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Article

Carbon Footprint Analyses of Mainstream Wastewater Treatment Technologies under Different Sludge Treatment Scenarios in China

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Abstract: With rapid urbanization and infrastructure investment, wastewater treatment plants (WWTPs) in Chinese cities are putting increased pressure on energy consumption and exacerbating greenhouse gas (GHG) emissions. A carbon footprint is provided as a tool to quantify the life cycle GHG emissions and identify opportunities to reduce climate change impacts. This study examined three mainstream wastewater treatment technologies: Anaerobic–Anoxic–Oxic (A–A–O), Sequencing Batch Reactor (SBR) and Oxygen Ditch, considering four different sludge treatment alternatives for small-to-medium-sized WWTPs. Following the life cycle approach, process design data and emission factors were used by the model to calculate the carbon footprint. Results found that direct emissions of CO_2 and N_2O, and indirect emissions of electricity use, are significant contributors to the carbon footprint. Although sludge anaerobic digestion and biogas recovery could significantly

contribute to emission reduction, it was less beneficial for Oxygen Ditch than the other two treatment technologies due to its low sludge production. The influence of choosing "high risk" or "low risk" N_2O emission factors on the carbon footprint was also investigated in this study. Oxygen Ditch was assessed as "low risk" of N_2O emissions while SBR was "high risk". The carbon footprint of A–A–O with sludge anaerobic digestion and energy recovery was more resilient to changes of N_2O emission factors and control of N_2O emissions, though process design parameters (*i.e.*, effluent total nitrogen (TN) concentration, mixed-liquor recycle (MLR) rates and solids retention time (SRT)) and operation conditions (*i.e.*, nitrite concentration) are critical for reducing carbon footprint of SBR. Analyses of carbon footprints suggested that aerobic treatment of sludge not only favors the generation of large amounts of CO_2, but also the emissions of N_2O, so the rationale of reducing aerobic treatment and maximizing anaerobic treatment applies to both wastewater and sludge treatment for reducing the carbon footprint, *i.e.*, the annamox process for wastewater nutrient removal and the anaerobic digestion for sludge treatment.

Keywords: carbon footprint; energy recovery; greenhouse gas emission; nitrous oxide; wastewater treatment; sludge treatment

1. Introduction

Environmental problems arising from urban areas have become critical issues facing human society. Globally, 50.5% of the world population was living in cities in 2010. While in China, 45.8% population dwells in urban areas and this number is expected to increase to 70% by 2050 [1]. Cities import water, energy and materials which are transformed into goods and services and ultimately returned to the environment in the form of emissions and waste. Among these urban infrastructures to facilitate public services, wastewater treatment plants (WWTPs) are recognized as a significant energy consumer and source of greenhouse gas (GHG) emissions [2–4], which are threatening global and regional environment and climate. Currently, there are 3513 WWTPs in China [5], and the electricity consumption for wastewater treatment is about 17.5 billion kilowatts/hour (kWh) in 2013, accounting for 0.4% of China's total electricity consumption. With rapid development of WWTPs in Chinese cities, more energy is expected to be consumed in the future for WWTPs construction and operation. According to the national GHG inventory, WWTPs were listed as the 6th largest contributors to methane (CH_4) emissions and 3rd largest sources of nitrous oxide (N_2O) emissions, respectively, accounting for 72% of total CH_4 emissions and 26% of total N_2O emissions [6]. Meanwhile, biosolids generated from the wastewater treatment process are likely to impose potential risks, polluting underground water and soil. In China, more than 60% of WWTPs send wasted sludge for landfill after dewatering and thickening [7], leaving most valuable energy and resources in biosolids untapped, but cause GHG emissions, land occupation and potential risks of underground water pollution from landfill.

Previous studies have highlighted the trade-offs between eutrophication and global warming impact categories caused mainly by effluent discharge, sludge treatment and disposal, and electricity use [2,8]. Driven by more stringent wastewater discharge standards aimed at improving the aquatic environment

by alleviating eutrophication arising from anthropogenic nutrient source, biological nutrient removal (BNR) is being increasingly applied at WWTPs. The overall trend, therefore, is toward increasing energy consumption and chemical dosage per unit of wastewater treated [9]. On the other hand, under the big backdrop of global and national actions to achieve carbon neutrality or even carbon negative development, the Chinese wastewater treatment sector will inevitably need to follow the lead to reduce GHG emissions and cut energy consumption to mitigate its climate change impact. Carbon footprint accounting thus should be adopted as a tool to measure climate change impact in developing technology roadmaps for wastewater treatment and in the decision-making process for taking precautionary measures [10]. With the development of urbanization in China, more WWTPs will be constructed and operated in small and medium cities in the near future; therefore, plants with an average flow rate of 20,000 cubic meters per day (m^3/day) will be the mainstream scale of newly built WWTPs. Assessment of carbon footprints of mainstream treatment technologies to be adopted in these new plants are therefore important for decision-making on process design, operation strategies and performance evaluation to achieve carbon neutrality. Meanwhile, the management of raw sludge is also a critical problem in close relationship with wastewater treatment. The treatment and disposal of raw sludge with increasing production from wastewater treatment process not only brings about potential pollution to the environment, *i.e.*, metals and trace pollutants contained might pollute underground water and soil, but also require energy and chemicals during the process. In previous studies on GHG accounting, most are focused on the GHG emissions from the liquor treatment [4,10–13] or sludge treatment [14–16] separately; less attention has been paid to quantify GHG emissions to a larger extent, with the integration of both wastewater and sludge treatment in the system boundary. The measurement of GHG emissions including both the wastewater and sludge treatment process could shed light on the water–energy–GHG nexus relationship and contribute to the GHG mitigation efforts in WWTPs with synergy effects. To meet this objective, this study aims to estimate and compare carbon footprints of mainstream liquid and sludge treatment portfolios to be considered for new WWTPs built in China, by using plant design data and following the life cycle approach. Extensive analyses and discussion of all treatment scenarios are presented to highlight the variation of carbon footprints of different treatment scenarios and factors that affect GHG emissions.

2. Materials and Methods

2.1. Definition of Carbon Footprint

Following the life cycle approach, the carbon footprint in this study was defined as direct and indirect GHG emissions caused by wastewater and sludge treatment within a defined system boundary. The accounted GHG emissions included carbon dioxide (CO_2), CH_4 and N_2O and were all converted into carbon dioxide equivalents (CO_2e) by global warming potentials (GWPs) over 100 years, namely, 1 for CO_2 25 for CH_4 and 298 for N_2O. It is to be noted that although CO_2 emissions from biological wastewater treatment is generally not considered in GHG inventory of wastewater treatment because of its biogenic origin, some studies pointed out that up to 20% of the carbon present in wastewaters can be of fossil origin [17] and fossil CO_2 emissions from wastewater treatment were underestimated [18]. They were therefore taken into account when quantifying the associated impact [19] in this study.

Within the defined system boundary (as shown in Figure 1), several flows of GHG emissions were estimated and compared among different treatment scenarios. The calculation included direct GHG emissions from wastewater treatment (e.g., CO_2 emissions from organic matters degradation and N_2O emissions from the nitrification/de-nitrification process) and sludge treatment (e.g., CH_4 and N_2O emissions from anaerobic digestion), indirect GHG emissions from sludge final disposal, indirect emissions from production and transportation of construction materials, electricity use and chemicals consumption during operation, and transport of sludge. Although previous studies find that environmental impacts of construction are much less than those of operation and usually are neglected in most life cycle assessment (LCA) studies, these case-specific studies are highly dependent on materials used in different countries and regions and the lifespan of each individual case [2,11,20]. Therefore, no generalization of the impacts of construction is possible and GHG emissions from construction materials are included in this study. The function unit is defined as the treatment of wastewater in one year.

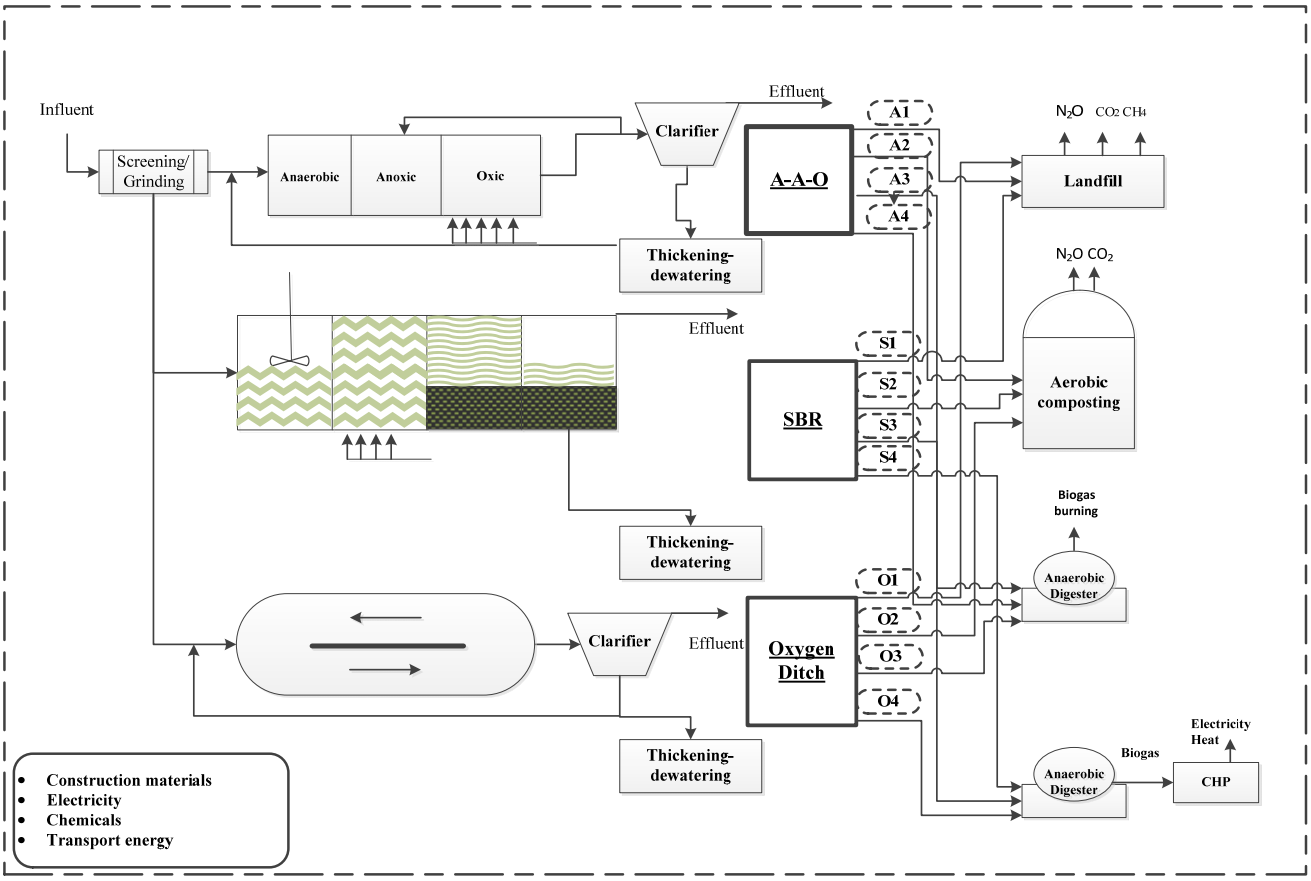


Figure 1. System boundary of this study and studied scenarios for wastewater and sludge treatment.

2.2. Scenarios Studied

In this study, three types of wastewater treatment technologies, Anaerobic–Anoxic–Oxic (A–A–O), Sequencing Batch Reactor (SBR) and Oxygen Ditch (OD), were selected for scenario analyses as these three processes are widely adopted in Chinese WWTPs as the best available technology (BAT) both in terms of plant numbers and treatment capacity [7]. The application of these three types of technologies

with low carbon footprint in small scale WWTPs (flow rate = 20,000 m^3/day) needed to be investigated in this study as sludge treatment and disposal not only concerns environmental pollution problems, but also plays a critical role in reducing the carbon footprint of the whole process. Simply put, the energy content in organic matters in the wastewater is either converted to CO_2 (or CH_4) or becomes wasted sludge through the biological process. Therefore, different pathways of sludge treatment and disposal could result in different levels of GHG emissions and energy utilization. In this study, anaerobic digestion and aerobic fermentation were considered for two sludge treatment alternatives, with landfill as the final sludge disposal method. Further, the utilization of biogas from anaerobic digestion was also studied to examine the contribution of energy recovery to GHG offsets. In addition to the pretreatment of biosolids, four scenarios of sludge treatment and disposal were studied: (1) Direct landfill, currently used by 63% of WWTPs in China; (2) Aerobic composting followed by land application (14% of Chinese WWTPs); (3) Anaerobic digestion with biogas combustion (for safety reasons assuming no CH_4 leakage and all CH_4 oxidized to CO_2 during complete combustion) and biosolids sent for land application; (4) Anaerobic digestion with energy recovery by using combined heat and power (CHP) units, and the biosolids are sent for land application. In all, 12 scenarios of wastewater treatment and sludge treatment and disposal, as shown in dotted circle in Figure 1, were studied: A–A–O with sludge landfill (A1), A–A–O with sludge composting and land application (A2), A–A–O with sludge anaerobic digestion and biogas combustion (A3) and A–A–O with sludge anaerobic digestion and energy recovery via CHP (A4); and SBR with the aforementioned four pathways of sludge treatment and disposal defined as S1, S2, S3 and S4, and lastly, Oxygen Ditch with the four sludge treatment alternatives defined as O1, O2, O3 and O4. Detailed inventory data and operative parameters of different scenarios of wastewater and sludge treatment are described in Table 1.

2.3. Data and Assessment Model

In this study, three treatment alternatives were used in treating 20,000 m^3/day of raw municipal wastewater, which is the mainstream treatment capacity for WWTPs built in China. Three treatment alternatives have a COD of 300–500 mg/L and a 5-day biological oxygen demand (BOD_5) of 180–300 mg/L, and contain 40–48 mg N/L and 3–5 mg P/L in order to meet the Chinese discharge standard (GB18918-2002) of level 1A; Being the most stringent emissions requirements limiting the COD to less than 50 mg/L, BOD_5 to less than 10 mg/L, total nitrogen (TN) to less than 8 mg N/L, NH_3-N to less than 10 mg N/L, and total phosphorus (TP) to less than 0.5 mg P/L. Current national GHG inventory methods used by government for estimating GHG emissions from wastewater treatment process are recommended in the 2006 IPCC Inventory Guidelines [21], which follows a top–down approach, for which technology-specific emission factors are not taken into consideration. In this study, total GHG emissions from wastewater and sludge treatment were calculated by using the method of emissions factors, as shown in Equation (1). Parameters of process configurations were acquired from WWTPs design documents and guidelines for municipal projects. Emission factors were mostly taken from peer-reviewed literature but emission factors of electricity used in this study is adjusted based on national electricity portfolio in China [22]. Emission factors used in this study are listed in Table 2.

Table 1. Operating parameters and inventory data for the twelve wastewater and sludge treatment scenarios of this study.

Sludge Treatment and Disposal Alternatives / **Wastewater Treatment Alternatives**	**A–A–O**	**SBR**	**OD**
	Flow rate: 20,000 m^3/day; CODin = 480 mg/L; CODeff = 50 mg/L; BODin = 216 mg/L; BODeff = 10 mg/L; TNin = 45 mg/L; TNeff = 15 mg/L; SRT = 15 days; MLR Rate = 200; Electricity Demand = 0.305 kWh/m^3; Raw Sludge Production = 5.2 t DS/day; PAC Demand = 138 t/year; PAM Demand = 5.6 t/year.	**Flow Rate: 20,000 m^3/day; CODin = 360 mg/L; CODeff = 50 mg/L; BODin = 180 mg/L; BODeff = 10 mg/L; TNin = 48 mg/L; TNeff = 15 mg/L; SRT = 15 days; MLR rate = 150; Electricity Demand = 0.249 kWh/m^3; Raw sludge Production = 4.3 t DS/day; PAM demand = 4.69 t/year.**	**Flow Rate: 20,000 m^3/day; CODin = 500 mg/L; CODeff = 50 mg/L; BODin = 300 mg/L; BODeff =10 mg/L; TNin = 40 mg/L; TNeff = 15 mg/L; SRT = 15 days, MLR Rate = 200; Electricity Demand = 0.343 kWh/m^3; Raw Sludge Production = 3.7 t DS/day; PAM demand = 4.04 t/year.**
Direct Landfill: Direct landfill after dewatering and thickening by truck transport; Transport distance =20 km; CO_2, CH_4 and N_2O emissions under anaerobic conditions during landfill were taken into account.	A1	S1	O1
Aerobic composting: Static aerobic composting applied; Electricity demand= 10 kWh/t biosolids (80% water content); N_2O and CO_2 were taken into account.	A2	S2	O2
Anaerobic digestion + biogas combustion: Mesophilic digester at 35 ± 2 °C; Assuming biogas, containing 65% (*v*/*v*) CH_4 and 32% (*v*/*v*) CO_2, was completely combusted and converted to CO_2; Electricity demand for heating and mixing = 40 kWh/t biosolids (80% water content).	A3: Biogas production = 1,517 m^3/day	S3: Biogas production = 1,270 m^3/day	O3: Biogas production = 1,094 m^3/day

Table 1. *Cont.*

Wastewater Treatment Alternatives / Sludge Treatment and Disposal Alternatives	A–A–O	SBR	OD
	Flow rate: 20,000 m^3/day; CODin = 480 mg/L; CODeff = 50 mg/L; BODin = 216 mg/L; BODeff = 10 mg/L; TNin = 45 mg/L; TNeff = 15 mg/L; SRT = 15 days; MLR Rate = 200; Electricity Demand = 0.305 kWh/m^3; Raw Sludge Production = 5.2 t DS/day; PAC Demand = 138 t/year; PAM Demand = 5.6 t/year.	**Flow Rate: 20,000 m^3/day; CODin = 360 mg/L; CODeff = 50 mg/L; BODin = 180 mg/L; BODeff = 10 mg/L; TNin = 48 mg/L; TNeff = 15 mg/L; SRT = 15 days; MLR rate = 150; Electricity Demand = 0.249 kWh/m^3; Raw sludge Production = 4.3 t DS/day; PAM demand = 4.69 t/year.**	**Flow Rate: 20,000 m^3/day; CODin = 500 mg/L; CODeff = 50 mg/L; BODin = 300 mg/L; BODeff =10 mg/L; TNin = 40 mg/L; TNeff = 15 mg/L; SRT = 15 days; MLR Rate = 200; Electricity Demand = 0.343 kWh/m^3; Raw Sludge Production = 3.7 t DS/day; PAM demand = 4.04 t/year.**
Anaerobic digestion + CHP: Mesophilic digester at 35 ± 2 °C; Electricity demand for heating and mixing = 40 kWh/t biosolids (80% water content); Biogas CHP unit to recover 70% energy, 30% for electricity and 40% for heat.	A4: Electricity production by CHP = 2,904 kWh/day; Heat production by CHP = 13,956 MJ/day.	S4: Electricity production by CHP=2,430 kWh/day; Heat production by CHP=11,681 MJ/day.	O4: Electricity production by CHP = 2,094 kWh/day; Heat production by CHP = 10,064 MJ/day.

Notes: CODin = influent COD concentration; CODeff = effluent COD concentration; BODin = influent BOD concentration; BODeff = effluent BOD concentration; TNin = influent TN concentration; TNeff = effluent COD concentration; MLR = Mix-liquor recycle; DS = dry sludge.

$$E_{i,j} = \sum AD_{i,j} \times f_{i,j} \quad (1)$$

where, $E_{i,j}$ Emissions of type i GHG from source j;

$AD_{i,j}$, activity data of type i GHG from source j, e.g., fuel consumption, electric power consumption, materials consumption;

$f_{i,j}$, emission factors of type i GHG from source j activity;

i, types of GHGs, three types of GHGs are considered in this study, CO_2, N_2O, CH_4;

j, categories of GHG emission sources, e.g., electric power consumption, organic matters degradation from wastewater treatment.

Table 2. Emission factors for greenhouse gas (GHG) calculation used in this study.

Parameters	Value	Unit	Reference
Wastewater Treatment			
CO_2 from OM oxidation	1.375	kg CO_2/kg $BOD_{removed}$	[23]
N_2O from denitrification	0.035	kg N_2O-N/kg $N_{denitrified}$	[24]
Sludge Treatment and Disposal			
N_2O from composting	0.700	g N_2O-N/kg DS	[25]
N_2O from landfill	8.200	g N_2O/kg $N_{applied}$	[9]
CH_4 from landfill	13.400	g CH_4/kg sludge	[20]
CO_2 from landfill	35.120	g CO_2/kg sludge	[20]
N_2O from biogas combustion	0.004	g N_2O/kg CH_4 burned	[26]
Chemicals			
Polymers (PAM) for dewatering	1.500	kg CO_2e/kg	[27]
Polyaluminium chloride (PAC)	22.700	g CO_2e/kg	[28]
Energy			
Electricity	0.681	kg CO_2e/kWh	[22]
Diesel	72.600	kg CO_2e/GJ	[6]
Coal	90.800	kg CO_2e/GJ	[6]
Construction Materials			
Cement	0.405	kg CO_2e/kg	Estimated
Steel	2.196	kg CO_2e/kg	Estimated
Timber	0.706	kg CO_2e/kg	Estimated
Sand	0.009	kg CO_2e/kg	Estimated
Gravel	0.009	kg CO_2e/kg	Estimated
Cast iron pipe	3.096	kg CO_2e/kg	Estimated
Steel pipe and fittings	3.096	kg CO_2e/kg	Estimated
Reinforced concrete pipe	0.095	kg CO_2e/kg	Estimated

Regarding emissions from the construction phase, material consumption was calculated based on project design documents and the Municipal Projects Investment Estimation Index [29]. GHG emission factors were determined based on the Inventory of Carbon and Energy, as presented in Table S1 in Supplementary Information [30]. Since the inventory data are estimated from the typical fuel mix of relevant UK industry, emission factors in this study were then adjusted based on the fuel mix of relevant industries in China [31–34], and details were described in Tables S2–S4 in Supplementary Information.

Meanwhile, in order to guarantee the transparency of the results, validation of emission factors of construction materials was discussed in Supplementary File and emission factors from Ecoinvent database were also provided in Table S5 for validation. It was assumed that unit energy consumption for transports of construction materials is 1836 kilojoules (kJ) per ton per kilometer (km) and the average distance is 25 km [11]. The lifespan of the wastewater treatment facilities is assumed to be 20 years.

In the operational phase, indirect GHG emissions from on-site electric power and chemicals consumption were considered in this study. On-site electric power is mainly required for aeration, sludge return, mixed liquid recirculation, sludge dewatering and mixers. The amount of electric power consumption was calculated based on the working loads of major power-consuming equipment, e.g., blowers, water pumps, sludge pumps and mixers. Meanwhile, flocculants such as Polyacrylamide (PAM) are consumed in sludge thickening and dewatering at a dosing rate of 3–4 kg per metric ton of dry sludge. The details of calculating the production of dry sludge were provided in Supplementary Information. With regard to N_2O emissions, as consensus on the main mechanism of N_2O emission during wastewater treatment has not yet been achieved and there are many process design parameters and operation conditions closely related to N_2O emission, the emission factor selected in this study was based on an average value of N_2O measurement from full-scale WWTPs [24]. Risks of choosing different emissions factors of N_2O are further discussed later.

In the scenario of sludge anaerobic digestion with biogas utilization for CHP, thickened sludge is sent to mesophilic digester for anaerobic digestion at a temperature of 35 ± 2 °C. Biogas from anaerobic digestion contains 65% of CH_4 and 32% of CO_2 [35], and then can be used in CHP Unit to generate electric power and heat via biogas turbines. Meanwhile, commercial-scale biogas CHP units can realize total energy conversion efficiency of 70%, 30% for electricity and 40% for heat, separately [20]. Thus, electricity and heat gains from CHP can be determined by biogas production (detailed information can be seen in the Supplementary Information), multiplying the assumed caloric value of biogas at 23 MJ/m^3 by respective energy conversion efficiencies. It is assumed that electricity could displace purchased grid power for WWTP operation and heat could be used to maintain the proper operating temperature of the digester instead of heat from a coal-fired boiler (assuming a heat conversion efficiency of 70%).

3. Results and Discussion

3.1. Carbon Footprint

Carbon footprints of twelve wastewater and sludge treatment alternatives at the flow rate of 20,000 m^3/day ranged from 5817–9928 t CO_2e per year (Table 3), with none of these treatment alternatives achieving carbon neutrality. As shown in Figure 2, SBR with sludge anaerobic digestion and energy recovery via CHP (S4) had the lowest carbon footprint (5817 t CO_2e/year), while A–A–O with sludge landfill (A1) had the highest carbon footprint (9928 t CO_2e/year). As a whole, for each of the three wastewater treatment alternatives, different sludge treatment pathways led to different levels of GHG emissions. For four different sludge treatment scenarios, direct landfill gave rise to highest GHG emissions due to its generation of potent CH_4 emissions, while anaerobic digestion brought about much lower emissions; in particular, the case of biogas utilization resulted in significant emission offsets (1186, 992, and 855 t CO_2e/year for A–A–O, SBR and Oxygen Ditch, respectively). Although aerobic

fermentation emitted less GHG emissions than direct landfill, higher energy consumption requirement for aeration (around 10 kWh/t biosolids) significantly contributed to the overall carbon footprint. Compared with sludge direct landfill in each wastewater treatment alternatives, sludge anaerobic digestion and biogas utilization with CHP could help to reduce 37% carbon footprint for A–A–O, 34% for SBR and 24% for Oxygen Ditch. Although sludge aerobic fermentation could also contribute to carbon footprint reduction, emissions reduction (8%–10%) is not as significant as anaerobic digestion with energy recovery. Sludge aerobic fermentation has a high degradation rate of organic matters and generates large amounts of CO_2 (1607–2259 t CO_2/year for three wastewater treatment alternatives), and the generation rate of fugitive N_2O (0.9–1.3 t N_2O/year for three wastewater treatment alternatives) is much larger than that of biogas combustion following sludge anaerobic digestion (0.7–1.0 kg N_2O/year for three wastewater treatment alternatives), so that is why the carbon footprint of sludge aerobic fermentation was still larger than that of sludge anaerobic digestion with biogas combustion and utilization. Meanwhile, for three of the sludge treatment alternatives (landfill, composting, anaerobic digestion and biogas combustion), the A–A–O process had the highest GHG emissions, Oxygen Ditch had the second highest, and SBR had the lowest GHG. However, for sludge anaerobic digestion with energy recovery via CHP, Oxygen Ditch had the largest carbon footprint, and A–A–O process had the second largest carbon footprint and SBR still had the lowest. This is because energy recovery via the biogas CHP system contributed to emission offsets and was significantly dependent on sludge production, biogas generation rate and biogas utilization efficiency. In this study, assuming other factors remain the same among different treatment alternatives, sludge production was the key determinant on energy recovery and emissions offsets, and sludge production from Oxygen Ditch was much lower than the other two wastewater treatment alternatives due to its process characteristics of extended aeration, longer solids retention time (SRT) and more stabilized biosolids.

According to the assessment of carbon footprints for the different treatment alternatives, as shown in Figure 3, direct emissions of CO_2 (22%–49%) and N_2O (23%–36%), and electricity (14%–26%) were significant contributors to GHG emissions. According to our study, CO_2 emissions, although traditionally not taken into account, can be of similar importance to electricity-associated ones if 50% are supposed not be of biogenic origin. In the scenario of sludge direct landfill, CH_4 from landfill process was also a significant source and contributed to 25%–32% of carbon footprints of three wastewater treatment alternatives. Although indirect emissions from construction materials (4%–6% of total GHG emissions) were not as significant as electricity and direct emissions of CO_2 and N_2O, they were still larger than indirect emissions from chemicals (0.07%–0.18% of total GHG emissions) and transports (0.1%–0.4% of total GHG emissions).

Table 3. Carbon footprints and GHG emissions from different sources for the twelve wastewater and sludge treatment scenarios.

Scenarios	Total GHG Emissions (t CO_2e/year)	GHG Emissions from Construction (t CO_2e/year)	GHG Emissions from Chemicals (t CO_2e/year)	GHG Emissions from Electricity Use (t CO_2e/year)	GHG Emissions from Transport (t CO_2e/year)	CH_4 Emissions (t CO_2e/year)	N_2O Emissions (t CO_2e/year)	CO_2 Emissions (t CO_2e/year)	GHG Offsets (t CO_2e/year)
A1	9928	364	11	1516	41	3127	2501	2367	n/a
A2	8870	332	7	1237	34.5	0	2683	1957	n/a
A3	9047	333	6.1	1705	31.3	0	2054	2662	n/a
A4	8912	364	11.4	1693	9.2	0	2649	4298	−1186
S1	7995	332	7	1384	8.4	2618	2806	3551	n/a
S2	8291	333	6.1	1831	8.8	0	2159	4033	n/a
S3	7466	364	11.4	1774	9.2	0	2253	3054	n/a
S4	6809	332	7	1451	8.4	0	2478	2532	−992
O1	7271	333	6.1	1889	8.8	2255	1878	3157	n/a
O2	6280	364	11.4	1774	9.2	0	2253	3054	n/a
O3	5817	332	7	1451	8.4	0	2478	2532	n/a
O4	6862	333	6.1	1889	8.8	0	1878	3157	−855

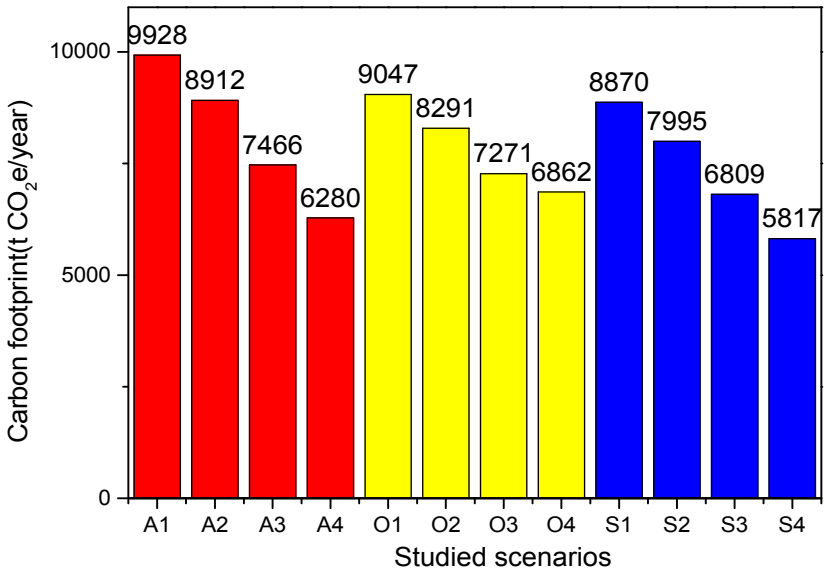


Figure 2. Carbon footprint of the twelve wastewater and sludge treatment scenarios.

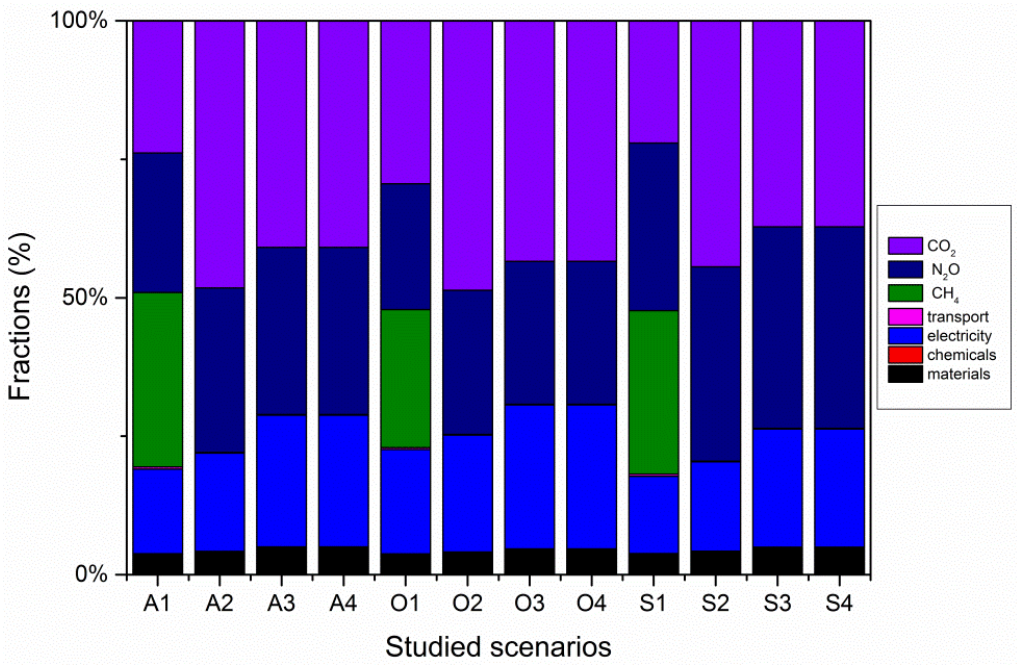
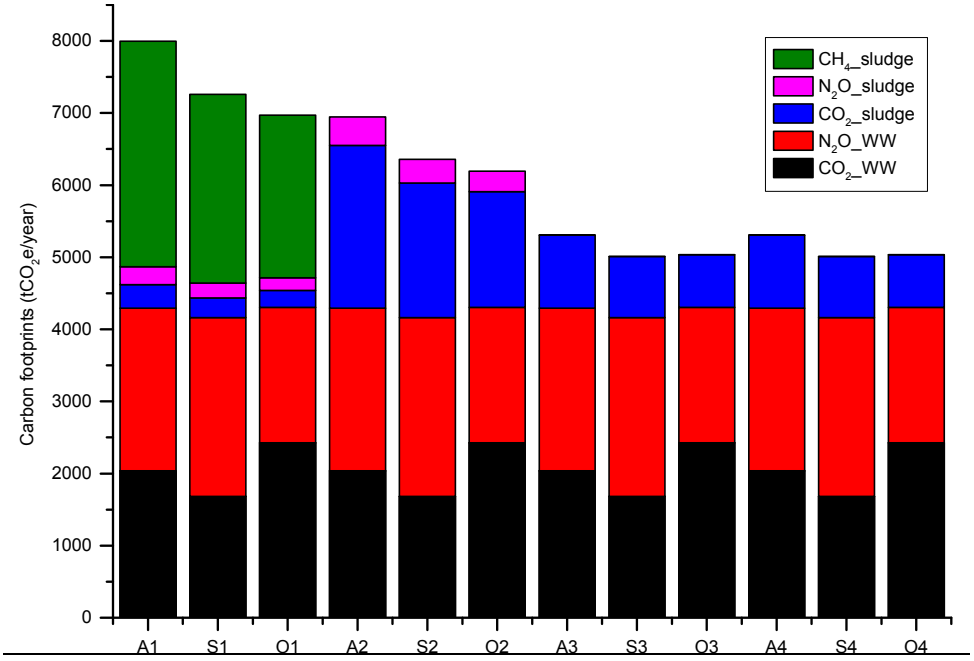


Figure 3. Proportions of different sources for GHG emissions of wastewater and sludge treatment.

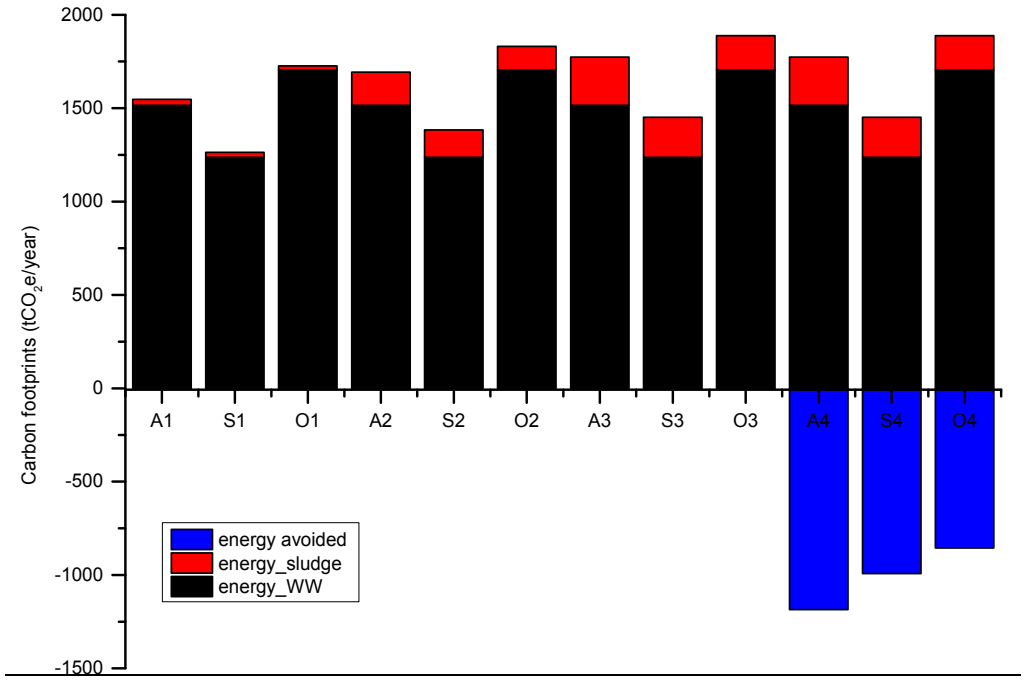
According to the depiction of direct GHG emissions from wastewater and sludge treatment in Figure 4a, CH_4 from landfill accounted for around a third (32%–39%) of the total direct GHG emissions in the sludge direct landfill scenario, but CO_2 and N_2O emissions from wastewater treatment dominated in most cases, ranging from 23%–48%, and 27%–49%, of total direct GHG emissions, respectively. N_2O emissions from sludge anaerobic digestion were very small (0.004%–0.006% of total direct emissions), while N_2O from landfill were ranging from 2%–3% of total direct emissions and that from aerobic fermentation ranged from 4%–6%. It is indicated that aerobic conditions was more favorable for N_2O emissions than anaerobic conditions in landfill and anaerobic digestion, and N_2O emissions from

sludge aerobic fermentation need to be controlled to lower the overall carbon footprint. Besides, CO_2 emissions from aerobic fermentation were also significant, accounting for 26%–33% of total direct emissions. While CO_2 emissions from anaerobic digestion were much less, only contributing to 15%–19% of total direct emissions. This is because under anaerobic conditions the degradation rate of biodegradable organic matter is much lower than aerobic conditions in fermentation. It suggests that aerobic condition was not only favorable for N_2O emissions but also CO_2 emissions during sludge treatment. Meanwhile, indirect emissions from electricity and transports during wastewater and sludge treatment and disposal in Figure 4b suggested that emission from electricity used for wastewater treatment significantly dominated the indirect GHG emissions, namely, 85%–99%. GHG emissions from electricity consumption during sludge anaerobic digestion, mainly for mixing and heating, were also significant, accounting for 10%–15% of total indirect GHG emissions, which is consistent with conclusions in previous studies that energy consumption in anaerobic digestion usually accounts for around 15% of total energy consumption in wastewater treatment [20]. It has to be noted that GHG emission offsets resulted from recovered electricity and heat from biogas CHP system contributed to 45%–68% of emissions reductions from energy consumption. Therefore, the results suggest that sludge anaerobic digestion is a very promising sludge treatment alternative to achieve energy recovery and lower carbon footprint. However, it should be realized that emission offsets from energy recovery in Oxygen Ditch were the lowest, 45% of reduction in indirect GHG emissions and SBR gained the highest emissions offsets, 68% of reduction in indirect GHG emissions. This is because electricity consumption for wastewater treatment was highest (1705 tCO_2e/year) for Oxygen Ditch, while emission offsets from recovered energy was the lowest (855 tCO_2e/year) for Oxygen Ditch due to its low sludge production and resulting biogas generation. Therefore, it could be concluded that sludge anaerobic digestion and energy recovery is less cost-effective and beneficial for Oxygen Ditch than that for SBR and A–A–O processes.



(a)

Figure 4. *Cont.*



(**b**)

Figure 4. The most important contributors to carbon footprint: (**a**) Direct emissions from wastewater treatment and sludge treatment; and (**b**) Indirect emissions from energy consumption (electricity and transport fuels) in wastewater treatment and sludge treatment.

3.2. Emission Factors of N_2O

As mentioned in Section 2.3, the consensus on the main mechanism of N_2O emission during wastewater treatment has not yet been achieved and there are many process design parameters and operation conditions closely related to N_2O emission, the emission factor selected in this study is based on an average value of the measurement of N_2O emissions from seven full-scale biological nutrient removal (BNR) WWTPs in Australia [24]. The measurements found that particular design parameters, e.g., high mixed-liquor recycle (MLR) rates and low effluent TN concentrations, potentially influence the generation of N_2O and higher MLR rates, and lower effluent TN concentrations were more likely to have higher and variable N_2O generation factors. The design of low effluent TN concentration, influent flow balancing, high MLR rate, larger bioreactor volume, and long SRT is more likely to lead to complete denitrification, which could result in low and stable N_2O emissions. For example, a high MLR rate tends to dilute the concentrations of all the intermediates of nitrification–denitrification, including nitrite (NO_2^-) and nitric oxide (NO), thus reducing their inhibitory effect. In light of the possible N_2O formation mechanisms, Foley, deHaas, Yuan and Lant [24] suggests that wastewater treatment process designed and operated for low effluent TN concentrations and that approach "ideal" well-mixed hydraulic conditions are expected to have relatively low N_2O generation factors, and *vice versa*.

The design parameters of three wastewater treatment alternatives are listed in Table 4. It was found that the designed effluent TN concentrations were all set at 15 mg/L, thus may lead to partial denitrification. Oxygen Ditch had the highest MLR rate and SBR had the lowest MLR rate, which indicates that higher MLR rate in Oxygen Ditch is more likely to depress the inhibitory effect by intermediates and reduce N_2O emissions than A–A–O and SBR. Besides, the wastewater treatment

alternatives all had relatively long SRT (*i.e.*, 18–22 days), which suggest relatively slow change in biomass inventory. It is suggested that plants that do not have a high degree of denitrification and approach more "plug flow" hydraulic conditions are more likely to occur nitrite accumulation and high N_2O generation factors [24]. While SBR which operate timed process sequences is similar to plug flow continuous reactors, thus resulting in higher N_2O generation factors. Therefore, in this study, Oxygen Ditch is at low risk of N_2O emission, while SBR is at high risk of N_2O generation. N_2O generation factors from measurement of seven full-scale BNR WWTPs varied in the range 0.006–0.253 kg N_2O-N/kg $N_{denitrified}$ with average value at 0.035 ± 0.027. In order to examine the influence on carbon footprint by different emission factors of N_2O, the study employed two levels of assessment at "high risk" and "low risk" of N_2O generation factors, together with the baseline estimation using a unified emission factor in abovementioned carbon footprint assessment. In the "low risk" estimation, the emission factors of N_2O were selected as the average value of the data range for each of the alternatives measured in [24]. And for the "high risk" estimation, emission factors were determined as the maximum value of the data range, shown as in Table 2.

Table 4. Design parameters of three wastewater treatment alternatives and adjustment of N_2O emission factor.

Treatment Technology	Effluent TN (mg/L)	MLR Rate (%)	SRT (days)	N_2O Emission Factor (kg N_2O-N/kg $N_{denitrified}$) [24]	N_2O Emission Factor (kg N_2O-N/kg $N_{denitrified}$) Baseline	N_2O Emission Factor (kg N_2O-N/kg $N_{denitrified}$) High Risk	N_2O Emission Factor (kg N_2O-N/kg $N_{denitrified}$) Low Risk
A–A–O	15	200	18	0.010–0.018 (ave. 0.014)	0.035	0.018	0.014
SBR	15	150	20	0.010–0.071 (ave. 0.033)	0.035	0.071	0.033
Oxygen Ditch	15	300	22	0.006–0.013 (ave. 0.008)	0.035	0.013	0.008

After the adjustment, SBR with sludge direct landfill (S1) occupied the largest carbon footprints and A–A–O with sludge anaerobic digestion and energy recovery (A4) showed the lowest carbon footprints for both "low risk" and "high risk" estimation of N_2O emission factors. As shown in Figure 5, compared with "baseline" estimation, carbon footprints of all treatment scenarios for "low risk" estimation were reduced, with scenario O4 reduced most of 28%. Therefore, in the "low risk" estimation, carbon footprint of O4 was more sensitive than other treatment scenarios in reducing carbon footprints. It is to be noted that except the scenarios with sludge anaerobic digestion and energy recovery, for other three sludge treatment and disposal alternatives (direct landfill, aerobic fermentation, anaerobic digestion and biogas combustion), SBR had the largest carbon footprint, A–A–O had the second largest carbon footprint, and Oxygen Ditch had the lowest carbon footprint. While for sludge anaerobic digestion and energy recovery, A–A–O was calculated for lowest carbon footprint due to its largest biogas production and recovered energy to avoid emissions from electricity and coal consumption, and Oxygen Ditch had the second largest carbon footprint and SBR still had the largest carbon footprint. While in the "high risk" estimation, due to the highest emission factor applied carbon footprint of SBR scenarios (S1, S2, S3 and S4) all increased significantly from the "baseline" level. Among them, carbon footprint of scenario S4

was increased most, namely, 43.8% from the level of baseline estimation. This is because N_2O emissions in scenario S4 accounted for 42.6% of total emissions, the largest fraction among all the scenarios. Therefore, carbon footprint of S4 was very sensitive to N_2O emission factor so that the control of N_2O emissions in SBR is crucial for reducing its carbon footprint. In both estimations of N_2O emission factors, scenario A4 displayed the lowest carbon footprint, which indicates its better performance on resilience to changes of N_2O emission factors and leads the way to reduce overall carbon footprint of wastewater treatment and sludge treatment process.

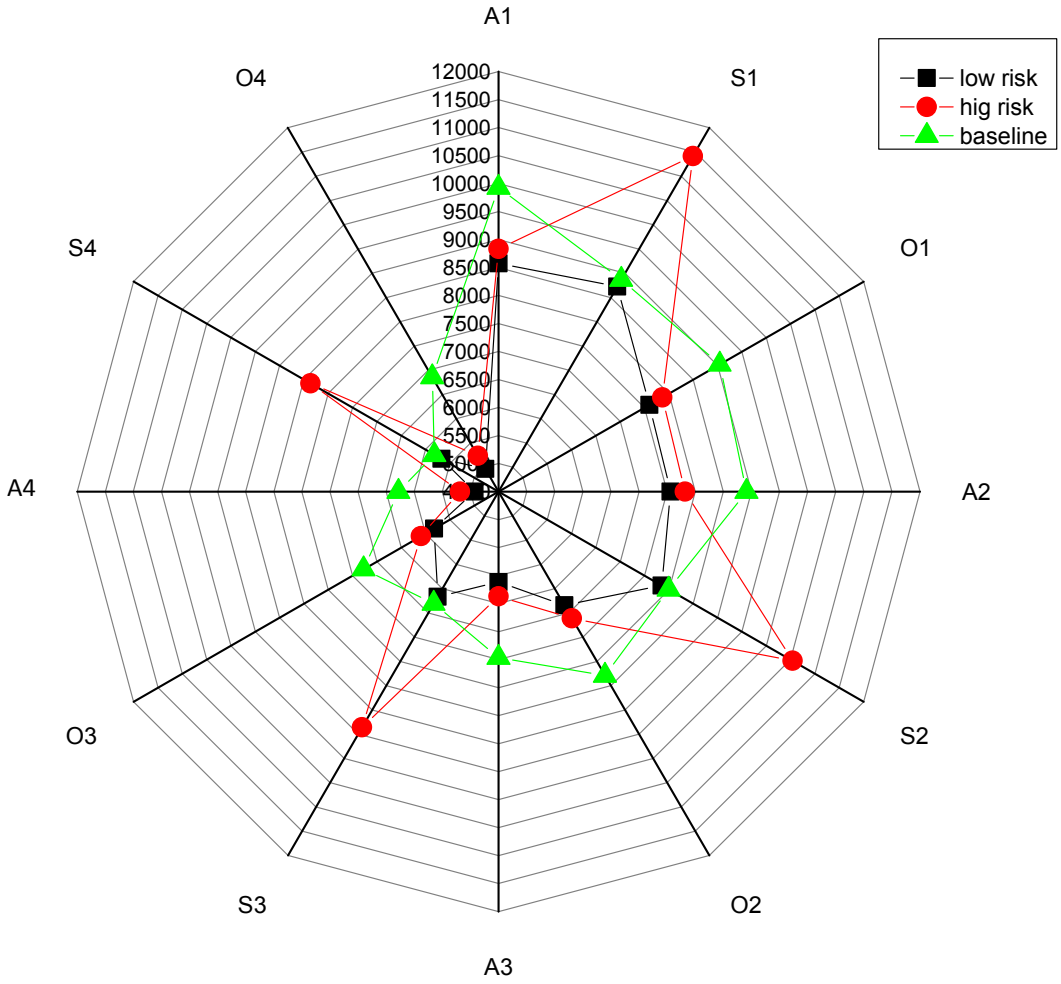


Figure 5. Carbon footprints under different N_2O emission factors at low risk, high risk and baseline estimation.

3.3. Measures to be Taken for Reducing Carbon Footprint

Carbon neutrality or negative carbon footprint of WWTPs has been widely discussed and investigated. Theoretically, the energy contained in the wastewater is far beyond the energy required [3], thus carbon neutrality should be possible. Energy content in wastewater, in the form of chemical oxygen demand (COD), is usually converted into CO_2 or CH_4 and biosolids through either aerobic treatment or anaerobic treatment. Therefore, decreasing the degree of aerobic treatment and maximizing energy recovery from CH_4 and biosolids are crucial to lower carbon footprint. From the perspective of energy consumption, anaerobic wastewater treatment is more favorable. However, municipal wastewater is characteristic of

low temperature and low concentrations, which make it difficult for efficient anaerobic treatment. An efficient precipitation in pre-settlers with sludge anaerobic digestion could contribute to the decrease of the degree of subsequent aerobic treatment, by removing certain amount of COD and reducing CO_2 emissions, and recover energy from CH_4 by anaerobic digestion. Nevertheless, denitrification requires COD and addition of external carbon source can increase carbon footprint. Anaerobic hydrolysis of sludge produces a favorable carbon source which can be used as external carbon source for denitrification.

The N_2O emissions from the wastewater treatment significantly affect the footprint of carbon emissions. Process design and operation condition can crucially have influences on the process of denitrification, thus determining the generation of N_2O emissions [24,36]. Parameters of process design that can facilitate complete denitrification, *i.e.*, low effluent TN concentration, high MLR rate, long SRT, and large reactor volume, can reduce the formation of N_2O. Meanwhile, annamox process could lead to less energy consumption and reduce N_2O emissions during the process. However, the dominant microbial of annamox preferred an environment of higher temperature, thus heat energy is required and a renewable energy source for heating is appropriate.

The rationale of reducing aerobic treatment and maximizing anaerobic treatment also applies to sludge treatment for reducing the carbon footprint. In this study, carbon footprints of sludge anaerobic digestion were much lower than sludge landfill and aerobic fermentation. In addition, the utilization of biogas generated from anaerobic digestion further contributed to offset GHG emissions by avoiding purchased electricity and coal for heating. Biogas production is also a key factor influencing the amount of recovered energy in the case of Oxygen Ditch, of which the characteristic of extended aeration resulted in less production of sludge fed into anaerobic digester. Therefore, improving biogas production and energy conversion efficiency is the subject of further research regarding their contribution to carbon emissions reduction.

3.4. Limitations

Although this study examined the carbon footprints of several mainstream wastewater treatment and sludge treatment technologies applied in Chinese WWTPs, several limitations exist and more research is needed for further study and application. First of all, because of the effect of scale economy, the results of carbon footprints in this study only apply to the studied treatment scale of 20,000 m^3/day. Different treatment scales could lead to a disproportionate decrease or increase in emissions and energy consumption. Secondly, by using the process design data, this study does not take into consideration the impacts of operational conditions on carbon footprints. The operational condition is a dynamic process, and any change will influence the emission factors, energy consumption and chemicals consumption. Therefore, additional research needs to be conducted investigating carbon footprints under various operation conditions.

4. Conclusions

In order to investigate the carbon footprints of future mainstream WWTPs at the flow rate of 20,000 m^3/day in China, 12 scenarios consisting of three wastewater treatment alternatives and four sludge treatment pathways were examined. The carbon footprints ranged from 5817–9928 t CO_2e per year, with the lowest carbon footprint from SBR with sludge anaerobic digestion and energy recovery

via CHP, and the largest carbon footprint was from A–A–O with sludge landfill. Sludge anaerobic digestion and biogas utilization with A–A–O, SBR and Oxygen Ditch helped to reduce the carbon footprints, by 37%, 34%, and 24%, respectively, from the sludge landfill scenarios. There were three significant sources for GHG emissions, namely, direct emissions of CO_2 (22%–49%) from aerobic treatment of wastewater and sludge, direct N_2O emissions (23%–43%) from wastewater treatment, and indirect emissions from electricity use (14%–28%). According to our study, CO_2 emissions, although traditionally not taken into account, can be of similar importance to electricity-associated ones if 50% are supposed to not be of biogenic origin. Whether or not to include direct CO_2 emissions in GHG accounting, is highly dependent on the sources of wastewater; the study results still highlighted that if no CO_2 emissions are considered at all during GHG accounting of wastewater and sludge treatment, it could, at a high probability, introduce bias to the result. Although indirect emissions from construction materials (4%–6%) were not as significant as direct GHG emissions and emissions from electricity, they were much larger than indirect emissions from chemicals (0.07%–0.18%) and transports (0.1%–0.4%). As N_2O emissions significantly influence the carbon footprint of wastewater treatment, emission factors of N_2O need to be deliberately chosen. Since SBR operates in timed sequences, and is similar to plug flow continuous reactors which is likely to have relatively steep concentration gradients throughout the reactor, incomplete denitrification is more likely to occur, thus resulting in higher N_2O emission factors. Therefore, the process design (*i.e.*, effluent TN concentrations, MLR rates, SRT, reactor volume) and operation conditions (*i.e.*, DO, nitrite concentration) are crucial for SBR to reduce N_2O emissions. More detailed research and onsite measurement are therefore needed in the future to investigate N_2O emission under different process configurations and operational conditions.

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Author Contributions

Yujie Feng conceived the paper and Man Sing Wong contributed to analysis tools and proofreading; Chunyan Chai performed data analysis and wrote the paper; Dawei Zhang and Yanling Yu contributed to technical expertise on wastewater treatment and sludge treatment and disposal.

Conflicts of Interest

The authors declare no conflict of interest.

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