Article

Process Energy and Material Consumption Determined by Reaction Sequence: From AAO to OHO

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Abstract: The anaerobic-anoxic-oxic (AAO) process is one of the most widely used processes for treating industrial organic wastewater, and it has shown significant effectiveness in the removal of organic compounds, as well as denitrification and phosphorus removal. However, for the treatment of industrial organic wastewater, this anaerobic preposition and aerobic postposition process has exposed various limitations. Therefore, for this type of wastewater, the oxic-hydrolytic and denitrification-oxic (OHO) treatment process has been proposed and developed based on the principles of three-sludge separation and fluidization. This study integrated operational data from 203 coking wastewater treatment plants worldwide, and the two-step nitrification-denitrification activated sludge model No.3 (TCW-ASM3) was used for comparative analysis of the pollutant removal efficiency and total operating cost of the AAO process and the OHO process in the face of characteristic pollutants in coking wastewater. The results indicate that the full-scale OHO process achieved removal efficiencies of up to 3784 mg/L for chemical oxygen demand (COD) and 297 mg/L for total nitrogen (TN). The theoretical total cost for OHO and AAO were 9.75 and 14.38 CNY/m³, respectively. The pre-treatment aerobic process effectively reduces the biological toxicity of high-toxicity and refractory industrial wastewater, and the three-sludge system provides a stable living space for functional microorganisms, the combination of multi-mode denitrification processes offers new possibilities for treating similar types of industrial wastewater.

Keywords: oxic-hydrolytic and denitrification-oxic process (OHO); coking wastewater; wastewater treatment technology; economic analysis

1. Introduction

Wastewater is a collection of metabolites, residual elements, and soluble substances produced by human activities that have no useful value. After undergoing appropriate wastewater treatment processes, it can be reused in human production activities or discharged into the ecological environment [1]. “Water resource reclamation” is a concept of circular utilization that involves the treatment and transformation of waste and by-products in water to achieve the reuse of food, animal by-products, resources, and energy [2]. The concept is intended to promote the sustainable development of wastewater treatment processes, reduce reliance on natural resources, and decrease environmental pollution [3,4]. Since the mid-18th century, the industrialization process of human society has developed rapidly, and the discharge volume and complexity of industrial wastewater have
also increased, causing serious pollution to rivers and lakes. As the industrialization process rapidly develops, wastewater treatment technology has also made corresponding progress [5]. In recent years, the emergence of technologies such as anaerobic-oxic (AO), anaerobic-anoxic-oxic (AAO), oxic-hydrolytic and denitrification-oxic (OHO) and oxic-hydrolytic and denitrification-hydrolytic and denitrification-oxic (OHHO) indicates that human understanding of the wastewater solution properties (WSPs) has become increasingly profound [6–9]. To ensure the effective treatment of industrial wastewater with complex, high-concentration, and highly toxic components, and to reflect the economic value of water resource reuse, it is urgent to design and adopt water treatment technologies that are compatible with the WSPs.

The AAO, as the most mainstream industrial wastewater and municipal wastewater treatment technology in the world, is primarily motivated by denitrification and phosphorus removal while also being accompanied by the removal of pollutants such as chemical oxygen demand (COD) and suspended solids (SS) [10]. In the anaerobic stage (A1), it mainly converts some refractory organic compounds and improves the biodegradability of wastewater while providing a carbon source for the subsequent anoxic stage (A2) denitrification process. In the aerobic stage (O), ammonia nitrogen and organic nitrogen are converted to nitrate nitrogen by nitrifying bacteria and recirculated to the anoxic stage for denitrification; at the same time, the organic matter is thoroughly degraded and mineralized. Under the condition of continuous inflow, the AAO regulates operational parameters such as hydraulic retention time (HRT) and organic loading rate (OLR) to create a favorable environment for the survival of functional bacteria, thereby removing pollutants from wastewater [11]. However, in many cases where AAO is used as the main process for treating industrial wastewater, several common issues have also been exposed and are worth attention [9,12]. The relevant engineering cases indicate that the removal efficiency of toxic and refractory organic compounds in industrial wastewater is very low in the anaerobic stage of the AAO [13]. When the effluent from the AAO process is discharged into the natural aquatic environment, the organic compounds/residual compounds carried in the high DO effluent enter the receiving water body, altering the aqueous solution properties (ASPs) and subjecting the ecological microorganisms to environmental stress. If the influent COD concentration exceeds 1500 mg/L and the OLR is over 0.8 kg COD/m³·d, significant toxicity inhibition occurs, resulting in a COD removal rate of less than 2% for coking wastewater (CWW) treatment. When there are high concentrations of organic nitrogen and inorganic compounds containing covalent bonds with nitrogen, such as CN⁻, SCN⁻, EDTA, etc., the AAO may face challenges and cannot effectively remove total nitrogen (TN) [14]. This will impose a severe burden on subsequent biochemical treatment processes. In addition, the functional division of sludge in the AAO is unclear, with carbon removal and nitrification functions overlapping in the aerobic stage. This overlap reduces the efficiency of nitrification and results in incomplete denitrification in the anoxic stage [15]. As a result, it is theoretically impossible to achieve the required standard discharge of total nitrogen. What is worth noting is that the AAO, which is a water treatment process relying on nitrified liquid reflux, unavoidably operates under conditions of high energy/material consumption [16]. This leads to the inefficient utilization and management of internal resources and external input energy in wastewater, making it difficult to reflect the economic value of the wastewater itself. At the same time, each unit and facility mechanically carries out reflux operations, making it difficult to reasonably control the denitrification pathway based on the WSPs. The entire process fails to demonstrate the editability of water treatment processes and is insufficient to deal with the treatment of complex industrial wastewater. Therefore, it is crucial to match the water treatment process according to the WSPs.

The rational design of water treatment processes relies on a profound understanding of the WSPs. The OHO, developed based on the principles of three sludge separation and fluidization, is highly competitive and successfully applicable in the treatment of industrial wastewater with high toxicity, high pollutant concentrations, and high organic loads.
(especially wastewater with a high proportion of organic nitrogen in TN) [17]. In the OHO, the O1 unit is responsible for COD removal, detoxification, and ammonification, ensuring the removal of organic pollutants and the effect of pre-nitrification. The H unit functions as a denitrification and hydrolysis unit, where the majority of TN is converted and removed. Through flexible process control, TN can be achieved to meet ultra-low discharge standards (like below 5 mg/L). The design of the O2 unit aims to ensure complete nitrification and carbon removal, ensuring compliance with effluent ammonia nitrogen limits and preventing COD exceedance. The design of OHO is creatively placing the aerobic unit as a pre-treatment step (O1); by utilizing aerobic microorganisms to selectively oxidize organic pollutants (reducing the biochemical oxygen demand to achieve a high N/C ratio) and other toxic substances under high loading conditions, this process effectively combats high toxicity loads and achieves significant removal of organic pollutants [18]. This, in turn, reduces water toxicity during the process and minimizes toxicity inhibition of functional bacteria such as ammonia-oxidizing bacteria and anaerobic ammonia-oxidizing bacteria. Furthermore, the aerobic process also consumes some carbon sources in the raw water, relieving the inhibitory effect of high-concentration carbon sources on microbial growth and metabolism in the anaerobic ammonia oxidation (anammox) process [19]. Additionally, this process facilitates the complete ammonification of nitrogenous organic compounds and toxic nitrogenous inorganic substances, which may potentially accumulate. This helps to achieve the initial conditions necessary for anammox, thereby ameliorating the aquatic solution properties (ASPs) and improving environmental suitability. Furthermore, the OHO is coupled with specific biological fluidized bed reactors (BFBR) (see Figure S1), creating its unique three-sludge treatment system (separating and expressing the functions of activated sludge in each unit reactor). This effectively avoids issues such as low nitrification efficiency caused by the confusion of functional bacterial populations in a single sludge system [17]. As a result, the active sludge microorganisms can make reasonable use of carbon sources, DO, and other resources within specific spatial constraints. In summary, the OHO process, combined with BFBR technology, allows for a more efficient and rational utilization of internal and external resources and energy in the treatment of industrial wastewater, such as CWW. Compared to mainstream CWW processes like AAO, it effectively reduces the biological toxicity of the wastewater and the concentration of target pollutants. It can achieve the rational distribution of internal elements in the wastewater under lower consumption conditions, thereby enhancing the biodegradability of the wastewater and its economic reuse value. Therefore, this study compared the reaction sequences on the AAO and OHO process platforms to investigate how “reaction sequences” affect pollutant removal efficiency and the overall energy/material consumption of the process, seeking a low-consumption, high-efficiency and less-discharge industrial wastewater treatment process is a highly necessary task [20].

The research is based on the actual engineering case of CWW treatment in Phase III of Shaoang (refer to Table S1 and Figure S2). The aim is to compare the mainstream CWW treatment process (using the AAO as an example) with a new CWW treatment process (the OHO) based on the CWW solution properties to determine the reaction sequence. The comparison focuses on the advantages and disadvantages of energy consumption, the removal efficiency of target pollutants, and environmental friendliness. Through this comparative study, we aim to gain a clearer understanding of the future water treatment process construction model and development trends [21]. The main research work includes (1) a quantitative comparison of AAO and OHO based on CWW treatment by collecting data from the literature to investigate their effectiveness in regulating the WSPs and resource utilization efficiency under the same HRT conditions; (2) an in-depth exploration of the practicability of the OHO compared to the AAO from various aspects, including wastewater elemental composition, WSPs, and reaction sequences; (3) using the two-step nitrification-denitrification activated sludge model No. 3 (TCW-ASM3) to simulate and optimize the operational conditions of the OHO, further seeking energy-saving potential
for this process; (4) and finally, through data comparative analysis, elucidation of biological denitrification principles, and model simulation optimization, the study provides an outlook on the operational potential of the OHO in reducing energy consumption, improving treatment efficiency, and reducing emissions. In conclusion, this study provides a systematic evaluation of the feasibility of using the OHO as a replacement for the AAO in industrial wastewater treatment, through an in-depth exploration of key indicators such as wastewater elemental transformation modes, material utilization efficiency, and development potential. This study promotes the scientific development of water treatment processes and provides scientific reference and technical support for the rational selection of industrial wastewater treatment processes in the future.

2. Materials and Methods

2.1. Data Collection and Analysis

To enhance the authority and representativeness of the AAO process-related data in this study, statistical analysis was conducted on wastewater data from a total of 203 coke wastewater treatment plants in 16 countries across 6 continents worldwide. The number of coking wastewater treatment plants (CWTP) in China included in the statistics is 108, and the quantity and specific distribution are shown in Figure 1a. The changes in COD and TN in the inflow and outflow of CWW treated by the AAO are illustrated in Figure 1b,c, respectively. The relevant data for the new OHO for treating CWW, which is based on the ASPs of CWW and determines the reaction sequence, were collected from the phase III of the CWW treatment project at Shaoguan Iron and Steel Co., Ltd., China, a subsidiary of China Baowu Steel Group. The sampling points for the wastewater were set at the outlets of each reactor (refer to Text S1 for details). Sampling was conducted at 9:00 am every day, and samples were collected every other day. After sampling, all physicochemical and wastewater quality indicators were immediately tested, followed by three analyses of distilled water as blank samples (refer to Figure S3). The required chemicals and instruments for the experimental process are described in detail in Text S2.
2.2. The Description of AAO and OHO

The AAO flow is shown in Figure 2a and can remove the target pollutants and improve wastewater biodegradability through nitrification liquid reflux and the addition of external chemicals (operating conditions are shown in Table S1). However, relying on the reflux of nitrification liquid and sludge in a single-sludge wastewater treatment system for denitrification inevitably leads to issues such as a single denitrification pathway, incomplete pollutant removal, high energy and resource consumption, and unstable system processing load. Therefore, an excellent CWW treatment process should start from the properties of the wastewater solution, clearly define the various functions of the sludge system, and design a reasonable process reaction sequence to achieve efficient reduction of pollutant concentration under conditions of low energy and less greenhouse gas emissions, and long-term stable operation. The OHO flow is shown in Figure 2b, the OHO process allows for flexible control of influent methods and operating conditions (Table S2) to meet the treatment requirements of different wastewater characteristics. The unique influent method in the OHO process is called “step-fed,” which refers to the direct pumping of raw water into the H-tank without passing through the first O-unit. Step-fed not only enables the direct utilization of some carbon sources in the raw water but also combines partial aeration in O1 to achieve short-distance denitrification. This process reduces excess nitrate to nitrite while eliminating residual dissolved oxygen, creating a suitable water-quality environment for subsequent anaerobic ammonia oxidation in the H-tank. As a result, the addition of external chemicals is significantly reduced, leading to clean and efficient nitrogen removal. We use R1 to represent the step-fed rate, typically with R1 ≤ 30%. The H unit used for hydrolysis and denitrification is more advanced and reasonable compared to the A2 unit in the AAO. It allows for the coupling of multiple denitrification pathways, including anammox coupled with autotrophic denitrification and heterotrophic denitrification. For example, research conducted by Li and others has found that when the organic load and dissolved oxygen (DO) in the influent of the OHO are within the ranges of 0.80–1.35 kg/m³·d and 3.0–5.0 mg/L, respectively, the toxic pollutant CN⁻/SCN⁻ in the wastewater can inhibit nitrite-oxidizing bacteria, thereby achieving stable partial nitrification [22]. Moreover, when combined with the flexible and controllable operational modes of the OHO, suitable reaction conditions such as substrate, pH, DO, and temperature can be provided for subsequent nanamox processes, enabling better wastewater treatment results and making it possible to achieve ultra-low TN discharge in the effluent. For a more detailed description of anammox, please refer to Text S3. Due to the significant relationship between the physicochemical techniques of water treatment
processes and the dosage of chemicals such as ferric ions and activated carbon, a greater investment in chemical dosage generally yields better physicochemical treatment results. Therefore, this study only compares and analyzes the operational stability, the removal of target pollutants, the ability to utilize internal and external resources in wastewater, and the environmental impact of the two processes during the biological stage.

![Figure 2. Process diagrams of AAO (a) and OHO (b).](image)

2.3. Analytic Method

Using a pH meter to measure the pH of the samples, the alkalinity of the solution was determined by acid titration. The concentrations of sulfate, cyanide, COD, TN, and ammonia nitrogen were determined using spectrophotometric methods. Additionally, we calculated and plotted the average concentrations of relevant indicators in the samples from the two processes, which operated continuously for 120 days with sampling every two days [23]. The calculation for the removal of target pollutants is as shown in Equation (1):

\[
\text{Pollutant removal from wastewater (\%)} = \frac{C_i^1 - C_i^e}{C_i^1} \quad (1)
\]

where \(C_i^1\) and \(C_i^e\), respectively, refer to the influent and effluent pollutant concentrations (mg/L) of unit a.

2.4. Establishment of Energy Consumption Calculation Model

The energy consumption of the water treatment process can be mainly divided into wastewater conveyance system (Wp), aerobic aeration system (Wa), mixing system (Wh), dosing system (Wd), and energy consumption of public facilities in the wastewater treatment plant (Wo). Wastewater conveyance system and aerobic aeration system account for over 85% of the energy consumption in the water treatment process. This study elaborately decomposed the energy consumption of the wastewater conveyance system and
aerobic aeration system in the AAO and O and established an energy consumption calculation model based on this (refer to Tables S3 and S4), as shown in Equation (2) (detailed derivation process is provided in Text S4).

\[ WT = W_p + W_a + W_a + W_d + W_o \]  

(2)

2.5. TCW-ASM3 Model Preparation

TCW-ASM3 is an enhanced version of the traditional ASM model, which incorporates two-step nitrification processes and includes components such as cyanide and thiocyanate along with their biological inhibition processes [24,25]. The reaction mechanism pathway of TCW-ASM3 is shown in Figure S4. It consists of 18 components and 25 reaction processes (as shown in Table S5). Other parameters, including kinetic parameters, stoichiometric coefficients, and component parameter values, can be found in Table S6. The chemical stoichiometric matrix and dynamic calculation formulas are provided in Tables S7 and S8, respectively. The entire system runs in MATLAB r2022a software, utilizing the ode15 equation to solve the system of partial differential equations. In addition, sensitivity analysis was performed on the model (detailed in Text S5), and relevant parameters were adjusted based on the analysis results to obtain a more accurate simulation of the CWTP [26]. In summary, the modeling approach in this study is illustrated in Figure 3. Firstly, field data is collected, and a new TCW-ASM3 model is established using MATLAB software. By inputting influent water quality parameters and process operating parameters, simulation outputs are obtained. Secondly, the model parameters are calibrated by matching the simulation outputs with actual measured data. Finally, the TCW-ASM3 model is used to simulate the impact of OHO process operating parameters (R1: step-feed/R2: reflux ratio, R1 ranging from 0 to 100%, R2 ranging from 0 to 1000%) on effluent water quality and operational costs. The aim is to seek ways to achieve effluent water quality that meets the discharge standards at lower costs.

![Figure 3. Metabolic pathways of TCW-ASM3.](image)

3. Results

3.1. Analysis of the Effect of Process Combinations on Pollutant Removal

To clarify the long-term stability and process performance of the current mainstream CWTP, namely the AAO and the OHO, as shown in Table 1, this study collected literature data from over 200 CWTPs worldwide to represent the process performance of the current mainstream CWW treatment processes (AAO). In addition, the OHO used in the Phase III project of Shaogang Wastewater Treatment Plant in China was monitored for 120 days to
qualitatively and quantitatively compare the pollutant removal efficiency, operating costs, and environmental friendliness of these two processes in CWW treatment.

From Figure 4a, it can be seen that the effluent treated by the AAO for CWW cannot meet the national discharge standards (COD < 80 mg/L, TN < 20 mg/L and NH$_4^+$ < 10 mg/L). This is because a single sludge system relying on reflux is difficult to effectively and thoroughly remove dissolved organic matter, TN, and persistent organic pollutants under high toxicity and high organic load conditions. Literature data from 203 regions worldwide have confirmed this problem (more data can be found in Table S9). There are data to show that thiocyanate above 200 mg/L seemed to inhibit nitrification, but it was due to the increased loading of ammonia produced from its biodegradation. Free cyanide above 0.2 mg/L seriously inhibited nitrification, and a similar conclusion was reached in the study by Chen et al. [26,27]. The influent of CWW contains various toxic substances such as cyanides, thiocyanates, and sulfides. However, the reaction sequence of the AAO process makes it difficult to selectively reduce these highly toxic inhibitory substances at the beginning of the biochemical treatment stage and thereby hard to reduce the toxicity inhibition on the subsequent microbial activity (Figure S5) [27]. However, the OHO is based on an understanding of the coking wastewater solution properties, innovative placement of aerobic unit in front, by utilizing aerobic conditions to rapidly propagate microorganisms (compared to anaerobic conditions), reducing the unit toxicity load of sludge. This effectively degrades and converts toxic substances and a significant portion of organic compounds in the incoming water. After treatment by the O1 unit, the BOD/COD ratio (B/C) of the raw water is reduced from 0.38 ± 0.04 to 0.10 ± 0.02, significantly lowering the B/C ratio of the solution (Figure S6). The B/C ratio reflects the comprehensive effect of endogenous toxicity and exogenous environment on the degradation adaptation process of microorganisms to organic pollutants and toxic substances in wastewater [28]. A low B/C value provides favorable water quality conditions for microbial growth under low-nutrient conditions, which is beneficial for microbial nitrification, anammox, and autotrophic denitrification. In addition to effectively removing toxic pollutants in the OHO, other characteristic pollutants also have a high removal rate (Figure 4b). Therefore, from the perspective of the solution properties of wastewater, the OHO is more suitable than the AAO for treating high toxicity and high-loading industrial wastewater.

![Figure 4](image_url). The relevant indicators of treating coking wastewater using AAO (a) and OHO (b) will change along the treatment process.
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Min</th>
<th>Max</th>
<th>Tolerance Limit</th>
<th>Partial reference</th>
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<td>5.5–6.0</td>
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<td>T, °C</td>
<td>34</td>
<td>52</td>
<td>/</td>
<td>Zhao et al., 2009 [35], Smol et al., 2018 [30], Chai et al., 2018 [36]</td>
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<td>COD</td>
<td>880</td>
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<td>DO</td>
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<td>/</td>
<td>3.5–4.5</td>
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<td>4400</td>
<td>/</td>
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<td>NH$_4^+$-N</td>
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<td>57</td>
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<td>108</td>
<td>37</td>
<td>Yang et al., 2018 [40], Vázquez et al., 2006 [41]</td>
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<td>TN</td>
<td>370</td>
<td>1820</td>
<td>144</td>
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<td>Total cyanides</td>
<td>16</td>
<td>416</td>
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<td>1400</td>
<td>45.2</td>
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<td>Chlorides</td>
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<td>/</td>
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<td>B/C</td>
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<td>O tank of AAO</td>
<td>Decarburization</td>
<td>COD</td>
<td>Organics + O2 → H2O + CO2</td>
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<td>Nitrification</td>
<td>TN</td>
<td>CN⁺ + O2 → NO₃⁻ + CO₂</td>
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<td>NO₂⁻ + O₂ → NO₃⁻</td>
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<td></td>
<td></td>
<td>CN⁻</td>
<td>SCN⁻ + O₂ → NO₃⁻ + CO₃⁻ + SO₄²⁻</td>
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<td>Partial nitrification</td>
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<td>SCN⁻ + O₂ → NO₃⁻ + CO₃⁻ + SO₄²⁻</td>
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<td>SCN⁻</td>
<td>NH₃ + O₂ → NO₂⁻</td>
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<td>SCN⁻ + O₂ → NO₃⁻ + CO₃⁻ + SO₄²⁻</td>
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Notes: (1) all values are in mg/L except for T (°C), B/C, ORP (mV), Conductivity (µS/cm), turbidity (NTU), pH, and color.

3.2. Unit Reaction Analysis and Economic Cost Accounting

The purpose of water treatment is to use processes to efficiently remove pollutants, but energy consumption and environmental friendliness are also crucial in this process. The unit reactor is the foundation of the combined process, determining the superior performance of the process. Based on the changes in CWW along the process in Section 3.1 of this article, combined with data collection from the literature, the coking wastewater solution properties, and a deep understanding of the operating parameters and design principles of the two processes, we have inferred the possible target pollutant degradation models in each reactor of the two processes (Tables S10 and S11) [16]. The chemical formulas that may occur in the aerobic units of the two processes are shown in Table 2. From the table, it can be seen that for the treatment of CWW, the OHO double-O function is more comprehensive and reasonable than the AAO single-O function. O1 reduces the toxicity and organic load and meets the inlet requirements of the H unit for different denitrification pathways according to the regulation of DO and other operating parameters. The existence of O2 ensures complete carbon removal and nitrification. Compared to single-O, the combination of double-O can meet the requirements for treating high-toxicity, high-concentration, and highly fluctuating wastewater [7].

Table 2. Proposed AAO and OHO biodegradation model of coking wastewater.

The consumption of chemicals and electricity accounts for over 80% of the operating costs of water treatment. Table in Figure 5 provides a detailed breakdown of the operating costs and cost components of the two processes per cubic meter of CWW. Taking the labor
cost as an example, using actual data from the Shaoguan Steel Coking Wastewater Treatment Plant, the labor cost of both processes is set at 0.79 CNY/m³ for discussion. This study theoretically calculated that under full-scale process operation conditions, the aeration cost of the AAO is 9.01 CNY/m³, which is 2.56 RMB higher than the cost of treating each cubic meter of CWW using the OHO. Although the OHO adopts a design with two aerobic units, it effectively degrades most of the COD and other target pollutants by regulating the pre-aerobic O₁ unit to operate under low DO (1.5–2.0 mg/L) and short HRT (40 h) conditions, thereby significantly reducing the aerobic degradation pressure of the O₂ unit. Relevant literature indicates that the AAO must ensure complete nitrification through a large amount of aeration in order to achieve good denitrification in subsequent stages. The gas/water ratio in the O tank often exceeds 50:1. As for OHO, after pre-aerobic treatment, the O₂ unit only needs to consolidate and achieve complete nitrification. Combined with the unique BFBR technology, the process efficiently removes various target pollutants by utilizing oxygen resources reasonably and fully within the reactor. This is achieved under conditions where the gas/water ratio is only (20–35):1. In the actual engineering of Shaogang Phase III, the aerobic unit of the AAO process often requires 80 h of aeration time to achieve good removal of target pollutants in the influent. The total aeration time of the OHO’s dual-aerobic units (70 h) is lower than 80 h, and it operates at a lower DO level, significantly reducing the operating cost of the aeration system.

Figure 5. The detailed operating cost and cost composition of the two processes.

The reflux of nitrified liquid in the AAO can cause a large amount of sludge loss in the O reactor, resulting in a decrease in the system’s sludge concentration and disruption of microbial metabolism. At the same time, the dilution effect of recirculation suppresses the biological reaction dynamics of the A2 tank. The high DO recirculation liquid also destroys the anaerobic environment of the A2 tank, which not only increases the operating cost (2.56 CNY/m³) but also interferes with the water treatment efficiency of the process to some extent.

Compared with the AAO, due to the partial nitrification in the aerobic pre-treatment stage, the OHO can also undergo denitrification reactions in the H reactor by adding chemicals without the need for nitrified liquid recirculation. In addition, OHO is a clear and independent multi-sludge system, with clearly defined and stable microbial functions in the reactor [5]. Combined with the screening and interception of microbial communities in the reactor by BFBR, the entire system can maintain sufficient microbial quantity in each reactor without the need for sludge recirculation, ensuring the treatment effect of the entire process.
During the operation of the OHO, in addition to efficiently utilizing oxygen through BFBR technology, there is also sufficient utilization of internal carbon sources and other resources in the wastewater. By flexibly adjusting the process mode and operating parameters, a small amount of raw water is bypassed to the H tank, maximizing the utilization of resources in the raw water to reduce the additional dosage of external chemicals. This article provides detailed information on the consumption of all chemicals during the operation of the AAO (Table S12) and the OHO (Table S13). Through calculation, the chemical consumption cost of the OHO is 0.59 CNY/m³, which is significantly lower than the chemical consumption of the AAO process (1.18 CNY/m³). This may be because the AAO requires more ferrous sulfate to reduce the toxicity of the raw water and does not fully utilize the carbon source in the raw water during the biochemical treatment process, resulting in the need for more external carbon source addition.

At the same time, the increase in chemical dosing leading to increased carbon emissions should not be overlooked. We categorize the sources of carbon emissions into those generated from pollutant removal, electricity consumption, and chemical dosing. The carbon emissions from pollutant removal can be further divided into CO₂ generated from COD degradation and NO₂ generated from TN removal. According to Table S14 and Figure S7, the carbon emissions of the OHO process are 18.025 kg CO₂-eq/m³, which is 2.911 kg CO₂-eq/m³ lower than those of the AAO. In summary, for CWW, conventional wastewater treatment processes (such as AAO) are difficult to effectively remove toxic pollutants, dissolved organic matter, and other target pollutants. Moreover, in the current era of advocating for environmentally friendly processes, it is even more challenging to realize the economic value of wastewater reuse through the sorting role of water treatment processes. In contrast, the OHO demonstrates its unique advantages and potential applications in these aspects.

### 3.3. Actual Engineering Verification

To verify the cost difference between the AAO and the OHO, the two processes were calculated for electricity cost, chemical addition cost, and labor cost at 10-day intervals under their respective optimal operating modes and process parameter conditions (see Tables S1 and S2). The Pearson equation was used for linear fitting, and the fitting results are shown in Figure 6. It can be seen that the total operating cost of the full-size AAO is 14.38 CNY/m³, which is 4.53 CNY/m³ higher than that of the OHO. The construction investment to transform the AAO into the OHO is about 12 million CNY. Based on the actual operation of the current OHO project and considering maintenance, wear and tear, etc., it is expected that the project net profit can be achieved in 7–8 years.

![Graph showing operating cost comparison between AAO and OHO](image-url)
Figure 6. Two kinds of process operation cost calculation.

Traditional CWW treatment processes tend to increase the aeration rate in the aerobic unit and raise the nitrification liquid reflux ratio (up to 5–7) to ensure that the treated effluent meets increasingly strict discharge standards. However, this leads to higher energy consumption for aeration and pumping and imposes more demanding requirements on plant operators. The operating costs of the OHO, determined by the reaction sequence, are theoretically lower than AAO. During the operation of the OHO, the deep understanding of the coking wastewater solution properties, advanced BFBR technology, and flexible control mode of full-scale processes contribute to the better water quality of the effluent and an increase in its reuse value. As a result, the total operating costs of the process can be reduced by over 35%.

3.4. OHO Performance Optimization Analysis

In order to further develop and optimize the performance of OHO, based on the TCW-ASM3 model developed by T. Wei, some parameters in the model were adjusted [22]. The calculation modules for energy consumption and other factors were also re-edited (Text S4) to analyze the total operating costs of the OHO and identify potential areas for optimization. For the sake of assessing the impact of each parameter on the overall model and facilitating the calibration of model parameters, it is necessary to conduct a sensitivity analysis on the TCW-ASM3 model. The results of parameter comparison and sensitivity analysis are shown in Figure S8. After calibrating the key parameters of the model, we adjusted the TCW-ASM3 model to simulate the total operating costs of the biological treatment stage in the OHO for CWW treatment.

The TCW-ASM3 model was validated by comparing the measured effluent values with the simulated values under the same influent conditions. During a 120-day operation of the OHO, the TCW-ASM3 model was run based on the real-time characteristics of the actual influent CWW, with the effluent COD concentration and TN concentration as the output variables. The measured values were taken every two days (60 data points). As shown in Figure S9, the R2 values for COD and TN between the measured and simulated values of CWW were 0.72 and 0.78, respectively. The simulated values closely matched the measured values, confirming the accuracy of the model. This result indicates that the TCW-ASM3 model has successfully simulated the total operating costs of the OHO for CWW treatment.

The TCW-ASM3 model was used to simulate the total operating costs of the biological stage in the OHO, so as to seek the lowest possible cost while meeting the effluent discharge standards. The total cost includes electricity consumption (mainly for aeration and pumping), chemical consumption, and labor costs. Changes in R1, R2, and DO will affect the cost and O2 effluent quality of the biological stage in the OHO process. In the TCW-ASM3 model, R1 varies from 0 to 100% and R2 varies from 0 to 1000%, with increments of 5%. DO is set at 3.0 mg/L, <0.1 mg/L, and 4.0 mg/L in the O1, H, and O2 units, respectively, which is the same as the actual operating conditions of the OHO in Shaoguan. This accurately simulates the total operating costs of the entire system during actual operation. Increasing R1 and R2 undoubtedly increases the pumping electricity consumption of the system, as shown in Figure 7a. Increasing R2 throughout the process leads to increased pumping electricity consumption for recirculation. Additionally, more recirculation liquid needs to be dosed with more chemicals in the H unit, resulting in an increase in the total cost of the entire process with increasing R2. When R1 is 55% and R2 is 1000%, the total treatment cost of the entire system reaches its maximum value of 7.59 CNY/m³. Beyond this point, further increasing R1 does not continue to increase the system cost. As R1 increases, although pumping electricity consumption increases, the aeration electricity consumption required for the O1 unit decreases. Typically, aeration electricity consumption accounts for about 60–80% of the total electricity consumption in a CWTP.
Therefore, increasing R1 within the range of 0–55% will also increase the total energy consumption of the system. Increasing R1 and R2 undoubtedly increases the pumping electricity consumption of the system, as shown in Figure 7a. Increasing R2 throughout the process leads to increased pumping electricity consumption for recirculation. Additionally, more recirculation liquid needs to be dosed with more chemicals in the H unit, resulting in an increase in the total cost of the entire process with increasing R2. When R1 is 0% and R2 is 100%, the total treatment cost of the entire system reaches its maximum value of 11.45 CNY/m³. Beyond this point, further increasing R1 does not continue to increase the system cost. This may be because more influent water enters the H unit through step-feed, and the carbon source in the influent raw water is more fully utilized, reducing the addition of external chemicals and slowly decreasing the total operating cost (see Tables S12 and S13 for details). When R1 is too large (R > 80%), most of the influent water is directly pumped into the H unit without being treated in the pre-aeration O1 unit. This causes the H unit to be unable to handle the high processing load of the influent water, leading to system failure and a sharp decrease in effluent water quality. In this case, although the pumping electricity consumption for R1 surpassing the influent water gradually increases, the recirculation of R2 and the dosing of chemicals for the entire system are gradually suspended, resulting in a significant decrease in the total cost of the system. When R1 is 100% and R2 is 0%, the overall cost of the system is the lowest (4.03 CNY/m³). However, the effluent water in this case does not meet the wastewater discharge standards. In the case of meeting the wastewater biological discharge standards (COD < 200 mg/L, TN < 50 mg/L), the total operating cost ranges from 10.31 CNY/m³ to 11.48 CNY/m³ (Figure 7b). It is worth noting that the most effective mode in actual wastewater treatment projects is Mode III (R1 = 20%, R2 = 100%). The simulation results show that the total treatment cost in this mode is 9.14 CNY/m³, which is lower than the actual total treatment cost of 10.48 CNY/m³, indicating that there is still room for optimizing the total treatment cost when the actual project operates under this condition. In summary, under the simulation optimization of TCW-ASM3, the OHO can have lower operating costs in engineering through parameter adjustment.

![Figure 7](image.png)

**Figure 7.** Full range operation cost (a) and energy consumption of reaching the emission target (b).

4. Conclusions

This study takes the treatment of phase three CWW in Shaoguan as an example and proposes a new process strategy using the OHO to replace the current mainstream wastewater treatment process (AAO), based on the design principles and actual engineering performance. A comprehensive comparison and analysis of the removal efficiency of characteristic pollutants in CWW and the total operating costs between the AAO and the OHO are conducted, the main conclusions are as follows: (1) Industrial wastewater commonly contains a high percentage of TN contributed by non-ammonia nitrogen, which
inhibits AAO from thoroughly removing TN. OHO, as a process feature, focuses on carbon and ammonia removal as well as alleviating toxic inhibition; it contributes to the establishment of water quality conditions for “nitrite accumulation-anammox”, providing theoretical support for low-consumption denitrification. (2) The in-situ sludge separation BFBR provides a platform for the three-sludge operation mode in the OHO, separating HRT and SRT; this eliminates functional conflicts among microorganisms, enabling them to perform specific functions. Additionally, there is no need for sludge or nitrification liquid reflux, resulting in a several-fold increase in reaction kinetics, which enhances the efficiency and thoroughness of the reactions. (3) The OHO changes the oxygen supply load, eliminates reflux power, and employs a dual-O cascade control to reduce the target pollutant concentration of effluent, with thorough nitrification and complete mineralization as its process characteristics. This eliminates the risk of exceeding the ammonia nitrogen limit. (4) The editability of the OHO denitrification pathway allows for the normalization of various industrial wastewater influent qualities and regulation of effluent targets, making it particularly suitable for extending the functionality of existing AAO, such as the application of the OHHO [7].

The low-consumption, high-efficiency, and lower-emission treatment of industrial wastewater is a highly attractive and challenging research area. The units and functions of complex wastewater treatment processes should aim to achieve the desired effluent water quality while minimizing temporal and spatial constraints and energy and resource consumption. In summary, this study proposes that OHO may be the most suitable biotechnology currently available for treating CWW. It also provides a new approach and platform for the treatment of highly toxic and refractory industrial wastewater, with the potential to become a mainstream treatment process in the field of industrial wastewater treatment.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w16131796/s1, Figure S1. Schematic diagram of biological fluidized bed reactor (a) and Practical engineering of biological fluidized bed reactor (b). Figure S2. Map of each process unit in Shaoguan III. Figure S3. Photographs of the sampling process and the project site. Figure S4. Metabolic pathways of TCW-ASM3. Figure S5. Toxicity changes after O1 treatment. Figure S6. BOD/COD ratio changes after O1 treatment. Figure S7. Equivalent carbon emissions in the two processes. Figure S8. Sensitivity analysis of TCW-ASM3. Figure S9. Simulated value and actual value of biological effluent in OHO process. Table S1. Operation conditions of AAO process. Table S2. Operation strategies of OHO process. Table S3. Energy-consuming equipment of the two process. Table S4. Electric motor safety coefficient. Table S5. Components in TCW-ASM3. Table S6. Stoichiometric coefficients and component parameter values in TCW-ASM3. Table S7. Stoichiometric and composition matrix of the TCW-ASM3. Table S8. Kinetic rate expression of TCW-ASM3 model. Table S9. Variation of coking wastewater concentration along the process (unit: mg/L). Table S10. Putative AAO biotransformation of coking wastewater. Table S11. Putative OHO biotransformation of coking wastewater. Table S12. The list of main chemicals used in the full-scale AAO processes. Table S13. The list of main chemicals used in the full-scale OHO process. Table S14. Carbon emissions of OHO and AAO process (unit: kg CO2-eq/m³).

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