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Emerging investigators series: revisiting greenhouse gas mitigation from conventional activated sludge and anaerobic-based wastewater treatment systems

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Recent literature on carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) emissions from wastewater treatment plants (WWTPs) has highlighted the poor consensus in total greenhouse gas (GHG) estimation (ranging from 0.243 to 2.4 kg CO_{2e} per m^3). In the present study, the major components of GHG emission variability in both conventional activated sludge (CAS) and mainstream anaerobic WWTPs are systematically investigated as a basis for delineating a roadmap to their future control and minimization. Through analysis of N_2O generation pathways, it was determined that additional research *via* isotope labelling is necessary to elucidate distinct generation mechanisms in CAS WWTPs (*e.g.*, nitrifier denitrification and hydroxylamine denitrification) and better predict N_2O contributions to total GHGs. Conversely, mainstream anaerobic processes, although a potentially more sustainable alternative to conventional aerobic treatment, introduce effluent dissolved CH_4 as a potentially significant GHG contributor. Sweep gas and vacuum driven membrane contactors are promising dissolved methane management strategies. However, further optimization of gas-to-liquid ratios and transmembrane pressures, respectively, are vital to balancing treatment efficiency with energy neutral/positive operation. Overall, a thorough elucidation of N_2O generation pathways in CAS WWTPs and the development of effective dissolved CH_4 management strategies for mainstream anaerobic processes will define their respective future roles in reducing wastewater-associated GHG emissions.

Water impact

Conventional and mainstream anaerobic wastewater processes remain difficult to compare in terms of their global warming potential (GWP) due to unresolved variability in high-impact GHG emissions. Here, we identify knowledge gaps in those GHG contributors, highlight potential mitigation strategies, and provide a basis for the direct comparison of CAS and anaerobic wastewater treatment.

1. Introduction

As the risks of climate change become increasingly acute, the necessity for accurate greenhouse gas (GHG) accounting has led to a renewed focus on wastewater management as an emissions source. The most widely employed wastewater treatment methods, namely aerobic (*i.e.*, activated sludge-based processes) and mainstream anaerobic processes, both significantly contribute to GHG generation in their current forms of implementation. Mainstream anaerobic processes, which lessen WWTP energy costs and biosolids generation, are receiving renewed interest as an alternative to aerobic processes.[1] The EPA Inventory of GHG Emissions and Sinks estimates that US wastewater treatment plants (WWTPs) accounted for approximately 0.3% of overall emissions in 2016, with CH_4 and N_2O accounting for 3 and 5 MMT CO_2 equivalents, respectively.[2] CH_4 and N_2O are of particular concern due to their relatively high 100-year global warming potentials (34 and 298 CO_{2eq}, respectively),[3] with WWTPs currently estimated to be the sixth largest contributor of N_2O emissions worldwide (approximately 3%).[4] Despite the magnitude of N_2O emissions from WWTPs, our understanding of formation mechanisms and ability to model or predict emissions remains lacking.[5,6]

Today, conventional activated sludge (CAS) processes coupled with anaerobic digestion are widely used in domestic wastewater treatment, despite their high energy requirements (up to 3% of overall US electricity consumption) and lack of large-scale energy and nutrient recovery.[7] Although anaerobic

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digestion significantly offsets WWTP energy demands and reduces sludge handling requirements, it is unclear whether this conventional approach will remain attractive in light of recent advances in mainstream anaerobic treatment.[8] Mainstream anaerobic systems directly recover energy *via* biogas production, produce drastically less sludge, and have been proven viable at a range of operational temperatures.[9] Taking advantage of their favorably warm climate, several Latin American countries have long incorporated mainstream anaerobic processes, specifically upflow anaerobic sludge blankets (UASBs), for full-scale domestic wastewater management.[10,11] The integration of membrane separation and anaerobic treatment (*i.e.*, anaerobic membrane bioreactors (AnMBRs)) has greatly expanded interest in mainstream anaerobic processes worldwide.[12,13] This has led to recent advances in the technology's application at ambient temperatures, as well as promising testing at the pilot-scale.[14,15] However, loss of dissolved CH_4 in effluents is an outstanding concern. Such losses not only reduce energy recovery, but also pose severe environmental impacts due to GHG emissions.[1] A direct comparison of GHG emissions between aerobic and anaerobic processes is vital to help stakeholders navigate a potential transition to anaerobic treatment.

Several review papers have been published to address GHG emissions from conventional WWTPs, many of which have focused specifically on N_2O emissions.[5,16–18] For mainstream anaerobic treatment, a recent review by Crone *et al.* evaluated dissolved effluent CH_4 while discussing technologies for recovery.[19] Despite the significant contributions of the aforementioned reviews, N_2O and CH_4 emission quantifications for the purpose of directly comparing CAS and mainstream anaerobic treatment systems' GWP remain unavailable. The primary purpose of the current study is to systematically focus on outstanding knowledge gaps in GHG emissions limiting direct comparability of CAS and mainstream anaerobic treatment. The issues specifically evaluated in this work include CAS WWTP total GHG estimation, pathway-associated N_2O generation mechanisms in aerobic-

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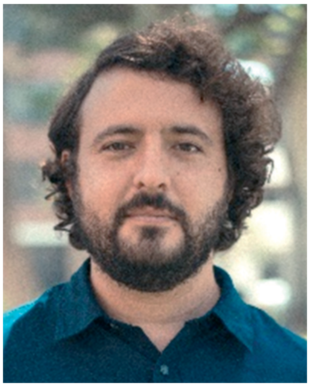

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based WWTPs, and dissolved CH_4 recovery efficiency for anaerobic system effluents.

2. Anthropogenic GHG emissions from CAS WWTPs

2.1. Overview of GHG generation in wastewater treatment processes

GHG emissions are attributable to essentially every unit process in conventional aerobic wastewater treatment coupled with anaerobic digestion (Fig. 1). Here, we categorize these emissions as either direct or indirect, where direct emissions include GHGs physically produced by either in-plant or downstream environmental processes and where indirect emissions include electrical energy demands and chemical inputs of the system. From the perspective of indirect emissions, aeration tanks comprise more than 40% of total plant energy demand[7,20,21] and are often reported as contributing most significantly to overall GHG emissions (approximately 0.298 kg CO_{2e} per m^3 based on reported U.S. energy carbon footprint of 0.472 kg CO_2 kW^{-1} h^{-1}).[22–25] Sidestream processes for primary and waste activated sludges also contribute indirect GHG emissions *via* energy demand and chemical addition during dewatering, transportation, land application, and landfilling,[26,27] which can account for between 0.134 to 0.167 kg CO_{2e} per m^3 of domestic wastewater.[28] It should be noted, however, that biogas production from anaerobic digestion can significantly offset indirect GHG emissions by lessening reliance on a potentially GHG emission heavy primary energy mix.[24,26]

Regarding direct GHG emissions, N_2O generated during denitrification, either in anoxic tanks (in the case of CAS with biological nitrogen removal) or in the receiving aquatic environment when nitrate-rich effluent is released (in the absence of on-site anoxic treatment), is considered the primary source of direct GHG emissions.[2] The EPA's Inventory of US Greenhouse Gases and Sinks reports this N_2O emission source as part of effluent emissions due to the majority of plants not employing biological nitrogen removal. However, in scenarios where nitrogen removal is achieved, these emissions are largely confined to within the plant footprint.[29] Aeration tanks are also responsible for direct N_2O generation as a result of incomplete nitrification, with their contribution to total N_2O footprint being recently identified as potentially much higher than previously considered.[2] In addition to N_2O, aeration tanks are also responsible for significant generation of CO_2 due to microbial degradation of organic carbon, however, these direct CO_2 emissions are not traditionally considered in GHG accounting because of their biogenic origin.[30] Despite this, recent research has shown that approximately 14% of total organic carbon in municipal wastewater is actually of non-biogenic origins due to domestic use of soaps and detergents, leading to an underestimation of direct GHG emissions.[31,32] Quantifying direct emissions of CAS WWTPs has proven to be the most challenging aspect of GHG estimation, as quantification methods and assumptions are wide ranging in existing literature and governmental reports. In the following, GHG emissions are normalized to volume of domestic wastewater (DWW) treated to better compare parallel studies, regardless of differences in treatment process.

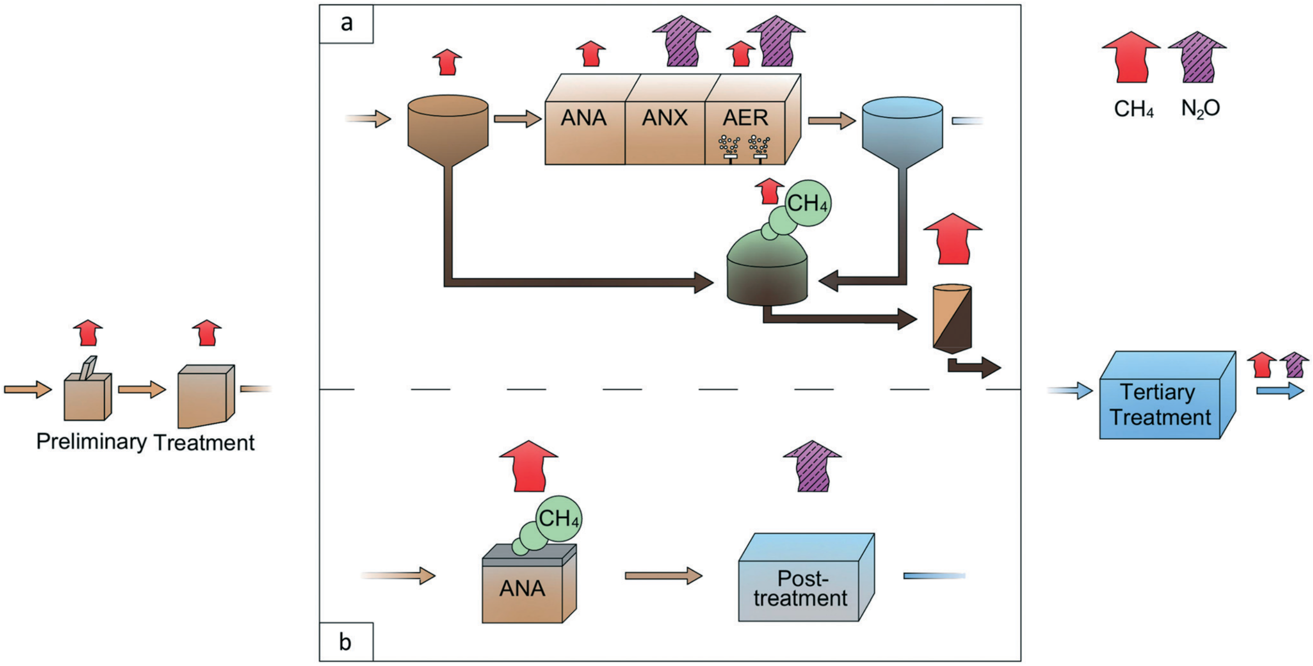

Fig. 1 (a) Direct greenhouse gas (CH_4 and N_2O) emissions from conventional wastewater treatment plants employing anaerobic/anoxic/oxic activated sludge processes and anaerobic sludge digestion (ana/anx/aer, anaerobic/anoxic/oxic tanks). (b) Direct greenhouse gas (CH_4 and N_2O) emissions from anaerobic-based wastewater treatment with post-treatment processes.

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2.2. Common findings of different quantification methods for plant-wide GHG emissions

Two common approaches have been reported for quantifying direct GHG emissions: (1) emission factor-based methods derived from dynamic modeling and (2) actual emission values determined from on-site measurement. Generally, model-based studies have reported high variability in overall plant emissions (from 0.24 to 2.4 kg CO_{2e} per m^3 DWW), with contradictory findings regarding the primary source of emissions.[20,33–35] A clear consensus has thus not yet been reached identifying the major contributors to total GHG emissions.[6] Models are typically constrained to specific plant configurations and feature inconsistent emission factors. Nonetheless, one common observation is that N_2O emissions contribute the greatest uncertainty in emissions estimation,[33,36] particularly due to its excessive GWP and the lack of a comprehensive mechanism-based model of formation.[6]

On-site quantification methods have included sampling and subsequent lab analysis,[22,23,37] on-line off-gas collection and analysis,[38–41] and tracer dispersion monitoring.[42] Normalization of on-site sampling methodology has enabled the comparison of different treatment processes, such as activated sludge, oxidation ditches, anaerobic/anoxic/aerobic processes (A^2O), and reverse A^2O.[22,37] Research taking advantage of covered treatment units and direct off-gas on-line analyzers showed correlation between operational conditions and GHG emissions. Such trends include emission increases with seasonal water temperature variation,[38] changes in aeration rates,[39] discharge of reject water to influent streams,[39] length of anoxic/oxic phases (in sequencing batch reactors (SBRs)),[40] and influent nitrite variations.[41] The use of less conventional approaches, such as tracer addition and dispersion monitoring, have generally been less accurate compared to on-site measurement. A study by Yoshida *et al.*, for example, that utilized tracers and mobile cavity ring-down spectroscopy sampling, found large variations in emissions over multiple campaigns, with CH_4 generation ranging from 4.99 to 92.3 kg h^{-1} and N_2O from 0.37 to 10.5 kg h^{-1}.[42]

Although aeration energy consumption is the primary contributor to indirect GHG emissions,[33,38,40] direct emission rates remain less clear and are a significant obstacle to achieving a plant-wide understanding.[22,23,37] Further, aeration control strategies impact both energy consumption and N_2O generation (affected by DO levels), implying that tradeoffs exist between direct and indirect GHG emissions.[20] Existing literature has reported between 0 and 14.6% of nitrogen entering WWTPs being converted to N_2O,[5,26,28,40,42] contributing 1% to 78.4% of overall plant carbon footprints.[23,24,28,38,40] Based on this extreme variability, a more thorough evaluation of literature addressing N_2O emissions in aerobic-based WWTPs is necessary.

2.3. Specific direct GHG emission sources in CAS WWTPs

2.3.1. Considering total CH_4 emissions. Unintentional methanogenic conditions in collections systems, influent piping, grit chambers, primary clarifiers, and anoxic/oxic tank dead zones all contribute to methane-based GHGs.[24,43,44] Existing studies have shown that this upstream-generated CH_4 is predominantly stripped from the liquid phase upon reaching the aeration tanks, serving as the primary source of CH_4 emissions in the mainstream portion of WWTPs (6–18 g CO_{2e} per m^3 DWW).[45–49] Further, the low organic carbon and high DO remaining in solution after aeration minimize the potential for any additional evolution and release of CH_4 in WWTP effluents (reportedly $<0.1\%$ of total CH_4 emissions).[50]

Only a few studies on GHG emissions have incorporated sidestream anaerobic digestion. Two studies by Daelman *et al.*[38,51] reported total methane-associated emissions in the range of 90–95 g CO_{2e} per m^3 DWW, showing that fugitive gasses associated with sludge handling, digester effluents, and cogeneration engine gas slip accounted for approximately three quarters (72 ± 23%) of WWTP CH_4 emissions. An analysis of studies reporting total digester CH_4 emissions (ranging from 17 to 72 g CO_{2e} per m^3 DWW) suggested that operational parameters such as WWTP SRT and anaerobic digester residence time likely play a significant role in CH_4 emission rates.[52] Digestion associated CH_4 losses, if fully recovered, could potentially increase energy recovery by 10–30%.[26] These results imply that although CH_4 is a relatively minor component of direct CAS emissions, reducing their losses in sludge treatment processes can significantly improve energy-associated GHG footprints.

2.3.2. N_2O emissions: taking generation pathways into account. N_2O generated during biological nutrient removal is one of the most variably reported phenomena known to occur in conventional WWTPs. The current US EPA guidance on national GHG inventories estimates that 0.5% of influent nitrogen will be converted to N_2O, primarily due to denitirification of effluent nitrate in receiving waterways.[30,53] This emission factor was originally developed as part of a study by Czepiel *et al.* that did not include in-plant denitrification.[54] More recent work on N_2O emissions from full-scale wastewater treatment systems, however, have reported values ranging from 0 to 14.6% of N. To elucidate source variability, a fundamental understanding of the factors affecting N_2O generation is necessary.

In CAS-based treatment, ammonium-containing wastewater is intentionally subjected to aerobic and anoxic conditions to convert nitrogen to dinitrogen gas *via* nitrification and denitrification. However, this process also has potential to contribute N_2O emissions through multiple distinct and complex pathways (Fig. 2). When autotrophic ammonium oxidizing bacteria (AOB) are present at low DO, high nitrite, or high ammonium conditions, AOB will perform denitrification, converting nitrite to N_2O (also known as nitrifier denitrification).[55] Nitrite can also independently react with coexisting organic or inorganic matter during the nitrification process to produce N_2O. Another intermediate during ammonium oxidation, known as a nitrosyl radical, has also been observed to convert to N_2O, either biologically or chemically.[56] During the denitrification process, N_2O serves as a necessary

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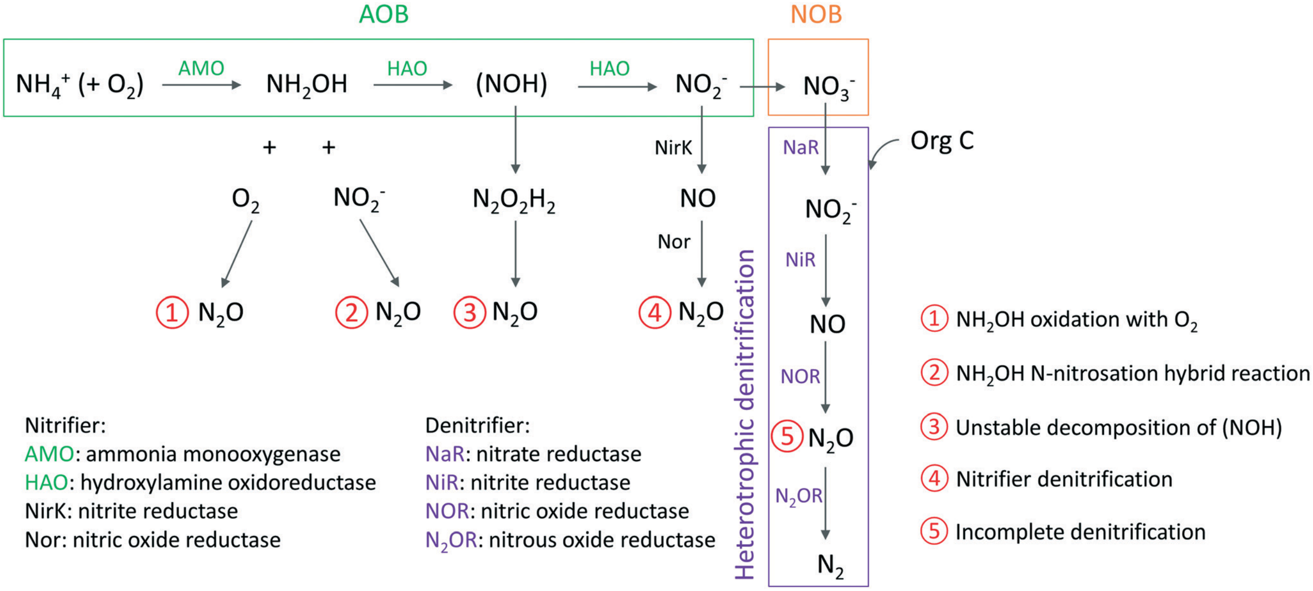

Fig. 2 Five distinct N_2O generation pathways (NH_2OH oxidation with O_2, NH_2OH *N*-nitrosation hybrid reaction, unstable decomposition of (NOH), nitrifier denitrification, incomplete heterotrophic denitrification) along with nitrification (ammonia oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB)) and denitrification pathways. Key enzymes are identified along each pathway.

intermediate and will accumulate as a result of oxygen intrusion into the anoxic environment, high nitrite concentrations, or limited carbon source availability. The cause of this accumulation is most commonly the inhibition of N_2O reductase. In other cases, the presence of a hydroxylamine intermediate during ammonium oxidizing conditions can promote N_2O generation through alternative pathways. This reaction can proceed with either oxygen as the electron acceptor (hydroxylamine oxidation) or with nitrite as the electron donor (*N*-nitrosation).

Although nitrifier and heterotrophic denitrification are considered the two main sources of N_2O, other less understood pathways likely play a significant role.[57–59] Recent studies have added inhibitors such as allylthiourea and chlorate to accredit N_2O emissions to nitrifier denitirification, heterotrophic denitrification, or NH_2OH oxidation pathways.[57,60,61] An underlying problem with this approach, however, is that these inhibitors also inhibit nitrification, in addition to the processes that generate N_2O *via* nitrifier denitrification. A promising alternative for N_2O emission source differentiation with high resolution is labelled isotope-based nitrogen species introduction and tracking.[62]

2.3.3. Operational factors affecting N_2O emissions. Environmental conditions, operational parameters, wastewater characteristics, and varying WWTP configurations can (individually or collectively) induce and/or increase N_2O generation. Further, numerous N_2O formation pathways have been identified across a range of microbially selective environments. Elucidating relationships between these variables and known N_2O generation mechanisms remains challenging. In the following, a critical analysis of the potential relationships between these two areas of literature is provided.

2.3.3.1. Dissolved oxygen control. Multiple reviews focusing on N_2O emissions from WWTPs have concluded that DO levels are primarily responsible for its generation—low DO during nitrification and high DO during denitrification.[5] However, existing literature that has investigated N_2O formation during partial and/or full nitrification has reached contradictory conclusions regarding the role of DO. For example, multiple studies on pure culture,[61,63] batch experiments,[57,64] lab-scale reactors (SBR and CSTR),[65–69] and pilot/full-scale wastewater treatment plants[68,70] have observed higher emissions at low DO conditions during nitrification. In most of these studies, the accumulation of NO_2^- was closely related to high N_2O emissions at low aeration rates. Conversely, other pure culture studies,[71,72] lab-scale experiments,[60,73–75] and a full-scale nitritation–anammox reactor investigation[76,77] have found elevated N_2O emissions under higher DO conditions. It is likely that these varying observations result from differences in microbial community structure and activity profiles leading to distinctly different formation mechanisms.

Recent studies have used nitrification inhibitor addition and/or isotope labelling of N-species to pair N_2O emissions with their specific generation pathways at varying DO levels. The use of nitrification inhibitors, specifically, has revealed decreasing relative contributions of AOB denitrification,[57,64] increasing NH_2OH oxidation contributions, and constant heterotrophic denitrification contributions to overall N_2O emissions at increasing DO.[60] Further insight provided in a study by Peng *et al.*,[74] which used isotopic site preference measurements, showed increases in NH_2OH oxidation-sourced N_2O and decreases in AOB denitrification-induced emissions with rising DO (from 0.2 to 3 mg L^{-1}). Based on the cumulative findings of these studies, it can be concluded

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that although AOB denitrification is commonly the dominant N_2O production pathway, the NH_2OH oxidation pathway could outcompete at high DO (*e.g.*, 3.5 mg O_2 L^{-1}) when combined with low NO_2^- (*e.g.*, <10 mg O_2 L^{-1}).[64] Ultimately, more applied research quantifying both gaseous and aqueous N_2O is needed, while taking into consideration all possible N_2O generation pathways.

2.3.3.2. pH. Studies investigating N_2O emissions during nitrification at different pH ranges have generally observed highest production at pH 8–8.5, independent of free ammonia and nitrous acid concentrations.[61,78] N_2O emissions during denitrification have been conversely observed to decrease with increasing pH across 5–8.5, with concurrent decreases in NO_2^-.[79–81] One study specifically found that no N_2O formation was detected at pH >6.8 and highest production occurred between 5 and 6.[80] Nonetheless, given that free nitrous acid is believed to exert a stronger inhibitory effect on N_2O reductase than pH and has been strongly correlated with N_2O production, it is possible that the relationship between pH and N_2O production during denitrification is purely incidental.[81]

2.3.3.3. Nitrite. In addition to its oxidation by O_2, NH_2OH can also serve as a precursor to N_2O formation *via* its reaction with nitrite (known as *N*-nitrosation). Even when NH_2OH is present as an intermediate in the ammonium oxidation process at low concentrations, the *N*-nitrosation hybrid reaction has still been observed to proceed in full-scale bioreactors (0.03 to 0.11 mg N L^{-1}).[82] Isotope labelled $N^{15}O_2^-$ and $N^{15}H_2OH$ have been used to distinguish respective contributions of nitrifier denitrification, the *N*-nitrosation hybrid reaction, and NH_2OH oxidation in a partial nitrifying bioreactor.[62] The *N*-nitrosation reaction was the prominent formation pathway in this study, possibly due to the relatively high DO levels. These results imply that high nitrite concentrations can result in significant N_2O formation in the nitrification process, even in the absence of nitrifier denitrification.

Increasing nitrite concentrations during denitrification have also been observed to limit the generation of NO reductase, leading to accumulation of nitric oxide (NO).[83] This can further impact N_2O emissions, as NO causes an inhibitory effect on enzymes involved in the denitrification process (*e.g.*, nitric acid and N_2O reductases). In a mixed microbial community of both nitrifiers and denitrifiers, for example, Tallec *et al.*[57] observed up to an eight fold increase in N_2O production with nitrite addition at 1 mg O_2 L^{-1}. Further, specific tests on oxidized nitrogen in an aerobic granule sludge system have shown specific N_2O generation to be approximately 44% higher in the presence of nitrite as compared to nitrate alone.[65] Although the mechanisms of N_2O formation in nitrification and denitrification processes are distinctly different, nitrite presence plays a significant role in both.

2.3.3.4. Carbon source availability/COD : N ratio. As has been reviewed,[5,16,17] limited availability of carbon sources increases N_2O production during denitrification. Although the exact mechanism by which this occurs is not fully understood, competition for electrons between various denitrification enzymes (*i.e.*, NO_3^-, NO_2^-, NO and N_2O reductase) is likely the cause.[17] Specifically, NO_3^- and NO_2^- reductases have relatively higher electron affinity than NO reductase and N_2O reductases, which induces incomplete denitrification under carbon limited conditions. Increased N_2O production in carbon source-limited environments can also be due to microbial consumption of internal storage compounds (*i.e.*, poly-β-hydroxybutyrate (PHB)).[5,17] In simultaneous nitrification/denitrification and phosphorus removal processes employing denitrifying phosphate accumulating organisms (DPAOs), N_2O generation has been observed to start immediately after the pulse addition of nitrite,[84] but further research is needed to determine the intrinsic mechanism of this phenomenon.

To maintain the minimum COD : N ratio necessary to accomplish full denitrification (typically considered to be >4), the addition of external substrate as a carbon source is often required.[17] This practice has been shown in certain instances to also significantly reduce N_2O production (by up to 95%).[75] As such, a range of external carbon source/substrate types (*e.g.*, acetate, methanol, mannitol, glucose, starch, acetic acid, sludge fermentation liquid) have been investigated for their effectiveness at curbing N_2O formation.[75,85–88] Resultantly, distinctive changes in both microbial diversity and N_2O production rates have been observed with different substrates. These differing microbial communities, which exert preferential consumption of each carbon source type, will ultimately dictate the enzymatic activity responsible for both NO and N_2O reduction.

2.3.3.5. Ammonium shock. Returning ammonium-rich reject water to the headworks can significantly contribute to N_2O emissions due to ammonium shock, especially during downstream transitions from anoxic to aerobic conditions/environments.[72,76] Given that this transition in redox conditions is often unavoidable, the accumulation of ammonium in anoxic environments should be closely monitored. Ammonium shock can also induce decreases in DO levels, potentially triggering nitrifier denitrification and subsequent elevated N_2O emissions.[89] Lab-scale work investigating this phenomenon has identified a critical ammonium loading rate of approximately 1.60 mg NH_3-N per g TSS, beyond which nitrite and N_2O increase significantly.[90]

2.3.4. Implications of N_2O emissions in CAS WWTPs. Optimization of key operational parameters (*i.e.*, sufficient carbon sourcing, pH, DO, and ammonium levels) is key to achieving predictable and minimized N_2O emission rates. A challenge associated with plant-level N_2O source identification is that nearly all N_2O is emitted from aeration tanks, regardless of formation pathway.[70,91] Therefore, more research employing isotope labelling is likely necessary to elucidate the underlying mechanisms and their contributions to overall N_2O emissions. With a better understanding of each N_2O generation pathway and its role within treatment systems, specific strategies can be devised to mitigate N_2O emissions and ultimately

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standardize operational guidelines to reduce nationwide GHG emissions.

3. GHG emission management for mainstream anaerobic treatment

Anaerobic processes are considered a sustainable and energetically favorable alternative to conventional aerobic processes. Anaerobic processes directly convert organics to methane-rich biogas and eliminate energy requirements associated with aeration.[9] However, the release of dissolved CH_4 along with discharged effluents remains a significant implementation concern, severely increasing GHG emissions while concomitantly reducing potential energy recovery.[1,19] Such losses, which in extreme cases account for up to 90% of total produced CH_4, pose a severe environmental threat if mainstream anaerobic treatment becomes the norm.[11] Still, successful mitigation of these emissions would enable anaerobic treatment with less GHGs than CAS processes, providing impetus for advancing dissolved CH_4 recovery technologies.[92]

3.1. GHG emission sources in anaerobic bioreactors

Given that the majority of GHG emission-related research on anaerobic treatment has been conducted at the bench- and pilot-scale,[93–106] full-scale indirect CO_2-based emissions estimates for electricity consumption remain largely unconfirmed. Nonetheless, energy balances of mainstream anaerobic treatment are generally expected to significantly improve upon current CAS.[107] In addition, an objective comparison of GHG emissions between CAS and mainstream anaerobic processes requires inclusion of downstream nutrient removal processes for anaerobic systems (*e.g.*, partial nitritation–anammox). Such nitrogen removal processes have been shown to emit even higher levels of N_2O than CAS, as reviewed by Massara *et al.*,[16] and necessitate further process optimization to be successfully mitigated.

Even accounting for these uncertainties, the most significant GHG-associated threat from mainstream anaerobic treatment remains effluent CH_4 losses. CH_4 saturation relative to Henry's law in anaerobic effluents has been observed to range

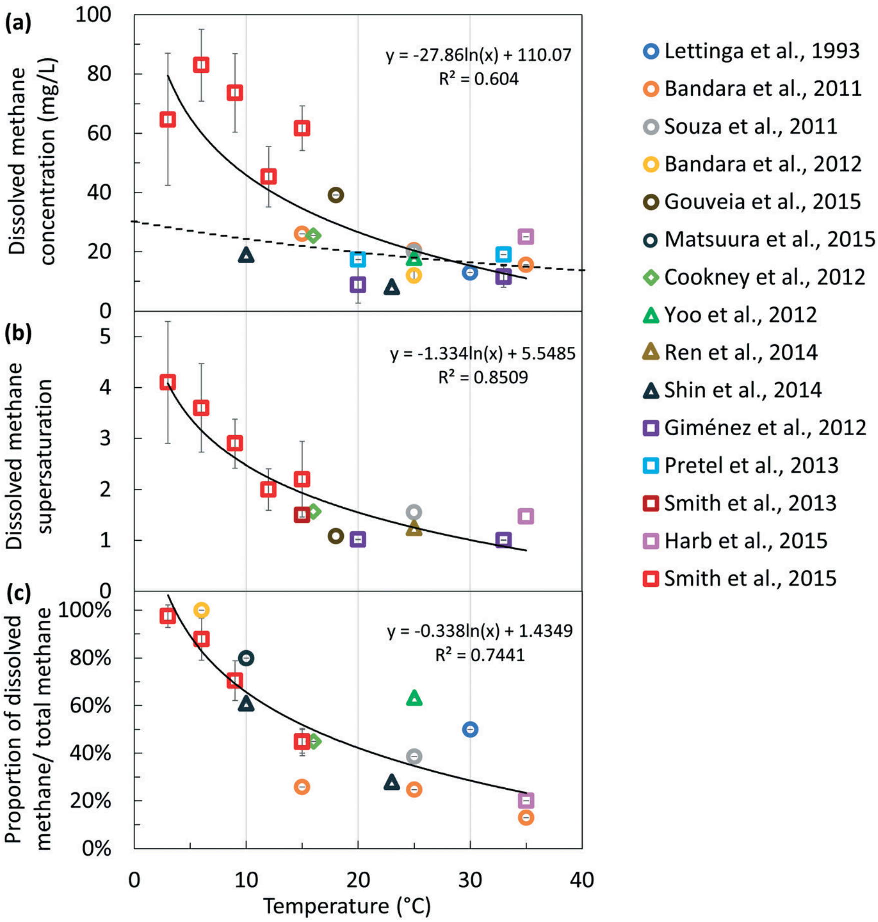

Fig. 3 (a) Dissolved methane concentration (mg l^{-1}) from anaerobic-based treatment processes across different studies at varying temperature. (b) Dissolved methane supersaturation ratio from anaerobic-based treatment processes across different studies at varying temperature. (c) Proportion of dissolved methane over total methane production from anaerobic-based treatment processes across different studies at varying temperature (○ represents upflow anaerobic blanket sludge bed reactor (UASB), ◊ represents expanded granular sludge bed reactor (EGSB), △ represents anaerobic fluidized membrane bioreactor (AFMBR), □ represents anaerobic membrane bioreactor (ANMBR); dashed line represents dissolved methane derived from Henry's law, with 80% gaseous methane in the headspace).

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between a factor of 1.0 to 5.2, resulting in the loss of 10–90% of total CH_4 produced.[19] Recent work has demonstrated robust operation (*i.e.*, COD removal) at temperatures as low as 6 °C. However, such low temperatures exacerbate GHG emission concerns by inherently increasing CH_4 solubility.[96] Overall, CH_4 solubility at low temperatures is largely responsible for increasing trends in dissolved CH_4 concentrations across all anaerobic bioreactor system types (Fig. 3a), despite reactor configuration and biogas composition also playing a role. Although the integration of membrane filtration in AnMBRs has improved effluent quality at such low temperatures, similar CH_4 oversaturation is still observed.[101,106] Studies by Smith *et al.*[106,108] on low-temperature AnMBR operation, specifically, have documented the likelihood that high methanogenic activity in membrane biofilms are responsible for dissolved CH_4 oversaturation. Experimental findings suggested that as systems increasingly relied on membrane biofilm-based treatment at decreased temperatures, biofilm methanogens directly emitted CH_4 into the effluent.[106]

Although limitations of gas–liquid transfer rates have been identified as an obstacle for recovery, *in situ* biogas sparging readily achieves gas–liquid equilibrium and maximizes CH_4 evolution to headspace. Several recent studies have demonstrated effluent CH_4 saturation factors of close to 1 and/or reduced dissolved CH_4 content by up to 50% by employing *in situ* biogas sparging.[98,103] AnMBRs operating at temperatures above 20 °C have also shown relatively low saturation factors (in the range of 1.0 to 1.1).[100,109,110] Yet, other work has shown CH_4 saturation exceeding a factor of 1.5 or greater, even when biogas sparging is sufficiently utilized.

The strongest deviation above CH_4 saturation level was observed for scenarios at 15 °C and lower (Fig. 3b). In some scenarios, no biogas CH_4 was produced, with all produced CH_4 being evolved in the effluents (Fig. 3c).[96,106] Given that this phenomenon is likely caused by disproportionate biofilm-based CH_4 production, it cannot be easily mitigated by reactor biogas sparging/stripping.[101,106] Using 34 as a standard GWP factor for CH_4, GHG emissions from dissolved CH_4 were calculated to be in the range of 0.281 to 2.82 kg CO_{2e} per m^3 DWW. This is generally comparable to the wide-ranging rates of CAS (0.24 to 2.4 kg CO_{2e} per m^3) and will ultimately necessitate downstream CH_4 recovery technologies.

3.2. Utilizing membrane contactors for dissolved CH_4 recovery

Of the physiochemical-driven methods examined for dissolved CH_4 removal and/or recovery from anaerobic effluents, the most widely tested involves membrane contactors for effluent CH_4 desorption. Relevant operational parameters of these systems include membrane properties, contact area, gas/liquid flow rates, flow direction, vacuum pressure, and gas/liquid supply sources (shell or lumen). In the following, we provide a comparative analysis of the energy use/recovery potential of the two primary modes of membrane contactor operation, namely sweep gas- and vacuum-based desorption.

3.2.1. Sweep gas membrane contactors. When operating membrane contactors in sweep gas mode, dissolved CH_4 removal in the liquid phase is driven by a concentration gradient across a gas permeable membrane into crossflowing nitrogen or air. Several studies have successfully demonstrated dissolved effluent CH_4 removal rates from UASB, AnMBR, and synthetic effluents of up to 98.9% and 92.6% using microporous and nonporous hollow fiber membrane contactors (HFMCs), respectively.[110,111] The aforementioned studies employed polydimethylsiloxane (PDMS) membranes (both microporous and nonporous types). Another recent study utilized fluorinated silica nanoparticle modified membranes to enhance surface hydrophobicity.[112] These modified membranes attained higher CH_4 recovery fluxes as compared to a commercial polypropylene microporous membranes (400–550 mg CH_4 m^{-2} h^{-} *vs.* 200–350 mg CH_4 m^{-2} h^{-1}) over 300 h of operation, implying that such surface modifications can alleviate long-term pore wetting issues.

3.2.2. Vacuum suction (degassing) membrane contactors. Membrane contactors operated in vacuum mode rely primarily on a pressure differential without significant gas cross-flow to achieve high-concentration CH_4 recovery. This allows for the direct use of captured CH_4 without further purification, but also requires additional energy input in the form of vacuum pressure. Multiple early studies by Bandara *et al.*[94,96,113] on membrane degasification for UASB effluents using a commercial multi-layer composite polyethylene hollow-fiber membrane contactor (HFMC) successfully desorbed 77% to 86% of dissolved CH_4 from UASB effluents into the lumen at vacuum pressures of 50 and 80 kPa.[94] Lumen-side liquid flow (as opposed to shell-side), has generally been observed as more effective at CH_4 desorption due to superior liquid to air transfer rates, however it can be limited by hollow fiber flow-path clogging over long-term operation.[19,114]

3.3. Analysis of energy demands and recovery by membrane contactors

Superior removal rates are achievable by vacuum degasification as compared to sweep gas operation.[115] Further, vacuum desorption has specific advantages associated with direct on-site CH_4 use, which are not achievable by sweep gas contactors. Such advantages, however, must be evaluated in comparison with the greater energy requirements of vacuum-driven transmembrane pressure (TMP). Given the knowledge gaps in literature from the perspective of energy use and recovery, a comparative analysis of the practical limitations of each CH_4 recovery method is necessary to assess each technology's economic feasibility and overall GWP.

As summarized in Table 1, CH_4 concentrations for sweep gas driven membrane contactors are generally less than 2.4% of total off-gas volume, with only one case demonstrating relatively high concentrations of 23.2% with a polypropylene HFMC operated at low gas to liquid (G/L) ratios.[97,110–112,114–116] In most cases, sweep gas driven membrane contactors exhibited increasing effluent removal

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Table 1 Dissolved methane concentration, recovery efficiency (RE), crossflow velocity ratio, sweep gas flowrate, recovered methane flowrate, and off-gas methane purity in sweep gas membrane contactors (PDMS = polydimethylsiloxane, PP = polypropylene)

Study	Dissolved methane concentration (mg L^{-1})	RE (%)	V_{gas}/V_{liquid} or Q_{gas}/Q_{liquid}	Sweep gas flowrate (N_2, m^3 s^{-1})	Recovered methane flowrate (m^3 s^{-1})	Ratio of methane/nitrogen in off-gas
Cookney *et al.* 2016, PDMS, nonporous[110]	21.0	92.6	825	3.10×10^{-2}	1.24×10^{-9}	4.01×10^{-8}
Cookney *et al.* 2012, PDMS, hollow fiber[97]	12.9	72.0	70.0	1.41×10^{-5}	6.21×10^{-9}	4.42×10^{-4}
Wongchitphimon *et al.* 2017, polymer-fluorinated silica composite, hollow fiber (Mo-MT-A)[112]	Tap water saturated with 60 : 40 CH_4/CO_2	NA	0.497	3.33×10^{-7}	1.15×10^{10}	3.46×10^{-4}
Rongwong *et al.* 2017, in-house fabricated hollow fiber membrane[116]	Hollow fiber anaerobic bioreactor pilot plant effluent bubbled with 60 : 40 CH_4/CO_2	<65.0	0.106	3.33×10^{-7}	4.58×10^{-9}	1.37×10^{-2}
Henares *et al.* 2016, PDMS, nonporous[114]	30.0	74.0	7.51×10^{-6}	7.51×10^{-7}	3.39×10^{-9}	4.52×10^{-3}
Henares *et al.* 2016, PP, microporous[114]	30.0	98.4	1.94×10^{-4}	2.20×10^{-7}	5.11×10^{-8}	2.32×10^{-1}
Henares *et al.* 2016, PDMS, nonporous[115]	30.0	75.0	6.94	6.94×10^{-7}	3.44×10^{-9}	4.95×10^{-3}
McLeod *et al.* 2016, PP, hollow fiber[111]	18.0	90.0	1.00	1.70×10^{-6}	4.21×10^{-8}	2.48×10^{-2}
Henares *et al.* 2018, PP, porous[165]	31.1 ± 3.1	98.0	6.34	7.22×10^{-6}	5.42×10^{-8}	7.50×10^{-2}

efficiencies at higher gas to liquid (G/L) ratios, which also lead to decreased CH_4 off-gas purity. The majority of studies to date investigating sweep gas membrane contactor use have had a primary objective of reducing effluent concentrations to eliminate combustion risks in downstream discharge piping. Therefore, the systems were not operated to achieve optimal off gas concentrations. Without post-removal purification of sweep gas, limited approaches are available for utilization, as CH_4 concentrations are generally too low for even basic off-gas flaring (*i.e.*, >5% CH_4). However, recent developments have shown that direct combustion *via* thermal/catalytic flow reversal reactors, regenerative/catalytic oxidation, or lean burn-gas turbine combustion can be achieved at CH_4 concentrations as low as 1%.[117] Implementing such air-based off-gas in on-site cogeneration plant engines has been proposed previously for anaerobic digester dewatering process gasses.[51] These applications, in combination with optimization of G/L ratios, could lead to sweep gas membrane contactors being a viable option for effluent CH_4 GHG mitigation.

A recent review by Crone *et al.*[19] calculated an energy input to recovery ratio of 1.0 for effluent CH_4 recovery using vacuum driven membrane contactors. However, given the high variability in existing literature associated with operational parameters of vacuum degasification, a more comparable evaluation of these variables is necessary. Table 2 provides a normalized summary of studies on vacuum-driven membrane contactors, their energy requirements, and the potential energy content of recovered CH_4 using a unified methodology. Results of multiple studies suggest that systems operated at TMPs between 14 and 50 kPa are generally energy positive while maintaining CH_4 recovery rates between 60 and 90%.[94,113–115] Further, CH_4 recovery in different scenarios did not improve substantially with increasing vacuum pressure, proving that low vacuum scenarios are generally effective. Based on these observations, operating vacuum driven membrane contactors at relatively low vacuum pressures (<50 kPa) can enable the entire HFMC system to be energy neutral/positive while achieving sufficient effluent GHG reduction.

3.4. Biological approaches

Biological oxidation is a promising alternative strategy for CH_4 removal from anaerobic effluents. The most common technique is the downflow hanging sponge (DHS). This method has proven highly effective by numerous studies, as previously reviewed.[19] In comparison, another potentially promising treatment system that has yet to be fully investigated is known as denitrifying anaerobic CH_4 oxidation (DAMO), and is reviewed herein.

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Table 2 Dissolved methane concentration, removal efficiency (RE), transmembrane pressure, suction flowrate, vacuum energy, electricity generated from desorbed gas, and electricity consumption/recovery ratio. The following constants were used in calculations: methane molecular weight, 16 g mol^{-1}; Ns, vacuum stage, 1; η, vacuum pump efficiency, 0.5;[166] atmospheric pressure, 101.325 kPa; methane energy density by combustion, 55.5 MJ kg^{-1}; electricity generation efficiency from methane combustion, 35%; P_2, vacuum pump discharging pressure, 101.325 kPa. Bold figures represent operational conditions that accomplish net energy recovery. (PDMS = polydimethylsiloxane, PP = polypropylene)

Study	Dissolved methane concentration (mg L^{-1})	RE (%)	Transmembrane pressure (kPa)	Suction flowrate, Q (assuming pure methane, m^3 s^{-1})	Vacuum pump/compressor energy $\left(\frac{\lambda \mathrm{RTW}}{\lambda-1}\left[\frac{P_2}{P_1}^{\left(\frac{\lambda}{\lambda-1}\right)}-1\right], \mathrm{J\,s^{-1}}\right)$[13]	Electricity generated from desorbed gas (J s^{-1})	Electricity converted from recovered methane/electricity consumption
Cookney *et al.* 2012, PDMS, nonporous[97]	12.9	77.0%	**30.8**	1.15×10^{-5}	**18.9**	$\mathbf{1.50 \times 10^{2}}$	**7.96**
Bandara *et al.* 2011, multi-layered composite hollow fiber membrane[94]	12.5–25.0	<89.0%	**50.0**	2.81×10^{-10}	**0.151**	**0.239**	**1.58**
			80.0	5.90×10^{-10}	14.7	0.502	0.0340
			80.0	9.73×10^{-10}	24.3	0.854	0.0352
			80.0	6.27×10^{-10}	15.7	0.570	0.0364
Henares *et al.* 2016, PP, microporous[114]	30.0	70.0%	**14.0**	3.64×10^{-8}	**0.0143**	**0.462**	**32.4**
		82.0%	**50.0**	4.26×10^{-8}	**0.331**	**0.541**	**1.64**
		94.0%	80.0	4.88×10^{-8}	17.6	0.620	0.0352
Luo *et al.* 2014, hollow fiber membrane[167]	~15.0	86.0%	94.0	6.77×10^{-10}	1.70×10^{3}	0.572	0.000337
			94.0	1.58×10^{-9}	3.96×10^{3}	1.33	0.000337
			94.0	2.31×10^{-9}	5.80×10^{3}	1.95	0.000337
Henares *et al.* 2016, PDMS, nonporous[115]	30.0	60.0%	**14.0**	2.75×10^{-9}	**0.00108**	**0.0350**	**32.4**
		70.0%	**50.0**	3.21×10^{-9}	**0.0249**	**0.0408**	**1.64**
		75.0%	80.0	3.44×10^{-9}	1.24	0.0437	0.0352
Bandara *et al.* 2013, 3-layer composite hollow fiber membrane[113]	17.3	90.0%	**50.0**	1.99×10^{-9}	**0.0155**	**0.0245**	**1.58**
		95.0%	70.0	2.80×10^{-9}	0.191	0.0344	0.180

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3.4.1. Effective CH_4 removal by the downflow hanging sponge (DHS). Several recent studies have utilized DHS bioreactors for the aerobic oxidation of dissolved CH_4 with relatively consistent removal results.[105,118–120] Through the optimization of operational parameters such as wastewater composition and air flowrates, systems have achieved removal of multiple residuals (*e.g.*, CH_4, ammonium, sulfur, *etc.*). While some have employed varying HRTs and aeration rates to achieve removals of up to 97% of dissolved CH_4 using single stage DHS reactors,[118] other work has shown that two-stage DHS systems are capable of both recovering CH_4 in off-gas at high concentrations (>30%) and oxidizing the remaining content to achieve near complete removal of dissolved CH_4 (>99%) from effluents.[119] Air flowrate is a critical operational parameter, as varying oxygen affinity and growth rates among different microbial communities significantly affect removal.[120]

3.4.2. The case for denitrifying anaerobic CH_4 oxidation (DAMO). A more recently proposed method, known as DAMO, provides a potential solution for CH_4 removal through its use as an electron donor. DAMO archaea are capable of reducing nitrate to nitrite while DAMO bacteria convert nitrite to nitrogen gas. Combining DAMO and anammox has recently been proposed as a means for simultaneous nitrogen and CH_4 removal from anaerobic effluents.[121] Nitrate reduction by DAMO archaea and nitrite reduction by DAMO bacteria with CH_4 oxidation are achieved through the sequential CH_4 oxidation processes below,

$$CH_4 + 4NO_3^- \rightarrow CO_2 + 4NO_2^- + 2H_2O$$

$$3CH_4 + 8NO_2^- + 8H^+ \rightarrow 3CO_2 + 4N_2 + 10H_2O$$

while nitrite reduction/ammonium oxidation by anammox concurrently produces nitrate:

$$NH_4^+ + 1.3NO_2^- \rightarrow N_2 + 0.3NO_3^- + 2H_2O$$

Recent studies by Chen *et al.*[122,123] have developed a system based on this model using a membrane biofilm reactor (MBfR). Multiple lab-scale investigations have shown that biofilms containing cocultures of DAMO and anammox microorganisms can achieve sufficient nitrate and nitrite reduction and ammonia oxidation.[124,125] Ultimately, the application of a combined anammox and DAMO process could offer significant economic and practical advantages over conventional practices if successfully combined with anaerobic systems. The implementation of this process for treatment of anaerobic bioreactor effluents, however, is highly dependent on the co-enrichment of specific DAMO and anammox organisms and the supplementation of nitrite to the system. Although magnetically stirred gas lift reactors (MSGLRs), MBfRs, and granular sludge reactors have all been identified as capable of supporting growth of DAMO microorganisms and retaining biomass effectively,[122–124,126–129] the most feasible option thus far for integration of anammox and DAMO is MBfRs.

Although such applications are still in their infancy, there are multiple practical advantages to applying DAMO as part of anaerobic effluent treatment processes.[130,131] With CH_4 as the sole electron donor for DAMO microorganisms, no additional organic carbon sources would be needed. Further, the slow growth rates of DAMO microorganisms such as *M. oxyfera* (doubling time of 1–2 weeks), and low yields of DAMO microorganisms in general,[122,128] alleviate the necessity of sludge disposal. Oxygen delivery *via* hollow fiber membrane units[123,127] or granular-based optimization of oxygen levels[128] require further investigation to practically alleviate the negative impacts of aeration on anammox/DAMO. Nonetheless, research thus far on MBfRs and granular sludge reactors suggests that they may soon be a feasible basis for post-treatment of anaerobic effluents.

3.4.3. MFCs as an alternative biological process. Microbial fuel cells (MFCs) have also been considered for effluent dissolved CH_4 management.[132–135] MFCs are bioelectrochemical systems where exoelectrogenic microorganisms oxidize organics and directly deposit electrons onto an anode.[136–138] Methane, as an organic substrate, can be used as an energy source to drive MFCs, converting it directly to electricity.[132–134,139–141] For example, a study by McAnulty *et al.*[133] manipulated engineered archaeal strains to produce acetate from CH_4 anaerobically *via* methyl coenzyme M reductase, subsequently generating electricity in a two-chamber MFC. Chen *et al.*[134] also reported electricity generation from CH_4 using a single-chamber MFC while observing microbial interactions between aerobic methanotrophs and exoelectrogenic *Geobacter*. It should be noted that these emerging biological processes, and specifically MFCs, require further optimization in terms of capital cost reduction and achieving consistent treatment performance before scaling up to pilot- and full-scale application.[142,143]

3.5. Implications of physical *vs.* biological systems for CH_4 mitigation

It should be noted that the aforementioned biological approaches, while potentially requiring less energy input than membrane contactors and still mitigating GHG emissions, do not capture CH_4 for energy recovery. Although this is a significant limitation for DHS systems, DAMO's use of CH_4 as an electron donor for nitrate reduction provides an alternative route to its utilization when nitrogen removal is required (*e.g.*, effluent discharge to nitrogen-sensitive waterways). MFCs, although only recently demonstrated for methane, could be advantageous over both physical CH_4 recovery (using membrane contactors) and other biological approaches due to their ability to directly recover energy.

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4. Future GHG management perspectives

4.1. N_2O as an energy recovery oxidant

N_2O, as a powerful oxidant, has the potential to be selectively produced in wastewater treatment processes (*e.g.*, *via* coupled aerobic–anoxic side-stream nitrogen removal) and serve as a combustion oxidant in combination with CH_4.[144,145] In conventional WWTPs, N_2O off-gas collection could be accomplished *via* the installation of covers on treatment unit processes, however this may be impractical for nitrification due to the large volume of gas produced by aeration. Although selective reduction of N_2O has been practiced industrially, similar processes may not be cost-effective in large application to low N_2O containing gases as they require catalysts and high reaction temperatures.[146] Nevertheless, with consistent N_2O production and improved collection efficiency in newly designed processes,[84] this could be a worthwhile future research topic.

4.2. Technologies for targeting CO_2 capture

Given the potential significance of non-biogenic CO_2 sources in municipal wastewater influents, it is important to also consider possible means for direct CO_2 sequestration or capture. However, considering the relatively high solubility of CO_2 in water and its potential cost of recovery, the treatment of dissolved CO_2 *in situ* using emerging technologies has recently become a topic of interest. For example, phototrophic technologies relying on algae and/or phototrophic bacteria could promote carbon fixation while simultaneously achieving nutrient removal.[147] Alternatively, carbonic anhydrase, a ubiquitous enzyme capable of catalyzing the hydration of CO_2 into bicarbonate and hydrogen at high rates, could potentially be incorporated into engineered systems to sequester carbon directly.[147] Microbially assisted electrolytic systems also have the potential to sequester and convert CO_2 to bicarbonate using either wastewater or seawater as the electrolyte while producing beneficial products such as H_2.[147,148]

4.3. Methanotroph-based recovery of high-value end products

Methanotrophs can be metabolically engineered to synthesize a range of high-value products including single cell proteins, biopolymers (*e.g.*, polyhydroxyalkanoates, PHB), soluble metabolites (*e.g.*, methanol, formaldehyde, formate), lipids, lycopene, C30 carotenoid, lactic acid and exopolysaccharides.[149–152] Methane oxidation is a multi-step process in which CH_4 is oxidized to methanol, formaldehyde, formate, and CO_2 sequentially. Given that methanol dehydrogenase is located in the periplasmic membrane of methanotrophs, methanol must be transported out of the cell membrane in order for subsequent processes to ensue. Based on this, methanotrophs can be genetically engineered or supplemented with inhibitors to suppress this dehydrogenase and stop CH_4 oxidation at methanol,[153] which can then be collected and enriched for use in MFCs (currently commercially available and used as portable electricity sources). The application of these processes, among others, could be especially useful when remote electricity generation is needed, the energy for which could be supplied exclusively from treated wastewater.

4.4. Methane adsorbent-related technology

Recent studies have demonstrated that structures with high adsorption capacity and packing density can be used to adsorb and store CH_4. For example, an investigation by Bagheri *et al.*[154] demonstrated that microporous activated carbon generated from corn cobs was capable of high levels of CH_4 adsorption (150 v/v). Other materials such as constructed multilayer graphene nanostructures (MGNs) with optimized layer distances were able to satisfy the U.S. department of engineering target for adsorbents (180 v/v).[155] Although the aforementioned materials require pressures of >100 psi to effectively sorb CH_4 into their structures (reducing their viability from an energy and GHG footprint perspective), the recent synthesis of a monolithic metal–organic framework has proven capable of reaching a CH_4 packing density of 259 v/v at pressures previously comparable to those of half of its capacity.[156] The continuous improvement of adsorptive materials and the potential for their exploration at lower sorption pressures could lead to viable use for CH_4 capture, purification, and transport from wastewater effluents.

5. Perspectives on the direct comparison of CAS- and anaerobic-based GHG emissions

Based on a normalized analysis of existing literature, herein we provide a parallel assessment of CAS-based and anaerobic-based mainstream wastewater treatment using their dominant GHG sources and assuming equivalent levels of treatment for effluents (*i.e.*, nitrogen and COD removal). Recent studies have shown that approximately 0.298 kg CO_{2e} per m^3 of GHG emissions come from electricity usage in conventional WWTPs (based on 0.472 kg CO_2 kW^{-1} h^{-2} energy carbon footprint),[23,24,40] with 50% of that energy demand being consumed by aeration.[7] It is estimated that roughly 25% of plant electricity use can be offset by energy produced from sludge digestion.[7] Taking these values into account, total fossil fuel-generated emissions from conventional treatment would be on the order of 0.224 kg CO_{2e} per m^3 DWW. Total fugitive CH_4 emissions were also included at an average of 0.095 kg CO_{2e} m^{-3}.[51]

Assuming full-scale mainstream anaerobic treatment energy demands are comparable to those of conventional WWTPs before considering aeration requirements, it can be anticipated that their electricity consumption accounts for approximately 0.149 kg CO_{2e} per m^3 DWW. The amount of energy achievable from direct biogas recovery (headspace)

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was further calculated based on methane loss values extracted from Fig. 3 at 25 °C and 10 °C (15 and 50 mg CH_4 L^{-1}, respectively). Assuming a 95% conversion of incoming COD (430 mg L^{-1}) to CH_4, energy density of 55.5 MJ kg^{-1}, conversion efficiency to electricity of 35%, and a CO_2 emission factor from electricity usage of 0.472 kg CO_2 kW^{-1} h^{-1},[157] it is estimated that anaerobic mainstream treatment electricity-associated GHG footprints could be reduced to below 0.02 kg CO_{2e} per m^3 at 10 °C, while actually achieving energy positive operation at 25 °C (−0.073 kg CO_{2e} per m^3). Based on these calculations, it can be concluded that mainstream anaerobic treatment has the potential to more significantly offset electricity-associated GHG emissions when compared to conventional WWTPs with anaerobic digestion (0.224 kg CO_{2e} per m^3). However, without downstream CH_4 recovery, anaerobic effluents would contribute GHG emissions of approximately 0.51 and 1.70 kg CO_{2e} per m^3 at 25 °C and 10 °C, respectively (using CH_4 GWP of 34 and dissolved methane-temperature relationship obtained from Fig. 3).

N_2O emissions from the nitrogen removal process in WWTPs have been identified as the most widely varying and least predictable of GHG sources (ranging from 0 to 14.6% of incoming nitrogen). Nonetheless, an analysis of several representative full-scale studies of conventional anoxic–oxic processes revealed an average emission factor of 1.5%,[37,38,42,158–160] resulting in N_2O emissions of 0.281 kg CO_{2e} per m^3 DWW for conventional WWTPs (assuming influent of 20 mg N L^{-1} and N_2O GWP of 298). If nitritation coupled with anammox is employed as the nitrogen removal process for mainstream anaerobic treatment and an average nitrogen to N_2O ratio of 2.8% is used (estimated from full-scale nitration–anammox studies),[76,77,161,162] N_2O emissions for anaerobic treatment can be calculated as 0.529 kg CO_{2e} per m^3 WW. The relatively higher emissions observed for nitration–anammox have been attributed to a lack of process optimization for N_2O mitigation,[16] which can likely be improved upon significantly in future research.

Therefore, total GHG emissions from CAS WWTPs are significantly lower than mainstream anaerobic systems, even at 25 °C (0.599 *vs.* 0.966 kg CO_{2e} per m^3 WW). Anaerobic treatment GHG footprints would likely be exacerbated at lower ambient temperatures, reaching up to 2.25 CO_{2e} per m^3 at 10 °C, if no effluent recovery was employed. As outlined in this review, however, emerging techniques for both nitrogen and dissolved CH_4 removal/recovery could effectively negate these outstanding issues. If, for example, energy-efficient dissolved CH_4 recovery is employed and comparable N_2O emissions are achieved through nitrogen removal processes optimization, mainstream anaerobic system GHG footprints would easily drop below those calculated for current CAS WWTP processes. Further, recent research has implicated mainstream anaerobic effluents as likely to become acceptable for direct irrigation reuse from a microbial safety perspective.[163,164] This application of nutrient-rich treated effluents could negate the necessity of nitrogen removal, essentially allowing for the elimination of direct N_2O emissions from mainstream anaerobic treatment and such systems to approach carbon neutrality.

6. Conclusions

Existing literature on WWTP GHGs has reported broadly varying total emissions ranging from 0.243 to 2.4 kg CO_{2e} per m^3 WW. A unified and comprehensive plant-wide approach inclusive of all direct and indirect emissions is necessary for accurate WWTP carbon footprint interpretation. Overall, the most significant obstacle facing GHG mitigation in CAS WWTPs is associated with understanding N_2O generation, whereas for the sustainability of mainstream anaerobic wastewater treatment, dissolved CH_4 emissions are of greatest concern. Other specific observations of this review are summarized as follows:

- N_2O emissions are both dominant and highly variable in conventional aerobic-based WWTPs (0 to 95% of N for lab-scale and 0 to 14.6% of N for full-scale), with several critical factors influencing this variability including: DO, pH, nitrite, carbon source availability and ammonium loading.
- More research is specifically needed in elucidating the pathways involved in N_2O formation (*i.e.*, nitrifier denitrification, heterotrophic denitrification, and hydroxylamine oxidation) at different operational conditions, which can be then used to correlate practical mitigation strategies with specific processes and configurations.
- Dissolved CH_4 contributions in mainstream anaerobic treatment account for the majority of GHG emissions. Anaerobic system GHG emissions are inversely correlated with operational temperature due to increasing CH_4 solubility and supersaturation (section 3.1).
- Analysis of membrane contactors for physical dissolved CH_4 removal showed that for sweep gas systems, gas to liquid (G/L) ratio is a critical parameter influencing CH_4 removal efficiency and off-gas CH_4 concentration. Vacuum driven membrane contactors, although capable of high-quality gas recovery, require operation at TMPs below 50 kPa to achieve energy neutrality.
- Several emerging methods for dissolved CH_4 recovery are likely to play significant roles in future management of dissolved CH_4. For example, DAMO combined with anammox could allow for the simultaneous removal of both nitrogen and CH_4 from anaerobic effluents.

To significantly reduce WWTP GHG emissions, future research on CAS must focus on N_2O management strategies to minimize emissions. For anaerobic systems, both efficient CH_4 and nitrogen resource recovery must be achieved without introducing incidental increases in N_2O generation. The accomplishment of this goal appears to be within reach, given the prospects of emerging CH_4 recovery processes and likelihood of effluent reuse.

Conflicts of interest

There are no conflicts to declare.

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