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Critical Review

Methane Emissions from Municipal Wastewater Collection and Treatment Systems

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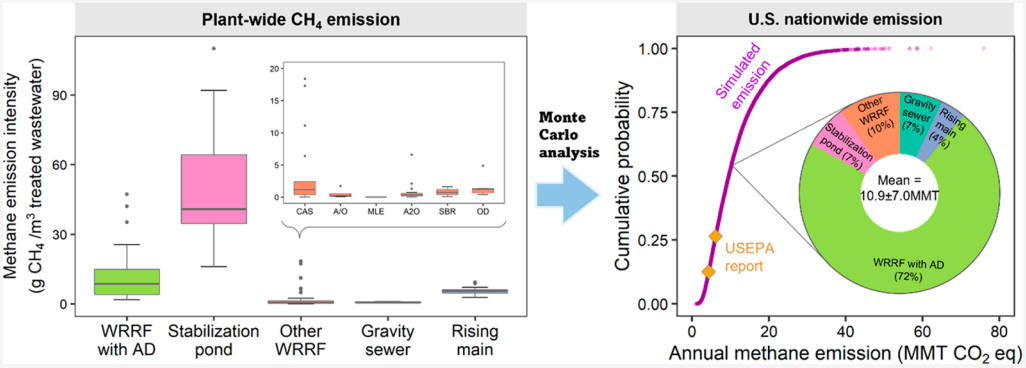

ABSTRACT: Municipal wastewater collection and treatment systems are critical infrastructures, and they are also identified as major sources of anthropogenic CH_4 emissions that contribute to climate change. The actual CH_4 emissions at the plant- or regional level vary greatly due to site-specific conditions as well as high seasonal and diurnal variations. Here, we conducted the first quantitative analysis of CH_4 emissions from different types of sewers and water resource recovery facilities (WRRFs). We examined variations in CH_4 emissions associated with methods applied in different monitoring campaigns, and identified main CH_4 sources and sinks to facilitate carbon emission reduction efforts in the wastewater sector. We found plant-wide CH_4 emissions vary by orders of magnitude, from 0.01 to 110 g CH_4/m^3 with high emissions associated with plants equipped with anaerobic digestion or stabilization ponds. Rising mains show higher dissolved CH_4 concentrations than gravity sewers when transporting similar raw sewage under similar environmental conditions, but the latter dominates most collection systems around the world. Using the updated data sets, we estimated annual CH_4 emission from the U.S. centralized, municipal wastewater treatment to be approximately 10.9 ± 7.0 MMT CO_2-eq/year, which is about twice as the IPCC (2019) Tier 2 estimates (4.3−6.1 MMT CO_2-eq/year). Given CH_4 emission control will play a crucial role in achieving net zero carbon goals by the midcentury, more studies are needed to profile and mitigate CH_4 emissions from the wastewater sector.

KEYWORDS: *Wastewater treatment, Methane, IPCC, Monitoring campaigns, Mitigation strategies, Literature text mining, Monte Carlo analysis*

1. INTRODUCTION

Methane is a potent greenhouse gas (GHG) with a much shorter average lifespan in the atmosphere (~12 years) than CO_2 (hundreds of years).[1] When measured over a 20-year period, its global warming potential is 84−86 times that of CO_2.[2,3] The total radiative forcing attributable to anthropogenic CH_4 is 0.54 ± 0.11 W/m^2, contributing around 16% of current atmospheric warming.[1] Studies have shown that CH_4 reduction can be more cost-effective than CO_2 when normalized to per tonne of abated CO_2-equivalent.[4] With many nations committed to achieving net zero carbon emission within the next 20−30 years, reducing CH_4 emission becomes a priority and a relatively quick way to reduce overall GHG emissions.[5]

The wastewater sector is a major source of CH_4 emission, contributing to 5−8% of global anthropogenic CH_4 emissions, just following livestock (32%), oil and gas (25%), landfills (13%), and coal mining (11%).[6] During wastewater collection and treatment, CH_4 is produced in anaerobic environments where methanogenic archaea convert acetate, H_2, or formate to CH_4 and CO_2 following anaerobic fermentation and acetogenesis. For a water resource recovery facility (WRRF), direct

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CH_4 emission alone can account for up to 79% of the Scope 1 emission (direct GHG emissions from a facility)[7] and up to 57% of the total carbon footprint (sum of direct and indirect emissions).[8,9] While many WRRFs promote managed CH_4 production via anaerobic digestion for energy recovery, fugitive emissions across the sewer and treatment systems remain a major challenge. It is mainly because CH_4 emissions have a diffusive and ebullitive nature, occur in many places, and vary depending on many factors such as process and configuration, wastewater characteristics, and operating conditions. In addition, CH_4 generated in one reactor may be emitted in downstream processes, which further complicates CH_4 management and mitigation.[10,11] For example, CH_4 generated in sewer lines and primary clarifiers is generally stripped during aeration in downstream activated sludge systems,[10,12,13] whereas CH_4 release from anaerobic reactors such as up-flow anaerobic sludge blanket (UASB) or anaerobic membrane bioreactor (AnMBR) is mainly associated with saturated or supersaturated dissolved CH_4 in the discharged effluent.[11,14]

Despite the recognition of CH_4 emissions from the wastewater sector, limited literature is available on its quantification and mitigation. Currently, the tier-based methodology proposed by the Intergovernmental Panel on Climate Change (IPCC) is commonly used at the city, region, and national levels for general emission estimates, and example protocols include the Australian National Greenhouse Accounts Factors and California Air Resources Board's Local Government Operations Protocol.[15,16] Successive tiers from Tier 1 to 3 provide increasing accuracy at the cost of increasing complexity in collecting plant- or country-specific data such as CH_4 emission factors (EFs) for prominent treatment systems and wastewater/sludge treatment activity data.[17] Given a lack of study compiling and analyzing CH_4 EFs associated with various treatment processes and operation conditions, almost all CH_4 emission estimates adopted the IPCC suggested EFs, which were obtained from 14 field measurements for aerobic treatment systems in conjunction with expert assessment for other treatment systems (e.g., anaerobic treatment).[17] However, this rough estimation may not accurately represent actual CH_4 emissions from wastewater treatment. Studies have reported that plant-level CH_4 emission estimated by simpler, lower-tier IPCC (2006) methodologies can be up to 2 orders of magnitude higher than the actual emission.[18] In addition, current tier-based IPCC methodology does not cover CH_4 emission from sewers due to a lack of data, which may significantly underestimate the overall emission from the wastewater sector as recent studies reported significant sewer network CH_4 production.[19,20] For example, Defratyka et al. (2021) measured the total CH_4 emissions using $\delta^{13}CH_4$ isotopes from ground covers of the sewer network in Paris, France to be 62,700 kg CH_4/year, and they found sewers contribute 33% of the total detectable emissions from the ground, only following the emission from natural gas distribution network (63%).[20]

The discrepancies between estimated and actual CH_4 emissions highlight the need for a global emission inventory based on real, full-scale measurements, whereby site-specific conditions should be taken into account.[21] Increasing monitoring campaigns have been carried out in recent years targeting facilities with different treatment capacities and processes. WRRFs from <1 million gallon per day (MGD) to >100 MGD have been monitored,[13,22−24] and popular processes such as conventional activated sludge, sequencing batch reactors, anaerobic/anoxic/oxic, and anaerobic digestion have been evaluated.[10,23,25−28] These studies provide valuable quantitative information on site-specific emissions, but the information is scattered, and comparison and assessment are difficult due to great variations and incomplete data sets. Moreover, different sampling methods and analytical techniques (e.g., flux chamber, tracer gas dispersion method (TDM), and optical gas imaging infrared (IR) video camera) were used in previous monitoring campaigns. These techniques provide varying degrees of spatial and temporal coverage and accuracy[29,30] and lead to significant variabilities in reported results.[31] For example, when on-site measurements using IR and TDM were compared for four biogas plants in Denmark, IR measured CH_4 emissions were found to be either lower (up to 58%) or higher (up to 120%) than results from TDM.[31] Lower results obtained from IR were mainly because of unidentified or unquantified CH_4 sources that are difficult to access (e.g., top of a reactor), variations in the true emission rate, and uncertainties in calculating the CH_4 emission rate from gas engines;[31] underestimated results from TDM were possibly caused by incomplete tracer and CH_4 gas mixing during measurement.[31]

In this study, we employed a literature mining method to collect all published literature that measured CH_4 emissions from the centralized, municipal wastewater sector. We then analyzed the results and extracted the findings to present a first comprehensive data analysis of CH_4 emissions across different sewer systems and treatment processes in WRRFs. We summarized and examined variations in CH_4 emissions associated with methods applied in different monitoring campaigns. More importantly, we identified the main CH_4 emitting sources and potential approaches to facilitate GHG emission reduction efforts in the wastewater sector. The findings offer a comprehensive and representative emission inventory covering all reports to date and provide a detailed comparison of CH_4 emission intensities associated with different treatment processes and emission sources. Using these updated data sets, we estimated nationwide CH_4 emissions from the U.S. municipal wastewater sector with consideration of treatment processes and site-specific conditions by performing a Monte Carlo analysis.

2. METHODS

2.1. Data Collection and Statistical Analysis. The literature text mining collected >310,000 full publication records between 1900 and 2022 from Web of Science based on organized keywords associated with wastewater, sludge, digestion, sewer, and other wastewater-related terms (refer to Section S1 of the Supporting Information (SI) for the detailed text mining method). Titles and abstracts were first word-tokenized by *n*-grams (one, two, three, and four adjacent words), and then all the tokens and author keywords were lowercased, stop-worded, and stemmed to develop a searchable database. Since we focused on actual CH_4 emissions of wastewater collection and treatment systems that were reported from monitoring campaigns, many studies about CH_4 recovery/conversion, dairy farm CH_4 emission, and other topics were excluded by applying a screening process based on both methane- and wastewater-related terms. In addition, further fine screening criteria were conducted to extract papers with explicit units of CH_4 emissions in the abstract and to exclude model-based and other domain-based (e.g., rivers and lakes) studies from the collected papers. The detailed text

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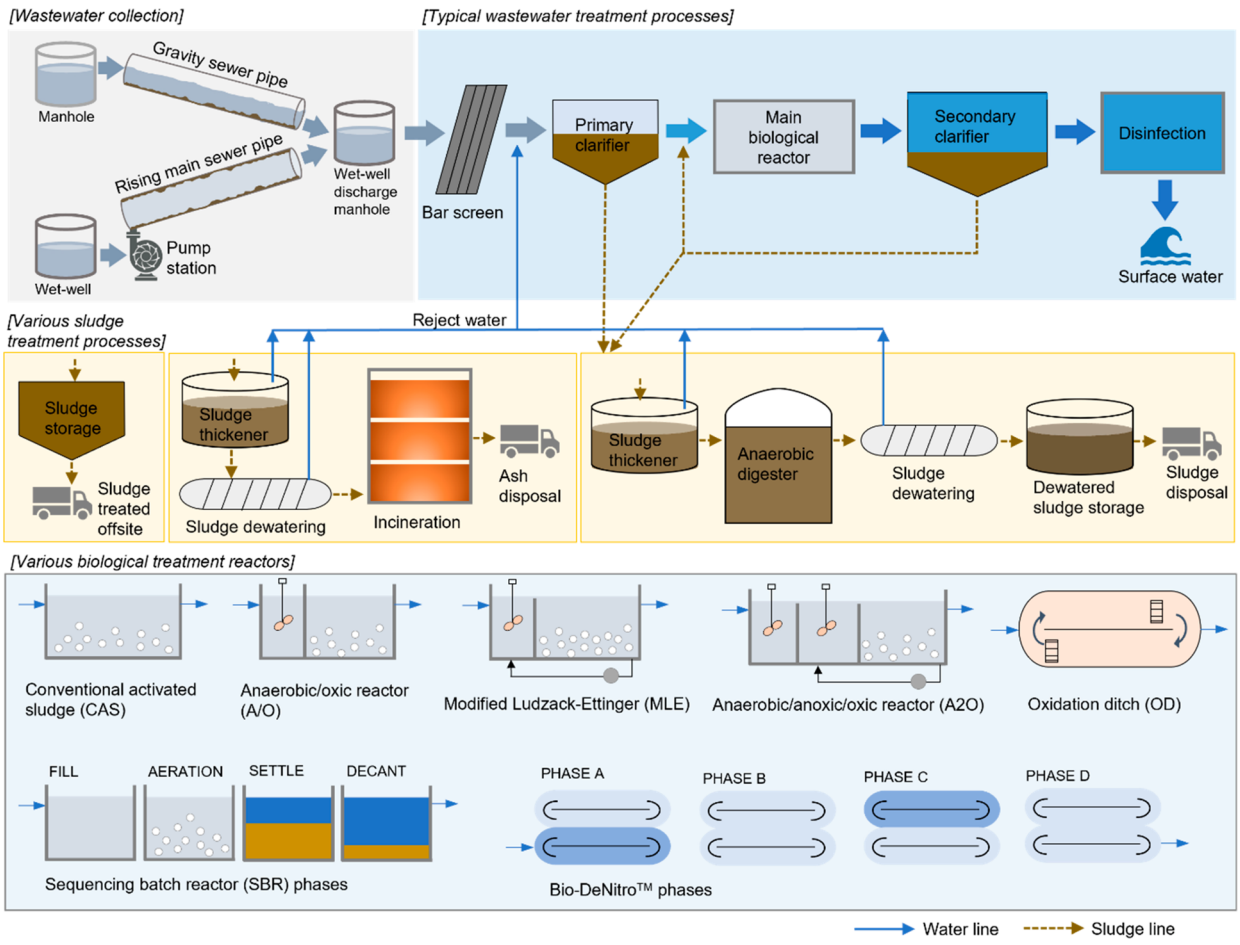

Figure 1. Typical wastewater collection and treatment trains with various biological treatment and sludge management processes.

mining methodology descriptions can be found in Section S1 of the SI and Zhu et al. (2021).[32]

We organized and normalized CH_4 emission intensity to gram CH_4 per volume of wastewater treated (g CH_4/m^3) using the average flow rate during the monitoring period. If the average flow rate was not provided, this information was obtained either from facility Web sites, state online repositories, or through cross reference of related peer-reviewed literature (see Section S2 for details). Temporal variations in CH_4 emissions were captured by collecting monthly or seasonal data reported in previous monitoring campaigns and given equal weight in the following analysis. For statistical analysis, we compared mean CH_4 emission and used the 95% confidence interval (95% CI, equaling the upper bound minus the lower bound at 95% confidence level) to estimate uncertainties of mean values. The 95% CI was calculated using the population standard deviation (σ) and sample size (n) assuming Student's t distribution and $\alpha = 1 - 0.95 = 0.05$.

2.2. Description of Methane Emission Data Sets. The general wastewater collection and treatment train with different variations is illustrated in Figure 1. Wastewater collection networks, including both gravity sewers and rising mains, carry sewage flows to nearby WRRFs which treat sewage to meet discharge requirements via various steps and processes. A typical train of wastewater treatment consists of primary (which for the purposes of this paper also includes preliminary treatment; e.g., influent screening, grit chamber, primary clarifier), secondary (e.g., aeration tank, secondary clarifier), tertiary (e.g., nutrient removal, disinfection), and sludge (e.g., thickening, anaerobic digestion, dewatering, and storage) treatments that aim to remove carbon, nitrogen, phosphorus, solids, pathogens, and other constituents, as well as reduce the volume and stabilize wastewater sludge. Figure 1 highlights the most common wastewater and sludge treatment processes summarized in this study. The variations include seven biological treatment options: 1) conventional activated sludge (CAS), 2) anaerobic/oxic process (A/O), 3) modified Ludzack-Ettinger (MLE), 4) anaerobic-anoxic-oxic process (A2O), 5) sequencing batch reactor (SBR), 6) oxidation ditch (OD), and 7) Bio-Denitro reactor. All of these biotreatment processes have a certain level of nutrient removal capability.[33,34] Besides, we also analyzed three sludge management processes: 1) untreated sludge storage, 2) sludge thickening, dewatering, and incineration for combined heat and power, and 3) sludge thickening, anaerobic digestion, dewatering, storage, and disposal.

2.2.1. Wastewater Collection Networks. Through literature mining and manual review, we collected 81 measurements for 21 sewer sites mainly located in the United States, Australia, Thailand, China, and France. While these sewer networks carry out the functions of sewage collection and conveyance, biological reactions may also occur in such pseudo plug-flow reactors. Generally, biofilms (also known as biological slime layers) grow on normally wetted pipe surfaces where hydrolyzing and methanogenic biology are a ubiquitous

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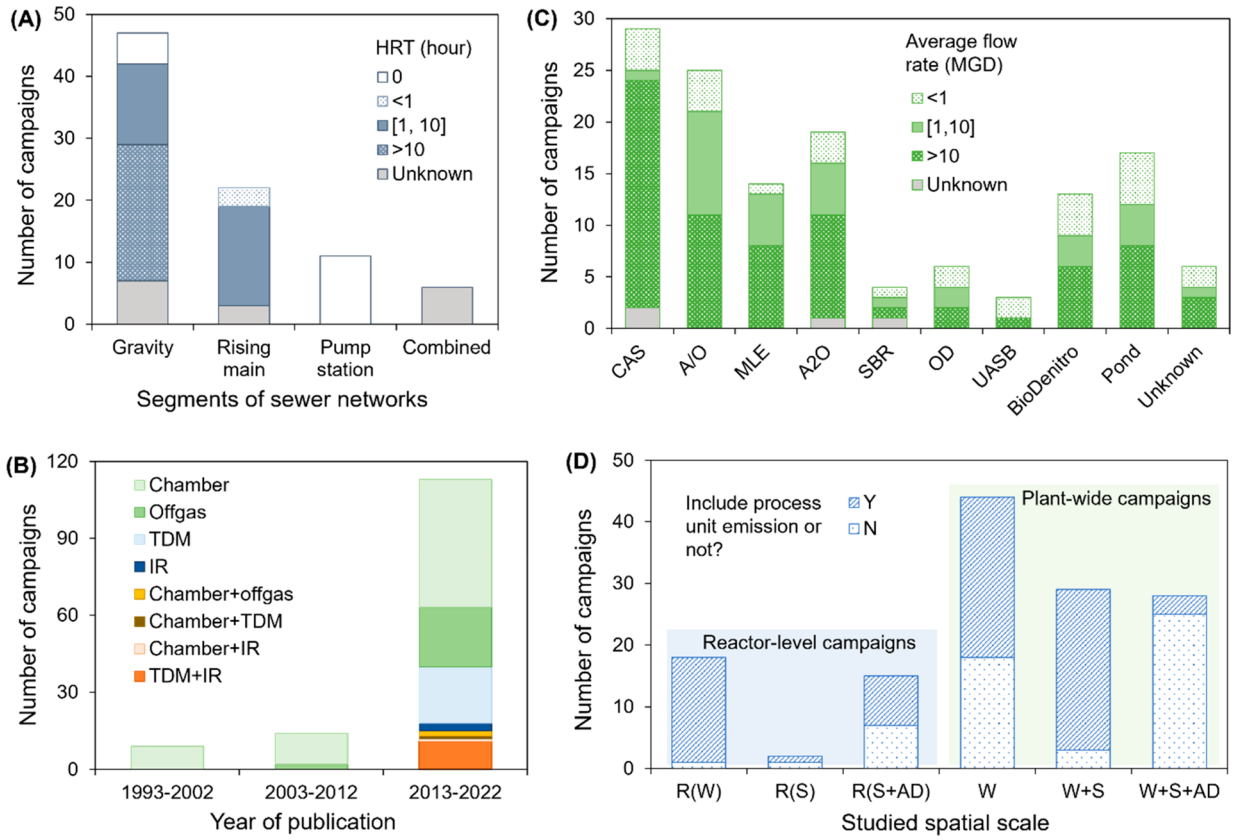

Figure 2. CH_4 monitoring campaigns for sewer networks (A) and WRRFs (B–D). (A) Sewer networks are shown in different types of sewer systems grouped by hydraulic retention time (HRT). WRRF data are shown for (B) the year of publication and applied measurement techniques, (C) plant configurations differentiated by main biological treatment processes, and (D) spatial scales of the studies. TDM: tracer gas dispersion method; IR: optical gas imaging infrared video camera; MGD: million gallon per day (1 MGD = 3785 m^3/day); pond refers stabilization pond; W, S, and AD are CH_4 emissions from wastewater line, sludge line (without anaerobic digestion), and anaerobic digestion, respectively.

primary source of CH_4 production, even in low-strength or rapidly moving systems. Wastewater flow often fluctuates substantially over time, and stagnant flow creates anaerobic environments and solids deposition that can heighten methanogenesis from bulk-phase or slime sources. Figure 2A shows the number of campaign data based on types of sewer networks (gravity or rising main sewer, pump station) and wastewater hydraulic retention time (HRT) of each monitoring campaign. Gravity sewer pipes are the most frequently studied sewer type (55% of the total measurements), followed by rising mains (26%) and pump stations (13%).

2.2.2. Water Resource Recovery Facilities. From literature mining we retrieved 136 measurements of CH_4 emissions from 90 WRRF sites. We focused only on centralized, engineered treatment systems and excluded studies on natural or constructed wetlands as well as decentralized systems such as septic and latrine systems during manual reviewing. Figure 2B presents the number of CH_4 monitoring campaigns reported over the past three decades and the measurement techniques used in these studies. The majority of the measurements (117 out of 136 total measurements) were conducted in the past decade, reflecting the increasing interest in emission studies and fast development in measurement techniques. The flux chamber method is the most common method used, which accounts for ~60% of the total measurements. It has high accuracy and works well for point sources and small areas, but for larger areas such as a whole plant or sewer network, chambers were found to underestimate the total flux, because emission hotspots are often not properly represented by a few measurement points.[35] The tracer gas dispersion method (TDM) was increasingly used in the past decade, representing ~24% of total measurements. TDM is a ground-based optical remote sensing method combining the controlled release of tracer gas with concentration measurements downwind of the facility, which can provide plant-integrated emissions. An optical gas imaging infrared (IR) video camera is another emerging method which is able to capture CH_4 emitted from a source as a gray-colored cloud, and can be used to detect leaks remotely from pipes, tanks, valves, and areas hard to access.[31] With recognition of the strengths and limitations of each technique, which are discussed thoroughly in Parravicini et al., (2022),[36] more studies started to apply multiple techniques to capture both reactor-scale and plant-level emissions.

Figure 2C shows the number of campaigns on various biological processes in WRRFs and the average flow rate of the monitored facilities. The most studied processes include CAS (21.5% of the total measurements), A/O (18%), A2O (14%), stabilization pond (12.5%), MLE (10%), and Bio-Denitro (10%). More than 80% of the monitoring campaigns focused on WRRFs with an average flow rate greater than 1 MGD (or 3785 m^3/day), whereas measurements on small plants (<1 MGD)[37] were limited. The most-commonly studied small WRRFs include UASB, stabilization pond, MLE, CAS, and A/O. Recent studies found that small plants may emit two times more CH_4 per volume of wastewater treated than large ones because small plants are often unable to adapt to wastewater

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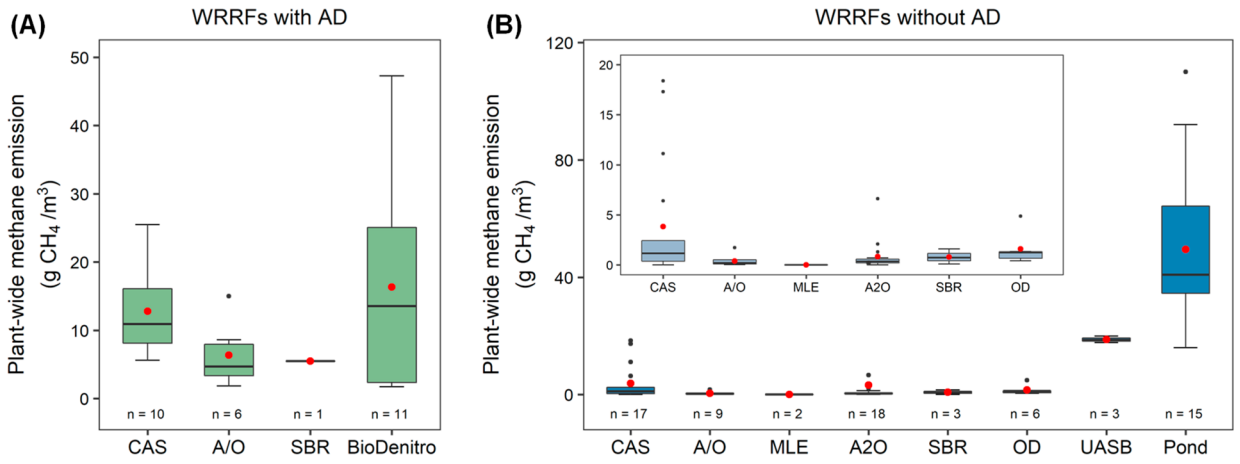

Figure 3. A summary of plant-wide CH_4 emissions with respect to different biological treatment processes for WRRFs with and without anaerobic digestion. The inset in (B) highlights those with relatively low CH_4 emissions. Boxplots show 25th, 50th, and 75th percentiles and outlier-bounds are based on 1.5 × IQR (interquartile range, equaling 75th percentile minus 25th percentile) of CH_4 emissions. Red dots represent the arithmetic mean. *n*: the number of monitoring data.

flow and quality changes due to aging infrastructure and inferior monitoring, which lead to suboptimal operating conditions and poor performance.[38] Considering a large number of small facilities do not have resources or work forces for efficient carbon management, more monitoring and analyses are needed for gaining better understanding of their emissions.[39,40]

The spatial scales of the monitoring data at either plant-wide or reactor-level are summarized in Figure 2D. The results indicate plant-wide CH_4 emissions are more intensively studied (101 out of the 136 measurements) than reactor-level emissions. We further grouped these measurements based on the distinct spatial scales involved, including wastewater line only (W), sludge line without anaerobic digestion (S), and sludge line with anaerobic digestion (S+AD). There are 44 plant-wide monitoring campaigns which only reported wastewater line emissions. We considered them as plant-wide measurements because some facilities do not have onsite sludge treatment,[7,10,41] or the contribution of sludge management on CH_4 emissions was excluded.[25] Notably, there are 51 plant-wide measurements and 33 reactor-level measurements that reported CH_4 emissions per process unit (e.g., primary clarifier, biological reactor, and sludge treatment), which provide key information for analyzing CH_4 emitting sources from WRRFs in Section 3.3.

3. RESULTS AND DISCUSSION

3.1. Plant-wide Methane Emissions Among Different Processes. Plant-wide CH_4 emissions are categorized based on the main biological treatment processes for WRRFs operating with and without AD and summarized from 101 measurements reported from the literature (Figure 3). By comparing WRRFs with similar treatment processes, the plants operating with AD showed >3 times higher CH_4 emissions than those plants without AD. For example, plant-wide CH_4 emissions from two A/O facilities operating with AD are in the range of 8.6−15.0 g CH_4/m^3,[18] which are much higher than A/O facilities without AD (0.1−1.7 g CH_4/m^3).[26,42−44] Elevated CH_4 emissions from WRRFs equipped with AD are mainly caused by biogas leakage from anaerobic digesters and incomplete flaring of excess CH_4.[45] It is commonly hypothesized that floating covers have higher emissions than fixed cover digesters, though no specific literature data were found. Other sources may include dewatering anaerobically treated solids, leaks at pressure relief valves, ventilation from biomass mixing tank, and gas engines.[31] Previous studies reported that CH_4 emissions from AD alone are in the range of 0.3−18.2 g CH_4/m^3 (exclude data from Mexico),[13,28,30,46,47] and as high as 4.7−42.9 g CH_4/m^3 for studies reported in Mexico.[24] While biogas production from AD followed by combined heat and power cogeneration (~150 g CO_2-eq/kWh) can offset GHG emissions otherwise provided by carbon-heavy grid electricity (national average of 373 g CO_2-eq/kWh based on eGRID data in 2020[48]), fugitive CH_4 emissions could complicate the net benefits of these AD operations.[49] This finding highlights the importance of efficient biogas capture in mitigating carbon emissions.

For WRRFs with AD, the most studied processes are Bio-Denitro and CAS (Figure 3A). Plant-wide CH_4 emissions are reported in the range of 1.7 to 47.3 g CH_4/m^3 with the mean (lower-upper 95% CI) value of 12.5 (7.9−17.2) g CH_4/m^3. Bio-Denitro process showed a higher emission and variation than other processes with mean (lower-upper 95% CI) at 16.3 (4.8−27.9) g CH_4/m^3. The Bio-Denitro process was designed for enhanced nitrogen removal capacity by minimizing dissolved oxygen transfers to the anoxic reactor via alternation of two tanks between anoxic and aerobic conditions without internal circulation.[34] Higher emissions were observed in such processes during foaming events in anaerobic digesters (up to 47 g CH_4/m^3) as compared to around 10 g CH_4/m^3 during normal operational conditions.[50] It indicates the importance of maintaining proper operational conditions of anaerobic digesters for carbon management. Besides, sludge treatment was also believed to be a main CH_4 emitting source for the studied Bio-Denitro facilities.[18,50] However, this qualitative statement still needs to be proven using measurements that aim to distinguish between emissions from various process units.

The most studied processes of WRRFs without AD include CAS, A/O, A2O, and stabilization pond (Figure 3B). For these facilities, plant-wide CH_4 emissions vary widely from 0.01 to 110 g CH_4/m^3 with the mean (lower-upper 95% CI) of 12.9 (7.5−18.2) g CH_4/m^3. High variabilities of CH_4 emissions are linked to the complexity of how CH_4 is generated and released

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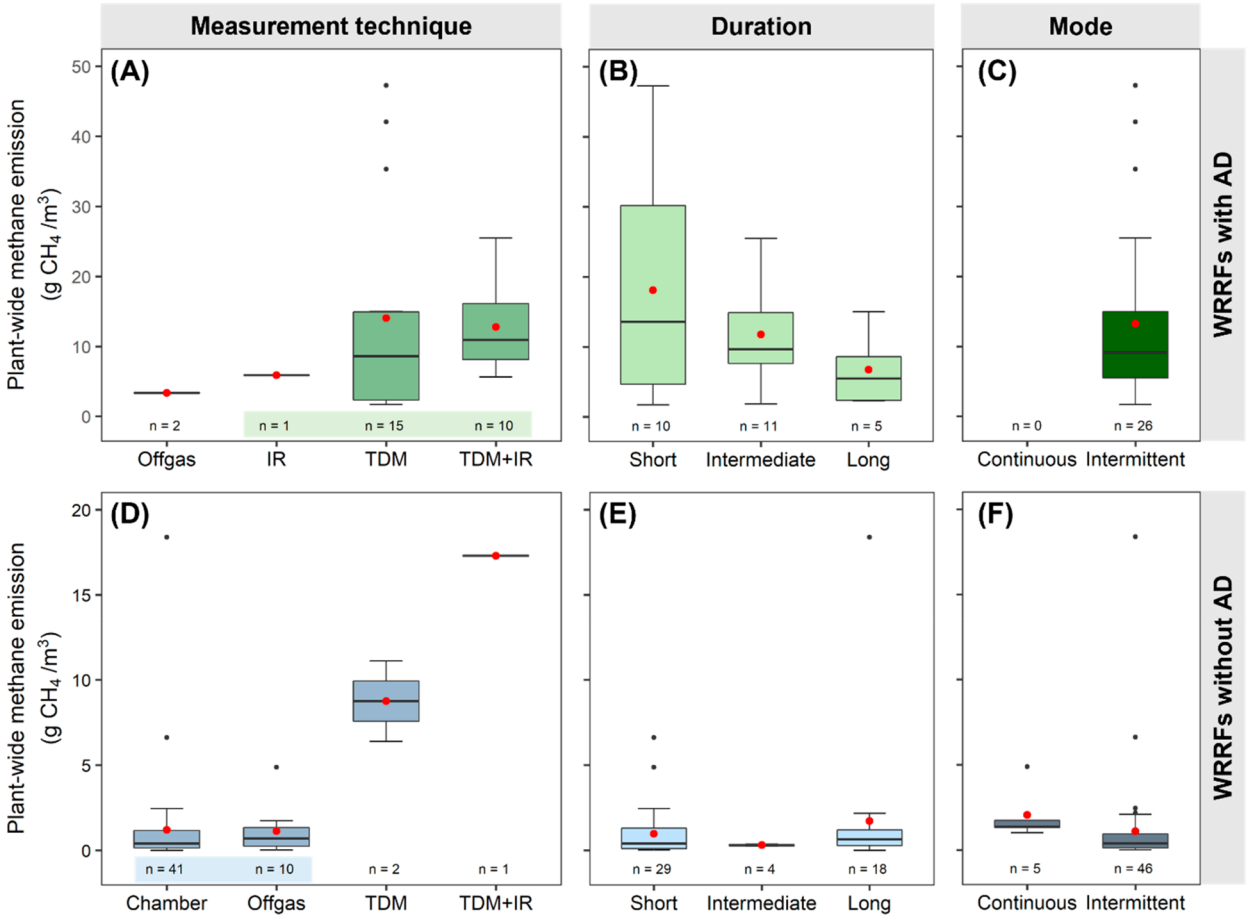

Figure 4. Plant-wide CH_4 emissions with respect to measurement techniques (IR: optical gas imaging infrared video camera, TDM: tracer gas dispersion method), the length of the monitoring period (short: less than one month, intermediate: a few months without capturing the whole spectrum of temperature changes, long: at least one year), and monitoring mode of gaseous sampling for WRRFs with AD (A–C) and those without AD (D–F). Boxplots show 25th, 50th, and 75th percentiles and outlier-bounds are based on 1.5 × IQR of CH_4 emissions. Red dots represent the arithmetic mean. *n*: the number of monitoring data. Data in the shaded areas in panels A and D are used to further analyze impacts of duration and mode on CH_4 emissions to avoid bias caused by various measurement techniques.

during each step of treatment of wastewater and sludge across various treatment processes. Among all studied WRRFs without AD, stabilization ponds and UASB were found to have higher mean emissions at 49.5 (35.1–63.9) g CH_4/m^3 and 18.8 (15.9–21.6) g CH_4/m^3, respectively, >4 times larger than the emissions from the remaining processes. Several factors are believed to lead to significantly higher emissions and variations in these processes. Stabilization ponds generally comprise an arrangement of open anaerobic, facultative, and/or maturation ponds in series. They are commonly used in community-level wastewater treatment, which are open to the environment and generally do not have accurate operational controls or sufficient and ubiquitous aeration. Consequently, studies have reported high CH_4 emissions in areas of anaerobic organic degradation.[7] For example, the maximum emissions were found around 110 g CH_4/m^3, and a large portion (>78%) of these emissions come from anaerobic ponds.[7,41] One thing to note is that the reported CH_4 emissions from stabilization ponds could underestimate actual emissions, because all data reported were measured using the flux chamber method. Such measurements focus on quantifying the diffusive flux of gases across the air–water surface but hardly capture sporadic bubble transmission (ebullition) by a limited number of gas hoods. Due to the relatively low solubility of CH_4 in water (e.g., 20.3 g/m^3 at 30 °C), CH_4 is often emitted in the form of bubbles that rise directly from the sediments.[14,29] Taking CH_4 emissions from inland freshwater ecosystems (e.g., lakes, rivers, and reservoirs) as an example, ebullition contributed up to 99% of the total diffusive and ebullitive flux.[29,51] However, there is no information on the contributions of ebullition to the overall CH_4 emission from stabilization ponds. Given stabilization ponds are popular wastewater treatment processes worldwide especially for decentralized and developing communities,[41] more efforts should be carried out to characterize and reduce CH_4 emissions from this type of facility.

The UASB process is widely used in treating high strength industrial wastewater. It has high organic loading, low sludge production, and energy and space saving advantages over aerobic treatment.[52] One benefit of UASB is that the reactors have a high rate in biogas generation, which can be used for energy recovery. Studies estimated that around 23–31% of the influent COD can be collected as CH_4.[53] However, dissolved CH_4 in the effluent has long been a challenge for UASB, and it was found 40–50% of the total generated CH_4 in UASB reactor is dissolved in the effluent,[14,54] which can be emitted to the atmosphere in effluent weirs and piping due to turbulence or agitation. Previous studies found that dissolved CH_4 in the

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effluent would result in 17.9−20.2 g CH_4/m^3 when influent COD ranges from 442 to 520 mg/L,[53] larger than some plant-integrated emissions. Different technologies have been tested for removal/recovery of dissolved CH_4 in the effluent, such as diffused aeration, packed or vacuum desorption chambers, closed downflow hanging sponge reactors, and membrane contactors.[11,55−57] To date, these methods have only been tested at the lab and pilot scales, with no full-scale applications yet reported.

The remaining types of WRRFs without AD showed relatively low CH_4 emissions. Their mean values vary in the range of 0.01 to 3.8 g CH_4/m^3 (inset figure of Figure 3B), suggesting that these process variations do not affect CH_4 emissions. For example, comparable plant-wide CH_4 emission intensities were observed from A/O (0.02−1.7 g CH_4/m^3),[8,12,58] A2O (0.04−6.6 g CH_4/m^3),[22,38,59,60] SBR (0.1−1.6 g CH_4/m^3),[10,23] and OD (0.4−4.9 g CH_4/m^3)[8] facilities with different flow rates and organic loadings. Even though a similar level of CH_4 emissions was found from these processes, different measurement techniques and analytical tools were used for short- and long-term monitoring campaigns. This makes direct comparison and benchmarking of CH_4 emissions from WRRFs challenging. The impacts of these factors will be discussed in detail in the following section.

3.2. Factors Affecting the Quantification of Methane Emissions. Figure 4 depicts factors influencing the quantification of plant-wide CH_4 emissions from WRRFs, including measurement techniques, the duration of monitoring campaigns, and sampling mode (continuous or intermittent). For WRRFs without AD, only processes shown in the inset of Figure 3B were compared because they are used in centralized plants with comparable CH_4 emission intensities. In this case, data from stabilization pond and UASB were excluded from this section's analysis to avoid biases caused by relatively high emissions from the two anaerobic processes.

Figure 4A,D compares plant-level CH_4 emissions of different measurement techniques for WRRFs with and without AD, respectively. The results shown that TDM and TDM related (i.e., TDM+IR) measurements are dominant methods for WRRFs with AD, whereas flux chamber and off-gas are mainly used for WRRFs without AD. When measuring the same emission source, TDM or TDM related measurements tend to have the highest mean CH_4 values, followed by other techniques such as IR, off-gas, and flux chamber. When previous studies compared the TDM and IR in CH_4 measurements from a biogas plant in Denmark, it was found the TDM based results (13.5 kg CH_4/h) showed more than double the amount measured by IR (6.5 kg CH_4/h).[31] Despite such a difference, the authors argued that TDM and IR measurements cannot replace each other for both accurate measurements of total CH_4 emissions and source identification.[31,61] Specifically, TDM provides a powerful and robust means to perform plant-integrated CH_4 measurements,[18,50] while IR and chamber techniques are more suitable in monitoring emissions at specific locations such as aeration tank or secondary clarifier, though the limitation of deployment numbers makes it difficult to cover all sources or capture the spatial and temporal variabilities of fluxes.[31,61]

High seasonal and diurnal variations in plant-wide CH_4 emissions were widely observed in previous studies.[25,49,58,62] Generally, CH_4 emissions tend to be high in spring/summer when the water temperature is relatively high, and lower emissions were observed in colder winter seasons.[50] Positive correlations between CH_4 emission and water temperature have been reported in previous studies (e.g., CH_4 emission rate in g CH_4/m^3 = 16.9 × water temperature −109).[12,58] Such a correlation can be attributed to more active microbial activities and therefore higher emissions in the high temperature environment.[25,26,58] Since the rate of reaction doubles for every 10 °C rise in temperature around room temperature,[58] the high production rate in conjunction with low solubility at high temperature (e.g., 29.9 g/m^3 at 10 °C, 23.2 g/m^3 at 20 °C, and 20.3 g/m^3 at 30 °C) intensifies gas stripping to the atmosphere and therefore also leads to higher emissions.[14,63] Regarding diurnal variations, studies reported the morning/evening peaks of influent flow were closely followed by an increase in CH_4 emission.[49,62] Consequently, the reported CH_4 emissions from the monitoring campaigns vary significantly depending on when, where, and how the campaign was carried out. To unveil how these factors affect measured CH_4 emissions, we further studied the impact of the length of the monitoring period and monitoring mode (continuous or intermittent) of gaseous sampling on reported plant-level CH_4 emissions. It should be noted that off-gas measurements were excluded for WRRFs with AD, and TDM related measurements were excluded for WRRFs without AD to avoid bias caused by various measurement techniques.

In Figure 4B,E, we categorized monitoring campaigns into short-term (less than one month), intermediate term (a few months without capturing the whole spectrum of temperature changes), and long-term samplings (lasting at least one year). For WRRFs with AD (Figure 4B), the mean CH_4 emissions decrease notably with the increase of duration of monitoring campaigns; e.g., the mean of short-term campaigns (18.1 g CH_4/m^3) is around 53% and 170% higher than intermediate term (11.8 g CH_4/m^3) and long-term (6.7 g CH_4/m^3) campaigns, respectively. This is mainly due to high emission events such as foaming in AD reactors captured by several short-term campaigns,[50] whereas intermediate and long-term campaigns neutralize the short-term fluctuations with no foaming events reported. When anaerobic digesters experienced foaming problems, excess pressure was created in the reactors and biogas may leak from the pressure relief valves that are installed at the top of the reactors to prevent over pressurization.[31,50] For WRRFs without AD (Figure 4E), short-term campaigns represent a slightly lower mean CH_4 emission (1.0 g CH_4/m^3) than long-term campaigns (1.7 g CH_4/m^3). It is likely because short-term monitoring periods fail to capture seasonal variations in CH_4 emissions.[33] In addition, short-term monitoring campaigns are easily affected by short-term perturbations which complicate the direct cross comparisons between different studies and their findings.

Figure 4C,F compares CH_4 emissions obtained from continuous campaigns versus intermittent campaigns for WRRFs with and without AD, respectively. For WRRFs with AD (Figure 4C), all measurements have been intermittent using TDM/IR methods, because the continuous TDM/IR measurements are difficult to deploy. For WRRFs without AD (Figure 4F), the mean CH_4 emission of continuous campaigns (2.0 g CH_4/m^3) is about two times that of intermittent campaigns (1.1 g CH_4/m^3), indicating intermittent samplings have a high probability to underestimate actual CH_4 emissions when flux chamber or off-gas technique was applied. It is worth noting that more continuous campaigns are needed to prove the above statement as current measurements are extensively dominated by intermittent campaigns.

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Overall, the campaign duration (from days to years) and mode (continuous monitoring or not) can move the mean and interquartile range of CH_4 emission significantly. Short-term and intermittent samplings likely induce under- or over-estimated results depending on the timing of the monitoring, the frequency of emissions, and the extent to which a one-time sampling (or few repeated samples) represents the emissions profile. High seasonal and diurnal variations along with high uncertainties of CH_4 emissions obtained from short-term and intermittent campaigns highlight the importance of implementing online, long-term monitoring campaigns to reliably identify CH_4 emission patterns and relative magnitudes for WRRFs.[9]

3.3. Unit-Level Methane Emissions in Typical Treatment Trains and Mitigation Strategies. To further quantify the contribution of CH_4 emissions from each process unit, we analyzed 51 plant-wide measurements and 33 reactor-level measurements that reported CH_4 emissions at process unit scale. We found that each process unit has the potential to become a major contributor in plant-wide CH_4 emissions, but the actual contributions vary widely from site to site. Figure 5

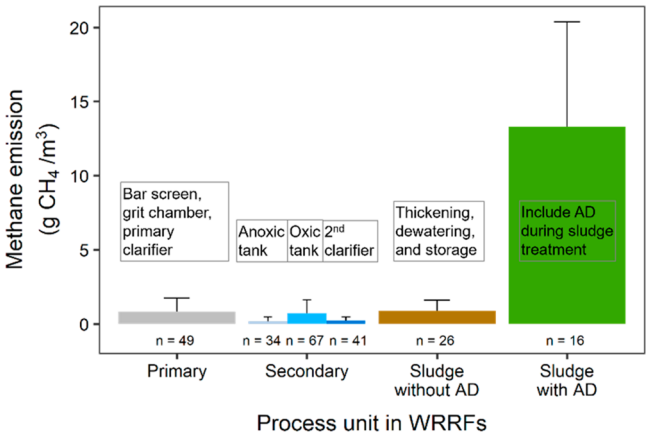

Figure 5. Mean CH_4 emissions of different process units in WRRFs. Error bars indicate upper 95% CI values.

summarizes mean CH_4 emissions of each process unit in WRRFs. It is apparent that sludge treatment with AD has the highest mean (lower-upper 95% CIs) values of 13.3 (7.3−20.1) g CH_4/m^3, which is one order of magnitude higher than other treatment stages, including secondary treatment at 1.1 (0.3−2.6) g CH_4/m^3 (sum of anoxic tank, oxic tank, and secondary clarifier), sludge treatment without AD at 0.9 (0.4−1.6) g CH_4/m^3, and primary treatment at 0.8 (0.2−1.7) g CH_4/m^3.

Generally, limited CH_4 is produced during the primary treatment process (i.e., grit chamber and primary clarifier) where biological activity is relatively low and HRT is short. Some studies reported high CH_4 emissions from primary treatment are closely linked to CH_4 production in the primary clarifier[26] and/or high dissolved CH_4 in the municipal wastewater arriving to the plant from the sewer network which is stripped out by the preaeration step before the primary clarifier.[64] Thus, reducing CH_4 production in sewer networks also facilitates carbon mitigation at WRRFs, which will be discussed in detail in Section 3.4.

The anoxic tank, oxic tank, and secondary clarifier are major units in secondary treatment that carry out most biological reactions. Despite a high DO concentration in the oxic tank that supposedly inhibits methanogenesis, studies found more CH_4 emissions occurred because a high aeration rate stripped dissolved CH_4 into the atmosphere (Figure 5).[23] Previous studies reported that the increase in DO in the range of 1 and 2 mg/L leads to higher agitation and increased gaseous CH_4 stripping from the aeration tank.[10,12,25] In addition, it was observed that CH_4 flux at the front part of the oxic tank (11.2 $g/m^2/day$) was orders of magnitude higher than its flux at the end of the tank (0.06 $g/m^2/day$).[9,23,65] Dissolved CH_4 arriving to oxic tank comes from various sources, including residual CH_4 generated in sewer networks, CH_4 generated in anoxic tank that resides before the oxic tank,[42] and dissolved CH_4 in the recycled sludge or side-stream lines pumped back to the front of the treatment train.[66] Schneider et al. (2015) reported facilities without sludge processing or return sludge flow showed low CH_4 emission rates in the oxic tank. Suggestions were made on separate treatment of return sludge or side-stream deammonification that may be a way to reduce CH_4 emission.[66] The secondary clarifier generally has negligible CH_4 emissions given the low turbulence environment, high DO, and limited dissolved CH_4 .[12,25] Relatively high CH_4 levels in the secondary clarifier were detected from several WRRFs, mostly attributed to conditions such as large air−water exchange surface area, low DO operation, or high organic loading if the secondary clarifier's influent is directly from the anoxic tank for denitrification purpose.[38,43,64,66]

For sludge treatment without AD (e.g., thickening, dewatering, and storage), CH_4 emissions occur when anaerobic conditions exist and sludge storage was identified as a dominant CH_4 emitting source for many WRRFs.[8,22,26] Some measures were tested to reduce CH_4 emissions during sludge treatment such as reducing sludge temperature or increasing the oxygen supply that makes the storage environment less favorable for CH_4 production.[45] Adding ammonia in sludge for sanitizing purposes also proved to result in low CH_4 emissions mainly due to ammonia inhibition of microbes.[67,68] When composting serves as the final step for sludge treatment, a 18% dosage of biochar was demonstrated to significantly reduce CH_4 emission by 95%.[69] Previous studies further suggested that certain facility configurations also play a role in abating CH_4 emissions, such as enclosed sludge treatment and storage units.[18,31,45]

For sludge treatment equipped with AD, elevated CH_4 emissions were detected from plants that have inefficient biogas capture or flaring systems as well as those that do not operate using good practices. As aforementioned, temperature plays a major role in CH_4 production and emission due to the interplay between microbial activities and CH_4 solubility. This may include operating digesters at suboptimal temperatures (i.e., 32 °C) or at an excessive sludge retention time in a mesophilic digester (i.e., 31 days).[24] In addition, storage and dewatering are also major sources of CH_4 emissions as the digestion process often continues in these steps, and small biogas bubbles in digested sludge cause supersaturation and CH_4 release.[46] Several approaches may help abate fugitive emissions from AD, such as covering the storage facility, installing vacuum degassing, switching digester feeding from in parallel to in series, or directly feeding the upgraded biogas (or biomethane) into the local gas grid instead of doing combined heat and power which is known to have minor CH_4 slippage due to incomplete combustion.[47,70] For example, negligible CH_4 emissions were detected in an on-site amine scrubber unit that was used for biogas upgrading unit, whereas CH_4

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emissions from combined heat and power were around 1.6−2.5 kg CH_4/h.[31] One should note that some of these methods are only tested in a small scale and still require more testing and optimization.

3.4. Methane Emissions from Sewer Networks. Limited literature on sewer CH_4 emissions is available, but recent results have clearly indicated that sewer networks contribute to fugitive CH_4 emissions.[20] Figure 6 summarizes the distributions of dissolved CH_4 concentrations in sewer networks with respect to operational and environmental conditions including wastewater temperature, geographical location, and HRT. The mean (lower-upper 95% CI) dissolved CH_4 concentrations from gravity, pump station, and rising main are 5.6 (4.2−7.1) mg/L, 1.0 (0.5−1.6) mg/L, and 5.3 (4.6−6.0) mg/L, respectively.[71] High dissolved CH_4 was observed from gravity sewers mainly because of extremely high dissolved CH_4 concentrations reported by one study carried out in Thailand. If that set of data was removed, the mean (lower-upper 95% CI) dissolved CH_4 concentration for gravity sewers is 0.7 (0.5−0.9) mg/L, on par or even lower than the pump station and rising main. It is worthwhile to note that the studied gravity sewers in Thailand are not representative of the majority of gravity sewers in developed countries. First, the sewers in Thailand accept large amounts of discharges from septic tanks, which have high loading of organics and methanogens that lead to more methanogenesis. Second, the high temperature (mean: 31 °C) and HRT (mean: 16 h) in Thailand compared with gravity sewer lines in other regions (temperature: 19 °C, HRT: 8 h) also contribute to high CH_4 production. For example, previous studies reported higher CH_4 production in the summer than in the winter from sewer networks.[72] A similar effect could explain the longer HRTs that lead to higher CH_4 emissions as long retention times lead to more anaerobic conditions and better growth of methanogens.[73−75] Except for temperature and HRT, many other factors also influence CH_4 production in gravity sewers such as the characteristics of the sewage, the ratio between biofilm area and wastewater volume, local pH, and nitrogen content (C:N ratio).[76,77] However, the correlations between CH_4 production and these factors are difficult to establish because of the lack of data.

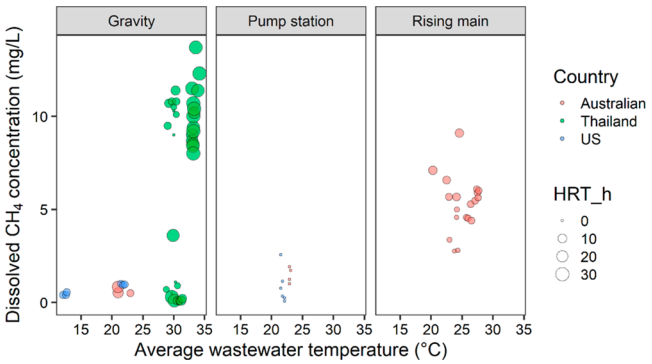

Figure 6. Dissolved CH_4 concentrations in different types of sewer networks with respect to wastewater temperature, geographical regions, and hydraulic retention time (HRT).

When comparing rising mains with gravity sewers under similar temperatures and HRT conditions, rising mains show much a higher (3−6 times) dissolved CH_4 concentration per unit length (Figure 6). This could be attributed to relatively low DO and fully wetted perimeters (and accordingly larger slime areas) in these pressurized sewer pipes. However, this does not mean the overall CH_4 emission from rising main is higher, because different from the ubiquitous emission from gravity sewer, dissolved CH_4 in rising main cannot release to the atmosphere until it reaches a pumping station or discharge manhole. Previous studies estimated that CH_4 production in three segments of rising mains (UC09, CO16, and C27 in Australia) is released to the atmosphere mainly at the downstream during preliminary treatment and the aeration process in WRRF, contributing around 18% of the plant-wide CH_4 emissions.[78] From a regional perspective, gravity sewers are still the most widespread type of sewer in most places which may dominate CH_4 production and emissions from sewer systems.[79] For example, United States has a network of over 800,000 miles of public sewers in which 92.5% are gravity sewers.[80] Nevertheless, there are still limited data and understanding on the sewer contribution to the overall CH_4 emissions so more studies are needed.

Due to the scale and complexity of sewer networks and their dynamic conditions, both manual and online sampling methods have faced many challenges. Manual sampling cannot capture all possible fluctuations of CH_4 concentrations in both liquid and gaseous phases. In addition, it is not feasible for long-term quantification over a large number of sampling sites in lengthy and sometimes hard to access sewer networks. Online sampling may provide more data availability, but current online sensors for gaseous CH_4 monitoring are not applicable in sewer conditions due to the high humidity (80−100% RH). In this context, better monitoring methods need to be developed, and mechanistic and machine learning models could be beneficial for converting wastewater operational and CH_4 data into information to inform decision making.[77]

Table 1. Characteristics of Annual Wastewater Being Treated, CH_4 Emission Intensity and the Best-Fit Distribution for Each Group of the Wastewater Sector

			CH_4 emission intensity of collected data set		
groups of wastewater sector	number or ratio	annual treated wastewater ($\times 10^9$ m^3/year)	mean ± s.d.	sample size	best-fit distribution
gravity	92.5%[80]	37.8[a]	0.7 ± 0.2mg/L	9	Lognormal (−0.4, 0.4)
rising main	7.5%[80]	3.1[a]	5.6 ± 1.8mg/L	22	Gamma (10.4, 1.8)
WRRF with AD	1223	22.6	12.5 ± 12.0 g CH_4/m^3	28	Gamma (1.4, 0.1)
stabilization pond	1394	0.5	49.5 ± 26.0 g CH_4/m^3	15	Weibull (2.1, 56.2)
other WRRFs	11572	17.7	2.5 ± 6.9 g CH_4/m^3	55	Weibull (0.5, 1.3)
total WRRFs	14189	40.9	-	-	-

[a]Note: Annual wastewater treated by gravity or rising main was calculated as the production of annual wastewater being treated by all WRRFs in the U.S. and ratio of each sewer pipe.

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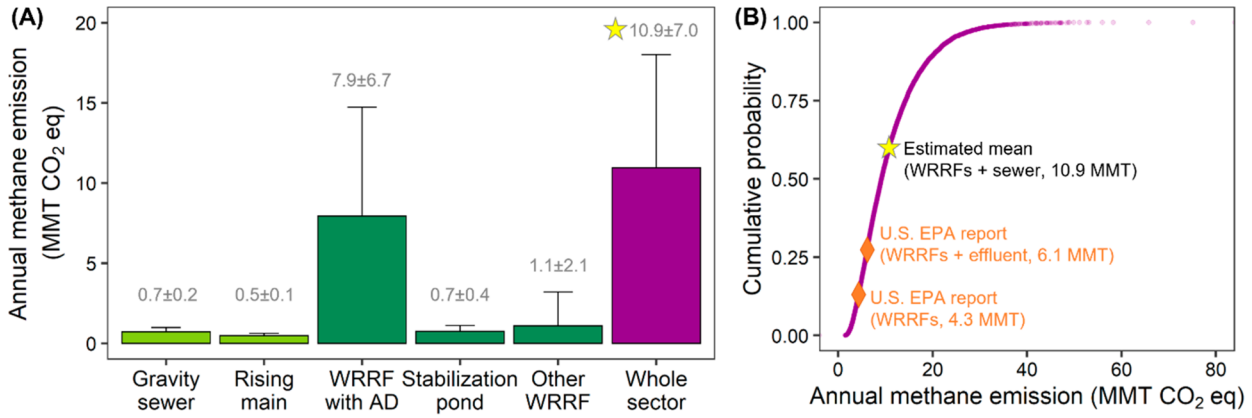

Figure 7. Nationwide CH_4 emissions from the U.S. wastewater sector. (A) Estimated annual mean (±s.d.) CH_4 flux of each group and (B) accumulative probability of CH_4 emissions from the whole wastewater sector.

Regarding CH_4 mitigation from sewer networks, a chemical dosing approach has been tested such as using oxygen, ferric salts, hydroxide, and free nitrous acid.[77] Besides these commonly used chemicals, a recent study showed that the addition of nitrite can also effectively reduce CH_4 emissions from sewers.[81] In addition, ideas have been proposed to develop engineered "end-of-pipe" CH_4 abatement solutions such as methanotrophic air biofilters, which promotes biological CH_4 consumption especially at potential CH_4 hotspots such as rising main gas relief points and headspace ventilation points.[82]

3.5. Nationwide Methane Emissions from the U.S. Wastewater Sector. The data sets curated in this study allowed us to perform Monte Carlo simulations to upscale the sewer and WRRF CH_4 emissions to the national scale with uncertainty analyses. We chose the U.S. as a case study as it has relatively high data availability on sewer type description, WRRF treatment train configurations, and wastewater flow at each facility. As described in previous sections, CH_4 emission intensities for different sewer types and WRRF configurations vary significantly. Thus, we classified sewer networks and WRRFs in the U.S. into five groups to reduce bias and minimize variance caused by different configurations (Table 1). Details about the identification of each type of WRRF and its corresponding flow rate, data sources/processing, and assumptions are provided in Section S3.1. Given the data of CH_4 emission intensities for each group are skewed, we then identified their best-fit distributions using the *fitdist()* function (R package fitdistrplus) and performed simulations separately. Detailed processes in determining which distribution fits the data best for each group in the wastewater sector are provided in Section S3.2. Nationwide CH_4 emission was the sum of CH_4 emissions from the five groups which were calculated as the production of CH_4 emission intensity and wastewater flow being treated (eq 1). A total of 10,000 random values were generated based on each group's best-fit distribution using Monte Carlo analysis (refer Section S3.3 for details). Uncertainty analyses associated with CH_4 emission differences in developing and developed countries as well as best-fit distributions are provided in Section S3.4.

$$E = \sum_{i=1}^{i=5} \mathrm{EI}_i \times Q_i \times \mathrm{GWP} \times 10^{-12} \quad (1)$$

where E is annual CH_4 emissions from the whole wastewater sector, million metric tonnes (MMT) CO_2-eq/year. EI_i and Q_i are CH_4 emission intensity and annual wastewater being treated for group i, respectively. GWP is 100-year global warming potential of CH_4, 28.[83] Conversion factor (10^{-12}) transfers units from g to MMT.

For the two groups belonging to a sewer network, their emission intensities are dissolved CH_4 concentration instead of gaseous emission intensity. Given the equilibrium solubility concentration for CH_4 in water is extremely low under ambient atmospheric conditions, there is a strong driving force for atmospheric ventilation of dissolved CH_4 from methane-supersaturated wastewater streams.[79] Thus, we assumed that all CH_4 concentrations in the sewer network are given as "excess" delta CH_4 (ΔCH_4) and represent the mass of dissolved CH_4 above the normal 100% saturation concentration (i.e., maximum potential CH_4 emission assuming 100% ΔCH_4 water-to-air mass transfer).

Figure 7A shows the estimated mean annual CH_4 emissions from each group within the centralized municipal wastewater. A total of 10.9 ± 7.0 MMT CO_2-eq/year of CH_4 was estimated to emit from the sewer networks and WRRFs in the U.S. Around 79% of the total emissions come from WRRFs with AD (7.9 ± 6.7 MMT CO_2-eq/year) and stabilization ponds (0.7 ± 0.4 MMT CO_2-eq/year), indicating they should be the priorities in monitoring and mitigation efforts. Notably, we only identified 1,223 WRRFs with AD (as of 2012, the most recent data available)[84] out of the total 14,189 municipal facilities in the U.S. Despite such a small fraction of WRRFs with AD, these are large facilities that provide the majority of the wastewater treatment services, treating 55.2% of all wastewaters as well as a slightly larger contribution (69.2%) to CH_4 emissions.

Figure 7B depicts the cumulative probability plot of nationwide CH_4 emissions from the wastewater sector obtained using the Monte Carlo simulations in comparison with the value proposed by the U.S. Environmental Protection Agency (EPA) based on the IPCC (2019) Tier 2 methodologies. The U.S. EPA reported CH_4 emission from centralized municipal WRRFs only is 4.3 MMT CO_2-eq in 2020 (refer Section S3.5 for details),[85] equivalent to 13% of the values simulated by Monte Carlo. It should be noted that the U.S. EPA recently updated the inventory on domestic wastewater CH_4 emissions and added a new category called "centrally-treated wastewater effluent", which counts the increase of CH_4

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emissions from aquatic environments (e.g., lakes, reservoirs) due to the release of dissolved CH_4 in effluent and/or the addition of organic matter from wastewater discharges.[85] With consideration of this new category (1.8 MMT CO_2-eq), the U.S. EPA reported CH_4 emission covers up to 28% of the values simulated by Monte Carlo, suggesting the IPCC (2019) Tier 2 methodologies for estimating CH_4 emissions are relatively low in relation to the measured data currently available in the literature.

The lower estimated results reported by the U.S. EPA could be largely be attributed to an underestimation of the actual CH_4 emissions from anaerobic sludge digesters as well as neglect of sewer emissions. Based on eq 7−15 in the U.S. EPA report,[85] CH_4 emission intensity of anaerobic sludge digesters equals 0.32 g CH_4/m^3 of treated wastewater, which is at least one order of magnitude lower than actual measured emission values from WRRFs with AD in the U.S. (5.6−25.5 g CH_4/m^3) or in other regions (1.7−47.3 g CH_4/m^3). Such a low CH_4 emission value calculated based on biogas generation rate per capita and destruction efficiency of flares only accounts for noncombusted CH_4 without considering other potential emissions sources (e.g., leaks at pressure relief valves, and ventilation from engine building and other tanks) which may not be accurate for estimating plant-specific variations as well as CH_4 emissions estimates. In terms of sewer systems including gravity sewers and rising main, their annual CH_4 emission is estimated to be 1.2 MMT CO_2-eq on average, which is not considered by the U.S. EPA and the IPCC methodologies. Given the high contribution and variation of national CH_4 emissions are mainly associated with WRRFs with AD, further continuous, long-term monitoring campaigns are necessary to characterize variations in CH_4 emissions from anaerobic digesters and to reduce uncertainties of countrywide emission estimations.

4. OUTLOOK AND RESEARCH NEEDS

This study presented a first comprehensive data analysis on CH_4 emissions across different wastewater collection systems and treatment processes. We identified main CH_4 sources, temporal and spatial variabilities, and potential mitigation methods. It is clear that there is still limited understanding on CH_4 emissions in the wastewater sector, and the current lower-tier IPCC methods cannot provide sufficient details for accurate emission estimates or pinpoint hot spots that require immediate attention and actions. The findings show that plant-wide CH_4 emissions for WRRFs vary greatly from 0.01 to 110 g CH_4/m^3, and different treatment processes have drastically different emissions. While most aerobic treatment units showed similar and relatively low emissions, open systems like stabilization ponds or intensified anaerobic processes such as AD and UASB are major contributors of fugitive emission. This brings a dilemma and an opportunity. Anaerobic technologies are moving to mainstream as they hold good potentials of energy savings and resource recovery, but their development and deployment need to be carefully designed and carried out with close monitoring, minimized GHG leaking, and optimized energy and resource recovery without loss.

From a life cycle perspective, anaerobic treatment with biogas recovery provides environmental benefits by reducing fossil fuel energy consumption and recovering renewable energy. Studies reported that net GHG emissions from AD could be 70−90% lower than other sludge management methods (e.g., incineration and composting).[86] However, large uncertainties of actual CH_4 emissions from different sludge management methods should be incorporated into future life cycle analysis. Considering that the U.S. EPA has qualified biogas from WRRFs as a cellulosic transportation biofuel (category D3) under the expanded Renewable Fuel Standard program in 2014, it provides financial incentives for WRRFs to increase biogas production as D3 fuels allowing for lower renewable volume obligations and hence a higher value than other forms of renewable fuel pathways.[87] As for fugitive emissions, the vacuum pump for dissolved biogas recovery has been commercialized and provides practical ways to further improve biogas capture efficiency and reduce direct emissions from AD effluent. In addition, developing guidance in retrofitting existing facilities or in designing new biogas production and utilization systems is needed to achieve carbon-neutrality and energy self-sufficiency.

Much more work needs to be done on CH_4 emission monitoring, and studies focusing on either detailed emission mapping for a plant or district, or national-multinational inventory building are urgently needed. The CH_4 emission monitoring campaigns do provide insightful information, but without site-specific data (e.g., wastewater characteristics, treatment processes, and operational conditions) it is difficult to provide accurate estimates for the wastewater sector compared to other well-studied sectors (like oil and gas). Direct comparative studies involving different measurement techniques and protocols should be conducted, and results should be analyzed, compared, and fed into the IPCC protocol considerations. Further continuous, long-term monitoring campaigns are required to characterize variations in CH_4 emissions from sewers and WRRFs, as the emissions can vary greatly depending on substrate, operation, weather, and other diurnal and seasonal changes. Development of tailored measurement approaches that aim to link CH_4 emissions to process parameters or performance indicators is beneficial compared with current labor- and time-intensive on-stie monitoring campaigns. New tools and technologies such as advanced sensors, machine learning models, and analytical methods will be essential to allow more accurate and comprehensive data collection, monitoring, and processing. Technologies and practices that address the root reasons for CH_4 emission reduction would be desired as well. These may range from covering the reactors (e.g., stabilization pond and digestate storage tank) and minimizing leakages to upgrading treatment processes, optimizing control, and increasing recovery from dissolved CH_4.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c04388.

Additional text mining methodology; normalization of methane emission intensity; upscaling to national methane emissions and uncertainty analysis; compiled CH_4 emission data from WRRFs and sewer networks; complete list of municipal WRRFs in the U.S. (PDF)

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Notes

The authors declare no competing financial interest.

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■ NOMENCLATURE

IPCC	Intergovernmental Panel on Climate Change
GHG	Greenhouse gas
GWP	Global warming potential
WRRF	Water resource recovery facility
AD	Anaerobic digestion
DO	Dissolved oxygen
COD	Chemical oxygen demand
BOD_5	5-day biological oxygen demand
HRT	Hydraulic retention time
MGD	Million gallon per day
A/O	Anaerobic/oxic reactor
A2O	Anaerobic/anoxic/oxic reactor
OD	Oxidation ditch
MLE	Modified Ludzack-Ettinger
SBR	Sequencing batch reactor
UASB	Up-flow anaerobic sludge blanket
TDM	Tracer gas dispersion method
IR	Optical gas imaging infrared video camera
EPA	Environmental Protection Agency

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