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Original Research

Methane and nitrous oxide emissions from municipal wastewater treatment plants in China: A plant-level and technology-specific study

Haiyan Li [a,*], Liangfang You [a], He Du [a], Bowen Yu [a], Lu Lu [a], Bo Zheng [b,**], Qiang Zhang [c], Kebin He [d], Nanqi Ren [a]

[a] School of Civil and Environmental Engineering, Harbin Institute of Technology, Shenzhen, 518055, China
[b] Shenzhen Key Laboratory of Ecological Remediation and Carbon Sequestration, Institute of Environment and Ecology, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen, 518055, China
[c] Ministry of Education Key Laboratory for Earth System Modeling, Department of Earth System Science, Tsinghua University, Beijing, 100084, China
[d] State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, 100084, China

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ABSTRACT

Wastewater treatment is an important source of greenhouse gases (GHGs). Yet large uncertainties remain in the quantification of GHG emissions from municipal wastewater treatment plants (MWWTPs) in China. A high-resolution and technology-specific emission inventory is still lacking to support mitigation strategies of MWWTPs. Here we develop a plant-level and technology-based MWWTP emission inventory for China covering 8703 plants and 19 treatment technology categories by compiling and harmonizing the most up-to-date facility-level databases. China's methane (CH_4) and nitrous oxide (N_2O) emissions from MWWTPs in 2020 are estimated to be 150.6 Gg and 22.0 Gg, respectively, with the uncertainty range of −30% to 37% and −30% to 26% at 95% confidence interval. We find an emission inequality across cities, with the richest cities emitting two times more CH_4 and N_2O per capita from municipal wastewater treatment than the poorest cities. The emitted CH_4 and N_2O are dominated by Anaerobic/Anoxic/Oxic-, Sequencing Batch Reactor-, Oxidation Ditch-, and Anoxic/Oxic-based MWWTPs of less than 20 years old. Considering the relatively young age structure of China's MWWTPs, the committed emissions highlight the importance of reducing on-site GHG emissions by optimization of operating conditions and innovation management. The emission differences among our estimates, previous studies, and the Intergovernmental Panel on Climate Change guidelines are largely attributed to the uncertainties in emission factors, implying the urgent need for more plant-integrated measurements to improve the accuracy of emission accounting.

1. Introduction

Municipal wastewater treatment plants (MWWTPs) are key infrastructures to alleviate water pollution but are also an important source of greenhouse gases (GHGs), which contribute to global warming [1–3]. Considerable amounts of the potent GHG methane (CH_4) and nitrous oxide (N_2O) can be emitted during wastewater treatment by anaerobic digestion and nitrification/denitrification processes, respectively [4–6]. Globally, around 12% of anthropogenic CH_4 and 4% of anthropogenic N_2O are generated by wastewater treatment and discharge at present (Emissions Database for Global Atmospheric Research (EDGAR) v6.0, https://edgar.jrc.ec.europa.eu/). Many countries have committed to achieving carbon neutrality by around the middle of the century. However, detailed and accurate information on GHG emission accounting for MWWTPs remains lacking, which impedes the design of effective mitigation measures.

China, the world's largest carbon emitter at present, has witnessed a rapid increase in the number and treatment capacity of MWWTPs during the last decade [7,8]. However, there remains a lack of consistent and accurate understanding regarding the extent of GHG emissions released from MWWTPs. For example, the estimation of CH_4 emissions from MWWTPs in China spans two orders

* Corresponding author.
** Corresponding author.
E-mail addresses: lihaiyan2021@hit.edu.cn (H. Li), bozheng@sz.tsinghua.edu.cn (B. Zheng).

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of magnitude from 6.2 to 1271.5 Gg [9–14], contributing 0.01–2.0% to China's anthropogenic CH_4 emissions according to EDGAR v6.0. To date, these reported emission estimates have been built at coarse spatial resolutions (e.g., country and province) where default emission factors have been universally applied. However, GHG emissions from wastewater treatment highly depend on the specific treatment technology of a MWWTP [15–18]. A lack of plant- and technology-based inventories of MWWTPs complicates the assignment of appropriate emission factors. Moreover, the coarse spatial resolution estimates hamper the development of spatially explicit emission maps — a fundamental input for atmospheric inversion modeling [19]. Recent studies, such as Wang et al. (2022) [20] and Du et al. (2023) [21], improved the GHG emission accounting from China's MWWTPs based on plant-level information, while the spatial distribution and detailed characteristics of CH_4 and N_2O emissions were not reported yet.

Here we develop a plant-level, technology-based MWWTPs emission inventory for China by compiling and harmonizing the most up-to-date facility-level databases. The CH_4 and N_2O emissions from biological treatment processes are estimated through a bottom-up approach and are located on a map according to the geographical location of each MWWTP. Additional information on treatment capacity and operation year is collected and summarized to characterize emission distribution patterns. The unprecedented details of emission features can imply mitigation challenges and opportunities, and the plant-level inventory can accurately support atmospheric inverse modeling. We compare our emission results with previous studies which are primarily based on the 2006 Intergovernmental Panel on Climate Change guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines). To understand the disagreeing parts with previous estimates, we also use the 2006 IPCC Guidelines and the 2019 refinement to estimate emissions at the country level. Our study aims to develop a high-resolution emission inventory for China's MWWTPs which represents our current best understanding.

2. Materials and methods

2.1. Development of plant-level database

We developed a plant-level database for MWWTPs in the Chinese mainland (excluding Hong Kong, Macao, and Taiwan) in 2020 by integrating three different plant-level datasets (Fig. 1). We started from the List of Centralized Sewage Treatment Facilities in China (DataSet1), which is maintained by the Ministry of Ecology and Environment (MEE) in China. This source provides essential information, including each plant's name, address, wastewater treatment capacity, and classification for municipal or industrial wastewater treatment. Subsequently, 8703 MWWTPs were identified in operation from a total of 10829 plants in this database. The information on wastewater treatment technology and the start year of operation were derived from another two datasets by matching plant name and physical address, including the List of National Municipal Sewage Treatment Facilities (DataSet2) from the MEE and the Directory of National Municipal Sewage Treatment Plants (DataSet3; Beijing Huahai Xindian Information Consulting Co., Ltd.). We further collected data from each facility's website and reports to supplement missing information where needed. The integration of the three datasets above (DataSet1, DataSet2, and DataSet3) provides complete plant-level information for most of China's MWWTPs, including facility name, physical address, treatment capacity, treatment technology, and start year of operation (Fig. 1). However, some small plants, which account for 8% of total wastewater treatment capacity, lack technology information. Notely, 70% of those small MWWTPs without technology information have a treatment capacity smaller than 2000 metric tons of wastewater per day.

Based on the newly developed plant-level database for MWWTPs, we estimated annual on-site emissions of CH_4 and N_2O from each of the 8703 MWWTPs in 2020 using equation (1):

$$E_{i,s} = A_{i,s} \times EF_{j,s} \tag{1}$$

where i represents a MWWTP; j represents wastewater treatment technology used by plant i; s represents emission species (i.e., CH_4 and N_2O); E represents annual on-site emissions (metric ton CH_4 or N_2O); A represents activity rate (metric ton), which is the annual removal of chemical oxygen demand (COD) for CH_4 estimation and total nitrogen (TN) removal for N_2O estimation; and EF represents emission factor (kg CH_4 per kg COD removed or kg N_2O per kg TN removed), depending on treatment technology j. The recovered CH_4 and N_2O emissions are regarded as being zero, considering the actual situation in China.

To map emissions at geological locations, we inferred the latitude and longitude coordinates of each MWWTP from the Baidu Map Platform [22] based on their physical addresses. The information on the physical address of each MWWTP contains province, city, county, town, and street number. The derived latitudes and longitudes were used to identify the administrative district where they are located and to cross-check the physical address. Discrepancies within the records highlighted the potential for erroneous locations, which were fixed through further inspection. To ensure the accuracy of locations of large MWWTPs, we visually checked the geolocations of MWWTPs with a treatment capacity larger than 200 thousand metric tons of wastewater per day. These plants comprise less than 2.3% of the number of MWWTPs but account for 28.2% of the total capacity.

2.2. Activity rates

We derived $A_{i,s}$ by combining our newly developed plant-level database with national and provincial statistical data based on several assumptions. The plant-level $A_{i,s}$ were estimated using equation (2):

$$A_{i,s} = A_{p,s} \times \frac{C_{i,p}}{\sum_i C_{i,p}} \tag{2}$$

where p represents the province where MWWTP i is located; and C represents wastewater treatment capacity (metric ton wastewater per day). In equation (2), the amount of pollutant removal by a MWWTP ($A_{i,s}$) is assumed proportional to its wastewater treatment capacity ($C_{i,p}$) within a province p. This assumption is confirmed by a good correlation between actual pollutant removal and wastewater treatment capacity at plant scales based on data from Zhejiang and Guangdong provinces (Fig. S1, Text S1).

The provincial activity rates $A_{p,s}$ were estimated based on equations (3) and (4):

$$A_{p,s} = A_s \times \frac{\overline{A}_{p,s}}{\sum_p \overline{A}_{p,s}} \tag{3}$$

$$A_s = P_s \times \frac{W_{\text{municipal}}}{W_{\text{municipal}} + W_{\text{industrial}}} \tag{4}$$

where A_s represents China's total activity rates in 2020; $\overline{A}_{p,s}$ represents the activity rates of province p averaged between 2011 and 2015 while data are not available after 2016; P_s represents the total

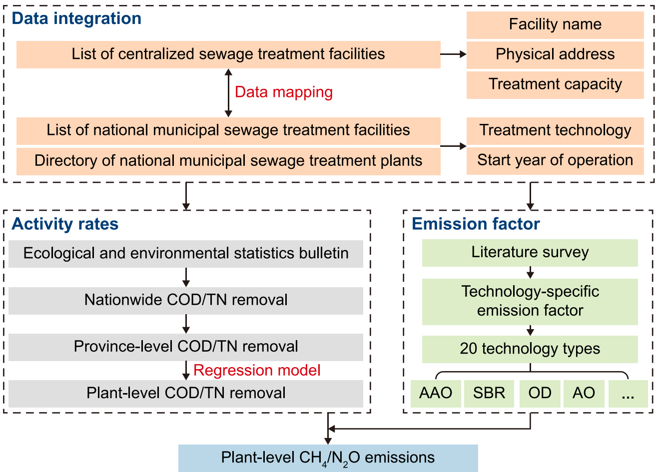

Fig. 1. Framework of the emission model in this study.

amount of pollutant removal by all centralized wastewater treatment plants in 2020; and W represents the amount of total treated wastewater in China. In equation (4), the amount of pollutant removal by municipal wastewater treatment plants (A_s) is assumed to be proportional to the amount of treated municipal wastewater ($W_{\text{municipal}}$).

The input data required by equations (2)–(4) are derived from several statistical yearbooks and reports. P_s, $W_{\text{municipal}}$, and $W_{\text{industrial}}$ are derived from Section 5.2.1 in the Annual Statistic Report on Ecology and Environment released by MEE (https://www.mee.gov.cn/hjzl/sthjzk/sthjtjnb/). The fraction of $W_{\text{municipal}}/W_{\text{municipal}} + W_{\text{industrial}}$ is estimated as 88.0% in 2020. $\overline{A}_{p,s}$ comes from the China Environment Yearbook. The sharing percentages of each province in the national total activity rates ($\overline{A}_{p,s}/\sum_{p}\overline{A}_{p,s}$) remain relatively stable between 2011 and 2015.

2.3. Emission factors

Emission factors ($EF_{j,s}$) are determined according to the biological treatment technology used in each MWWTP. We classified the MWWTPs in our plant-level database into 19 technology categories according to their biological treatment processes (Table 1). Each technology category was assigned an average *EF* derived from field measurement data, which were based on a thorough literature review of 48 peer-reviewed papers. For those small MWWTPs where information on treatment technology was lacking, we utilized emission factors that were averaged over the most commonly used technologies in China. For *EF* data selection and processing, we accorded high priority to the measurement of plant-integrated emission factors because it can reliably represent overall emissions from MWWTPs compared to reactor-scale measurements (Text S2).

2.4. Comparison with previous estimates

To evaluate our emission results, we compared our estimates with previous studies that calculated emissions from MWWTPs in the Chinese mainland [9–14,20,23]. Although the years for emission estimation diverged among studies (all after 2010), the inconsistency, especially the large discrepancy shown in the data comparison, still helped investigate major differences in our estimates and previous studies. Activity rates and emission factors used in different studies were collected and compared to reveal the sources of uncertainty in input data (Tables S3–S4). Moreover, we noted that most previous studies have used methods based on the 2006 IPCC Guidelines [9–14,23]. To dig deep into the methodology differences, we also used the 2006 IPCC Guidelines (IPCC 2006) and the 2019 refinement (IPCC 2019) to estimate CH_4 and N_2O emissions from China's MWWTPs in 2020 independently (Text S3; Tables S1–S2), which helped us understand the differences between the IPCC-based methods and our plant- and technology-based methods developed in this study.

2.5. Uncertainty analysis

To assess the uncertainties of emission estimates, Monte Carlo analysis was performed by estimating the 95% confidence interval of CH_4 and N_2O emissions. We collected uncertainty information on activity data and emission factors that can be described as a probability density function and built a Monte Carlo framework. The plant-level activity rates were assumed to follow a normally distributed pattern with a coefficient of variations (CV) of 70% for COD removal and 100% for TN removal, according to data sources. The emission factors were assumed to follow triangular

Table 1
Emission factors (EF) of different biological treatment technologies in this study.

No.	Biological treatment technology	EF(CH_4) (kg CH_4 per kg COD removed)	References	EF(N_2O) (kg N_2O per kg TN removed)	References
1	AAO	0.0091	[29]	0.0081	[37]
2	Reverse AAO	0.0091	Same as No. 1	0.0081	Same as No. 1
3	AO	0.0138	[38–40]	0.0209	[38–40]
4	SBR	0.0098	[31]	0.0196	[31]
5	OD	0.0094	[15,24,25,37,41]	0.0111	[37]
6	Membrane bio-reactor	0.0027	[37]	0.0141	[37]
7	Activated sludge	0.0123	[31,42]	0.0178	[31,42]
8	Biological aerated filter	0.0029	Same as No. 12	0.0102	Same as No. 12
9	Rotating biological contactor	0.0029	Same as No. 12	0.0102	Same as No. 12
10	Biofilter	0.0029	Same as No. 12	0.0102	Same as No. 12
11	Biological contact oxidation	0.0029	Same as No. 12	0.0102	Same as No. 12
12	Biofilm	0.0029	[37]	0.0102	[37]
13	Aerobic biological treatment	0.0123	Same as No. 7	0.0178	Same as No. 7
14	Anaerobic hydrolysis	0.2	IPCC 2019 [43]	0	IPCC 2019 [43]
15	Anaerobic biological treatment	0.2	IPCC 2019 [43]	0	IPCC 2019 [43]
16	Biological treatment	0.0095	The average of No. 1–7	0.0142	The average of No. 1–7
17	Stabilization pond	0.0571	Same as No. 18	0.0065	Same as No. 18
18	Constructed wetland	0.0571	[37]	0.0065	[37]
19	Others	0.0095	The average of No. 1–7	0.0142	The average of No. 1–7
20	Unrecognized	0.0095	The average of No. 1–7	0.0142	The average of No. 1–7

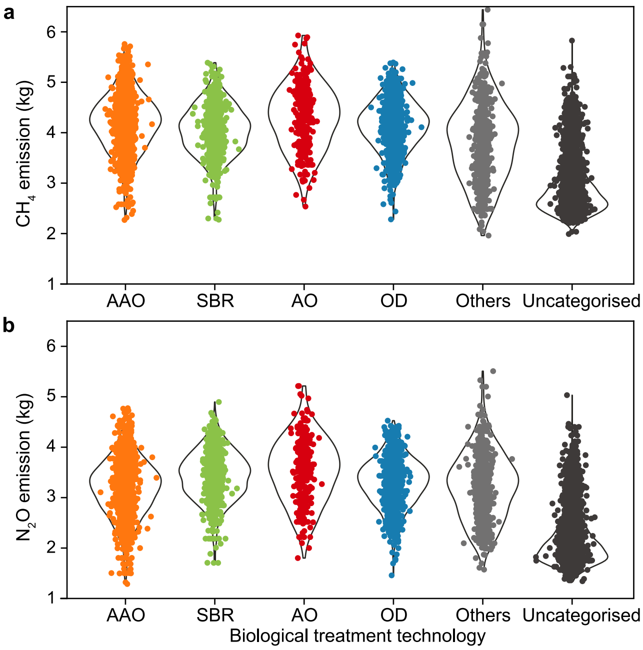

Fig. 2. Plant-level and technology-specific CH_4 (**a**) and N_2O (**b**) emissions from MWWTPs in China, with each dot representing a wastewater treatment facility.

distributions for technologies with mean values and ranges. For other technologies, 100% deviations were applied as the lower and upper bounds, considering the large variations of emission factors. 100000 trials were conducted to estimate the uncertainty range at a 95% confidence interval.

3. Results

3.1. Magnitude and distribution of emissions from MWWTPs

The emissions from MWWTPs in China are estimated to be 150.6 Gg CH_4 and 22.0 Gg N_2O in 2020, over 70% of which are dominated by four widely used wastewater treatment technologies, namely anaerobic/anoxic/oxic (AAO), sequencing batch reactor (SBR), anoxic/oxic (AO), and oxidation ditch (OD) (Fig. 2). Specifically, the AAO-, OD-, SBR-, and AO-based MWWTPs account for 39.1%, 18.8%, 11.2%, and 10.6%, of CH_4 emissions, respectively, and account for 27.3%, 18.2%, 17.8%, and 12.7% of N_2O emissions, respectively. The GHG emissions of different treatment processes can vary widely due to different operational characteristics, site-specific influent concentrations, control parameters, and measurement methods. Previous studies show that the CH_4 and N_2O emission factors of different treatment technologies vary widely by orders of magnitude, and generally, SBR has higher emission intensities than AAO [15–18,24–26]. The CH_4 emission factor of AAO applied in this study is comparable to those of OD, SBR, and AO, while its N_2O emission factor is lower than OD, SBR, and AO. That explains why MWWTPs using the AAO technology generate the largest share of CH_4 emissions in China but have a relatively smaller contribution to N_2O emissions.

Fig. S2 presents the geographical distribution of the 8703 MWWTPs in China, their emissions in 2020 (dot size), and wastewater treatment technology (dot color). MWWTPs and their emissions are unevenly distributed in space and are mainly located in East China, where most of China's population and Gross Domestic Product (GDP) are concentrated. The emissions from all China's MWWTPs vary by a wide data range across orders of magnitude, with the top 1000 MWWTPs accounting for approximately two-thirds of total CH_4 and N_2O emissions. These emission hotspots are mostly concentrated in densely populated cities. The plant hotspots of CH_4 emissions mainly use the treatment technology of AAO (pink dots in Fig. S2a and pink part in Fig. S3a), while the hotspots of N_2O emissions come from all the four widely used treatment technologies, AAO, SBR (green dots in Fig. S2b and green part in Fig. S3b), OD (blue dots in Fig. S2b and blue part in Fig. S3b), and AO (red dots in Fig. S2b and red part in Fig. S3b).

3.2. Socioeconomic factors driving emission distribution

Emission totals of each city are integrated to investigate the socioeconomic factors that affect emission distributions. MWWTP emissions vary widely by city, ranging from 0.5 to 8602.0 metric tons for CH_4 and from 0.08 to 1020.0 metric tons for N_2O. The top three cities with the largest emissions from MWWTPs are Shanghai, Beijing, and Tianjin (Fig. S4), which account for 11.0% of CH_4 emissions and 12.1% of N_2O emissions, surpassing their proportions in China's population (4.3%) and GDP (8.8%). The other top 20 cities of MWWTP emissions are also shown in Fig. S4. These biggest emission cities produce the most municipal wastewater due to their large population and economic production, and large amounts of COD and TN are removed by MWWTPs in these cities (Fig. S4).

As shown in Fig. 3, we further examined the variations in emissions from MWWTPs among cities on a per capita basis, with the cumulative emissions on the *y*-axis and the cumulative population on the *x*-axis ranked by GDP per capita of different cities. Cities with higher GDP per capita tend to emit more CH_4 and N_2O from MWWTPs than cities with lower GDP per capita for the same population. The most economically influential cities, where 20.0% of China's population resides, account for 34.3% and 30.7% of CH_4 and N_2O emissions from MWWTPs in 2020, respectively (Fig. 3). In contrast, in the cities with the lowest GDP per capita, 20.0% of China's population only accounts for 10.7% and 11.6% of CH_4 and N_2O emissions, respectively, in 2020 (Fig. 3). The contrast of city emission distributions indicates an emission inequality across cities, i.e, people living in the richest cities emit two times more CH_4 and N_2O per capita than those living in the poorest cities, probably due to a two-time larger amount of wastewater and pollutant produced. Such a high correlation between wastewater production and socioeconomic growth has been reported globally [27,28]. The emission inequality among cities indicates that cities of different development levels should formulate differentiated emission mitigation policies for wastewater treatment. Besides, we observed a significant difference in technology preferences for wastewater treatment among cities, with AAO systems more widely used in cities with higher GDP per capita.

3.3. MWWTPs emissions by years of service

The CH_4 and N_2O emissions in 2020 are mostly generated by MWWTPs with years of service less than 20 years (i.e., built after 2000) (Fig. 4), especially within 10–20 years (i.e., built between 2000 and 2010), while fewer emissions come from older or newer MWWTPs. Such an emission distribution pattern reflects the development trend of MWWTPs in China over the last two decades. In 2000, 66.7% of China's wastewater was discharged into the environment untreated, while this percentage declined to 17.7% in 2010 (Ministry of Housing and Urban-Rural Construction of the People's Republic of China, https://www.mohurd.gov.cn/) due to the large-scale construction of MWWTPs, which increased wastewater treatment capacity by 383.6% during the ten years [29]. The construction of MWWTPs continued but slowed down after 2010,

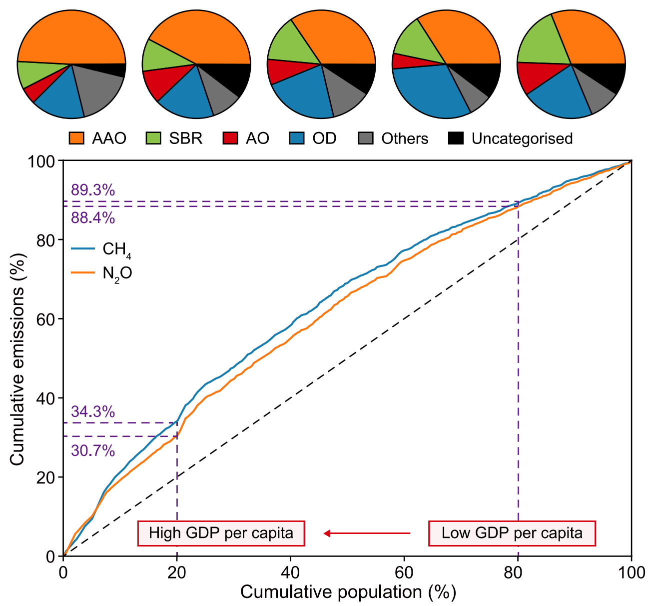

Fig. 3. Variations of cumulative CH_4 and N_2O emissions with the cumulative population of different cities. Cities are ranked from left to right based on the descending order of GDP per capita. Pie charts on the top panel show the contribution of different technologies to wastewater treatment capacity across cities and populations.

further increasing wastewater treatment capacity by 84.6% from 2010 to 2020 and reducing the percentage of untreated wastewater to 2% during the same period [30]. The slower growth during 2010–2020 explains why MWWTPs built after 2010 contribute to a lower share, amounting to less than half, of emissions than MWWTPs built between 2000 and 2010.

We further explored the MWWTP emission distributions by treatment technology and age group (Fig. S5). The removal of COD and TN is mostly dominated by AAO-based MWWTPs less than 20 years old, followed by OD- and SBR-based MWWTPs in the same age group (Figs. S5a and b). The CH_4 emission distribution patterns across treatment technologies and age groups broadly agree with the COD removal (Fig. S5c), while the distributions of N_2O emissions differ from those of TN removal (Fig. S5d). Unlike the TN removal, AAO-, SBR-, OD-, and AO-based MWWTPs of less than 20 years old all contributed significantly to N_2O emissions. This is mainly due to the lower plant-integrated N_2O emission factors of AAO processes than the other treatment technologies used in this study (Table 1). Previous studies using reactor-scale measurements have also observed higher N_2O emission rates of SBR systems than AAO systems [15–18,25,26].

Since the life expectancy of MWWTPs is typically 40–50 years on average, the relatively young age structure of China's MWWTPs suggests that most of China's MWWTPs could continue to operate and generate CH_4 and N_2O emissions in the coming 20–30 years. Those operational MWWTPs in 2020, even those emission hotspots, are not expected to retire shortly. Therefore, any emission mitigation measures must be designed considering the committed emissions from the current MWWTPs in China. To reduce the climate impacts of MWWTPs, we may need to pay attention to the measures that can reduce on-site emissions by optimizing operating conditions and introducing innovative management.

4. Discussion

4.1. Comparison with previous studies

Our estimates of CH_4 (150.6 Gg; uncertainty range of −30% to 37% at a 95% confidence interval) and N_2O (22.0 Gg; uncertainty range of −30% to 26% at a 95% confidence interval) emissions from MWWTPs in China in 2020 are consistent with the latest study of Wang et al. (2022) [20], which also used plant-level information and estimated emissions of 69.1 Gg CH_4 and 17.8 Gg N_2O for 2019. We give a larger estimate of CH_4 emissions because the plant-integrated CH_4 emission factors of the most commonly used technologies, AAO, SBR, OD, and AO, derived from field measurement in this study are 2.1–18.7 times the emission factors used by Wang et al. (2022) [20], coming from reactor-scale measurements. Previous studies also suggest that plant-integrated emission factors tend to be larger than those aggregated from reactor-scale measurements [31], probably because plant-integrated emission factors are more appropriate to represent total emissions from a wastewater plant [32].

Other previous studies show a large spread of annual CH_4 emission estimates for MWWTPs in China, which spans from 6.2 to 1271.5 Gg for the years before 2016 (Fig. 5). While our results lie within this range, the difference of two orders of magnitude shown here suggests potential large uncertainties in our current understanding of MWWTPs emissions. Comparisons across different studies (Table S3) suggest that the large discrepancies are mainly due to the usage of different emission factors, while activity rates are broadly consistent based on statistical data. The studies that provide lower emission estimates mostly use emission factors derived from reactor-scale measurements, such as Cai et al. (2015) [10], Wang et al. (2022) [20], Guo et al. (2019) [23], and Yan et al. (2018) [33]. The studies that give higher estimates of emissions tend to use emission factors based on the 2006 IPCC Guidelines [11,12,14,34]. The IPCC approaches have been optimized and combined with local data if available in these studies. However, the studies based on the 2006 IPCC guidelines still tend to give larger emission estimates than those based on field-measured emission factors. In our study, we conducted a thorough literature review to obtain plant-integrated emission factors, which, to our knowledge, could reflect the current best understanding of MWWTP emission levels. However, the measurement of plant-integrated emission factors is still insufficient at present to constrain uncertainties in emission inventories, which deserves more attention in the future.

Few studies have estimated N_2O emissions from MWWTPs in China, which provides a large spread between 1.3 and 93.5 Gg (Fig. 5). Our emission results fall within this wide data range. Like CH_4, the substantial differences in the estimates of N_2O emissions reflect large uncertainties in current emission inventories, mainly attributed to a lack of reliable N_2O emission factors (Table S4). Our study here adopts the latest field measurement of N_2O emission factors to build our emission inventory, aiming to improve emission estimates as much as possible, which could benefit from more measurement data if available.

4.2. Comparison with IPCC methods

Our emission approach differs from the IPCC methods in estimating activity data and emission factors. The IPCC methods multiply population by per capita organic matter generation and consumption to roughly represent activity data. For example, population and country-specific per capita biochemical oxygen demand (BOD)/COD are combined to calculate BOD/COD removal for CH_4 emission estimation. For N_2O, the IPCC 2019 refinement method estimates activity data by combining the amount of wastewater plant-served population with per capita protein consumption. Our study uses the amount of COD and TN removal by MWWTPs derived from actual statistics as activity data, representing more direct measures of the activities related to CH_4 and N_2O emissions during wastewater treatment.

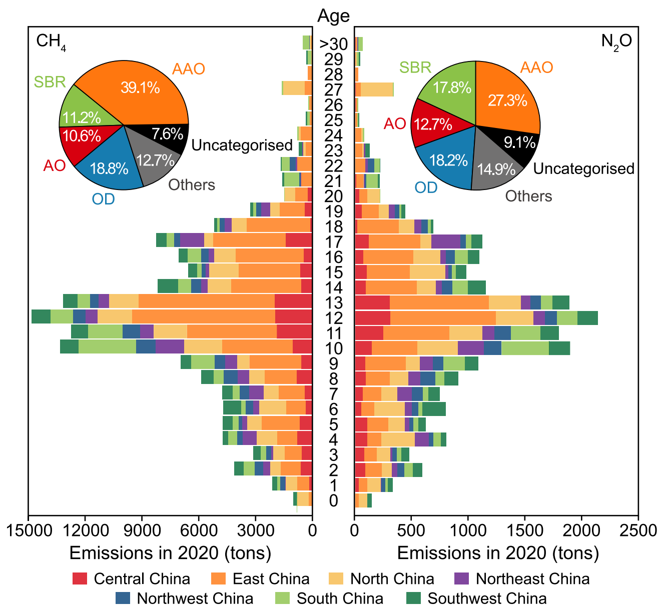

Fig. 4. Age structure of CH_4 and N_2O emissions from MWWTPs in China. Bars indicate the emission distribution from different regions for each age group. Note: 0 years old means the WWTP began operating from 2020 in this study. Pie charts show the contribution of different biological treatment technologies to total CH_4 and N_2O emissions.

The emission factors recommended by the IPCC guidelines are mostly derived from expert judgment based on a literature review, failing to distinguish different wastewater treatment technologies. CH_4 emission factors are calculated as the product of the default maximum CH_4-producing capacity and country-specific methane correction factor, both of which do not distinguish treatment technologies. For N_2O, the 2006 IPCC Guidelines estimate emissions based on population and N_2O emissions per capita, and the IPCC 2019 refinement method further recommended emission factors based on full-scale plant measurement. Distinct from the IPCC methods, our study collects technology-specific emission factors, which are derived from plant-integrated measurement techniques, and combines them with our newly developed plant-level database for MWWTPs to depict emission characteristics in more detail.

Due to the methodological differences described above, our emission results are largely different from the estimation based on the IPCC methods (Fig. 5). For CH_4, the total COD removal from the Annual Statistic Report on Ecology and Environment released by MEE (https://www.mee.gov.cn/hjzl/sthjzk/sthjtjnb/), used as activity data in our method, is 62.9% larger than that estimated by the IPCC methods. The average CH_4 emission factor used in our estimation is 76.7% lower than that of the 2006 IPCC Guidelines but 28.0% higher than that from the IPCC 2019 refinement. Therefore, our CH_4 emission results are lower than the estimate based on the 2006 IPCC Guidelines by around −246 Gg (−369 Gg contributed by emission factor and +123 Gg caused by activity level) but higher than the estimate based on the IPCC 2019 refinement method by around +78 Gg (+26 Gg contributed by emission factor and +52 Gg by activity level; Fig. 5). For N_2O, the activity data and the average emission factor in our study are 43.8% and 62.5%, respectively, lower than those estimated based on the IPCC 2019 refinement method. Our estimation of N_2O emissions is thus lower than the estimation based on the IPCC 2019 refinement method by around −54 Gg (−31 Gg contributed by emission factor and −23 Gg by activity level). The N_2O emissions estimated in this study are 3.2 times higher than the 2006 IPCC Guidelines-based estimate. This finding is consistent with Xi et al. (2021) [18], which suggests that N_2O emissions calculated using firm-level operation data are lower than the estimate of the IPCC 2019 refinement method but higher than the estimate based on the 2006 IPCC Guidelines.

4.3. Limitations and future work

The comprehensive comparisons with previous studies and the IPCC methods indicate the necessity of developing plant-level emission inventories for MWWTPs, especially considering the uneven distribution of emissions in space and the dependence of emission factors on wastewater treatment technology. Our study is one of the first attempts to establish plant-level emission inventories for MWWTPs in China using plant- and technology-based information. Based on the high-resolution emission accounting, our analysis reveals unprecedented emission features and details not shown before. However, the large discrepancies in the comparison still suggest large uncertainties underlying our current understanding of GHG emissions from MWWTPs. To address these uncertainties and harmonize different estimates, further investigations are imperative, including improvements in the following aspects.

Firstly, acquiring data on the COD or TN removal of each MWWTP becomes imperative to reduce uncertainties in activity data, which are caused by downscaling country-level activity data to plant scales in this study. Secondly, measurements of plant-integrated emission factors are urgently needed to extend to cover all treatment technologies with more reliable samples. This can help improve the accuracy of the emission factor, which is one of the most uncertain parameters in our current emission estimates for MWWTPs. Furthermore, the emission factors of MWWTPs are also affected by operating conditions, such as wastewater temperature, dissolved oxygen, and COD and TN concentrations [15,35,36]. Developing a data-driven or process-based emission factor model can further improve dynamic estimates of MWWTP emissions, which is beneficial to accurate emission estimates, emission management, and mitigation measure design. More datasets should be established and integrated to develop a more comprehensive understanding of MWWTP emissions.

5. Conclusions and implications

In conclusion, we have developed a high-resolution inventory of CH_4 and N_2O emissions from biological treatment processes of MWWTPs in China in 2020. Using the most up-to-date facility-level databases, our inventory offers an unprecedented level of detail on emission features. The national CH_4 and N_2O emissions from MWWTPs are 150.6 Gg and 22.0 Gg, with the uncertainties estimated to be −30% to 37% and −30% to 26% (at a 95% confidence interval), respectively. Our bottom-up analyses reveal an emission inequality among different cities, with the most economically developed cities emitting two times more CH_4 and N_2O per capita than the poorest cities. China's MWTTPs exhibit a relatively youthful age structure, with most CH_4 and N_2O emitted by plants less than 20 years old. This study shows that the wastewater sector is an important source of GHGs, especially for CH_4 and N_2O. The wastewater sector can make valuable contributions to realizing carbon neutrality in China through technological innovation and integration, though this transformation requires both time and investment. In addition, the emission inequality across cities implies that economically developed cities bear more responsibility to reduce carbon emissions from wastewater treatment. Taking all these factors into consideration, there stands a good chance that technological innovation in wastewater treatment will be implemented first in more developed regions, such as East China. It

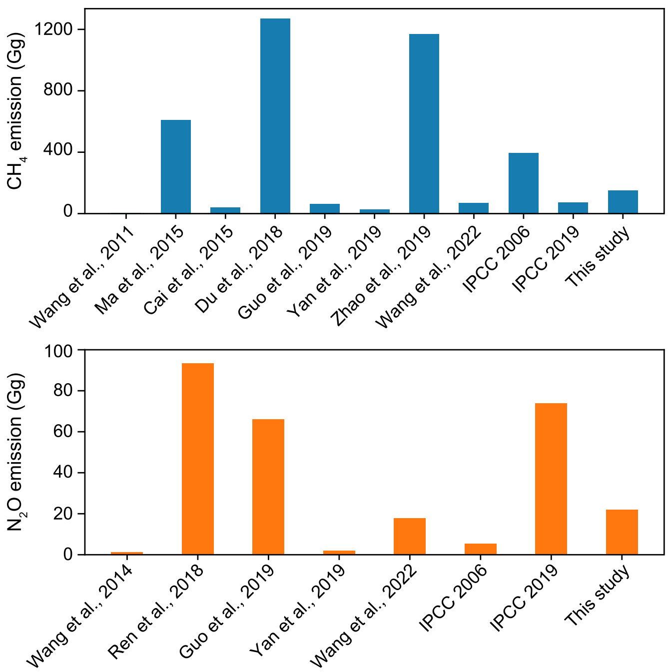

Fig. 5. CH_4 and N_2O emission evaluation by facility-based data in this study, IPCC 2006 method, IPCC 2019 method, and previous studies.

should be noted that innovative, low-carbon technologies won't replace the current conventional treatment technologies massively in the near future, especially considering the young-age structure of China's MWWTPs. Therefore, attention should also be paid to reducing carbon emissions from treatment processes by optimizing the operating conditions of current technologies.

Data availability

The dataset in this study is available from the corresponding author (lihaiyan2021@hit.edu.cn) upon reasonable request.

CRediT authorship contribution statement

Haiyan Li: Conceptualization, Methodology, Visualization, Writing - Original Draft, Writing - Reviewing & Editing. **Liangfang You:** Data Curation, Validation. **He Du:** Data Curation. **Bowen Yu:** Data Curation. **Lu Lu:** Writing - Reviewing & Editing. **Bo Zheng:** Conceptualization, Supervision, Writing - Reviewing & Editing. **Qiang Zhang:** Writing - Reviewing & Editing. **Kebin He:** Writing - Reviewing & Editing. **Nanqi Ren:** Writing - Reviewing & 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.2023.100345.

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