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Key Points:
- The methane emissions from municipal wastewater treatment plants of 229 Chinese cities were 29.2 $MtCO_2e$ in 2014
- Large cities located in the prosperous eastern China had larger methane emissions in absolute and per capita terms
- Cities with higher GDP, household food consumption expenditure, or household consumption expenditure tend to emit more methane

Supporting Information:
- Supporting Information S1
- Table S1

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China's Urban Methane Emissions From Municipal Wastewater Treatment Plant

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Abstract The increased number and capacity of municipal wastewater treatment plants (WWTPs) in China has driven the emission of methane (CH_4). Few studies have focused on quantification of CH_4 emissions from municipal WWTPs of different cities and analysis of socioeconomic factors influencing the quantity of emissions. Here we estimated CH_4 emissions from WWTPs in China for 229 prefectural-level cities, based on data from 2,019 working municipal WWTPs. The results show the total CH_4 emissions to be 1,169.8 thousand tons (29.2 $MtCO_2e$) in 2014, which is over three times that of the municipal WWTPs in the United States in 2016. Large cities along the east coast regions had larger CH_4 emissions in absolute and per capita terms. Correlation analysis shows that cities with higher gross domestic product, household food consumption expenditure, or household consumption expenditure produced more degradable organics in wastewater, thus more CH_4 emissions. Measures to control the sources of degradable organics and regulate WWTP processes with less emission factor are key to mitigate CH_4 emissions. In addition to aerobic or anaerobic wastewater treatment systems, factors such as wastewater temperature, length of sewer, and the addition of nitrate that influencing emission factor are suggested to be involved in CH_4 emission modeling.

Plain Language Summary The increased number and capacity of municipal wastewater treatment plants (WWTPs) in Chinese cities has driven the emission of methane, a potent greenhouse gas. Understanding and balancing the trade-offs between increased municipal wastewater treatment capacity and the demands for greenhouse gas emissions reduction is a big challenge for cities in developing countries like China. We estimated methane emissions from 2,019 working municipal WWTPs in China for 229 cities. The results show the total methane emissions to be 1,169.8 thousand tons in 2014, which is over three times that of the municipal WWTPs in the United States in 2016. Large and wealth cities along the east coast regions had larger methane emissions in absolute and per capita terms. Cities with higher gross domestic product, household food consumption expenditure, or household consumption expenditure produced more degradable organics in wastewater, thus more methane emissions. Measures to control the sources of degradable organics and regulate WWTP processes are key to mitigate methane emissions.

1. Introduction

Lack of treatment of municipal wastewater presents a serious environmental and public health problem, particularly in developing countries where 80–90% wastewater is either untreated or poorly treated prior to discharge (van Loosdrecht & Brdjanovic, 2014). Hence, municipal wastewater treatment plants (WWTPs) will become one of the major urban infrastructure in most developing countries in the coming decades, due to rapid urbanization and the need to treat large volume of wastewater (Singh et al., 2016). WWTPs are a significant source of greenhouse gas (GHG) emissions, generating two potent GHGs, that is, methane (CH_4) and nitrous oxide (N_2O; Mannina et al., 2018). After carbon dioxide (CO_2), methane is the second most important GHG from anthropogenic sources, which has a global warming potential of 25 CO_2 equivalents over a horizon of 100 years (Miller et al., 2013). Methane is mainly generated when organic matter is decomposed in anaerobic conditions (Mannina et al., 2018). Compared to N_2O, CH_4 emissions from WWTPs have received less attention from researchers (Daelman et al., 2012).

In the last four decades, China has experienced an unprecedented process of urbanization (Liu et al., 2015). Between 1978 (the start of economic reform in China) and 2012, the percentage population living in cities has increased from 17.9% to 52.6% (Bai et al., 2014). One of the by-products of such rapid and unplanned urbanization is the increasing discharge of untreated wastewater and accompanying and serious deterioration of freshwater bodies (Luo et al., 2018). Indeed, municipal wastewater discharge in China has increased by 230% during 2001–2014 (Society of Chinese Urban Water Supply and Drainage, 2015). Accordingly, this presents a need to collect and treat the increased volumes of urban wastewater, which inevitably drives process-related CH_4 emissions from the expansion of municipal WWTPs. Indeed, China's two latest national reports on climate change to the United Nations Framework Convention on Climate Change have revealed the increasing CH_4 emissions for wastewater treatment (NDRC, 2012, 2016). The national CH_4 emissions for wastewater treatment were 1,620 thousand tons in 2005, accounting for 3.64% of total CH_4 in that year. In 2012, the CH_4 emissions for wastewater treatment increased to 2,892 thousand tons and also took a larger share (5.17%) of total CH_4 emissions. Hence, understanding and balancing the trade-offs between increased municipal wastewater treatment capacity and the demands for GHG emissions reduction is a big challenge for cities, particularly those in developing countries such as China.

Existing peer-reviewed studies relating to CH_4 emissions from WWTPs are limited and almost all focus on measuring CH_4 emissions from specific WWTPs. Czepiel et al. (1993) quantified CH_4 emissions from a typical WWTP located in Durham, United States, and investigated the impact of wastewater temperature on CH_4 emissions. Wang et al. (2011) monitored CH_4 emissions from a full-scale anaerobic/anoxic/oxic WWTP in China and found that dissolved oxygen concentration and wastewater temperature were the two main factors influencing methane emissions in the monitored WWTP. Evaluating CH_4 emissions of a WWTP with anaerobic sludge digestion, Daelman et al. (2012) demonstrated the amount of CH_4 emissions exceeded CO_2 emissions through utilization of the resulting biogas. The monitoring results from Rodriguez-Caballero et al. (2014) showed that CH_4 emissions from a plug-flow bioreactor located in a municipal WWTP accounted for 0.016% of the influent chemical oxygen demand (COD). An investigation by Masuda et al. (2015) showed that 86.4% of CH_4 emissions were derived from anaerobic treatment tanks. These authors also evaluated three different treatment processes, oxidation ditch, double circulated anoxic-oxic, and anoxic-oxic, and found that substantial CH_4 emissions were derived from sewer transfer (Masuda et al., 2018).

GHG emissions from WWTPs at national or regional levels are not measured directly but are estimated using a mass balance approach. *The 2006 IPCC Guidelines for National Greenhouse Gas Inventories* (the 2006 IPCC Guidelines for short; IPCC, 2006) provide the accounting methodology based on the fact that CH_4 production depends primarily on the amount of degradable organic material in the wastewater. This approach has been adopted by different countries for reporting the inventory of their GHG emissions. The latest application of the IPCC approach can be found in the annual report of U.S. GHG emissions and sinks developed by the Environmental Protection Agency (United States Environmental Protection Agency, 2018). The report estimated that 357 thousand tons (8.9 MMT CO_2 Eq.) of CH_4 was emitted during domestic wastewater treatment in the United States in 2016. In China, the National Development and Reform Commission of China has issued the *Guidelines for Provincial Greenhouse Gas Emission Inventories* (*Draft*) in 2010 (the Guidelines for China for short), which aims to assist the compilation of national GHG emission inventories (NDRC, 2010). The Guidelines for China also adopted the IPCC approach and its emission factors. Using the IPCC approach, there have been two peer-reviewed studies quantifying China's CH_4 emissions from both municipal and industrial WWTPs at national (Ma et al., 2015) and provincial (Du et al., 2018) levels based on national/provincial statistics.

In summary, investigations of CH_4 emissions from municipal WWTPs have either focused on specific sites in order to assess the factors related to features of treatment and sewerage systems, that is, WWTPs and sewers, or were implemented at national/regional levels to compile an inventory of national GHG emissions. Although the features of urban drainage systems and inflow of wastewater are subject to urban planning and influenced by socioeconomic development of cities, few studies have carried out CH_4 emissions assessments from municipal WWTPs at urban level. As a result, how socioeconomic features of cities along with

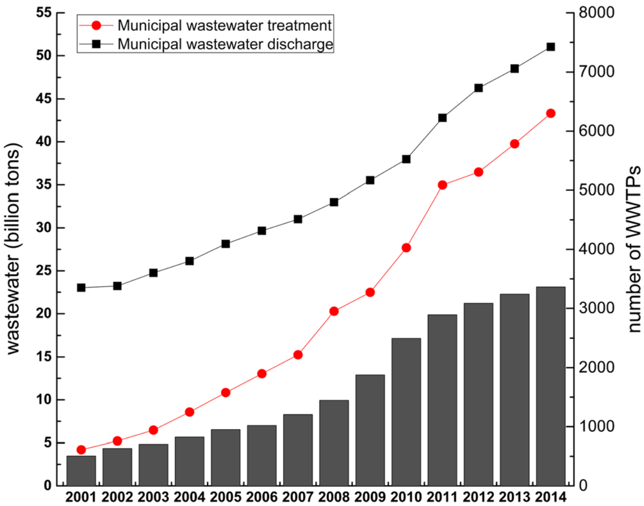

Figure 1. Municipal wastewater discharge, treatment, and numbers of WWTPs in China from 2001 to 2014. WWTP = wastewater treatment plant.

WWTP technologies have affected CH_4 emissions from municipal wastewater treatment remains largely unexplored.

Using the IPCC approach, the present study quantifies China's urban CH_4 emissions from municipal WWTPs covering 229 prefectural-level cities out of 288 in mainland China (referred to as cities for simplicity). The total population of these cities was 1.05 billion in 2014 accounting for 77.1% of the total population in China, with a gross domestic product (GDP) of 90.2% of the national total (National Bureau of Statistics of China, 2015). CH_4 emissions of 2,019 municipal WWTPs were first quantified based on data from the *Urban Drainage Statistic Yearbook* (Society of Chinese Urban Water Supply and Drainage, 2015), the results of which were aggregated to different cities. The spatial characteristics of urban CH_4 emissions and the socioeconomic factors that affect CH_4 emissions were then analyzed.

2. Overview of China's Municipal WWTPs

Municipal wastewater discharges in China have more than doubled during the 21st century (Figure 1). During this time, the amount of treated municipal wastewater has grown even faster, increasing 10.3 times between 2001 and 2014. As a result, the treatment efficiency, that is, the rate of treated municipal wastewater to discharged wastewater, has increased from 18% to 85%. Such an increase demonstrates that China's capacity to treat sewage has undergone rapid development in a relatively short period of time. The number of municipal WWTPs has grown from only 506 in mainland China in 2001 to 3,362 at the end of 2014 (Figure 1; Society of Chinese Urban Water Supply and Drainage, 2015).

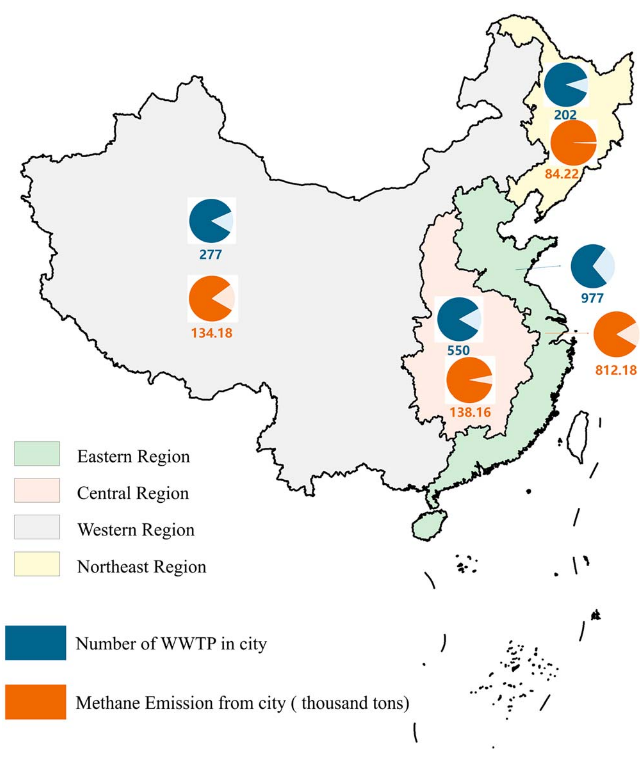

Figure 2. Methane emissions and number of municipal WWTPs in four regions accounted for by cities and regional totals (regional totals are represented as the full pie and calculated through adding up provincial totals in that region). WWTP = wastewater treatment plant.

In terms of treatment processes, anaerobic-anoxic-oxic and oxidation ditch were the two most popular wastewater treatment processes, accounting for 31% and 21% of total WWTPs in China (Zhang et al., 2016). The main treatment technologies in descending order were conventional activated sludge, sequencing batch reactors, anoxic-oxic, biological film, and chemical and physicochemical treatments. Spatially, municipal WWTPs are unevenly distributed. Officially, China can be divided into four economic regions, that is, eastern, central, western, and northeastern China (Figure 2; Long et al., 2011). In 2014, about half of the WWTPs (1,707) were located in eastern China, with the number of WWTPs in central, western, and northeastern China being 802 (23.8%), 575 (17.1%), and 278 (8.3%), respectively (Figure 2).

3. Methodology and Data

3.1. Method to Estimate CH_4 Emissions

Our study utilized the approach described in the Guidelines for China, which followed the basic framework and emission factors provided in the 2006 IPCC Guidelines. The IPCC approach calculates the maximum amount of methane from a given amount of degradable organics, which is commonly expressed through biochemical oxygen demand (BOD) or COD (IPCC, 2006). The equations for CH_4 emissions from a municipal WWTP are as follows:

$$E_{CH_4} = (TOW \times EF) - R, \tag{1}$$

where E_{CH4} is the CH_4 emissions in the inventory year (kg CH_4/year); *TOW* is the total organics in wastewater in the inventory year measured through BOD (in this study kgBOD per year); *EF* is the emission factor (kg CH_4/kg BOD); and R is the amount of CH_4 reclamation in the inventory year (kg CH_4/year). Since there is still no large-scale recovery of CH_4 in China (Hu et al., 2014), the amount of R is assumed to be zero.

The formula of the emission factor (EF) is shown as follows:

$$EF = B_0 \times MCF, \tag{2}$$

where B_0 is the maximum CH_4 producing capacity (kg CH_4/kg BOD), using the recommended value of 0.6 (NDRC, 2010); MCF is the methane correction factor, which according to the 2006 IPCC Guidelines (IPCC, 2006) is zero for well-managed aerobic systems, 0.3 for not well-managed aerobic systems, and 0.8 for anaerobic systems. The MCF of detailed WWTP is based on expert judgement as recommended by the 2006 IPCC Guidelines. The MCF was selected for each WWTP in our study based first on whether the WWTP belongs to an aerobic or anaerobic system through expert judgement (details in supporting information Table S1) and second, for aerobic systems, whether the WWTP is organically overloaded, that is, not well-managed, or is otherwise considered to be operating within design parameters and well-managed based on data. Finally, for those WWTPs with no data on their treatment processes, we applied the national average MCF of 0.165.

The CH_4 emissions of each WWTP were thus quantified utilizing equations (1) and (2), and the CH_4 emissions of each city were acquired by summing up the CH_4 emissions of all WWTPs for that city. In addition, we used a Pearson correlation analysis to investigate the relationship between CH_4 emissions and several socioeconomic factors.

We also calculated the national total CH_4 emissions from municipal WWTPs. First, we added up provincial level CH_4 emissions from our existing data of 2,583 municipal WWTPs (${CH_4}^{pe}$). Second, because our data did not cover all municipal WWTPs of China, the missing part of CH_4 emission at provincial level (${CH_4}^{pm}$) is calculated using national average EF.

$${CH_4}^{pm} = B_0 \times MCF_{\text{average}} \times (TOW^p - TOW^w), \tag{3}$$

where MCF_{average} is the national average MCF of 0.165, TOW^p is the total BOD influent to municipal WWTPs at provincial level (National Bureau of Statistics of China, 2015), and TOW^w is the total provincial BOD influent from the municipal WWTPs of our existing data. Hence, the CH_4 emissions at provincial level can be acquired as ${CH_4}^p = {CH_4}^{pe} + {CH_4}^{pm}$, and the national total CH_4 emissions are the sum of provincial totals.

3.2. Data

The information from specific WWTPs to quantify CH_4 emissions, that is, the BOD content of wastewater and wastewater treatment process, were acquired from *Urban Drainage Statistic Yearbook* (Society of Chinese Urban Water Supply and Drainage, 2015). This Yearbook covers the above information for 2,583 municipal WWTPs, which accounted for approximately 77% of national municipal WWTPs. Among these municipal WWTPs, there are 2,019 belonging to prefectural-level cities and other 564 belonging to county-level cities. We then selected these 2,019 municipal WWTPs from 229 Chinese cities in 29 provinces to do the quantification. The data used for correlation analysis including urban GDP, population, household consumption expenditure, food consumption expenditure, and sewer length for the 229 cities were collected from the Provincial Statistic Year Book 2015 (National Bureau of Statistics of China, 2015). The urban water quality stress of the cities used for our correlation analysis was calculated according to Zhao et al. (2016), which was acquired as the ratio of gray water footprint to annual renewable freshwater for that city. Gray water footprint here means the volume of freshwater required to assimilate the pollutant load based on its ambient water quality standard and natural background concentration (Hoekstra et al., 2011).

4. Results and Discussion

4.1. Spatial Distribution of CH_4 Emissions From Chinese Cities

The total CH_4 emissions from municipal WWTPs in the 229 Chinese cities contained in our study amounted to 1,169.8 thousand tons (29.2 $MtCO_2e$) in 2014. This volume amounted to 83.8% of the national total for WWTP CH_4 emissions of 1,395.8 thousand tons. The ratio of CH_4 emissions and the number of WWTPs in our study to that of the national total are different among the four economic regions and can be found in

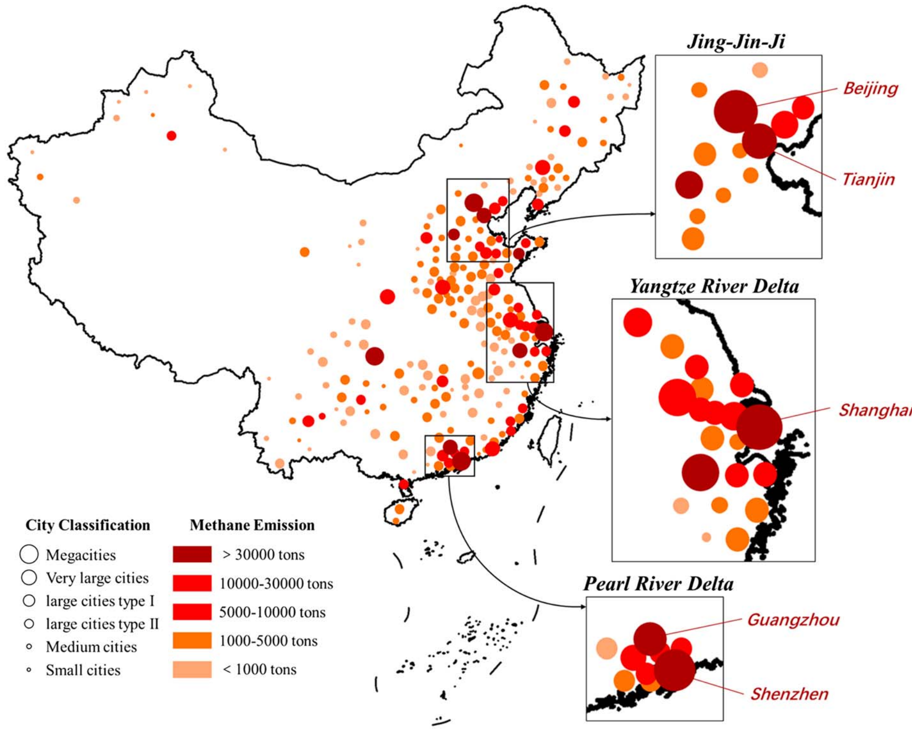

Figure 3. CH_4 emissions from municipal wastewater treatment plants in 229 cities.

Figure 2. CH_4 emissions for different cities showed a large difference, ranging from 0.028 to 97.8 thousand tons. The top five cities with the largest CH_4 emissions were Shanghai, Shenzhen, Beijing, Guangzhou, and Tianjin; emissions from these five cities accounted for 26.8% of the total CH_4 emissions of the 229 study cities. Shanghai alone generated 97.8 thousand tons of CH_4 in 2014, accounting for 8.4% of total emissions. Among these top cities, Shanghai, Beijing, and Shenzhen are classified as megacities (urban population in excess of 10 million), and Tianjin and Guangzhou are the two most populous cities in the very large category (urban population between 5 and 10 million). A common feature of these cities is that they are all located in the prosperous eastern China. In addition, they are the economic centers of China's three biggest Metropolises, that is, Beijing and Tianjin in the so-called Jing-Jin-Ji Metropolis, Shanghai in the Yangtze Delta Metropolis, and Shenzhen and Guangzhou in the Pearl River Delta Metropolis (Figure 3). These three Metropolises, consisting of 42 cities, emitted 609.8 thousand tons of CH_4 accounting for 43.7% of total emissions of the 229 study cities (Table 1).

Apparently, bigger cities with bigger populations emit more CH_4 from their WWTP's since they tend to generate more municipal wastewater. We found that most of these larger cities also had greater per capita CH_4 emissions. In 2014, mean per capita CH_4 emissions for the 229 study cities was 1.1 kg per capita, of which Shenzhen had the largest CH_4 emission per capita (5.6 kg per capita), followed by Shihezi (5.4 kg per capita), Hangzhou (4.9 kg per capita), Qingdao (4.3 kg per capita), Shanghai (4.0 kg per capita), and Guangzhou (3.9 kg per capita). With the exception of Shihezi, which has a relatively small population of only 0.64 million, the other four cities all have populations in excess of 9 million people.

We classified our study cities into six population-based groupings in order to reveal the pattern of per capita CH_4 emissions. Such classification follows the standard issued by the State Council of China (http://www.gov.cn/zhengce/content/2014-11/20/content_9225.htm), which is based on the permanent population in urban areas (Figure 4). Our analysis shows that per capita CH_4 emissions reduce when the scale of the cities gets smaller, from 2.9 to 0.3 kg per capita, from the highest to the lowest. This sharp decline in CH_4 emissions occurs between Larger city type I (population of 3–5 million people) and Larger city type II (population of 1–3 million people). As shown in Table 1, a similar trend was found for per capita GDP and the ratio of anaerobic to aerobic treatment systems in cities, which may provide an explanation for CH_4 emission

Table 1
Comparison Between Different City Groupings and Metropolises (Note That There Are Seven Cities Not Included in the List of City Groupings due to the Absence of Population Data)

	Number of cities	CH_4 emissions (thousand tons)	Per capita wastewater treatment capacity (L per capita per day)	Ratio of anaerobic to aerobic systems	Per capita GDP
City groups					
Megacities	4	252.0	234	1.77	86.9
Very large cities	8	179.0	195	1.43	96.1
Large cities type I	14	239.3	163	1.73	81.6
Large cities type II	91	338.7	87	0.94	47.7
Medium cities	86	122.1	65	0.75	37.1
Small cities	19	14.8	50	0.72	31.3
Metropolises					
Jing-Jin-Ji	13	181.4	111	1.02	60.2
Yangtze River Delta	20	252.6	158	1.83	91.1
Pearl River Delta	9	175.8	311	1.18	100.0

Note. GDP = gross domestic product.

patterns in the different population groupings. In other words, larger city groupings with greater populations are more developed and apply more anaerobic treatment solutions to their WWTPs, which may contribute to greater CH_4 emissions.

4.2. Socio-Economic Factors Affecting CH_4 Emissions From Municipal WWTPs

According to the IPCC approach, large discrepancies in CH_4 emissions in different cities are mainly determined by two factors, that is, EF and the degradable organic fraction in the municipal wastewater. We explored the impact of degradable organics on CH_4 emission in this section and the EF in next section. To exclude the impact of the EF, we used China's national average EF of 0.099 kg CH_4/kg BOD provided by *the Guidelines for China* for all WWTPs contained in our study so as to recalculate CH_4 emissions for each city. When the EF was fixed, CH_4 emissions were solely determined by degradable organics in wastewater, and the higher the ratio of degradable organics in the wastewater, the more CH_4 was emitted from the municipal WWTP (El-Fadel & Massoud, 2001). Since the degradable organics were measured through BOD, the quantity of BOD in municipal wastewater was the direct factor affecting CH_4 emissions using the fixed EF. To go a step further, the difference in BOD content in municipal wastewater may be attributed to varied socioeconomic factors. Here we propose several factors which may potentially affect urban degradable organic fraction in wastewater and investigate the correlation between CH_4 emissions (under the fixed EF) and the proposed socioeconomic factors. The selection of the factors are based on the following considerations: First, higher GDP, household food consumption expenditure (the amount of final consumption expenditure made by resident households to meet their food needs), and household consumption expenditure (the amount of final consumption expenditure made by resident households to meet their everyday needs, such as clothing, food, housing, energy, transport, durable goods, health costs, leisure, and services) represent higher standards of living in cities, which may increase the degradable organic fraction in wastewater (Ma et al., 2015). Second, in the previous section, population was shown to influence CH_4 emissions. Third, higher water quality stress in cities suggests greater discharge of BOD into watercourses. Hence, we chose urban GDP (covering 229 cities), household consumption expenditure (205 cities), household food consumption expenditure (114 cities), population (229 cities), and water quality stress (82 cities) as the socioeconomic factors influencing the biodegradable organic fraction in wastewater.

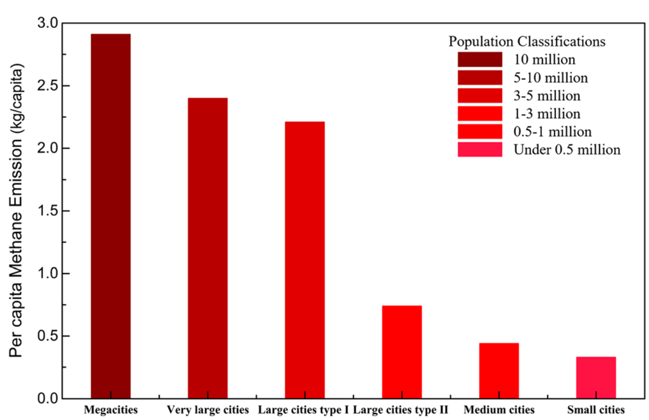

Figure 4. Per capita methane emissions from different city groupings.

The resulting correlation analysis showed three factors, that is, GDP, household food consumption expenditure, and household consumption expenditure, were very strongly correlated with CH_4 emissions using

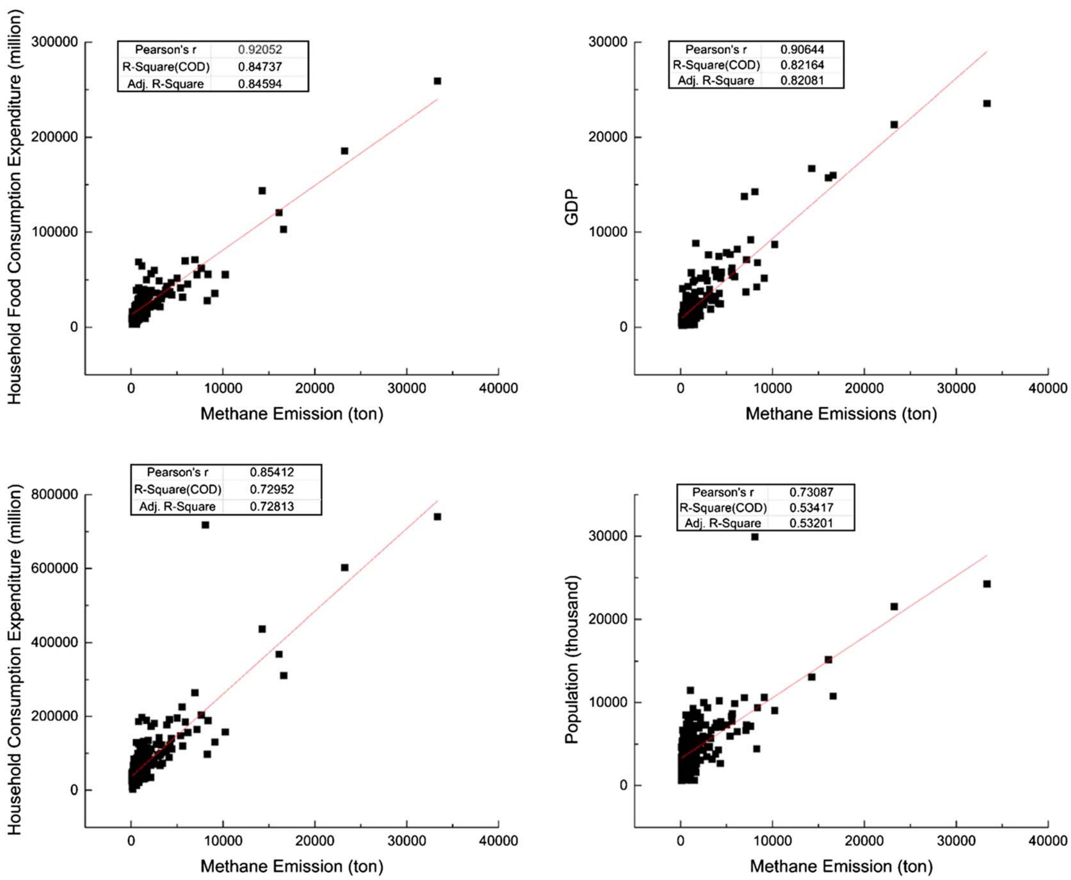

Figure 5. Correlation between CH_4 emissions using fixed emission factor and socioeconomic factors.

fixed EF ($r > 0.8$, $p < 0.01$; Figure 5). Previously, Ma et al. (2015) found the quantity of domestic wastewater effluent grew annually with stable GDP growth, thus suggesting anthropogenic CH_4 emissions might be highly correlated with levels of economic development. Our findings support this argument through examination of the relationship between degradable organics in municipal WWTPs and living standards. Two explanations may be postulated: First, cities with populations enjoying higher living standards consume greater quantities of food with higher protein content, such as meat, egg, and dairy products, the waste of which results in higher degradable organic fractions in wastewater. Second, GDP is a reflection of the degree to which a city has more highly developed infrastructure including municipal WWTPs. In turn, this means it has increased capacity to collect wastewater, thereby greater inflow to municipal WWTPs. Evidence for the above explanation is that CH_4 emissions using fixed EF are highly correlated to the extent of sewerage in 169 of the study cities (Figure S1).

In Figure 4, we found a strong correlation ($0.6 < r < 0.8$, $p < 0.01$) between population and CH_4 emissions from WWTPs. As previously mentioned, bigger cities with bigger populations tend to generate more municipal wastewater, thus more CH_4 from their municipal WWTPs. In addition, population is also highly correlated with living standards in China. Large cities with greater populations tend to be more developed due to the agglomeration effect of cities. Our correlation analysis showed that urban GDP is very strongly associated with urban population ($r = 0.81$, $p < 0.01$).

Furthermore, the result of the correlation analysis indicated very weak correlation ($r = 0.05$) between CH_4 emissions and water quality stress (Figure S2). Some cities, such as Pingdingshan and Anyang in Henan Province, suffer extreme water stress with less WWTP derived CH_4 emissions, which may be explained by poor density of WWTPs. In this case, expansion of WWTP's may help reduce urban water quality stress but may increase CH_4 emissions. Conversely, WWTPs deployed in cities such as Beijing, Tianjin, and Shijiazhuang in Hebei Province as a result of high water stress result in higher CH_4 emissions. In both

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Table 2
Emission Factors From Different Wastewater Treatment Plants

Location	Emission factor (kgCH$_4$/kg COD)	References
229 Chinese cities	0.017–0.24	Current study
Durham, United States	0.0016	Czepiel et al. (1993)
Jinan, China	0.0008	Wang et al. (2011)
Capelle aan den Ijssel, Netherland	0.0113	Daelman et al. (2012)
Valence, France	0.0175	Yver Kwok et al. (2015)
Bellheim, Germany	0.0001	Tumendelger et al. (2019)

cases, source pollution control is recommended as a way of reducing urban water quality stress and CH_4 emissions.

4.3. Analysis Toward Improvement of Emission Factor

In addition to degradable organics, EF is the other important factor in determining CH_4 emissions. The EF used in this paper for different WWTPs was obtained from the 2006 IPCC Guidelines, ranging from 0.034 to 0.48 kg CH_4/kg BOD for different cities. In equation (2), a default value was used for maximum CH_4 producing potential (B_0), and EF was solely determined by the methane correction factor (*MCF*) which indicated the degree of anaerobic treatment in the wastewater system (IPCC, 2006). As shown in Table 1, larger city groupings tend to have more WWTPs based on anaerobic processes, suggesting that they have larger EF. Such an observation suggests EF may also be correlated with higher standards of living (higher GPD, household food consumption expenditure, and household consumption expenditure). This observation is supported from correlation analysis using variable EF (i.e., the effect of the EF is not excluded). Compared to the previous section using fixed EF, similar correlation relationships were obtained between CH_4 emissions using variable EF and the proposed socioeconomic factors (Figure S3).

The results from the IPCC approach imply that cities utilizing more aerobic WWTPs have less or no CH_4 emissions (*MCF* is zero for well-managed aerobic systems). However, such a conclusion simplifies the impact of wastewater treatment processes and other on-site factors on EF and CH_4 emissions. Different WWTPs have different scales and locations and also utilize different biological, physical, and chemical technologies during wastewater treatment, all of which are related to CH_4 emissions. Hence, estimating the EF based on on-site influencing factors and simulating CH_4 generation from WWTPs based on the mathematical model using more specific EF need to be developed. Until recently, data describing CH_4 emissions from on-site processes were very limited. There are only a few peer-reviewed studies reporting on their on-site measurements of EF in the form of CH_4 emissions per unit of COD influent (Table 2). These results, derived from specific WWTPs, are various and not enough to be used to represent the EF of multiple cities or larger regions. In addition, the on-site measurement itself has a lot of uncertainties due to the variation of different measurement methods and conditions (Yver Kwok et al., 2015). Hence, the IPCC approach using EF is currently more suitable to quantify CH_4 from municipal WWTPs of multiple cities/regions. It should be noted that a refinement of the IPCC approach is underway to incorporate new knowledge on data for EF development (IPCC, 2016). As for the WWTPs, efforts to obtain better data reflecting emissions from various types of WWPTs are being developed (United States Environmental Protection Agency, 2018). We suggest that a more wide-ranging comparison of CH_4 emissions with different treatment processes, scale, and geophysical location is a promising research avenue to provide more accurate EF for WWTPs.

4.4. Uncertainty Analysis

The overall uncertainty associated with CH_4 emission estimates from municipal WWTPs was quantified using Approach 1 methodology in the 2006 IPCC Guidelines, that is, an error propagation method (IPCC, 2006). Uncertainty associated with the parameters used to estimate CH_4 emissions in this study includes methane correction factor (*MCF*), maximum CH_4 producing capacity (B_0), and the data of BOD contents. According to the 2006 IPCC Guidelines, the uncertainty range for methane correction factor (*MCF*) and maximum CH_4 producing capacity (B_0) are $\pm$10% and $\pm$ 30%, respectively. We take the uncertainty in the BOD data to be 10%, because the uncertainty of statistical data in China is 5–10% according to Du et al.

(2018). Combining the above uncertainty together by multiplication (equation S1), the uncertainty associated with CH_4 emission of a single municipal WWTP is ±33.17%. While combining the uncertainties of 2,019 municipal WWTPs by addition (equation S2), the overall uncertainty is ±2.47%. Although the data at plant level reduced the overall uncertainty, this uncertainty estimates overlook an important uncertainty, that is, the EF-based IPCC approach itself. Overall, using EF to estimate GHG emissions from WWTPs leads to great uncertainty (Mannina et al., 2018).

Model improvement is a practical way to reduce the uncertainty. We summarize several on-site influencing factors which could be considered in model development to improve the EF estimation. During wastewater treatment, CH_4 is typically generated in areas of high BOD and low oxygen concentration (Czepiel et al., 1993), and the dissolved CH_4 is stripped from wastewater mainly through aeration (Wang et al., 2011). Other studies have shown CH_4 is mainly generated in sewers and through the anaerobic digestion of sewage sludge (Daelman et al., 2012; Masuda et al., 2015, 2018) and is then stripped in open tanks with high dissolved oxygen concentrations, such as aerated grit chambers and oxic tanks (Wang et al., 2011). According to the 2006 IPCC Guidelines, anaerobic digester of sludge was considered in the EF estimation, but the impact of sewerage on CH_4 emissions was not included in EF estimation since "wastewater in closed underground sewers is not believed to be a significant source of CH_4" (IPCC, 2006). However, more recent studies have found that a large proportion of CH_4 is generated in sewers by methanogenic organisms during anaerobic biological nutrient decomposition processes (Guisasola et al., 2008). This generated CH_4 is dissolved in the sewage and later emitted at the WWTP. Indeed, monitoring results from Wang et al. (2011) showed significant CH_4 was emitted from aerated grit chambers and influent pumping stations due to influent wastewater from sewers. An investigation on CH_4 emissions from a full-scale WWTP showed that 18.4% of CH_4 was produced in sewers and later emitted at the WWTP (Masuda et al., 2015). Masuda et al. (2018) suggested that, because the hydraulic retention time in grit chambers is generally only a few minutes, almost all CH_4 emissions from grit chambers must originate in sewers. Moreover, temperature in wastewater was reported to be one of the most significant factors influencing CH_4 emissions. Czepiel et al. (1993) found CH_4 emissions from grit tanks were highly correlated to wastewater temperature. Masuda et al. (2015) showed that CH_4 emissions were higher in summer and lower in winter due to seasonal temperature fluctuations. In addition, a study by Jiang et al. (2010) found that addition of nitrite can substantially inhibit CH_4 production in a laboratory based gravity sewer system, which could be considered as a way of mitigating CH_4 emissions. Therefore, a correction factor could be added to the EF estimating model to include the impact of sewers, wastewater temperature, and nitrite in wastewater.

5. Conclusions

Water pollution and carbon emissions are two great environmental challenges faced by urbanization. On one hand, wastewater treatment ranks the fifth in terms of anthropogenic CH_4 emissions (United States Environmental Protection Agency (USEPA), 2014), while, on the other, rapid urbanization and requirement for better water environment in developing countries inevitably drive greater CH_4 emissions from municipal WWTPs. There is potential for mitigating CH_4 emissions from municipal wastewater treatment, but these depend on socioeconomic factors such as economic development, government, and technology selection (United States Environmental Protection Agency (USEPA), 2014). From a modeling perspective, we have reported on the quantification of CH_4 emissions from municipal WWTPs for 229 cities in China based on the IPCC approach. Such quantification facilitates comparison of different urban CH_4 emissions, thus assisting in identification of spatial features and key socioeconomic factors influencing these emissions. Our results show the largest CH_4 emissions occur in the more economically developed region of eastern China, with its greater and more affluent population. Socio-economic factors such as GDP, household consumption expenditure, household food consumption expenditure, and population were found to highly correlate with urban CH_4 emissions from municipal WWTPs. Such findings suggest that controlling residential discharge of municipal wastewater is a promising avenue for control of wastewater CH_4 emissions. Based on the IPCC framework, installing more WWTPs with aerobic systems is a recommendation for reducing EF (United States Environmental Protection Agency (USEPA), 2014). However, other factors such as length of and in-sewer conditions, wastewater temperature, and nitrate concentrations in WWTPs were also important on-site factors in determining CH_4 emissions. We would recommend these are included in future EF estimation work. Due to data limitation, we are unable to develop multiyear CH_4 inventory for municipal WWTPs. Analyzing the evolution of CH_4 emissions from municipal WWTPs and the associated driving forces would be our next goal.

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