C/N consumption of section A was relatively high, but the removal effect of TN was poor. The analysis showed that, with the increase of feed water ratio of pre-anoxic section (A1), the carbon source in the influent of pre-anoxic section (A1) could meet the consumption of partial denitrification and phosphorus accumulating bacteria in pre-anoxic section (A1), and part of carbon source was consumed in aerobic section (O2). However, most of the carbon sources are consumed in the first half of the system, resulting in the remaining carbon sources unable to meet the demand of nitrogen and phosphorus removal in the latter half of the system. Moreover, due to the decrease of the proportion of influent in the anaerobic section (A3), the available carbon sources in the anaerobic section (A3) and the anoxic section (A4) continued to decrease. This could be seen from the COD consumption in Figure 3, which leaded to the shortage of carbon sources in the latter half of the system. The effect of nitrogen and phosphorus removal was affected. In conclusion, the total nitrogen removal rate was the highest when the influent ratio was 5:5.

3.3 Removal performance of TP with different inflow ratio

According to the classical theory of phosphorus release, anaerobic unit was the main place of phosphorus release. Under anaerobic conditions, the phosphorus accumulating bacteria released polyphosphates from the body,at the same time, organic substances such as PHA were absorbed, and the PHA stored in the body was decomposed and released energy under aerobic conditions. Meanwhile, dissolved phosphorus was absorbed in the environment and stored in the body in the form of polyphosphates, and then discharged from the system in the form of phosphorous-rich sludge to achieve the purpose of phosphorus removal (Yoshitaka 1994; Hu et al. 2003).However, According to the theory of denitrifying phosphorus removal, denitrifying phosphorus accumulating bacteria (DPAOs) released phosphorus under anaerobic conditions, and then in anoxic or aerobic conditions, DPAOs absorbed excessive phosphorus and denitrify with nitrate or nitrite nitrogen as electron acceptor to realize simultaneous nitrogen and phosphorus removal(Kuba et al. 1993; Pochana et al. 1999; Kuba et al. 1999).In this study, the sludge retention time (SRT) was set as 7.5 d, and the phosphorus removal was achieved by sludge discharge. The contradiction of SRT between nitrifying bacteria and phosphorus accumulating bacteria was solved by adding suspended biological carrier into the system, and the problem of carbon source competition was solved by multi-point influent.

It could be seen from Fig. 8 that the TP concentration of influentwas1.80-3.82 mg/L, the average influent concentration was 2.90 mg/L, and the influent C/P (BOD_5/ TP) ratio was 26.31–70.50. Under the operating conditions of Run1-Run5, the TP concentration in effluent of the system were0.44 mg/L, 0.33 mg/L, 0.34 mg/L, 0.45 mg/L and 0.51 mg/L, and the average removal rates were 84.23%, 88.77%, 88.12%, 84.34% and 82.34%, respectively. The TP removal rate of the system was significantly affected by the proportion distribution of influent, and the removal rate increased at first and then decreased. The concentration of total phosphorus in the influent changed greatly, but the concentration of total phosphorus in the effluent was relatively stable, indicating that the process had strong impact resistance.

It could be seen from Fig. 8 that in Run1-Run5, Run1, Run4 and Run5 (when the influent ratios were 3:7, 6:4 and 7:3), the effluent concentration of total phosphorus was higher and the removal rate was poor. Through calculation, the C/P(BOD_5/TP) consumption ratio under each working condition were 38.00, 36.08, 36.54, 39.54, 39.31, respectively. Under Run1 operation condition, although the C/P consumption ratio was large, the residence time of anaerobic section (A3) was correspondingly shorter due to the large influent ratio in anaerobic stage (A3), and the phosphorus uptake in the aerobic section(O5) was insufficient due to the insufficient phosphorus release of the phosphorus accumulating bacteria in the anaerobic phase (A3).Moreover, it could be seen from Fig. 5 that the concentration of nitrate nitrogen in the effluent of Run1 was very high, and a large amount of nitrate nitrogen (NO_3^--N = 15 mg/L) entered the pre-anoxic section (A1) with the sludge return, resulting in the competition of denitrifying bacteria and phosphorus accumulating bacteria for carbon source. Some carbon sources were used for denitrification, affecting phosphorus release and PHB synthesis of phosphorus accumulating bacteria, and then affecting phosphorus absorption in aerobic section (O2). Furumai (1999) and Garzon-Zuniga and Gonzalez-Martinez (1996) Also found that when the nitrate nitrogen concentration exceeded 10 mg/L, PAOs could not release sufficient phosphorus in the anaerobic stage. Under Run2 condition, although the C/P consumption ratio of the system was only 36.08, the phosphorus removal efficiency of the system was the best, with an average removal rate of 88.77%, and the average phosphorus concentration in the effluent was as low as 0.33 mg/L. The results showed that with the decrease of influent ratio in the anaerobic stage (A3), the residence time of each reaction unit after the anaerobic stage (A3) was extended accordingly, so the phosphorus accumulating bacteria could fully release phosphorus in the anaerobic phase (A3) and absorbed phosphorus in the aerobic section (O5). At the same time, with the increase of influent in pre-anoxic stage (A1), more phosphorus could be removed by alternating changes of pre-anoxic stage (A1) and aerobic stage (O2), thus reducing the pressure on phosphorus treatment in the latter half stage. Denitrifying phosphorus removal was a kind of facultative anaerobic microorganism with denitrification and phosphorus removal under the condition of alternating operation of anaerobic/anoxic environment. The phosphorus accumulating bacteria could use NO_3^-as electron acceptor to complete the process of excessive phosphorus absorption and denitrification at the same time through their metabolism, so as to minimize the carbon source demand and realize the double saving of energy and resources. Denitrifying phosphorus removal could save about 50% of COD and 30% of oxygen, and reduce about 50% of excess sludge. Under Run3 condition, the C/P consumption ratio was very low (C/P = 36.54), and the system lacked carbon source, but the phosphorus removal capacity was very good. As the system operates alternately in anaerobic/anoxic environment, as mentioned above, the possible reason for this phenomenon was denitrifying phosphorus removal in the system, so the system had good removal effect on nitrogen and phosphorus under Run3 condition. The reason why the phosphorus removal efficiency of Run3 was slightly lower than that of Run2 might be due to the high content of nitrate nitrogen from aerobic stage (O2) to anaerobic stage (A3) under Run3 condition, which leaded to carbon source competition between denitrifying heterotrophic bacteria and phosphorus accumulating bacteria in anaerobic section (A3), which was basically consistent with the research conclusion of Banu et al. (2009). Under the conditions of Run4 and Run5, the phosphorus concentration in the effluent of the system reached 0.45 mg/L and 0.51 mg/L, respectively, which further reduced the phosphorus removal effect of the system. Although the C/P consumption ratio were 39.54 and 39.31 under Run4 and Run5 conditions, when the influent ratio of pre-anoxic section (A1) was further increased, a large number of carbon sources were consumed in the pre-anoxic section (A1) and aerobic section (O2), resulting in the shortage of carbon source in the influent of the second half of the reactor, which affected the anaerobic phosphorus release and aerobic phosphorus absorption of the subsequent process, and greatly limited the phosphorus removal effect of the system. At the same time, a large amount of ammonia nitrogen was converted to nitrate nitrogen in the aerobic section (O2), resulting in a large amount of nitrate nitrogen accumulation. Although higher concentration of nitrate nitrogen could promote the enrichment of denitrifying phosphorus accumulating bacteria, excessive nitrate nitrogen would also cause denitrifying bacteria and phosphorus accumulating bacteria competing for carbon source (Wang et al. 2018). The results showed that the system had a good phosphorus removal effect under Run2 and Run3 conditions, which indicated that the influent ratio had a certain impact on the phosphorus removal effect of the system. The appropriate influent ratio could reasonably distribute the carbon source in the influent to meet the requirements of anaerobic phosphorus release, aerobic phosphorus absorption and denitrifying phosphorus removal by phosphorus accumulating bacteria. It had good phosphorus removal effect in the process of domestic sewage treatment with a low C/N ratio

3.4 Influence of temperature on removal performance of COD, NH_4^+-N, TN, TP

The influence of temperature on the removal effect of COD, NH_4^+-N, TN, TP was shown in Fig. 9. It could be seen from Fig. 9 that temperature had a great influence on COD, NH_4^+-N, TN, TP. When the operating temperature raise from below 15℃ to 35℃, the removal rate of COD, NH_4^+-N, TN, TP in the system also kept increasing. The removal rate of COD increased from 78.6–87.4%; the removal rate of NH_4^+-N increased from 90.5–98.7%; the removal rate of TN increased from 71.1–77.6%and the TP increased from 82.7–89.3%. As could be seen from Fig. 9, the system also had a good removal effect for pollutants under low temperature conditions (12–15℃). The average concentration of COD, NH_4^+-N, TN, TP in the effluent were 37.8 mg/L, 4.2 mg/L, 12.7 mg/L and 0.4 mg/L, respectively, and the effluent quality could meet the effluent standard of National Class A discharge standard of China (GB 18918 – 2002), indicating that the system had a good low temperature resistance.

3.5 Effect comparison with traditional A^2O

It could be seen from Fig. 10 (b) that the concentration of COD, NH_4^+- N, TN and TP in the influent of traditional A^2O were 167.97 (± 1.4) mg/L, 44.55 (± 0.2) mg/L, 49.28 (± 1.6) mg/L, 49.28 (± 1.6) mg/L, 2.93 (± 0.5) mg/L, respectively, the concentration of COD, NH_4^+- N, TN and TP in the effluent were 26.37(± 1.33) mg/L, 7.59(± 0.56) mg/L, 17.64(± 1.47) mg/L, 0.82(± 0.06) mg/L, respectively, and the removal rates of COD, NH_4^+- N, TN and TP were 84.3(± 3.7)%, 82.9(± 5.6)%, 64.2(± 2.9)%, 72.1(± 5.9)%. While, in this study, the removal of pollutants by A1/O2/A3/A4/O5 process in the treatment of wastewater with low C/N ratio was improved to different degrees compared with the traditional A^2O. Under the same inlet condition, the concentrations of COD, NH_4^+- N, TN and TP in the effluent from A1/O2/A3/A4/O5 process were l21.32(± 1.51) mg/L, 0.60(± 0.38) mg/L, 12.76(± 1.15) mg/L, 0.34(± 0.04) mg/L, respectively. The removal rates of COD, NH_4^+- N, TN and TP were 87.3 (± 1.2)%, 98.7 (± 0.4)%, 74.1 (± 1.4)%, 88.1 (± 0.4)%, respectively. Compared with the traditional A^2O process, the removal rates of COD, NH_4^+- N, TN and TP were increased by 3.6%, 19.1%, 15.4% and 22.2%, respectively. Compared with traditional A^2O, this study has great advantages in pollutant removal. By adding suspended biological carrier into the system, the contradiction between nitrifying bacteria and phosphorus accumulating bacteria in traditional A^2O on sludge age (SRT) was solved, and the nitrification capacity of the system was strengthened. The phosphorus removal effect of the system was improved. The problem of carbon source competition between denitrifying bacteria and phosphorus accumulating bacteria was solved by the way of step-feeding and the carbon source allocation was optimized. The denitrifying and phosphorus removal performance of the system was strengthened. Therefore, compared with traditional A^2O, the removal of COD, NH_4^+- N, TN and TP and other pollutants had been greatly improved in this study. To sum up, the process could effectively remove pollutants on the basis of effective utilization of carbon source. Even in the treatment of wastewater with low C/N ratio, it also had a good effect of nitrogen and phosphorus removal. It hoped that this process could play a certain reference role in the upgrading and transformation of traditional wastewater treatment plants.

4. Conclusion

The system showed good performance of pollutant removal under optimal operating conditions. The process overcame the problem that phosphorus was difficult to be less than 0.5 $mg\cdot L^{-1}$ in the treatment of domestic sewage with low C/N ratio by traditional A^2O process. Under the low temperature of 12-15°C, the effluent quality could also meet the discharge standard. Compared with the traditional A^2O process, the process had better removal effect on pollutants, and the removal rates of COD, NH_4^+-N, TN and TP could be increased by 3.6%, 19.1%, 15.4% and 22.2% respectively. It was of great significance in the treatment of domestic sewage with low C/N ratio, which provided a new solution and beneficial enlightenment for the treatment of urban low C/N ratio sewage.

Declarations

Ethics approval and consent to participate. (Not applicable)

Consent for publication.

Written informed consent for publication was obtained from all participants.

Availability of data and materials

The data that support the findings of this study are available from the corresponding author upon reasonable request and I declare that [the/all other] data supporting the findings of this study are available within the article.

Competing interests

The authors declared that they have no conflicts of interest to this work. We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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Authors' contributions

ZDW modified the paper; ZDW and ZJT wrote the manuscript; ZJ reviewed the rationality of the structure of the paper; ZMK, WML and ZSH analyzed the data of the paper.

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Figures

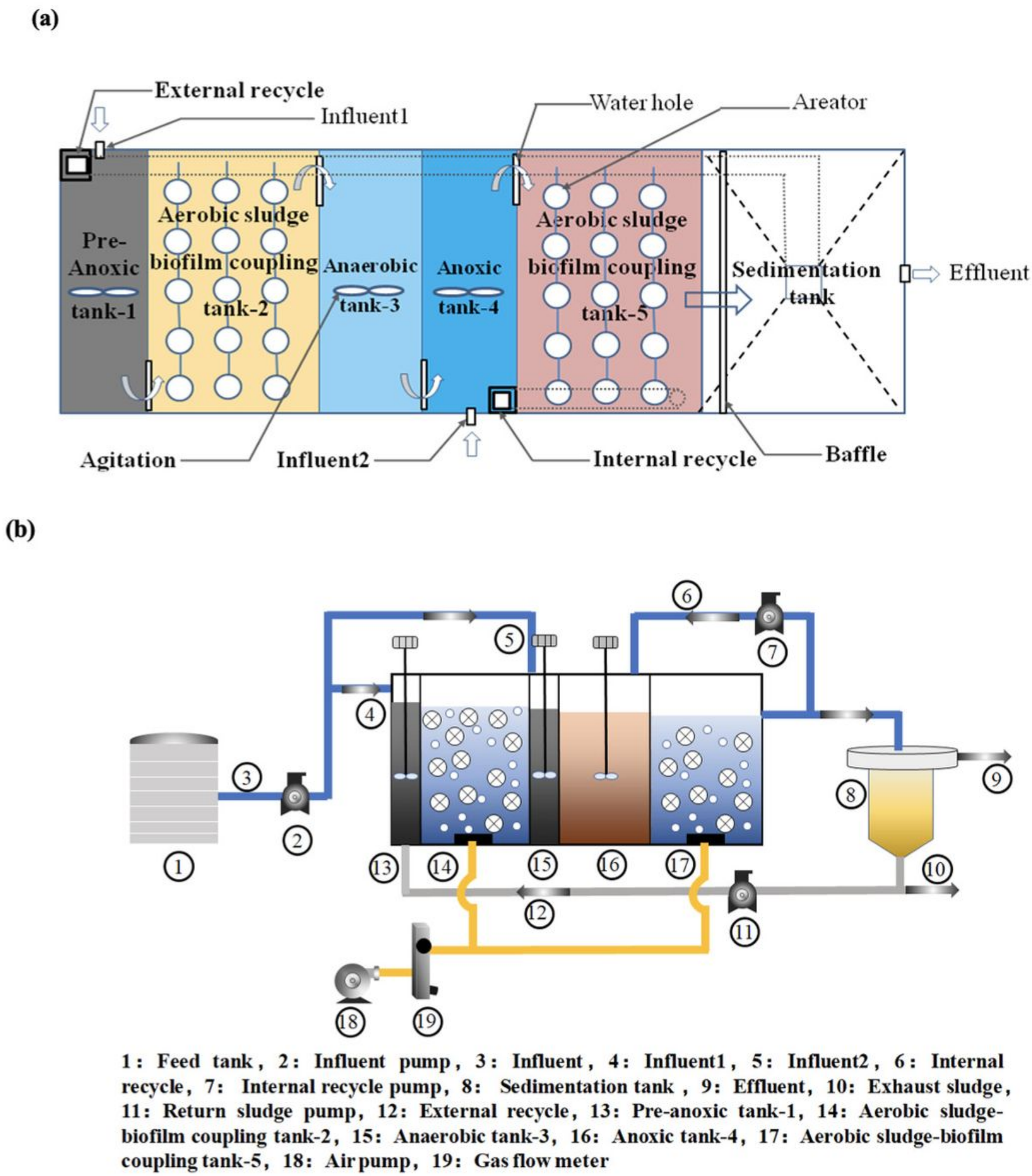

Figure 1

(a) Schematic plan diagram of the A1/O2/A3/A4/O5 sludge-membrane coupling reactor. (b) New A1/O2/A3/A4/O5 sludge-membrane coupling process flow chart for multi-point water

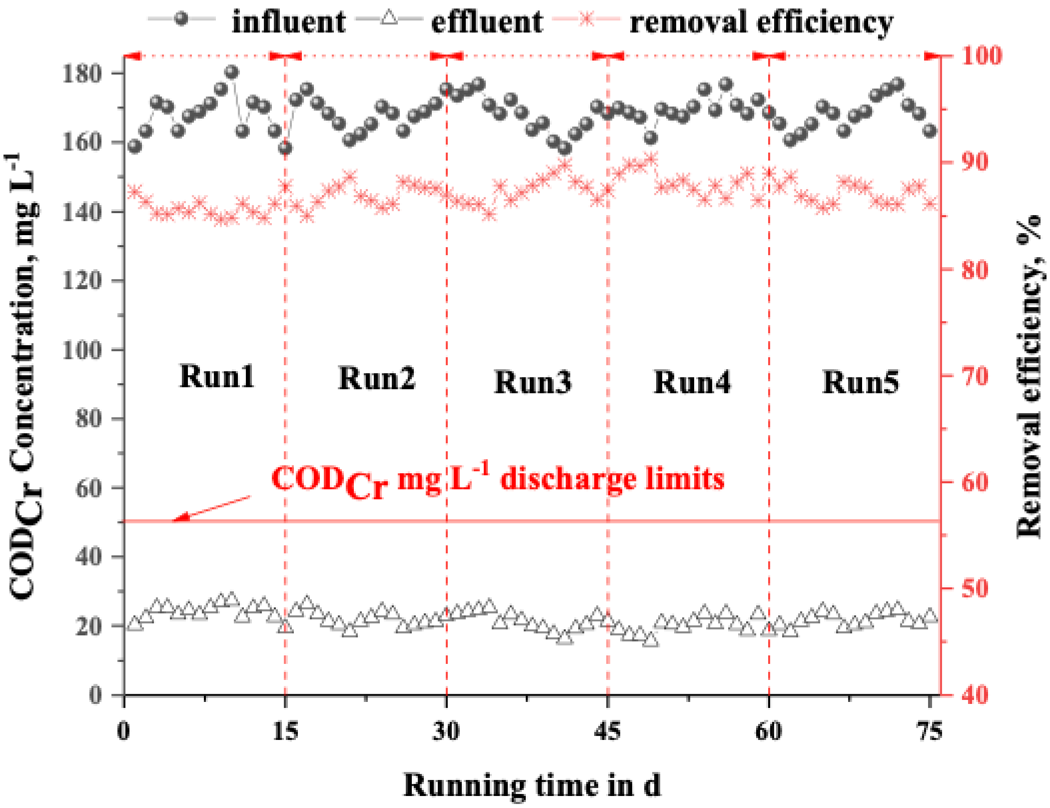

Figure 2

Influence of different inflow ratio on removal efficiency of COD

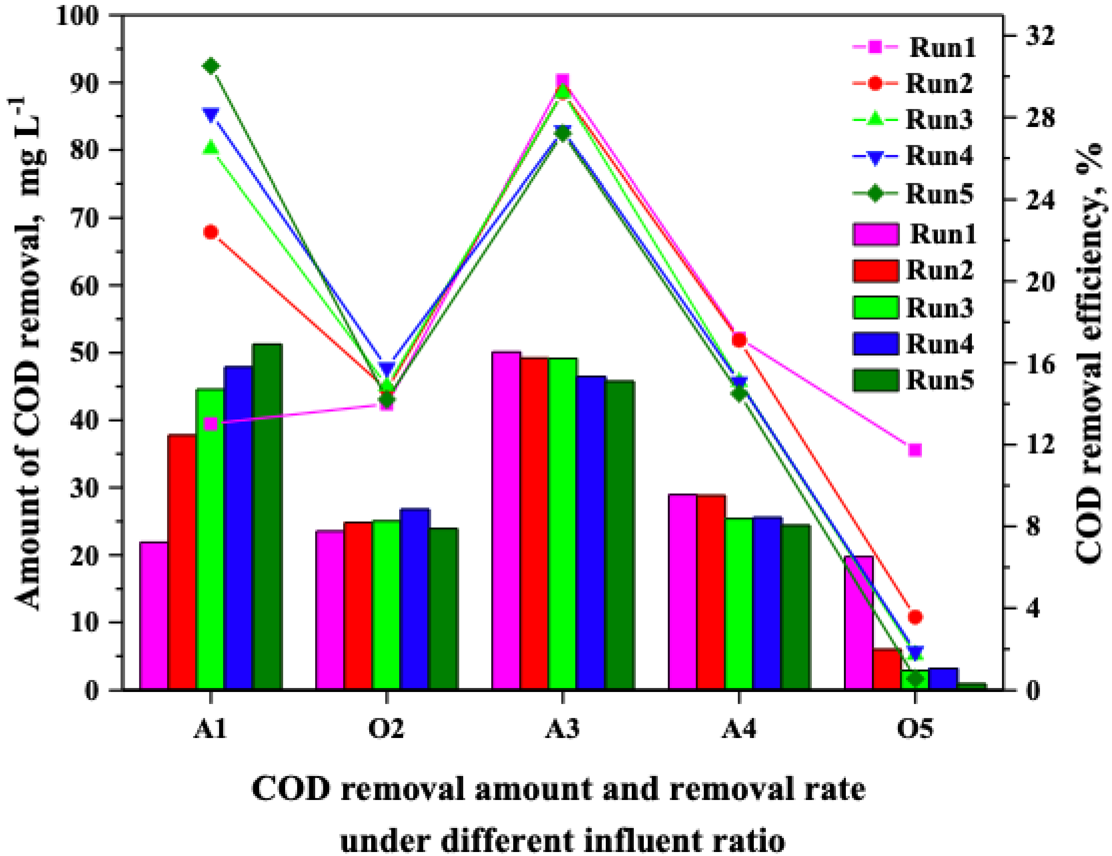

Figure 3

Removal of COD along with Different inflow ratio

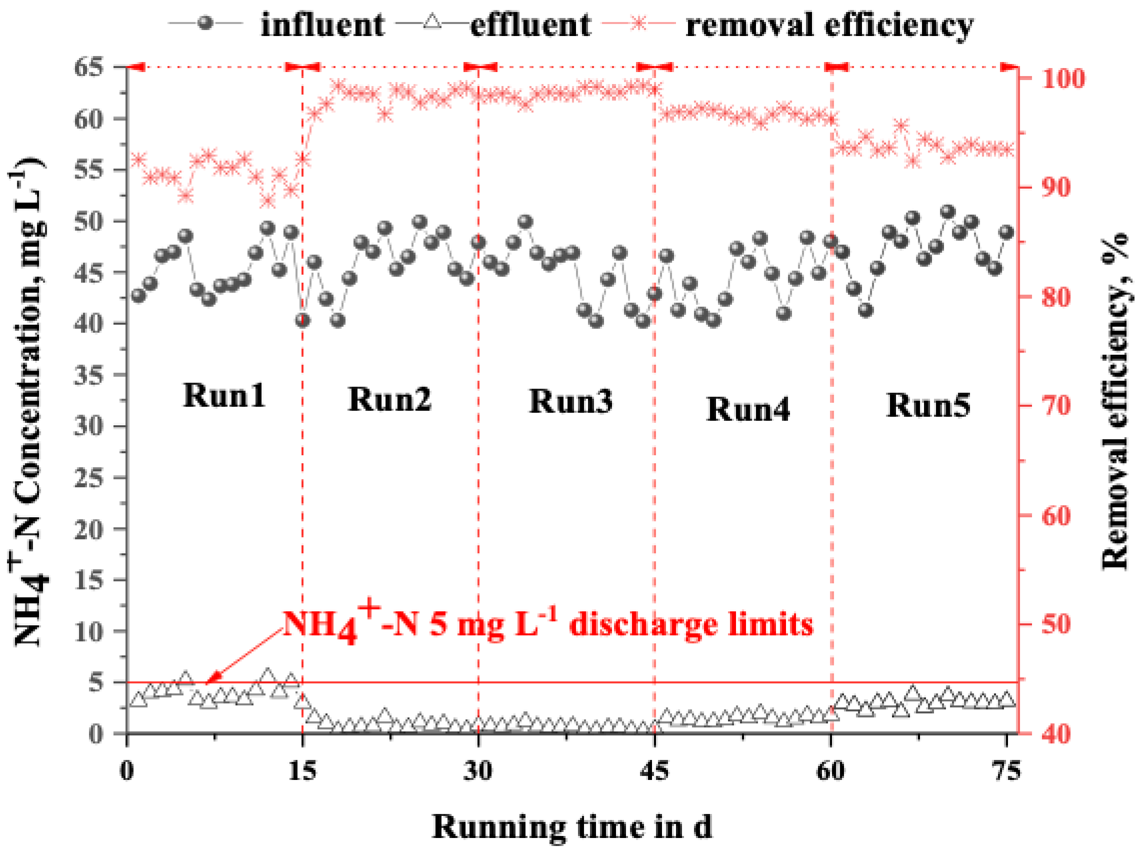

Figure 4

Removal effect of NH4+-N with different inflow ratio

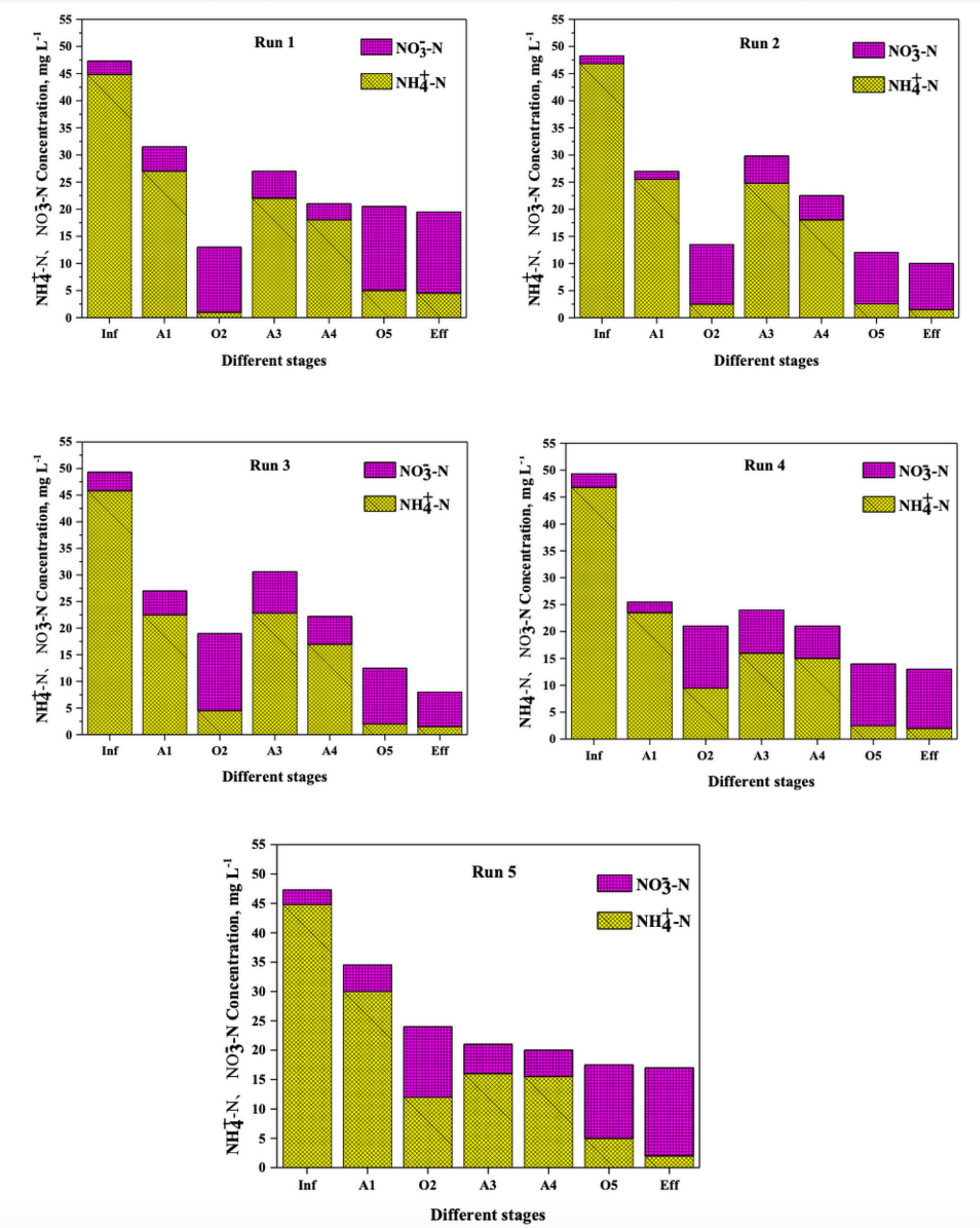

Figure 5

NH4+-N、NO3–N concentration in different stage under different operational conditions

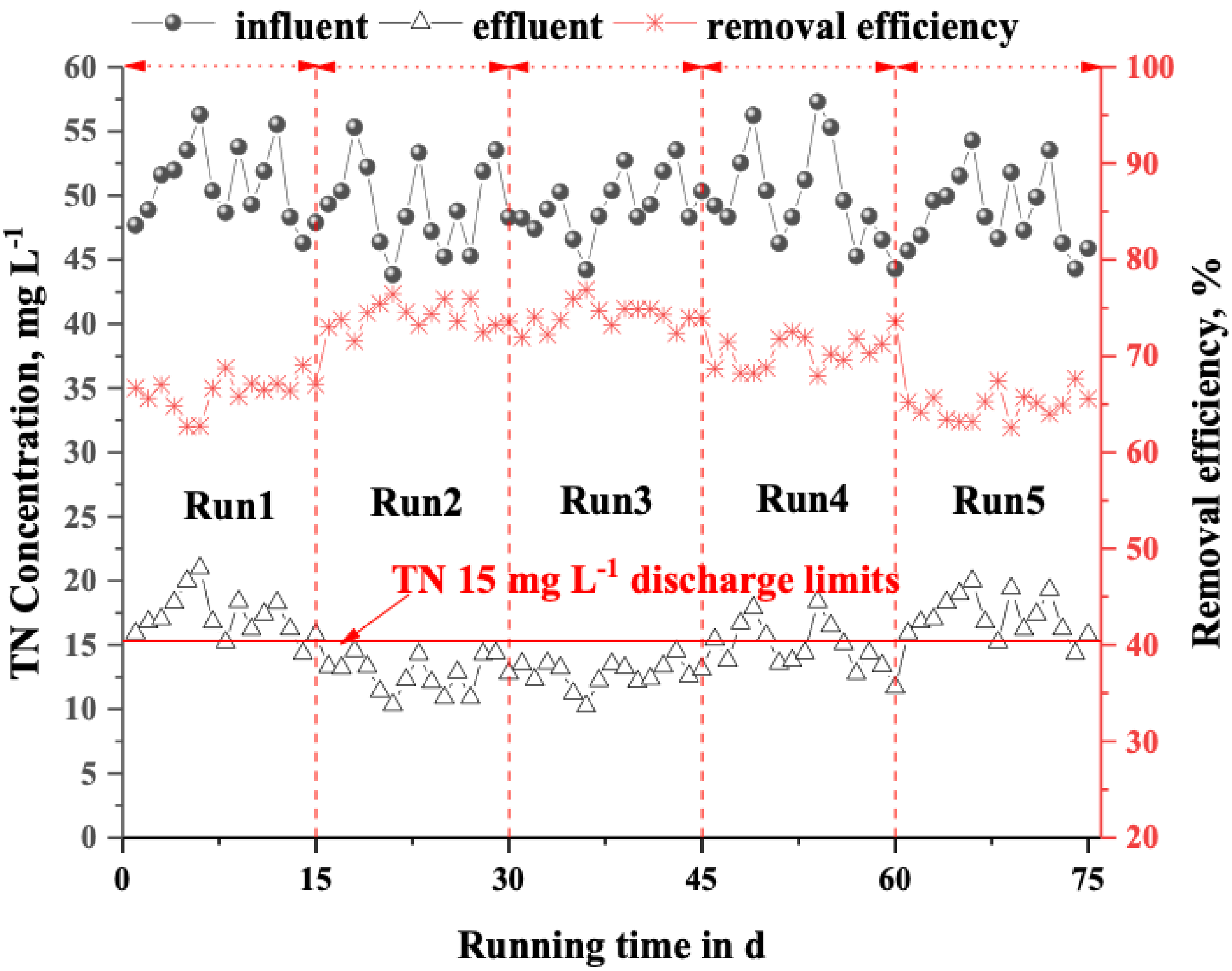

Figure 6

Removal efficiency of TN with different inlet water ratio

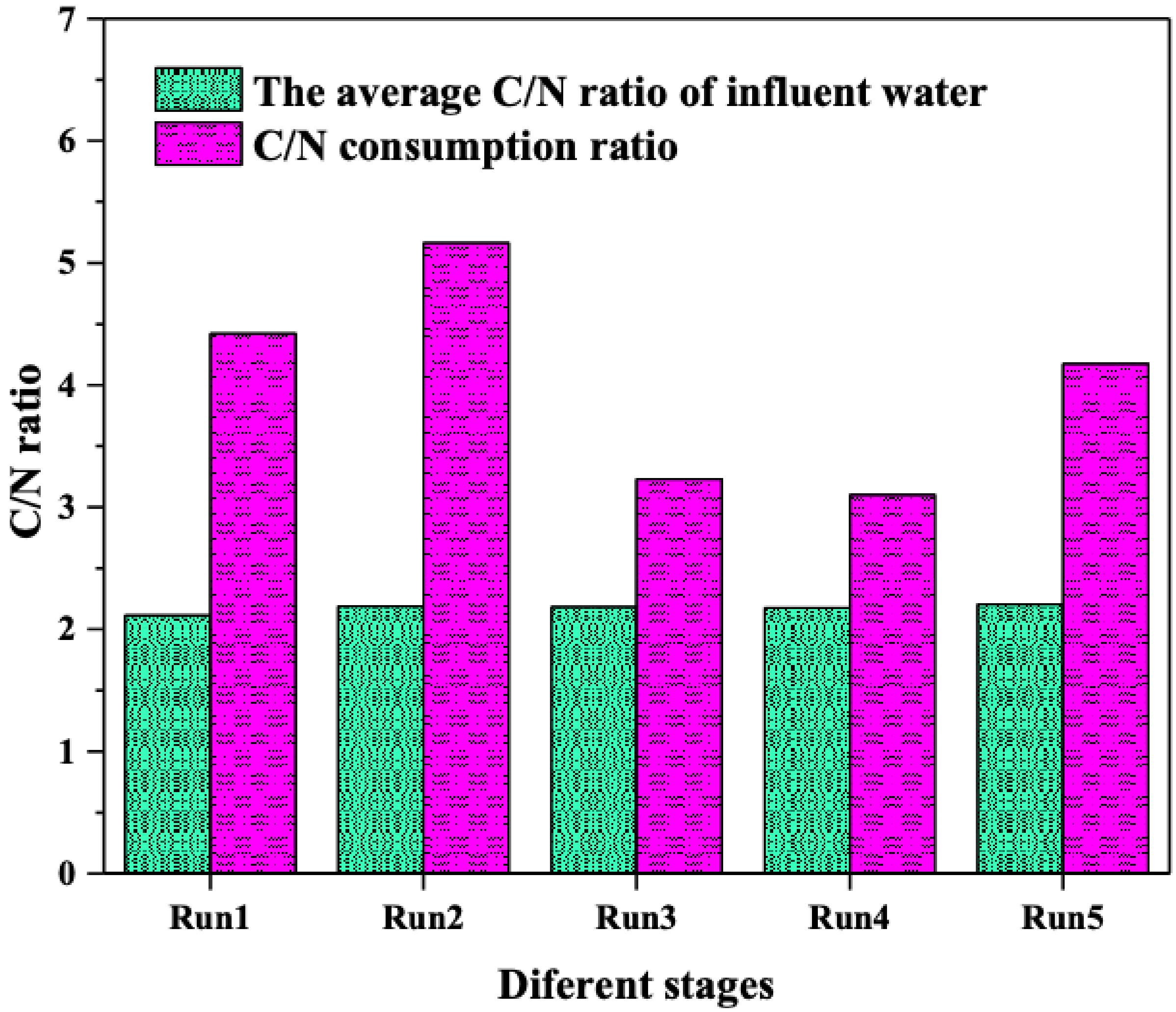

Figure 7

The C/N ratio of influent and its consumption in section A under various operating conditions

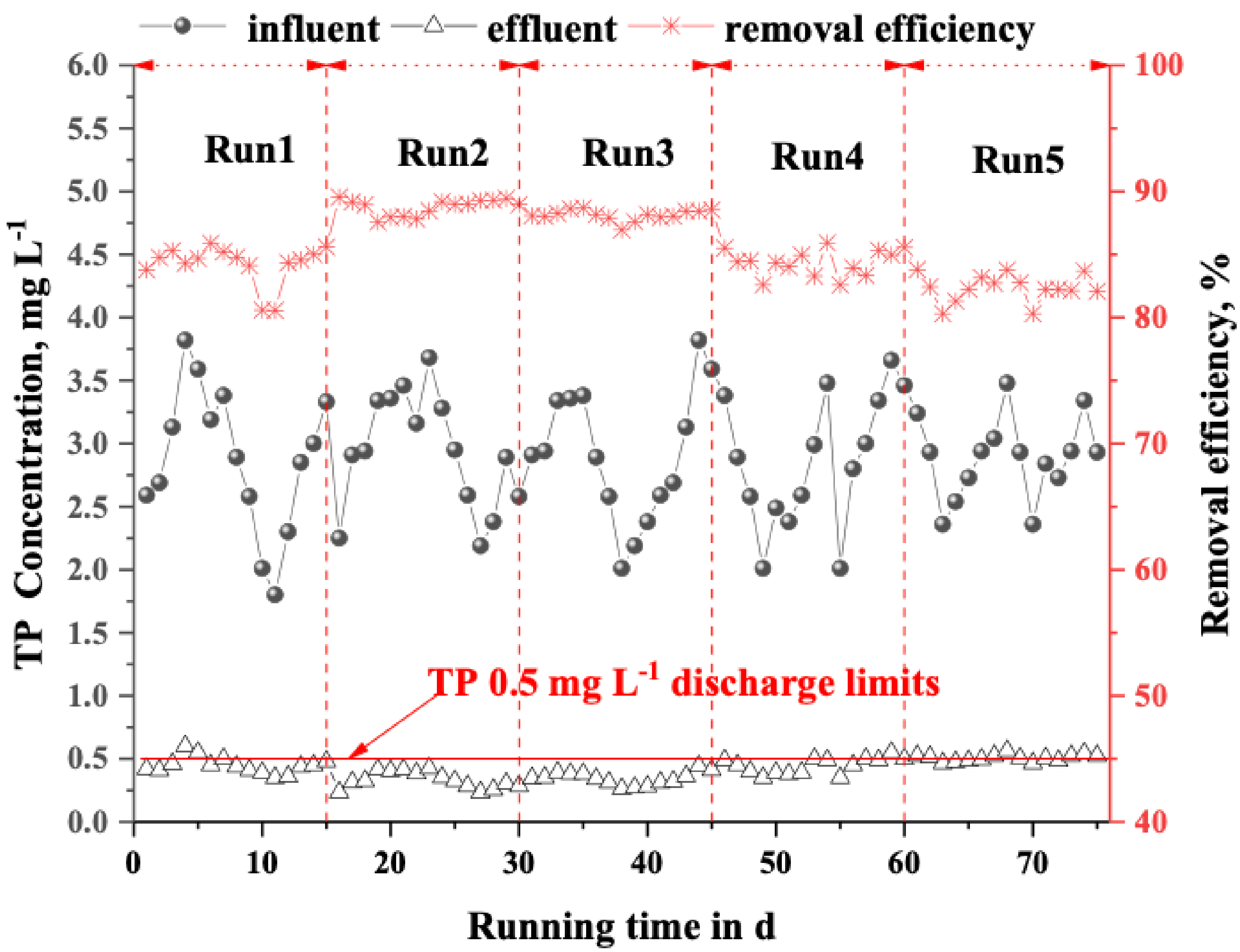

Figure 8

The removal effect of TP with different inflow ratio

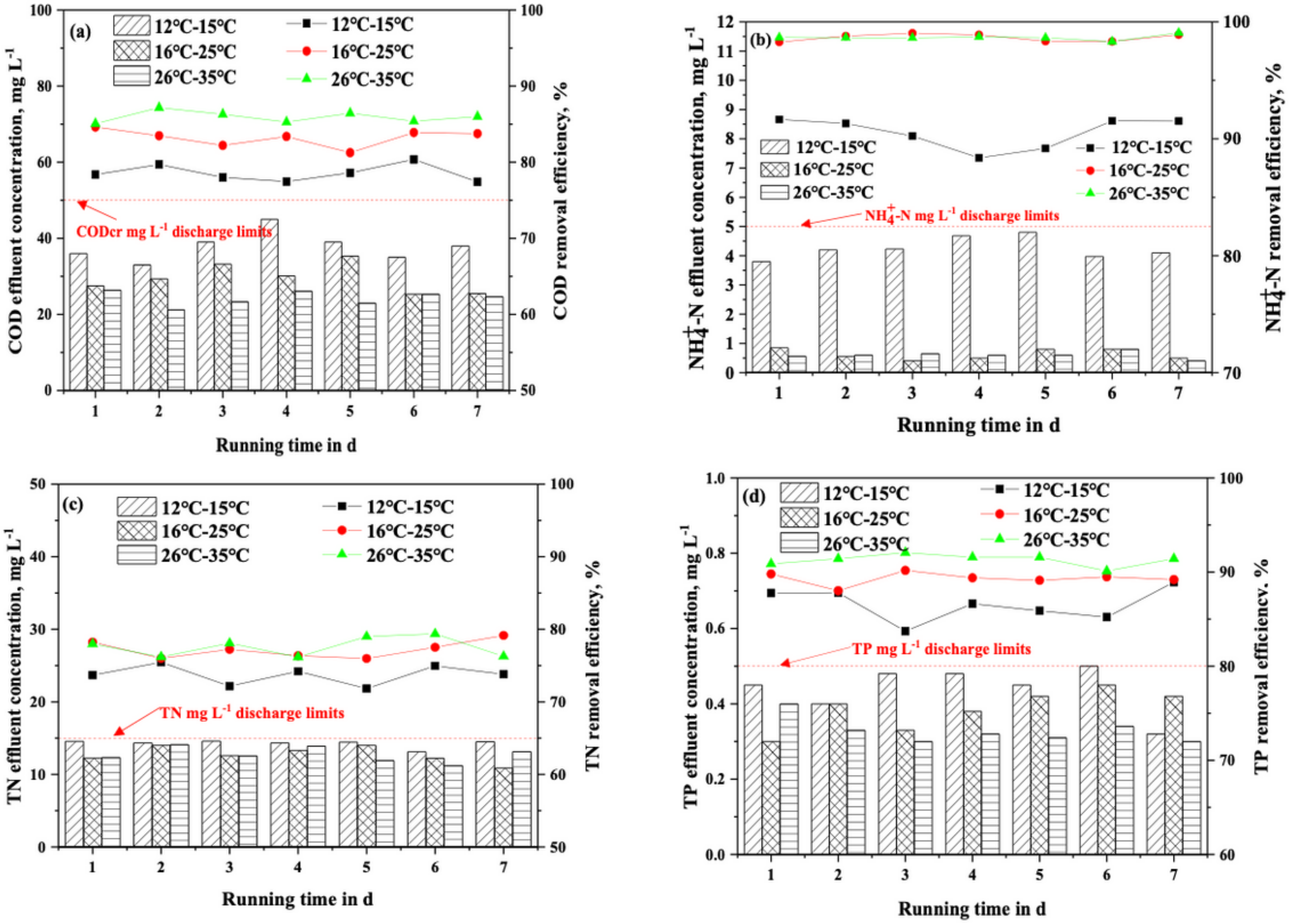

Figure 9

(a) The effect of temperature on COD removal effect, (b) the effect of temperature on NH4+-N removal effect, (c) the effect of temperature on TN removal effect, and (d) the effect of temperature on TP removal effect

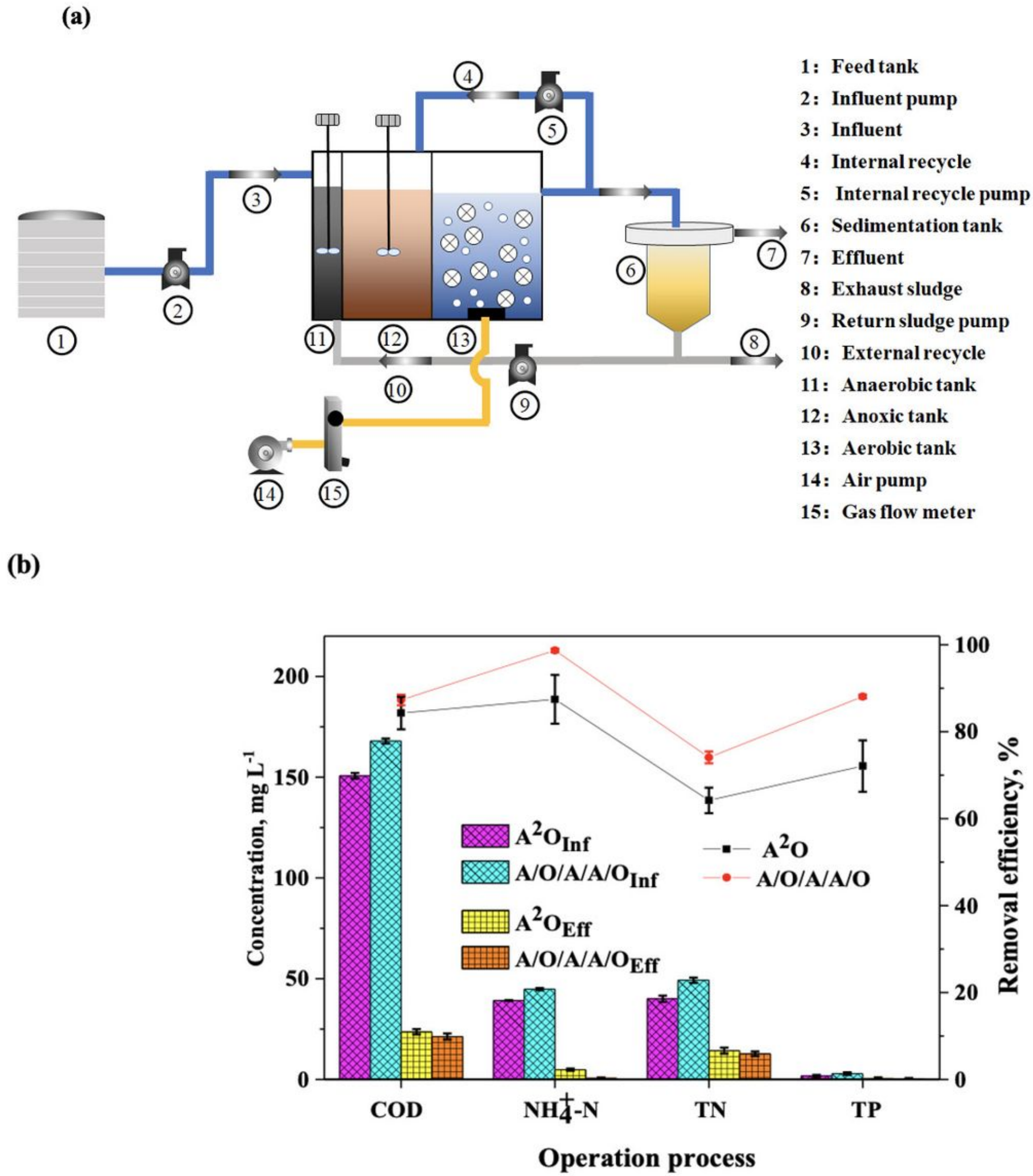

Figure 10

(a) Traditional A2O process flow chart. (b) Comparison between A1/O2/A3/A4/O5 process and traditional A2O in terms of pollutant removal