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Discussion

Mitigating greenhouse gas emissions from municipal wastewater treatment in China



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ABSTRACT

Municipal wastewater treatment plays an indispensable role in enhancing water quality by eliminating contaminants. While the process is vital, its environmental footprint, especially in terms of greenhouse gas (GHG) emissions, remains underexplored. Here we offer a comprehensive assessment of GHG emissions from wastewater treatment plants (WWTPs) across China. Our analyses reveal an estimated 1.54 (0.92–2.65) \times 10 4 Gg release of GHGs (CO₂-eq) in 2020, with a dominant contribution from N₂O emissions and electricity consumption. We can foresee a 60–65% reduction potential in GHG emissions with promising advancements in wastewater treatment, such as cutting-edge biological techniques, intelligent wastewater strategies, and a shift towards renewable energy sources.

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1. Introduction

The urban water sector has been identified as a significant contributor to greenhouse gas (GHG) emissions. Recent updates in emission inventories underscore the pivotal role of GHG emissions stemming from urban water supplies and wastewater treatments in the global carbon budgets [1–6]. It is estimated that the degradation of organics in wastewater treatments contributed ~0.77 Gt CO₂-equivalent (CO₂-eq) GHG emissions in 2010, equivalent to 1.57% of the global GHG emission (i.e., 49 Gt CO₂-eq) [7]. In particular, non-CO₂ GHG emissions (i.e., N₂O and CH₄) in wastewater treatments and discharge doubled from 1970 to 2015, occupying ~9.6% of global non-CO₂ GHG emissions in 2015 [8].

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Rapid urbanization and increasing levels of affluence have led to a large increase in GHG emissions from urban water sectors. As China aims to reach its carbon peak by 2030 and achieve carbon neutrality by 2060, it is crucial to examine the contribution of urban water sectors to the national GHG emissions budget.

Urban wastewater systems are a complex source of GHG emissions, characterized by intricate interactions between water and energy [9-11]. Recognizing the urban water sector's role in GHG emissions is imperative for addressing the challenges posed by global warming [6,12]. GHG emissions from wastewater treatments are primarily derived from direct productions (e.g., N₂O and CH₄) and indirect CO₂ emissions from energy and chemical consumptions [13]. While microbial processes are commonly used to reduce nutrients and organic matter in wastewater treatments, they can also produce GHGs such as CH₄ and N₂O [14]. On a global scale, wastewater treatment is estimated to contribute around 2% to the total anthropogenic GHG emissions [2,15]. It is estimated that between 2005 and 2030, annual non-CO2 GHG emissions from wastewater treatment processes could range from 0.56 to 0.71 Gt of CO₂-eq per year [16]. Historically, direct CO₂ emissions from degradations of wastewater organics were considered a carbon-neutral

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process in GHG accounting [17,18]. On the other hand, non- CO_2 GHG emissions are increasingly recognized as significant due to their much higher global warming potentials than CO_2 . They are 25–298 times stronger (in a 100-year time horizon) in global warming potentials than CO_2 [2,19].

China has undergone one of the largest and most rapid urbanization processes in human history, with over 60% of the population, approximately 900 million people, living in urban regions in 2020 compared to less than 20% in the 1970s [20,21]. To improve water quality and sanitation, there has been significant investment in the construction of wastewater treatment plants (WWTPs) in many countries. In the specific case of China, the percentage of treated municipal wastewater increased from approximately 40% in 2005 to over 90% in 2017 [20,22]. Nowadays, China has the world's largest municipal wastewater treatment capacity (~60 billion m³ year⁻¹) and continues to make large investments in expanding wastewater treatment facilities. As such, the urban wastewater sector has the potential to make substantial contributions to GHG emissions [23-27]. Our previous studies have demonstrated that WWTP constructions have effectively reduced nutrient inputs into waters and improved water quality [22,28,29]. For instance, we find that improved treatments of both rural and urban domestic wastewater have resulted in large-scale declines in lake phosphorus (P) concentrations in the most populated area of China [28]. In some regions, due to the higher removal efficiency of P compared with nitrogen (N), the N/P mass ratio in the total municipal wastewater discharges has continued to increase from the median of 10.7 in 2008 to 17.7 in 2017 [22]. Thus, WWTP constructions have been successful in improving water quality. However, GHG emissions associated with the operation of these plants have received limited attention. Earlier studies have explored GHG emissions stemming from wastewater treatment processes. For example, Yan et al. applied an emission factor method to determine the spatial and temporal distribution characteristics of GHG emissions from municipal WWTPs from 2005 to 2014. Their study included CH₄ and N₂O emissions, disregarding the potentially substantial contributions of indirect CO₂ emissions [24]. Huang et al. established plant-level monthly operational emissions inventories of China's WWTPs from 2009 to 2019. They showed that urban wastewater treatment has been enhanced, with 80% more chemical oxygen demand (COD) removed annually. They concluded that the enhanced urban wastewater treatment increases GHG emissions and regional inequality [30]. Hua et al. developed a framework to obtain multi-level GHG emission factors of WWTPs. They found that GHG emission factors of different technologies may range widely from 180.0 to 615.7 g CO₂-eq per ton wastewater [23].

In this study, we focus on quantifying GHG emissions in wastewater treatment processes in China. To achieve this goal, we reviewed the development of WWTPs in China from 2000 to 2020. A systematic investigation was conducted on pollutant concentrations, such as total nitrogen (TN), ammonia nitrogen (NH₃-N), and chemical oxygen demand, in influents and effluents of WWTPs as well as on electricity consumptions, wastewater treatment volume, treatment technologies, and location. These efforts aimed to improve the accuracy of GHG emission estimates associated with WWTP operations. Based on the technique-specific GHG emission factors for the WWTPs [23], direct and indirect GHG emissions have been estimated for WWTPs for 31 provinces of China. Subsequently, we analyzed feasible strategies to mitigate GHG emissions stemming from WWTPs, considering their practical applicability. Improving nutrient removal efficiency in wastewater treatment processes while reducing GHG emissions is an important goal for developing wastewater treatment technologies in China and other nations. This target should be emphasized as a crucial part of achieving carbon neutrality.

2. Methods and materials

2.1. Data sources

The investigation encompassed 3444 centralized WWTPs distributed across 31 provinces in China, drawing data from China's Environmental Statistics Databases, China's Urban Construction Yearbooks, and previously researched by the author [22,31,32]. These investigated WWTPs represented approximately 70% of the total WWTPs in China and about 55% of the total volumes of wastewater treated in the country in 2020. The investigation of each WWTP included the following information: (1) geographic locations (latitudes and longitudes); (2) the concentrations for TN, NH₃-N, and COD in the influent and effluent; (3) the volumes of treated wastewater; (4) energy usages; and (5) employed treatment techniques. Wastewater treatment technologies applied in the investigated WWTPs were classified into eleven types, including anaerobic-anoxic-oxic (A/A/O), anaerobic-oxic (A/O), improved activated sludge (IAS), conventional activated sludge (CAS), BIOLAK, oxidation ditch (OD), sequencing batch reactor (SBR), membrane bioreactor (MBR), biological filter (BF), ecological treatment (ET) and others. The average yearly wastewater treatfor investigated **WWTPs** ment volume all $(1183.7 \pm 1973.6) \times 10^4 \, \text{m}^3$ (Fig. S3). Among them, 27% had applied A/A/O, and 31% had applied OD. The estimated operating cost for treating 1 m³ of wastewater by each different technique is provided in Fig. S1.

2.2. Estimation of GHG emissions from WWTPs

GHG emissions in wastewater treatments were classified into direct and indirect emissions [13]. Direct GHG emissions from the treatment process included CH₄ and N₂O emissions from biological nutrient removal. However, direct CO₂ emissions were typically regarded as 100% biogenic and not included in the GHG emission inventory for wastewater treatments [17,23]. Indirect GHG emissions emphasized in this study were the part incurred from energy usage, while the contributions from chemical consumption were not considered due to data scarcity. GHG contributions from chemical consumption could account for between 5% and 15% of the total GHG emissions in wastewater treatments [33]. A threestep procedure was performed to estimate GHG emissions from WWTPs. First, a comprehensive examination encompassed parameters such as treated wastewater volume, treatment techniques, nutrient concentrations in the influents and effluents of WWTPs, and energy usage. Subsequently, estimates were derived for the masses of organic material and nutrient removal. Secondly, a literature review was conducted to source GHG emission factors for different treatment techniques. This study adopted GHG emission factors by different treatment technologies from a previous study (Table S1) [23]. CH₄, N₂O, and indirect CO₂ emissions by treating 1 m³ were provided in Table S1, and these GHG emissions could range among different treatment technologies. The median and 95% confidence intervals (95% CI) were provided in Table S1 and were applied to estimate the ranges of GHG emissions in each WWTP. Thirdly, it is observed that the number of WWTPs included in some provinces in this assessment was lower than the count reported in the national statistics [32]. In order to correct this bias, this study applied a correction factor to re-adjust the estimated GHG emissions in the WWTPs when the volume of treated wastewater derived from the investigation was less than 90% of the corresponding national statistics. CH₄ and N₂O emissions from the WWTPs were estimated as follows:

$$GHG_{\rm D} = \sum V_{\rm T} \times EF_{\rm CH_4/N_2O/CO_2} \times 10^{-5} \tag{1}$$

where GHG_D represents direct GHG emissions in the form of CH₄ and N₂O and the indirect CO₂ emission (Gg); V_T refers to the masses of wastewater treated by each WWTP (Gg); EF represents the GHG emissions (i.e., direct CH₄ and N₂O emissions and indirect CO₂ emission) by treating one-ton wastewater. EF varies depending on applied treatment technologies (see details in Table S1) [23]. Nutrient removal by WWTPs in each province could be estimated as follows:

$$N_{\rm R} = \sum (C_{\rm I} - C_{\rm E}) \times V_{\rm E} \times 10^{-5}$$
 (2)

where N_R represents pollutant removal by WWTP in each province (Gg); C_E represents the pollutant concentrations (i.e., COD and TN) in the effluents of WWTPs (mg L⁻¹); C_I refers to the pollutant concentration in the influents of WWTPs (mg L⁻¹). This information was derived from the investigation described in section 2.1. V_E represents the volume of treated wastewater (10⁴ m³) for each WWTP. Indirect GHG emissions from WWTPs were estimated based on electricity consumption in wastewater treatments as follows:

$$GHG_{\rm I} = \sum V_{\rm E} \times EF_{\rm El} \times 10^{-5} \tag{3}$$

where GHG_I represents indirect GHG emissions by electricity consumption (Gg); V_W refers to the volume of treated wastewater (10^4 m³); EF_{EI} represents indirect GHG emissions by electricity consumption in treating one ton of wastewater. For some provinces, the number of WWTPs included in the investigation was lower than the number of WWTPs in the national statistics. A correction factor for the province was calculated as follows:

$$C_{\rm R} = \frac{V_{\rm N}}{V_{\rm F}} \tag{4}$$

where V_N refers to the volume of treated wastewater in the province from the national statistical data (10⁴ m³); V_E represents the volume of treated wastewater estimated in the investigation (10⁴ m³).

2.3. Statistical analysis

This study analyzed the statistical differences in nutrient removal efficiencies by different treatment technologies or provinces using a one-way ANOVA. Post hoc multiple comparisons were conducted according to Tukey's least significant difference procedure. Statistical analyses were conducted using the SPSS 23.0 statistical package for personal computers (IBM, USA), with a level of significance P < 0.05 for all tests.

3. Results and discussion

3.1. Estimating GHG emissions from WWTPs in China

Analyses of national, regional, and local census data revealed a substantial increase in municipal wastewater discharge in China from 33 billion m³ in 2000 to 57 billion m³ in 2020 (Text S1). This increase coincided with rapid developments of WWTPs in the country, with the number of WWTPs increasing from only 506 in 2000 to 4326 in 2020 and the percentage of treated municipal wastewater increasing from 36% in 2000 to 95% in 2020 in urban areas (Fig. 1). In addition, the national average concentration of total nitrogen and ammonia-nitrogen in the influents were

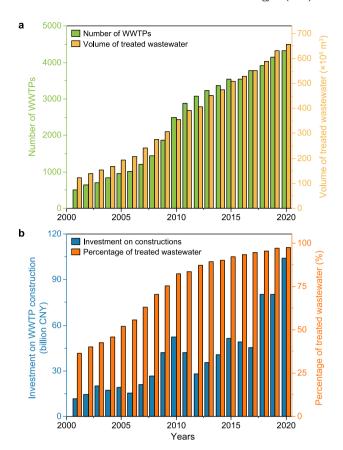


Fig. 1. a, Changes in WWTPs and volumes of treated wastewater during 2000–2020. **b**, Changes in investment in WWTPs' construction and percentage of wastewater being treated (%) during 2000–2020.

 34.3 ± 18.2 and 26.1 ± 15.6 mg L⁻¹, respectively, and their corresponding effluents showed a large reduction in concentration to 9.8 ± 5.3 and 2.3 ± 4.3 mg L⁻¹, respectively (see more details in Text S2).

GHG emissions from the WWTPs in China were estimated based on treated wastewater volumes and GHG emission factors by different treatment techniques. Nationally, around 1.54 $(0.92-2.65) \times 10^4$ Gg of GHGs (CO₂-eq, including both direct and indirect CO2 emissions) had been conservatively estimated to be released from wastewater treatment in China (Table 1). The majority of emissions come from N2O and indirect contributions from electricity consumption, which accounts for around 90% of the total (N₂O: 4917 [1254-12808] Gg CO₂-eq; CH₄: 1166 [485-2504] Gg CO₂-eq, and indirect emission: 9268 [7470-11157 on a national scale]). Due to large variations in the volume of the wastewater being treated, large differences in GHG emissions were observed among different provinces (Table 1). The largest total GHG emission from WWTPs was observed in Guangdong province with a value of 2219.6 (1313.6-3781.7) Gg CO₂-eq, and the lowest GHG emission was observed in Xizang Zizhiqu with a value of only 31.2 (15.5-58.4) Gg. Notely, the estimation in this study was primarily based on the volume of wastewater being treated, emission factors from different techniques, and applied treatment techniques, while other factors like local climates and WWTP management levels had not been considered [12]. These differences could also result in variations in estimated GHG emissions from the wastewater treatment.

Table 1GHG emissions from WWTPs in 31 provinces of China. Taiwan, Hong Kong, and Macao are not displayed due to the lack of data.

Province	N ₂ O emission (Gg CO ₂ -eq)	CH ₄ emission (Gg CO ₂ -eq)	Indirect emission (Gg CO ₂ -eq)	Total emission (Gg CO ₂ -eq)	Per capita GHG emission (kg CO ₂ -eq)
Anhui	155.6 (35.7-401.3)	61.0 (29.1–98.3)	331.4 (267.1-339.0)	548.0 (331.9-898.6)	15.4 (9.3–25.2)
Beijing	147.5 (48.6-490.6)	30.4 (13.6-60.3)	451.8 (364.1-543.8)	629.7 (426.2-1094.7)	32.8 (22.2-57.1)
Fujian	121.4 (30.9-318.8)	37.0 (17.6-69.8)	231.5 (186.6-278.7)	389.9 (235.2-667.4)	13.6 (8.2–23.3)
Gansu	49.0 (13.3-121.7)	10.1 (4.9-22.5)	84.5 (68.1-101.7)	143.6 (86.3-245.9)	11.0 (8.2–23.3)
Guangdong	723.4 (175.4–1827.8)	175.0 (73.2-363.4)	1321.2 (1064.9-1590.5)	2219.6 (1313.6-3781.7)	23.7 (14.0-40.4)
Guangxi	153.6 (42.2-391.8)	26.0 (11.4-72.5)	229.8 (185.2-276.6)	409.4 (238.7-740.9)	15.0 (8.7–27.2)
Guizhou	94.3 (26.0-239.6)	21.5 (9.3-49.3)	159.9 (128.8-192.4)	275.6 (164.2-481.3)	13.4 (8.0-23.5)
Hainan	33.4 (10.9-80.3)	7.3 (2.4–17.3)	70.2 (56.5-84.5)	110.8 (69.9-182.1)	18.2 (11.5-29.9)
Hebei	153.9 (42.8-413.7)	36.5 (15.9-78.1)	318.9 (257.0-383.9)	509.3 (315.7-875.7)	11.4 (7.0-19.5)
Henan	147.3 (28.9-384.4)	52.4 (21.5-90.4)	292.2 (235.6-351.8)	491.9 (286.0-826.7)	8.9 (5.2-15.0)
Heilongjiang	134.6 (35.5-336.9)	21.2 (6.2-64.6)	188.4 (151.8-226.8)	344.2 (193.5-628.2)	16.5 (9.3-30.2)
Hubei	213.0 (48.0-545.0)	52.2 (20.2-64.6)	402.6 (324.5-226.8)	667.9 (392.7-628.2)	18.5 (10.9-31.5)
Hunan	191.6 (49.1-496.7)	57.1 (27.1-105.4)	395.5 (318.8-476.1)	644.2 (395.1-1078.3)	16.5 (10.1–27.61)
Jilin	147.5 (29.5-355.1)	23.9 (6.0-67.6)	173.1 (139.5-208.4)	344.6 (175.0-631.1)	22.9 (11.6-42.0)
Jiangsu	393.7 (116.1-1030.1)	82.6 (41.1-187.6)	778.2 (627.2–936.8)	1254.5 (784.4-2154.4)	20.2 (12.6-34.6)
Jiangxi	78.8 (16.9-210.7)	31.3 (14.0-50.5)	176.0 (141.9-211.9)	286.1 (172.8-473.1)	10.5 (6.32-17.3)
Liaoning	311.5 (83-784.4)	53.8 (15.3-149.2)	517.2 (416.8-622.5)	882.5 (515.1-1556.1)	28.8 (16.8-50.7)
Nei Mongol	58.7 (13.8-156.3)	11.8 (3.9-28.8)	104.3 (84.0-125.5)	174.7 (101.7-310.6)	10.8 (6.3-19.2)
Ningxia	21.4 (4.6-54.8)	7.7 (3.5-13.0)	43.0 (34.7-51.8)	72.2 (42.8-119.6)	15.4 (9.1-25.2)
Qinghai	12.8 (3.1-33.9)	3.7 (1.6-6.2)	31.2 (25.1-37.50	47.7 (29.8-77.6)	13.4 (8.4–21.8)
Shandong	257.6 (62.2-701.0)	65.7 (26.0-132.8)	558.9 (450.4-672.8)	882.2 (538.6-1506.5)	13.8 (8.4–23.5)
Shanxi	74.6 (21.7-206.4)	23.6 (11.4-43.3)	176.1 (141.9-212.0)	274.3 (175.1-461.7)	12.6 (8.0-21.2)
Shaanxi	135.5 (37.8-335.2)	28.3 (14.0-64.8)	224.5 (180.9-270.2)	388.3 (232.8-670.2)	15.7 (9.4-27.4)
Shanghai	152.7 (50.7-483.5)	43.1 (24.0-90.8)	410.1 (330.5-493.6)	605.9 (401.5-1068.0)	27.3 (18.1-48.1)
Sichuan	244.1 (62.5-596.8)	50.1 (20.0-119.5)	389.3 (313.7-468.6)	683.5 (396.3-1184.9)	14.4 (8.3-25.0)
Tianjin	91.2 (19.3-258.9)	20.5 (4.6-51.0)	162.7 (131.2-195.9)	274.4 (155.0-505.8)	23.4 (13.2-43.1)
Xizang	15.8 (3.8-35.5)	1.8 (0.7-6.5)	13.6 (11.0-16.4)	31.2 (15.5-58.4)	23.9 (11.8-44.7)
Xinjiang	70.2 (12.8-167.4)	13.1 (2.9-29.9)	110.6 (89.2-133.2)	193.9 (104.9-330.5)	13.2 (7.2–22.6)
Yunnan	122.6 (36.9-306.3)	20.6 (11.0-53.6)	201.2 (162.1-242.2)	344.4 (210.0-602.1)	14.6 (8.9-25.5)
Zhejiang	297.2 (74.3-769.1)	64.4 (24.9-148.0)	533.3 (429.9-224.8)	894.9 (529.0-1559.1)	19.2 (11.3-33.4)
Chongqing	112.2 (17.3-274.3)	32.4 (10.6-60.5)	186.7 (150.5-224.8)	331.3 (178.5-559.6)	14.9 (8.0-25.1)
National	4916.5 (1253.6-12808.2)	1166.3 (484.5-2503.9)	9267.9 (7469.7-11156.7)	15350.7 (9207.8–26468.8)	17.1 (10.2–29.4)

3.2. Direct and indirect contributions of GHG emissions in WWTPs

Improving sanitation access and wastewater treatments benefits public health and environmental protections [28]. However, the urban water sector is a high-energy consumption sector [6,12,23], with the contribution of the urban water sector to GHG emission in the USA ranging from 1% to 3% [34]. In China, the estimated total GHG emissions from WWTPs were 15.4 (9.2-26.5) Tg CO₂-eq (Table 1), and the percentage in the national GHG emission (up to $\sim 1.4 \times 10^4$ Tg The USA) is lower than in the other high-income countries probably due to the substantial contribution by energyintensive industry [35]. China's WWTPs were estimated to account for about 0.2% of the 15 billion kWh used in China every year [36]. Most electricity consumed in conventional wastewater treatments is used in pumping water and aeration. Total electricity consumption for advanced treatment technologies varies, surpassing the energy demands of secondary treatment processes [23]. The average electricity usage in WWTPs applying different treatment technologies is provided in Fig. S2. For most treatment techniques, 0.3 kWh of electricity will be consumed to treat 1 m³ of wastewater, while MBR requires around 0.5 kWh of electricity. Additionally, the primary and secondary stages of sewage treatment are designed to perform various functions such as thickening, conditioning, and dewatering of sewage sludge. Among these processes, dewatering usually incurs the highest energy consumption [37]. It's worth noting that the total annual electricity consumption of individual WWTPs in China has significantly decreased from 3.1 million kWh in 2006 to 2.0 million kWh in 2015, owing to the development and applications of new low-energy techniques, such as biological filter and membrane biological fluidized bed [38,39].

3.3. Strategies to reduce GHG emissions from WWTPs

WWTPs are increasingly recognized as an important source of GHG emissions. Following this recognition, several potential strategies have been explored to reduce the GHG emissions from WWTPs. Our estimations indicate that most GHG emissions from WWTPs (Table 1), are attributed to N₂O and indirect CO₂ emissions, aligning with prior research findings [40,41]. Therefore, the reductions of direct N₂O emissions and indirect off-site CO₂ emissions from electricity usage are needed for GHG emission reduction from WWTPs. Accordingly, technology optimization to reduce GHG emissions and energy recovery to offset electricity usage are two dominant mitigation strategies at the WWTPs. In this section, we review and identify mitigation strategies that have the potential to contribute to GHG emission reduction in China's WWTPs (Fig. 2).

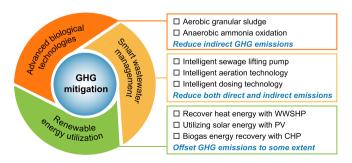


Fig. 2. Proposed strategies for the GHG mitigation from the WWTPs. WWSHP: Wastewater source heat pump; PV: Photovoltaic power generation; CHP: Combined heat and power technology.

3.3.1. Advanced biological technologies to reduce indirect GHG emissions

Despite large GHG emissions in current wastewater treatment processes, emerging evidence shows that, if properly tailored, biological techniques can provide low-cost and environmentally friendly measures to reduce GHG emissions from WWTPs. Aerobic granular sludge (AGS) has become a competitive alternative for biological N removal (BNR) in WWTPs [42]. Compared to CAS. which has been widely adopted in China, AGS has 2-3 times more microbial enrichment, together with a higher biochemical reaction efficiency, better effluent quality, requiring fewer land areas, lower chemical dosage, and energy usages [43,44]. China's largest AGS wastewater treatment project (80000 m³ day⁻¹) could save up to 1.46 million kWh year⁻¹ in electricity consumption and reduce carbon emissions by 882 tons year⁻¹ compared to the CAS process [44]. A WWTP in Henan province, China, adopted AGS technology to upgrade the scale and expand the treatment capacity by more than three times, following which energy consumption and chemical dosages were reduced by more than 20% [43]. In AGS, there are both aerobic and anoxic environments where the anoxic denitrification possibly serves as a sink to reduce N₂O production. However, a wide range of N2O emission factors (0.33-22%) had been reported for AGS under different operation conditions, introducing uncertainty regarding whether AGS yields lower N2O emission compared to CAS [45].

Anaerobic ammonia oxidation (Anammox) is an autotrophic BNR process in which Anammox microorganisms apply the nitrite as the electron acceptors and oxidize ammonium to nitrogen under anaerobic conditions [46]. Anammox-based process is a more costeffective and sustainable method to remove reactive N from wastewater [47,48]. Compared to the CAS process, carbon emissions from the whole Anammox process can be reduced by more than 50% due to lower oxygen consumption (reduced by 63%) and external carbon source (reduced by 100%), as well as theoretically having zero N_2O emission [49,50]. This will greatly support the "energy-neutral" and even "energy-positive" urban wastewater treatment goal in the future [43]. The construction of Beijing's Anammox denitrification project, which boasts the world's largest sludge digestion capacity at 15900 m³ day⁻¹, anticipates an annual reduction in carbon emissions estimated at 10500 ton CO₂-eq per year [43]. Nevertheless, due to the nitrite demand and oxygenlimited conditions, Anammox is usually applied for BNR in the presence of nitrifiers and heterotrophic denitrifiers, which would produce N₂O during the BNR process [51,52]. The partial nitritation/ anammox system did have a higher N2O emission factor than CAS in some studies [53]. Inspiringly, it is still possible to significantly reduce N₂O emission when utilizing anammox technology for low N₂O generating alternatives for nitrifiers (e.g., ammonia-oxidizing archaea). In summary, the advanced biological technologies (i.e., AGS and Anammox) proposed in this study mainly contribute to the mitigation of indirect GHG emissions. Considering the remaining challenges of full-scale application in mainstream wastewater treatment processes, approximately 7% and 16% of indirect GHG emissions at WWTPs in China are expected to be reduced by AGS and Anammox technologies, respectively (Table S4).

3.3.2. Smart wastewater management to reduce the GHG emissions

Indirect GHG emissions from wastewater treatment processes come mainly from energy and chemical consumption. Energy consumption that accounts for about 73.4% of total indirect GHG emissions is mostly used in the sewage-lifting pumps and aeration systems [54], while indirect GHG emissions from the chemical consumption could account for 4.8% [55]. Improving the management of these contributing factors through smart systems could help to reduce overall contributions to GHG emissions. Upgrading

the sewage-lifting pumps or adopting an intelligent operational mode that optimizes energy consumption could effectively reduce energy consumption. Specifically, the use of frequency conversion technology to transform old sewage lifting pumps and the direct application of new equipment or facilities (e.g., digital pumps) are two major pathways to reduce the energy consumption needed for the sewage lifting pumps [56]. Optimizing the operations and improving energy efficiency can also contribute to reductions in energy consumption. The evolution of the Internet of Things and mobile Internet technology can be harnessed to create smart systems and intelligent management platforms to support remote control, centralized management, and digital operation of sewage lifting pumps to increase energy savings and reduce overall energy consumption [57].

Improving oxygen management throughout the wastewater treatment process can also help reduce GHG emissions. During the aerobic stage of wastewater treatment, oxygen supply is an important determinant. Insufficient oxygen levels result in reduced microbial growth rates and low reproduction rates of microorganisms, thereby leading to poor wastewater treatment performance. However, excessive oxygen supply wastes energy and increases material consumption. The key to minimizing GHG emissions is balancing biological oxygen consumption and supply. An intelligent system that controls the aeration quantity in biochemical pools with feedback controls and intelligent models could be deployed to maintain a balance of aeration [58]. Real-time optimization and control of operational parameters can be performed by online instruments to control biological processes in the reaction tanks and improve overall biological treatment efficiency. It is estimated that intelligent aeration technology can save energy consumption by 20-40% [58]. In addition to reducing indirect GHG emissions, intelligent aeration technology is important in N2O mitigations. A significant reduction of N₂O emission by 35-90% was obtained in lab-scale and full-scale BNR systems by applying various aeration control strategies, such as reducing the DO set point, reducing the aeration rate, and changing the aeration scheme [59].

Due to low C/N ratios, especially in south China, and high requirements for nitrogen and phosphorus removal, many WWTPs rely on substantial external carbon sources and chemical phosphorus removal agents to maintain nitrogen and phosphorus removals, which is generally overdosage. Overdosing chemicals inevitably increases operation costs, energy consumption, and carbon emissions. Therefore, intelligent dosing technology is of great significance for GHG mitigation. For instance, with implementations of intelligent dosing systems assisted by biological modeling technology, carbon source addition at Changzhi WWTP was reduced by more than 50%. The amount of carbon source dosing and phosphorus removal agent dosing at the Linyi Qinglonghe Water Purification Plant decreased by 13% and 27%, respectively [43]. More importantly, over 60% of N₂O productions were determined to be reduced by the controlled supplies of chemicals in some laboratory studies [59,60]. According to the international experience and successful domestic cases, it was estimated that adopting intelligent management technologies can achieve a GHG emission reduction potential of 4% in the sewage lifting system, 18% in the aeration system, and 13% in the chemical feeding system, respectively [25]. In conclusion, enhancing the overall efficiency of sewage treatment via intelligent wastewater management can result in a 60% reduction in direct GHG emissions, with a 25% potential reduction in indirect GHG emissions (Table S4).

3.3.3. *Renewable energy utilization to offset GHG emissions*Low-carbon operational strategies for wastewater treatments

are also largely dependent on energy sources. The developments and utilization of renewable energy sources, such as heat, solar, and biogas energy, throughout the wastewater process, play important roles in realizing the low-carbon operation of WWTPs [61–63]. One major way to use heat energy at WWTPs is to extract waste heat energy using wastewater source heat pump (WWSHP) technology. Numerous WWSHP systems have been constructed and implemented in China (Table S3). However, heat energy generated through wastewater treatments is relatively low-graded, and its reuse is limited by geographical distance, as effective heat transfer is restricted to only 3–5 km [64]. A case study indicated that the use of WWSHP for winter heating in the surrounding regions of a WWTP in Shenyang province led to significant reductions in coal consumption, as well as substantial decreases of 71000 tons in SO₂ emissions, 727533 tons in soot emissions, and 140000 tons in CO₂ emissions [65]. CO₂ has the largest reduction, measuring 111464 and 287846 kg d⁻¹ under refrigeration and heating, respectively. WWSHP has also been actively used in 17 reclaimed water plants in Beijing [66]. From 2016 to 2020, these reclaimed water plants accumulated a heating capacity of up to 5.3 million GJ, saving 160 million m³ of natural gases. The applications of WWSHP technology could help WWTPs offset their CO2 emissions through energy conservation or emission reduction within a certain period, realizing a proportion of carbon-neutral operation up to 487.63% [67].

Photovoltaic power generation (PV), which harnesses the photo-generating voltage effects at semiconductor interfaces to convert solar energy into electricity, can potentially reduce GHG emissions from WWTPs [68]. However, due to the characteristics of WWTPs, including the long spans of WWTP pools, many underground pipelines, frequent obstacles, difficult construction, and some other factors, the "sewage plant + PV" projects have not been implemented widely [66]. This notwithstanding, several attempts have been made to install PV equipment above the reaction pools, such as primary sedimentation tanks, aeration tanks, and clarifiers, to increase contributions to clean power generation and effective thermal insulation [66]. By the end of the 13th Five-Year Plan, several PV projects have been constructed in Xiaohongmen, Qinghe, and Jiuxianqiao WWTPs, with a total installed capacity of 18.7 MW. The total installed capacity is expected to be increased by another 17 MW by 2025, with an annual power generation reaching 18 million kWh, contributing to a reduction in CO₂ emission by up to 11000 tons per year [43]. Solar energy could be utilized to improve the energy self-sufficiency of WWTPs. Bailong WWTP in Shanghai plans to implement a PV system and is expected to replace 25% of the current total electricity consumption with renewable energies [67].

Sludge is the main by-product of sewage treatment. Recently, anaerobic digestion has been widely employed in sludge treatments for its ability to convert organic matter to methane gas, which is then captured as a valuable product [69,70]. Methane produced during the sludge digestion accounts for about 60% of biogas [71]. Coupling with the combined heat and power (CHP) technology, biogas can be utilized in biogas engines to generate renewable power in electricity and heat, powering the surrounding equipment or being exported to the national grids [66]. Such combined technology has been applied in many projects in China with superior carbon reduction capacity. For example, Beijing Drainage Group's five sludge treatment centers are expected to replace about 18%–20% of the total electric energy in the whole treatment process of sewage and sludge. The energy selfsufficiency rate of anaerobic digestion had reached 100% in Bailonggang WWTP, Shanghai. By adopting the advanced sludge anaerobic digestion process, Xiaohongmen reclaimed water plant had realized an annual methane output of 13–15 million m³, which could replace around 30–33 million kWh of electric energy [43]. With the rapid development of sludge pretreatment technologies, the biogas production efficiency will be further improved in the future [72,73]. In addition, sludge anaerobic digestion and methane capturing could reduce the unorganized CH₄ emission of sludge landfills, thus reducing the amount of CH₄ discharged directly into the atmosphere. Significantly, a model analysis indicates that due to the low influent organic loads in WWTPs in China, it is difficult to achieve the target of "carbon neutralization" even after implementing biogas energy recovery (i.e., CH₄), but it can completely make up for more than half of the energy consumption and reduce indirect CO₂ emissions by at least 50% [74]. Eventually, for renewable energy utilization strategy, the implementation plans to promote synergy on pollution reductions and carbon mitigations [75] and the Outline of Development Plan 2035 for Urban Water Sector [76] both encourage renewable energy such as thermal, solar, and biogas energy at WWTPs. However, it should be noted that these mitigation strategies are more likely to be applied at large-scale WWTPs [43]. Overall, the renewable energy utilization strategy is expected to reduce the indirect GHG emissions by 10%, 3%, and 4% through extracting the thermal, solar, and biogas energy, respectively, at WWTPs in China (Table S4.

Below are certain constraints that require elucidation. First, the investigation of 3444 centralized WWTPs located in 31 provinces across China was conducted by combining multiple primary data sources, and the investigated WWTPs accounted for about 70% of the total number of WWTPs in China and about 55% of the total volumes of wastewaters treated in the country in 2020. In order to correct this bias, this study applied a correction factor to re-adjust the estimated GHG emissions in WWTPs. This may induce uncertainty in the calculation. Second, we adopted a GHG emission factors data set established by Hua et al. In that study, the GHG emission data set was established through an extensive collection of published on-site GHG monitoring data, and they reported the GHG emission factors from different treatment technologies [23]. However, GHG emission factors could also depend on other factors, such as climate conditions and wastewater management levels. This information has not been emphasized in this study.

4. Conclusion

In this study, we comprehensively investigated GHG emissions from WWTPs in China. Using an extensive database encompassing WWTPs nationwide, we evaluated potential contributions to GHG emission from influent and effluent pollutant concentrations, wastewater volume, and electricity consumption. Our analysis revealed that approximately $1.54 (0.92-2.65) \times 10^4$ GHGs (CO₂-eq) were released annually from WWTPs in China. Most GHG emissions are from N2O and indirect emissions from electricity consumption, accounting for about 90% of emissions from China's WWTPs. Furthermore, we observed a significant upsurge in municipal wastewater discharge in China, escalating from 33 billion m³ in 2000 to 57 billion m³ in 2020, and the number of WWTPs increased from 506 to 4326 during the same period. This surge in WWTPs, coupled with the burgeoning pace of urbanization and the imposition of more stringent effluent discharge standards, is poised to amplify the contribution of WWTP to national GHG emissions.

We propose several strategies to mitigate GHG emissions originating from WWTPs. Firstly, we suggest the application of advanced biological technologies, such as Anammox and AGS, to reduce indirect GHG emissions. Secondly, we recommend adopting smart wastewater management practices to optimize treatment processes and increase energy efficiency, such as upgrading sewage-lifting pumps or adopting intelligent operation modes. Finally, we propose the integration of renewable energies into WWTP operations, such as utilizing heat in wastewater treatment,

applying photovoltaic systems, and recovering biogas energy. Overall, this study underscores the significant GHG emissions from WWTPs in China and the importance of effective strategies to mitigate these emissions. By implementing the proposed strategies, WWTPs in China can reduce their contribution to GHG emissions by 64%, thereby promoting sustainable wastewater management practices.

CRediT authorship contribution statement

Yindong Tong: Conceptualization, Methodology, Writing - Original Draft, Funding Acquisition. Xiawei Liao: Visualization, Formal Analysis. Yanying He: Investigation, Validation, Writing - Original Draft. Xiaomei Cui: Investigation. Marcus Wishart: Formal Analysis. Feng Zhao: Formal Analysis. Yulian Liao: Visualization. Yingxin Zhao: Writing - Review & Editing. Xuebin Lv: Writing - Review & Editing. Jiawen Xie: Software. Yiwen Liu: Methodology, Writing - Review & Editing, Supervision, Funding Acquisition. Guanyi Chen: Writing - Review & Editing. Li'an Hou: Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ese.2023.100341.

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