Greenhouse gas emissions from municipal wastewater treatment facilities in China from 2006 to 2019

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Wastewater treatment plants (WWtPs) alleviate water pollution but also induce resource consumption and environmental impacts especially greenhouse gas (GHG) emissions. Mitigating GHG emissions of WWtPs can contribute to achieving carbon neutrality in China. But there is still a lack of a high-resolution and time-series GHG emission inventories of WWtPs in China. In this study, we construct a firm-level emission inventory of WWtPs for CH4, N2O and CO2 emissions from different wastewater treatment processes, energy consumption and effluent discharge for the time-period from 2006 to 2019. We aim to develop a transparent, verifiable and comparable WWTP GHG emission inventory to support GHG mitigation of WWtPs in China.

Background & Summary

Municipal wastewater treatment facilities are the main technical solution to mitigating water pollution. But wastewater purification in WWtPs and other treatment facilities always comes at the cost of energy consumption, use of chemicals and environmental impacts12, among which, GHG emissions are of most concern14. Even though GHG emissions from wastewater make only a small contribution to global anthropogenic GHG emissions, it is still important to map GHG emissions from wastewater treatment systems, and to set reasonable targets for mitigation of GHG emissions5,6. To achieve these purposes, a comprehensive GHG inventory is a prerequisite. There have been numerous studies establishing GHG accounts of WWtPs7–13, but there still exist challenges and problems.

Current GHG accounts often do not consider differences of treatment processes/technologies. The accounting of GHG emissions of WWtPs at the regional level mainly uses IPCC emission factors, where the centralized biological treatment processes are only categorized into aerobic and anaerobic processes but neglect the differentiation of sub-categories of aerobic or anaerobic technologies7,10–14, leading to large uncertainties of GHG emission factors. To accurately account GHG emissions in WWtPs, detailed processes/technologies should be considered and analysed.

Frequently, only CH4 and/or N2O are accounted for, excluding CO2 emissions of biological treatment processes as these are generally derived from modern (biogenic) organic matter in human excreta or food waste and should not be included in national total emissions (IPCC 2019, Volume 5, Chapter 6, Page 7)15. But intensive research has shown that a significant amount of fossil CO2 are directly emitted from WWtPs, and assuming that all direct CO2 emissions are biogenic may underestimate GHG emissions16–20.

Dissolved GHG in the treated effluent themselves have the potential to be released. In addition, many waterways are in eutrophic or nutrient-rich conditions, which can further induce discharged wastewater to increase
GHG emissions. However, GHG emissions from receiving waters are rarely accounted for, due to a lack of data of the water quality of the recipient body of water and downstream discharge pathways. Even though some studies considered off-site emissions from the treated effluent, only one discharge pathway of entering rivers, lakes or oceans was assumed. To account emissions from different discharge pathways (such as direct discharge into rivers, lakes, reservoirs, seas, soil, and sewage irrigated farmland) is essential for identifying key emission sources, GHG composition and their contribution to the whole wastewater treatment system.

Existing regional- or national-level studies on GHG emissions accounting of wastewater treatment systems are not comparable. This is mainly due to different emission factors and data sources in different studies. For example, Zhao, et al. used firm-level activity data and IPCC 2006 emission factors to calculate CH₄ emissions, while emission factors of Yan, et al. were obtained from the average of four references excluding IPCC emission factors, and provincial-level activity data from China Environment Yearbook and China Statistical Yearbook. Differences in applied methodology and data sources contribute to a factor 38 difference in calculated CH₄ emissions for the same year.

To solve the above gaps, we constructed a high-resolution (firm-level) and time series (from 2006 to 2019) GHG emission inventory of WWTPs in China. Emission sources include on-site emissions from biological treatment processes and off-site emissions from energy consumption and discharge pathways of the WWTP. We distinguished between 10 potential pathways: direct and indirect (after sewers) discharge into seas; direct and indirect discharge into rivers, lakes, reservoirs etc.; municipal WWTPs; direct discharge onto sewage irrigated farmlands; discharge onto land; other facilities (decentralized wastewater treatment facilities); centralized industrial WWTPs and other discharge pathways. To account for the different emission potentials of different treatment technologies, we calculated emissions based on 48 separate biological, physical, chemical and physicochemical technologies and their combinations. GHG emission factors of different biological treatment technologies in line with China’s conditions were obtained from the literature. Three GHG were estimated in this research, i.e., CO₂, N₂O and CH₄. We did not distinguish between fossil CO₂ and biogenic CO₂ emissions from biological treatment but regarded CO₂ emission as the sum of fossil CO₂ and biogenic CO₂ emissions.

**Methods**

We include GHG emissions from domestic wastewater treated by municipal WWTPs and other facilities in this paper. The other facilities mainly collect and treat wastewater discharged from residential areas, tourist facilities, resorts, nursing homes, airports, railway stations, and other public places. Domestic wastewater collected by both municipal WWTPs, and other facilities maybe mixed with industrial wastewater under certain conditions. In this case, IPCC 2019 suggests that the mixed domestic and industrial wastewater can be considered as domestic wastewater.

GHG emissions of a WWTP result from on-site and off-site emissions. On-site emissions are usually defined as emissions induced by wastewater and sludge treatment processes of WWTPs. In our study, the system boundary excludes GHG emissions from sludge treatment and disposal processes in a WWTP due to lack of data, even though it is reported that sewage sludge treatment and disposal processes account for about 40% GHG emissions in wastewater systems. On the other hand, generated CH₄ emissions from a WWTP are rarely recovered or flared in China, we regard recovered or flared CH₄ emissions as being zero. Therefore, on-site emissions only refer to emissions from wastewater treatment procedures in this research. For various wastewater treatment technologies, biological treatment technologies generate on-site GHG emissions during wastewater treatment processes, but physical, chemical, and physicochemical treatment technologies do not. Off-site emissions refer to emissions from effluent, electricity consumption, production and transportation of chemicals. But we exclude off-site emissions generated by chemicals’ production and transportation due to lack of data for each WWTP, and they being negligible compared with electricity consumption. CO₂ emissions from electricity consumption are fossil CO₂, because they come from coal-fired power generation, but CO₂ emissions generated by on-site wastewater treatment and off-site effluent are mixed with fossil CO₂ and biogenic CO₂, as influent and effluent COD may contain both fossil and biogenic carbon.

Figure 1 shows a flowchart of the construction of the firm-level GHG emission inventory of wastewater treatment facilities from 2006 to 2019 in China. The first step to quantify GHG emissions of a WWTP is to judge the applied treatment technology. If the WWTP adopts biological treatment, on-site emissions from the biological treatment process are calculated. Otherwise, off-site emissions from electricity consumption and the discharge pathway for each WWTP are quantified. Calculation of GHG emissions from each emission source was based on the multiplication of emission factors and activity data. The activity data for each WWTP was collected from China Environmental Statistics Database (CESD).

**Classification of wastewater treatment technologies and its priority.** To examine GHG emissions of different wastewater treatment processes, we need to decide the category of technology applied in each WWTP. In most cases, a WWTP has a primary, secondary or tertiary treatment process, and for each process, especially in secondary treatment, more than one technology may be applied. It is impossible to quantify on-site GHG emissions for each technology, since we only collected data on concentration of influent and effluent pollutants for the whole WWTP, rather than for each technology or process. Therefore, to simplify the calculations of on-site GHG emissions, we first need to judge the main category of treatment technology of a WWTP, and then choose the corresponding emission factors of CH₄, N₂O and CO₂ to calculate GHG emissions generated by biological treatment processes. The technology classification is presented in Table 1. A decision tree for determining the category of treatment technology of a WWTP is shown in Fig. 2.

**On-site emissions from biological treatment.** CH₄, N₂O and CO₂ emissions estimated by this study. WWTPs or other treatment facilities, which have biological treatment processes, emit CH₄, N₂O and
CO2 directly, which were calculated by Eq. 1.1, 1.2 and 1.3, respectively. The CH4, N2O and CO2 emission factors of different biological treatment processes adopted in this study were obtained from the literature, and most were studies on GHG emission factors of existing Chinese WWTPs. On the other hand, some emission factors were adopted from the IPCC 2019 report, laboratory-based studies or other models, because of a lack of studies on emission factors of full-scale wastewater treatment processes. Detailed CH4, N2O and CO2 emission factors from the literature were summarised in Table S1, Table S2 and Table S3, respectively. We obtained the minimum, maximum and average values of emission factors for each biological treatment process. The average values of emission factors were defined as the default emission factors in this study, and they are shown in Table 2. We also list IPCC 2019 emission factors of biological treatment processes in Table 2 for comparison. For those WWTPs or other treatment facilities adopted by the combined enhanced activated sludge treatment technology, their emissions factors are mean of emission factors of specific enhanced activated sludge treatment technologies (i.e., AO, A2O, OD or SBR). CH4 and N2O are converted to the CO2 equivalent by Global Warming Potential (GWP) values for 100 years. The GWP of N2O, CH4 and CO2 are 265, 28 and 1, respectively.

\[
CH4_{bio, i} = EF_{bio, CH4, j} \times AD_{bio, CH4, i} \times 28 \tag{1.1}
\]

\[
N2O_{bio, i} = EF_{bio, N2O, j} \times TN_{in, i} \times 265 \tag{1.2}
\]

\[
CO2_{bio, i} = EF_{bio, CO2, j} \times COD_{removed, i} \tag{1.3}
\]

Where, \( CH4_{bio, i}, N2O_{bio, i} \) and \( CO2_{bio, i} \) refer to CH4, N2O and CO2 emissions (g CO2eq/year) from biological treatment processes in the \( i \) th WWTP. \( EF_{bio, CH4, j} \) (g CH4/kg COD removed or g CH4/kg COD), \( EF_{bio, N2O, j} \) (g N2O/kg TN influent) and \( EF_{bio, CO2, j} \) (g CO2/kg COD removed) are three GHG emission factors of the process \( j \) in the \( i \) th WWTP. \( AD_{bio, CH4, i} \) is activity data of biological CH4 emissions. There are two types of \( AD_{bio, CH4, i} \). When the unit of \( EF_{bio, CH4, j} \) for the process \( j \) is g CH4/kg COD removed, \( AD_{bio, CH4, i} \) is the removed COD per year (kg COD removed/year) in the \( i \)th WWTP. But \( AD_{bio, CH4, i} \) refers to the difference between influent COD mass and COD transferred to sludge if the unit of \( EF_{bio, CH4, j} \) is g CH4/kg COD. In the section of ‘Calculation of COD removed in the form of sludge’, we described how to estimate the COD transferred in the form of sludge.
for each process. $TN_{in,i}$ is the annually influent TN mass (kg TN influent/year) in the $i$th WWTP, and $COD_{removed,i}$ is the annually removed COD (kg COD removed/year) in the $i$th WWTP.

$CH_4$ and $N_2O$ emissions estimated by IPCC 2019. To make a comparison with our study, we also used the method of IPCC 2019 to calculate $CH_4$ and $N_2O$ emissions from biological treatment processes. $CH_4$ and $N_2O$ emission factors for each wastewater treatment process are from IPCC 2019 (Table 2).

\[
CH_4_{IPCC\_bio,i} = EF_{IPCC\_bio,CH4j} \times (COD_{in,i} - SCOD_{in,i}) - R_{CH4,i} \times 28
\]

(1.4)

\[
N2O_{IPCC\_bio,i} = EF_{IPCC\_bio,N2Oj} \times TN_{in,i} \times 265
\]

(1.5)

where, $CH_4_{IPCC\_bio,i}$ and $N2O_{IPCC\_bio,i}$ refer to $CH_4$ and $CO_2$ emissions (g CO2eq/year) from biological treatment processes in the $i$th WWTP. $EF_{IPCC\_bio,CH4j}$ ($CH_4$/kg COD) and $EF_{IPCC\_bio,N2Oj}$ ($N_2O$/kg TN influent) are IPCC 2019 $CH_4$ and $N_2O$ emission factors of the process $j$ in the $i$th WWTP. $COD_{in,i}$ is the annually influent COD mass (kg COD influent/year) in the $i$th WWTP. $SCOD_{in,i}$ (kg COD removed as sludge/year) is the COD removed in the form of sludge in the $i$th WWTP. $R_{CH4,i}$ is amount of $CH_4$ recovered or flared from the $i$th WWTP. This value was regarded as being zero because there are very few $CH_4$ recovered or flared in China. $TN_{in,i}$ is the annually influent TN mass (kg TN influent/year) in the $i$th WWTP.

Table 1. Classification of treatment processes of WWTPs in China. Note: Wastewater treatment technologies of Conventional Activated Sludge (4), Enhanced Activated Sludge Process (5), Biofilm (6), Anaerobic Biological Method (7), Stabilization Pond, Constructed Wetland and Land Treatment (8) all belong to subcategories of biological treatment processes. But for some WWTPs, their subcategories of biological treatment processes were not reported in the original dataset. In this case, their treatment technologies were named as Biological Treatment (10), and their GHG emissions are estimated by emission factors of the technology of activated sludge treatment (4.2 in Table 1), as it is recognized as the most popular wastewater treatment technology around the world.
Calculation of COD removed in the form of sludge.

\[ S_{COD,j} = COD_{removed, i} \times Y_{obs,j} \times 1.42 \]  

(1.6)

\[ COD_{removed, i} = (COD_{in} - COD_{out}) \times V_{wastewater, i} \]  

(1.7)

where, \( S_{COD,j} \) (g COD removed as sludge/year) is the COD removed in the form of sludge in the \( i \)th WWTP, \( COD_{removed, i} \) (g COD/year) is the COD removed of the \( i \)th WWTP, \( Y_{obs,j} \) (g VSS/ g COD) is the observed sludge yield of process \( j \) in the \( i \)th WWTP, 1.42 (g COD/ g VSS) is the conversion factor that determine biomass concentration in terms of COD\(_{in}\). \( COD_{in} \) and \( COD_{out} \) are influent and effluent COD concentration of the \( i \)th WWTP. \( V_{wastewater} \) is the volume of treated wastewater in the \( i \)th WWTP. The coefficient of \( Y_{obs,j} \) (g VSS/ g COD) for each...
The process is from Chen et al.40. Since a membrane bioreactor (MBR) is the combination of an enhanced activated sludge process and a membrane process, its $Y_{obsj}$ was estimated by the average value of observed sludge yield of an enhanced activated sludge process and a biofilm process. Coefficients $Y_{obsj}$ of different treatment processes are shown in Table 3.

Treated wastewater was discharged in one of 10 different pathways. Table 5 shows emission factors of CO$_2$, N$_2$O and CH$_4$ of each discharge pathway. The effluent emission factors of CH$_4$ and N$_2$O were adopted from IPCC 2019, while the CO$_2$ emission factors of the treated effluent were derived from the appendix of IPCC 2019 (IPCC 2019, Volume 5, Chapter 6, Page 59-Page 60)15. The detailed derivation process of CO$_2$ emission factor of effluent discharge refers to Supplementary Information 'CO$_2$ emission factor of effluent discharge'. Emissions from discharge pathways were calculated by Eq. 2.1–2.3:

$$CH_4_{eff,i} = E_{eff,CH_4,i} \cdot COD_{out,i} \cdot 28$$  \hspace{1cm} (2.1)

$$N_2O_{eff,i} = E_{eff,N_2O,i} \cdot TN_{out,i} \cdot 265$$  \hspace{1cm} (2.2)
\[
\text{CO}_2\text{eff}_{i,j} = EF_{\text{eff,CO}_2,j} \times \text{COD}_{\text{out},i}
\]  
(2.3)

where, \(CH_4\) \(_{\text{eff,j}}\), \(N_2O\) \(_{\text{eff,j}}\) and \(CO_2\) \(_{\text{eff,j}}\) are \(CH_4\), \(N_2O\) and \(CO_2\) emissions (g CO2eq/year) from the discharge pathway \(j\) in the \(i\)th WWTP, \(EF_{\text{eff,CH}_4,j}\) (g CH4/g COD effluent), \(EF_{\text{eff,N}_2O,j}\) (g N2O/kg TN effluent) and \(EF_{\text{eff,CO}_2,j}\) (g CO2/kg COD effluent) are effluent emission factors of the discharge pathway \(j\) of the \(i\)th WWTP. COD\(_{\text{out},i}\) (kg COD effluent/year) and TN\(_{\text{out},i}\) (kg TN effluent/year) are annually effluent COD and TN mass of the \(i\)th WWTP.

Off-site emissions from electricity consumption. The calculation of GHG emissions from electricity consumption is shown in Eq. 3.1. Baseline emission factors for regional power grids in China\(^{41-44}\) were used in this study. Only CO2 is considered for emission factors for regional power grids without considering \(N_2O\) and \(CH_4\) due to their small contributions. China’s baseline emission factors for regional power grids are presented in Table 4.

\[
\text{CO}_2\text{ele}_{i,j} = EF_{\text{ele,CO}_2,j} \times \text{Ele}_{\text{con},i}
\]  
(3.1)

where, \(CO_2\) \(_{\text{ele,i,j}}\) is the \(CO_2\) emission from electricity consumption (kg CO2/year). \(EF_{\text{ele,CO}_2,j}\) (kg CO2/kWh) denotes the \(CO_2\) emission factor of province \(j\) of the studied WWTP. \(\text{Ele}_{\text{con},j}\) (kWh/year) refers to the electricity consumption of the \(i\)th WWTP.

Uncertainty analysis. The uncertainty of GHG emissions was mainly caused by emission factors. Since calculation of activity data of each WWTP was based on annual on-site monitored data of the volume of treated
wastewater, influent and effluent concentration of pollutants and electricity consumption, there is no uncertainty for activity data. We analysed GHG emissions uncertainty induced by biological treatment processes and discharge pathways. The uncertainty caused by electricity consumption was not considered, because China’s regional power grid baseline emission factors are based on specific values rather than ranges.

For the emission factors of biological treatment processes, we acquired the minimum, maximum and average emission factors of each technology from the literature. Then, we used the following Eq. 4.1 and 4.2 to calculate the uncertainty of emission factors.

\[
\text{Uncertainty lower bound } U_{lb} = \left( \frac{EF_{\text{min}} - EF_{\text{ave}}}{EF_{\text{ave}}} \right) \times 100\% \quad (4.1)
\]

\[
\text{Uncertainty upper bound } U_{ub} = \left( \frac{EF_{\text{max}} - EF_{\text{ave}}}{EF_{\text{ave}}} \right) \times 100\% \quad (4.2)
\]

Since the CH₄ emission factor was determined by the multiplication of the maximum producing potential (B₀) and the methane correction factor (MCF), its uncertainty was measured by Eq. 4.3. The uncertainty of B₀ (U_{B₀}) is ±30% in IPCC 2019, and the uncertainty of MCF (U_{MCF}) was determined by Eq. 4.1 and 4.2. The uncertainties of N₂O and CO₂ emission factors of discharge pathways were calculated by Eq. 4.1 and 4.2.

\[
U_{\text{CH}_4} = \pm \sqrt{U_{B_0}^2 + U_{\text{MCF}}^2} \quad (4.3)
\]

We applied Monte Carlo simulations to analyse the combined uncertainty of emission factors and activity data. Emission factors of CH₄, N₂O and CO₂ of biological treatment processes and discharge pathways all follow triangular distributions, because ‘upper and lower and a preferred value are provided (IPCC 2006, Volume 1, Chapter 3, Page 22)\(^{15}\) in this study. Random sampling on emission factors was performed 100,000 times, then multiplied by activity data of each GHG in each WWTP, generating 100,000 values for GHG emissions. Finally, uncertainty ranges of 95% confidence intervals of GHG emissions were adopted.

Other causes that may induce uncertainties include ‘Measurement error’, ‘Lack of completeness’ and ‘Misreporting or misclassification’. With regard to the measurement error in a real WWTP, the measured influent and effluent concentration of pollutants and electricity consumption may be incorrect. But this uncertainty is difficult to quantify and control in this study. In terms of lack of completeness, the original data was incomplete for all WWTPs. For instance, data of some indicators was lacking, e.g., volume of treated wastewater, influent, or effluent concentrations of COD. When a WWTP does not have sufficient indicators, the WWTP was removed, and its emissions were not calculated. For the misreporting or misclassification, accurate classification of treatment technologies is the basis for calculating GHG emissions of secondary biological treatment processes, but uncertainties caused by misreporting and/or misclassification of treatment technologies are possible and cannot be easily rectified.

**Data Records**

The dataset of “Greenhouse gas emissions of wastewater treatment plants in China from 2006 to 2019” is made public under Figshare\(^{45}\). There are 400,512 data records in the dataset. These include:

| Discharge Pathway | CH₄ (g CH₄/kg COD effluent) | N₂O (g N₂O/kg TN effluent) | CO₂ (g CO₂/kg COD effluent) |
|-------------------|-----------------------------|----------------------------|-----------------------------|
| 1. Discharge into seas directly | 8.75 | 7.90 | 570.90 |
| 2. Discharge into rivers, lakes, reservoirs etc. directly | 47.50 | 7.90 | 570.90 |
| 3. Enter sewers first, then discharge into rivers, lakes, and reservoirs | 47.50 | 7.90 | 570.90 |
| 4. Enter sewers first, then discharge into seas | 8.75 | 7.90 | 570.90 |
| 5. Enter municipal WWTPs | 0.00 | 0.00 | 0.00 |
| 6. Discharge into sewage irrigated farmlands directly | 0.00 | 8.00 | — |
| 7. Discharge into soil | 0.00 | 8.00 | — |
| 8. Enter other facilities (decentralized wastewater treatment facilities) | 0.00 | 0.00 | 0.00 |
| 9. Centralized industrial WWTPs | 0.00 | 0.00 | 0.00 |
| 10. Other discharge pathways | 27.50 | 7.90 | 570.90 |

**Table 5.** Emission factors of different GHG emissions from discharge pathways. Note: CH₄ and N₂O emission factors of discharge pathways of 5, 8, and 9 are zero, as they belong to the pathway of ‘flowing sewer’, and CH₄ and N₂O emission factors for the discharge pathway of ‘flowing sewer (open or closed)’ are zero in IPCC 2019. We also assumed that there was no CO₂ generation under the pathway of ‘flowing sewer’. Discharge pathway 6 and 7 were regarded as discharge into soil in this study. From IPCC 2019, default CH₄ emission factor of the pathway of discharge into soil was 0 g CH₄/kg COD effluent. We did not consider CO₂ emissions of discharge into soil, because of a lack of data on the CO₂ emission factor of discharge into soil.
In this study, the firm-level GHG emission inventory provides a foundation for the remaining emission inventories. Based on the firm-level GHG emission inventory, annual CH₄, N₂O and CO₂ emission inventories of biological treatment processes, effluent and electricity consumption are presented, and annual total CO₂eq emissions of different technologies from biological treatment processes, electricity consumption and discharge pathways are also quantified.

Figure 3 presents annual CH₄, N₂O and CO₂ emissions from different emission sources and annual treated wastewater from 2006 to 2019. The pie charts in Fig. 4 show the structure of treatment technology in total CO₂eq emissions in 2006, 2010, 2015 and 2019, respectively. Treatment technologies are classified by main categories of processes based on the classification in Table 1. Since the enhanced activated sludge process is the main wastewater treatment technology in China and it includes many sub-categories, the emission structure of sub-categories (i.e., AO, A²O, OD and SBR) of the enhanced activated sludge process is also shown in pie charts.

Technical Validation

Uncertainty analysis. The uncertainty of emission factors. The uncertainty of CH₄, N₂O and CO₂ emission factors of biological treatment technologies is presented in Table 6. For comparison, we also list the uncertainty of CH₄ and N₂O emission factors based on IPCC 2019. The analysis by IPCC 2019 shows higher uncertainty in terms of CH₄ and N₂O emission factors of a majority of biological treatment technologies, due to its less detailed classification of technologies. For instance, different activated sludge technologies in IPCC 2019 possess the same emission factors and uncertainties, because IPCC 2019 classifies all activated sludge processes into one aerobic process category. However, processes of AO, A²O, SBR and OD are different and have different ranges (Table 6). However, on-site emission factors of certain processes are rarely reported in the literature, and we cannot obtain their emission factors based on detailed process classification. For example, we applied a CH₄ emission factor (200 g CH₄/kg COD) of the anaerobic process from IPCC 2019 to four different anaerobic processes (i.e., anaerobic hydrolysis, typical anaerobic reactors, anaerobic biofilter, and other anaerobic biological treatment), due to a lack of their on-site emission factors from references. Therefore, reported uncertainties (−30%,39%) for CH₄ emission factors of the four anaerobic processes are the same. Overall, the uncertainties of GHG emission factors of different biological treatment technologies were
One of the main reasons is that GHG emission factors are strongly affected by different operational parameters (temperature, pH, dissolved oxygen (DO), sludge retention time (SRT), hydraulic retention time (HRT), influent chemical oxygen demand (COD) to total nitrogen ratio (C/N), influent chemical oxygen demand (COD) to total phosphorus ratio (C/P), etc.) of these WWTPs.

Since CH4 and N2O emission factors for the discharge pathway of 'flowing sewer (open or closed)' are zero in IPCC 2019, we assumed that there was no CO2 generation under this flowing condition. We regarded discharge pathways via municipal WWTPs, centralized industrial WWTPs and other facilities (decentralized wastewater treatment facilities) as discharge pathways of 'flowing sewer'. Therefore, we do not report any uncertainty of CH4, N2O and CO2 emission factors of entering municipal WWTPs, industrial WWTPs and other facilities. We considered the discharge pathway of 'other discharge pathways' in this study as 'discharge to aquatic environments (Tier 1)' in IPCC 2019, and its uncertainties of CH4 (−100%, 148%) and N2O emission factors (−90%, 1394%) are the largest compared with other discharge pathways. Because there are very few studies on the CO2 emission factor of the treated effluent, we derived CO2 emission factors of lakes, rivers and reservoirs from the appendix of IPCC 2019 (IPCC 2019, Volume 5, Chapter 6, Page 59-Page 60) and we assumed that pathways of discharging into sea and 'others' also have the same CO2 emission factors. Thus, their CO2 emission factor uncertainties were all the same, with the uncertainty of (−12%, 20%).

Fig. 4 Structure of treatment technology in total CO2eq emissions in 2006, 2010, 2015 and 2019 (in million tons CO2eq). GHG emissions from enhanced activated sludge processes and conventional activated sludge accounted for a large proportion (>80%) in 2010, 2015 and 2019. While the percentage from biological treatment process was very high (58%) in 2006, because for some WWTPs, their subcategories of biological treatment processes were not reported in the original dataset. In this case, their treatment technologies were named as biological treatment, and their GHG emissions were estimated by emission factors of the process of activated sludge treatment in this study.
Table 6. Uncertainty of CH₄, N₂O and CO₂ emission factors of biological treatment technologies. Note: The symbol ‘—’ indicates the uncertainty of CH₄, N₂O or CO₂ emission factor of a biological treatment technology is not existed when the default emission factor of a treatment process is zero.

| Technology                      | IPCC 2019 CH₄ | IPCC 2019 N₂O | IPCC 2019 CO₂ | This study CH₄ | This study N₂O | This study CO₂ |
|---------------------------------|---------------|---------------|---------------|---------------|---------------|---------------|
| Aerobic Biological Treatment    | −95% 202%     | −99% 184%     | −43% 57%      | −83% 158%     | −4% 0%        |
| Activated Sludge                | −95% 202%     | −99% 184%     | −43% 57%      | −83% 158%     | −4% 0%        |
| AO                              | −95% 202%     | −99% 184%     | −68% 351%     | −13% 15%      | −86% 86%      |
| A’O                             | −95% 202%     | −99% 184%     | −93% 324%     | −97% 456%     | −57% 54%      |
| OD                              | −95% 202%     | −99% 184%     | −33% 22%      | −50% 228%     | −71% 49%      |
| SBR                             | −95% 202%     | −99% 184%     | −86% 28%      | −40% 50%      | −82% 82%      |
| AB                              | −95% 202%     | −99% 184%     | −43% 57%      | −83% 158%     | −4% 0%        |
| Biotrilm                        | −99% 184%     | −99% 184%     | −99% 119%     | −120%         |
| Biotrilm                        | −99% 184%     | −99% 184%     | −99% 119%     | −120%         |
| Rotating Biological Contactor   | −99% 184%     | −99% 184%     | −99% 119%     | −120%         |
| Biological Contact Oxidation    | −99% 184%     | −99% 184%     | −99% 119%     | −120%         |
| Anaerobic Biological Treatment  | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Anaerobic Hydrolysis            | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Typical Anaerobic Reactors      | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Anaerobic Biofilter             | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Other Anaerobic Biological      | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Treatment                      | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Stabilization Pond, Constructed | −54% 58%      | −97% 173%     | −54% 58%      | −97% 173%     | −17% 14%      |
| Wetland and Land Treatment      | −47% 52%      | −99% 186%     | −47% 52%      | −99% 186%     | −9% 6%        |
| Stabilization Lagoon            | −95% 202%     | −99% 184%     | −95% 202%     | −99% 184%     | −4% 0%        |
| Anamorphic Lagoon               | −30% 39%      | −30% 39%      | −30% 39%      | −30% 39%      |
| Facultative Lagoon              | −100% 58%     | −99% 184%     | −100% 58%     | −99% 184%     | −4% 0%        |
| Anamorphic Lagoon               | −95% 202%     | −99% 184%     | −95% 202%     | −99% 184%     | −4% 0%        |
| Constructed Wetland             | −77% 73%      | −80% 80%      | −77% 73%      | −80% 80%      | −30% 30%      |
| Subsurface Flow Constructed Wetland | −45% 45% | −79% 79% | −45% 45% | −79% 79% | −30% 30% |
| Surface Flow Constructed Wetland | −85% 81%     | −90% 90%      | −85% 81%      | −90% 90%      | −30% 30%      |
| Land Infiltration               | −39% 53%      | −100% 129%    | −39% 53%      | −100% 129%    | −17% 14%      |
| Biological Treatment            | −95% 202%     | −99% 184%     | −95% 202%     | −99% 184%     | −4% 0%        |

Table 7. Uncertainty of emission factors of different discharge pathways. Note: The symbol ‘—’ indicates the uncertainty of CH₄, N₂O or CO₂ emission factor of a discharge pathway is not existed when the default emission factor of a discharge pathway is zero or is not existed.

| Pathway                             | CH₄ | N₂O | CO₂ |
|-------------------------------------|-----|-----|-----|
| 1 Direct discharge into seas        | −94% 80% | −90% 1394% | −12% 20% |
| 2 Direct discharge into rivers, lakes, reservoirs etc. | −65% 52% | −90% 1394% | −12% 20% |
| 3 Enter sewers first, then discharge into rivers, lakes, and reservoirs | −65% 52% | −90% 1394% | −12% 20% |
| 4 Enter sewers first, then discharge into seas | −94% 80% | −90% 1394% | −12% 20% |
| 5 Enter municipal WWTPs              | —   | —   | —   |
| 6 Direct discharge into sewage irrigated farmland | 100% 116% | —   | —   |
| 7 Discharge into soil                | —   | —   | —   |
| 8 Enter other facilities (decentralized wastewater treatment facilities) | —   | —   | —   |
| 9 Enter centralized industrial WWTPs | —   | —   | —   |
| 10 Other discharge pathways          | −100% 148% | −90% 1394% | −12% 20% |

Combined uncertainty of GHG emissions. The combined uncertainty of GHG emissions of biological treatment processes is presented in Table 8 and Fig. 5(a–c). The shadow areas shown in Fig. 5 indicate the 95% confidence interval of GHG emissions. For comparison, CH₄ and N₂O emissions calculated by emission factors of IPCC 2019 are also shown in Fig. 5(a,b). From 2006 to 2019, the uncertainties of CH₄, N₂O and CO₂ emissions in this study were (−57%, 124%), (−63%, 184%) and (−43%, 38%), respectively. But uncertainties of CH₄ and N₂O emissions calculated by the methodology of IPCC 2019 were (−91%, 189%) and (−99%, 184%).
minimum and maximum CH₄ and N₂O emissions calculated by IPCC 2019 were all outside of the shadow areas in Fig. 5(a,b), reflecting larger uncertainties than in our study.

The combined uncertainty of effluent GHG emissions is presented in Table 9 and Fig. 5(e–g). The overall uncertainties of the effluent N₂O were very high (−33%, 1161%), mainly resulting from high uncertainty of the effluent N₂O emission factor (−100%, 1394%). N₂O emission factors vary substantially between WWTPs, due to different process designs and operational conditions. Effluent CH₄ and CO₂ emission uncertainties were relatively low, with values of (−52%, 29%) and (−9%, 16%), respectively. The uncertainty of total GHG emissions of WWTPs are shown in Fig. 5(h) and Table S4. The uncertainties of total GHG emissions from WWTPs were about (−27%, 97%).

### Comparison with existing estimations

Several studies on CH₄ or N₂O emissions of WWTPs at the national level in China have been reported. In Table S5, we list wastewater GHG estimations in the literature for comparison. In most cases, the current estimation results are not comparable. The use of different system boundaries across studies is one of the main reasons. For instance, CH₄ emissions (76.2 Mt CO₂eq) of wastewater from China's second biennial update report on climate change in 2014 refer to emissions from both industrial and domestic wastewater at the national level and activity data was obtained from the Environmental Statistics Yearbook, while Zhao et al. considered CH₄ emissions (29.2 Mt CO₂eq) from 2019 WWTPs at the firm level in 229 cities in 2014 and the data was from the Urban Drainage Statistics Yearbook. Their results are not comparable, since 2019 WWTPs in Zhao et al.’s study contained mainly prefecture-level municipal WWTPs but excluded county-level and industrial WWTPs in China, and it is not clear how many WWTPs/wastewater treatment facilities are included in China's second biennial update report. Therefore, the activity data and CH₄ emissions were not comparable in these two studies, although they all used IPCC 2006 method for their inventories. In our paper, on-site CH₄ emissions from 4455 WWTPs and 718 other treatment facilities were estimated to be 2.55 Mt CO₂eq in 2014, which were about one tenth of Zhao et al.’s results. This discrepancy was caused by using different system boundaries and the use of different emission factors.

Most studies used emission factors from the IPCC, but even CH₄ emission factors from IPCC 2006 and IPCC 2019 are quite different. The default methane correct factor (MCF) in IPCC 2019 is 0.165, while this value is 0.3 in IPCC 2006 for overloaded WWTPs, and it may differ by one order of magnitude for CH₄ emissions. Our uncertainty analysis shows that CH₄ emissions calculated by IPCC 2019 are about 20%–62% larger than our results. This discrepancy was caused by using different system boundaries and the use of different emission factors.

### Table 8. The combined uncertainty of GHG emissions from biological treatment.

| Year | CH₄ emissions | IPCC 2019 | N₂O emissions | IPCC 2019 | CO₂ emissions | IPCC 2019 |
|------|---------------|-----------|---------------|-----------|--------------|-----------|
| 2006 | −48% 98%      | −75% 147% | −63% 131%     | −99% 184% | −18% 16%     |
| 2007 | −52% 111%     | −83% 166% | −60% 126%     | −99% 184% | −22% 21%     |
| 2008 | −57% 124%     | −91% 189% | −57% 118%     | −99% 184% | −22% 21%     |
| 2009 | −52% 113%     | −85% 176% | −55% 116%     | −99% 184% | −24% 24%     |
| 2010 | −29% 59%      | −59% 113% | −37% 121%     | −99% 184% | −41% 38%     |
| 2011 | −30% 64%      | −60% 115% | −43% 154%     | −99% 184% | −41% 37%     |
| 2012 | −29% 59%      | −57% 108% | −43% 146%     | −99% 184% | −41% 37%     |
| 2013 | −30% 62%      | −60% 114% | −43% 149%     | −99% 184% | −41% 37%     |
| 2014 | −30% 62%      | −58% 110% | −44% 154%     | −99% 184% | −39% 36%     |
| 2015 | −30% 65%      | −59% 113% | −43% 157%     | −99% 184% | −41% 37%     |
| 2016 | −35% 88%      | −72% 143% | −42% 164%     | −99% 184% | −40% 37%     |
| 2017 | −39% 103%     | −76% 155% | −45% 169%     | −99% 184% | −41% 36%     |
| 2018 | −37% 96%      | −73% 146% | −44% 180%     | −99% 184% | −42% 35%     |
| 2019 | −39% 104%     | −75% 151% | −44% 184%     | −99% 184% | −43% 36%     |
COD (or BOD) mass minus COD (or BOD) removed in the form of sludge means that organic components transferred to sludge do not generate direct CH$_4$, but only the remaining organic matter in the wastewater has potential to emit CH$_4$. Therefore, the unit (kg CH$_4$/kg BOD or kg CH$_4$/kg COD) of CH$_4$ emission factor in IPCC indicates CH$_4$ emissions per unit remaining organic mass in the influent after considering COD (or BOD)
transferred to the sludge, rather than CH4 emissions per unit influent COD (or influent BOD) or per unit COD (or BOD) removed. In addition, organic matter removed in the form of sludge was assumed as being zero for all treatment technologies. The reasons for the incorrect assumption may be the lack of data on sludge generation, and the method to estimate organic components removed in the form of sludge is not mentioned in IPCC 2006, or the lack of background on wastewater treatment. The assumption may overestimate CH4 emissions as most aerobic biological treatment technologies generate sludge during wastewater treatment. However, IPCC 2019 updated the method to account CH4 emissions based on IPCC 2006, especially providing equations and detailed information to estimate COD (or BOD) transferred to sludge, which provides guidance for accurate CH4 accounting.

Limitations. We have four main limitations in this study. (1) A WWTP may have one or more wastewater treatment streams, and for each treatment stream, it may contain primary, secondary or tertiary treatment processes, while for each process (normally for a secondary treatment process), it has multiple treatment technologies. But to simplify GHG emissions estimation of biological treatment technologies of the secondary treatment process of a WWTP, the decision tree (Fig. 2) was applied to determine the main category of treatment technology and its corresponding emission factors, especially when a WWTP has several secondary treatment technologies. (2) Our emission factors of different biological treatment technologies were not based on the monitoring of each wastewater treatment plant. But we used emission factors in line with Chinese conditions. The emission factors were acquired from different references, such as on-site monitoring of specific biological technologies or modelling estimations in the literature, which was based on case studies of WWTPs in China. However, emission factors of some biological technologies, such as CH4 and CO2 emission factors of anaerobic technologies and constructed wetlands, were missing for China, thus we used IPCC emission factors for these technologies instead. On the other hand, given that emission factors of a specific biological treatment technology are greatly affected by operational conditions, different WWTPs with the same biological technology may have different emission factors. Therefore, GHG emission factors of a biological technology obtained from references are not representative for real emission factors of all WWTPs with the same technology. (3) GHG emissions from industrial WWTPs are not available and thus not included in our study although being important GHG emission sources of wastewater treatment systems. For instance, Xing et al. reported that CH4 emissions from on-site industrial wastewater treatment were always higher than that of domestic wastewater treatment between 2003 and 2008 in China. CH4 emissions from industrial and domestic wastewater treatment were 0.95 Mt and 0.91 Mt respectively in 2008. (4) Anthropogenic CO2 emissions (or fossil CO2 emissions) from biological treatment processes and discharge pathways are of main concern compared with biogenic CO2 emissions, but we did not calculate fossil CO2 emissions separately, because the CO2 emission factors available in the literature are only reported as total CO2, rather than separate fossil and biogenic CO2.

Table 9. The combined uncertainty of GHG emissions from effluent.

|       | CH4 emissions | N2O emissions | CO2 emissions |
|-------|---------------|---------------|---------------|
| 2006  | −52% 29%      | −33% 1160%    | −9% 16%       |
| 2007  | −52% 29%      | −33% 1151%    | −9% 16%       |
| 2008  | −52% 29%      | −33% 1148%    | −9% 16%       |
| 2009  | −52% 29%      | −33% 1152%    | −9% 16%       |
| 2010  | −52% 29%      | −33% 1148%    | −9% 16%       |
| 2011  | −52% 28%      | −33% 1149%    | −9% 16%       |
| 2012  | −52% 28%      | −33% 1149%    | −9% 16%       |
| 2013  | −52% 28%      | −33% 1159%    | −9% 16%       |
| 2014  | −52% 28%      | −33% 1150%    | −9% 16%       |
| 2015  | −52% 28%      | −33% 1158%    | −9% 16%       |
| 2016  | −52% 28%      | −33% 1145%    | −9% 16%       |
| 2017  | −52% 28%      | −33% 1159%    | −9% 16%       |
| 2018  | −52% 28%      | −33% 1160%    | −9% 16%       |
| 2019  | −52% 28%      | −33% 1161%    | −9% 16%       |

The scripts used to calculate firm level GHG emissions of wastewater treatment facilities are available in the Zenodo repository: https://doi.org/10.5281/zenodo.60528156.

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Competing interests

The authors declare no competing interests.

Additional information

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