Optimizing external carbon source addition in domestic wastewater treatment based on online sensoring data and a numerical model
Qibin Wang, Qiuwen Chen and Jing Chen

ABSTRACT
The removal of total nitrogen in wastewater treatment plants (WWTPs) is often unsatisfactory for a variety of reasons. One possible measure to improve nitrogen removal is the addition of external carbon. However, the amount of carbon addition is directly related to WWTP operation costs, highlighting the importance of accurately determining the amount of external carbon required. The objective of this study was to obtain a low nitrate concentration in the anoxic zone of WWTPs efficiently and economically by optimizing the external carbon source dosage. Experiments were conducted using a pilot-scale pre-denitrification reactor at a Nanjing WWTP in China. External carbon source addition based on online monitoring of influent wastewater quality and a developed nitrification–denitrification numerical model was investigated. Results showed that carbon addition was reduced by 47.7% and aeration costs were reduced by 8.0% compared with those using a fixed-dose addition mode in the pilot reactor. The obtained technology was applied to the full-scale Jiangxinzhou WWTP in Nanjing with promising results.

Key words | aeration costs, carbon source addition, nitrogen removal, pilot reactor

INTRODUCTION
Biological wastewater treatment systems have denitrification capabilities (Biesterfeld et al. 2003; Peng et al. 2007). However, nitrogen removal efficiency is often limited by available carbon in the influent, which can lead to effluent nitrogen concentrations exceeding mandatory standards, especially when high nitrogen loads enter the plant. The addition of an external carbon source to wastewater treatment systems has been proven effective in enhancing nitrogen reduction in low C/N ratio wastewater (Isaacs & Henze 1993; Kampas et al. 2007; Park et al. 2011; Zubrowska-Sudol & Walczak 2015), but can also increase both the cost and carbon footprint (Flores-Alsina et al. 2011; Gustavsson & Tumlin 2013). Therefore, external carbon source dosage should be based on estimation of the denitrification potential of wastewater.

Kujawa & Klapwijk (1999) estimated the denitrification potential for pre-denitrification systems using nitrate utilization rate (NUR) batch tests. Thus, the minimum obtainable nitrate effluent quality could be predicted and the treatment process could be improved by the addition of external carbon. However, this is difficult to apply under real-time conditions. Some researchers (Kim et al. 2004; Queinnec & Sperandio 2004) found that the duration of the anoxic phase could be distributed optimally by using oxidation-reduction potential (ORP) as denitrification control parameter, and constant effluent quality could be obtained with the optimum external carbon source in the sequencing batch reactors. However, in a continuous system for treating wastewater, the nitrate breakpoint using ORP may be difficult to determine because of the interaction between mixed liquid recycling from oxic zones and returned activated sludge from the secondary clarifier.

In recent years, some studies have applied advanced control strategies to improve the activated sludge process for nitrogen removal (Lindberg & Carlsson 1996; Yuan et al. 2002). Meyer & Popel (2005) described fuzzy-control for
improving nitrogen removal in wastewater treatment plants (WWTPs) with pre-denitification, showing that fuzzy-control systems can be accepted by operating personnel. Lindberg & Carlsson (1999) proposed and applied an adaptive carbon flow rate controller to a pre-denitrifying pilot-scale plant and achieved low effluent nitrate concentration. Yuan & Keller (2003) investigated the integrated control of nitrate recirculation and external carbon addition for a pre-denitrification biological wastewater treatment system. Their proposed control structure consisted of four feedback control loops, which regulated nitrate recirculation and carbon dosage flows in a highly coordinated manner such that the consumption of external carbon was minimized, while nitrate discharge limits were met. The control system required the measurement of nitrate concentrations at the end of both the anoxic and aerobic zones. Distinct from ordinary control systems, which typically minimize variation in the controlled variables, their proposed system essentially maximized the diurnal variation of effluent nitrate concentration, and thereby maximized the use of influent chemical oxygen demand (COD) for denitrification and minimized the requirement for an external carbon source. However, few studies have attempted to develop methods to improve nitrogen removal and reduce operation costs through optimizing carbon source addition according to online sensoring data and influent substrate availability. Determining the minimum amount of external carbon required based on effluent quality is still a problem to be solved.

The present study explored the effectiveness of nitrogen removal from domestic wastewater with the addition of an external carbon source in different operation modes. The objectives of this study were to: (1) optimize carbon source addition through an online monitoring system and a developed nitrification–denitrification model in a continuous wastewater treatment process; (2) examine the efficiency of using the combined monitoring and modeling approach. A pilot-scale plant configured as a pre-denitrification reactor was operated for more than 3 months to demonstrate the robustness of the proposed approach with daily varying municipal wastewater.

**MATERIALS AND METHODS**

**Diagram for optimizing external carbon source addition**

A systematic optimization method was developed to determine optimal external carbon source addition for enhancing biological nitrogen removal from domestic wastewater. It consisted of data collection, external carbon source estimation, implementation and nitrogen removal optimization (Figure 1). Firstly, process parameters and operating parameters were collected. Then a mathematical nitrification–denitrification model was developed to describe the nitrogen removal process (Henze et al. 2008). The model calibration and verification was carried out using historical operating data of the reactor in order to ensure model performance. Influent characteristics (i.e. influent COD \(S_{COD,inf}\), total nitrogen (TN), total Kjeldahl nitrogen (TKN) and water temperature) were obtained using online sensors. The main operating variables (i.e. nitrate recirculation flow rate, returned sludge flow rate) were determined through the numerical model, which estimated denitrification potentials based on the adequacy of influent organic materials. If the influent organic material was sufficient and the predicted TN concentration of the effluent

---

**Figure 1** | Diagram for enhanced biological nitrogen removal by optimizing external carbon source addition.
could meet the discharge requirements (90% of standard limit as the maximum value), the operating variables were applied to the reactor. Otherwise, an external carbon source was added, and the accurate dosage was calculated according to the discharge requirement and the availability of influent substrate. If effluent nitrogen did not meet discharge requirements, the main operation variables were redetermined and the optimization procedure was reiterated until effluent nitrogen concentration met discharge requirements. Since the influent water quality changed continuously, the above procedures were conducted at each period (2 h in this study) using the averaged online monitoring data within the time.

**Experimental design and operation**

This study was conducted at the Jiangxinzhou WWTP (640,000 m³/d) in Nanjing, China, which employs an activated sludge process. Configuration of the full-scale WWTP was described in detail in our previous study (Wang & Chen 2016). A pilot-scale plant was installed at the WWTP, with the pre-denitrification process shown in Figure 2. The pilot reactor had a maximum working volume of 1.4 m³. Other design parameters are listed in Table 1. In the process, influent was continuously introduced into a four-chambered anoxic tank by a peristaltic pump, which then flowed into a 12-chambered aerobic tank. Mechanical mixing and air compression were conducted separately to provide anoxic and aerobic conditions, respectively.

Returned sludge and nitrate were transported to the first compartment of the anoxic tank by peristaltic pumps. In addition, online sensors were installed at the entrance of the full-scale WWTP to monitor the concentrations of COD and ammonium online. Acetate as a carbon supplement was added in the form of a sodium acetate (CH₃COONa·3H₂O) solution (25,000 mg COD/L) fed into the first compartment of the anoxic tank by a peristaltic pump.

Raw wastewater was collected from the outlet of an aerated grit chamber from the full-scale WWTP. It was first pumped into an intermediate storage tank before being pumped into the bioreactor. Activated sludge from the aeration tank of the full-scale WWTP was used as an inoculum for the system. The total hydraulic retention time (HRT) of the wastewater in the system (including anoxic and aerobic tanks) was controlled within 6 h. A solid retention time (SRT) of 12–15 days was controlled by excess sludge discharge during the experiments. Mixed liquor suspended solids (MLSS) in the oxic tanks were maintained between 2,500 and 4,000 mg/L. Dissolved oxygen (DO) concentrations in the oxic zone and anoxic zone were controlled at 1.0–2.0 mg/L and ≤0.5 mg/L, respectively, during operation of the reactor. Water temperature was approximately 13 °C during the experimental period. As many WWTPs lack flexibility (Jeppsson et al. 2002), sludge recycling ratio was set constant to 80% in this pre-denitrification plant. The nitrate recirculation ratio could be adjusted from 60 to 100% with an interval of 10% in order to change denitrification potential of the system. The DO (5540D0A, HACH) and pH (PD1P1, HACH) sensors were placed in the fifth and eleventh aerobic zones. Experiments on the effectiveness of nitrogen removal were carried out in three phases: Phase 1 was from day 1 to day 40, without any external carbon source; Phase 2 was from day 41 to day 70, with carbon source addition in fixed mode; Phase 3 was from

![Figure 2](https://iwaponline.com/wst/article-pdf/75/11/2716/453284/wst075112716.pdf)  
*Schematic diagram of pilot-scale pre-denitrification process.*
day 71 to day 95, with carbon source addition in optimized mode.

### Wastewater characteristics

Raw domestic wastewater was collected from the outlet of the full-scale WWTP aerated grit chamber. The experimental operation lasted for more than 3 months. Influent wastewater characteristics of the treatment system are given in Table 2.

Samples were collected regularly from the influent and effluent of the pilot-scale reactor to measure concentrations of COD, ammonium (NH$_4^+$-N), nitrate (NO$_3^-$-N), nitrite (NO$_2^-$-N), TKN, TN, MLSS and volatile suspended solids (VSS) according to Standard Methods (APHA 2005). Temperature and DO were measured in situ using an YSI 6600 V2 Multi-Parameter Water Quality Sonde (USA).

The organic material could be divided into particulate non-biodegradable fraction, soluble non-biodegradable fraction and biodegradable fraction, which could be determined using the method by Brouwer et al. (1998). The nitrogen content included TKN and total oxidized nitrogen. Measured values for COD and nitrogen fractionation in wastewater characterization for the wastewater treatment system are presented in Table 3.

### Determination of carbon source addition

In the study, the Class-1A (COD < 50 mg/L, NH$_4^+$-N < 5 mg/L, TN < 15 mg/L) Discharge Standards of Pollutants for Municipal WWTPs (MEP 2002) were set as the target discharge qualities.

For Phase 1, daily monitoring of the reactor performance was conducted from November 20, 2013 (day 1) to December 29, 2013 (day 40). Data showed that effluent TN did not always meet the Class-1A standards. Therefore, an external carbon source dosage for Phase 2 was applied to achieve the optimal COD/TN ratio under the worst conditions (i.e. lowest COD/TN ratio) in Phase 1. Previous studies indicate that the optimal COD/TN ratio for good performance of nitrogen removal was about 5 (Wang et al. 2009), 4 (Zou et al. 2014), 6.96 (Jia et al. 2015) and 3.7 (Carrera et al. 2003). In this study, the average value 4.92 of the COD/TN ratios (5, 4, 6.96 and 3.7) was used as the limit for the external carbon source addition.

For Phase 3, the nitrate production and nitrogen required for sludge are defined in Equations (1)–(3) as described in the literature (Henze et al. 2008):

\[
N_{ne}(mg N/L) = N_{hi} - N_s - N_{te}
\]

\[
N_{te}(mg N/L) = N_{ae} + f_{Nouts} \times N_{hi}
\]

where \(N_{ne}\) (mg N/L) is nitrate production; \(N_{hi}\) (mg N/L) is influent TKN concentration; \(N_s\) (mg N/L) is nitrogen concentration required for sludge production (mg N/L); \(N_{te}\) is

### Table 1 | Design parameters for pilot-scale reactor

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Configuration size</strong></td>
<td></td>
</tr>
<tr>
<td>Storage tank</td>
<td>1.5 m (L) × 1.5 m (W) × 1.0 m (H)</td>
</tr>
<tr>
<td>Anoxic tank</td>
<td>1.2 m (L) × 0.4 m (W) × 1.0 m (H)</td>
</tr>
<tr>
<td>Oxic tank</td>
<td>3.6 m (L) × 0.4 m (W) × 1.0 m (H)</td>
</tr>
<tr>
<td>Secondary clarifier</td>
<td>1.0 m (D) × 0.75 m (H)</td>
</tr>
<tr>
<td><strong>Secondary clarifier unit</strong></td>
<td></td>
</tr>
<tr>
<td>HRT</td>
<td>3 h</td>
</tr>
<tr>
<td>Surface loading</td>
<td>0.29 m$^3$/(m$^2$ h)</td>
</tr>
</tbody>
</table>

### Table 2 | Influent wastewater quality characteristics from the Jiangxinzhou WWTP in Nanjing, China

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Unit</th>
<th>Range</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>71.2–168.3</td>
<td>114.5</td>
</tr>
<tr>
<td>NH$_4^+$-N</td>
<td>mg/L</td>
<td>12.7–28.6</td>
<td>21.8</td>
</tr>
<tr>
<td>NO$_2^-$-N</td>
<td>mg/L</td>
<td>0–0.12</td>
<td>0.04</td>
</tr>
<tr>
<td>NO$_3^-$-N</td>
<td>mg/L</td>
<td>0.04–1.21</td>
<td>0.60</td>
</tr>
<tr>
<td>TN</td>
<td>mg/L</td>
<td>16.4–34.2</td>
<td>25.0</td>
</tr>
<tr>
<td>C/N ratio</td>
<td>–</td>
<td>2.80–7.56</td>
<td>4.62</td>
</tr>
</tbody>
</table>

*Particulate non-biodegradable fraction of total influent COD.
Soluble non-biodegradable fraction of total influent COD.
Fraction of nitrogen in sludge.
Soluble non-biodegradable organic nitrogen fraction of total influent TN.
Total TKN fraction of total influent nitrogen.
Total oxidized nitrogen fraction of total influent nitrogen.

### Table 3 | COD and nitrogen fractionation in wastewater characterization for the wastewater treatment system

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_{\text{SUN}}$</td>
<td>–</td>
<td>0.16</td>
<td>This study (measured)</td>
</tr>
<tr>
<td>$f_{\text{SU}}$</td>
<td>–</td>
<td>0.07</td>
<td>This study (measured)</td>
</tr>
<tr>
<td>$f_{\text{N}}$</td>
<td>mg N/mgVSS</td>
<td>0.1</td>
<td>Marais &amp; Ekama (1976)</td>
</tr>
<tr>
<td>$f_{\text{Nouts}}$</td>
<td>–</td>
<td>0.03</td>
<td>Marais &amp; Ekama (1976)</td>
</tr>
<tr>
<td>$f_{\text{TKN}}$</td>
<td>–</td>
<td>0.97</td>
<td>This study (measured)</td>
</tr>
<tr>
<td>$f_{\text{NO}}$</td>
<td>–</td>
<td>0.03</td>
<td>This study (measured)</td>
</tr>
</tbody>
</table>
TKN in the effluent (mg N/L); \( N_{ae} \) (mg N/L) is effluent ammonium concentration; and \( f_{NVus} \) is the soluble non-biodegradable organic nitrogen fraction of total influent TKN.

\[
\frac{N_s}{S_{H}} (mgN/mgCOD) = f_h \left[ \frac{(1 - f_{sus} - f_{sup})}{1 + b_H SRT} (1 + f_h b_H SRT) + \frac{f_{sup}}{f_{co}} \right]
\]

where \( N_s/S_H \) (mg N/mg COD) is the concentration of nitrogen required per influent for sludge production per mg COD/L organic load in the wastewater treatment system; \( f_n \) (mg N/mg VSS) is the non-biodegradable particulate VSS nitrogen content; \( f_{sus} \) is the soluble non-biodegradable fraction of total influent COD; \( f_{sup} \) is the particulate non-biodegradable fraction of total influent COD; \( f_{CV} \) (mg COD/mg VSS) is the COD to VSS ratio of the sludge; \( f_h \) is the non-biodegradable fraction of the ordinary heterotrophic organisms (OHs); \( b_H \) is the specific rate of endogenous mass loss of denitrification bacteria; and \( SRT \) (day) is the solid residence time.

Denitrification potential (\( DN_{Recycle} \)) can be expressed by:

\[
DN_{Recycle} (mgN/d) = \frac{R_1 + R_2}{1 + R_1 + R_2} \times N_{ne} \times Q_i
\]

where \( R_1 \) is the nitrate recirculation ratio; \( R_2 \) is the sludge recycling ratio; and \( Q_i \) (m\(^3\)/d) is the influent flow rate.

It should be noted that Equation (4) only holds true on the assumption that there is no organic substrate limitation occurring in the anoxic zone. Under operation conditions, influent substrate may not be sufficient to denitrify the entire amount of nitrate formed.

The influent COD available for denitrification (\( S_{COD, DN} \)) in the anoxic zone can be expressed by Equation (5):

\[
S_{COD, DN} (mgCOD/L) = S_{COD, int} \times (1 - f_{sus} - f_{sup})
\]

Denitrification in a pre-denitrification reactor can be estimated based on the stoichiometric relationships between \( NO_3^-N \) and COD concentrations. Compared with the substrate from the influent, the contribution of organics from endogenous respiration is negligible (Tan & Ng 2008). The availability of COD (\( S_{COD, DNP} \)) demand for denitrification is calculated in Equation (6):

\[
S_{COD, DNP} (mgCOD/L) = m \times DN_{Recycle}
\]

where \( m \) (mg COD/mg \( NO_3^-N \)) is the amount of COD consumed per unit of \( NO_3^-N \).

Equation (5) calculates the availability of COD present in the influent wastewater for denitrification, i.e. the denitrification capacity of a pre-denitrification plant; Equation (6) calculates available COD demand for the denitrification potential, which is limited by the nitrate recirculation ratio and sludge recycling ratio. Thus, for a particular set of operating conditions, there were two possible outcomes for comparison between \( S_{COD, DN} \) and \( S_{COD, DNP} \) in the present study:

1. \( S_{COD, DN} \geq S_{COD, DNP} \) indicating influent organic substrate was adequate and effluent TN generally met discharge standards.
2. \( S_{COD, DN} < S_{COD, DNP} \), suggesting carbon source in the influent was insufficient and external carbon should be added to the anoxic tank to enhance nitrogen removal.

The amount of sodium acetate as the external carbon source was calculated by Equation (7):

\[
D(mgCOD/d) = \frac{S_{COD, DNP} - S_{COD, DN}}{0.47}
\]

where \( D \) (mg COD/d) is the dosage of sodium acetate; and 0.47 is the conversion coefficient from sodium acetate to COD (Isaacs & Henze 1995).

It should be noted that TN was not monitored directly, but was converted from the monitored NH\(_4^+\)-N concentrations. Long-term (March–October 2013) continuous observation showed that TN was linearly related to the concentration of NH\(_4^+\)-N (\( R^2 = 0.91 \), Figure 3). As the concentrations of influent
nitrate and nitrite were very low (sum was less than 1.0 mg/L, Table 2), and could be ignored, the concentration of TKN in the influent was approximately equal to that of influent TN.

**Determination of total oxygen demand**

The calculation equations for total oxygen demand in a biological nitrogen removal system were as per Henze et al. (2008) (Equations (8)–(12)).

Daily average oxygen demand for organic material removal ($FO_c$) is given by Equation (8) and for nitrification ($FO_n$) is given by Equation (10):

$$FO_c (kg O_2/d) = FS_{bi} \times \left[ (1 - f_{cV}Y_{He}) + (1 - f_{H})b_H \times \frac{Y_{He}SRT}{1 + b_HSRT} \right]$$

$$FS_{bi} (mg COD/d) = FS_{bi} (1 - f_{ru} - f_{ur}) + FS_{ai}$$

where $FS_{bi}$ (mg COD/d) is the readily biodegradable organics; $Y_{HV}$ (mgVSS/mgCOD) is the VSS yield of OHOs; and $FS_{ai}$ (mg COD/d) is the daily carbon source addition dosage.

$$FO_n (kg O_2/d) = 4.57 \times N_{ne} \times Q_i$$

where $FO_n$ (kg O_2/d) is the daily oxygen demand for nitrification; and 4.57 mg O_2/mg NH$_4^+$-N nitrified to NO$_3^-$-N is a constant.

$$FO_d (kg O_2/d) = 2.86 \times (N_c - N_{ne}) \times Q_i$$

where $FO_d$ (kg O_2/d) is the daily recovery oxygen for denitrification; $N_c$ is the nitrification capacity (mg N/L); $N_{ne}$ is the effluent nitrate concentration (mg N/L); and 2.86 mg O_2/mg NO$_3^-$-N transferred to gaseous nitrogen is a constant.

$$FO_d (kg O_2/d) = FO_c + FO_n - FO_d$$

where $FO_d$ (kg O_2/d) is the total daily oxygen demand in a biological nitrogen removal system.

The activated sludge model parameters are given in Table 4 based on previous studies (Marais & Ekama 1976; Henze et al. 2008).

**RESULTS AND DISCUSSION**

**Reactor performance**

Figure 4 shows the characteristics of the influent and effluent, and the removal efficiencies of COD, NH$_4^+$-N and TN in each period of the study. The COD concentration in the influent ranged from 71.2 to 168.3 mg/L, and was less than 50 mg/L in the effluent. The average removal efficiency of COD was 78.6%. Thus, the COD removal efficiencies were found to be independent of the addition modes in this study.

The NH$_4^+$-N concentration in the influent ranged from 14.1 to 28.6 mg/L. After treatment, the average effluent concentration and removal efficiency were 0.24 mg/L and 98.6%, respectively. There were obvious differences in TN removal between carbon source addition and no addition. In Phase 1, influent TN varied from 16.4 to 32.1 mg/L, and effluent TN concentration was mostly lower than 15 mg/L (Class-1A discharge standard), with a few exceptions. In Phase 2, sodium acetate was used as the external carbon source.

**Table 4** Stoichiometric and kinetic constants and their temperature dependency in the steady-state activated sludge model

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Temperature dependency</th>
<th>$\theta$</th>
<th>Unit</th>
<th>Value at 20 $^\circ$C</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Y_{HV}$</td>
<td>Remains constant</td>
<td>1</td>
<td>mgCOD/mgCOD</td>
<td>0.67</td>
<td>Henze et al. (2008)</td>
</tr>
<tr>
<td>$Y_{HV}$</td>
<td>Remains constant</td>
<td>1</td>
<td>mgVSS/mgCOD</td>
<td>0.45</td>
<td>Henze et al. (2008)</td>
</tr>
<tr>
<td>$b_H$</td>
<td>$b_H = b_{H20}(T - 20)$</td>
<td>1</td>
<td>mgCOD/mgCOD</td>
<td>0.2</td>
<td>Henze et al. (2008)</td>
</tr>
<tr>
<td>$f_{cV}$</td>
<td>Remains constant</td>
<td>1</td>
<td>mgCOD/mgVSS</td>
<td>1.48</td>
<td>Henze et al. (2008)</td>
</tr>
<tr>
<td>$m^t$</td>
<td>Remains constant</td>
<td>1</td>
<td>mgCOD/mgNO$_3^-$-N</td>
<td>8.6</td>
<td>Marais &amp; Ekama (1976)</td>
</tr>
</tbody>
</table>

*COD yield of heterotrophic bacteria.

$^a$VSS yield of heterotrophic bacteria.

$^b$Specific rate of endogenous mass loss of heterotrophic bacteria.

$^c$Don-biodegradable fraction of the heterotrophic bacteria.

$^d$COD to VSS ratio of the sludge.

$^e$Amount of COD consumed per unit of NO$_3^-$-N.
organic carbon source to raise the influent COD/TN ratio (more than 4.92) in the fixed mode, and effluent TN concentration was always lower than 15 mg/L. In Phase 3, the added sodium acetate dosage was estimated by the proposed approach. The TN concentration in the effluent was stable and always below 15 mg/L.

According to Figure 4, variation in the influent had little effect on the removal efficiencies of COD and NH$_4^+$-N, which meant that the COD and NH$_4^+$-N removals were not influenced by an external carbon source addition. Although the concentrations of ammonium in the influent showed great variability, they were quite low in the effluent, which indicated that nitrification in the reactor was excellent irrespective of influent variations. Similar results were reported by Kumar et al. (2012). However, the removal of TN through denitrification in our system was highly dependent on the influent substrate. This could be due to insufficient available organic material for the growth of heterotrophic microbes during denitrification. Sodium acetate as an external carbon source was added to the anoxic tank in the fixed and optimized modes, respectively. The TN concentration in the effluent was stable and met the Class-1A limits under both modes. Moreover, high COD removal efficiency was observed throughout the whole experimental period, implying that the sodium acetate was almost completely consumed.

The averaged mass balance of nitrogen in different operation phases of the pilot-scale treatment system is shown in Figure 5. The exit routes of nitrogen in influent comprised five pathways: gaseous nitrogen, TKN, nitrate, nitrite and nitrogen in the excess sludge. In Phases 2 and 3, the eliminated proportions of nitrogen increased by more than 10% compared with that in Phase 1, because carbon source addition improved nitrogen removal in the anoxic zone. The fractions of nitrogen increased accordingly with the excess sludge because carbon source addition resulted in greater sludge production. There were no significant differences in the proportions of gaseous nitrogen between Phase 2 and 3.

**Carbon source dosage in different addition modes**

Nitrogen removal is usually conducted by nitrification and denitrification. Figure 4 shows that almost 100% NH$_4^+$-N removal efficiency was achieved, proving that complete oxidation of ammonium nitrogen in the first step was obtained. In Phase 1, effluent TN was not consistently satisfactory, implying that nitrogen elimination was mainly confined to the second step. Liu et al. (2013) proved that COD was
mostly consumed in the anoxic tank and anaerobic unit, and the remaining COD was mainly non-biodegradable or difficult-to-degrade organic matter. The lack of available carbon in the anoxic chamber led to low TN removal efficiency.

In Phase 1, raw municipal wastewater was fed into the reactor without the addition of external carbon. In the fixed mode (Phase 2), the carbon source dosage was 88.0 g (sodium acetate)/m³ (influent wastewater) for 30 days, which was sufficient to ensure effluent quality under any circumstance. In the optimized mode (Phase 3), carbon source was added according to online monitoring of influent conditions and operation parameters. No external carbon was added when the influent substrate was adequate; however, when it was insufficient for denitrification, the external carbon dose was calculated according to Equation (7). The average dose was 46.0 g (sodium acetate)/m³ (influent wastewater) under the optimized control mode. Compared with the fixed mode, the carbon source amount was reduced by 47.7% in Phase 3.

**Control variables**

The performance of the reactor was significantly related to the control variables. In order to reduce operation cost, the optimization on the reactor was mostly carried out by only adjusting the controllable parameters. When effluent quality was not satisfactory, the optimal dosage of external carbon was calculated by the model and added accordingly. Since the influent nitrogen concentrations were within a narrow range and the pilot reactor had a small storage (2.0 m³), the control variables were set constant during a running cycle, which lasted approximately 7 h. For example, the nitrate recirculation ratio was set to 80% in this study. However, if the influent nitrogen concentration varies rapidly, it is necessary to adjust the nitrate recirculation ratio timely, as the denitrification potential is influenced by the nitrate recirculation ratio. Therefore, accurate and flexible control variables have to been determined by the model using the data from online sensors.

**Benefits of carbon source addition**

The NUR under different operational conditions was measured during the experiment. In Phase 1, without a carbon source addition, the average NUR of the reactor was 2.42 mg NO₃-N/(gVSS·h). The low nitrogen removal efficiencies were mainly due to the relatively low COD/TN ratios in the influent, which affected the removal of NO₃-N through denitrification. In Phase 2, an external carbon source was added into the anoxic tank in a fixed mode. Results showed that the average NUR increased to 3.93 mg NO₃-N/(gVSS·h). In Phase 3, carbon was added according to the optimized mode. Results showed that the average NUR was 3.52 mg NO₃-N/(gVSS·h), which was higher than that in Phase 1, but slightly lower than that in Phase 2.

If the dose of carbon is higher than the amount required for denitrification, it needs to be removed with oxygen in the oxic zone, which can lead to additional aeration requirements. The comparison of average total oxygen demand for COD and nitrogen removal in 13-day-old sludge is shown in Table 5. During operation, the influent flow rate was constant. From Phase 2 to Phase 3, carbonaceous oxygen demand declined remarkably from 341.2 to 292.4 g O₂/d due to the reduction in carbon source addition; nitrification oxygen demand exhibited only slight variations due to the concentration of influent TKN being almost the same. Oxygen recovered by denitrification presented no significant difference between the last two phases, implying that carbon source was not a limiting factor for denitrification. However, there were substantial differences in net total oxygen demand due to the different control strategies of carbon addition. Optimizing external carbon source addition as determined by the model obviously saved aeration consumption.

Many domestic WWTPs still suffer from a lack of available carbon for enhanced nutrient removal (Park et al. 2011). It is known that nitrate plays an important role in triggering the activity of OHOs, which remove nitrate using organic substrates as electron donors. However, excessive carbon loading not only causes additional aeration consumption,

**Table 5** Comparison of average total oxygen demand for COD and nitrogen removal for 13-day-old sludge

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Unit</th>
<th>Phase 2</th>
<th>Phase 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Influent flow</td>
<td>m³/d</td>
<td>5.4</td>
<td>5.4</td>
</tr>
<tr>
<td>Influent COD concentration</td>
<td>mg/L</td>
<td>152ᵃ</td>
<td>135ᵇ</td>
</tr>
<tr>
<td>Influent TKN concentration</td>
<td>mg/L</td>
<td>24.9</td>
<td>24.0</td>
</tr>
<tr>
<td>Effluent TKN</td>
<td>mg/L</td>
<td>5.3</td>
<td>5.1</td>
</tr>
<tr>
<td>Carbonaceous oxygen demand</td>
<td>g O₂/d</td>
<td>341.2</td>
<td>292.4</td>
</tr>
<tr>
<td>Nitrification oxygen demand</td>
<td>g O₂/d</td>
<td>346.7</td>
<td>349.2</td>
</tr>
<tr>
<td>Oxygen recovered by denitrification</td>
<td>g O₂/d</td>
<td>182.0</td>
<td>176.3</td>
</tr>
<tr>
<td>Net total oxygen demand</td>
<td>g O₂/d</td>
<td>505.9</td>
<td>465.3</td>
</tr>
</tbody>
</table>

ᵃIncluding carbon source addition under fixed mode.
ᵇIncluding carbon source addition under optimized control mode.
but can also lead to poor denitrifier communities, resulting in low nitrogen removal efficiency (Wang et al. 2013). Therefore, an appropriate carbon amount is vital to nitrate removal and energy savings. In the fixed mode, the carbon source dosage ensured that effluent nitrate and TN met the discharge standards under the most unfavorable situations. In the optimized control mode, the addition of carbon was optimized to achieve nitrogen removal and avoid excess carbon. Results indicated that the addition of carbon source was dramatically reduced during Phase 3. External carbon addition is a major operational cost of wastewater treatment systems; therefore, minimizing carbon addition directly reduced costs. Another benefit of this advanced carbon source control strategy was the reduction of aeration expenditure. Net oxygen demand decreased from 505.9 g/d in Phase 2 to 465.3 g/d in Phase 3, resulting in an 8.0% saving in aeration costs (Table 5). In summary, the optimized control mode for carbon source addition provided a feasible way to improve treatment efficiency while minimizing costs.

Implications of the results and future study

Biodegradable oxygen demand over 5 days (BOD₅) is mostly lower than 75 mg/L in domestic wastewater (Shao 1999); therefore, the lack of an available carbon source is a widely limiting factor for nitrogen removal in WWTPs. External carbon source addition is an effective approach for enhanced nitrogen removal. Moreover, removal rates and economic costs should be taken into account when operating a WWTP in practice. This study proposed a feasible method to enhance nitrogen removal and reduce operational costs using an optimized control mode for treating low-carbon wastewater. The practical application of an online sensor system in the Jiangxinzhou WWTP showed that optimal carbon source addition coupled with optimized nitrate and sludge recirculation ratios significantly improved removal efficiency. In future research, a guide to the optimized control of the Jiangxinzhou WWTP will be developed for operators based on real application results.

During the overall operation period, ammonium was completely oxidized, even at the low temperature of 13 °C in winter, as shown in Figure 5. However, low temperature affects the biochemical reaction rates of denitrification (Gujer et al. 1999). Gujer et al. (1999) reported that heterotrophic maximal growth rates were 1.0 and 2.0 d⁻¹ at 10 and 20 °C, respectively. With the increase in temperature, the activity of denitrifying bacteria was enhanced. Therefore, the carbon source dose could be further reduced in summer through optimized control, and the proposed method could be applied throughout the year.

As the basic principle of biological nitrogen removal process has been well recognized and the active sludge model has been widely accepted by international water industries, the proposed control system is applicable to optimize external carbon source dosage when the specific operating parameters, such as SRT, sludge recycling ratio, nitrate recirculation ratio, are obtained. Therefore, the developed optimization method in this study could be broadly implemented in other WWTPs using activated sludge processes. It is well-known that the equivalent nitrate load from the aerobic tanks and secondary settler are related to the nitrate recirculation flow rate and sludge recycling flow rates. Therefore, carbon source addition combined with the optimization of nitrate, sludge recirculation ratios and aeration control could improve overall system performance.

CONCLUSIONS

This study showed that denitrification could be increased by supplementing the process with sodium acetate. Application of online sensors combined with a numerical model provided an efficient and economical way to determine the dosage of carbon required. Using the proposed approach, carbon source addition and aeration costs were significantly reduced compared with the fixed-dose mode. Preliminary application of the method to the Jiangxinzhou WWTP showed promise with regards to improving nitrogen removal in a full-scale plant.

ACKNOWLEDGEMENTS

The authors are grateful for support from the Jiangsu Water Protection Project (2015005), Jiangsu Science Fund (BE2016617) and the Yangzhou Water Authority. We thank Catharine Rice for proofreading the English.

REFERENCES

American Public Health Association (APHA) 2005 Standard Methods for the Examination of Water and Wastewater, 21st edn. APHA, Washington, DC, USA.


