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Identification of Key Basic Parameters Involved in Carbon Emissions in Full-Scale Wastewater Treatment Plants

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Abstract: In this study, carbon emissions in three full-scale wastewater treatment plants were determined by the emission factor method. Moreover, the correlation between basic parameters (influent water parameters and pollutant removal efficiency) and carbon emissions was examined via a structural equation model (SEM). The results showed a significant variation in the total carbon emission intensity of plants over time. The average total carbon emission intensity of plants A, B and C were 0.314, 0.404 and 0.363 kg eqCO_2/m^3, respectively. Meanwhile, the indirect carbon emission caused by energy and chemical agent consumption accounts for the majority of total carbon emissions (about 85%). Generally, statistical analysis results show that carbon emission intensity is positively correlated with pollutant removal efficiency. Notably, RTN showed the highest positive correlation with E_{ind}, followed by RTN > $RCOD_{Cr}$ > RTP > TN > RNH_3-N > NH_3-N > TP. Moreover, capacity showed the greatest negative contribution to E_{ind}, followed by COD_{Cr}. In contrast, the positive contribution to E_{dir} was followed by the sequence of RTN > $RCOD_{Cr}$ > TN > RNH_3-N > NH_3-N. Notably, COD_{Cr} showed a significantly negative correlation with E_{dir}, while TP and its removal showed little correlation with E_{dir}.

Keywords: carbon emission; structural equation model; anaerobic/anoxic/oxic oxidation ditch process

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1. Introduction

The ongoing emission of anthropogenic greenhouse gas (GHG) is triggering changes in many climate hazards that can impact humanity [1]. Wastewater treatment plants (WWTPs), as essential units of the urban water system, can contribute nearly 1~2% of the total global anthropogenic carbon emission [2]. More critically, carbon emissions continually increase due to the increased discharge of pollutants [3]. Generally, carbon emissions in WWTPs can be divided into direct carbon emissions and indirect carbon emissions [4]. Direct carbon emissions are mainly the GHGs (CO_2, CH_4 and N_2O) discharged during biological reaction processes. In addition, indirect carbon emissions mainly refer to the GHG discharged during the production of consumed energy and chemical agents in wastewater treatment. Briefly, GHG emissions from WWTPs are major contributors to the overall anthropogenic GHG emission, which should be taken seriously [5].

Currently, carbon emission reduction has become an urgently prioritized objective in the wastewater industry [6,7]. Many effective measures such as appropriate process selection, technologies with low carbon emission and precision management have been applied in order to pursue environmental sustainability [8–10]. However, the essential prerequisite of carbon emission reduction strategies, which involves the identification of key parameters involved in carbon emissions, still requires further comprehensive research. Generally, the carbon emission of WWTPs varies greatly among influent quality, the discharge standard of pollutants and operation conditions [11]. Moreover, the first two conditions, influent conditions and discharge standards, comprise the criteria of working condition adjustments and process selection [12,13]. The study of the influence of these conditions on carbon emissions is conducive to adjusting operation strategies. Generally, low influent COD_{Cr} and TN with a relatively stable C/N ratio demonstrated high carbon emission intensity. By evaluating the carbon emissions of 50 WWTPs in Shanghai, Jiarui, Xi et al. pointed out that the lowest carbon emission was obtained when influent COD_{Cr} was 150~250 mg/L and NH_3-N was 15~25 mg/L [14]. In contrast, stricter discharge limits led to a higher emission intensity [15]. Highly influent nutrients consume more oxygen and chemical agents, while poorly influent nutrients need extra carbon sources to support the growth of microorganisms, which can remove nitrogen. In particular, the influent C/N (COD_{Cr}/TN) ratio is considered one of the most significant parameters because it can markedly affect carbon emissions from both nitrification and denitrification processes [16]. By adjusting the influent C/N, Chen et al. pointed out that an influent C/N ratio of 10 would be optimal for simultaneously achieving relatively higher pollutant removal efficiency and lower GHG emissions in constructed wetlands [17]. However, there are still few comprehensive studies on the correlation between basic parameters (influent water parameters and pollutant removal efficiency) and carbon emissions in full-scale plants.

Usually, the same parameter shows different contributions to different types of carbon emission intensity. Direct emission intensity shows no significant difference between different WWTP scale groups, while indirect carbon emission intensity shows a significant scale effect. The WWTPS at small scales always obtains higher indirect emissions [14]. Meanwhile, some researchers pointed out that the improved scheduling of the influent load can reduce the energy costs of the wastewater treatment plant [18]. Xi Jiarui et al. pointed out that the indirect emission intensity of WWTPs with low COD_{Cr} and NH_3-N concentrations was twice that of other WWTPs [14]. Notably, the previous study mainly focused on the influence of influent conditions and discharge standards on carbon emissions. Unfortunately, the influence of different processes is not properly excluded.

In addition, the treatment process also has a significant impact on carbon emission intensity. The highest emission intensity is usually reported in the process using membranes with higher energy and consumed chemical agents; for example, 0.79 $kgCO_2\text{-eq}/m^3$ was reported by a previous study at the Shenzhen MBR plant [19]. Via the remolding process (anaerobic fixed-film MBR reverse osmosis–chlorination process), the carbon emission of the municipal wastewater reclamation could be 0.31 $kgCO_2\text{-eq}/m^3$ [20]. Nguyen et al. pointed out that the GHG emission intensity of AAO was one-fifth that of SBR in the WWTPs of Australia [21]. In another study, AAO and oxidation ditch processes showed a lower carbon emission per ton of water in WWTPs in Shanghai [14]. Notably, the technologies mostly used in WWTPs in China are AAO and oxidation ditches, which account for over 50% of the existing WWTPs [22]. With the implementation of the Class A Discharge Standard of Pollutants (GB18918-2002) [23] for municipal wastewater treatment plants (WWTPs), treatment processes have been upgraded and rebuilt in many WWTPs in China. An AAO oxidation ditch with larger influent loads, improved TN removal performance and lower energy costs is a typical selected treatment process for implementing upgrades [24]. However, there are few studies on the carbon emission characteristics of the AAO oxidation ditch process. A more comprehensive and in-depth study should be carried out.

In this study, data from three full-scale WWTPs that adopted the AAO oxidation ditch process were collected for conducting operation performance evaluation and carbon emission characteristics analyses. Moreover, structural equation models (SEMs) were adopted to analyze the correlation between carbon emission and the main influencing factors, including capacity, influent water quality and pollutant removal efficiency. This study provides a valuable reference to identify the key parameters involved in the carbon emission of WWTPs.

2. Materials and Methods

2.1. Wastewater Treatment System Description

Carbon emission accounting was carried out at three full-scale tertiary municipal WWTPs in Lu'an, Anhui province. Additionally, the satellite images of studied WWTPs and general information are listed in Table S1.

The influent water in the studied plants comprised mainly domestic sewage, and the effluent can meet the first class, A (from Discharge Limits of Pollutants for Municipal Wastewater Treatment Plant, China (GB18918-2002): $COD_{Cr} < 50$ mg/L, $BOD_5 < 10$ mg/L, $TN < 15$ mg/L and $TP < 0.5$ mg/L). As Figure 1 shows, the plants adopt the anaerobic/anoxic/oxic (A/A/O) oxidation ditch process for extensive nitrogen and phosphorus removal. The wastewater and externally returned active sludge first flow into an anaerobic pool, where organic matter is hydrolyzed and acidified to improve biodegradability, and phosphorus accumulation organisms (PAOs) can gain a competitive advantage in the following stage via the synthesis of poly-β-hydroxybutyrate (PHB). Secondly, wastewater flows into the oxidation ditch, and the transformation from an anoxic zone to an aerobic zone is realized by adjusting the guide wall and aeration device. Then, the solid–liquid separation of the effluent is performed in a secondary sedimentation tank. The residual nutrient will be further removed in an activated sand filter. Finally, the effluent is disinfected and discharged into the natural water body.

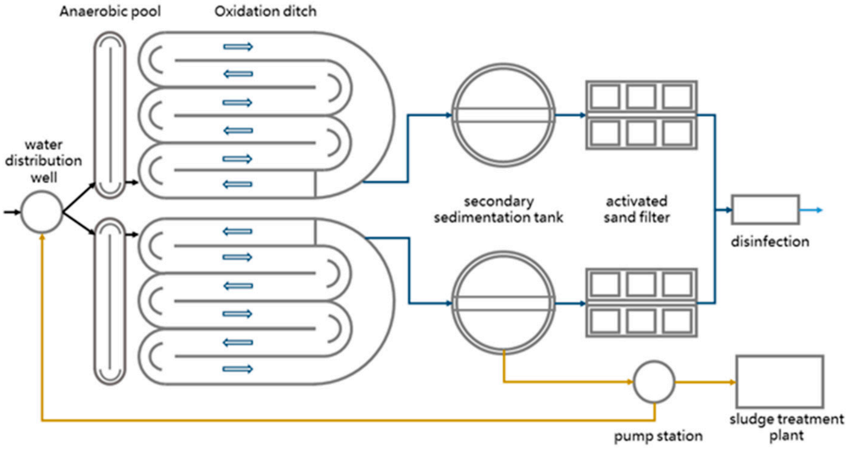

Figure 1. Simplified wastewater treatment process diagram of studied WWTPs.

2.2. Carbon Emission Accounting

Carbon emission accounting is carried out based on the emission factor method, and the parameters were selected by referring to previous studies [25,26]. Carbon emission was calculated with respect to two parts: direct carbon emission and indirect carbon emission. Direct carbon emission, E_{dir}, is the equivalent CO_2 emission that is mainly from CH_4 ($E_{dir}^{CH_4}$) and N_2O ($E_{dir}^{N_2O}$) emissions and from biochemical processing units in WWTP. In addition, indirect carbon emission, E_{ind}, refers to the equivalent CO_2 emission during the production of consumed energy and chemical reagents during wastewater treatment. They can be calculated as the following equations, which are provided by the Intergovernmental Panel on Climate Change (IPCC) [25]:

$$E_{dir} = \frac{GWP_{CH_4} \times E_{dir}^{CH_4} + GWP_{N_2O} \times E_{dir}^{N_2O}}{Q} \tag{1}$$

$$E_{dir}^{CH_4} = Q \times (COD_i) \times EF_{CH_4} \tag{2}$$

$$E_{dir}^{N_2O} = Q \times (TN_i) \times EF_{N_2O} \tag{3}$$

$$E_{ind} = EC_{ind} \times EF_{CO_2} \tag{4}$$

where GWP refers to the global warming potential within a certain future time period of the GHG. In this study, GWP_{CH_4} and GWP_{N_2O} are 25 and 298, separately [27]. Additionally, the adopted emission factor (EF_{N_2O}, EF_{CH_4} and EF_{CO_2}) in this study can be found in Tables 1 and 2.

Table 1. Description and emission factor value of GHG in previous studies.

This Study		Previous Studies		
Emission Factor	**Value**	**Value**	**Description**	**Reference**
EF_{CH_4}	0.0030 kg CH_4/kg $COD_{influent}$	0.00306	Orbal oxidation ditch in Beijing WWTPs	[28]
		0.0079	pre-anaerobic carrousel oxidation ditch in Jinan	[29]
		0.00133	oxidation ditch in the Akiu sewage treatment plant in Sendai city, Japan	[30]
EF_{N_2O}	0.0017 kg N_2O/kg $TN_{influent}$	0.00571	aeration oxidation ditch in Brisbane, Queensland	[31]
		0.00014	oxidation ditch in Akiu sewage treatment plant in Sendai city, Japan	[30]
		0.00173	Orbal oxidation ditch in Beijing WWTPs	[28]
		0.000295	plug-flow AS tank	[32]
		0.00037~0.0015	Orbal oxidation ditch in Xi'an, No.3 WWTP	[33]

Table 2. Equivalent CO_2 emission factor value for consumed energy and chemical reagent production.

Consumed Energy and Chemical Reagent	Value	Reference
electricity	0.7921 kg CO_2-eq/kWh	[34]
polyaluminium chloride	0.53 kg CO_2-eq/kg	[26]
sodium acetate	0.623 kg CO_2-eq/kg	[26]
ferric chloride	0.26 kg CO_2-eq/kg	[26]
sodium hypochlorite	0.99 kg CO_2-eq/kg	[26]

2.3. Statistical Analysis

R 4.2.1 was used for data processing and figure drawing in this study. The Pearson correlation coefficient between WWTP performance and influent parameters was calculated by using the following equation:

$$\mathrm{r}_{(X,Y)} = \frac{\sum_{i=1}^{n}\left(X_i - X\right)\left(Y_i - Y\right)}{\sqrt{\sum_{i=1}^{n}\left(X_i - X\right)^2}\sqrt{\sum_{i=1}^{n}\left(Y_i - Y\right)^2}} \tag{5}$$

where X and Y refer to the corresponding observed variables (capacity, COD_{Cr}, TN, etc.), $\mathrm{r}_{(X,Y)}$ refers to the Pearson correlation coefficient between X and Y, and subscript i represents the number of observations.

Additionally, the structural equation model (SEM) was used to analyze the relationship between basic parameters (influent water parameters and pollutant removal efficiency) and carbon emission intensity. The SEM was constructed using the lavaan 0.6–12 package in R [35], and the model parameter was estimated by the maximum likelihood method. Carbon emissions during the process are related to the capacity, influent water quality (COD_{Cr}, TN, NH_3-N, TP, etc.) and their removal efficiencies ($RCOD_{Cr}$, RTN, RNH_3-N, RTP, etc.). All plausible paths between carbon emissions and influent quality were tested. The path coefficient was used to measure the degree of influence or effect between variables. The regression associations implied by the model can be represented by the following:

$$E_{ind} = \gamma_{11} capacity + \gamma_{12} COD_{Cr} + \gamma_{13} TN + \gamma_{14} NH_3N + \gamma_{15} TP + \gamma_{16} RCOD_{Cr} + \gamma_{17} RTN + \gamma_{18} RNH_3 - N + \gamma_{19} RTP + \alpha_1 + \epsilon_1 \quad (6)$$

$$E_{dir} = \gamma_{21} capacity + \gamma_{22} COD_{Cr} + \gamma_{23} TN + \gamma_{24} NH_3 - N + \gamma_{25} TP + \gamma_{26} RCOD_{Cr} + \gamma_{27} RTN + \gamma_{28} RNH_3 - N + \gamma_{29} RTP + \alpha_2 + \epsilon_2 \quad (7)$$

$$E_{total} = \gamma_{31} E_{ind} + \gamma_{32} E_{dir} + \alpha_3 + \epsilon_3 \quad (8)$$

where γ refers to the path coefficients, α refers to the constant terms, and ϵ refers to the error terms.

The model evaluation of SEM first involved examining whether the results of parameters estimated in the model had statistical significance, testing the significance of the path coefficient and selecting the path in the model. The chi square/degrees of freedom, non-normed fit index (NNFI), adjusted goodness-of-fit index (AGFI), normed fit index (NFI) and standardized root mean square residual (SRMR) were used to evaluate the successful fit of the model.

3. Results

3.1. Characteristics of Influent Quality

The average daily concentrations of four crucial water quality parameters that correspond to the wastewater influent quality at the studied WWTPs in 2021 are shown in Figure S1. As Figure S1 shows, the monthly median influent COD_{Cr} varied from 73.5 to 175.0 mg/L in Plant A, from 88.0 to 170.0 mg/L in Plant B and from 87.0 to 145.0 mg/L in Plant C, which is close to the influent water quality of WWTPs in China reported in previous studies [36,37]. Notably, the sources of wastewater influents are mainly municipal sewage, as well as a possible mixture of stormwater and surface water. In July and August, the COD_{Cr} concentration reached an obvious "valley" and the capacity showed a dramatic increase (Figure S1a), which coincided with the food season in Lu'an.

The monthly median influent TN varied from 22.5 to 37.0 mg/L in Plant A, from 17.0 to 38.5 mg/L in Plant B and from 27.0 to 41.5 mg/L in Plant C (Figure S1b). Meanwhile, the monthly median influent NH_3-N varied from 18.5 to 34.3 mg/L in Plant A, from 11.8 to 18.9 mg/L in Plant B and from 22.7 to 34.8 mg/L in Plant C (Figure S1c). In this study, NH_3-N in influent wastewater accounted for about 48~90%, which is consistent with a previous study [37]. The high proportion of NH_3-N in influent wastewater is closely related to the expansion of human activities and industrial processes.

Additionally, the monthly median influent TP was around 2.7~4.6 mg/L in Plant A, 2.3~5.2 mg/L in Plant B and 2.4~4.2 mg/L in Plant C (Figure S1d). Notably, the monthly median capacity varied from 3210 to 4000 m^3/h in Plant A, 748 to 1255 m^3/h in Plant B and 1880 to 2160 m^3/h in Plant C, which showed a different trend compared with the influent water quality. In accordance with COD_{Cr}, the TN, NH_3-N and TP exhibited their minimum value in August. The obvious "valley" is mainly for the rainfall effects on inflow and infiltration in wastewater treatment systems [38], and mixed rainwater may greatly influence the subsequent wastewater treatment process. The impact of influent wastewater quality on carbon emissions will be discussed in detail in Section 3.4.

3.2. The Correlation between WWTP Performance and Influent Parameters

The major pollutant removal rate is shown in Figure S2. The monthly median COD_{Cr} removal rate differed from 82.0% to 92.5% in Plant A, from 87.5% to 92.5% in Plant B and from 88.0% to 94.0% in Plant C (Figure S2a). The monthly median TN removal rate varied from 64.5% to 80% in Plant A, from 68.0% to 85.0% in Plant B and from 63.0% to 74.8% in Plant C (Figure S2b). Additionally, the monthly median NH_3-N removal rate differed from 90.5% to 99.3% in Plant A, from 92.5% to 98.5% in Plant B and from 96.8% to 99.6% in Plant C (Figure S2c). In addition, the monthly median TP removal rate differed from 89.0% to 94.2% in Plant A, from 91.6% to 97.5% in Plant B and from 89.2% to 93.5% in Plant C (Figure S2d). Notably, the major nutrient removal rate was consistent with the influent

COD_{Cr}. In the influent "valley" (July and August), nutrient removal became unstable and exhibited poor performance.

The correlation between the removal rate and influent parameters is shown in Figure 2. Apparently, there is a negative correlation between capacity and influent water quality, which is further evidence of the rainfall effects on inflow and infiltration in wastewater treatment systems. Additionally, the influent TN showed a significant negative correlation with the removal of TN (RTN). This is mainly because the higher influent TN needs more carbon sources for denitrification, which is the main denitrification path in this study. Thus, RTN will decrease if there is no sufficient carbon source in the influent wastewater or the external carbon source is not obtained in a timely and precise manner. Interestingly, the TP removal rate (RTP) increased with the increase in influent TP, which was mainly due to the fluctuation of influent C/P from 23 to 33, and the variation in influent phosphorus concentrations had little effect on biological phosphorus removal efficiency. With the subsequent chemical phosphorus removal and the increase in phosphorus mass concentration, the mass concentration of iron–phosphorus precipitates in the effluent increases, and the chance of dissolved phosphorus being complexed by precipitate adsorption increases greatly, which will improve the phosphorus removal rate.

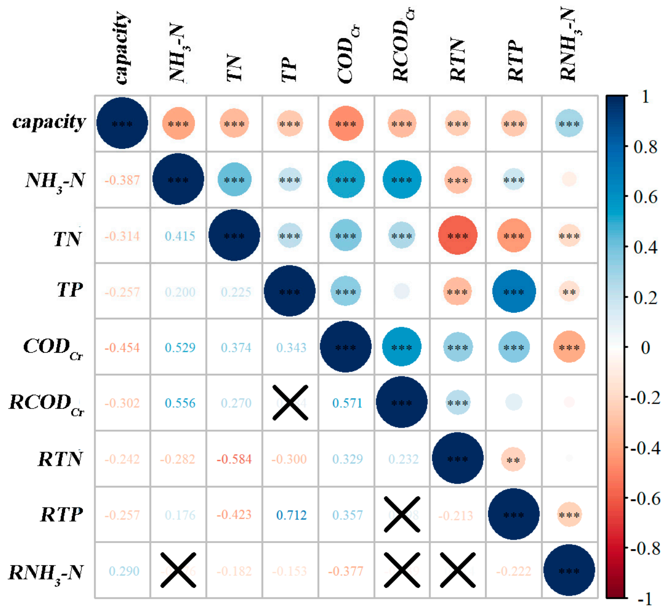

Figure 2. The correlation heatmap between influent parameters, operation parameters and (data with *p*-value > 0.05 are already hidden) influent performance parameters: capacity, COD_{Cr}, TN, NH_3-N and TP; removal efficiency: $RCOD_{Cr}$, RTN, RTP and RNH_3-N. "**"and "***" suggest that P value is less than 0.01 and 0.001, respectively. And the "×" suggests that the correlation is not significant.

In addition, there was a negative correlation between RTP and RTN, which is mainly for the competition between phosphate-accumulating organisms (PAOs) and denitrifying bacteria for carbon sources [39]. If the carbon source, which is the electron donor during denitrification, is insufficient due to the interference of biological phosphorus removal, various denitrifying enzymes will compete for electrons, leading to a decrease in denitrification efficiency and the accumulation of N_2O [40].

3.3. Carbon Emission Characteristics of WWTPs

The different types of carbon emissions of the studied WWTPs in Lu'an were accounted, and the results are shown in Figure 3. The total carbon emission intensity (E_{total}) reached a relative peak in the summer, which is due to the sudden increase in influent load and relatively low influent nutrients during the rainy season. The average monthly total carbon emission intensity is about 0.273~0.334 kg $eqCO_2/m^3$ in Plant A, 0.301~0.514 kg $eqCO_2/m^3$ in Plant B and 0.322~0.413 kg $eqCO_2/m^3$ in Plant C, which is close to previous studies [14,41]. Plant B, with a lower capacity (20,000 m^3/d), exhibits a higher total carbon emission intensity, which should be due to the scale effect [42].

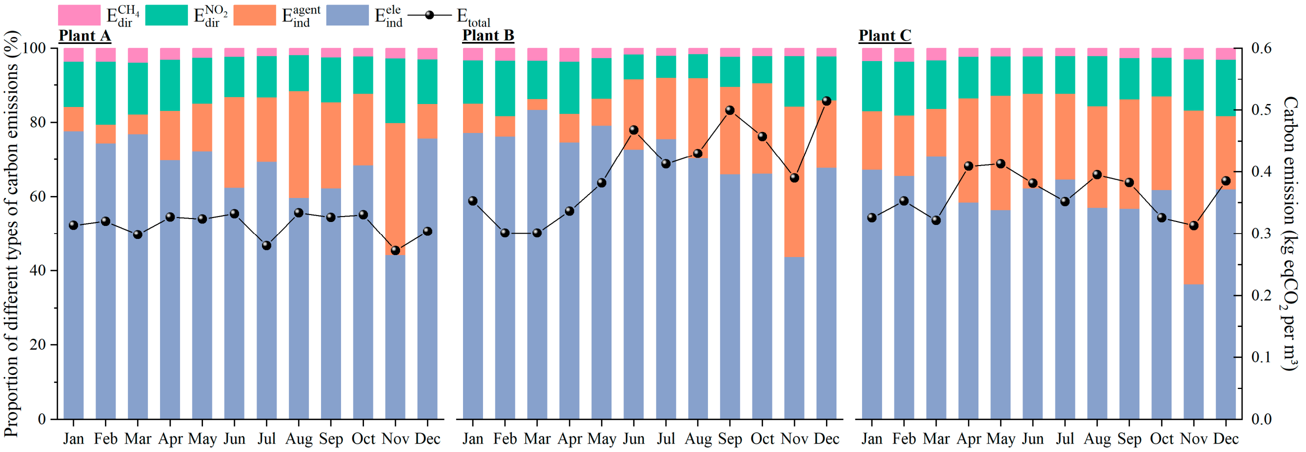

Figure 3. The variation in total carbon emission intensity and the proportion of different types of carbon emissions. Plants A–C are three full-scale wastewater treatment plants in Anhui province and the detailed information can be found in Table S1.

As Figure 3 shows, major carbon emission is caused by electricity consumption (E_{ind}^{ele}), which accounts for 44.3~77.7% in Plant A, 43.7~83.4% in Plant B and 36.4~70.7% in Plant C. Then, carbon emissions introduced by chemical agent consumption account for 4.9~35.6% in Plant A, 2.9~40.6% in Plant B and 12.9~46.9% in Plant C. It is important to note that there is an excessive variation in chemical agent consumption relative to influent water quality and organic removal efficiency, which indicates a further improvement in the management of chemical agent dosing. That is, the refined management of chemical agent dosing will be one of the priorities of carbon emission reduction in these WWTPs. Collectively, the indirect carbon emission accounts for 79.4~88.4% in Plant A, 81.7~92.1% in Plant B and 81.7~87.7% in Plant C.

While direct carbon emission accounts for 11.6~20.6% in Plant A, 7.8~18.3% in Plant B and 12.2~18.3% in Plant C, it should be noted that direct carbon emissions due to N_2O emissions, $E_{dir}^{N_2O}$, comprise a major part of direct carbon emissions. This is mainly related to the adopted treatment process, which is mainly based on the aerobic environment.

Notably, the proportion of different types of carbon emissions also shows clear differences over time. E_{ind}^{ele} comprises a relatively large portion during the winter. At lower ambient temperatures, the activity of microorganisms decreased significantly, and increasing dissolved oxygen (DO) to maintain stable performance is the most common and preferred strategy [43], which leads to more power consumption. Meanwhile, carbon emissions caused by chemicals consumption $\left(E_{ind}^{chemicals}\right)$ account for 2.9~46.9%. The high chemical cost mainly occurs in the summer and autumn seasons, which can be attributed to the low C/N and TP in influent water [44]. Significantly, Plant B exhibited far more carbon emissions (0.404 kg $eqCO_2/m^3$) in September than in other months. This is mainly attributed to the high hydraulic load (~130% design capacity). In addition, the high hydraulic loading rate is a common problem in WWTPs in China [36]. Under a higher hydraulic load, WWTPs would have little room for regulation and cannot effectively cope with changes in wastewater influent quality and capacity, eventually leading to more carbon emissions.

3.4. SEM Analysis on Basic Parameters Involved in Carbon Emissions

Regarding the wide range of parameters related to carbon emissions, SEM was used to identify the degree of correlation. SEM (Figure 4) shows the influence of the influent water quality and pollutant removal rate on carbon emissions. The non-normed fit index (NNFI) = 0.907, adjusted goodness-of-fit index (AGFI) = 0.997 and chi square/degrees of freedom = 1~2 indicate a good fit of the model to the pure data [45,46].

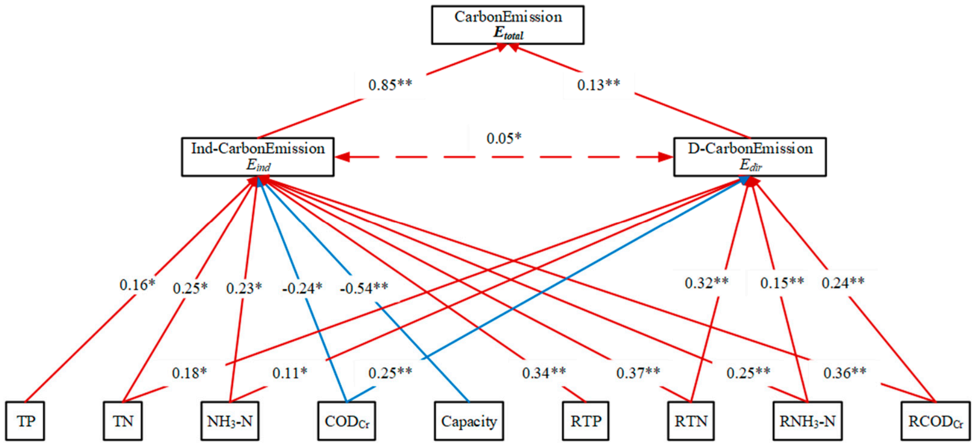

Figure 4. The path diagram of SEM analysis. Single arrows denote the causal relationship from one variable to another, and double-headed arrows denote the association between two variables. Numbers adjacent to arrows are path coefficients, which represent the strength of the relationship. Red lines indicate that the correlation is positive, and dark blue lines indicate that the correlation is negative. * represents $p < 0.05$; ** represents $p < 0.01$. The SEM model fits well (chi square = 16.615; degrees of freedom (DF) = 9; standardized root mean square residual (SRMR) = 0.013).

In line with previous studies, it can be observed that a significantly strong relationship (0.85 **) exists between indirect carbon emission (E_{ind}) and total carbon emission (E_{total}), which indicates that indirect carbon emissions account for large proportions of the total carbon emissions of WWTPs [47]. Notably, E_{dir} and E_{ind} have a significant positive interrelationship. That is, when E_{ind} is higher, E_{dir} is usually higher as well. An improper operational control strategy results in useless energy and chemical agent consumption besides more greenhouse gas emissions. For instance, excessive dissolved oxygen (DO) requires more electricity, which results in more E_{ind}. Moreover, it can inhibit the production and activity of denitrification enzymes, and nitrous oxide reductase (NOR) is more sensitive to the fluctuations of DO than other denitrification enzymes, which leads to increased nitrous oxide accumulation [48].

Collectively, influent parameters have different impacts on direct carbon emissions (E_{dir}), indirect carbon emissions (E_{ind}) and total carbon emissions (E_{total}). Higher concentrations of TN result in higher carbon emission intensity, which involves both E_{ind} and E_{dir}. In contrast, higher concentrations of COD_{Cr} during the study period are more beneficial for pollutant removal [49], which decreases E_{ind} and E_{dir}. Capacity has a significant negative effect on E_{ind} and E_{total}, which should be attributed to the scale effect [42,50]. In this study, Plant B shows a higher E_{total} with a significant fluctuation under the lowest capacity (Figure 3). Notably, TP and its removal RTP are mainly related to the chemical agent's dosage during tertiary treatments, which is an important part of E_{ind}. Meanwhile, they showed little correlation with E_{dir} in the SEM analysis. Notably, phosphorus removal is linked to nitrogen removal (Figure 2). Murnleitner et al. pointed out that there is competition between phosphate-accumulating organisms (PAOs) and denitrifying bacteria for carbon sources [39], which will further impact direct carbon emissions. All concerned influent parameters and pollutant removal efficiency have a direct positive contribution to carbon emissions during wastewater treatment processes, with the exception of capacity. Influent parameters show different influences on direct and indirect carbon emissions. Among these, capacity is significantly negatively associated with E_{ind} (−0.54 **) and has no significant correlation with E_{dir}. This indicates that capacity is a key factor affecting the carbon emissions of WWTPs due to the scale effect. Conclusively, RTN and $RCOD_{Cr}$

are singled out as the significant positive influential factors for E_{dir} and E_{ind}, while COD_{Cr} shows a negative influence on E_{dir} and E_{ind}. The positive contribution to E_{dir} followed the sequence of RTN > $RCOD_{Cr}$ > TN > RNH_3-N > NH_3-N, while the positive contribution to E_{ind} followed the sequence of RTN > $RCOD_{Cr}$ > RTP > TN > RNH_3-N > NH_3-N > TP.

4. Discussion

Generally, the average influent concentrations of the COD_{Cr}, NH_3-N, TN and TP of the studied WWTPs in 2021 were 124.13, 22.07, 29.69 and 3.40 mg/L, respectively. In this study, the average removal rates of COD_{Cr}, NH_3-N, TN and TP were 88.7%, 97.2%, 71.9% and 93.0%, respectively, which showed good performance for pollutant removal. Additionally, the average total carbon emission intensity of the AAO oxidation ditch was about 0.314~0.404 kg $eqCO_2/m^3$, which is slightly above the total carbon emission intensity of the oxidation ditch (0.318 kg $eqCO_2/m^3$) in the previous study [14]. This is mainly due to the seriously insufficient carbon source in wastewater influents with a C/N ratio of around 4.2, which is lower than the minimum value in the study by Sun et al. [37]. The insufficient carbon source is crucial to BNR processes as COD_{Cr} acts as a limiting factor for phosphorus release and denitrification. This leads to additional carbon sources for nitrogen and phosphorus removal, which introduces higher carbon emissions caused by chemical agent costs (0.04 kg $eqCO_2/m^3$).

Although there have been many studies focused on carbon neutrality and energy self-sufficiency in WWTPs [51], most WWTPs still require considerable energy and chemical agents to remove pollutants. Among them, RTN shows the highest positive correlation with E_{dir} and E_{ind}. Nitrogen removal has been known as an important challenge for WWTPs' performance [52]. Moreover, there is difficulty in coping with nitrogen removal during the WWTPs' carbon emission reduction. The removal of nitrogen in an efficient and low-carbon manner is an urgent problem that needs to be overcome. Additionally, phosphorus removal has also been another key challenge for WWTPs. In our study, RTP is mainly positively related to E_{ind} but shows little correlation with E_{dir}.

The SEM results indicated that there was a significant correlation between pollutant removal and carbon emission. Guo et al. pointed out that there should be a trade-off between carbon emission and the pollutant removal of activated sludge processes, which needs further research [53]. In particular, the removal of TN has the greatest impact on carbon emissions (Figure 4), and the adoption of low carbon emission nitrogen removal processes is also one of the important ways to realize carbon emission reductions in the wastewater industry [54]. Notably, the capacity also shows a significant scale effect on the carbon emission per ton of water, which was also proved in a previous study on 217 wastewater treatment plants in the Valencia region [42]. In general, the AAO oxidation ditch has a high application potential, but its application conditions still need to be further studied according to influent conditions and pollutant removal requirements.

5. Conclusions

The carbon emissions of WWTPs are rather complicated due to the variety of GHGs, their generation and the wide range of influencing parameters. The main objective of this study is to identify key basic parameters involved in full-scale WWTPs, such as capacity, influent water quality and pollutant removal efficiency. This study is inspiring in a certain sense with respect to forming clear judgments and thus creating appropriate designs, operations and the management of carbon emission reductions in wastewater treatment processes. The primary conclusions have been summarized as follows:

1. In 2021, the average removal rates of COD_{Cr}, NH_3-N, TN and TP were 86.1%, 97.4%, 71.0% and 91.5% in Plant A; 89.6%, 96.1%, 75.8% and 95.1% in Plant B; and 90.3%, 98.1%, 69.0% and 92.5% in Plant C. These showed good performances for the AAO oxidation ditch with respect to pollutant removal.
2. Carbon emissions during wastewater treatment processes mainly consist of indirect carbon emissions (~90%). The SEM results show that influent COD_{Cr} and TN and

their removal should be key indicators related to carbon emissions. For domestic sewage, a higher influent organic matter concentration helps reduce energy and agent consumption and greenhouse gas emissions.

3. The SEM results indicated that the positive contribution to E_{ind} followed the sequence of RTN > $RCOD_{Cr}$ > RTP > RNH_3-N > TN > NH_3-N > TP. Notably, capacity showed a significant negative contribution to E_{ind}. Additionally, capacity showed the highest negative correlation with E_{ind}, followed by COD_{Cr}, while the contribution to E_{dir} followed the sequence of RTN > $RCOD_{Cr}$ > TN > RNH_3-N > NH_3-N. Notably, COD_{Cr} showed a significantly negative correlation with E_{dir}.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/su15097225/s1, Table S1. Images of the WWTPs (from Tiandi Maps) and general information. Figure S1. The water quality parameters variation of influent water: (a) CODCr, (b) TN, (c) NH3-N, (d) TP and (e) capacity. Figure S2. Removal rate of major pollutants: (a) CODCr, (b) TN, (c) NH3-N and (d) TP.

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