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Combined treatment of domestic wastewater with landfill leachate by using A^2/O process

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ABSTRACT

A set of anaerobic–anoxic–aerobic (A^2/O) bioreactor system was designed and used to treat domestic wastewater mixed with landfill leachate in Datansha Sewage Treatment Plant in Guangzhou, south China. The optimal mixing proportion of combined treatment of domestic wastewater with landfill leachate and the optimal operating conditions for the removal efficiencies of nitrogen by using Taguchi orthogonal array test was conducted to evaluate the influence of parameters. The results showed that: the optimal volume ratio of landfill leachate and domestic wastewater in the A^2/O process was 1:500. The average removal efficiencies of NH₄⁺-N, TN and COD was achieved to be 96.5%, 61.0% and 81.7%, respectively in the case of the hydraulic retention time (HRT) of 11 h, dissolved oxygen (DO) of 3 mg L⁻¹, the mixed-liquid return ratio (r) of 200% and sludge return ratio (R) of 80% in the case of the confirmatory experiment. The pilot scale (3.8 m³) investigation results were applied in the large-scale (220,000 m³/d) combined treatment of sewage wastewater with landfill leachate in Guangzhou Datansha Domestic Sewage Wastewater Treatment Plant. The removal efficiencies of COD, NH₄⁺-N, T-N and T-P were 82.65%, 92.69%, 57.10% and 76.55%, respectively.

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1. Introduction

Landfilling, compared to other technologies such as incineration and composting, is a common way to dispose of solid waste. It is reported that about 90% of the municipal solid waste (MSW) is disposed of in landfills in China. Landfill leachate, comes from the waste degradation in landfill sites, especially those from aged landfill sites, has been a challenge for complete treatment economically by both biological or combined treatment with physico-chemical methods. The focuses of old landfill leachate treatments are the significant amount of ammonium, high organic matter contents with non-biodegradable organic substances, such as humic-type constituents, etc. As a result, the surface water pollution caused by landfill leachate may appear to be significant. It was reported that the surface water which was 4 km far from a landfill site was seriously polluted [1]. Moreover, the quality of groundwater over 60 m depth was reported to be influenced by landfill leachate. The removal of organic matters (measured as chemical oxygen demand

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COD and biological oxygen demand BOD) and ammonium nitrogen (NH₄⁺-N) in landfill leachate are essential before it is discharged into natural environments.

There were mainly two methods for the combined treatment of landfill leachate. One was the physical-chemical treatments of landfill leachate, such as coagulation flocculation [2], chemical precipitation [3], ammonium stripping [4], membrane filtration [5] and activated carbon adsorption [6], etc. Another was biological methods including anaerobic treatment by upflow anaerobic sludge blanket (UASB) [7], aerobic treatment by sequencing batch reactor (SBR) [8] and membrane bioreactor [9]. Furthermore, the post-treatment of physico-chemical method, such as reverse osmosis (RO) and Fenton oxidation [10] methods were the necessary options for the subsequent treatment of biological (both anaerobic and aerobic including anoxic) processes for the treatment of landfill leachate. RO has been widely and successfully used in secondary treatment of leachate effluent from biological processes in many countries owing to its ability to retain both organic and inorganic contaminants [11-14]. However, some 30% of total concentrated leachate effluent could be generated after treatment by using RO technology in engineering scale [14]. It was very expensive and difficult to perform the further treatment and purification [15-17] for the concentrated leachate effluent generated by RO process because of the large amounts of recal-

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citrant and non-biodegradable organic substances it contained [18–20].

Guangzhou is a metropolitan in South China with the population of over 13 million. The average daily municipal solid wastes in 2008 were estimated to be 9476 t/d. However, there is only one landfill site, Xingfeng Municipal Solid Waste Sanitary Landfill (XFL), is available for the landfilling of solid wastes. The designed capacity for landfilling solid wastes was 2500 t/d before 2005, and the Landfill Leachate Treatment Plant in XFL was designed on the basis of the above-mentioned capacity, by using combined treatment of UASB and SBR together with RO processes [17]. The effluent of leachate from the Landfill Leachate Treatment Plant has been kept to meet the strict discharge limits. However, the capacity designed for the landfill leachate treatment was much lower than the amount of leachate generated because of the increase of solid wastes accepted by XFL. Therefore, the combined treatment of landfill leachate with sewage wastewater was recommended in Guangzhou. As a result, Guangzhou Datansha Domestic Sewage Wastewater Treatment Plant (total capacity: 550,000 m³/d) with anaerobic-anoxic-aerobic (A²/O) process (capacity: 220,000 m³/d) was recommended for the combined treatment of landfill leachate generated from XFL in recent years.

Biological nitrogen removal is the major concern for the mixture and treatment of domestic sewage wastewater with landfill leachate because of the high concentration of ammonium in leachate. The anaerobic–anoxic–oxic (A^2/O) process is a good option for achieving complete biological nitrogen removal by simultaneous/coupled nitrification and denitrification processes. The mechanisms of nitrification and denitrification can be explained by the stoichiometric reactions derived on the basis of electron flow balance, as listed by Eqs. (1)–(4) [21–23].

• Nitrification (nitrite pathway):

$$\frac{10+3f_s}{60}\mathrm{NH_4}^+ + \frac{1-f_s}{4}\mathrm{O_2} + \frac{f_s}{4}\mathrm{HCO_3}^-$$
$$= \frac{f_s}{20}\mathrm{C_5H_7O_2N} + \frac{1}{6}\mathrm{NO_2}^- + \frac{5-3f_s}{15}\mathrm{H}^+ + \frac{10+9f_s}{60}\mathrm{H_2O} \qquad (1)$$

• Nitrification (nitrate pathway):

$$\frac{5+2f_s}{40}NH_4^+ + \frac{1-f_s}{4}O_2 + \frac{f_s}{4}HCO_3^-$$
$$= \frac{f_s}{20}C_5H_7O_2N + \frac{1}{8}NO_3^- + \frac{5-4f_s}{20}H^+ + \frac{5+6f_s}{40}H_2O$$
(2)

where f_s is the fraction of electron donors coupled to cell (C₅H₇O₂N) synthesis.

Denitrification (nitrate pathway):

$$\frac{1}{4}(\text{COD}) + \frac{28 - 23f_s}{140}\text{NO}_3^- = \frac{f_s}{28}\text{C}_5\text{H}_7\text{O}_2\text{N} + \frac{1 - f_s}{10}\text{N}_2 \uparrow + (\text{others})$$
(3)

• Denitrification (nitrite pathway):

$$\frac{1}{4}(\text{COD}) + \frac{26 - 23f_s}{78}\text{NO}_2^- = \frac{f_s}{26}\text{C}_5\text{H}_7\text{O}_2\text{N} + \frac{1 - f_s}{6}\text{N}_2 \uparrow + (\text{others})$$
(4)

The amount of chemical oxygen demand (COD) required for denitrifications by nitrate or nitrite to nitrogen gas varies in an range associated with the yield of microorganisms (Y), as Eqs. (5) and (6) shown, respectively.

$$\frac{\text{COD}}{\text{N-NO}_3} = \frac{2.86}{1 - 1.628Y}$$
(5)
COD 1.714 (6)

$$\frac{\text{COD}}{\text{N}-\text{NO}_2} = \frac{1.714}{1 - 1.628\text{Y}} \tag{6}$$

The amount of COD required by denitrification of nitrate to N₂ is associated with the yield of microorganisms ($Y_{x/c}$) or f_s .

Nevertheless, a number of affective factors such as the ratio of leachate to sewage waste water, the concentration of dissolved oxygen (DO), the hydraulic retention time (HRT), the mixed-liquid return ratio (r), the sludge return ratio (R) as well as the removal efficiencies of COD, NH₄⁺-N, total nitrogen (T-N), need to be considered for achieving high performance and the optimal operation of A²/O process for the combined treatment of sewage wastewater with leachate. However, it is difficult to find any related published report for consideration. Therefore, in this study, an A²/O bioreactor (3.8 m³) system simulating the available engineering scale A²/O system (220,000 m³/d) was designed and used for an experimental scale investigation of the mixture treatment. The design of experimental methodology using Taguchi orthogonal array was applied to evaluate the influence of four factors (HRT, DO, *r*, *R*).

The objective of this paper was to evaluate the feasibility and optimal mixture proportion for combined treatment of landfill leachate and domestic wastewater in engineering scale properly.

2. Materials and methods

2.1. Experimental apparatus

The experimental apparatus consisted of an A²/O bioreactor, a regulating tank, three submersible pumps, an air compressor and a setting tank (Fig. 1). The bioreactor was made of steel-plate, with total volume of 3800 L, have four compartments.

The first compartment was typically operated as an anaerobic zone, followed by an anoxic zone, with the remaining two compartments with separate aeration control as aerobic zones. The volume ratio of anaerobic, anoxic and aerobic zone was 1:1.6:5. The settling tank was also made of steel-plate, with a volume of 900L capacity. The effective depth of water was 1.1m. The on-line sensors for measuring DO and temperature was installed in the system. The mixed-liquid return ratio, sludge return ratio and influent flow can be adjusted by the submersible pumps.

2.2. Microorganisms and substrate

Activated sludge added to the experimental bioreactor was obtained from the concentrated sludge vessel at Datansha Sewage Treatment Plant. The mixed liquid suspended solids (MLSS) were about $12,000 \text{ mg L}^{-1}$.

The influent consisted of domestic wastewater mixed with landfill leachate at proper ratios. The leachate used in the experiment was obtained from the XFL landfill site, which received about 7500 t of domestic solid wastes per day. Leachate samples were collected, and stored in a regulating reservoir at Datansha Sewage Wastewa-



I.Anaerobic II.Anoxic III.Aerobic1 IV.Aerobic2 V.Settling tank 1.influent 2.stirrer 3.diffuser 4.air pressure 5.mixed-liquid return 6.sludge return 7.effluent

Fig. 1. Schematic diagram of the A²/O process. (I) Anaerobic; (II) anoxic; (III) aerobic1; (IV) aerobic2; (V) Settling tank. (1) Influent, (2) stirrer, (3) diffuser, (4) air pressure, (5) mixed-liquid return, (6) sludge return, and (7) effluent.

Table 1

Characteristics of domestic sewage and landfill leachate (mg L⁻¹).

	Domestic sewage	Landfill leachate
BOD ₅	70–150	4000-12000
COD	100-300	8000-20000
NH4 ⁺ -N	15-35	1500-3800
T-N	20-40	2000-8000
NO ₃ ⁻ -N	0.0	4–20
NO ₂ N	0.0	2-12
T-P	1.5–2.8	0.86-56.0

ter Treatment Plant. The characteristics of the landfill leachate and domestic wastewater are listed in Tables 1 and 2.

According Table 1, it was not difficult to find that landfill leachate have a much more high concentrations of organic compounds and nitrogen, which would result in an impulse loading to the operation of the A^2/O process for treating domestic wastewater if the mixture was not manipulated properly. Based on Table 2, the mass proportion of landfill leachate, such as COD, BOD₅, SS, T-N, NH₄⁺-N and NOx-N, in the mixture of landfill leachate with sewage wastewater accounts for 21.2%, 13.9%, 13.9%, 18.4%, 18.4% and 100%, respectively. However, the influent concentrations of nitrite and nitrate are not very high.

2.3. Analytical methods

The measurements of influent and effluent COD, NH_4^+-N , T-N, nitrite nitrogen (NO_2^--N), nitrate nitrogen (NO_3^--N), and alkalinity were performed on the basis of the standard methods [24]. MLSS was determined by drying the sludge sample at 105 °C for 24 h. Dissolved oxygen (DO) was measured with a DO instrument (YSI-5000, YSI Company, America). Temperature and pH were measured with ordinary mercury thermometer and pH meter (pHS-25, Jingke Company, China), respectively.

Design of experiments (DOE) methodology by Taguchi orthogonal array (OA) was used to facilitate the optimal operation of A^2/O system by a set of independent variables (factors) which could make the process performance insensitive to variation by proper design of parameters. As Table 3 shown, there are four individual factors, such as HRT, DO, *r* and *R*, etc., are very important for being considered in this study.

Optimized levels of variables were designed according to the Taguchi fractional design method that considers only the important combined effects of the parameters in the experimental plan [25]. Performance was measured by the deviation of a characteristic from the target value and a less function [L(y)] was developed for the deviation [26]:

$$L(y) = k(y - m)^2$$
(7)

where 'k' denotes the proportionality constant, 'm' represents the target value and 'y' is the experimental value obtained for each trial. In case of 'bigger is better' quality characteristics, the loss function can be written as

$$L(y) = k\left(\frac{1}{y^2}\right) \tag{8}$$

Table 3	
Factors in orthogonal array experimental	design.

Level Factor A-HRT(h) B-DO $(mg L^{-1})$ C-r(%) D-R (%) 1 11 2 100 60 2 80 3 200 9 3 7 4 300 100

3. Results and discussion

3.1. Optimal mixture ratio of landfill leachate to domestic wastewater

The practical mixture ratio of landfill leachate and domestic wastewater in Datansha Sewage Treatment Plant has been about 1:700 since 2005 without experimental study because of the urgent situation of treatment of landfill leachate in Guangzhou XFL. The investigation of this study was required and started for a better and optimal operation of the mixture of landfill leachate with sewage wastewater in order to treat landfill leachate as possible as it could.

The designed mixture ratios of landfill leachate to sewage wastewater were 1:250, 1:350, 1:500 and 1:700, respectively. All the operating conditions were performed according to the Datansha Sewage Treatment Plant: the temperature was kept $24 \,^{\circ}$ C, mixed-liquid return ratio was 200%, sludge return ratio was 80%, HRT was 9 h, DO in the aerobic tank was $3 \,\text{mg L}^{-1}$, MLSS was $3000 \,\text{mg L}^{-1}$ and SRT was 12 days. The operation times for each case was 7 days, the total operation time was 35 days. The results of removal efficiencies of NH₄⁺-N, T-N and COD at different mixture ratios are shown in Table 4.

All the influent and effluent concentrations of NH₄-N, T-N and COD with addition of landfill leachate were higher than the control one (in Table 4). The concentrations of NH₄⁺-N, T-N and COD increased with the increase of mixture ratios. The influent concentrations of NH4+-N, T-N and COD increased 36.5%, 30.8% and 37.5% respectively in comparison with the control one in the case of the mixture ratio of 1:500 (7 days operation), while the effluent concentrations of NH₄⁺-N, T-N and COD increased 750%, 113.7% and 75% respectively in comparison with the control one in this case. Correspondingly, the removal efficiencies of NH4⁺-N, T-N and COD increased 4.2%, 16.2% and 4.9% respectively. However, most of the effluent concentration values of NH4+-N, T-N and COD listed in Table 4 are below the limits (level 1B) of China Discharge Standard (COD \leq 40 mg L⁻¹, NH₄⁺-N \leq 8 mg L⁻¹, T-N \leq 20 mg L⁻¹, T- $P \le 1.0 \text{ mg L}^{-1}$) for sewage wastewater except for the influent ratio at 1:250. According to the principle of mixing landfill leachate with domestic wastewater as much as possible while meeting the effluent standard, the higher mixture ratio of landfill leachate and domestic wastewater was 1:500 which could enhance the treatment of landfill leachate from 315 t/d to 440 t/d.

3.2. Influence of individual factors

According to the above-described experimental performance, the A^2/O process efficiency has been found to be very much depen-

Table 2

Characteristics of mixture wastewaters at volume mixture ratio of 1:500.

	COD	BOD ₅	SS	T-N	NH4 ⁺ -N	NO_3^N	NO_2^N
Landfill leachate(mg L^{-1})	16191.1	5667.5	5280.8	5096.3	3691.8	6.0	3.0
Sewage wastewater (mg L ⁻¹)	121.1	71.2	65.6	45.1	32.7	0.0	0.0
Ratio of features	133.7	79.6	80.5	113.0	112.9		
Pollutants in leachate (kg/m ³)	16.2	5.7	5.3	5.1	3.7	0.006	0.003
Pollutants in sewage wastewater (kg/(500 m ³))	60.6	35.6	32.8	22.6	16.4	0.0	0.0
Mass proportion of landfill leachate in the mixture (%)	21.2	13.8	13.9	18.4	18.4	100	100

Table 4	
Removal efficiency of NH $_4^+$ -N, T-N and COD at different ratios (mean data of 7 days for each case)	

Ratios of leachate:	NH4 ⁺ -N			T-N			COD	COD		
domestic sewage	Influent (mg L ⁻¹)	Effluent (mg L ⁻¹)	Removal efficiency (%)	Influent (mg L ⁻¹)	Effluent $(mg L^{-1})$	Removal efficiency (%)	Influent (mg L ⁻¹)	Effluent $(mg L^{-1})$	Removal efficiency (%)	
1:700	31.8	0.6	98.3	34.5	13.1	62.1	108.0	21.6	79.9	
1:500	34.4	1.7	95.0	37.4	15.6	58.3	118.0	26.6	77.4	
1:350	36.3	5.1	85.9	39.5	17.3	56.3	134.7	30.4	77.3	
1:250	40.3	9.1	77.3	43.1	20.2	53.1	150.0	33.1	77.9	
0 (control)	25.2	0.2	99.2	28.6	7.3	74.5	85.8	15.2	82.3	
Difference between Control one and the mixture (1:500)	36.5%	750%	4.2%	30.8%	113.7%	16.2%	37.5%	75%	4.9%	

dent on the selected conditions. The effect of the factors is DO concentration in aerobic tank, the HRT, the mixed-liquid return ratio (r) and the sludge return ratio (R). The focus was on the removal efficiency of nitrogen and the test mixture ratio of landfill leachate to sewage wastewater was kept at 1:500 with the operation period of time 7 days for each run.

3.2.1. Effect of DO on nitrogen removal

The effect of DO on the removal efficiency of nitrogen was investigated at DO concentrations of 2 mg L^{-1} , 3 mg L^{-1} and 4 mg L^{-1} , respectively. Other experimental conditions were kept as following: the temperature $T=28 \circ \text{C}$, r=200%, R=80%, HRT=9 h. The removal efficiencies of NH₄-N and T-N at different DO are listed in Table 5.

The removal of ammonium was rather high when DO ranged from 2 mg L^{-1} to 3 mg L^{-1} , all removal efficiencies were more than 90%, the A²/O process achieved 97.4% and 97.5% of ammonium removal when the DO concentrations were 2 mg L^{-1} and 3 mg L^{-1} , respectively.

However, the removal efficiency of T-N was different. The removal efficiency of T-N increased with the increase of DO from 2 mg L^{-1} to 3 mg L^{-1} , but when the DO was further increased to 4 mg L^{-1} , the removal efficiency of T-N decreased from 58.2% to 42.5%. The A²/O process achieved the highest removal efficiency (58.2%) of T-N in the case of DO 3 mg L⁻¹ in the aerobic zone. In the aerobic zone, one of the main biochemical reaction was nitrification, as Eqs. (1) and (2) shown, nitrate concentration accumulated with the decrease of ammonium concentration on the condition of DO was higher than 2 mg L^{-1} . The removal efficiencies of NH₄⁺⁻N and T-N in case of DO 3 mg L⁻¹ showed that this A²/O process achieved better both in nitrification and denitrification. Nevertheless, as we know, the total nitrogen means the sum of total Kjeldahl

nitrogen (organic and ammonia nitrogen) nitrate–nitrogen and nitrite–nitrogen, the removal efficiency of T-N depends on the removal of nitrate by denitrification, as Table 5 shown, the effluent concentrations of NO₃-N accounted for 63.1%, 79.5% and 86.4% of T-N, respectively. It was beneficial for denitrification to control low concentration of DO. When the DO increased to 4 mg L^{-1} , it had a negative impact on the performance in the anoxic tank, by inhibiting the activity of denitrifying bacteria. So the DO in the aerobic tank must be controlled within an optimal range for high removal efficiency of T-N.

3.2.2. Effect of HRT on nitrogen removal

The effect of HRT on the removal efficiency of nitrogen was investigated in case of HRT of 7 h, 9 h and 11 h, respectively. The experimental conditions were kept as: temperature $T = 28 \,^{\circ}$ C, r = 200%, R = 80%, DO = 3 mg L⁻¹. The results are listed in Table 6, which demonstrated the variation of removal efficiencies of NH₄⁺⁻N and T-N at each HRT. Nitrification and denitrification were enhanced when the HRT was increased from 7 h to 11 h. Better nitrogen removal was achieved in case of HRT greater than 9 h. The effluent concentrations of NO₃-N accounted for 80.6%, 79.5% and 51.2% of T-N, respectively. However, the effluent concentration of T-N revealed that higher removal efficiency of T-N was achieved in the case of higher HRT because of the higher removal of ammonium by nitrification and denitrification.

3.2.3. Effect of r on nitrogen removal

The effect of *r* (the mixed-liquid return ratio) on the removal efficiency of nitrogen was investigated in case of *r* equaled to 100%, 200% and 300%, respectively. The experimental conditions were: temperature $T = 28 \degree C$, HRT = 9 h, R = 80%, DO = 3 mg L⁻¹. The removal efficiencies of NH₄-N and TN at each *r* is shown in Table 7.

Table 5

Removal efficiency of NH4⁺-N, NOx-N and T-N at different DO concentrations (mean data of 7 days for each case).

$DO(mgL^{-1})$	NH4 ⁺ -N (m	$NH_4^+-N(mgL^{-1})$			$NO_2^{-}-N (mg L^{-1})$ $NO_3^{-}-N (mg L^{-1})$		(mg L ⁻¹)	T-N (mg L ⁻¹)		
	Inf.	Eff.	RE (%)	Inf.	Eff.	Inf.	Eff.	Inf.	Eff.	RE (%)
2.0	33.7	3.3	90.2	0.0	2.6	0.0	11.8	36.3	18.7	48.6
3.0	30.6	0.8	97.4	0.0	0.4	0.0	11.6	34.9	14.6	58.2
4.0	29.2	0.7	97.5	0.0	0.7	0.0	16.4	33.1	19.0	42.5

Inf.: influent; Eff.: effluent; RE: removal efficiency.

Table 6

Removal efficiency of NH4+-N, NOx-N and T-N at different HRT (mean data of 7 days for each case).

HRT (h)	NH4 ⁺ -N (m	$NH_4^+-N (mg L^{-1})$			$NO_2^{-}-N(mgL^{-1})$		$NO_3^{-}-N(mgL^{-1})$		$T-N(mgL^{-1})$	
	Inf.	Eff.	RE (%)	Inf.	Eff.	Inf.	Eff.	Inf.	Eff.	RE (%)
11	31.0	0.5	98.4	0.0	0.3	0.0	11.2	36.3	13.9	61.6
9	30.6	0.8	97.4	0.0	0.4	0.0	11.6	34.9	14.6	58.2
7	32.3	8.9	72.6	0.0	1.7	0.0	12.8	36.9	25.0	32.1

Inf.: influent; Eff.: effluent; RE: removal efficiency.

Table 7	
Removal efficiency of NH4 ⁺ -N, NOx-N and T-N at different return ratios (mean data of 7 days for each case).

r (%)	%) $NH_4^+ - N (mg L^{-1})$		NO ₂ N	$NO_2^{-}-N (mg L^{-1})$		$NO_3^{-}-N(mgL^{-1})$		$T-N(mg L^{-1})$		
	Inf.	Eff.	RE (%)	Inf.	Eff.	Inf.	Eff.	Inf.	Eff.	RE (%)
100	30.8	1.7	94.5	0.0	0.5	0.0	12.5	33.6	16.2	51.7
200	30.6	0.8	97.4	0.0	0.4	0.0	11.6	34.9	14.6	58.2
300	32.1	1.9	94.0	0.0	0.8	0.0	12.2	33.7	16.0	52.5

Inf.: influent; Eff.: effluent; RE: removal efficiency.

Table 8

Removal efficiency of NH4⁺-N, NOx-N and T-N at different sludge return ratio (mean data of 7 days for each case).

R (%)	NH4 ⁺ -N (m	$NH_4^+ - N (mg L^{-1})$		$NO_2^{-}-N(mgL^{-1})$		$NO_3^{-}-N (mg L^{-1})$		$T-N(mgL^{-1})$		
	Inf.	Eff.	RE (%)	Inf.	Eff.	Inf.	Eff.	Inf.	Eff.	RE (%)
60	31.6	1.5	95.4	0.0	0.5	0.0	13.5	35.7	16.4	54.1
80	30.6	0.8	97.4	0.0	0.4	0.0	11.6	34.9	14.6	58.2
100	30.8	0.7	97.8	0.0	0.7	0.0	12.0	33.9	13.7	59.7

Inf.: influent; Eff.: effluent; RE: removal efficiency.

The removal efficiency of NH4⁺-N was more than 94% for all three tested values of r. The results in Table 6 showed that the removal efficiency of T-N increased from 51.7% to 58.2% in case of *r* equals to 100% and 200%, respectively. However, the removal efficiency decreased from 58.2% to 52.5% when r was changed to be 300%. The function of the mixed-liquid return is to provide nitrate nitrogen for the anoxic tank as electron acceptor for denitrification [27]. When *r* is too low, it will result in insufficient nitrate nitrogen in the anoxic tank and affects the removal efficiency of T-N. When *r* is too high, a higher recycle ratio is not recommended because it implies a high flow of recycle mixed-liquid containing some oxygen to the anoxic reactor. As a result, this excess of oxygen will be used for organic matter oxidation, thus the denitrification capacity of the system will be decreased [28]. Moreover, the effluent concentrations of NO₃-N accounted for 77.2%, 79.5% and 76.3% of T-N, respectively. The optimal removal efficiency of nitrogen was achieved at r = 200% in this investigation.

3.2.4. Effect of R on nitrogen removal

The effect of *R* (the sludge return ratio) on the removal efficiency of nitrogen was conducted at *R* equalled to 60%, 80% and 100%, respectively. The experimental conditions were: the temperature $T = 28 \degree$ C, r = 200%, HRT = 9 h, DO = 3 mg L⁻¹. The removal efficiencies of NH₄-N and T-N in case of *R* is shown in Table 8.

Maintaining the quantity of active sludge in the A^2/O system by continuous return ensures a high removal efficiency of nitrogen. Compared with enhancing the ratio of mixed-liquid return (r), increasing R was more beneficial for nitrogen removal [29]. It can be confirmed from Tables 6 and 8 that the removal efficiency of T-N increased from 54.1% to 58.2% with the increase of R from 60% to 80%, the removal efficiency of T-N continued to increase to 59.7% when R = 100%. In addition, the effluent concentrations of NO₃⁻-N accounted for 82.3%, 79.5% and 87.6% of T-N, respectively. These results proved that the increase of *R* would be more favorable for nitrogen removal.

3.2.5. Effect of individual factors on COD removal

The effect of individual factors (DO, HRT, r, R) on the removal efficiency of COD is shown in Table 9. The experimental conditions were: the temperature $T = 28 \,^{\circ}$ C, the mixture ratio of landfill leachate to sewage wastewater was 1:500, the operation time was 7 days for each run period, totally 84 days.

According to Table 8, the influent COD at different cases changed a lot with the variable practical characteristics of landfill leachate and sewage wastewater everyday. However, the effluent COD, especially the removal efficiencies of COD at different cases had a little change, except for the minimum and maximum removal efficiency values of 65.5% and 84.0% in the case of HRT=7 h and R=100%, respectively.

Therefore, the removal efficiencies of COD were more stable than those of ammonium and total nitrogen. This suggests that there are complex mechanisms among the removal of COD and ammonium nitrogen through nitrification and denitrification. This should be further investigated.

3.3. Influence of multiple factors on nitrogen removal

3.3.1. Design and results of an orthogonal array test

As Table 3 summarized and the above-described results of individual factors on nitrogen removal, the effects of HRT, DO, *r* and *R*

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Removal of COD at different affective factors (mean data of 7 days for each case).

Affective factor	Inf. COD $(mg L^{-1})$	Inf. T-N (mg L^{-1})	Influent COD/N	Eff. COD ($mg L^{-1}$)	Eff. T-N (mg L^{-1})	COD RE (%)	T-N RE (%)
$DO = 2 mg L^{-1}$	150.2	36.3	4.1	39.8	18.7	72.8	48.6
$DO = 3 \text{ mg } L^{-1}$	132.8	34.9	3.8	33.4	14.6	74.9	58.2
$DO = 4 \text{ mg } \text{L}^{-1}$	125.7	33.1	3.8	32.9	19.0	73.7	42.5
HRT = 11 h	122.1	36.3	3.4	35.4	13.9	70.7	61.6
HRT=9h	150.2	34.9	4.3	40.1	14.6	72.6	58.2
HRT = 7 h	128.3	36.9	3.4	44.1	25.0	65.5	32.1
r = 100%	131.4	34.6	3.8	34.5	16.2	73.3	51.7
r=200%	126.6	34.9	3.6	29.4	14.6	76.6	58.2
r = 300%	129.0	33.7	3.8	33.7	16.0	73.6	52.5
R = 60%	121.5	35.7	3.4	30.2	16.4	75.0	54.1
R = 80%	126.6	34.9	3.6	29.4	14.6	76.6	58.2
R = 100%	135.5	33.9	3.9	20.9	13.7	84.0	59.7

Inf.: influent; Eff.: effluent; RE: removal efficiency.

	Factor				COD removal efficiency (%)	NH ₄ -N removal efficiency (%)	T-N removal efficiency (%)	
	A	В	С	D				
	Column		_					
	1	2	3	4				
1	1(11)	1(2)	1(100)	1(60)	72.2	91.3	55.9	
2	1	2	2	2	81.7	98.8	63.8	
3	1	3	3	3	79.6	99.6	57.1	
4	2(9)	1	2(200)	3	71.4	85.3	48.4	
5	2	2(3)	3	1	74.2	90.3	51.2	
6	2	3	1	2(80)	75.8	92.2	46.1	
7	3(7)	1	3(300)	2	64.8	72.2	39.9	
8	3	2	1	3(100)	66.9	78.2	41.8	
9	3	3(4)	2	1	65.5	77.0	36.3	

on the removal efficiency of nitrogen in A^2/O process were strong. However, the overall process of A^2/O system for nitrification and denitrification together with the removal of COD depend upon a few individual factors and their interactions.

The determination of optimal multiple factors for the optimal operation of A^2/O process in this study was based on the maximum removal efficiency of COD, NH_4^+ -N and TN. A $L_9(3^4)$ orthogonal array was used in this experimental design [30,31] according to Table 3. The results of orthogonal experiment are listed in Table 10.

Calculating the average value of each column for horizontal K_1 , K_2 , K_3 and the range of each column. The analysis of the orthogonal experiment is shown in Table 11.

The removal efficiency of NH_4^+ -N and T-N obtained by considering interactive factors ranged from 72.2% to 99.6% and from 36.3% to 63.8%, respectively, while the removal efficiency of COD ranged from 64.8% to 81.7%. The optimal combination of the experiment is the tested factor levels resulting in the largest values of the percentage removal of NH_4^+ -N, TN, and COD. Based on this principle, the influential sequence of the four factors for the three criteria was ABDC and the optimal formulation was A1B2C2D2. As a result, the optimal values of individual factors in this study were HRT = 11 h, DO = 3 mg L⁻¹, r = 200% and R = 80%.

3.3.2. Confirmatory experiment under near optimal condition

A confirmation experiment was carried out to verify the optimal parameters obtained from the orthogonal array test. The experimental results were shown in Fig. 2 after operated for 45 days.

The average removal efficiencies of NH_4^+ -N, T-N, COD and T-P were 96.5%, 61.0%, 81.7% and 80.9%, respectively. The A^2/O system achieved high and stable removal efficiencies of NH_4^+ -N, T-N, COD and T-P after 15 operation days. Moreover, the removal efficiency for COD was over 82% during all the experiments, indicating that COD removal was only slightly affected by the variation of all indi-

Table 11	
The analysis of the orthogonal experiment	•

		А	В	С	D
COD	K_1	77.8	69.5	71.6	70.6
	K_2	73.8	74.2	72.8	74.1
	K_3	65.7	73.6	72.8	72.6
	R	12.1	4.8	1.2	3.5
T-N	K_1	58.9	48.1	47.9	47.8
	K_2	48.6	52.3	49.5	49.9
	K_3	39.3	46.5	49.4	49.1
	R	19.6	5.8	1.6	2.1
NH4 ⁺ -N	K_1	96.6	82.9	87.2	86.2
	K_2	89.3	89.1	87.0	87.8
	K_3	75.8	89.6	87.4	87.7
	R	20.8	6.7	0.3	1.6

vidual factors. The removal efficiency of NH₄-N and T-N increased at the beginning and was stable after 15 days, maintaining the high removal efficiencies of 98% and 63%, respectively, while the removal efficiency of T-P decreased gradually from 84% to 79%.

3.3.3. Application in engineering scale

Guangzhou Datansha Domestic Sewage Wastewater Treatment Plant was the first constructed large-scale (total capacity: $550,000 \text{ m}^3/\text{d}$) sewage treatment plant in Guangdong Province, South China. The concentrations of COD, NH4⁺-N, T-N and T-P in sewage wastewater are relatively low because of the subtropical and rainy climate of the city. As listed in Table 3, the concentrations of COD, NH4⁺-N, T-N for control of this experimental study were 85.8 mg L^{-1} , 25.2 mg L^{-1} , 28.6 mg L^{-1} , respectively. Generally, the concentration of T-P in Guangzhou sewage wastewater ranges from 1.5 mg L^{-1} to 2.8 mg L^{-1} . Therefore, combined treatment of sewage wastewater with high organic loading wastewaters, such as landfill leachate and fecal wastewater, etc. was considered. As mentioned above, the anaerobic–anoxic–aerobic (A^2/O) system (capacity: 220.000 m³/d) in Guangzhou Datansha Domestic Sewage Wastewater Treatment Plant was recommended to be constructed for the combined treatment of landfill leachate generated from XFL because of the significant insufficient treatment capacity of landfill leachate in XFL

We applied the pilot scale investigation results in the large-scale combined treatment of sewage wastewater with landfill leachate. As described above, the optimal operation parameters obtained by the orthogonal experiment, i.e., HRT=11 h, DO=3 mg L⁻¹, r=200% and R=80% was applied in the optimal operation of the 220,000 m³/d of A²/O system. Though it was very difficult to control the mixture ratio exactly to be 1:500 in engineering scale, very good removal efficiencies were achieved, as shown in Table 12, the mean data for a whole year of 2008 (365 days) indicated that the pilot scale experimental results were valuable and effective.



Fig. 2. The removal efficiencies of NH4⁺-N, T-N, COD and T-P of the system.

Year 2008	$COD (mg L^{-1})$		$NH_4^+-N(mgL^{-1})$		T-N (mg L ⁻¹)		$T-P(mg L^{-1})$	
Month	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent
1	182.00	31.20	34.14	3.38	37.09	17.23	2.34	0.60
2	141.97	32.86	29.99	4.40	32.77	19.21	1.87	0.63
3	226.73	36.33	45.85	6.32	60.27	18.88	2.74	0.73
4	249.92	34.69	52.57	4.46	55.29	18.75	2.31	0.24
5	170.54	32.99	37.55	4.45	40.16	19.39	1.79	0.53
6	159.47	30.08	28.78	1.25	32.57	15.53	1.28	0.12
7	191.21	29.80	35.72	1.39	39.30	17.39	1.60	0.12
8	178.00	31.20	36.33	0.89	39.74	18.47	1.65	0.28
9	171.10	34.20	36.69	1.12	40.08	17.94	1.66	0.49
10	171.71	33.02	41.1	1.58	43.79	18.97	1.98	0.65
11	179.74	31.36	46.43	2.55	49.59	17.76	2.15	0.68
12	214.91	30.52	29.21	1.42	33.79	16.89	2.51	0.53
Mean	186.44	32.35	37.86	2.77	42.04	18.03	1.99	0.47
Removal efficiency		82.65%		92.69%		57.10%		76.55%
China Discharge Standard (GB18918-2002), level 1B		40.0		8.0		20.0		1.0

Concentrations of COD, NH4+-N, T-P, T-P in engineering scale operation of sewage wastewater with leachate (mean data of 365 days).

It is evident that the mean data of concentrations of effluent COD, NH₄⁺-N, T-N and T-P in 2008 (365 days) have been kept below the discharge limits of China Standard (COD \leq 40 mg L⁻¹, NH₄⁺- $N \le 8 \text{ mg } L^{-1}$, $T-N \le 20 \text{ mg } L^{-1}$, $T-P \le 1.0 \text{ mg } L^{-1}$). Furthermore, the removal efficiencies of COD, NH4+-N, T-N and T-P were 82.65%, 92.69%, 57.10% and 76.55%, respectively. These high removal efficiencies of pollutants in mixed wastewater were achieved among the ranges obtained by the orthogonal experiment (Table 10). The removal efficiency (mean data of 365 days) of COD (82.65%) in engineering scale $(220,000 \text{ m}^3/\text{d})$ was greater than that of the pilot scale $(3.8 \text{ m}^3, 77.4\% \text{ in case of mixture ratio} = 1:500 \text{ in Table 4})$ and was similar to the control one (82.3%). However, the removal efficiencies of NH₄⁺-N and T-N in engineering scale (92.69% and 57.10%, respectively) were a little bit lower that those of pilot scale experiment (95.0% and 58.3% in case of mixture ratio=1:500 in Table 4). Therefore, the main problem for the removal of pollutants in combined sewage wastewater with landfill leachate was how to enhance the removal efficiency of nitrate by denitrification and promote the removal efficiency of total nitrogen accordingly. We have been trying to improve the optimal operation and control of the processes, the denitrification process was improved. We measured the concentrations of nitrate and nitrite for anaerobic, anoxic and aerobic process in the engineering vessels for the latest period of 32 days (from November to December 2009). The mean data for nitrite were less than 0.1 mg L^{-1} , while the mean data of effluent nitrate was 5.35 mg L^{-1} , as shown in Fig. 3.

Table 12



Fig. 3. The concentrations of NO_3^- at different processes in the system.

It was illustrated in Fig. 3 that the nitrate concentration in anaerobic and anoxic processes were much lower that that in aerobic process, the mean data of nitrate concentrations in anaerobic, anoxic and aerobic processes were 0.54 mg L^{-1} , 1.77 mg L^{-1} and 5.96 mg L^{-1} , respectively. The effluent concentration of nitrate was much lower that those obtained in the pilot scale experiments.

4. Conclusions

A pilot scale of A²/O bioreactor was designed and used for investigation of combined treatment of mixed sewage wastewater with landfill leachate. The results revealed that the optimal mixture ratio of landfill leachate and domestic wastewater was 1:500. The average effluent concentrations of COD, NH₄-N and TN could be maintained below the discharge limit for Chinese Discharge Standard (level 1B: COD \leq 40 mg L⁻¹, NH₄⁺-N \leq 8 mg L⁻¹, TN < 20 mg L⁻¹,T-P \leq 1.0 mg L⁻¹).

The orthogonal array test was designed and used to investigate the optimal combination of four individual factors for the nitrogen removal in the mixture wastewater. The results demonstrated that HRT was the most major factor. The optimal conditions for the A^2/O system were HRT = 11 h, DO = 3 mg L⁻¹, r = 200% and R = 80%.

A confirmatory experiment was carried out using the optimal formulation. This combination of parameter values enabled the system to achieve high removal efficiencies for nitrogen, phosphorus and organic compounds. The average removal efficiencies of NH_4 -N, T-N, COD and T-P were 96.5%, 61.0%, 81.7% and 80.9% respectively.

The pilot scale (3.8 m^3) investigation results were applied in the large-scale $(220,000 \text{ m}^3/\text{d})$ combined treatment of sewage wastewater with landfill leachate in Guangzhou Datansha Domestic Sewage Wastewater Treatment Plant. An 1-year period of operation results showed that the optimal orthogonal array test results were valuable and effective. The removal efficiencies of COD, NH₄⁺-N, T-N and T-P were 82.65%, 92.69%, 57.10% and 76.55%, respectively. The effort for enhancing removal efficiency of nitrate was successful through the optimal operation and control of the A²/O processes.

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